ATTO-X

10th International Conference on Attosecond Science and Technology

July 6th-11th, 2025
The Loop at Science Village, Lund, Sweden
Stadshallen, Lund, Sweden

www.attox.se

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Hosted by

Lund University | LLC Lund Laser Centre

Sponsored by

Carl Tryggers Stiftelse för Vetenskaplig Forskning
Crafoordska stiftelsen, grundad av Holger Crafoord 1980
Lund University | LTH Faculty of Engineering
The Royal Physiographic Society of Lund, Academy for the Natural Sciences, Medicine and Technology
LINXS Institute of Advanced Neutron and X-ray Science
WACQT Wallenberg Centre for Quantum Technology
City of Lund
ASML
Optica

PROGRAM AT A GLANCE

ATTO-X

10th International Conference on Attosecond Science and Technology

Time	Sunday 6 July	Monday 7 July	Tuesday 8 July	Wednesday 9 July	Thursday 10 July	Friday 11 July
		The Loop - Science Village			Stadshallen - City Center	
08:30		Opening				
08:45		Attosecond physics and spectroscopy	Laser technology for attoscience	Attosecond chemistry		Attosecond science at FELs
09:00		Tutorial: Marc Vrakking	Tutorial: Zenghu Chang	Tutorial: Morgane Vacher	Excursion	Invited: Carlo Callegari
09:15						
09:30		Invited: Luca Argenti	Yoann Pertot	Invited: Vincent Wanie		Invited: Robin Santra
09:45			Laszlo Veisz	Rafael Menezes Ferreira		
10:00		Federico Vismarra	Yuval Kern	Arnab Sen		Ulf Saalmann
10:15		Sizuo Luo	Martin Kretschmar			Praveen Kumar Maroju
10:30		Coffee	Coffee	Coffee		Matthieu Guer
10:45						Coffee
11:00		Invited: Yann Mairesse	Invited: Christian Brahms	Invited: Alicia Palacios		Invited: James Cryan
11:15						
11:30		Noa Yaffe	Gaspard Beaufort	Jia-Bao Ji		Juliette Leroux
11:45		Gergana D. Borisova	Bruno Schmidt	Jason Greenwood		Yu He
12:00		Jonathan Dubois	Nikoleta Kotsina	Andrés Ordóñez		Felix Egun
12:15		Lunch (1st IPC meeting)	Lunch	Lunch (2nd IPC meeting)	Lunch	Veronica Guo
12:30						
12:45						Closing
13:00		Poster session 1	Poster session 2	Poster session 3	Invited: Angana Mondal	Lunch to-go & transfer to the Physics Department
13:15						
13:30					Invited: Mohammed Hassan	
13:45						
14:00					Matteo Lucchini	
14:15						
14:30		Attosecond quantum science	Attosecond HHG sources	Attosecond science in condensed materials	Boyang Ma	Lab visits
14:45		Invited: Emilio Pisanty	Invited: Balazs Major	Invited: Stefanie Gräfe		
15:00		Invited: Kenichi Ishikawa	Invited: Caterina Vozzi	Invited: Mauro Nisoli	Yiqi Fang	
15:15						
15:30	Registration	Gabriel Granveau	Thierry Ruchon	Tristan Kopp	Coffee	
15:45		Gustav Arvidsson	Rodrigo Martin-Hernandez	Alexander Magunia		
16:00		Jonathan Pölloth	Melvin Redon	Coffee	Invited: Tran Trung Luu	
16:15						
16:30		Coffee	Coffee	Attosci. in cond. mat.	Invited: Álvaro Jiménez-Galán	
16:45		Invited: Marcus Dahlström	Invited: Bernd Schütte	Invited: Nirit Dudovich	Mette Gaarde	
17:00						
17:15		Invited: Hamed Merdji	Matthias Kübel	Invited: Peter Hommelhoff	Jiro Itatani	
17:30			Parnia Bastani	Giacomo Inzani		
17:45		Giulio Vampa	Miguel Angel Silva-Toledo	Nicola Di Palo	Mingle	
18:00		Marcelo Ciappina	Eric Constant	Romain Cazali		
18:15		Matan Even Tzur	Nataliia Kuzkova	Zhaopin Chen	Nobel session	
18:30			Sponsor Night			
18:45	Welcome reception				Transfer	
19:00						
19:15			The origins of time-dependent theory in attosecond physics: The science of Kenneth Kulander			
19:30						
19:45						
20:00			Sponsor Night		Conference dinner	
20:15						
20:30						
20:45						
21:00						
21:15						
21:30						
21:45						
22:00						

WELCOME MESSAGE

We welcome you to the 10th International Conference on Attosecond Science and Technology (ATTO X), held from July 6th to July 11th, 2025 in the city of Lund, Sweden.

ATTO is the largest and most prestigious international conference in the field of attosecond science and technology, traditionally held every two years. At ATTO, state-of-the-art research works are shared among scholars from academia, research institutes and industries from all over the world.

Lund is a student-filled city in the southern part of Sweden. The university was established in 1666 and is located in the heart of the city, but is now extending to the outskirts with major research facilities: MAX IV, the first 4th generation synchrotron radiation light source, and ESS, the new European Spallation Source.

We are excited to welcome you in the new scientific and innovation center, called Science Village, during the first four days (July 6th-9th) of the conference before moving to Stadshallen to enjoy the atmosphere of the historical city center (July 10th-11th).

Looking forward to meeting all of you at ATTO X!

Per Eng-Johnsson and Anne-Lise Viotti (chairs)
On behalf of the local organizing committee

10 EDITIONS OF ATTO CONFERENCES

ATTO stands as the most prestigious global conference in attosecond science, encompassing a wide range of topics including attosecond physics and spectroscopy, quantum science, laser technology, high-order harmonic generation sources, attosecond chemistry, attoscience in condensed matter and at Free Electron Lasers.

The ATTO conference series started August 1st-5th, 2007 in Dresden, Germany, under the name of "Attosecond Physics" coordinated by Paul Corkum (National Research Council of Canada, Ottawa, Canada), Ferenc Krausz (Max Planck Institute for Quantum Optics, Garching, Germany), and Jean-Michel Rost (Max Planck Institute for the Physics of Complex Systems, Dresden, Germany).

The following 9 ATTO conferences were held as follows:

2009, July 29th-August 1st
Kansas State University, Manhattan, Kansas, USA
Co-chaired by Chii-Dong Lin and Zenghu Chang

2011, July 6th-8th
Hokkaido University, Sapporo, Japan
Co-chaired by Katsumi Midorikawa and Kaoru Yamanouchi

2013, July 8th-12th
Institut Pasteur, Paris, France
Co-chaired by Pascal Salières and Eric Constant

2015, July 5th-10th
Manoir Saint-Sauveur, Saint-Sauveur, Quebec, Canada
Co-chaired by Paul Corkum and François Légaré

2017, July 2nd-7th
Shaanxi Zhangbagou Guesthouse, Xi'an, China
Co-chaired by Zhiyi Wei and Zenghu Chang

2019, July 1st-5th
University of Szeged Congress Center, Szeged, Hungary
Co-chaired by Katalin Varjú and Dimitris Charalambidis

2022, July 11th-15th
University of Central Florida, Orlando, Florida, USA
Co-chaired by Luca Argenti and Michael Chini

2023, July 9th-14th
Booyoung Hotel and Resort, Jeju Island, Jeju, Korea
Co-chaired by Chang Hee Nam and Kyung Taec Kim

This year, the 10th edition of ATTO is taking place on July 6th-11th in Lund, Sweden. The conference has two locations: The Loop at Science Village and Stadshallen in the city center. The conference format is designed to foster extensive and open discussions. Three tutorials have been arranged and the conference features 23 invited talks, 49 contributed talks and an impressive number of 300 poster presentations. In keeping with the tradition of the ATTO conferences, there are no parallel sessions. Moreover, 21 exhibitors are present to showcase technologies and strenghten collaborations with industries and large-scale facilities.

COMMITTEES

International Program Committee

The International Program Committee is responsible for the governance and the scientific program of the ATTO conference. Its members oversee the conduction of ATTO, decide the conference program sessions and the list of invited speakers, scrutinize the abstracts submitted to the conference, select the contributed speakers, and appoint the organizers of future ATTO conferences.

Luca Argenti, University of Central Florida, USA
Andrius Baltuška, Technical University of Vienna, Austria
Dimitris Charalambidis, University of Crete, Greece
Michael Chini, Ohio State University, USA
Oren Cohen, Technion Institute of Technology, Israel
Per Eng-Johnsson, Lund University, Sweden (not active for ATTO X)
Davide Faccialà, CNR-IFN, Italy
Mette Gaarde, Louisiana State University, USA
Meng Han, Kansas State University, USA
Jiro Itatani, University of Tokyo, Japan
Kyung Taec Kim, Gwangju Institute of Science and Technology, Korea
Matthias Kling, SLAC Stanford University, USA
François Légaré, INRS, Canada
Eva Lindroth, Stockholm University, Sweden (not active for ATTO X)
José Antonio Pérez-Hernández, CLPU, Spain
Thierry Ruchon, CEA Saclay, France
Olga Smirnova, Max Born Institute, Germany
Vasily Strelkov, Russian Academy of Sciences, Russia (not active for ATTO X)
Richard Taïeb, Sorbonne University, France
John Tisch, Imperial College London, UK
Giulio Vampa, Ottawa University, Canada
Katalin Varjú, ELI-ALPS, Hungary
Hans Jakob Wörner, ETH Zurich, Switzerland
Kun Zhao, Institute of Physics, Chinese Academy of Sciences, China
Michael Zürch, University of California, USA

International Advisory Board

The Local Organizing Committee relies on the expertise and advice of all colleagues working in attosecond science to prepare an event that meets the needs of the international attosecond community at large. A selected group of researchers, representative of the various geographic regions and scientific interests of the attosecond community, has agreed to serve in an International Advisory Board on the occasion of ATTO X. The members are asked to bring to the attention of the Local Organizing Committee, and the International Program Committee, any new and relevant research trend in their region and field of interest.

Jens Biegert, ICFO, Spain
Francesca Calegari, CFEL DESY, Germany
Paul Corkum, National Research Council, Canada
Louis DiMauro, Ohio State University, USA
Nirit Dudovich, Weizmann Institute of Science, Israel
Feng He, Shanghai Jiaotong University, China
Ursula Keller, ETH Zurich, Switzerland
Igor Litvinyuk, Griffith University, Australia
Agostino Marinelli, SLAC, USA
Chang Hee Nam, Institute of Basic Science, Korea
Alicia Palacios, Universidad Autónoma de Madrid, Spain
Thomas Pfeifer, Max Planck Institute for Nuclear Physics, Germany
Françoise Remacle, University of Liège, Belgium
Pascal Salières, CEA Saclay, France
Giuseppe Sansone, Freiburg University, Germany
Caterina Vozzi, CNR-IFN, Italy
Zhiyi Wei, Chinese Academy of Sciences, China
Amelle Zaïr, King's College London, UK

Local Organizing Committee

The Local Organizing Committee is responsible for all the activities necessary for the successful conduction of the conference, as well as for facilitating and coordinating the activities of the International Program Committee.

Per Eng-Johnsson, Lund University, Sweden (chair)
Anne-Lise Viotti, Lund University, Sweden (chair)
Cord L. Arnold, Lund University, Sweden
David Busto, Lund University, Sweden
Marcus Dahlström, Lund University, Sweden
Raimund Feifel, University of Gothenburg, Sweden
Mathieu Gisselbrecht, Lund University, Sweden
Anne L'Huillier, Lund University, Sweden
Eva Lindroth, Stockholm University, Sweden
Johan Mauritsson, Lund University, Sweden
László Veisz, Umeå University, Sweden

CONFERENCE INFORMATION

Registration hours

July 6th: 15:30 ~ 21:00
July 7th-8th: 8:00 ~ 18:30
July 9th: 8:00 ~ 16:00
July 10th: 12:00 ~16:00
July 11th: 08:00 ~12:00

Certificate of attendance

If you need a certificate of attendance, please give your name at the registration desk.

Events

Welcome reception
Sunday, July 6th, 18:30 - 21:00
The Loop Atrium, Science Village

Opening ceremony
Monday, July 7th, 8:30 - 8:45
The Loop lecture hall, Science Village

Sponsors Night: Mingling with exhibitors
Tuesday, July 8th, 18:30 - 21:00
The Loop Atrium, Science Village

Special evening session
The origins of time-dependent theory in attosecond physics: The science of Kenneth Kulander
Tuesday, July 8th, 19:15 - 20:00
The Loop lecture hall, Science Village

Excursion at Kulturen
Thursday, July 10th, 9:00 - 12:00
Kulturen, Green Gate at the crossing of Adelgatan/Tegnérsplatsen

Nobel session and mingle
Thursday, July 10th, 16:45 - 18:45
Stadshallen, Lund

Conference dinner
Thursday, July 10th, 19:00 - 22:00
Bar and music until ~24:00
AF-borgen, Sandgatan 2, Lund

Closing remarks
Friday, July 11th, 12:45 - 13:00
Stadshallen, Lund

Lab visits: High-Power Laser Facility
Friday, July 11th, 13:30 - 17:00
Physics department, Professorsgatan 1B, Lund

CONFERENCE PROGRAM

Tutorials



Marc Vrakking

Max Born Institute Berlin, Germany

"Attosecond science: from adolescence to adulthood"



Zenghu Chang

University of Ottawa, Canada

"Attosecond X-ray light sources driven by mid-infrared lasers"



Morgane Vacher

University of Nantes, France

"Attosecond chemistry: tutorial and theoretical perspectives"

Sunday, July 6th, The Loop

15:30 - 18:30 Registration

18:30 - 21:00 Welcome reception

Monday, July 7th, The Loop

08:30 - 08:45 Opening ceremony in the lecture hall

08:45 - 10:30 [Mo1] Attosecond Physics and Spectroscopy (I)

Session chair: Kyung Taec Kim, Institute for Basic Science, Korea

08:45 - 09:30 **Attosecond science: from adolescence to adulthood [Mo1.1 tutorial]**
Marc Vrakking
Max-Born-Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Str. 2A, 12489 Berlin, Germany

09:30 - 10:00 **Time-dependent close coupling on the heels of attosecond electron dynamics [Mo1.2 invited]**
Luca Argenti
University of Central Florida, Department of Physics, USA

10:00 - 10:15 **Laser-assisted dynamical interference of photoelectrons driven by chirped extreme-ultraviolet pulses [Mo1.3]**
Federico Vismarra*, Mattias Bertolino, Elisa Appi, Marius Plach, Lenard Gulyas Oldal, Timea Grosz, Gian Luca Dolso, Vénus Poulain, Daniele Mocci, Giacomo Inzani, Chinmoy Biswas, Massimo De Marco, Gabriele Zeni, Fabio Frassetto, Luca Poletto, Maurizio Reduzzi, Rocío Borrego-Varillas, Hans Jakob Wörner, Zoltan Filus, Imre Seres, Peter Jojart, Balázs Major, Tamas Csizmadia, Mauro Nisoli, Per Eng-Johnsson, Jan Marcus Dahlström, Matteo Lucchini
**Department of Physics, Politecnico di Milano, Italy; Institute of Photonics and Nanotechnologies, IFN-CNR, Italy; Laboratorium für Physikalische Chemie, ETH Zurich, Switzerland*

10:15 - 10:30 **Attosecond spectroscopy reveals electron correlation in the photoionization of Argon [Mo1.4]**
Sizuo Luo*, Mingxuan Li, Huiyong Wang, Rezvan Tahouri, Robin Weissenbilder, Jialong Li, Wentao Wang, Jiaao Cai, Xiaochun Hong, Xiaosen Shi, Liangwen Pi, David Busto, Mathieu Gisselbrecht, Philipp. V Demekhin, Kiyoshi Ueda, Anne L'Huillier, Jan Marcus Dahlström, Eva Lindroth, Dajun Ding
**Institute of atomic and molecular physics, Jilin University, Changchun, 130012, China.*

10:30 - 11:00 Coffee break in Atrium

11:00 - 12:00 [Mo2] Attosecond Physics and Spectroscopy (II)

Session chair: Igor Litvinyuk, Griffith University, Australia

11:00 - 11:30 **Chiral molecules in strong laser fields [Mo2.1 invited]**
Yann Mairesse
Université de Bordeaux, CNRS, CEA, CELIA, UMR5107, F33405 Talence, France

11:30 - 11:45 **Vectorial attosecond transient spectroscopy [Mo2.2]**
Noa Yaffe*, Omer Kneller, Chen Mor, Yann Mairesse, Nirit Dudovich
**Weizmann Institute of Science, Rehovot, 7630031, Israel*

11:45 - 12:00 **Attosecond XUV pump-control spectroscopy for tracking and modification of excited neutral molecules [Mo2.3]**
Gergana D. Borisova*, Paula Barber Belda, Shuyuan Hu, Paul Birk, Veit Stoo, Maximilian Hartmann, Daniel Fan, Robert Moshammer, Alejandro Saenz, Christian Ott, Thomas Pfeifer
**Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany*

12:00 - 12:15 **Clocking the tunneling time in strong-field ionization [Mo2.4]**
Jonathan Dubois*, Leonardo Rico, Letizia Fede, Camille Levêque, Jeremie Caillat, Yann Mairesse, Richard Taïeb
**Sorbonne Université, CNRS, Laboratoire de Chimie Physique - Matière et Rayonnement, LCPMR, 75005 Paris, France*

12:15 - 13:00 Lunch break

13:00 - 14:30 [MoP] Poster Session (I) - 1st floor

14:30 - 16:15 [Mo3] Attosecond Quantum Science (I)

Session chair: Daniel Finkelstein-Shapiro, Uni. Nac. Aut. de Mexico, Mexico

14:30 - 15:00 **Quantum-orbit dynamics in strongly-polychromatic laser fields [Mo3.1 invited]**
Emilio Pisanty
Attosecond Quantum Physics Laboratory, King's College London, London WC2 R2LS, UK

15:00 - 15:30 **Ion-photoelectron entanglement and coherence controlled by two-color laser pulses [Mo3.2 invited]**
Kenichi Ishikawa
Department of Nuclear Engineering and Management, The University of Tokyo, Tokyo 113-8656, Japan

15:30 - 15:45 **Measurement of photoelectron decoherence and attosecond dynamics of unobserved ions [Mo3.3]**
Gabriel Granveau*, Morgan Berkane, David Bresteau, Rafael Menezes-Ferreira, Gabriele Crippa, Hugo Marroux, Thierry Ruchon, Camille Levêque, Richard Taïeb, Jeremie Caillat, Pascal Salières, Charles Bourassin-Bouchet
**Université Paris-Saclay, CNRS, IOGS, Laboratoire Charles Fabry, 91127 Palaiseau, France*

15:45 - 16:00 **Testing quantum mechanics with photoelectron interferometry [Mo3.4]**
Gustav Arvidsson*, Edoardo Alberto Boati, Robin Weissenbilder, Mattias Ammitzböll, Christoph Dittel, Richard James Squibb, Raimund Feifel, Mathieu Gisselbrecht, Cord Louis Arnold, Anne L'Huillier, David Busto
**Department of Physics, Lund University, Box 118, 221 00 Lund, Sweden*

16:00 - 16:15 **Strong-field dynamics of electrons photoemitted with non-classical light [Mo3.5]**
Jonathan Pölloth*, Jonas Heimerl, Andrei Rasputnyi, Francesco Tani, Maria Chekhova, Peter Hommelhoff
**Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany*

16:15 - 16:45 Coffee break in Atrium

16:45 - 18:30 [Mo4] Attosecond Quantum Science (II)

Session chair: Richard Taïeb, LCPMR-CNRS-SU, France

16:45 - 17:15 **Giant counter-rotating oscillations and quantum entanglement from time-dependent strong coupling in the extreme ultraviolet [Mo4.1 invited]**
Jan Marcus Dahlström
Department of Physics, Lund University, P.O. Box 118, SE-22100, Lund, Sweden

17:15 - 17:45 **Experimental evidence of the quantum-optical nature of high-harmonic generation in semiconductors [Mo4.2 invited]**
Hamed Merdji
Laboratoire d'Optique Appliquée, CNRS, ENSTA, Institut Polytechnique de Paris, Palaiseau, 91120, France

17:45 - 18:00 **Photon bunching in high-harmonic emission controlled by quantum light [Mo4.3]**
Samuel Lemieux, Sohail A Jalil, David N Purschke, Neda Boroumand, TJ Hammond, David Villeneuve, Andrei Naumov, Thomas Brabec, Giulio Vampa*
**Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, Ottawa, K1N 0R6, Canada*

18:00 - 18:15 **Non-classical effects drive recombination in high-harmonic generation using circularly polarized fields [Mo4.4]**
Javier Rivera-Dean, Philipp Stammer, Maciej Lewenstein, Marcelo Ciappina*
**Guangdong Technion-Israel Institute of Technology, Physics Program, China; Technion - Israel Institute of Technology, Israel*

18:15 - 18:30 **Measuring and controlling the birth of quantum attosecond pulses [Mo4.5]**
Chen Mor, Matan Even Tzur*, Noa Yaffe, Michael Birk, Andrei Rasputnyi, Omer Kneller, Ido Nisim, Ido Kaminer, Michael Krüger, Oren Cohen, Nirit Dudovich
**Department of Physics, Technion—Israel Institute of Technology, Israel; Solid State Institute, Technion—Israel Institute of Technology, Israel; Helen Diller Quantum Center, Technion—Israel Institute of Technology, Israel*

Tuesday, July 8th, The Loop

08:45 - 10:30 [Tu1] Laser Technology for Attoscience (I)

Session chair: Lukas Gallmann, ETH Zurich, Switzerland

08:45 - 09:30 **Attosecond X-ray light sources driven by mid-infrared lasers [Tu1.1 tutorial]**
Zenghu Chang
University of Ottawa, Canada

09:30 - 09:45 **High power CEP stable Ti:Sa amplifier seeded by OPCPA [Tu1.2]**
Jingfeng Chen, Raman Maksimenka, Solene Favier, Philippe Demengeot, Yoann Pertot*
**Amplitude, 2-4 rue du Bois Chaland - CE 2926, 91029 Evry, France*

09:45 - 10:00 **Waveform-controlled near-single-cycle pulses at the 100 TW level for intense attosecond sources [Tu1.3]**
László Veisz*, Peter Fischer, Sajjad Vardast, Fritz Schnur, Alexander Muschet, Aitor De Andres, Sreehari Kaniyeri, Hang Li, Roushdey Salh, Karpat Ferencz, Gergely Norbert Nagy, Subhendu Kahaly
**Department of Physics, Umeå University, Linnaeus väg 24, 90187, Umeå, Sweden*

10:00 - 10:15 **Single-shot pulse retrieval of femtosecond bright squeezed vacuum [Tu1.4]**
Yuval Kern*, Ido Nisim, Michael Birk, Andrei Rasputnyi, Zhaopin Chen, Pavel Sidorenko, Ido Kaminer, Oren Cohen, Michael Krüger
**Department of Physics, Technion - Israel Institute of Technology, 32000 Haifa, Israel; Solid State Institute and Helen Diller Quantum Center, Technion - Israel Institute of Technology, 32000 Haifa, Israel*

10:15 - 10:30 **Characterization of sub-3-fs tunable VUV pulses [Tu1.5]**
Martin Kretschmar*, Jose Andrade, Rostyslav Danylo, Stefanos Carlström, Tobias Witting, Alexandre Mermillod-Blondin, Serguei Patchkovskii, Misha Ivanov, Marc Vrakking, Arnaud Rouzée, Tamas Nagy
**Max Born Institute, Max-Born-Str. 2A, 12489 Berlin, Germany*

10:30 - 11:00 Coffee break in Atrium

11:00 - 12:15 [Tu2] Laser Technology for Attoscience (II)

Session chair: Zhiyi Wei, Institute of Physics, CAS, China

11:00 - 11:30 **New ultrafast sources and science enabled by advanced nonlinear optics in hollow-core waveguides [Tu2.1 invited]**
Christian Brahms
School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, EH14 4AS, United Kingdom

11:30 - 11:45 **Multi-mirror multi-pass cells for compression of energetic narrowband laser pulses into the femtosecond regime [Tu2.2]**
Gaspard Beaufort*, Nayla Jimenez, Gunnar Arisholm, Victor Hariton, Ayhan Tajalli, Ingmar Hartl, Anne-Lise Viotti, Marcus Seidel
**Deutsches Elektronen-Synchrotron DESY, Hamburg, 22607, Germany; Department of Physics, Lund University, Lund, SE-22100, Sweden*

11:45 - 12:00 **Pulse compression of 300 W, 12 mJ with 83% transmission in hollow-core fiber [Tu2.3]**
Maksym Ivanov, Etienne Doiron, Marco Scalia, Pedram Adbolghader, François Légaré, Carlos Trallero, Bruno Schmidt*
**few-cycle Inc, Varennes, J3X 1P7, Canada*

12:00 - 12:15 **Terawatt optical attosecond pulses and 100 GW-scale far-ultraviolet pulses through extreme soliton dynamics [Tu2.4]**
Nikoleta Kotsina*, Michael Heynck, Joleik Nordmann, Martin Gebhardt, Teodora Grigorova, Christian Brahms, John Travers
**School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh, EH14 4AS, United Kingdom*

12:15 - 13:00 Lunch break

13:00 - 14:30 [TuP] Poster Session (II) - 1st floor

14:30 - 16:15 [Tu3] Attosecond HHG Sources (I)

Session chair: Pascal Salières, CEA Paris-Saclay, France

14:30 - 15:00 **Recent developments of attosecond high-harmonic generation sources in and outside the Extreme Light Infrastructure [Tu3.1 invited]**
Balázs Major
ELI-ALPS, Hungary

15:00 - 15:30 **Microfluidic-based sources for efficient XUV generation and manipulation [Tu3.2 invited]**
Caterina Vozzi
Istituto di Fotonica e Nanotecnologie CNR, Italy

15:30 - 15:45 **High harmonic generation with two non-collinear drivers: a unique gateway to extreme nonlinear phenomena [Tu3.3]**
Martin Luttmann, Mekha Vimal, Titouan Gadeyne, Matthieu Guer, Romain Cazali, Celine Chappuis, David Bresteau, Fabien Lepetit, Olivier Tcherbako, Jean-Francois Hergott, Thierry Auguste, Thierry Ruchon*
**Université Paris-Saclay, CEA, LIDYL, 91191 Gif-sur-Yvette, France*

15:45 - 16:00 **High-order harmonic up-conversion of spatiotemporal and spatiospectral optical vortices into the extreme ultraviolet [Tu3.4]**
Rodrigo Martin-Hernandez*, Guan Gui, Luis Plaja, Henry K. Kapteyn, Margaret M. Murnane, Chen-Ting Liao, Miguel Angel Porras, Carlos Hernandez-Garcia
**Grupo de Investigacion en Aplicaciones del Laser y Fotonica, Universidad de Salamanca, 37008, Salamanca, Spain; Unidad de Excelencia en Luz y Materia Estructuradas (LUMES), Universidad de Salamanca, Salamanca, Spain*

16:00 - 16:15 **Hollow gaussian beams for low-divergence high harmonic generation and attosecond pulse optimization [Tu3.5]**
Melvin Redon*, Rodrigo Martin-Hernandez, Ann-Kathrin Raab, Luis Plaja, Anne L'Huillier, Carlos Hernandez-Garca, Cord Arnold
**Department of Physics, Lund University, P.O. Box 118, 22100, Lund, Sweden*

16:15 - 16:45 Coffee break in Atrium

16:45 - 18:30 [Tu4] Attosecond HHG Sources (II)

Session chair: Katalin Varjú, ELI-ALPS, Szeged, Hungary

16:45 - 17:15 **Table-top all-attosecond transient absorption spectroscopy [Tu4.1 invited]**
Bernd Schütte
Max-Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Berlin, 12489, Germany

17:15 - 17:30 **Coincidence measurements of two-photon double ionization driven by a high-harmonic generation source [Tu4.2]**
Matthias Kübel*, Sebastian Hell, Julian Späthe, Robert Klas, Jan Rothhardt, Jens Limpert, Gerhard Paulus, Morten Førre, Christian Ott, Stephan Fritzsche
**Institute of Optics and Quantum Electronics, Friedrich-Schiller University, 07743 Jena, Germany; Helmholtz Institute Jena, 07743 Jena, Germany*

17:30 - 17:45 **Using nanostructured dielectric surfaces for the generation of nanofocused XUV radiation [Tu4.3]**
Parnia Bastani*, Aleksey Korobenko, Vedran Jelic, David N. Purschke, Shima Gholam Mirzaeimoghadar, Md Wazedur Rahman, Mathieu de Lafontaine, Andre Staudte, Paul B. Corkum, François Légaré, Giulio Vampa
**Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, Otawa, Ontario K1N 0R6, Canada; Advanced Laser Light Source (ALLS), Centre Energie Matériaux Télécommunications, Institut National de la Recherche Scientifique (INRS-EMT), 1650 Boulevard Lionel-Boulet, Varennes, Quebec J3X 1P7, Canada*

17:45 - 18:00 **Efficient water-window isolated attosecond pulse generation with tailored sub-cycle fields [Tu4.4]**
Miguel Angel Silva-Toledo*, Fabian Scheiba, Maximilian Kubullek, Rafael de Q. Garcia, Roland E. Mainz, Giulio Maria Rossi, Franz X. Kärtner
**Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, Hamburg, 22607, Germany; Physics Department and The Hamburg Centre for Ultrafast Imaging (CUI), University of Hamburg, Notkestrasse 9-11, Hamburg, 22607, Germany*

18:00 - 18:15 **Optimizing polarization gated generation of isolated attosecond pulses without CEP stabilization [Tu4.5]**
Corentin Picot, Lucas Perez, Emilien Prost, Franck Lepine, Eric Constant*
**Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière, UMR5306, Villeurbanne 69100, France*

18:15 - 18:30 **Direct measurement of the dipole phase in solid-state high-harmonic generation via XUV attosecond interferometry [Tu4.6]**
Nataliia Kuzkova*, Pieter J. van Essen, Brian de Keijzer, Roy van der Linden, Alvaro Jimenez-Galan, Rui E. F. Silva, Peter M. Kraus
**Advanced Research Center for Nanolithography, the Netherlands; Department of Physics and Astronomy, and LaserLaB, Vrije Universiteit Amsterdam, the Netherlands*

18:30 - 21:00 Sponsors Night:
Mingling with exhibitors in Atrium

Including drinks and food

19:15 - 20:00 Special Evening Session

Memorial - The origins of time-dependent theory in attosecond physics: The science of Kenneth Kulander

With Kenneth Schafer, Anne L'Huillier, Louis DiMauro, ...

Wednesday, July 9th, The Loop

08:45 - 10:30 [We1] Attosecond Chemistry (I)

Session chair: Hans Jakob Wörner, ETH Zurich, Switzerland

08:45 - 09:30 **Attosecond chemistry: tutorial and theoretical perspectives [We1.1 tutorial]**
Morgane Vacher
University of Nantes, France

09:30 - 10:00 **Advances in ultrafast ultraviolet spectroscopy of molecules [We1.2 invited]**
Vincent Wanie
The Hamburg Centre for Ultrafast Imaging, Universit at Hamburg, Germany; Physics Department, Universität Hamburg, Germany

10:00 - 10:15 **Probing iodine chemical environment with ionization delays [We1.3]**
Rafael Menezes Ferreira*, Gabriele Crippa, Lou Barreau, Constant Schouder, Georey Carneiro, Nicolas Gueneaux, Fabien Lepetit, Jean François Hergott, Francis Penent, Jerôme Palaudoux, Pascal Salières, Hugo Marroux
**Université Paris-Saclay, CEA, LIDYL, 91190 Gif-sur-Yvette, France*

10:15 - 10:30 **Probing the ultrafast vibronic wavepacket dynamics of ethylene with sub-4fs VUV pulses [We1.4]**
Arnab Sen*, Martin Kretschmar, Martha Jouybari, Rostyslav Danylo, Jose Andrade, Phillippe Burden, Simon Neville, Marc Vrakking, Albert Stolow, Tamas Nagy, Michael Schuurman, Arnaud Rouzée
**Max-Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Berlin, 12489, Germany*

10:30 - 11:00 Coffee break in Atrium

11:00 - 12:15 [We2] Attosecond Chemistry (II)

Session chair: Francesca Calegari, CFEL DESY, Germany

11:00 - 11:30 **Ab initio attosecond chemistry: strong electron-electron-nuclei correlations in multiphoton single and double ionization [We2.1 invited]**
Alicia Palacios
Departamento de Quimica, Universidad Autonoma de Madrid, UAM, Madrid, 28049, Spain; Condensed Matter Physics Center (IFIMAC), Universidad Autonoma de Madrid, Madrid, 28049, Spain

11:30 - 11:45 **Attosecond coherent control and measurement of the photoionization dynamics of chiral molecules [We2.2]**
Meng Han, Jia-Bao Ji*, Alexander Blech, Esteban Goetz, Corbin Allison, Loren Greenman, Christiane Koch, Hans Jakob Wörner
**ETH Zürich, Laboratorium für Physikalische Chemie, Switzerland*

11:45 - 12:00 **Control of photo-ion circular dichroism using orthogonal laser beams [We2.3]**
Jason Greenwood*, Leah Donnelly
**School of Mathematics and Physics, Queen's University Belfast, Belfast, United Kingdom*

12:00 - 12:15 **All-optical coherent control of chiral electronic transitions for highly enantioselective photochemistry using femtosecond lasers [We2.4]**
Andrés Ordóñez*, Patricia Vindel-Zandbergen, David Ayuso
**Imperial College London, Department of Physics, London, SW7 2BW, UK*

12:15 - 13:00 Lunch break

13:00 - 14:30 [WeP] Poster Session (III) - 1st floor

14:30 - 16:00 [We3] Attosecond Chemistry (III)

Session chair: Fernando Martin, Uni. Aut. de Madrid & IMDEA Nano, Spain

14:30 - 15:00 **Attosecond time-resolved electronic and vibrational dynamics in furan [We3.1 invited]**
Stefanie Gräfe
Friedrich-Schiller University Jena, Germany

15:00 - 15:30 **Attosecond dynamics in molecules: probing ultrafast charge motion with EUV/IR and UV/EUV spectroscopy [We3.2 invited]**
Mauro Nisoli
Department of Physics, Politecnico di Milano, Italy; Institute of Photonics and Nanotechnologies, IFN-CNR, Italy

15:30 - 15:45 **Time-resolved x-ray absorption spectroscopy in thiophene covering sulfur L and carbon K edges [We3.3]**
Tristan Kopp*, Federico Vismarra, Valentina Utrio Lanfaloni, Leonardo Redaelli, Tadas Balciunas, Emir Ardali, Hans Jakob Wörner
**Laboratory of Physical Chemistry, ETH Zurich, Zurich, Switzerland*

15:45 - 16:00 **Time-resolving molecular dissociation with FEL-pump and HHG-probe pulses [We3.4]**
Alexander Magunia*, Elisa Appi, Christina C. Papadopoulou, Hannes Lindenblatt, Florian Trost, Severin Meister, Thomas Ding, Michael Straub, Gergana D. Borisova, Junhee Lee, Rui Jin, Alexander von der Dellen, Christian Kaiser, Markus Braune, Stefan Düsterer, Skirmantas Alisauskas, Tino Lang, Christoph M. Heyl, Bastian Manschwetus, Sören Grunewald, Ulrike Frühling, Ayhan Tajalli, Ammar bin Wahid, Laura Silletti, Francesca Calegari, Philip Mosel, Uwe Morgner, Milutin Kovacev, Uwe Thumm, Ingmar Hartl, Rolf Treusch, Robert Moshammer, Christian Ott, Thomas Pfeifer
**Max-Planck-Institut für Kernphysik, Germany*

16:00 - 16:30 Coffee break in Atrium

16:30 - 18:30 [We4] Attosecond Science in Condensed Materials (I)

Session chair: Amelle Zaïr, King's College London, UK

16:30 - 17:00 **Observation of the Berry phase via attosecond interferometry [We4.1 invited]**
Nirit Dudovich
Weizmann Institute, Israel

17:00 - 17:30 **Strong-field physics inside of (light-dressed) graphene and at the surface of metal needle tips [We4.2 invited]**
Peter Hommelhoff
Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

17:30 - 17:45 **Subcycle band-structure videography of lightwave-driven graphene [We4.3]**
Giacomo Inzani*, Vincent Eggers, Manuel Meierhofer, Jakob Helml, Lasse Münster, Robert Wallauer, Sarah Zajusch, Suguru Ito, Leon Machtl, Hao Yin, Christian Kumpf, Francois C Bocquet, Changhua Bao, Jens Güdde, F Stefan Tautz, Rupert Huber, Ulrich Höfer
**Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, Regensburg, 93040, Germany*

17:45 - 18:00 **Discovery of a hybrid exciton state by attosecond spectroscopy [We4.4]**
Simone Bonetti, Nicola Di Palo*, Giacomo Inzani, Gian Luca Dolso, Matteo Talarico, Martin Zukerstein, Giacomo Fiorentini, Rocío Borrego-Varillas, Mauro Nisoli, Marco D'Alessandro, Nicolas Tancogne-Dejean, Umberto De Giovannini, Davide Sangalli, Matteo Lucchini
**Department of Physics, Politecnico di Milano, Milano, 20133, Italy*

18:00 - 18:15 **Correlations drive the attosecond response of strongly-correlated insulators [We4.5]**
Romain Cazali*, Amina Alic, Matthieu Guer, Christopher Kaplan, Fabien Lepetit, Olivier Tcherbako, Stephane Guizard, Angel Rubio, Nicolas Tancogne-Dejean, Gheorghe Chiuzbaian, Romain Geneaux
**Université Paris-Saclay, CEA, LIDYL, France*

18:15 - 18:30 **Excitonic high harmonics and attosecond time delays in GaN [We4.6]**
Zhaopin Chen*, You Wu, Christopher Ayala, Mark Levit, Ido Nisim, Yuanpeng Wu, Zetian Mi, Steven Cundiff, Mackillo Kira, Michael Krüger
**Department of Physics and Solid State Institute, Technion - Israel Institute of Technology, Haifa 3200003, Israel*

Thursday, July 10th, Stadshallen

09:00 - 12:00 Excursion at Kulturen

12:00 - 13:00 Lunch at Stadshallen

13:00 - 14:45 [Th1] Attosecond Science in Condensed Materials (II)

Session chair: Jan Vogelsang, Universität Oldenburg, Germany

13:00 - 13:30 **Probing ultra-fast electron dynamics in liquids and solutions using high-harmonic spectroscopy [Th1.1 invited]**
Angana Mondal
Laboratory of Physical Chemistry, ETH Zürich, Zürich, Switzerland

13:30 - 14:00 **Attomicroscopy imaging and control of electron motion in solid-state [Th1.2 invited]**
Mohammed Hassan
Arizona University, USA

14:00 - 14:15 **Attosecond spectroscopy of virtual charge dynamics [Th1.3]**
Gian Luca Dolso, ShunSuke A Sato, Giacomo Inzani, Nicola Di Palo, Bruno Moio, Rocío Borrego-Varillas, Mauro Nisoli, Matteo Lucchini*
**Department of Physics, Politecnico di Milano, Piazza Leonardo da Vinci, 20133 Milano, Italy*

14:15 - 14:30 **Two-color control of attosecond scanning tunneling microscopy [Th1.4]**
Boyang Ma*, Daniel Davidovich, Adi Goldner, Michael Krüger
**Physics Department and Solid State Institute and Helen Diller Quantum Center, Technion-Israel Institute of Technology, Israel*

14:30 - 14:45 **Attosecond electron pulses with chiral mass and charge [Th1.5]**
Yiqi Fang*, Joel Kuttru, David Nabben, Peter Baum
**Universität Konstanz, Fachbereich Physik, 78464 Konstanz, Germany*

14:45 - 15:15 Coffee break in Foyer

15:15 - 16:45 [Th2] Attosecond Science in Condensed Materials (III)

Session chair: Michael Krüger, Technion, Israel

15:15 - 15:45 **High-harmonic spectroscopy probes lattice dynamics: extension to bandgap dynamics and structural phase transitions [Th2.1 invited]**
Tran Trung Luu
Hong Kong University, Hong Kong

15:45 - 16:15 **Ultrafast control of valley physics in 2D materials [Th2.2 invited]**
Álvaro Jiménez-Galán
Instituto de Ciencia de Materiales de Madrid, Spain

16:15 - 16:30 **High harmonic generation from solutions [Th2.3]**
Mette Gaarde*, Eric Moore, Sucharita Giri, Andreas Koutsogiannis, Tahereh Alavi, Greg Mc-Cracken, Kenneth Lopata, John Herbert, Louis DiMauro
**Louisiana State University, Department of Physics and Astronomy, USA*

16:30 - 16:45 **Enhancement of harmonic generation in liquid water jet by resonant excitation in the mid-infared pulses [Th2.4]**
Tianqi Yang, Yangyang Hua, Tomoya Mizuno, Teruto Kanai, Satoshi Ashihara, Yoshihisa Harada, Jiro Itatani*
**The Institute for Solid State Physics, The University of Tokyo, Japan*

16:45 - 17:45 Mingle with drinks in Foyer

17:45 - 18:45 Nobel Session

Moderator: Mikhail Ivanov
Max-Born-Institut, Berlin, Germany

Panelists: Anne L'Huillier and Pierre Agostini
Lund University, Sweden and *Ohio State University, USA*

Live Q&A - Ask your question(s)!

19:00 - 22:00 Conference Dinner at AF-borgen

Bar and Music from 22:00!

Friday, July 11th, Stadshallen

09:00 - 10:45 [Fr1] Attosecond Science at FELs (I)

Session chair: Louis DiMauro, Ohio State University, USA

09:00 - 09:30 **Nonlinear science with multi-harmonic FEL at sub-femtosecond resolution [Fr1.1 invited]**
Carlo Callegari
FERMI ELETTRA, Italy

09:30 - 10:00 **Dephasing in attosecond science and the collective nature of structural quantum fluctuations [Fr1.2 invited]**
Robin Santra
Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany

10:00 - 10:15 **Coherent control with two photons at a seeded free-electron laser [Fr1.3]**
Ulf Saalmann
Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Str. 38, 01187 Dresden, Germany

10:15 - 10:30 **Complex attosecond waveform synthesis using regular- and irregularly-spaced harmonic combs at seeded free-electron laser FERMI [Fr1.4]**
Praveen Kumar Maroju*, Miguel Benito de Lama, Michele Di Fraia, Oksana Plekan, Matteo Bonanomi, Barbara Merzuk, David Busto, Ioannis Makos, Marvin Schmoll, Ronak Shah, Primož R Ribič, Luca Giannessi, Enrico Allaria, Giovanni De Ninno, Carlo Spezzani, Giuseppe Penco, Alexander Demidovich, Miltcho Danailov, Marcello Coreno, Marco Zangrando, Alberto Simoncig, Michele Manfredda, Richard J Squibb, Raimund Feifel, Samuel Bengtsson, Emma R Simpson, Tamás Csizmadia, Mathieu Dumergue, Sergei Kühn, Kiyoshi Ueda, Jianxiong Li, Kenneth J Schafer, Fabio Frassetto, Luca Poletto, Kevin C Prince, Johan Mauritsson, Johannes Feist, Alicia Palacios, Carlo Callegari, Giuseppe Sansone
**Physikalisches Institut, University of Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany; Department of Physics, Lund University, PO Box 118, SE-221 00 Lund, Sweden*

10:30 - 10:45 **Magnetization dynamics probed with helicoidal dichroism [Fr1.5]**
Mauro Fanciulli, Matteo Pancaldi, Anda-Elena Stanciu, Matthieu Guer*, Emanuele Pedersoli, Dario De Angelis, Primoz Rebernic-Ribic, David Bresteau, Martin Luttmann, Pietro Carrara, Arun Ravindran, Benedikt Rösner, Christian David, Carlo Spezzani, Michele Manfredda, Ricardo Sousa, Laurent Vila, Ioan Lucian Prejbeanu, Liliana D. Buda-Prejbeanu, Bernard Dieny, Giovanni De Ninno, Flavio Capotondi, Thierry Ruchon, Maurizio Sacchi
**Université Paris-Saclay, CEA, LIDYL, Gif-sur-Yvette, 91191, France*

10:45 - 11:15 Coffee break in Foyer

11:15 - 12:45 [Fr2] Attosecond Science at FELs (II)

Session chair: Arnaud Rouzée, Max Born Institute, Germany

11:15 - 11:45 **Ultrafast electron motion probed with attosecond X-ray Free Electron Lasers [Fr2.1 invited]**
James Cryan
SLAC National Accelerator Laboratory, USA

11:45 - 12:00 **Characterization of polarized attosecond soft X-ray pulses at Eu-XFEL [Fr2.2]**
Juliette Leroux*, Sadia Bari, Thomas Baumann, Rebecca Boll, Francesca Calegari, Alberto De Fanis, Filippa Dudda, Danilo Enoque Ferreira de Lima, Lars Funke, Andreas Galler, Gianluca Geloni, Wolfram Helml, Akhila Jose, Alice Judt, Tommaso Mazza, Moritz Mogilowski, Terry Mullins, Thorsten Otto, Yevheniy Ovcharenko, Gunnar Peterson, Dirk Raiser, Johan Ribbing, Matthew Robinson, Sara Savio, Philipp Schmidt, Bjorn Senftleben, Svitozar Serkez, Sergey Usenko, Vincent Wanie, Niclas Wieland, Lasse Wulng, Michael Meyer, Markus Ilchen
**Department of Physics, University of Hamburg, 22607 Hamburg, Germany*

12:00 - 12:15 **Redirection and reshaping of intense extreme-ultraviolet radiation [Fr2.3]**
Yu He*, Alexander Magunia, Harijyoti Mandal, Muwaffaq Ali Mourtada, Carlo Kleine, Arikta Saha, Marc Rebholz, Gergana D. Borisova, Lina Hedewig, Hannes Lindenblatt, Florian Trost, Ulrike Frühling, Christina C. Papadopoulou, Elisa Appi, Stefan Düsterer, Tino Lang, Skirmantas Alisaukas, Christoph M. Heyl, Steffen Palutke, Markus Braune, Christina Bömer, Dietrich Krebs, Doriana Vinci, Philip Mosel, Peer Biesterfeld, Ingmar Hartl, Robert Moshammer, Milutin Kovacev, Kiyoshi Ueda, Mette B. Gaarde, Christian Ott, Thomas Pfeifer
**Max Planck Institute for Nuclear Physics, Germany*

12:15 - 12:30 **Impulsive stimulated X-ray Raman scattering in the condensed phase with attosecond X-ray Free Electron Laser pulses [Fr2.4]**
Felix Egun*, Oliver Alexander, Laura Rego, Ana M Gutierrez, Douglas Garratt, Stefan Moeller, Ming-Fu Lin, Georgi Dakovski, James P Cryan, Agostino Marinelli, Antonio Picón, Jonathan P Marangos and the LX52 Collaboration
**Department of Physics, Imperial College London, South Kensington, SW7 2AZ, United Kingdom*

12:30 - 12:45 **Single-shot measurements of soft x-ray attosecond pulses at kilohertz repetition rate [Fr2.5]**
Veronica Guo*, Kurtis Borne, Mat Britton, David Cesar, James Cryan, Taran Driver, Paris Franz, Erik Isele, Xiang Li, Ming-Fu Lin, Agostino Marinelli, Razib Obaid, River Robles, Nicholas Sudar, Emily Thierstein, Jun Wang
**Stanford University, USA; SLAC National Accelerator Laboratory, USA*

12:45 - 13:00 Closing Remarks

13:00 - 13:30 Lunch to-go & transfer to the Physics Department

13:30 - 17:00 Lab Visits - Lund High Power Laser Facility

Poster Sessions

13:00 - 14:30 [MoP] Poster Session (I) - Monday, July 7th

Poster Session sponsored by: ASML

[MoP.1] Characterizing and controlling lifetimes of doubly-excited states directly in the time domain
Rupprecht, Patrick
[MoP.2] RABBITT-HOP: RABBITT with higher-order processes
Brown, Andrew
[MoP.3] Ultrathin liquid flat jet system for attosecond spectroscopy of solvated molecules
Rockenstein, Sabine
[MoP.4] Conditions for Mollow triplets in ultrafast absorption and dynamical fluorescence
Stenquist, Axel
[MoP.5] Absolute photoemission timing and asymmetry parameter of the I4d giant resonance of the Iodoalkanes
Forster, Maximilian
[MoP.6] Attosecond resonant Auger spectroscopy
Kimberg, Victor
[MoP.7] Attosecond partial wave meter and its application in resolving the photoemission time delays in atoms and molecules
Gong, Xiaochun
[MoP.8] Nonlocal resonance theory for electron–nuclear dynamics in molecules
Bertolino, Mattias
[MoP.9] Ultrafast time-varying nonlinearity at epsilon-near-zero
Li, Yan
[MoP.10] Symmetries, dominance and quantum interference in below-threshold nonsequential double ionization with linearly polarized two-color fields
Hashim, Sufia
[MoP.11] Attosecond Fourier transform spectroscopy
Kneller, Omer
[MoP.12] Electronic-vibrational dynamics in x-ray transient absorption of N2+ induced by strong-field ionization
Zhao, Jing
[MoP.13] Photoemission timing of surface-oriented Iodoalkanes on Pt(111)
Paul, Sven-Joachim
[MoP.14] Attosecond ionization delay arising from Feshbach-resonance of N2
Wang, W

[MoP.15] Quantifying centrifugal-potential effects: channel-resolved attosecond time delays in nondipole photoionization
Liao, Yijie
[MoP.16] Phase measurements beyond strong-field approximation in non-perturbative single-color high-harmonic spectroscopy
Avnery, Keren
[MoP.17] Analytical expression for continuum-continuum transition amplitude of Hydrogen-like atoms
Ji, Jia-Bao
[MoP.18] HHG-based all-attosecond transient absorption spectroscopy
Svirplys, Evaldas
[MoP.19] Hole alignment in noble gas atoms using RTDCIS method
Tahouri, Rezvan
[MoP.20] Modelling sequential ionisation within time-dependent R-Matrix Theory
Lavery, Holly
[MoP.21] SoftMeter: Ultrafast molecular imaging driven by a tunable two-color field
Ghaffari, Fatemehsadat
[MoP.22] Core ionization of positronium chloride: a numerical study
Camper, Antoine
[MoP.23] Attosecond transient absorption spectroscopy of laser-dressed dense helium: Spectral lineshape modification and weak transitions brought to light
He, Yu
[MoP.24] Observation of Bloch oscillation phenomena via high harmonic generation spectroscopy
Cahana, Noa
[MoP.25] Electron recollisional excitation on dissociative ionization of OCS in phaselocked two-color intense laser fields
Endo, Tomoyuki
[MoP.26] Ro-vibronic coupling in vibrational resolved attosecond photoelectron spectroscopy of acetylene
Li, Jialong
[MoP.27] Attosecond dynamics of direct electrons in optical near-fields
Heimerl, Jonas
[MoP.28] Influence of catastrophes and hidden dynamical symmetries on ultrafast photoelectron physics
Rook, Thomas
[MoP.29] Interference of direct and resonant two-photon ionization in helium
Weissenbilder, Robin
[MoP.30] Estimation of ZnO thickness from high harmonic spectrum using machine learning algorithms
Nagyillés, Balázs

[MoP.31] Anisotropy induced electronic coherence signal in attosecond transient absorption spectroscopy
Yuen, CH Isaac
[MoP.32] Probing the attosecond dynamics of high-energy-radiation-induced collisional ionization cascades in condensed-phase samples
Ferté, Anthony
[MoP.33] Modeling angularly resolved phase-jumps in heavy atoms
Mankov, Elisei
[MoP.34] Neural network assisted denoising in attosecond transient spectroscopy beyond correlated source noise
Hollm, Marko
[MoP.35] Multi-photon and strong-field ionization with a twist
Rajak, Debobrata
[MoP.36] From megabarns to attoseconds: How to relate the cross-section with the time delay
Kheifets, Anatoli
[MoP.37] Quantum trajectory selector: a novel method to clock attosecond dynamics within recollision physics
Raab, Ann-Kathrin
[MoP.38] Orientation-dependent production of electron spirals from multi-orbital heteronuclear diatomic molecules
Ngoko Djiokap, Jean Marcel
[MoP.39] Guiding synthetic chiral light
Loehr, Alexander
[MoP.40] Attosecond transient absorption in the water-window regime driven by self-compressed light-transient pulses
Vismarra, Federico
[MoP.41] Tuning the Fano resonance line shape in hundred zeptosecond timescale by using two attosecond pulses
Jin, Cheng
[MoP.42] Heterodyne analysis of high-order partial waves in attosecond photoionization of Helium
Han, Lulu
[MoP.43] Exciting autoionizing states with multiple photons
Majety, Vinay Pramod
[MoP.44] Imaging ultrafast structural rearrangement in ammonia molecule with high harmonic spectroscopy
He, Lixin
[MoP.45] New 1D atomic model potential for more accurate HHG spectra
Sallai, Krisztina
[MoP.46] Attosecond electron dynamics in two-color multiphoton processes in helium
Benito De Lama, Miguel
[MoP.47] Retrieving the density matrix form experimental KRAKEN data
Evensen, Andreas

[MoP.48] Partial-wave transitions in attosecond time delays
Mašín, Zdeněk
[MoP.49] Wigner vs. Smith: Time delays in anisotropic systems
Saalmann, Ulf
[MoP.50] Extreme-ultraviolet optical response of atomically thin MoS2 crystals
Di Palo, Nicola
[MoP.51] Effect of attosecond electronic coherences on conical-intersection dynamics in core-excited states
Rodriguez Cuenca, Emilio
[MoP.52] Cross-process interference in strong-field ionization: insights from CEP-dependent photoelectron spectra
Herzig, Anne
[MoP.53] All-attosecond transient absorption spectroscopy of gas-phase molecules
Chiang, Kuo-Yang
[MoP.54] Attosecond Raman time-delay in resonant transitions of ions
Zhao, Zengxiu
[MoP.55] Imaging in the water window with coherence tomography using high-harmonic generation
Fuchs, Silvio
[MoP.56] Attosecond optical orientation
Drescher, Lauren B
[MoP.57] Simulation of transient-grating high-harmonic spectroscopy from dissociating molecules
Trieu, Doan-An
[MoP.58] Probing of ultrafast nuclear vibrational and rotational dynamics with X-ray
Liu, Ji-Cai
[MoP.59] Unifying two manifestations of multielectron polarization in high harmonic generation: peak's frequency shift and odd-even intensity modulation
Phan, Ngoc-Loan
[MoP.60] State-resolved femtosecond phase control in dense-gas laser-atom interaction enabled by attosecond XUV interferometry
Hedewig, Lina
[MoP.61] Controlling the orbital angular momentum of electron in neutral atom
Hu, Hongtao
[MoP.62] Chiral Optical Tweezers - efficient enantioseparation of molecules
Jones, Robert Michael
[MoP.63] Time delays in photodetachment of atomic and molecular anions at low energies
Dvořák, Jan
[MoP.64] Ultrafast atomic physics between the perturbative and strong-field regimes
Marshallsay, Sean
[MoP.65] Multidimensional spectroscopy protocols with attosecond pulses
Finkelstein-Shapiro, Daniel

[MoP.66] Enhancing chiral recognition in gas-phase molecules using chiral topological light
Haram, Nida
[MoP.67] Chiral topological light for detection of robust enantiosensitive observables
Mayer, Nicola
[MoP.68] Complete retrieval of photoelectron dynamics from partially-coherent measurements in entangled photoemission
Caillat, Jérémie
[MoP.69] Photoelectron momentum distributions using crossed linearly polarized attosecond pulses
Kjærsdam Telléus, Emilia M
[MoP.70] Ultrafast photoionization dynamics in the vicinity of CO autoionizing states
Faria, Felipe
[MoP.71] Time-resolved Auger decay of an O2 intruder-state
Short, Andrew
[MoP.72] Probing attosecond dynamics of chiral bound wave packets with an attoclock
Ge, Peipei
[MoP.73] Extended RPAE method solves the Argon delay-puzzle
Lindroth, Eva
[MoP.74] Signature of nuclear wavepacket interference in high-order harmonic generation during HCN/HNC isomerization process
Le, Van-Hoang
[MoP.75] Quantum path interference in high harmonic generation driven by highly bichromatic fields
Zou, Xiaozhou
[MoP.76] Decoherence phenomena of electron dynamics in liquid water
Crippa, Gabriele
[MoP.77] A general approximator for strong-field ionization rates
Agarwal, Manoram
[MoP.78] State- and phase-resolved vibrational wavepacket dynamics in N2 studied using all-attosecond transient absorption spectroscopy
Vibudh, Vibudh
[MoP.79] Chiral measures make chiral moments
Khokhlova, Margarita
[MoP.80] Observing the Jahn-Teller dynamics of the propadiene cation using interferometric strong-field ionization
Brupbacher, Katherine
[MoP.81] Photon momentum transfer and partitioning: from one to many
Ni, Hongcheng
[MoP.82] Subcycle conservation law in strong-field ionization
Ni, Hongcheng
[MoP.83] Ultrafast spin migration in the argon dimer cation
Carlström, Stefanos

[MoP.84] Attosecond coincidence spectroscopy in small molecules
Makos, Ioannis
[MoP.85] Investigating photoionization delays using an attosecond source synchronized with an infrared non-collinear optical parametric amplifier
Jahanzeb, Muhammad
[MoP.86] Nonlinear spectroscopic studies using intense gas high-order harmonics at the SYLOS COMPACT beam line, ELI-ALPS
Mukhopadhyay, Sourin
[MoP.87] Rotational pseudo-Doppler effect in solid-state high harmonic generation
Zuo, Zitan
[MoP.88] Quantum beating and cyclic structures in the phase-space dynamics of the Kramers-Henneberg atom
Aynul, Anika
[MoP.89] Transient extreme ultraviolet scatterometry for nanostructure morphology re-construction and probing material dynamics
Kechaoglou, Emmanouil
[MoP.90] Attosecond ionic photoionization spectroscopy
Xu, Yidan
[MoP.91] Interplay of locally chiral rotational and electronic currents in chiral molecules
Mirahmadi, Marjan
[MoP.92] Ultrafast TACOS
Terentjevas, Justas
[MoP.93] Strong-field theory for attosecond scanning tunneling microscopy
Davidovich, Daniel
[MoP.94] Ultrafast and topological chiral sensing with vector beams
Rodriguez, Aude
[MoP.95] Theoretical study of photoemission time delay for two-photon transition via Multiple Scattering approach
Miura, Yugo
[MoP.96] Ultrafast dissociation dynamics of alkyl iodides induced by few-fs UV pulses
Ryabchuk, Sergey
[MoP.97] Reconstruction of the ionization and recombination times from the spectral phase of the emitted HHG radiation
Laurent, Guillaume
[MoP.98] Spin-orbit delays in argon: tracking ionization delays across an autoionizing Rydberg resonance
Berglitsch, Thomas
[MoP.99] A highly accurate semiclassical method for atoms and molecules in ultrafast intense laser pulses
Le, Anh-Thu

13:00 - 14:30 [TuP] Poster Session (II) - Tuesday, July 8th

Poster Session sponsored by:







[TuP.1] Laser-driven high-flux source of coherent quasi-monochromatic XUV radiation for coincidence spectroscopy
Späthe, Julian

[TuP.2] Ionization-induced blue-shift of the driving pulse leads to increased harmonic yield and single attosecond pulse
Schütte, Bernd

[TuP.3] Polarization anisotropy of high harmonic generation in silicon controlled by coherent combination of ω-3ω fields
Gindl, Adam

[TuP.4] Complete polarization measurements on broadband high-harmonic radiation
Gulyás Oldal, Lénárd

[TuP.5] Beamline development optimized for STARPES : a highly tunable extreme ultraviolet lightsource
Nonnon, Pierre

[TuP.6] Enhancement of vacuum-ultraviolet laser generation through near-threshold harmonics driven by a few-cycle two-color laser field
Wang, Bincheng

[TuP.7] High-power and high-energy post-pulse-compression of a thin-disk laser
Gong, Xiaochun

[TuP.8] Generating visible-ultraviolet sub-femtosecond pulses in a single fiber and sampling their fields
Dassie, Marco

[TuP.9] 200-kHz laser system delivering few-cycle near-infrared pulses for attosecond strong-field experiments
Meier, Katrin

[TuP.10] Intensity-control of few-cycle pulses
Broughton, Joseph

[TuP.11] Spectral modulation and double attosecond pulse emission from relativistic laser-plasma interactions
Liu, Aihua

[TuP.12] High-intensity attosecond beamline for XUV pump-XUV probe investigations
Vardast, Sajjad

[TuP.13] High-harmonic spectroscopy probes the directional charge-density-wavestrengths of TiSe2
Tyulnev, Igor

[TuP.14] XUV yield optimization of two-color high-order harmonic generation in gases
Redon, Melvin

[TuP.15] Tracing electronic coherences at conical intersections with heterodyned attosecond four-wave-mixing spectroscopy
Rupprecht, Patrick
[TuP.16] High power sub-20-fs laser based on gas multi-pass cell
Li, Pinbin
[TuP.17] Filamentation-assisted isolated attosecond pulse generation
Chien, Yu-En
[TuP.18] Attosecond X-ray core-level chronoscopy of aromatic molecules
Ji, Jia-Bao
[TuP.19] Double-blind holography of ultrashort free-electron laser pulses
Azzolin, Agata
[TuP.20] Attosecond plasma lens
Svirplys, Evaldas
[TuP.21] Resolving attosecond time-delay in strong-field ultrafast process of valence-shell electrons of argon
Han, Lin
[TuP.22] Exploring the effects of pulse duration on high-order harmonic generation with tunable pulse post-compression
Westerberg, Saga
[TuP.23] Ultrafast bright non-Gaussian state of light
Rasputnyi, Andrei
[TuP.24] Enhancement of extreme ultraviolet pulse generation by orthogonally polarized laser-controlled spontaneous emission of Ar atoms
Wang, Zhi-Bin
[TuP.25] High harmonic generation from a solid-gas dual-target configuration
Xie, Baichuan
[TuP.26] Laser-assisted X-ray second harmonic generation
Xie, Xinhua
[TuP.27] High repetition rate, high average power XUV sources based on high harmonic generation
Manschwetus, Bastian
[TuP.28] Apparatus for attosecond spectroscopy driven by dual-color few-cycle lasers
Zhong, Shiyang
[TuP.29] Stimulated Raman transitions below and above the ionization threshold of neon using a free-electron laser
Carlström, Stefanos
[TuP.30] Attosecond pulses produced by a few-cycle laser
Ouahioune, Nedjma
[TuP.31] High harmonic generation from a high-repetition-rate liquid plasma mirror
Park, Seong Cheol
[TuP.32] Carrier-envelope phase-stabilized sub-10-fs multiple-plate compression for MHz isolated attosecond pulses
Okamoto, Takuya

[TuP.33] Improving absorption measurements in attosecond XFEL experiments using machine learning
Hu, Kaiyu
[TuP.34] Using AttosecondRayTracing to design a beam transport setup for attosecond pulses from surface plasma relativistic HHG
Kalouguine, Andre
[TuP.35] The effect of driving laser properties on the coherent extreme-ultraviolet wave-front from high harmonic generation
Körmöczi, Andor
[TuP.36] Continuous relativistic high-harmonic generation from a liquid-leaf plasma mirror at kHz repetition rate
Haessler, Stefan
[TuP.37] High harmonic polarization shaping around a Cooper minimum
Chen, Ming-Chang
[TuP.38] Asymmetric polarization gating for spectral tuning and temporal confinement of high-order harmonics
Vábek, Jan
[TuP.39] Isolation of bright soft x-ray attosecond pulses in a highly ionized medium using two-color synthesized driving laser
Jin, Cheng
[TuP.40] The generation of photoelectron vortices by chirped attosecond pulses
Zhang, Bingshuang
[TuP.41] Compact attosecond XUV source for the investigation of Raman-induced attosecond electronic coherences
Danylo, Rostyslav
[TuP.42] Single-stage multipass cell spectral broadening and compression of 90 fs, 300 µJ laser down to 8.5 fs
Pronin, Oleg
[TuP.43] Few-cycle isolated attosecond pulses generation with NIR driving fields
Wang, Xiaowei
[TuP.44] Few-femtosecond soft X-ray transient absorption spectroscopy with tuneable deep ultraviolet-visible pump pulses
Lee, Jacob
[TuP.45] A high repetition HHG source of ultrafast XUV pulses for the SXP instrument at the European XFEL
Grychtol, Patrik
[TuP.46] Tuneable few-femtosecond far-ultraviolet laser pulses generated by Yb-laser-driven resonant dispersive wave emission
Satpathy, Deepjyoti
[TuP.47] Few-cycle optical vortices for strong field physics
Guer, Matthieu

[TuP.48] Towards a new HHG driver laser platform: bridging Q-switched and mode-locked laser technology via multi-pass cell post-compression
Seitz, Marc
[TuP.49] Batch Bayesian optimization of attosecond betatron pulses from laser wakefield acceleration
Maslarova, Dominika
[TuP.50] High repetition rate deep ultraviolet and infrared beamline for time-resolved experiments on the few-femtosecond timescale
Oliveira e Silva, Ana
[TuP.51] Soft X-rays beyond 600 eV from a 45 W average-power, few-cycle, short-wave infrared driver
Walke, Daniel
[TuP.52] Shaping attosecond pulses using the minimum in high-order harmonic generation spectrum of argon atoms
Li, XiaoYong
[TuP.53] Nonlinear spin-orbit coupling of light in harmonic generation at solid surfaces
Nagai, Kohei
[TuP.54] High-power, few-cycle, short-wave infrared source for high-flux, soft X-ray generation at 200 kHz repetition rate
Juliano, Caroline
[TuP.55] Dark pulse interferometry for attosecond pulse characterization
Garcia-Hernandez, Mario
[TuP.56] A beamline with efficient isolated attosecond pulse generation and detection for time-resolved spectroscopy driven by synthesized sub-cycle pulses
Kubullek, Maximilian
[TuP.57] Efficient high-order harmonic generation in the overdriven regime in a sub-mm glass chip
Cannelli, Oliviero
[TuP.58] Hyper Spectral Ultrafast Source - HYPUS
Schmidt, Bruno
[TuP.59] Attosecond electron beams for XFEL and MeV-UED
Wang, Xijie
[TuP.60] Bright isolated XUV attopulse generation through high-order frequency mixing and ellipticity gating techniques
Batelić Janković, Mateja
[TuP.61] Towards 300 mJ, 1 kHz, < 1 ps thin-disk regenerative amplifiers
Osolodkov, Mikhail
[TuP.62] An in-line setup for the generation of a cross-polarized double attosecond pulse with controllable delay
Díaz Rivas, Daniel
[TuP.63] Control of attosecond XUV pulses using an intermediate-field spatial filtering
Valentin, Constance

[TuP.64] Attosecond metrology of vacuum-ultraviolet high-order harmonics generated in semiconductors via laser-dressed photoionization of alkali metals
Farkas, Balazs
[TuP.65] XUV-pump IR-probe resonant shake-up ionization of helium
Lewis, Nicholas
[TuP.66] SNAIL: A library for fast SFA calculation for HHG in gas targets
Benitez, Alex
[TuP.67] Xenon photoionization in the vicinity of 4d giant resonance and Cooper minimum using an XUV-NIR pump-probe experiment at FLASH
Litvinyuk, Igor
[TuP.68] First principles simulation of attosecond XUV pump-XUV probe spectra for small organic molecules
Martin, Fernando
[TuP.69] Optical parametric MPC: a hybrid approach for high-power few-cycle laser pulse generation
Rupp, Nikolas
[TuP.70] High-order harmonic generation driven by vector necklace beams
Oakley, Séraphine
[TuP.71] Direct optical characterisation of a driving field with a time-dependent ellipticity for high harmonic generation
Miller, Benjamin
[TuP.72] Attosecond pulse trains generation in hollow core waveguides
Vozzi, Caterina
[TuP.73] XUV chromatic aberrations in high order harmonic generation controlled by flat-top spatial shaping of the laser driver
Constant, Eric
[TuP.74] Tailoring XUV sources for ultrafast science: advances at ELI Beamlines
Hort, Ondřej
[TuP.75] High harmonic generation control by sub-cycle multi-color waveform synthesis of an ultraviolet to near-infrared driver
van der Linden, Roy
[TuP.76] Generation and characterisation of optical attosecond pulses with terawatt peak power
Nordmann, Joleik
[TuP.77] Generation of ultra-broadband millijoule-level radial vector beams
Heynck, Michael
[TuP.78] Commercial table-top beamline for attosecond science
Reiger, Simon
[TuP.79] High-power and ultrashort IR-driver platform for industrial XUV metrology
Guichard, Florent
[TuP.80] Post-compression of an Yb laser in two steps for attosecond transient absorption spectroscopy
Lericheux, Nicolas

[TuP.81] The effect of laser pulse duration on the yield of high harmonic generation
Nagyilles, Balazs
[TuP.82] Single-shot carrier-envelope phase measurement at 586 kHz using optical Fourier-Transform interferometry
Guo, Chen
[TuP.83] Over-barrier ionization dynamics studied by backpropagation
Ni, Hongcheng
[TuP.84] Optimizing harmonic generation yield and refocusing conditions
Poulain, Vénus
[TuP.85] KeV attosecond pulse generation driven by MIR lasers through waveform control and phase matching
Shim, Bonggu
[TuP.86] Spatial features of high-order harmonic generation from liquids
Tao, Wan Chen
[TuP.87] Driving the high harmonic process using a multi-pass cell
Steiner, Benjamin
[TuP.88] Short attosecond pulse trains for the time-slit experiments
Maroju, Praveen Kumar
[TuP.89] Dispersion-scan (re)meets neural networks
Canhota, Miguel
[TuP.90] Angular asymmetry of double ionization of Helium by ultrashort extreme ultraviolet laser pulses
Yang, Yugang
[TuP.91] Cumulative effect on extreme ultraviolet high harmonics generated by high repetition rate Yb-based amplifier
Yamada, Hotaka
[TuP.92] Highly efficient enantiosensitive low-order harmonic generation
Heilemann, Rico
[TuP.93] Time-resolved X-ray absorption spectroscopy of strong-field ionized water
Thierstein, Emily
[TuP.94] Synthesis of Tesla-scale attosecond magnetic pulses with ring-current gating
de las Heras, Alba
[TuP.95] Probing many-body effects in strongly correlated materials via reflectivity using infrared laser fields
Brown, Graham Gardiner
[TuP.96] Orbital angular momentum control in attosecond pulses of light
de las Heras, Alba
[TuP.97] Combining tunable few cycle UV and soft x-ray pulses at the European XFEL
Mullins, Terry
[TuP.98] In-situ waveform sampling in a reaction microscope
Gholam-Mirzaei, Shima
[TuP.99] Attohallen - A new attosecond science facility in Sweden
Zaki, Massume

13:00 - 14:30 [WeP] Poster Session (III) - Wednesday, July 9th

Poster Session sponsored by:

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[WeP.1] Rydberg states generation driven by quantum light
Liu, Jinlei
[WeP.2] Probing coherent phonon in BaF2 through time-resolved high harmonic generation spectroscopy
Tran, Tien-Dat
[WeP.3] Observation of an evanescent field-induced harmonic generation in preionized liquid water
Mizuno, Tomoya
[WeP.4] Tracking bandgap dynamics in solids with extreme ultraviolet high harmonic interferometry
Juergens, Peter
[WeP.5] Decoding the ring currents in organic molecules using tr-ARPES
Chordiya, Kalyani
[WeP.6] Phase evolution of strong-field ionization
Brown, Andrew
[WeP.7] Dynamical study of the competition between rotamerization and tautomerization of O-Hydroxybenzaldehyde
Vigneau, Jean-Nicolas
[WeP.8] Predicting ultrastrong coupling effects in extreme ultraviolet fields with combined Floquet and effective Hamiltonian theory
Bruhnke, Jakob Nicolai
[WeP.9] Linear dichroism of core excitons in LiF
Gannan, Kylie
[WeP.10] Rise and demise of entanglement in strongly coupled photoionization via quantum relay to spontaneous emission
Stenquist, Axel
[WeP.11] Non-classical bright states of light generated via light-matter entanglement in strong laser fields
Yi, Sili
[WeP.12] Semiconductor Wannier equations: a real-time, real-space approach to the nonlinear optical response in crystals
Ferreira Da Silva, Rui Emanuel
[WeP.13] Ultrafast imaging below the diffraction limit with high harmonic deactivation microscopy
Murzyn, Kevin

[WeP.14] Origin of sub-optical-cycle dephasing in high-order harmonic generation in crystalline solids
Kartashov, Daniil
[WeP.15] High harmonic generation from localized electronic states in solids
Liang, Shiqi
[WeP.16] Theoretical description of attosecond momentum-resolved resonant x-ray scattering of photo-excited molecules
Radionov, Maksim
[WeP.17] Quantitative characterization and manipulation of quantum entanglement in multiphoton ionization
Mao, Yi-Jia
[WeP.18] Ultrafast optical control of multi-valley states in 2D SnS
Hashmi, Arqum
[WeP.19] Sub-cycle switchable light-induced Floquet topological flat bands in Dirac system and its dynamics
Li, Gefei
[WeP.20] Pseudospin-driven polarimetric singularities in high harmonic generation of black phosphorus
Choi, Youngchan
[WeP.21] Revealing the Berry phase under the tunneling barrier
Faeyrman, Lior
[WeP.22] Polarization-dependent high-harmonic generation in organic molecular crystals
Schöpa, Samuel
[WeP.23] Impact of solvation on the ultrafast ring-opening dynamics of Furfural
Trester, Joel
[WeP.24] All-attosecond transient reflection spectroscopy
Drescher, Lauren B
[WeP.25] Efficient quantum state tomography of photoelectrons via polychromatic infrared pulses
Boati, Edoardo Alberto
[WeP.26] Quantum tomography of nonperturbative harmonic light from solids
Nisim, Ido
[WeP.27] Photoelectron emission from silver clusters on substrates
Bednov, Mikhail
[WeP.28] Element-specific phonon dynamics in Molybdenum Dioxide revealed by transient absorption spectroscopy
Burri, Florence
[WeP.29] Excitation and deexcitation dynamics in high harmonic generation of correlated electron systems
Bae, Gimin
[WeP.30] First-principles simulation for subcycle reciprocal image of electron density
Shinohara, Yasushi

[WeP.31] Novel quantum states of exciton-Floquet composites: electron-hole entanglement and information
Park, Hyosub
[WeP.32] Diatomic molecular vibrations in intense laser fields: an analytical perspective
Hack, Szabolcs
[WeP.33] Probing band structure properties through HHG in MgO: the role of ellipticity and Bloch confinement
Weiss, Roni
[WeP.34] Beyond electric-dipole treatment of light-matter interactions in materials: nondipole harmonic generation in bulk Si
Jensen, Simon Vendelbo Bylling
[WeP.35] Attosecond-resolved non-dipole photoionization dynamics
Ji, Jia-Bao
[WeP.36] Rabi oscillations, photo-emission and entanglement: A time-resolved picture
Rico, Léonardo
[WeP.37] Strong-field control over valley polarization and topology
Tyulnev, Igor
[WeP.38] Reduction of chromatic aberration in high order harmonic XUV beams using a temporal flat top
Miller, Benjamin
[WeP.39] Progress towards revealing attosecond dynamics in strongly correlated systems
Klingner, Jan Maximilian
[WeP.40] Electron trajectory analysis through carrier-envelope phase effects in high harmonic generation from ZnO
Allegre, Hortense
[WeP.41] Phase transitions induced by quantum vacuum
Klimkin, Nikolai
[WeP.42] Time-resolved coupled electron-nuclear dynamics of Methane cation upon photoionization using an XUV attopulse
Pandey, Gaurav
[WeP.43] Loss of contrast in attosecond temporal double slit interferometry from atomic photoionization
Ngoko Djiokap, Jean Marcel
[WeP.44] Ultrafast charge migration in solvated molecules
Giri, Sucharita
[WeP.45] Update on the Modular Multiscale Approach (MMA) to simulate high harmonic generation in gases
Vábek, Jan
[WeP.46] Proposal for high-energy cutoff extension of optical harmonics of solid materials using the example of a one-dimensional ZnO crystal
Lang, Yue

[WeP.47] Primary events of the MLCT dynamics of photoexcited Iron(II)-Tris(2,2'-Bipyridine) in solution probed by transient soft X-ray absorption spectroscopy
Rouzée, Arnaud
[WeP.48] Coherent phonon-driven modifications of orbital-specific local fields
Neb, Sergej
[WeP.49] Photoionization time delays in H_2O
Pranjal, Prateek
[WeP.50] First-principles calculations of high-harmonic generation in amorphous Si and SiO
Gushiken, Eiyu
[WeP.51] Ultrafast correlation dynamics during high-harmonic generation in multielectron atoms
Buczolich, Katharina
[WeP.52] Electron-nuclear energy sharing through low-energy inelastic recollisions in dissociative strong-field ionization of deuterium
Hell, Sebastian
[WeP.53] Experimental realisation of synthetic chiral light
Picciuto, Rose
[WeP.54] Ab initio calculation of real-space light-driven current in graphene under strong laser fields
Li, Siyuan
[WeP.55] Photoionization of chiral molecules by sub-cycle shaped laser fields
Fede, Letizia
[WeP.56] Ga3d and Se3d spin orbit delays in GaSe solids
Schabbehard, Tillmann
[WeP.57] High-order harmonic generation in Pentacene
Wiechmann, Falk-Erik
[WeP.58] A universal approach to saddle-point methods in attosecond science
Weber, Anne
[WeP.59] Relativistic calculations of electron-parent ion entanglement using the KRAKEN protocol
Petersson, Leon
[WeP.60] Probing the final state continuum on atomic length scales by attosecond time-resolved photoelectron spectroscopy
Gebauer, Andreas
[WeP.61] Ultrafast charge carrier dynamics in semiconductor nanostructures resolved in time, space and energy
K. Jensen, Thomas
[WeP.62] Scaling of the fractional angular momenta of a light polarization Möbius strip in extreme nonlinear optics
Ruchon, Thierry
[WeP.63] Calculations of impulsive stimulated X-ray Raman scattering in liquid Water
Rego, Laura

[WeP.64] On the use of a self-homodyning experiment to study the quantum nature of high harmonics generated in solids
Cotte, Viviane
[WeP.65] Spatial mode transformations of high-order harmonic generation in transition metal dichalcogenides
Schlemmer, Leon
[WeP.66] Attosecond transient reflectivity and absorption from an Ytterbium laser driven high harmonic generation source
Tyagi, Akansha
[WeP.67] Two-color high harmonic spectroscopy of ultrafast Dirac electron dynamics
Krüger, Michael
[WeP.68] Electron-ion covariance spectrometry for molecular dynamics
Månsson, Erik
[WeP.69] Control of Graphene by two-colour laser fields
Boyt Archibald, Elliott
[WeP.70] A nonlinear optical chirality for high-harmonic generation
Pisanty, Emilio
[WeP.71] Enantiosensitive exceptional points of open chiral systems
Mayer, Nicola
[WeP.72] Topological valley control of graphene by tailored two-colour fields using a light and fast SBE solver
Tomaselli, Anna
[WeP.73] Competition between proton transfer and resonant intermolecular Coulombic decay in water dimers
Calzolari, Alessandro
[WeP.74] Reconstruction and characterization of single particle entanglement dynamics in photoionization
Berkane, Morgan
[WeP.75] Attosecond resolved intramolecular charge transfer in neutral donor-acceptor organic molecules
Martin, Fernando
[WeP.76] Relativistic core-level molecular photoionization in heavy molecules
Zapata, Felipe
[WeP.77] Observation of nonadiabatic coupling induced by Floquet-Bloch states in high harmonic generation from large-bandgap materials
Zhao, Cong
[WeP.78] Towards attosecond photoemission electron microscopy: spatially-resolved strong-field electron acceleration from a nanostructure
Vogelsang, Jan
[WeP.79] Ultrafast ethylene dication formation through resonant enhanced multiphoton ionization
Marante, Carlos

[WeP.80] Adapting polarization gating for generation of isolated attosecond pulses after multiple mirror reflexions
Constant, Eric
[WeP.81] High-order frequency mixing: harvesting bright XUV from propagation
Khokhlova, Margarita
[WeP.82] Observing coherent electronic wavepackets in molecules using attosecond transient absorption spectroscopy
Bredehoeft, Jona
[WeP.83] eTraj.jl: Trajectory-based simulation for strong-field ionization
Ni, Hongcheng
[WeP.84] Self-consistent extraction of photoabsorption time delays in attosecond streak camera
Ni, Hongcheng
[WeP.85] Analysis of low-energy electron interferences in Helium
Eriksson, Samuel
[WeP.86] Probing laser dressing-induced dynamics in Acetonitrile with soft X-ray attosecond transient absorption spectroscopy
Ardali, Emir
[WeP.87] Generation of high-order harmonics with hybrid spin and transverse orbital angular momentum from a crystalline solid
Zhang, Jianing
[WeP.88] Ultrafast light-induced spin and orbital magnetization in two-dimensional semiconductors
Muraleedharan, Mrudul
[WeP.89] Manipulating molecular dynamics with polarization-shaped laser pulses
Pan, Shengzhe
[WeP.90] Non-cascade random walk in solid high harmonic generation
Wang, Yiwen
[WeP.91] Robust discrimination between solid-state high harmonic generation mechanisms based on real-space dynamics
Brown, Graham Gardiner
[WeP.92] Electronic dynamics created at conical intersections and its dephasing in aqueous solution
Chang, Yi-Ping
[WeP.93] In search of lost tunneling time
Maier, Pablo Martin
[WeP.94] Enantiosensitive geometric phases in multi-photoexcitation processes
Roos, Aycke
[WeP.95] Ultra-fast nonlinear optical response of chiral molecules with a focus on conformer sensitivity
Christou, Elena Aethra

[WeP.96] Spin and orientation locking in photoionization of chiral molecules
Flores, Philip Caesar
[WeP.97] An entanglement perspective on molecular Auger decay
Östberg, E
[WeP.98] Photoelectron spectra calculated from absorption
Steinsvåg Dalen, Bendik
[WeP.99] Non-adiabatic dynamics investigation of Cr(CO)4(2,2'-bipyridine)
Ciborowski, Bartosz

NOTES

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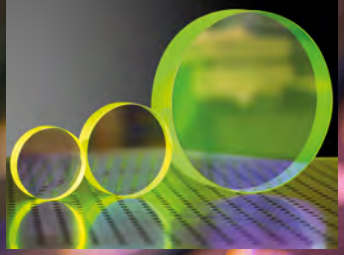
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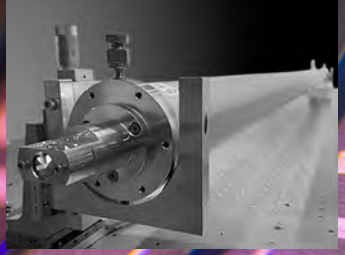
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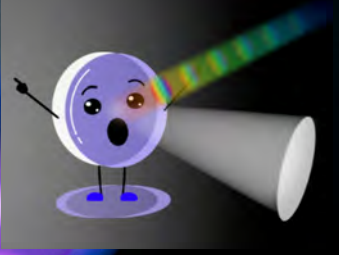
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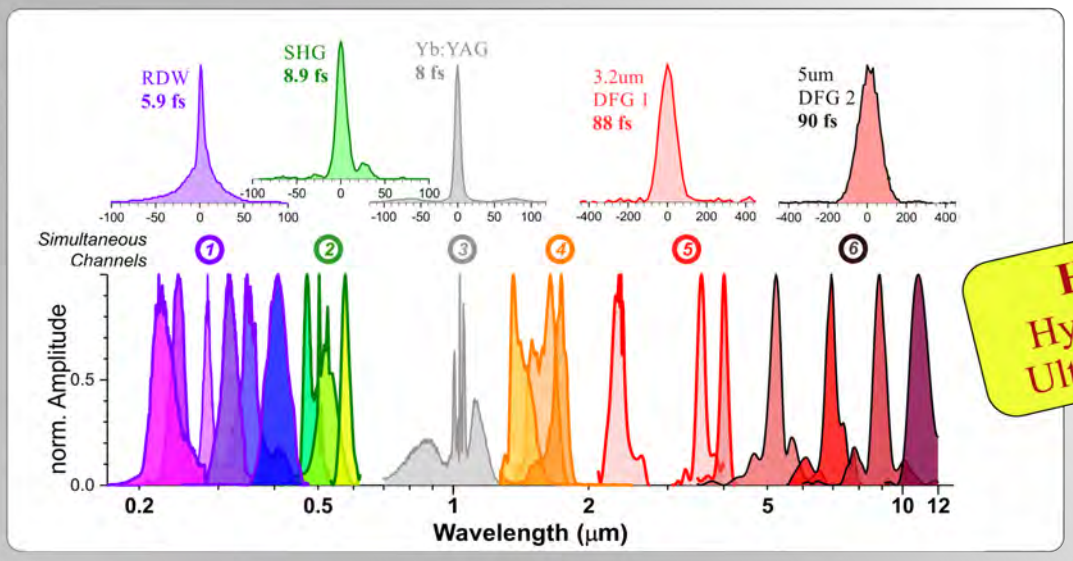
Moonlander HHG

Attosecond Beamlines

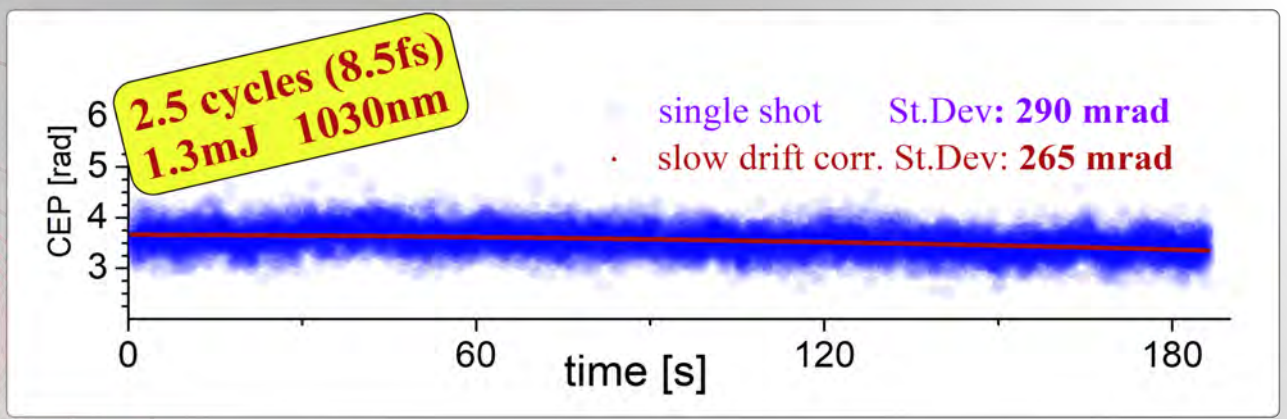


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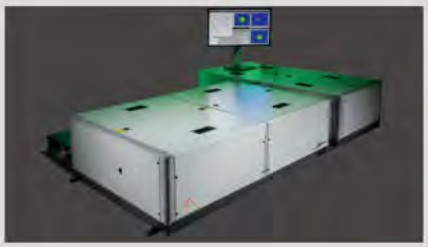


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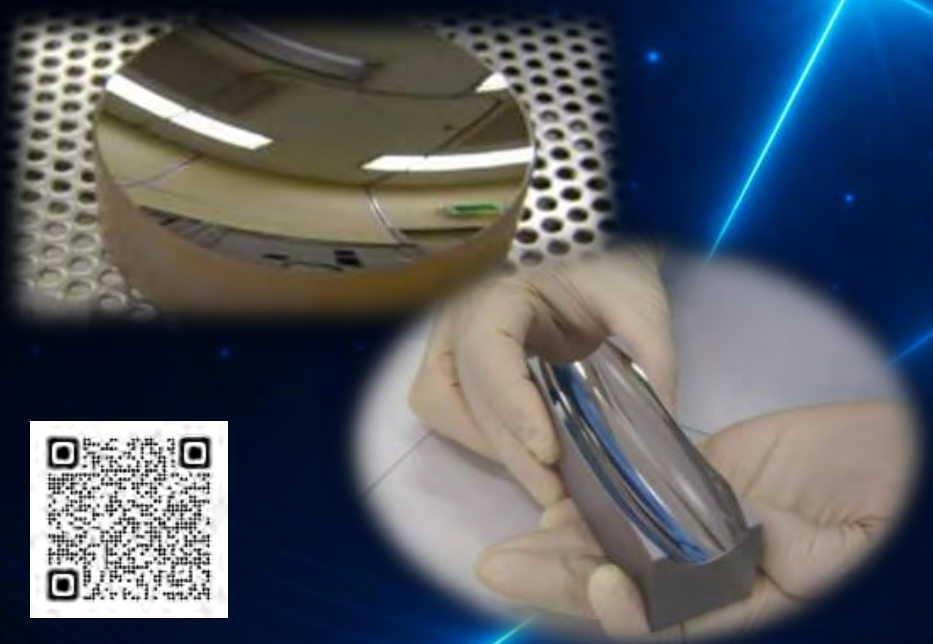
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- High-harmonic generation
- Plasma-based sources
- Free-electron lasers
- Synchrotrons



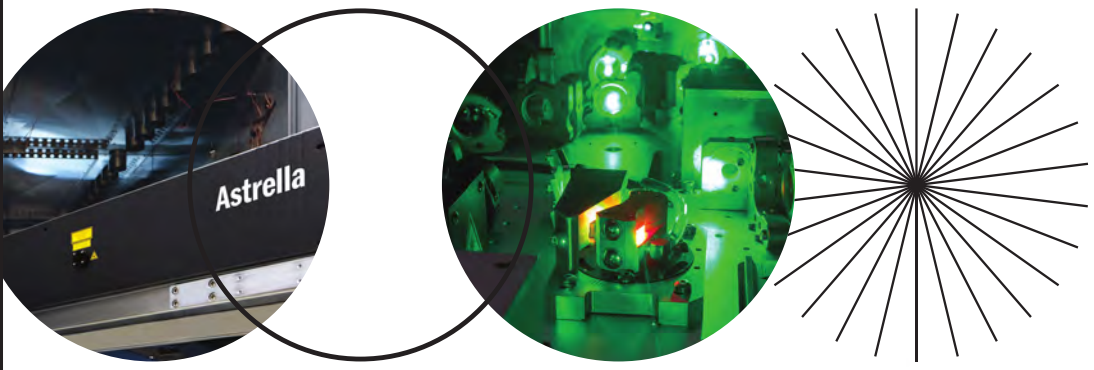
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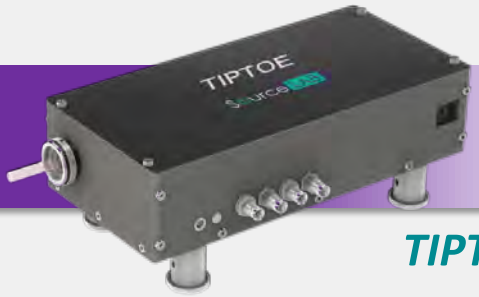
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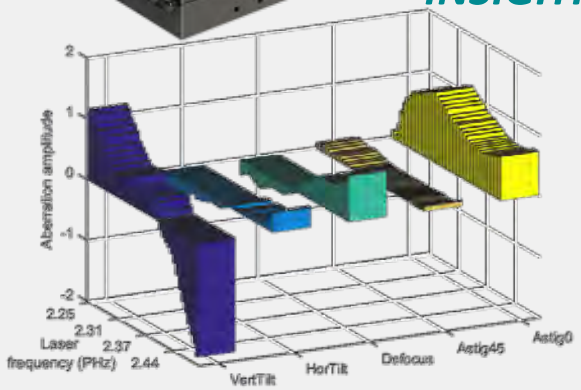
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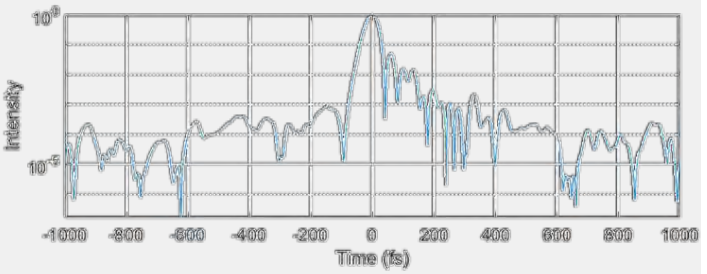
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TIPTOE



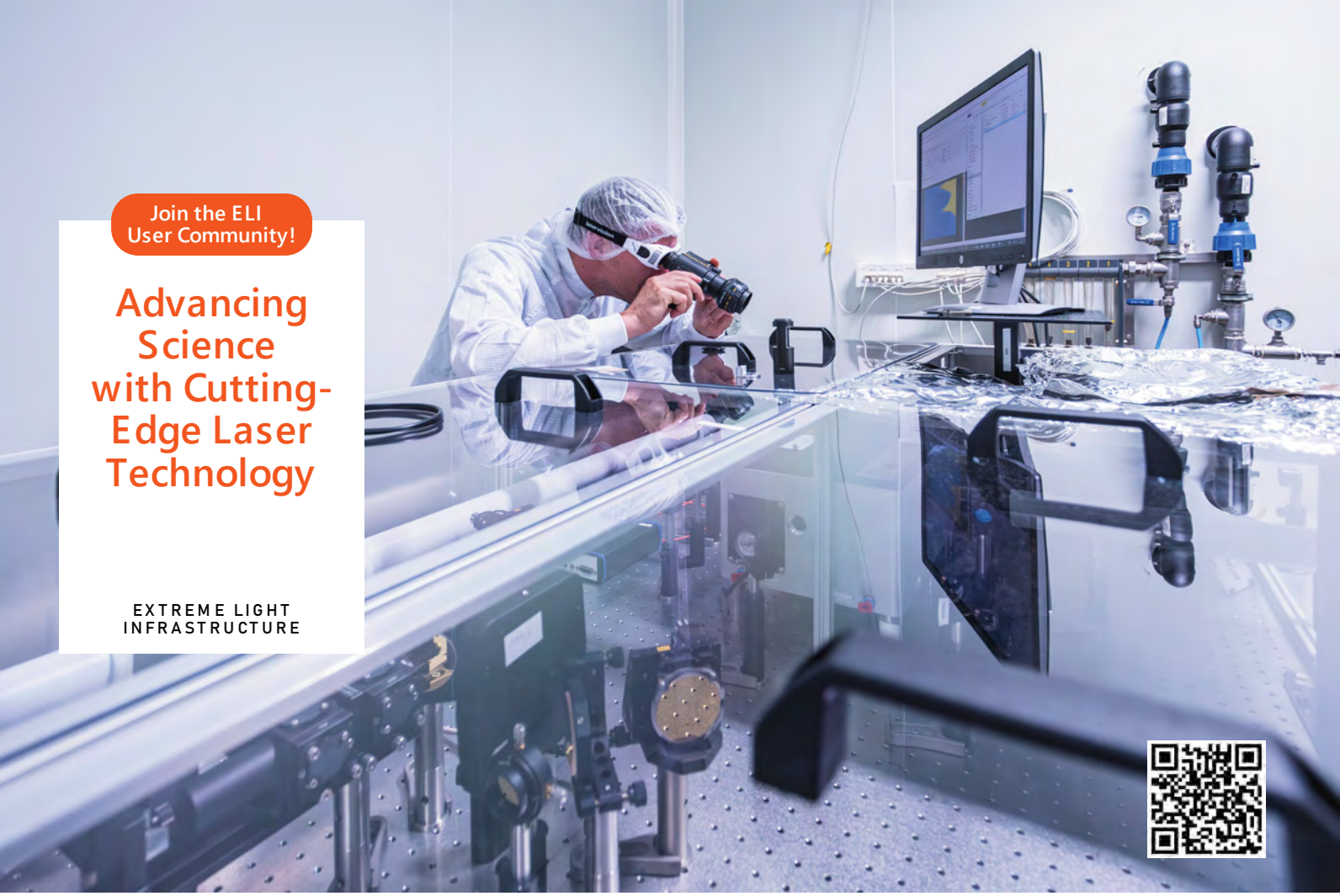
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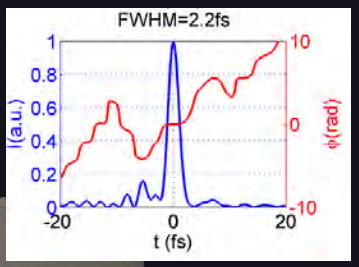
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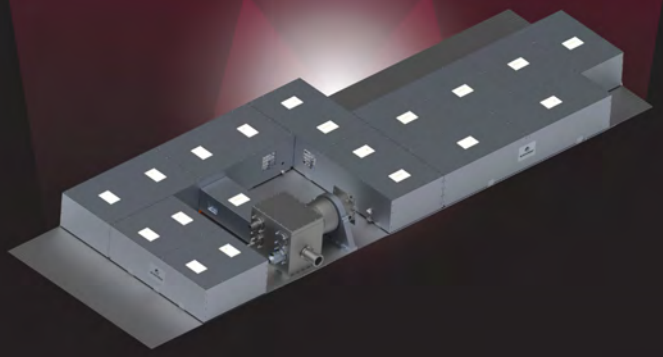




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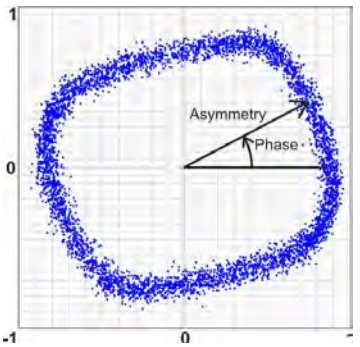
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Book of Abstracts

ATTO X

The 10th International Conference on Attosecond Science and Technology

2025-07-06 — 2025-07-11 in Lund, Sweden

PRESENTATIONS:

Attosecond Physics and spectroscopy (I)

[Mo1.1] **Attosecond science: from adolescence to adulthood** *(Tutorial)*
Vrakking, Marc , Max Born Institut (MBI), Max-Born Strasse 2a, 12489 Berlin, Germany

[Mo1.2] **Time-Dependent Close Coupling on the heels of attosecond electron dynamics** *(Invited)*
Argenti, Luca , Department of Physics, Chemistry, and CREOL, University of Central Florida, Orlando, FL 32816 (U.S.A.)

[Mo1.3] **Laser-assisted dynamical interference of photoelectrons driven by chirped extreme-ultraviolet pulses** *(Contributed)*
Vismarra, Federico , Department of Physics, Politecnico di Milano, Italy; Institute of Photonics and Nanotechnologies, IFN-CNR, Italy; Laboratorium fur Physikalische Chemie, ETH Zurich, Switzerland

[Mo1.4] **Attosecond spectroscopy reveals electron correlation in the photoionization of Argon** *(Contributed)*
Luo, Sizuo , Institute of atomic and molecular physics, Jilin University, Changchun, 130012, China.

Attosecond Physics and spectroscopy (II)

[Mo2.1] **Chiral molecules in strong laser fields** *(Invited)*
Mairesse, Yann , Université de Bordeaux - CNRS - CEA, CELIA, UMR5107, Talence, France

[Mo2.2] **Vectorial attosecond transient spectroscopy** *(Contributed)*
Yaffe, Noa , Weizmann Institute of Science, Rehovot, 7630031, Israel

[Mo2.3] **Attosecond XUV pump-control spectroscopy for tracking and modification of excited neutral molecules** *(Contributed)*
Borisova, Gergana D. , Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

[Mo2.4] **Clocking the tunneling time in strong-field ionization** *(Contributed)*
Dubois, Jonathan , Sorbonne Université, CNRS, Laboratoire de Chimie Physique - Matière et Rayonnement, LCPMR, 75005 Paris, France

Attosecond Quantum Science (I)

[Mo3.1] **Quantum-orbit dynamics in strongly-polychromatic laser fields** *(Invited)*
Pisanty, Emilio , Attosecond Quantum Physics Laboratory, King's College London, WC2R 2LS London, UK

[Mo3.2] **Ion-photoelectron entanglement and coherence controlled by two-color laser pulses** *(Invited)*
Ishikawa, Kenichi L. , Department of Nuclear Engineering and Management, Graduate School of Engineering, The University of Tokyo, Tokyo 113-8656, Japan; Photon Science Center, Graduate School of Engineering, The University of Tokyo, Tokyo 113-8656, Japan; Research Institute for Photon Science and Laser Technology, The University of Tokyo, Tokyo 113-0033, Japan; Institute for Attosecond Laser Facility, The University of Tokyo, Tokyo 113-0033, Japan

[Mo3.3] **Measurement of photoelectron decoherence and attosecond dynamics of unobserved ions** *(Contributed)*
Granveau, Gabriel , Université Paris-Saclay, CNRS, IOGS, Laboratoire Charles Fabry, 91127 Palaiseau, France

[Mo3.4] **Testing quantum mechanics with photoelectron interferometry** *(Contributed)*
Arvidsson, Gustav , Department of Physics, Lund University, Box 118, 221 00 Lund, Sweden

[Mo3.5] **Strong-field dynamics of electrons photoemitted with non-classical light** *(Contributed)*
Pölloth, Jonathan , Department of Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

Attosecond Quantum Science (II)

[Mo4.1] **Giant counter-rotating oscillations and quantum entanglement from time-dependent strong coupling in the extreme ultraviolet** *(Invited)*
Dahlström, J. Marcus , Department of Physics, Lund University, 22100 Lund, Sweden

[Mo4.2] **Experimental evidence of the quantum-optical nature of high- harmonic generation in semiconductors** *(Invited)*
Merdji, Hamed , Laboratoire d'Optique Appliquée, École Polytechnique, Palaiseau, 91120, France

[Mo4.3] **Photon bunching in high-harmonic emission controlled by quantum light.** *(Contributed)*
Vampa, Giulio , Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, Ottawa, K1N 0R6, Canada

[Mo4.4] **Non-classical Effects Drive Recombination in High-Harmonic Generation Using Circularly Polarized Fields** *(Contributed)*
Ciappina, Marcelo , Guangdong Technion-Israel Institute of Technology, Physics Program, China; Technion - Israel Institute of Technology, Israel,

[Mo4.5] **Measuring and controlling the birth of quantum attosecond pulses** *(Contributed)*
Even Tzur, Matan , Department of Physics, Technion–Israel Institute of Technology, Israel; Solid State Institute, Technion–Israel Institute of Technology, Israel; Helen Diller Quantum Center, Technion–Israel Institute of Technology, Israel

Laser Technology for Attoscience (I)

[Tu1.1] **Attosecond X-ray light sources driven by mid-infrared lasers** *(Tutorial)*
Chang, Zenghu , Laboratory for Infrared-driven Intense-field Science (LIRIS), Department of Physics, University of Ottawa, 25 Templeton St. Ottawa, ON K1N 7N9, Canada

[Tu1.2] **High power CEP stable Ti:Sa amplifier seeded by OPCPA** *(Contributed)*
Pertot, Yoann , Fastlite by Amplitude, 165 rue des Cistes, 06600 Antibes, France

[Tu1.3] **Waveform-controlled near-single-cycle pulses at the 100 TW level for intense attosecond sources** *(Contributed)*
Veisz, Laszlo , Department of Physics, Umeå University, Linnaeus väg 24, 90187, Umeå, Sweden

[Tu1.4] **Single-shot pulse retrieval of femtosecond bright squeezed vacuum** *(Contributed)*
Kern, Yuval , Department of Physics, Technion - Israel Institute of Technology, 32000 Haifa, Israel; Solid State Institute and Helen Diller Quantum Center, Technion - Israel Institute of Technology, 32000 Haifa, Israel

[Tu1.5] **Characterization of sub-3-fs tunable VUV pulses** *(Contributed)*
Kretschmar, Martin , Max Born Institute, Max-Born-Str. 2A, 12489 Berlin, Germany

Laser Technology for Attoscience (II)

[Tu2.1] **New ultrafast sources and science enabled by advanced nonlinear optics in hollow-core waveguides** *(Invited)*
Brahms, Christian , School of Engineering and Physical Sciences, Heriot-Watt University, Edinburgh EH14 4AS, United Kingdom

[Tu2.2] **Multi-Mirror Multi-Pass Cells for Compression of Energetic Narrowband Laser Pulses into the Femtosecond Regime** *(Contributed)*
Beaufort, Gaspard , Deutsches Elektronen-Synchrotron DESY, Hamburg, 22607, Germany; Department of Physics, Lund University, Lund, SE-22100, Sweden

[Tu2.3] **Pulse compression of 300 W, 12 mJ with 83% transmission in hollow-core fiber** *(Contributed)*
Schmidt, Bruno , few-cycle Inc

[Tu2.4] **Terawatt optical attosecond pulses and 100 GW-scale far-ultraviolet pulses through extreme soliton dynamics** *(Contributed)*
Kotsina, Nikoleta , Heriot-Watt University, School of Engineering and Physical Sciences, United Kingdom

Attosecond HHG Sources (I)

[Tu3.1] **Recent developments of attosecond high-harmonic generation sources in and outside the Extreme Light Infrastructure** *(Invited)*
Major, Balázs , ELI ALPS, ELI-HU Non-Profit Ltd., Wolfgang Sandner utca 3., Szeged, H-6728, Hungary

[Tu3.2] **Microfluidic-based sources for efficient XUV generation and manipulation** *(Invited)*
Vozzi, C. , CNR Istituto di Fotonica e Nanotecnologie, Milano, 20133, Italy

[Tu3.3] **High Harmonic Generation with Two Non-Collinear Drivers: A Unique Gateway to Extreme Nonlinear Phenomena** *(Contributed)*
Ruchon, Thierry , Université Paris-Saclay, CEA, LIDYL, 91191 Gif-sur-Yvette, France

[Tu3.4] **High-order harmonic up-conversion of spatiotemporal and spatioespectral optical vortices into the extreme ultraviolet** *(Contributed)*
Martin-Hernandez, Rodrigo , Grupo de Investigación en Aplicaciones del Láser y Fotónica, Universidad de Salamanca, 37008, Salamanca, Spain; Unidad de Excelencia en Luz y Materia Estructuradas (LUMES), Universidad de Salamanca, Salamanca, Spain

[Tu3.5] **Hollow Gaussian Beams for Low-Divergence High Harmonic Generation and Attosecond Pulse Optimization** *(Contributed)*
Redon, Melvin , Department of Physics, Lund University, P.O. Box 118, 22100, Lund, Sweden

Attosecond HHG Sources (II)

[Tu4.1] **Table-top all-attosecond transient absorption spectroscopy** *(Invited)*
Schütte, Bernd , Max-Born-Institut, Max-Born-Str. 2A, 12489 Berlin, Germany

[Tu4.2] **Coincidence measurements of two-photon double ionization driven by a high-harmonic generation source** *(Contributed)*
Kübel, Matthias , Institute of Optics and Quantum Electronics, Friedrich-Schiller University, 07743 Jena, Germany; Helmholtz Institute Jena, 07743 Jena, Germany

[Tu4.3] **Using nanostructured dielectric surfaces for the generation of nano-focused XUV radiation** *(Contributed)*
Bastani, Parnia , Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, Otawa, Ontario K1N 0R6, Canada; Advanced Laser Light Source (ALLS), Centre Énergie Matériaux Télécommunications, Institut National de la Recherche Scientifique (INRS-EMT), 1650 Boulevard Lionel-Boulet, Varennes, Quebec J3X 1P7, Canada

[Tu4.4] **Efficient water-window isolated attosecond pulse generation with tailored sub-cycle fields** *(Contributed)*
Silva-Toledo, Miguel Angel , Center for Free-Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, Hamburg, 22607, Germany; Physics Department and The Hamburg Centre for Ultrafast Imaging (CUI), University of Hamburg, Notkestrasse 9-11, Hamburg, 22607, Germany

[Tu4.5] **Optimizing polarization gated generation of isolated attosecond pulses without CEP stabilization** *(Contributed)*
Constant, Eric , Universite Claude Bernard Lyon 1, CNRS, Institut Lumiere Matiere, UMR5306, Villeurbanne 69100, France

[Tu4.6] **Direct measurement of the dipole phase in solid-state high-harmonic generation via XUV attosecond interferometry** *(Contributed)*
Kuzkova, Nataliia , Advanced Research Center for Nanolithography, the Netherlands; Department of Physics and Astronomy, and LaserLaB, Vrije Universiteit Amsterdam, the Netherlands

Attosecond Chemistry (I)

[We1.1] **Attosecond chemistry: tutorial and theoretical perspectives** *(Tutorial)*
Vacher, Morgane , Nantes Université, CNRS, CEISAM, UMR 6230, F-44000 Nantes, France

[We1.2] **Advances in ultrafast ultraviolet spectroscopy of molecules** *(Invited)*
Wanie, Vincent , Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, 22607, Germany

[We1.3] **Probing iodine chemical environment with ionization delays** *(Contributed)*
Menezes Ferreira, Rafael , Universtié Paris-Saclay, CEA, LIDYL, 91190 Gif-sur-Yvette, France

[We1.4] **Probing the ultrafast vibronic wavepacket dynamics of ethylene with sub-4fs VUV pulses** *(Contributed)*
Sen, Arnab , Max-Born Institute

Attosecond Chemistry (II)

[We2.1] **Strong electron-electron-nuclei correlations in multiphoton single and double ionization** *(Invited)*
Palacios, Alicia , Universidad Autónoma de Madrid, Campus Cantoblanco, 28049, Madrid, Spain

[We2.2] **Attosecond Coherent Control and Measurement of the Photoionization Dynamics of Chiral Molecules** *(Contributed)*
Ji, Jia-Bao , ETH Zürich, Laboratorium für Physikalische Chemie, Switzerland

[We2.3] **Control of Photo-Ion Circular Dichroism Using Orthogonal Laser Beams** *(Contributed)*
Greenwood, Jason , School of Mathematics and Physics, Queen's University Belfast, Belfast, United Kingdom

[We2.4] **All-optical coherent control of chiral electronic transitions for highly enantioselective photochemistry using femtosecond lasers** *(Contributed)*
Ordóñez, Andrés , Imperial College London, Department of Physics, UK

Attosecond Chemistry (III)

[We3.1] **Attosecond time-resolved electronic and vibrational dynamics in furan** *(Invited)*
Gräfe, Stefanie , Friedrich-Schiller-Universität Jena, 07743 Jena, Germany; Fraunhofer Institute for Applied Optics and Precision Engineering, 07745 Jena, Germany

[We3.2] **Attosecond dynamics in molecules: probing ultrafast charge motion with EUV/IR and UV/EUV spectroscopy** *(Invited)*
Nisoli, Mauro , Department of Physics, Politecnico di Milano, Milano, 20133, Italy; Institute for Photonics and Nanotechnologies, IFN-CNR, Milano, 20133, Italy

[We3.3] **Time-resolved x-ray absorption spectroscopy in thiophene covering sulfur L and carbon K Edges** *(Contributed)*
Kopp, Tristan , Laboratory of Physical Chemistry, ETH Zurich, Zurich, Switzerland

[We3.4] **Time-resolving molecular dissociation with FEL-pump and HHG-probe pulses** *(Contributed)*
Magunia, Alexander , Max-Planck-Institut für Kernphysik, Germany

Attosecond Science in Condensed Matter (I)

[We4.1] **Observation of the Berry phase via attosecond interferometry** *(Invited)*
Dudovich, Nirit , Weizmann Institute of Science, Rehovot, Israel

[We4.2] **Strongfield physics inside of (light-dressed) graphene and at the surface of metal needle tips** *(Invited)*
Hommelhoff, Peter , Physics Department, Friedrich-Alexander University Erlangen-Nürnberg (FAU), Erlangen, 91058, Germany; Max Planck Institute for the Science of Light, Erlangen, 91058, Germany; Physics Department, Ludwig-Maximilian University München, Munich, 80799, Germany

[We4.4] **Subcycle band-structure videography of lightwave-driven graphene** *(Contributed)*
Inzani, Giacomo , Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, Regensburg, 93040, Germany

[We4.5] **Discovery of a Hybrid Exciton State by Attosecond Spectroscopy** *(Contributed)*
Di Palo, Nicola , Department of Physics, Politecnico di Milano, Milano, 20133, Italy

[We4.6] **Correlations drive the attosecond response of strongly-correlated insulators.** *(Contributed)*
Cazali, Romain , Université Paris-Saclay, CEA, LIDYL, France

[We4.7] **Excitonic high harmonics and attosecond time delays in GaN** *(Contributed)*
Chen, Zhaopin , Department of Physics and Solid State Institute, Technion-Israel Institute of Technology, Haifa 3200003, Israel

Attosecond Science in Condensed Matter (II)

[Th1.1] **Probing ultra-fast electron dynamics in liquids and solutions using high-harmonic spectroscopy** *(Invited)*
Mondal, Angana , Laboratorium für Physikalische Chemie, ETH Zürich, Zurich, 8093, Switzerland

[Th1.2] **Attomicroscopy imaging and control of electron motion in solid-state** *(Invited)*
Hassan, Mohammed Th. , Department of Physics, University of Arizona, Tucson, AZ 85721, USA

[Th1.3] **Attosecond spectroscopy of virtual charge dynamics** *(Contributed)*
Lucchini, Matteo , Department of Physics, Politecnico di Milano, Piazza Leonardo da Vinci, 20133 Milano, Italy; Institute for Photonics and Nanotechnologies, IFN-CNR, 20133 Milano, Italy

[Th1.4] **Two-color control of attosecond scanning tunneling microscopy** *(Contributed)*
MA, BOYANG , Physics Department and Solid State Institute and Helen Diller Quantum Center, Technion-Israel Institute of Technology, Israel

[Th1.5] **Attosecond electron pulses with chiral mass and charge** *(Contributed)*
Fang, Yiqi , Universität Konstanz, Fachbereich Physik, 78464 Konstanz, Germany

Attosecond Science in Condensed Matter (III)

[Th2.1] **High-harmonic spectroscopy probes lattice dynamics: extension to bandgap dynamics and structural phase transitions** *(Invited)*
Luu, Tran Trung , Department of Physics, The University of Hong Kong, Hong Kong SAR

[Th2.2] **Ultrafast control of valley physics in 2D materials** *(Invited)*
Jiménez-Galán, Álvaro , Instituto de Ciencia de Materiales de Madrid (ICMM), CSIC, Madrid 28049, Spain

[Th2.3] **High harmonic generation from solutions** *(Contributed)*
Gaarde, Mette , Louisiana State University, Department of Physics and Astronomy, USA

[Th2.4] **Enhancement of harmonic generation in liquid water jet by resonant excitation in the mid-infared pulses** *(Contributed)*
Itatani, Jiro , The Institute for Solid State Physics, The University of Tokyo

Attosecond Science at FELs (I)

[Fr1.1] **Nonlinear science with multi-harmonic FEL at sub-femtosecond resolution** *(Invited)*
Callegari, Carlo , Elettra - Sincrotrone Trieste, 34149 Basovizza, Trieste, Italy

[Fr1.2] **Dephasing in attosecond science and the collective nature of structural quantum fluctuations** *(Invited)*
Santra, Robin , Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, 22607 Hamburg, Germany; Department of Physics, University of Hamburg, 22607 Hamburg, Germany

[Fr1.3] **Coherent control with two photons at a seeded free-electron laser** *(Contributed)*
Saalmann, Ulf , Max-Planck-Institut für Physik komplexer Systeme, Nöthnitzer Str. 38, 01187 Dresden, Germany

[Fr1.4] **Complex attosecond waveform synthesis using regular- and irregularly spaced harmonic combs at seeded free-electron laser FERMI** *(Contributed)*
Maroju, P K , Physikalisches Institut, University of Freiburg, Hermann-Herder-Straße 3, 79104 Freiburg, Germany; Department of Physics, Lund University, PO Box 118, SE-221 00 Lund, Sweden

[Fr1.5] **Magnetization Dynamics probed with Helicoidal Dichroism** *(Contributed)*
Guer, Matthieu , Université Paris-Saclay, CEA, LIDYL, 91191 Gif-sur-Yvette, France

Attosecond Science at FELs (II)

[Fr2.1] **Ultrafast Electron Motion Probed with Attosecond X-ray Free Electron Lasers** *(Invited)*
Cryan, James , Stanford PULSE Institute; Linac Coherent Light Source SLAC National Accelerator Laboratory, Menlo Park, CA, 94070, USA

[Fr2.2] **Characterization of polarized attosecond soft X-ray pulses at EuXFEL** *(Contributed)*
Leroux, Juliette , Department of Physics, University of Hamburg, 22607 Hamburg, Germany

[Fr2.3] **Redirection and reshaping of intense extreme-ultraviolet radiation** *(Contributed)*
He, Yu , Max Planck Institute for Nuclear Physics, Germany

[Fr2.4] **Impulsive stimulated X-ray Raman scattering in the condensed phase with attosecond X-ray Free Electron Laser pulses** *(Contributed)*
Egun, Felix , Felix Egun, Department of Physics, Imperial College London, South Kensington, SW7 2AZ, United Kingdom

[Fr2.5] **Single-shot measurements of soft x-ray attosecond pulses at kilohertz repetition rate** *(Contributed)*
Guo, Veronica , Stanford University; SLAC National Accelerator Laboratory

POSTERS:

Poster session 1

[MoP.1] **Characterizing and controlling lifetimes of doubly-excited states directly in the time domain**
— *Rupprecht, Patrick* —

[MoP.2] **RABBITT-HOP: RABBITT with Higher-Order Processes**
— *Brown, Andrew* —

[MoP.3] **Ultrathin liquid flat jet system for attosecond spectroscopy of solvated molecules**
— *Rockenstein, Sabine* —

[MoP.4] **Conditions for Mollow Triplets in Ultrafast Absorption and Dynamical Fluorescence**
— *Stenquist, Axel* —

[MoP.5] **Absolute photoemssion timing and asymmetry parameter of the I4d giant resonance of the Iodoalkanes**
— *Forster, Maximilian* —

[MoP.6] **Attosecond resonant Auger spectroscopy**
— *Kimberg, Victor* —

[MoP.7] **Attosecond partial wave meter and its application in resolving the photoemission time delays in atoms and molecules**
— *Gong, Xiaochun* —

[MoP.8] **Nonlocal resonance theory for electron–nuclear dynamics in molecules**
— *Bertolino, Mattias* —

[MoP.9] **Ultrafast Time-varying Nonlinearity at Epsilon-near-zero**
— *Li, Yan* —

[MoP.10] **Symmetries, dominance and quantum interference in below-threshold nonsequential double ionization with linearly polarized two-color fields**
— *Hashim, Sufia* —

[MoP.11] **Attosecond Fourier Transform Spectroscopy**
— *Kneller, Omer* —

[MoP.12] **Electronic-vibrational dynamics in x-ray transient absorption of N2+ induced by strong-field ionization**
— *Zhao, Jing* —

[MoP.13] **Photoemission Timing of Surface-Oriented Iodoalkanes on Pt(111)**
— *Paul, Sven-Joachim* —

[MoP.14] **Attosecond ionization delay arising from Feshbach-resonance of N2**
— *Wang, W* —

[MoP.15] **Quantifying Centrifugal-Potential Effects: Channel-Resolved Attosecond Time Delays in Nondipole Photoionization**
— *Liao, Yijie* —

[MoP.16] **Phase Measurements Beyond Strong-Field Approximation in Non-Perturbative Single-Color High-Harmonic Spectroscopy**
— *Avnery, Keren* —

[MoP.17] **Analytical Expression for Continuum-continuum Transition Amplitude of Hydrogen-like Atoms**
— *Ji, Jia-Bao* —

[MoP.18] **HHG-based all-attosecond transient absorption spectroscopy**
— *Svirplys, Evaldas* —

[MoP.19] **Hole alignment in noble gas atoms using RTDCIS method**
— *Tahouri, Rezvan* —

[MoP.20] **Modelling sequential ionisation within time-dependent R-Matrix Theory**
— *Lavery, Holly* —

[MoP.21] **SoftMeter: Ultrafast molecular imaging driven by a tunable two-color field**
— *Ghaffari, Fatemehsadat* —

[MoP.22] **Core ionization of positronium chloride: a numerical study**
— *Camper, Antoine* —

[MoP.23] **Attosecond transient absorption spectroscopy of laser-dressed dense helium: Spectral lineshape modification and weak transitions brought to light**
— *He, Yu* —

[MoP.24] **Observation of Bloch Oscillation phenomena via High Harmonic Generation Spectroscopy**
— *Cahana, Noa* —

[MoP.25] **Electron recollisional excitation on dissociative ionization of OCS in phase-locked two-color intense laser fields**
— *Endo, Tomoyuki* —

[MoP.26] **Ro-vibronic coupling in vibrational resolved attosecond photoelectron spectroscopy of acetylene**
— *Li, J* —

[MoP.27] **Attosecond dynamics of direct electrons in optical near-fields**
— *Heimerl, Jonas* —

[MoP.28] **Influence of catastrophes and hidden dynamical symmetries on ultrafast photoelectron physics**
— *Rook, Thomas* —

[MoP.29] **Interference of direct and resonant two-photon ionization in helium**
— *Weissenbilder, Robin* —

[MoP.30] **Estimation of ZnO thickness from high harmonic spectrum using machine learning algorithms**
— *Nagyillés, Balázs* —

[MoP.31] **Anisotropy Induced Electronic Coherence Signal in Attosecond Transient Absorption Spectroscopy**
— *Yuen, CH Isaac* —

[MoP.32] **Probing the attosecond dynamics of high-energy-radiation-induced collisional ionization cascades in condensed-phase samples**
— *Ferté, Anthony* —

[MoP.33] **Modeling angularly resolved phase-jumps in heavy atoms**
— *Mankov, Elisei* —

[MoP.34] **Neural Network Assisted Denoising in Attosecond Transient Spectroscopy Beyond Correlated Source Noise**
— *Hollm, Marko* —

[MoP.35] **Multi-photon and strong-field ionization with a twist**
— *Rajak, Debobrata* —

[MoP.36] **From megabarns to attoseconds: How to relate the cross-section with the time delay**
— *Kheifets, Anatoli* —

[MoP.37] **Quantum trajectory selector: a novel method to clock attosecond dynamics within recollision physics**
— *Raab, Ann-Kathrin* —

[MoP.38] **Orientation-dependent production of electron spirals from multi-orbital heteronuclear diatomic molecules**
— *Ngoko Djiokap, Jean Marcel* —

[MoP.39] **Guiding synthetic chiral light**
— *Loehr, Alexander* —

[MoP.40] **Attosecond transient absorption in the water-window regime driven by self-compressed light-transient pulses**
— *Vismarra, Federico* —

[MoP.41] **Tuning the Fano resonance line shape in hundred zeptosecond timescale by using two attosecond pulses**
— *Jin, Cheng* —

[MoP.42] **Heterodyne analysis of high-order partial waves in attosecond photoionization of Helium**
— *Han, Lulu* —

[MoP.43] **Exciting autoionizing states with multiple photons**
— *Majety, Vinay Pramod* —

[MoP.44] **Imaging ultrafast structural rearrangement in ammonia molecule with high harmonic spectroscopy**
— *He, Lixin* —

[MoP.45] **New 1D atomic model potential for more accurate HHG spectra**
— *Sallai, Krisztina* —

[MoP.46] **Attosecond electron dynamics in two-color multiphoton processes in helium**
— *Benito De Lama, Miguel* —

[MoP.47] **Retrieving the density matrix form experimental KRAKEN data**
— *Evensen, Andreas* —

[MoP.48] **Partial-wave transitions in attosecond time delays**
— *Mašín, Zdeněk* —

[MoP.49] **Wigner vs. Smith: Time delays in anisotropic systems**
— *Saalmann, Ulf* —

[MoP.50] **Extreme-Ultraviolet Optical Response of Atomically Thin MoS2 Crystals**
— *Di Palo, Nicola* —

[MoP.51] **Effect of attosecond electronic coherences on conical-intersection dynamics in core-excited states**
— *Rodriguez Cuenca, Emilio* —

[MoP.52] **Cross-Process Interference in Strong-Field Ionization: Insights from CEP-Dependent Photoelectron Spectra**
— *Herzig, Anne* —

[MoP.53] **All-attosecond transient absorption spectroscopy of gas-phase molecules**
— *Chiang, Kuo-Yang* —

[MoP.54] **Attosecond Raman time-delay in resonant transitions of ions**
— *Zhao, Zengxiu* —

[MoP.55] **Imaging in the water window with coherence tomography using high-harmonic generation**
— *Fuchs, Silvio* —

[MoP.56] **Attosecond Optical Orientation**
— *Drescher, Lauren B* —

[MoP.57] **Simulation of transient-grating high-harmonic spectroscopy from dissiociating molecules**
— *Trieu, Doan-An* —

[MoP.58] **Probing of ultrafast nuclear vibrational and rotational dynamics with X-ray**
— *Liu, Ji-Cai* —

[MoP.59] **Unifying two manifestations of multielectron polarization in high harmonic generation: peak's frequency shift and odd-even intensity modulation**
— *Phan, Ngoc-Loan* —

[MoP.60] **State-resolved femtosecond phase control in dense-gas laser-atom interaction enabled by attosecond XUV interferometry**
— *Hedewig, Lina* —

[MoP.61] **Controlling the Orbital Angular Momentum of Electron in Neutral Atom**
— *Hu, Hongtao* —

[MoP.62] **Chiral Optical Tweezers - efficient enantioseparation of molecules**
— *Jones, Robert Michael* —

[MoP.63] **Time Delays in Photodetachment of Atomic and Molecular Anions at Low Energies**

— *Dvořák, Jan* —

[MoP.64] **Ultrafast atomic physics between the perturbative and strong-field regimes.**
— *Marshallsay, Sean* —

[MoP.65] **Multidimensional spectroscopy protocols with attosecond pulses**
— *Finkelstein-Shapiro, Daniel* —

[MoP.66] **Enhancing Chiral Recognition in Gas-Phase Molecules Using Chiral Topological Light**
— *Haram, Nida* —

[MoP.67] **Chiral topological light for detection of robust enantiosensitive observables**
— *Mayer, Nicola* —

[MoP.68] **Complete retrieval of photoelectron dynamics from partially-coherent measurements in entangled photoemission**
— *Caillat, Jérémie* —

[MoP.69] **Photoelectron momentum distributions using crossed linearly polarized attosecond pulses**
— *Kjærsdam Telléus, Emilia M* —

[MoP.70] **Ultrafast photoionization dynamics in the vicinity of CO autoionzing states**
— *Faria, Felipe* —

[MoP.71] **Time-resolved Auger decay of an O2 intruder-state.**
— *Short, Andrew* —

[MoP.72] **Probing attosecond dynamics of chiral bound wave packets with an atto-clock**
— *Ge, Peipei* —

[MoP.73] **Extended RPAE method solves the Argon delay-puzzle**
— *Lindroth, E* —

[MoP.74] **Signature of nuclear wavepacket interference in high-order harmonic generation during HCN/HNC isomerization process**
— *Le, Van-Hoang* —

[MoP.75] **Quantum path interference in high harmonic generation driven by highly bichromatic fields**
— *Zou, Xiaozhou* —

[MoP.76] **Decoherence phenomena of electron dynamics in liquid water**
— *Crippa, Gabriele* —

[MoP.77] **A general approximator for strong-field ionization rates**
— *Agarwal, Manoram* —

[MoP.78] **State- and Phase-Resolved Vibrational Wavepacket Dynamics in N2 Studied using All-Attosecond Transient Absorption Spectroscopy**
— *Vibudh, Vibudh* —

[MoP.79] **Chiral Measures make Chiral Moments**
— *Khokhlova, Margarita* —

[MoP.80] **Observing the Jahn-Teller dynamics of the propadiene cation using interferometric strong-field ionization**
— *Brupbacher, Katherine* —

[MoP.81] **Photon Momentum Transfer and Partitioning: From One to Many**
— *Ni, Hongcheng* —

[MoP.82] **Subcycle Conservation Law in Strong-Field Ionization**
— *Ni, Hongcheng* —

[MoP.83] **Ultrafast Spin Migration in the Argon Dimer Cation**
— *Carlström, Stefanos* —

[MoP.84] **Attosecond coincidence spectroscopy in small molecules**
— *Makos, Ioannis* —

[MoP.85] **Investigating Photoionization Delays Using an Attosecond Source Synchronized with an Infrared Non-Collinear Optical Parametric Amplifier.**
— *Jahanzeb, Muhammad* —

[MoP.86] **Nonlinear spectroscopic studies using intense Gas High-order Harmonics at the SYLOS COMPACT beam line, EL-ALPS**
— *Mukhopadhyay, Sourin* —

[MoP.87] **Rotational Pseudo-Doppler Effect in Solid-State High Harmonic Generation**
— *Zuo, Zitan* —

[MoP.88] **Quantum beating and cyclic structures in the phase-space dynamics of the Kramers-Henneberger atom**
— *Aynul, Anika* —

[MoP.89] **Transient extreme ultraviolet scatterometry for nanostructure morphology re-construction and probing material dynamics**
— *Kechaoglou, Emmanouil* —

[MoP.90] **Attosecond Ionic Photoionization Spectroscopy**
— *Xu, Yidan* —

[MoP.91] **Interplay of locally chiral rotational and electronic currents in chiral molecules**
— *Mirahmadi, Marjan* —

[MoP.92] **Ultrafast TACOS**
— *Terentjevas, Justas* —

[MoP.93] **Strong-field theory for attosecond scanning tunneling microscopy**
— *Davidovich, Daniel* —

[MoP.94] **Ultrafast and topological chiral sensing with vector beams**
— *Rodriguez, Aude* —

[MoP.95] **Theoretical study of photoemission time delay for two-photon transition via Multiple Scattering approach**
— *Miura, Yugo* —

[MoP.96] **Ultrafast dissociation dynamics of alkyl iodides induced by few-fs UV pulses**

— *Ryabchuk, Sergey* —

[MoP.97] **Reconstruction of the ionization and recombination times from the spectral phase of the emitted HHG radiation.**

— *Laurent, Guillaume* —

[MoP.98] **Spin-Orbit Delays in Argon: Tracking Ionization Delays Across an Autoionizing Rydberg Resonance**

— *Berglitsch, Thomas* —

[MoP.99] **A highly accurate semiclassical method for atoms and molecules in ultrafast intense laser pulses**

— *Le, Anh-Thu* —

Poster session 2

[TuP.1] **Laser-driven high-flux source of coherent quasi-monochromatic XUV radiation for coincidence spectroscopy**
— *Späthe, Julian* —

[TuP.2] **Ionization-induced blue-shift of the driving pulse leads to increased harmonic yield and single attosecond pulse**
— *Schütte, Bernd* —

[TuP.3] **Polarization anisotropy of high harmonic generation in silicon controlled by coherent combination of ω-3ω fields**
— *Gindl, Adam* —

[TuP.4] **Complete polarization measurements on broadband high-harmonic radiation**
— *Gulyás Oldal, Lénárd* —

[TuP.5] **Beamline development optimized for STARPES : a highly tunable extreme ultraviolet lightsource**
— *Nonnon, Pierre* —

[TuP.6] **Enhancement of vacuum-ultraviolet laser generation through near-threshold harmonics driven by a few-cycle two-color laser field**
— *Wang, Bincheng* —

[TuP.7] **High-power and high-energy post-pulse-compression of a thin-disk laser**
— *Gong, Xiaochun* —

[TuP.8] **Generating visible-ultraviolet sub-femtosecond pulses in a single fiber and sampling their fields**
— *Dassie, Marco* —

[TuP.9] **200-kHz laser system delivering few-cycle near-infrared pulses for attosecond strong-field experiments**
— *Meier, Katrin* —

[TuP.10] **Intensity-Control of Few-Cycle Pulses**
— *Broughton, Joseph* —

[TuP.11] **Spectral modulation and double attosecond pulse emission from relativistic laser-plasma interactions**
— *Liu, Aihua* —

[TuP.12] **High-intensity attosecond beamline for XUV pump - XUV probe investigations**
— *Vardast, Sajjad* —

[TuP.13] **High-harmonic spectroscopy probes the directional charge-density-wave-strengths of TiSe2**
— *Tyulnev, Igor* —

[TuP.14] **XUV yield optimization of two-color high-order harmonic generation in gases**
— *Redon, Melvin* —

[TuP.15] **Tracing electronic coherences at conical intersections with heterodyned attosecond four-wave-mixing spectroscopy**
— *Rupprecht, Patrick* —

[TuP.16] **High power sub-20-fs laser based on gas multi-pass cell**
— *Li, Pinbin* —

[TuP.17] **Filamentation-Assisted Isolated Attosecond Pulse Generation**
— *Chien, Yu-En* —

[TuP.18] **Attosecond X-ray Core-level Chronoscopy of Aromatic Molecules**
— *Ji, Jia-Bao* —

[TuP.19] **Double-blind holography of ultrashort free-electron laser pulses**
— *Azzolin, Agata* —

[TuP.20] **Attosecond plasma lens**
— *Svirplys, Evaldas* —

[TuP.21] **Resolving attosecond time-delay in strong-field ultrafast process of valence-shell electrons of argon**
— *Han, Lin* —

[TuP.22] **Exploring the effects of pulse duration on high-order harmonic generation with tunable pulse post-compression**
— *Westerberg, Saga* —

[TuP.23] **Ultrafast bright non-Gaussian state of light**
— *Rasputnyi, Andrei* —

[TuP.24] **Enhancement of extreme ultraviolet pulse generation by orthogonally polarized laser-controlled spontaneous emission of Ar atoms**
— *Wang, Zhi-Bin* —

[TuP.25] **High harmonic generation from a solid-gas dual-target configuration**
— *Xie, Baichuan* —

[TuP.26] **Laser-Assisted X-ray Second Harmonic Generation**
— *Xie, Xinhua* —

[TuP.27] **High repetition rate, high average power XUV sources based on High Harmonic Generation**
— *Manschwetus, Bastian* —

[TuP.28] **Apparatus for attosecond spectroscopy driven by dual-color few-cycle lasers**
— *Zhong, Shiyang* —

[TuP.29] **Stimulated Raman transitions below and above the ionization threshold of neon using a free-electron laser**
— *Carlström, Stefanos* —

[TuP.30] **Attosecond pulses produced by a few-cycle laser**
— *Ouahioune, Nedjma* —

[TuP.31] **High harmonic generation from a high-repetition-rate liquid plasma mirror**
— *Park, Seong Cheol* —

[TuP.32] **Carrier-envelope phase-stabilized sub-10-fs multiple-plate compression for MHz isolated attosecond pulses**
— *Okamoto, Takuya* —

[TuP.33] **Improving absorption measurements in attosecond XFEL experiments using machine learning**
— *Hu, Kaiyu* —

[TuP.34] **Using AttosecondRayTracing to design a beam transport setup for attosecond pulses from surface plasma relativistic HHG**
— *Kalouguine, Andre* —

[TuP.35] **The effect of driving laser properties on the coherent extreme-ultraviolet wave-front from high harmonic generation**
— *Körmöczi, Andor* —

[TuP.36] **Continuous Relativistic High-Harmonic Generation from a Liquid-Leaf Plasma Mirror at kHz Repetition Rate**
— *Haessler, Stefan* —

[TuP.37] **High Harmonic Polarization Shaping Around a Cooper Minimum**
— *Chen, Ming-Chang* —

[TuP.38] **Asymmetric polarization gating for spectral tuning and temporal confinement of high-order harmonics**
— *Vábek, Jan* —

[TuP.39] **Isolation of bright soft x-ray attosecond pulses in a highly ionized medium using two-color synthesized driving laser**
— *Jin, Cheng* —

[TuP.40] **The generation of photoelectron vortices by chirped attosecond pulses**
— *Zhang, Bingshuang* —

[TuP.41] **Compact attosecond XUV source for the investigation of Raman-induced attosecond electronic coherences**
— *Danylo, Rostyslav* —

[TuP.42] **Single-stage multipass cell spectral broadening and compression of 90 fs, 300 μJ laser down to 8.5 fs**
— *Pronin, Oleg* —

[TuP.43] **Few-cycle Isolated Attosecond Pulses Generation with NIR Driving Fields**
— *Wang, Xiaowei* —

[TuP.44] **Few-femtosecond soft X-ray transient absorption spectroscopy with tuneable deep ultraviolet-visible pump pulses**
— *Lee, Jacob* —

[TuP.45] **A high repetition HHG source of ultrafast XUV pulses for the SXP instrument at the European XFEL**
— *Grychtol, Patrik* —

[TuP.46] **Tuneable few-femtosecond far-ultraviolet laser pulses generated by Yb-laser-driven resonant dispersive wave emission**
— *Satpathy, Deepjyoti* —

[TuP.47] **Few-cycle optical vortices for strong field physics**
— *Guer, Matthieu* —

[TuP.48] **Towards a New HHG Driver Laser Platform: Bridging Q-Switched and Mode-Locked Laser Technology via Multi-Pass Cell Post-Compression**
— *Seitz, Marc* —

[TuP.49] **Batch Bayesian optimization of attosecond betatron pulses from laser wakefield acceleration**
— *Maslarova, Dominika* —

[TuP.50] **High repetition rate deep ultraviolet and infrared beamline for time-resolved experiments on the few-femtosecond timescale**
— *Oliveira e Silva, Ana* —

[TuP.51] **Soft X-rays beyond 600 eV from a 45 W average-power, few-cycle, short-wave infrared driver**
— *Walke, Daniel* —

[TuP.52] **Shaping attosecond pulses using the minimum in high-order harmonic generation spectrum of argon atoms**
— *Li, XiaoYong* —

[TuP.53] **Nonlinear spin-orbit coupling of light in harmonic generation at solid surfaces**
— *Nagai, Kohei* —

[TuP.54] **High-power, few-cycle, short-wave infrared source for high-flux, soft X-ray generation at 200 kHz repetition rate**
— *Juliano, Caroline* —

[TuP.55] **Dark Pulse Interferometry for Attosecond Pulse Characterization**
— *Garcia-Hernandez, Mario* —

[TuP.56] **A Beamline with Efficient Isolated Attosecond Pulse Generation and Detection for Time Resolved Spectroscopy Driven by Synthesized Sub-Cycle Pulses.**
— *Kubullek, Maximilian* —

[TuP.57] **Efficient High-order Harmonic Generation in the Overdriven Regime in a Sub-mm Glass Chip**
— *Cannelli, Oliviero* —

[TuP.58] **Hyper Spectral Ultrafast Source - HYPUS**
— *Schmidt, Bruno* —

[TuP.59] **Atto-seconds electron beams for XFEL and MeV-UED**
— *Wang, Xijie* —

[TuP.60] **Bright isolated XUV attopulse generation through high-order frequency mixing and ellipticity gating techniques**
— *Batelić Janković, Mateja* —

[TuP.61] **Towards 300 mJ, 1 kHz, ¡ 1 ps thin-disk regenerative amplifiers**
— *Osolodkov, Mikhail* —

[TuP.62] **An in-line setup for the generation of a cross-polarized double attosecond pulses with controllable delay**

— *Díaz Rivas, Daniel* —

[TuP.63] **Control of attosecond XUV pulses using an intermediate-field spatial filtering**
— *Valentin, Constance* —

[TuP.64] **Attosecond metrology of vacuum-ultraviolet high-order harmonics generated in semiconductors via laser-dressed photoionization of alkali metals**
— *Farkas, Balazs* —

[TuP.65] **XUV-pump IR-probe resonant shake-up ionization of helium**
— *Lewis, Nicholas* —

[TuP.66] **SNAIL: A library for fast SFA calculation for HHG in gas targets**
— *Benitez, Alex* —

[TuP.67] **Xenon photoionization in the vicinity of 4d giant resonance and Cooper minimum using an XUV-NIR pump-probe experiment at FLASH**
— *Litvinyuk, Igor* —

[TuP.68] **First principles simulation of attosecond XUV pump - XUV probe spectra for small organic molecules**
— *Martin, Fernando* —

[TuP.69] **Optical Parametric MPC: A Hybrid Approach for High-Power Few-Cycle Laser Pulse Generation**
— *Rupp, Nikolas* —

[TuP.70] **High-order Harmonic Generation driven by Vector Necklace Beams**
— *Oakley, Séraphine* —

[TuP.71] **Direct Optical Characterisation of a Driving Field with a Time-Dependent Ellipticity for High Harmonic Generation**
— *Miller, Benjamin* —

[TuP.72] **Attosecond pulse trains generation in hollow core waveguides**
— *Vozzi, Caterina* —

[TuP.73] **XUV chromatic aberrations in high order harmonic generation controlled by flat-top spatial shaping of the laser driver**
— *Constant, Eric* —

[TuP.74] **Tailoring XUV Sources for Ultrafast Science: Advances at ELI Beamlines**
— *Hort, Ondřej* —

[TuP.75] **High harmonic generation control by sub-cycle multi-color waveform synthesis of an ultraviolet to near-infrared driver**
— *van der Linden, Roy* —

[TuP.76] **Generation and characterisation of optical attosecond pulses with terawatt peak power**
— *Nordmann, Joleik* —

[TuP.77] **Generation of ultra-broadband millijoule-level radial vector beams**
— *Heynck, Michael* —

[TuP.78] **Commercial table-top beamline for attosecond science**
— *Reiger, Simon* —

[TuP.79] **High-power and ultrashort IR-driver platform for industrial XUV metrology**
— *Guichard, Florent* —

[TuP.80] **Post-compression of an Yb laser in two steps for attosecond transient absorption spectroscopy**
— *Lericheux, Nicolas* —

[TuP.81] **The Effect of Laser Pulse Duration On The Yield Of High Harmonic Generation**
— *Nagyilles, Balazs* —

[TuP.82] **Single-Shot Carrier-Envelope Phase Measurement at 586 kHz Using Optical Fourier-Transform Interferometry**
— *Arnold, Cord* —

[TuP.83] **Over-Barrier Ionization Dynamics Studied by Backpropagation**
— *Ni, Hongcheng* —

[TuP.84] **Optimizing Harmonic Generation Yield and Refocusing conditions.**
— *Poulain, Vénus* —

[TuP.85] **KeV attosecond pulse generation driven by MIR lasers through waveform control and phase matching**
— *Shim, Bonggu* —

[TuP.86] **Spatial features of high-order harmonic generation from liquids**
— *Tao, Wan Chen* —

[TuP.87] **Driving the high harmonic process using a multi-pass cell**
— *Steiner, Benjamin* —

[TuP.88] **Short attosecond pulse trains for the time-slit experiments**
— *Maroju, P K* —

[TuP.89] **Dispersion-scan (re)meets neural networks**
— *Canhota, Miguel* —

[TuP.90] **Angular Asymmetry of Double Ionization of Helium by Ultrashort Extreme Ultraviolet Laser Pulses**
— *Yang, Yugang* —

[TuP.91] **Cumulative effect on extreme ultraviolet high harmonics generated by high repetition rate Yb-based amplifier**
— *Yamada, Hotaka* —

[TuP.92] **Highly Efficient Enantiosensitive Low-Order Harmonic Generation**
— *Heilemann, Rico* —

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Abstracts for Presentations:

Attosecond science: from adolescence to adulthood

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Synopsis Attosecond science is an interdisciplinary research field, encompassing and connecting applications in physics, chemistry, biology and quantum information science. In my tutorial, I will provide an overview of the development of attosecond science, and will show how some of the early demonstrations of the use of attosecond pump-probe spectroscopy as an analytical technique have more recently paved the way towards the use of attosecond science as a tool to investigate fundamental quantum mechanical phenomena.

The birth of attosecond science was recognized by the 2023 Physics Nobel Prize that was awarded to Anne L'Huillier, for her discovery of the process of High-Harmonic Generation (HHG) in the late 1980's and Pierre Agostini and Ferenc Krausz, for their first demonstrations of the generation of attosecond pulses in 2001.

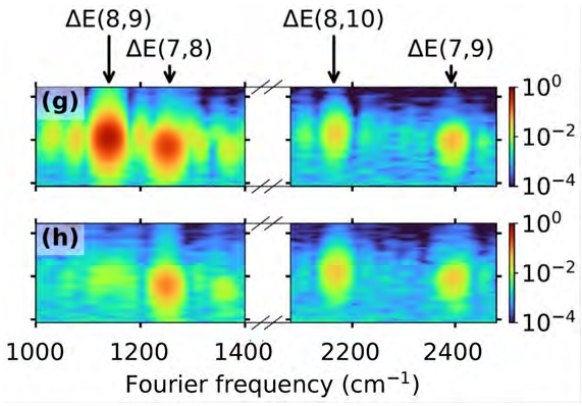


Figure 1. Fourier Transform Power spectra revealing vibrational coherences that occur in a pump-probe experiment on H_2 molecules. The upper and lower data set was recorded using two different delays between the attosecond pulse trains that were used to ionize the molecule. The absence of a number of vibrational coherences in the lower figure is due to ion-photoelectron entanglement.

Presently, a little more than two decades later, attosecond science has developed into an interdisciplinary research field, encompassing and connecting applications in physics, chemistry, biology and quantum information science, with recent applications as diverse as the determination of attosecond delays in X-ray photoionization [1] and the observation of Berry phase phenomena in the condensed phase [2]. In my tutorial, I will provide an overview of the development of attosecond science, and will show how some of the early demonstrations of the use of attosecond pump-probe spectroscopy as an analytical technique [3-5] have more recently paved the way towards the use of attosecond science as a tool to investigate fundamental quantum mechanical phenomena [6-8].

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Time-Dependent Close Coupling on the heels of attosecond electron dynamics

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Synopsis I will present recent results from TDCC simulations with NewStock and from Astra, a new *ab initio* molecular ionization code. Applications include the radiative stabilization of polaritonic resonances, spin-resolved phase jumps in Rainbow RABBITT, attosecond transient absorption in CO and O_2, and XUV-pump IR-probe double ionization of C_2H_4 with coupled electron-nuclear dynamics.

Attosecond science has been instrumental to resolve in time the photoionization and coherent electronic excitation of atoms, molecules, and solids, recently extending its application to increasingly more complex molecules, to monitor coupled electron-nuclear motion during chemical reactions [1]. In this context, *ab initio* time-dependent close-coupling (TDCC), wavefunction-based methods are essential as they can describe processes influenced by multiple excitations as well as entanglement between photofragments. In the first part of the talk, I will illustrate the capabilities of NewStock, an atomic ionization code, in reproducing RABBITT, attosecond transient absorption (ATAS), and non-collinear four-wave mixing atomic spectra. I will show how radiative coupling can stabilize autoionizing states [2] and how Rainbow RABBITT [3] reveals a full 2π phase excursion near the argon $3s^{-1}4p$ state [4], characteristic of complex-q Fano profiles [5].

Despite significant progress [6–11], molecular scaling remains a major challenge when treating ionic correlation, large targets, or fragmentation. In the second part of the talk, I will present recent results obtained with Astra [12] (AttoSecond TRAnsitions), an *ab initio* CC molecular ionization code based on high-order transition density matrices between correlated ionic states of arbitrary multiplicity [13], and hybrid Gaussian-B-spline integrals [11, 14]. Astra accurately reproduces benchmark photoionization cross sections and photoelectron angular distributions for molecules such as N_2, CO, H_2CO, and Pyrazine. I will highlight recent studies of ATAS in CO and O_2, charge migration, as well as the sequential XUV-pump IR-probe double ionization of C_2H_4. For the latter, we combined Astra with surface-hopping methods [15] to account for nuclear dynamics and reproduce multiphoton observables.

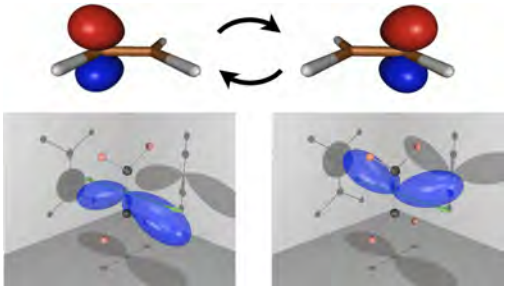


Figure 1. Time-dependent molecular-frame photoelectron angular distributions from a coherently excited ethylene molecule, computed with Astra.

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Laser-assisted dynamical interference of photoelectrons driven by chirped extreme-ultraviolet pulses

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Synopsis By combining chirped femtosecond XUV pulses with moderate-intensity IR fields, we experimentally demonstrate chirped-Laser-Assisted Dynamical Interference (cLADI), a new regime of light-matter interaction where multi-pathway interference of photoionized electrons modifies the photoemission process.

Quantum interference in photoionization is a powerful probe of fundamental light-matter interaction with significant implications for ultrafast spectroscopy and quantum control. Here, we introduce and experimentally demonstrate chirped-Laser-Assisted Dynamical Interference (cLADI). The LADI mechanism arises from the interference of electron pathways, where photoelectrons, generated by an extreme ultraviolet (XUV) pulse, experience a kinetic energy (KE) modulation due to the ponderomotive shift (U_p) induced by an additional dressing infrared (IR) field [1]. Despite its generality, high-KE resolution requirements and competing multiphoton processes (MP) have hindered its unambiguous observation [2]. By introducing a quadratic spectral phase to the XUV, our cLADI scheme relaxes these constraints, enabling verification at moderate IR intensities (about 2 TW/cm^2). The chirped XUV pulse expands the range of interference pathways, which emerge when the time-varying ponderomotive energy intersects the instantaneous XUV frequency, allowing tunability via the chirp magnitude and IR intensity, Fig. 1a.

Experimentally, we validated cLADI at the ELI ALPS facility by generating chirped XUV pulses centered at 42 eV with a 33-fs duration and overlapping them with a 15-fs IR field on an atomic target. As shown in Fig. 1b, the measured photoelectron spectra exhibit a pronounced splitting of the main peak at moderate IR intensities, in excellent agreement with simulations incorporating the measured pulses. Crucially, crossed polarizations (orange) of the XUV and IR suppress the MP interference channels observed with linear polarization (blue), ensuring that the observed pattern predominantly arises from cLADI rather than from sideband formation (SB).

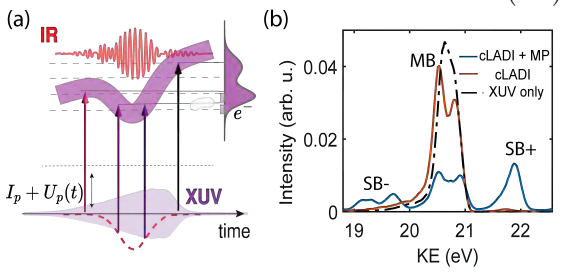


Figure 1. **(a)** Sketch of cLADI, I_p is the ionization potential. **(b)** Experimental photoelectron distribution showing a splitting arising from cLADI.

By establishing cLADI as a novel interference regime, we enable its use in ultrafast spectroscopy, free from spectral overlap and multiphoton constraints, paving the way for high-precision control of free-electron wavepacket and enhanced probing of ultrafast processes in complex systems.

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Attosecond spectroscopy reveals electron correlation in the photoionization of Argon

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Synopsis Attosecond spectroscopy with high-energy resolution presents great power on tracing electron dynamics in matters. Here, the electron correlation in Argon has been studied by using RABBIT spectroscopy, which confirms the correlation advances the emission of 3s electron up to ~ -250 as at its minima, and unveils the electron dynamics in multielectron atoms.

Electron dynamics in matter govern its physical and chemical properties, making it a central focus in the development of experimental techniques to track its motion. Attosecond photoelectron spectroscopy has emerged as a powerful tool for investigating attosecond electron correlation dynamics in atoms and tracing electron-nuclear coupling in molecules.

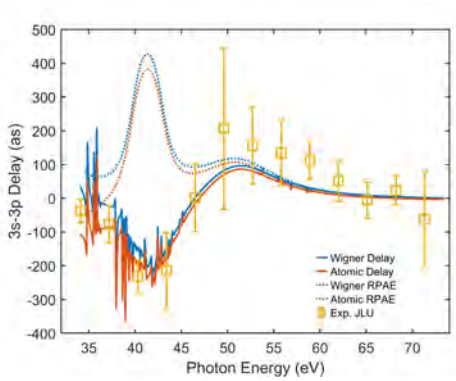


Figure 1. Measured and calculated relative ionization delay between 3s and 3p electrons of Argon.

In this work, we will present our recent work on the ionization dynamics of argon atoms, offering insights into their unique electron behavior. Using high-energy resolution ($\sim$ 100 meV) RABBIT spectroscopy, we measured the ionization time delay between the 3s and 3p valence shells across the Cooper minimum of argon [1, 2, 3]. The ionization delay of the 3s cooper minimum has been debated for years, and we finally came to an agreement between theory and experiment. Our results reveal that, in the case of argon, electron correlation accelerates the emission of the 3s electron by up to ~ -250 as at its minima cross-section. This suggests that an electron wavepacket propagating through half of argon's Coulomb potential exits much earlier than if it had traveled the same distance in a vacuum.

With high-energy resolution measurements and newly developed many-body perturbation theory calculations, we demonstrated that the ionization delay of the 3s Cooper minimum is negative, as shown in Fig. 1, resolving discrepancies between theoretical models. We identify key correlations, mediated by shake-up configurations of the type $3p^{-2}n/\epsilon' d;\epsilon'' p$, that are essential for understanding the 3s ionization delay in argon. Our findings provide deeper insight into correlation dynamics in multielectron systems and have broader implications for testing fundamental theories of ns subshells in closed-shell atoms and molecules.

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Chiral molecules in strong laser fields

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Synopsis When chiral molecules are photoionized by a circular, elliptical or polarization-shaped strong laser field, the combined action of the chiral potential and sub-cycle rotation of the field results in forward-backward asymmetries in the photoelectron angular distributions along the laser propagation direction. By shaping appropriately the ionizing field, it is possible to reveal the influence of chirality on the various steps of the ionization process, from tunneling to rescattering.

Circularly polarized light has been used since the 19th century to distinguish the two mirror images of a chiral molecule, through various forms of circular dichroisms (CD) in photoabsorption. In 2001, a new type of CD was experimentally observed, in photoionization: PhotoElectron Circular Dichroism (PECD) [1]. This effect, observed in the photoelectron angular distribution of chiral molecules ionized by circularly polarized light, consists of a forward/backward asymmetry along the light propagation direction. Unlike other CDs which rely on magnetic dipole or electric quadrupole interactions, PECD results from pure electric-dipole effects, which makes it intense, in the 10% range. In the past two decades, PECD has been shown to be a universal effect in photoionization – in single, multiphoton and strong field ionization – and to be a remarkably sensitive probe of structure and dynamics of chiral molecules (for a review see [2]). In this talk we will focus on the strong-field ionization regime and see how manipulating the laser polarization state enables probing different aspects of the chiral photoionization dynamics.

The main steps of strong-field ionization are schematized in Fig. 1. First, electrons escape the molecular potential by tunneling through the barrier lowered by the laser field. Then they are accelerated by the field, gaining kinetic energy. If the field is not circularly polarized, some electrons may rescatter onto the ionic potential and interfere with the direct electrons, while some may experience an elastic collision. All of these processes have been investigated in-depth in strong-field ionization through various techniques such as the attoclock [3], attosecond delay measurements [4], photoelectron interferomety [5], or laser-induced electron diffraction (LIED) [6]. We will see that these techniques can be transposed to chiral photoionization, to reveal the enantiosensitivity of tunneling [7, 8] and electron emission delays [9], or to resolve chiral structures by LIED [10]. By adding a perturbative field to the strong laser field, as recently demonstrated in atoms [11], it becomes possible to resolve and control the electron dynamics in chiral LIED. Last, we will show that chirality can be used to reveal non-dipole effects in the strong-field ionization process.

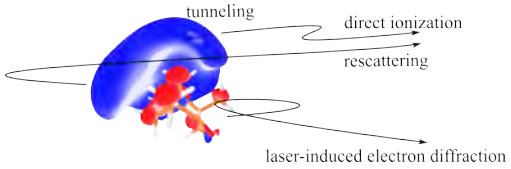


Figure 1. Schematic view of electron trajectories in strong-field ionization.

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Vectorial attosecond transient spectroscopy

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Synopsis We introduce vectorial attosecond transient spectroscopy to probe ultrafast polarization dynamics in light-driven systems. By integrating vectorial measurement and control, we resolve the transient polarization state of transmitted attosecond pulses. This new scheme provides heterodyne amplification of the weak vectorial signal, offering a powerful tool to investigate transient symmetry breaking in quantum systems with sub-cycle resolution.

Vectorial light-matter interactions reveal the fundamental symmetries of quantum systems, imprinting structural properties into the polarization state of an interacting field. In the nonlinear regime, these symmetries can undergo dramatic modifications due to interaction with a strong driving-field. Applying attosecond metrology, dynamical symmetry breaking can be both induced and resolved within the optical cycle. Attosecond transient absorption is one of the most successful experimental techniques in attosecond science, maping the dynamics of a laser-driven system onto the absorption spectrum of attosecond pulses [1]. However, the underlying quantum dynamics are encoded in the amplitude, phase, and polarization state of the transmitted attosecond pulses. Recently, attosecond transient interferometry was introduced, resolving the dynamical phase [2,3]. So far, these schemes remain scalar, while the sub-cycle vectorial properties of the transient interaction are yet to be revealed.

Here, we establish vectorial attosecond transient spectroscopy, measuring ultrafast nonlinear polarization rotation of attosecond pulses transmitted through field-driven helium. In this work, we integrate a vectorial driving field with XUV-XUV interferometry. We achieve two-dimensional control by introducing a driving-field composed of a strong IR and its orthogonaly polarized second-harmonic. This field induces dynamical symmetry breaking in the target [4], modifying both the spectrum and polarization state of the transmitted attosecond pulses. Interfering these pulses with an appropriate vectorial XUV reference enables heterodyne detection of the weak vectorial signal, while resolving the transient amplitude and phase encoded in two orthogonal polarization components. By measuring the full complex response, we reconstruct the transient polarization of the transmitted attosecond pulses, visualizing dynamical symmetry breaking on a sub-cycle timescale. Vectorial attosecond transient spectroscopy offers a powerful method for probing the field-driven two-dimensional dynamics of quantum wavefunctions in complex systems, from chiral molecules [5] to topological materials [6].

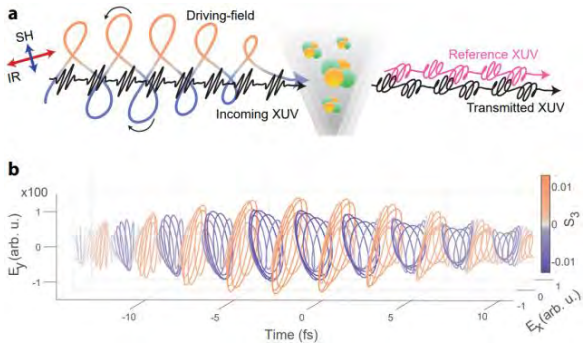


Figure 1. (**a**) Schematic description of vectorial attosecond transient spectroscopy. (**b**) Reconstruction of ultrafast polarization dynamics in field-driven helium. The transmitted polarization is elliptical, dynamically switching its handedness. The colorbar represents the Stokes parameter S_3, with measured ellipticity values on the order of 1%.

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Attosecond XUV pump-control spectroscopy for tracking and modification of excited neutral molecules

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Synopsis We demonstrate the modification of wave-packet evolution in the neutral H_2 molecule. The vibronically (electronically and vibrationally) excited states promoted coherently by an attosecond XUV laser pulse were probed and spectrally resolved while the molecules were interacting with an intense near-infrared laser field. The vibrational wave-packet revival in the $D\,^1\Pi_u$ excited state exhibits NIR-intensity speed-up to earlier times, confirmed by a multi-level TDSE simulation. The observed revival time-shift is attributed to state-specific NIR-induced phase shifts.

In a newly introduced pump-control scheme for attosecond XUV spectroscopy, Fig. 1(a), we focus on the time-domain interpretation of absorption spectra in correlated molecular systems [1]. Our novel experimental approach for tracking excited wave packets in neutral molecules allows access to the fastest nuclear motion, down to the 10-fs-timescale in the lightest of all molecules, H_2. Here, the molecular ground state reveals the system's dynamics through the molecular dipole emission, thus acting as a self-probe for the intact neutral molecule. Experimentally measured absorption spectra with high spectral resolution (4 meV at 15 eV, Fig. 1(b)) allow for the reconstruction of this time-dependent dipole response through a Fourier-based technique, Fig. 1(c), previously developed for isolated atomic states [2]. The accessible ultrafast dynamics of the $D^1\Pi_u$ wave packet in H_2 reaches up to the first vibrational revival at ~ 270 fs. An experimental study of the intensity dependence of the revival and a supporting multi-level simulation solving the time-dependent Schrödinger equation show: the higher the NIR intensity, the earlier the revival time, Fig. 1(c). Furthermore, we extract the state-specific NIR-induced phase-shifts, which are a key contributor to the observed time shifts.

The presented pump-control approach complements charge-particle-based pump-control-probe experimental schemes, typically targeting the ultrafast dynamics in molecular ions, and opens a straightforward route to control the molecular dynamics in neutral systems.

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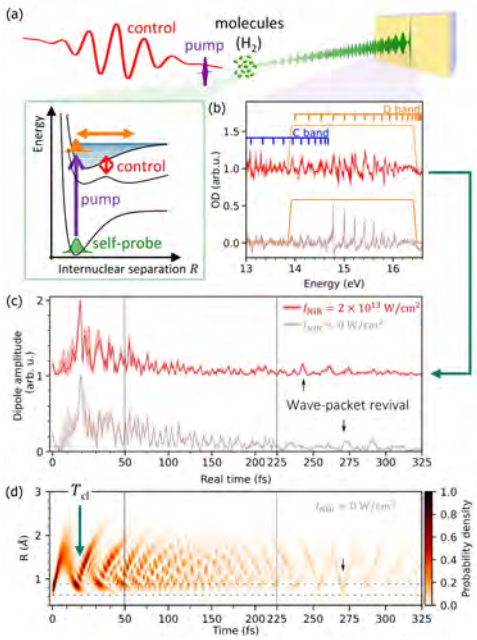


Figure 1. (a) Experimental scheme for attosecond XUV pump-control spectroscopy. (b) Detected optical density (OD) spectra showing vibronic excitations in neutral H_2. (c) Reconstructed time-dependent dipole amplitude of the $D^1\Pi_u$ vibrational wave packet in an XUV-only configuration (lower curve) and for an NIR control field with $I_{NIR} \approx 2\times10^{13}$ W/cm^2 (upper curve), respectively. (d) Calculated wave-packet evolution.

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Clocking the tunneling time in strong-field ionization

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Synopsis We will show that a photoelectron wavepacket created through tunneling carries information on its own tunneling time, making it an unambiguously measurable quantity in the context of highly non-linear strong-field physics.

When a quantum particle undergoes tunneling, the time spent under the barrier, referred to as the tunneling time, is subject to several debated questions regarding both its definition and its measurability [1, 2]. In particular, the measurement of the tunneling times in the context of strong-field ionization [3] is still controversial [4] and the latest conclusions remain in conflict with the results from so-called Larmor clock experiments [5]. Here, we will show that a photoelectron wavepacket created through tunneling carries information on its own tunneling time, making it an unambiguously measurable quantity in the context of highly non-linear strong-field physics.

For this, we identify a dynamical process, the "transversal squeezing", according to which the longer the time a 3D wavepacket spends "under" a barrier, the narrower its transverse momentum distribution at the tunnel exit. Our approach has been rigorously established analytically within an SFA-like framework and constitutes an alternative theory for the interpretation of "attoclock" experiments [3]. Its practical implementation also uses a circularly polarized laser which, at the atomic scale, creates a tunneling barrier that both oscillates and rotates within the pulse. Therefore, a given photoemission direction is associated with a single barrier configuration. By fully exploiting this mapping and the transversal squeezing, we can thus attribute a given tunneling time to each direction in the photoelectron moment distribution (PMD), in a more comprehensive way than the original attoclock.

Using numerical ab initio simulations on H initially in its 1s ground state, we show that a proper analysis of the PMD gives access to orientation-dependent tunneling times lying in the ~ 100 as range, in excellent agreement with the SFA-based predictions. Furthermore, an extension to initial states carrying ring currents ($m \neq 0$) demonstrates that electrons counter-rotating with the laser undergo shorter tunneling times than co-rotating ones [6] and provides an additional improvement of the original attoclock.

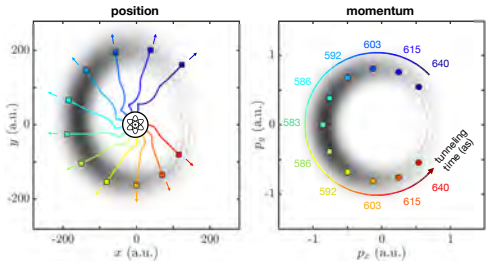


Figure 1. Amplitude probability (left panel) at the end of the laser pulse and trajectories (colored lines) guiding wavepackets ionized at different times. Photoelectron momentum distribution (right panel), asymptotic momentum (colored dots) associated with each emission angle and tunneling time.

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Quantum-orbit dynamics in strongly-polychromatic laser fields

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Synopsis Strong-field driven quantum systems can typically be described using highly-oscillatory Feynman path integrals, which reduce to a discrete sum of contributions from individual 'quantum orbits'. I will present a broad overview of the advantages provided by quantum-orbit methods, including the rich phenomena caused by the imaginary parts – not just of the ionization times, but of the trajectories themselves, the applications and challenges in strongly polychromatic fields, and a robust and universal approach for identifying and calculating these quantum orbits in arbitrary temporal waveforms.

When quantum matter is subjected to a strong-field laser driver, it will generally produce an ultrafast response in the attosecond regime, from high-harmonic generation to above-threshold ionization and everything in between. These responses are governed by a complex wave equation, the Schrödinger equation, but they are often well modelled by relatively simple classical dynamics. In between those two extremes, and linking them together, sits a quantitative model known as the 'quantum orbit' approach [1]: using complex-valued trajectories derived from Feynman-path-integral oscillatory integrals, and providing both intuitive handles to understand the dynamics as well as quantitative predictions that are a good match to experimental outcomes.

These trajectories often involve complex-valued times, and, sometimes, positions; complex-valued times are at the core of essential timing debates in attoscience [2], while complex-valued positions often provide substantial clarity in understanding experimental observables [3, 4]. Indeed, leaning into the complex-valuedness theme often allows one to simplify, in one fell swoop, significant chunks of the theory [5].

One emerging challenge for this formalism is the increasing availability of strongly-polychromatic light sources: that is, laser systems that provide, in addition to a non-perturbative fundamental oscillation, high intensities of its harmonics, at comparable or even higher intensity [6]. Strongly-polychromatic sources offer the prospect of substantial control over sub-cycle strong-field dynamics, but they are hampered by the complexity of the response, and by our ability to understand and predict its features. This is normally the area where quantum-orbit methods are most helpful, but in complex waveforms they become challenging, as it becomes (at best) labour-intensive to determine which trajectories contribute to the response and which ones are excluded.

I will present recent breakthroughs in the analytical theory of quantum orbits, which allow us to identify the relevant quantum orbits in arbitrarily complex tailored driver waveforms [7]. Moreover, our theory provides an alternative, robust, and efficient method to calculate oscillatory integrals, based on the mathematical framework of Picard-Lefschetz theory.

As an example of the key questions that need to be tackled going forward, I will focus on the 'colour switchover': what happens to strong-field phenomena if a strong laser driver is steadily replaced by its second harmonic? The dynamics is rich and complex, including caustics, saddle-point coalescences, branch cuts, and intricate topologies; perhaps most intriguingly, it gives rise to a quantum orbit that encodes optical tunnelling at a time when the electric field vanishes – quantum tunnelling without a barrier [8].

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Ion-photoelectron entanglement and coherence controlled by two-color laser pulses

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Synopsis We report a theoretical investigation of photoionization by a pair of coherent, ultrashort, fundamental and second-harmonic extreme-ultraviolet pulses, where the photon energies are selected to yield the same photoelectron energy for ionization of Ne $2p$ and $2s$ subshells. By the essential-states approach as well as multiconfiguration time-dependent Hartree-Fock calculations, we show that the Rabi coupling converts ion-photoelectron entanglement to coherent superposition. This creates coherence between the two photoelectron wave packets, which would otherwise be incoherent.

With longitudinally coherent pulses of two different wavelengths, e.g., from FERMI [1, 2], one can coherently control the outcome of experiments by varying their relative phase [3, 4]. Consider a Ne atom under collinear z-polarized bichromatic light ($\hbar\omega$ and $2\hbar\omega$). Ionization thresholds for Ne $2p$ and $2s$ are 21.56 and 48.47 eV, respectively. If $\hbar\omega$ matches their difference (26.91 eV), $2\hbar\omega$ ionizes $2s$ and $\hbar\omega$ ionizes $2p$, yielding photoelectrons with 5.35 eV energy and opposite parities. The two photoelectron wave packets do not interfere, since they leave different ionic states ($2s^{-1}$ and $2p^{-1}$); in other words, the photoelectron and ionic subsystems are entangled. Since $\hbar\omega$ equals the energy difference between $2s^{-1}$ and $2p^{-1}$, the fundamental pulse couples the two ionic states, causing Rabi oscillations. If $2p^{-1}$ transitions to $2s^{-1}$ or vice versa, the two photoelectron states can now interfere.

In this work, using the essential-states approach and multiconfiguration time-dependent Hartree-Fock calculations, we show that the Rabi coupling indeed converts entanglement to coherent superposition. This creates coherence between the two, otherwise incoherent, photoelectron wave packets, leading to, e.g., coherent control of the photoelectron angular distribution through the ω-2ω relative phase (Fig. 1) [5]. We analyze how the asymmetry of the photoelectron angular distribution is affected by the parameters such as relative phase, pulse width and intensity, delay between the two pulses, Rabi coupling strength, and photoelectron energy.

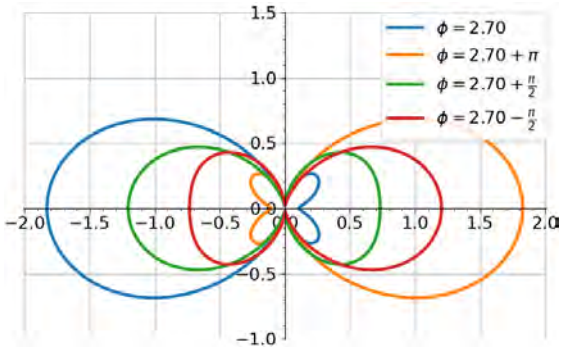


Figure 1. Photoelectron angular distribution at the spectral center vs. the ω-2ω relative phase ϕ.

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Measurement of photoelectron decoherence and attosecond dynamics of unobserved ions

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Synopsis Xenon atoms were photoionized with high-order harmonics in order to create an entangled photoelectron+photoion system. We measured the partially coherent quantum state of the *photoelectron* wavepacket using the Mixed-FROG scheme, from which we reconstructed the attosecond dynamics of the unobserved but entangled *ion*.

Laser-dressed photoelectron spectroscopy has allowed the measurement of the quantum state of photoelectrons. In ideally coherent situations, this state can be fully described using a wave function which can be retrieved with well-established techniques such as RABBIT and attosecond streaking. However in a general situation, the photoelectron can be entangled with unresolved degrees of freedom and exhibit a partially coherent behavior. Its state should then be described using a density matrix [1], which takes into account the loss of information when performing incomplete measurements. New experimental schemes have recently been developed to access such photoelectron density matrix [2, 3].

Using the Mixed-FROG technique, we measured the reduced density matrix of photoelectrons resulting from the ionization of xenon atoms by high harmonics, see Fig. 1. This density matrix $\rho_{\mathrm{PE}}(t \to +\infty)$ represents the asymptotic quantum state of the photoelectron from which we reconstructed its transient evolution $\rho_{\mathrm{PE}}(t)$ through time-frequency analysis [4, 5]. In our experimental conditions, the electron can reach the continuum through two ionization channels, leaving the Xe^+ ion in a superposition of $5p_{3/2}$ and $5p_{1/2}$ states. These two quantum paths are fully distinguishable in the photoelectron spectrum, thus forming a maximally entangled ion+electron system. We show that this entanglement can become a vector of information about the unobserved ion. Indeed enough information is preserved in the electron density matrix $\rho_{\mathrm{PE}}(t)$ to access the transient ion density matrix $\rho_{\mathrm{ion}}(t)$ up to an absolute phase. This experimental reconstruction reveals that even though no asymptotic coherence is left in the ion, some transient ionic coherence can exist during the ionization process. In this work we therefore demonstrate that entanglement can be used to access experimentally the attosecond dynamics of unobserved quantum subsystems.

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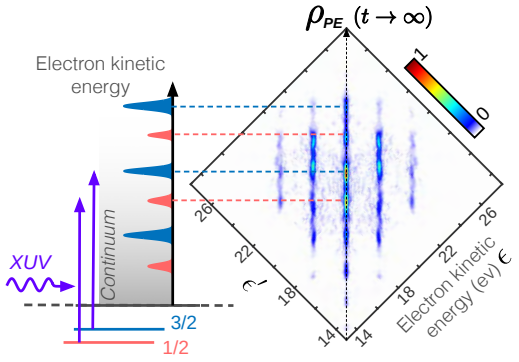


Figure 1. Experimental density matrix (only modulus shown) of photoelectrons obtained by the ionization of Xe through the $5p_{3/2}$ and $5p_{1/2}$ channels (left: photoionization scheme)

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Testing quantum mechanics with photoelectron interferometry

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Synopsis Born's rule is a postulate of quantum mechanics that gives the probability that a measurement on a quantum system will give a certain result. When applied to multi-slit experiments, Born's rule implies that only pair-wise interference can be observed. Here we test for the first time this postulate using photoelectron interferometry based on ionization of argon atoms using an ultrashort extreme ultraviolet pulse and a tunable trichromatic IR pulse.

Quantum mechanics is used to describe the world at the microscopic scale. This theory relies on a set of postulates that lay the foundations for all its predictions. One of these postulates is Born's rule, which defines the probability to measure a quantum system in a given quantum state. This postulate has profound implications for the observation of quantum interference since it implies that in the presence of multiple quantum paths interfering, the total interference pattern is described by pairwise interference terms. In other words, quantum mechanics does not allow for interference between more than two paths. The strength of third order interference can be experimentally measured using the Sorkin test [1]. Sorkin's test was first implemented by Sinha et al. using photons [2], confirming Born's rule within error bars. Following [3], we here report on the first implementation of Sorkin's test using photoelectrons.

Our measurements are based on the ionization of argon atoms in the presence of ultrashort extreme ultraviolet (XUV) light pulses and a tunable trichromatic infrared (IR) pulse. The XUV pulse generates an electron wave packet in the continuum followed by further absorption of a photon from the trichromatic IR pulse, resulting in three interference paths from the ground state to the final state as shown in Fig. 1a). The three IR components act as spectral slits that can be open or closed by blocking or letting through the different spectral components. By recording photoelectron spectra for different path configurations the strength of third order interference can quantified [1, 3]. Examples are presented in Fig. 1b), together with the corresponding IR spectra shown in Fig. 1c). Our results indicate that a nonlinear evolution of the photoelectron in the IR field can significantly affect the outcome of the Sorkin test.

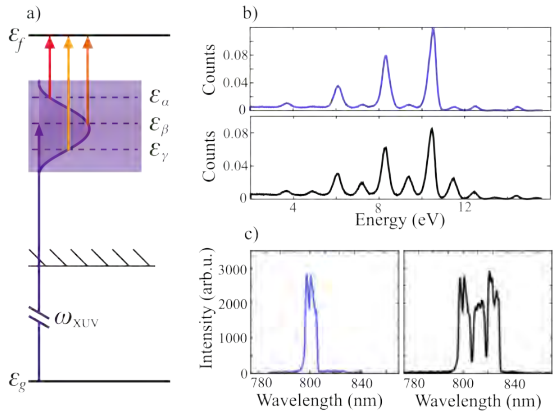


Figure 1. a) Energy diagram for the three-path interferometer. b) Photoelectron spectra from two different IR spectra. c) IR spectra corresponding to the two different photoelectron spectra.

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Strong-field dynamics of electrons photoemitted with non-classical light

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Synopsis We present the measurement of strong-field energy spectra of electrons photoemitted from a nanometric needle tip driven with bright squeezed vacuum. A broad electron energy spectrum and the generation of high-energetic electrons result from driving electrons in this strong non-classical light field. We discuss how the formation of these spectra can be understood in terms of an incoherent sum of coherent-state electron energy spectra for different intensities, where the relative contribution of each coherent-state spectrum is determined by the Husimi distribution of the driving light.

The investigation of electron dynamics at the surface of metal needle tips in the strong optical field of ultrashort laser pulses allows us to understand light-matter interaction processes on the attosecond timescale [1]. Since reaching the strong-field regime of light-matter interaction usually requires high intensities, the driving light fields were commonly described as classical electromagnetic waves in the past. However, in recent years, theoretical studies have investigated the influence of the quantum-optical state of the driving light on strong-field processes such as high harmonic generation [2]. The development of intense sources of non-classical light, such as bright squeezed vacuum (BSV), has made it possible to study these effects also experimentally [3]. For the case of the photoemission of electrons from a nanometric needle tip using BSV, it has been shown that the emitted electrons inherit the number statistics of the driving state of light [4]. However, strong-field processes such as rescattering driven by BSV have not yet been measured in this case. Here, we present the first measurements of strong-field electron energy spectra using non-classical light. We focus single-mode BSV light with a central wavelength of 1600 nm onto a metal needle tip to trigger non-linear photoemission and measure the final energy of the electrons by a home-built spectrometer. We observe energy spectra as broad as 60 eV for BSV compared to 20 eV for coherent light with similar mean intensities (Fig. 1). We explain the broad spectra by the incoherent sum of classical spectra and the broad Husimi distribution of BSV. Our experiments represent a step towards understanding how strong non-classical light interacts with matter on sub-cycle timescales [5].

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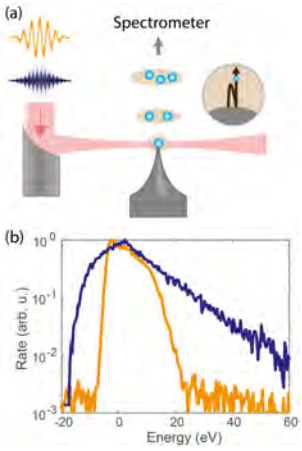


Figure 1. (a) Pulses of classical coherent (orange) or BSV (blue) light are focused on a metal needle tip. The emitted electrons are driven in the light field and their final energy is measured. (b) Measured electron energy spectra for coherent light (orange, pulse energy ≈ 31 nJ) and BSV (blue, mean pulse energy ≈ 14 nJ).

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Giant counter-rotating oscillations and quantum entanglement from time-dependent strong coupling in the extreme ultraviolet

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Synopsis Five key questions about time-dependent strong coupling on ultrafast time scales are addressed: A novel type of ultrastrong coupling is proposed, a new regime of two-photon Rabi oscillations is predicted, and time-symmetry of laser envelopes is proposed to control ultrafast entanglement, light absorption, and entanglement transfer via fluorescence. These ideas may inspire future works that utilize seeded XUV FELs.

Recent advances in free-electron laser (FEL) technology have intensified interest in ultrafast light-matter interactions in the extreme ultraviolet (XUV) regime. Our group has been at the forefront of theoretical developments in this field and working closely with experimentalists to predict nonlinear quantum interference effects in photoionization [1] and ultrafast generation of quantum entanglement [2] in strongly coupled atoms and ions, respectively. While previous studies have focused on traditional strong-coupling processes, enforcing energy conservation via the rotating-wave approximation (RWA) with laser pulses of traditional forms (such as flattops and Gaussians), we now explore novel regimes that challenge these traditional approximations.

Five key questions in strong coupling:

1. Can intense XUV light drive non-traditional processes beyond the RWA, while preserving strong coupling?
2. Can same-parity states in atoms be strongly coupled by intense XUV light?
3. How does the shape of the pulse envelope alter the generation of entanglement?
4. How does the envelope affect the associated fluorescence and absorption processes?
5. What is the ultimate fate of quantum entanglement produced between electrons and ions in photoionization?

We demonstrate that a novel *ultrastrong coupling* regime emerges, where atomic states hybridize across excitation numbers. This leads to giant counter-rotating oscillations, on the *attosecond timescale*, in the excited-state population [3] — an effect strikingly distinct from conventional two-level dynamics [1]. Furthermore, we identify the $1s^2$–1s3d transition in helium as a promising, and previously unexplored, candidate for *two-photon Rabi oscillations*, marking the first prediction of such dynamics driven by intense XUV light [3].

Quantum entanglement is a fascinating concept in physics, which was recently demonstrated to emerge as a result of strong coupling between electrons and ions [2]. Here, we show how *time-symmetry considerations*, more precisely odd envelope functions, can be used to control the entanglement process in time, within the strong-coupling regime [4]. Interestingly, electrons associated with different final ion states now "avoid each other", opening up for *coincidence detection of entanglement* from strong coupling between free electrons and ions [4]. Going further, the shape of the envelope influences absorption of laser light [5] and how fluorescence leads to an *entanglement transfer* from a photoelectron–ion pair to a photoelectron–photon pair via a transient state of *three entangled particles* [6].

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Experimental evidence of the quantum-optical nature of high-harmonic generation in semiconductors

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Synopsis One critical showstopper for realizing the fantastic prospects promised by quantum technology is upscaling entangled quantum states of light. I will present how attosecond laser-driven electron recollision in the process of high harmonic generation in semiconductors can produce multipartite entangled states of light.

One critical showstopper for realizing the fantastic prospects promised by quantum technology is upscaling entangled quantum states of light. However, the scaling problem arises from the intrinsic probabilistic nature of quantum mechanics and the usual strategy to manipulate arduously isolated quantum systems with weakly interacting optical and electric fields. This is far from easy, and after decades of research and tremendous efforts, the generation of entangled states of light is limited to a few modes and relatively small photon numbers. High harmonic generation (HHG) has the potential to overcome the bottleneck of generating many modes, high dimension quantum states of light [1]. Instead of freezing temperature, our strategy is based on freezing the system in time using ultrafast strong optical fields. Indeed, high harmonics of the driving light are emitted as attosecond burst of light, well before main decoherence channels of the system occur.

I will present how attosecond laser-driven electron recollision in the process of high harmonic generation in semiconductors can produce multipartite entangled states of light. Recently, independent theoretical investigations predicted non-classical effects in this strong field laser driven process [1-3]. After few years of debate, the first experimental demonstration confirmed the presence of squeezing and entanglement in HHG generated on various semiconductor samples (Si, ZnO, GaAs, CdTe) [4,5]. Thorough photon statistics analysis shows that harmonics are emitted as near single-mode displaced squeezed state. Further, the high harmonic generation process can be finely controlled in spatial, spectral phase with high and complex dimensionality such as topology from beams that carry orbital angular momentum. This semiconductor-based quantum light source is a highly promising candidate for applications in the broad field of quantum information science and engineering, gaining possible quantum advantage. As an example, I will report on perspectives in condensed matter systems such as spin sensitivity of the quantum light states in cadmium telluride.

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Photon bunching in high-harmonic emission controlled by quantum light.

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Synopsis We perturb the emission of high-order harmonics from a semiconductor with a quantum-optical state, bright squeezed vacuum, to generate sidebands of the high harmonics that exhibit super-Poissonian (i.e. *bunched*) photon statistics. Our results suggest that perturbing strong-field dynamics with quantum-optical states is a viable way to coherently control the generation of these states at short wavelengths, such as extreme ultraviolet or soft X-rays. Quantum correlations will be instrumental to advance attosecond spectroscopy and imaging beyond the classical limits.

Recent works[1,2] have laid the foundations of "attosecond quantum optics", e.g. the understanding of how quantum-optical (QO) properties affect high-field photonics, such as strong-field ionization and acceleration of electrons in QO fields, and how entanglement between the field modes arises during the interaction[3]. Understanding QO properties of attosecond interactions is well justified for it may enable the creation of QO states at short wavelengths, such as deep and extreme ultraviolet and X-rays, which could enable more precise sensors in a spectral region where detector noise is negligible, as well as quantum-enhanced spectroscopy and imaging with chemical or elemental sensitivity. Yet, experimental investigations of "attosecond quantum optics" are still just beginning.

Here we investigate, experimentally, how strongly driven electron-hole pairs responsible for high-harmonic emission from a semiconductor (ZnO) react to a QO perturbation, a Bright Squeezed Vacuum (BSV) beam. The BSV beam creates sidebands of the unperturbed odd-harmonic spectrum that exhibit shot-to-shot power fluctuations with super-Poissonian statistics (Fig. 1), indicative of *photon bunching* and of the generation of a non-coherent state at the short high-harmonic wavelength. In addition, the bandwidth of the sidebands suggests the creation of an attosecond-lived gate for the BSV field. Moreover, the mere emission of sidebands elicits questions about the nature of the BSV field, whose electric field polarity is not defined at any given time, yet the generation of sidebands at nearly the frequency of the even harmonics suggests a well defined time translation symmetry. Our theoretical model based on a photon mixing picture reveals that the quadrature variance of the sidebands can be coherently controlled with the relative phase between the BSV and mid-infrared fields. Finally, we also demonstrate the generation of high harmonics of BSV alone.

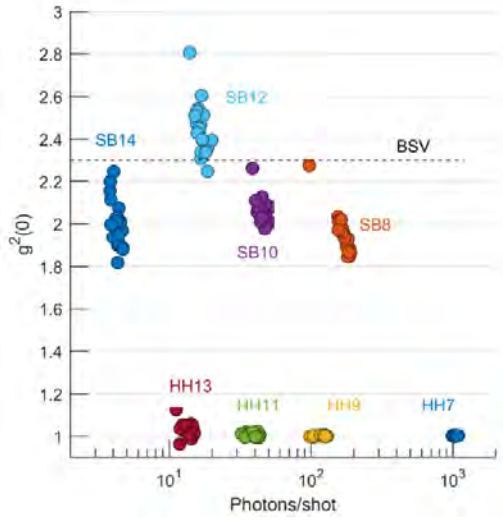


Figure 1. $g^{(2)}$ of the high harmonics is close to 1.0 (as expected of coherent beams), while for the sidebands $g^{(2)}$~2.6, comparable to that of the BSV beam (black dashed line).

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Non-classical Effects Drive Recombination in High-Harmonic Generation Using Circularly Polarized Fields

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Synopsis High-harmonic generation (HHG) is a nonlinear optical process in which a strong field interacts with a material, generating high-order harmonics of the driving field. Although circular polarization typically suppresses HHG, we demonstrate that non-classical structured squeezed light enables HHG even in these classically forbidden regimes. We show that non-classical fluctuations act as an effective force, guiding electrons to recombine and emit high-harmonic radiation. This work bridges quantum optics and HHG, offering a new approach to controlling light-matter interactions.

High-harmonic generation (HHG) is a nonlinear optical process where a strong field interacts with a material to produce high-order harmonics of the driving frequency, enabling ultrafast studies of matter at attosecond timescales. While HHG with circularly polarized light has traditionally been ineffective due to the electron being driven away from the ion core, i.e. no recombination step is possible, this research demonstrates how non-classical structured light, specifically circularly polarized light with squeezing applied along one polarization direction, overcomes this limitation. Quantum fluctuations introduced by squeezing create an effective force [1] that guides the electron back to the ion, enabling recombination and harmonic emission [2].

The spectral characteristics of the emitted harmonics depend on the type of squeezing (see Fig. 1): amplitude squeezing produces a single plateau with an extended cutoff, while phase squeezing creates a double-plateau structure with reduced yield in higher frequencies. These effects stem from the influence of quantum fluctuations on the field's effective ellipticity, allowing for recombination trajectories that are inaccessible in classical scenarios [2].

This work bridges quantum optics and strong-field physics, demonstrating how non-classical light can extend the boundaries of HHG, particularly by enabling harmonic emission in classically forbidden regimes. Beyond atomic systems, these findings pave the way for exploring quantum-driven ultrafast processes in solid-state [3] and molecular [4] systems. In these, it has been already shown that the use of classical structured light crucially shapes the HHG response of the material, depending on properties such as chirality [4]. Thus, adding non-classical features on top introduce an additional knob to manipulate light-matter interactions.

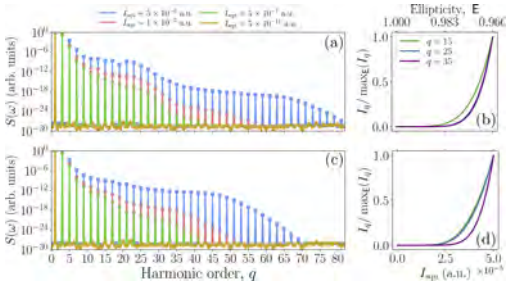


Figure 1. (a), (c) HHG spectra computed for amplitude and phase squeezing; (b), (d) scaling of selected harmonic intensities with the ellipticity.

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Measuring and controlling the birth of quantum attosecond pulses

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Synopsis In this work we transfer fundamental concepts in quantum optics into attosecond science. By driving a high harmonic generation process with bright squeezed vacuum (BSV, a non-classical state of light), and a strong coherent field, we imprint the quantum correlations of the input BSV onto both the electron wavefunction and the harmonics' field. We develop the ability to measure and control quantum correlations in both electrons and attosecond pulses, establishing a foundation for attosecond electrodynamics and manipulating the quantum state of electrons and photons with sub-cycle precision.

The generation and control of extreme ultraviolet (XUV) radiation by high harmonic generation (HHG) have advanced ultrafast science, providing direct insights into electron dynamics on their natural time scale and resolving ultrafast quantum phenomena in matter. HHG spectroscopy was, so far, based on the characterization and control of the classical properties of the high harmonics. Recent theoretical proposals[1,2] introduced novel schemes for generating and manipulating XUV HHG with quantum features.

Here, we demonstrate the generation of an attosecond pulse train in the XUV regime exhibiting controllable quantum properties. Combining a strong coherent field centered at 800nm with a BSV centered at 1600nm, we transfer the statistical properties of the quantum driving field into the sub-cycle dynamics of the field driven wavepacket. The wavepacket dynamics are subsequently imprinted onto the quantum properties of the emitted attosecond pulses, as illustrated in Fig. 1a. The two-color field acts as a sub-cycle interferometer[3], revealing attosecond scale correlations of the electronic wavefunction, mirroring the quantum correlations on the input BSV. The interferometric nature of our scheme enables homodyne-like quantum state tomography, allowing the reconstruction of the quantum state of XUV harmonics (represented by a Wigner function) in Fig. 1c. Our study transfers quantum optics to new extremes by enabling unique phase-resolved correlation measurements on attosecond time scales.

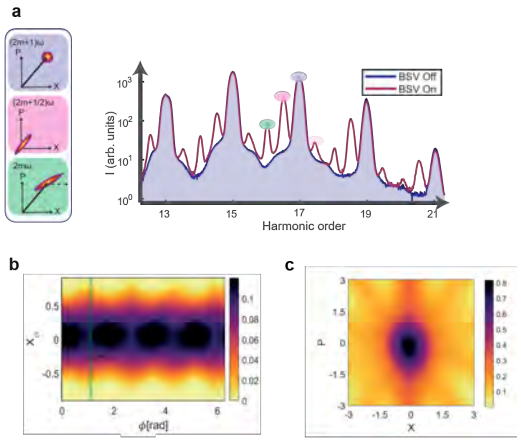


Figure 1. a, Measured spectrum with and without the BSV, and predicted quantum states of the different harmonics. **b,c** Quadrature and Wigner function of harmonic 14.5, exhibiting squeezing properties.

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Attosecond X-ray light sources driven by mid-infrared lasers

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Synopsis. Significant progress has been made in developing few-cycle, carrier-envelope phase-stabilized, high-peak-power lasers in the 1.6–2 μm range for generating attosecond X-ray sources in the water window. Recent breakthroughs in mid-infrared lasers have laid the foundation for further extending the high harmonic cutoff.

The development of chirped-pulse amplification (CPA) lasers in 1980s significantly transformed the study of intense laser–matter interactions. While most strong-field experiments have been confined to the near-infrared range of 0.8–1 μm, theoretical predictions indicate that many physical processes become more favorable at longer laser wavelengths [1]. For instance, as illustrated in Fig. 1, the cutoff photon energy in high harmonic generation (HHG) increases advantageously with the driving laser wavelength.

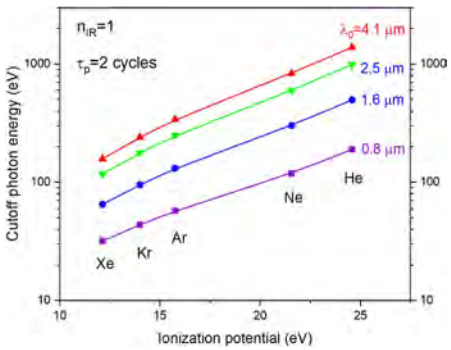


Figure 1. The calculated cutoff photon energy of phase matched HHG driving by lasers at different wavelengths [2].

Initial efforts to extend the high harmonic cutoff utilized optical parametric amplifiers (OPAs) [3] and optical parametric chirped pulse amplifiers (OPCPAs) pumped by Ti:Sapphire lasers, covering wavelengths from 1.1 to 1.8 μm. These developments enabled attosecond transient absorption spectroscopy experiments within the water window spectral region [4]. Subsequently, employing Yb-based lasers for pumping facilitated OPCPA operation at high repetition rates.

In recent years, high-power mid-infrared lasers have emerged as valuable tools for exploring wavelength scaling laws in strong-field atomic, molecular, and plasma physics. We demonstrated a femtosecond chirped-pulse amplifier based on cryogenically cooled Fe:ZnSe [5]. Its long upper-state lifetime allows the use of free-running, diode-pumped Er:YAG lasers as pump sources. Few-cycle pulses were generated by passing the CPA output, operating at a 400 Hz repetition rate, through a gas-filled hollow-core fiber, followed by dispersion compensation using bulk CaF_2. When filled with krypton, the fiber produced 1.14-mJ, 42-fs pulses centered at 4.07 μm [6].

Additionally, we developed a compact OPCPA system utilizing $ZnGeP_2$ crystals, which generates 3.2-mJ, 92-fs pulses centered at 3.1 μm at a 1-kHz repetition rate [7]. Pumped by a 2-μm chirped-pulse amplifier with a flat-top beam profile, the system achieves an overall efficiency of 16.5%, the highest reported efficiency for OPCPA at this wavelength to date.

These high-power, high-repetition-rate CPA and OPCPA mid-infrared sources will be employed for generating isolated attosecond keV X-ray pulses, air filamentation, and other strong-field physics experiments.

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High power CEP stable Ti:Sa amplifier seeded by OPCPA

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Synopsis We present our work on the unique architecture of a CEP stable Ti:Sa amplifier system seeded by a ytterbium pumped optical parametric chirped pulse amplifier. Passive CEP is obtained by difference frequency generation in the OPCPA seeder. An active correction loop for slow variations introduced in the Ti:Sa amplifier is done with a Dazzler device located in the stretcher.

We present the work on a unique and original Ti:Sa amplifier architecture intended for generating a CEP stable high average and peak power pulses for attosecond science.

The system is a grating based Ti:Sa amplifier from Amplitude (ARCO) delivering 10 mJ pulses at a repetition rate of 1 kHz and an output pulse duration below 30 fs (see Figure 1). The amplifier is seeded by a custom OPCPA system delivering pulses centered at 800 nm with a 150 nm bandwidth and passive CEP stability obtained through a difference frequency scheme.

The seeder pulse energy of several microjoules allows for robust operation of the broadband Ti:Sa amplifier. The temporal contrast is enhanced due to the very high pulse contrast from the OPCPA seeder and higher seeding pulse energy.

Passive single-shot CEP stability at the output of the seeder is obtained with a 1-hour measurement interval ($3.6 \cdot 10^7$ pulses at 10 kHz) with a record value as low as 300 mrad rms without active correction [1]. Only a low drift due to thermal issues and air fluctuations is observed. Local CEP stability over 1000 consecutive shots at 10 kHz is of the order of 200 mrad.

An active correction loop is added in the stretcher of the Ti:Sa system, in order to compensate for "slow" fluctuations and other noise contributions (e.g., acoustic noise) in the large amplifier. The loop consists of the measurement setup - an f-to-2f interferometer followed by a fast spectrometer (Fringeezz, Fastlite), and a phase actuator - pulse shaper (Dazzler, Fastlite). The latter is also used for spectral phase optimization. The active stabilization allows for reaching a 10 shots-averaged CEP stability of 230 mrad rms over one hour (see Figure 1).

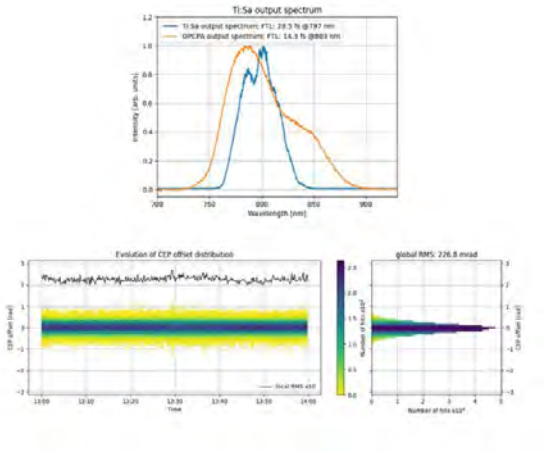


Figure 1. (Top) Output spectra of the OPCPA seeder and the Ti:Sa amplifier. (Bottom) Actively stabilized CEP with a 1-hour measurement interval averaged over 10 consecutive shots at 1 kHz.

This unique system architecture combines the best capabilities of both Ti:Sa laser amplification and OPCPA technologies in a single system leading to a cost-efficient high average and peak power with CEP stability for attosecond science such as high harmonic generation. While current performances are already compatible with attosecond applications, we believe to significantly reduce the CEP noise level in the future.

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Waveform-controlled near-single-cycle pulses at the 100 TW level for intense attosecond sources

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Synopsis We report on an optical parametric synthesizer system providing waveform-controlled 4.3 fs light pulses with 100 TW peak power. The carrier envelope phase is stabilized to <300 mrad. Utilizing temporal super-resolution the pulses are shortened to sub-4-fs duration. This light source is dedicated to advance the frontiers of intense attosecond x-ray sources based on high-harmonic generation in gases as well as on relativistic plasma surfaces.

Ultrafast laser technology had a significant development in the past few decades, resulting in ultrahigh peak power lasers (from 100 TW to multi PW) as well as systems with near single cycle pulse duration. The combination of these parameters was not available until now, though such a laser has promising applications.

We present a light source, termed Light Wave Synthesizer 100 (LWS100), that merges a near-single-cycle pulse duration and a peak power of 100 TW and extends these with a stabilized carrier-envelope phase (CEP). This is achieved by a non-collinear optical parametric amplifier frontend and three optical parametric synthesizer (OPS) [1] stages resulting in a pulse energy of 460-500 mJ. Each OPS stage involves two optical parametric chirped pulse amplifiers that boost two complementary spectral ranges to reach an almost octave spanning spectrum (580-1030nm).

The CEP is measured at the end of the system and a feedback loop corrects for slow changes in the amplifiers behind the CEP-stable frontend. Longer term (> 1 hour) stabilization to 300 mad or below is routinely reached (see Fig. 1). Relativistic plasma physics experiments are sensitive to the high-dynamic range temporal contrast. Therefore, the system has been developed to achieve an extraordinary contrast, which reaches 8 and > 11.5 (detection limit) orders of magnitude at 2 ps and 20 ps before the main pulse.

Utilizing temporal super-resolution [2] via spectral amplitude shaping the pulses are shortened to 3.9 fs. This technique can increase the continuum in the high-order harmonic generation (HHG) spectrum.

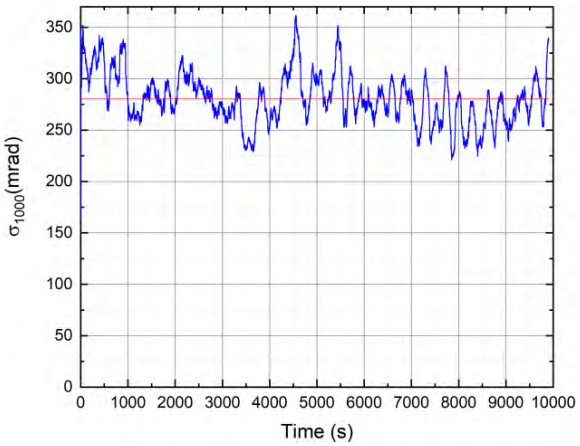


Figure 1. RMS CEP stability of LWS100 over 1000 shots operating at 100TW and 10 Hz over almost 3 hours. The RMS over the full time span is 282 mrad (red line).

LWS100 and the underlying technology opens up a myriad of exciting and unique applications, among others creation of intense isolated attosecond x-ray pulses via HHG from gases as well as relativistic plasma surfaces [3].

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Single-shot pulse retrieval of femtosecond bright squeezed vacuum

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Synopsis We report the first full single-shot characterization of femtosecond single-mode bright squeezed vacuum (BSV), a novel driver for attosecond quantum physics. Applying spectral interferometry to 1030-nm BSV, we find an average duration of 44fs and consistent shot-to-shot behavior of the group delay.

Bright squeezed vacuum (BSV) takes the phenomenon of squeezed vacuum to the extreme [1]. Extremely high photon numbers of up to 10^{13} can be found inside a single BSV pulse due to the antisqueezed quadrature, with effective pulse durations down to tens of femtoseconds [2]. BSV is sufficiently intense to drive nonperturbative HHG from solids [2] and multiphoton electron emission from nanotips [3]. Intriguingly, BSV has the potential to explore attosecond light-matter interactions above the conventional damage threshold [2]. However, the full temporal properties (mean and distribution) of BSV pulses have remained elusive. Here demonstrate full temporal characterization (amplitude and phase) of single-shot BSV pulses, for the first time.

In our experiment, we produce spatially single-mode 1030-nm BSV pulses from 515-nm pulses using a double-crystal configuration, resulting in a mean pulse energy of ~100 nJ. To reconstruct an isolated BSV pulse, we interfere it with a broadband 1030-nm coherent-state reference pulse. We fully characterize the reference pulse by frequency-resolved optical gating (FROG). We record single-shot spectral interferograms with a BSV-coherent delay of ~2 ps and extract the relative spectral phase and amplitude of each BSV shot. Post-selecting on the fundamental temporal BSV mode, we obtain spectral intensity and group delay (Fig. 1(a)) as well as the temporal pulse shape (Fig. 1(b)). Averaging over 125 single-pulse reconstructions, we find a spectral width of 14 THz.

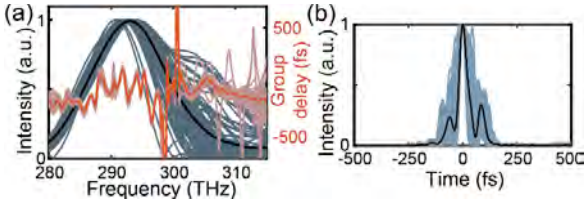


Figure 1. (a) Normalized single-shot 1030-nm BSV spectra (grey) and average spectrum (black), as well as group delays (grey/orange) and their average (orange). (b) Normalized pulse intensity in time (black: average).

The averaged group delay shows oscillations as a function of frequency, which we attribute to imperfections in the FROG retrieval of the SPM reference pulse. The shot-resolved group delay is fluctuating around the average but displays a largely consistent behavior. The reconstructed pulses possess an average duration of 44 fs with a standard deviation of 17 fs, with most of the energy contained in the central burst along with side peaks. Remarkably, the average duration is much shorter than the duration of the 1030-nm femtosecond laser pulses that drive the experiment (~180 fs).

In conclusion, we demonstrated the first single-shot pulse retrieval of BSV pulses. This ability will open the door to sub-cycle spectroscopic measurements of attosecond dynamics and time-domain quantum state tomography.

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Characterization of sub-3-fs tunable VUV pulses

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Synopsis : We generate and in-situ characterize ultrashort pulses in the vacuum ultraviolet. The VUV source relying on soliton dynamics in hollow waveguides delivers tunable radiation below 200 nm with multi-microjoule energy and sub-3-fs duration. The VUV pulses are fully characterized using a two-photon photoelectron FROG scheme.

Vacuum ultraviolet (VUV) radiation has a large variety of applications ranging from spectroscopy, material science and photolysis to quantum chemistry. The utilization of complex spatio-temporal soliton-dynamics in hollow-core fibers (HCF), a phenomenon called resonant dispersive wave generation (RDW), allows for the generation of highly energetic (µJ level) VUV-pulses with a broad spectral tunability, yielding pulse durations in the few-fs regime [1]. So far, the technique has proven to deliver sub-3-fs pulses in the deep-UV, which were directly characterized via an all-optical-detection setup [2].

In this work we expand the direct characterization of the RDW pulses to the VUV spectral region below 200 nm – where an all optical characterization of the the few-fs broadband pulses becomes impossible. We show that the tunable pulses retain the extremely short duration while carrying a few µJ of energy.

The VUV-pulses are generated through RDW generation in a hollow-core fiber and are fully characterized by a photoelectron FROG scheme which is displayed in Fig. 1. The pressure dependent spectrum of the ultrashort tunable VUV pulses is shown in Fig. 1 a) and shows a large tunability. The pulse characterization was achieved by two-photon ionization of a rare gas atoms. To do so, the VUV pulse was split into two pulse replica and then focused into an atomic gas target at the center of a velocity map imaging spectrometer. The resulting spectrogram is displayed in Fig. 1 b) and closely resembles common FROG traces relying on second-harmonic generation. This allows for a reconstruction of the amplitude and phase of the vaccum-ultraviolet pulses, as shown in Fig. 1 b).

Furthermore, pulses with different central frequencies have been characterized and we show

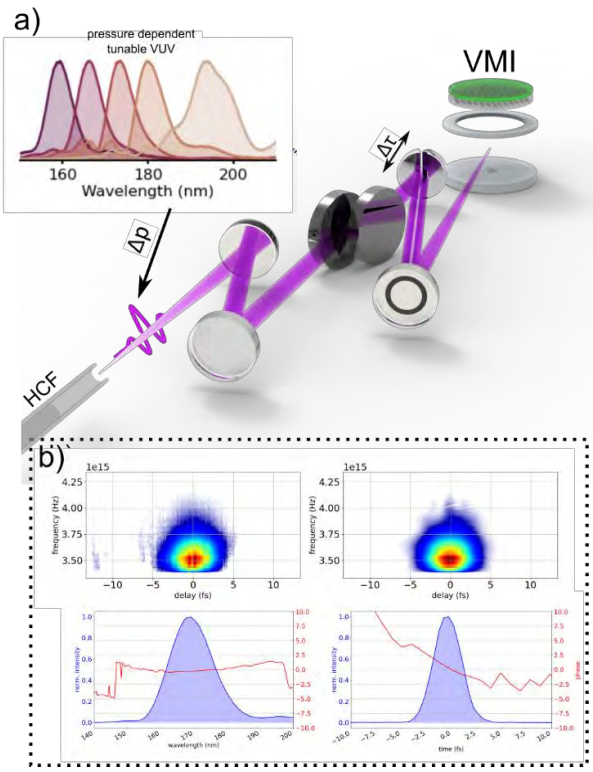


Figure 1 : Sketch of the photoelectron FROG scheme used to characterize VUV-pulses with a tunable spectrum, shown in panel a). The measured and reconstructed FROG trace together with the retrieved pulse and spectrum is shown in b).

that in each case sub-3 fs pulses with an energy exceeding 1 µJ can be be generated [3]. The pulses have further been applied for the investigation of ultrafast relaxation dynamics occurring in photoexcited molecules.

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New ultrafast sources and science enabled by advanced nonlinear optics in hollow-core waveguides

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Synopsis Ultrafast light sources based on soliton dynamics in gas-filled waveguides can fill several important technology gaps in ultrafast science. We will discuss our work developing these sources and applying them to cutting-edge experiments in ultrafast science, as well as future directions.

The generation of ever shorter pulses at a wider variety of central wavelengths has been a core goal of ultrafast optics since its inception, and new advances in light sources have always enabled new insights in ultrafast science. State-of-the-art ultrafast spectroscopy and imaging experiments demand previously inaccessible parameter regions, such as highly energetic sub-cycle field transients [1] and extremely short far-ultraviolet pulses [2].

Ultrafast laser sources based on soliton dynamics in gas-filled hollow-core fibres can address several of the most pressing technology gaps in ultrafast science. For the correct combination of parameters, the intricate interplay between optical nonlinearity and anomalous dispersion can cause a pulse coupled into such a waveguide to undergo dramatic self-compression. This process can create fully compressed, sub-cycle-duration field transients without any external compression optics. If the process is pushed further, the self-compressed soliton breaks up and emits a resonant dispersive wave (RDW), a secondary pulse at a wavelength which depends on the dispersion landscape, and which can therefore be tuned simply by changing the gas pressure in the fibre. The RDW pulse can be of few-femtosecond duration and its tuning range extends from the vacuum ultraviolet across all of the visible and to the near infrared, depending on the driving conditions [3]. Sub-cycle self-compression and ultraviolet RDW emission were first studied in microstructured fibres [4]. The larger core size of capillary fibres, which have been a core technology in attosecond science for several decades [5, 6], allows for significant up-scaling of the pulse energy and peak power [7].

Here, we will provide an overview of our work on soliton-based ultrafast light sources to date, addressing both fundamental aspects and scientific applications. Soliton-based sources have now been applied in experiments in ultrafast spectroscopy [8, 9] and photoelectron imaging [10], with more applications in development. We will also address current and future directions, including improvements in the repetition rate and average power, further dramatic scaling of the pulse energy and peak power to the terawatt regime, and extensions to exotic polarisation states such as radial vector beams.

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Multi-Mirror Multi-Pass Cells for Compression of Energetic Narrowband Laser Pulses into the Femtosecond Regime

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Synopsis We propose using multi-mirror multi-pass cells to compress energetic narrowband lasers into the femtosecond regime. Spectral broadening is demonstrated in air as a proof of principle, using an 11-mirror cell providing 297 passes. With input pulses of 1 ps duration, 280 μJ energy and <300 MW peak power, a 15-fold spectral broadening is achieved, corresponding to an output Fourier limited duration of 56 fs.

Post-compression of (sub)-ps laser pulses to sub-100-fs durations is now standard for high-peak-power, high-average-power sources [1]. We propose a new post-compression geometry for extreme temporal compression of energetic narrowband lasers, such as diode or electrically pumped four-level systems which can deliver kW-average powers [2], but have pulse durations from a few to hundreds of picoseconds. Their direct compression into the femtosecond regime will further enhance the average power of high-peak-power sources, critical for next-generation laser-plasma accelerators [3] and attosecond science.

The key to achieving the required extreme spectral broadening factors >1000 is a power-scalable, virtually lossless waveguide, enabling nonlinear propagation over km-scale lengths. Multi-pass cells (MPCs) offer a compact and efficient solution for nonlinear pulse compression, combining high transmissions, large compression factors, and handling high pulse energies [4]. To extend their nonlinear propagation length beyond the typical 20 to 30 round-trips in Herriott configurations, we implemented an MPC with 11 mirrors (2-inch diameter) arranged in a circle (Fig. 1b), allowing 297 passes within a 1 m^2 footprint. Similar setups have been used for absorption spectroscopy [5], only up to 5 mirrors. As a proof of principle, we demonstrate spectral broadening in air using a burst-mode Yb:YAG laser producing 1-ps, 1-mJ energy pulses. At 280 μJ input energy and <300 MW peak power, a spectral broadening factor of 15 is achieved (Fig. 1a), corresponding to a Fourier-limited duration of 56 fs, with >85% transmission. Beam quality remains stable at low energies (M^2 = 1.2 ×1.1).

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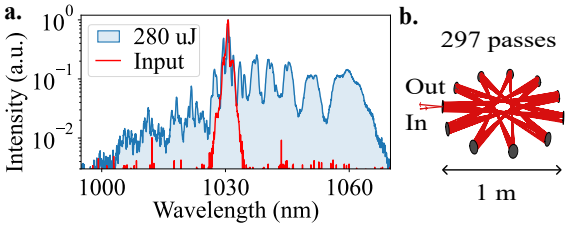


Figure 1. a. Input and output spectra at 280 μJ input pulse energy, broadened in air. b. Simulated beam trajectory in the 11-mirrors MPC.

Using 11 mirrors of 3-inch diameter, an MPC supporting 1300 passes could be arranged with the same footprint. For this configuration, simulations with the SISYFOS package [6] predict a >1000 broadening factor for 300 ps long, 100 mJ input pulses and a cell filled with 13 bars of krypton. We infer that the new approach can enable compact, cost-efficient light sources with TW-level peak power at multi-kHz repetition rates, which would be groundbreaking for upscaling the average power of ultraintense lasers.

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Pulse compression of 300 W, 12 mJ with 83% transmission in hollow-core fiber

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Synopsis We present the power scaling of pulse post-compression in a stretched hollow-core fiber (HCF) setup. An air-cooled HCF setup with up to 300 W of average power reaches a transmission of 83%. More than a ten-fold compression of 1.3 ps pulses down to 100 fs in a single HCF setup was demonstrated. The versatile HCF setup supports variable repetition rates from 100 kHz (3 mJ) to 25 kHz (12 mJ), while maintaining a constant 300 W average power.

Laser technology based on ytterbium doped gain media is enjoying increasing popularity and gave a huge push to the development of pulse post-compression schemes. Such, post-compression of 70 mJ of input energy at 1030 nm [1] and of 580 W average power with 5.8 mJ energy [2] have been demonstrated. Recently, we have demonstrated pulse compression down to few-cycle pulses (6 fs at 1030nm) at average power of 80 W, 2mJ [3] in a double stage HCF compression setup. In this contribution, we demonstrate that by utilizing largely the same hardware as in the 80 W experiment, it is possible to raise both the input average power to 300 W as well as the pulse energy to 12 mJ.

The driving laser is an Yb:YAG InnoSlab laser (Amphos GmbH) delivering 1.26 ps long pulses with a variable combination of pulse energy (3-12 mJ) and repetition rate (25-100kHz) while keeping the average power constant at 300 W. With this input we achieved simultaneous upscaling of average power and pulse energy to 250 W and 10 mJ at the HCF output. This denotes to date the highest energy pulse compression in HCFs filled with atomic gases at the multi-hundred-Watt level. Similar results, was achieved with molecular (N_2, O_2, N_2O) post-compression [4].

The beam was coupled into 3.1 m long HCF using a lens with f=2.8 m. To have less heat deposited to the fiber tip we chose a high throughput - large mode fiber of 1 mm inner diameter.

For 300 W input power and energy of 12 mJ HCF was filled with Krypton at a relative pressure of 0.17 bar (or 1.5 bar for 3 mJ, 300W). Pulse compression was performed using 36 bounces (40 bounces for 3 mJ case) on chirped mirrors (−500 fs^2 at 1010–1060 nm, few-cycle Inc). The temporal characterization by autocorrelation shows that the pulse was compressed down to 103 fs (99 fs for 3 mJ case) at FWHM (Fig. 1 magenta curve), which corresponds to 12-fold pulse compression at 300 W of input average power. The spatial quality of the beam after the HCF was verified by refocusing the beam after the HCF to a symmetric focal spot. A stable 1-hour long power measurement demonstrates a remarkably low standard deviation of 0.58% at 250 W of average power at 25 kHz.

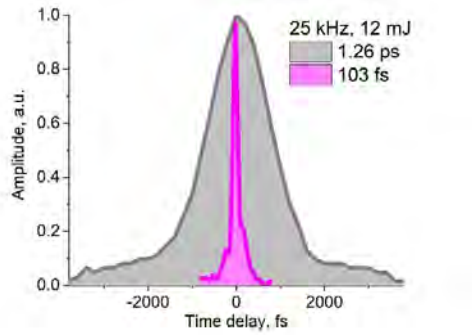


Figure 1. Autocorrelation showing compressed pulse duration after the HCF, 103 fs (magenta), versus input pulse duration of 1.26 ps (gray) at 25 kHz.

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Terawatt optical attosecond pulses and 100 GW-scale far-ultraviolet pulses through extreme soliton dynamics

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Synopsis Soliton-driven light sources can directly deliver optical attosecond pulses in the visible-infrared range and few-cycle deep and vacuum ultraviolet pulses. Here we describe our new XSOL (Extreme Soliton) beamline designed to up-scale soliton dynamics and deliver sources for driving strong-field physics. Initial results include the generation of terawatt sub-cycle self-compressed pulses and deep ultraviolet pulses with greater than 170 µJ energy through resonant dispersive-wave (RDW) emission. RDW tunability down to 140 nm has been achieved.

Optical soliton dynamics in gas-filled hollow capillaries enable extreme pulse self-compression in the visible-infrared region and efficient, tuneable, few-femtosecond pulse generation in the deep and vacuum ultraviolet (DUV/VUV) through resonant dispersive-wave emission (RDW) [1]. This technique is scalable in peak power and energy by adjusting the fibre core size and length. The first demonstration achieved 40 GW peak power, 1.2 fs self-compressed pulses, and tuneable DUV/VUV generation with tens of µJ energy. Similar experiments have shown the DUV pulses to have a sub-3 fs pulse duration [2]. Here we describe our new XSOL (Extreme Soliton) beamline that has been designed to up-scale soliton dynamics, delivering a ten-fold peak power and energy upgrade for driving strong-field physics.

We utilise three stretched capillary fibres (4.7 m long, 530 µm core diameter) as illustrated in Fig. 1(a). The first two stages pre-compress 40 fs input pulses through spectral broadening and phase compensation with chirped mirrors to obtain 5.5 fs, ~ 3 mJ pulses with the option for pre-pulse contrast enhancement. The third stage is for the soliton dynamics. The input and output of all three fibre stages are maintained in large vacuum chambers to avoid free-space nonlinearity and ionisation, and to allow for long in-vacuum focusing distances to account for the low-divergence high-fluence beams emerging from larger core fibres. The helium gas used in all three stages is therefore injected by side ports along the fiber length.

We have achieved self-compression to 0.77 fs (envelope full-width at half-maximum), 2.1 mJ pulses with 1.36 TW peak power (Fig. 1(b)), characterised using an in-vacuum TIPTOE setup. Driving RDW emission yields tuneable deep-ultraviolet pulses with energy exceeding 170 µJ. When accounting for expected pulse duration, these DUV pulses are at the 100 GW level. Extended tuneability down to 140 nm (Fig. 1(c)) has also been obtained, with full in-vacuum energy and temporal characterisation in progress. Our next goal is to utilise these extreme pulses for strong-field physics experiments.

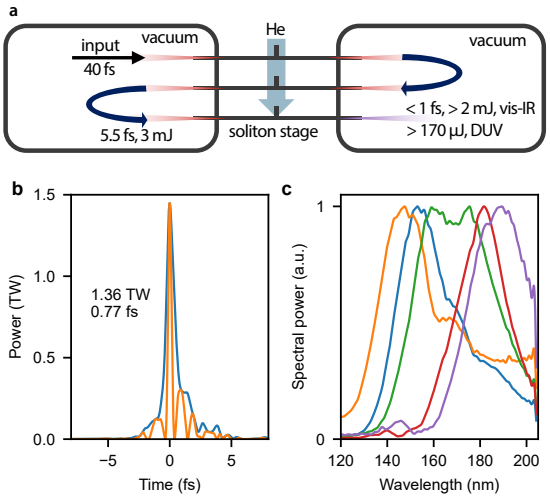


Figure 1. (a) Experimental setup used for extreme soliton dynamics. (b) Measured sub-femtosecond TW-scale pulse. (c) Tuneable VUV generation.

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Recent developments of attosecond high-harmonic generation sources in and outside the Extreme Light Infrastructure

B Major

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Synopsis In my talk I will present recent developments of high-harmonic generation sources at the Extreme Light Infrastructure (ELI) European Research Infrastructure Consortium (ERIC) facilities along with some highlights of user experiments carried out wit them. Additionally, I will include some current advancements in the topic achieved in laboratories outside ELI.

Attosecond sources based on high-harmonc generation (HHG) are the key equipment for attosecond science and technology, using either gas-based or solid-phase targets for obtaining coherent extreme-ultraviolet (XUV) radiation [1]. Accordingly, the Extreme Light Infrastructure (ELI) European Research Infrastructure Consortium (ERIC) offers multiple of these equipment to its users at its two research institutes, at ELI Beamlines [2] and at ELI Attosecond Light Pulse Source (ALPS) [3]. Since ELI is dedicated to providing cutting edge equipment to its user communitie(s), substantial efforts are made to develop existing sources to facilitate experiments defining the forefront of science.

As an example of advancements, the High Repetition rate laser system-based Gas High-order Harmonic Generation attosecond beamline for Gas targets (HR Gas) beamline of the ELI ALPS facility has recently been extended with a unique, optically-locked versatile double interferometer for pump and probe delay stabilization, operatiomal with the high-average power laser beams of the HR laser system family (>1 mJ, 100 kHz, >100 W [4,5]), and allowing for multiple-day long attosecond pump-probe measurements with the Reaction Microscope end-station [6]. The twin setup – designated to surface science thanks to the NanoESCA end-station (a photoemission electron microscope (PEEM) with energy and spin resolving capabilities [7]) –, the HR Condensed beamline [8], has also been demonstrated to access attosecond time-scale dynamics in solid phase [8].

Another recent advancement, also part of the HR Gas beamline, is the installation of a unqie, two-stage polarimeter, giving access to the full polarization state of the generated broadband XUV radiation [10]. This capability, combined with the generation of circularly polarized XUV light, is enabling access to novel chiro-optical phenomena, as highlighted by others [11].

The ELI ALPS and Beamlines facilities provide additional opportunities to those needing coherent XUV sources at lower (<= 1 kHz) repetition rates [2,3], and terahertz (THz) – XUV pump-probe capabilities are also being currently developed.

As additional outlook, I will also briefly go throught recent developments at universities and research institutes of the world where possible new trends are being pursued in the field of HHG-based attosecond sources and their blooming topics of applications.

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Microfluidic-based sources for efficient XUV generation and manipulation

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Synopsis: We report on the recent experimental development for the study of ultrafast electron dynamics in materials and complex molecules using our efficient XUV radiation source based on HHG inside a microfluidic device.

Understanding molecular dynamics is crucial in ultrafast spectroscopy due to its importance in physics, biochemistry, and quantum technologies. High Harmonic Generation (HHG) using intense femtosecond lasers and noble gases produces tabletop coherent Extreme Ultraviolet (EUV) pulses. These ultrafast emissions offer attosecond resolution and chemical selectivity, revealing insights into electronic dynamics and light-matter interactions [1]. However, challenges such as setup complexity and low efficiency, particularly at higher photon energies, exist. We present a versatile beamline for EUV and soft-X-ray spectroscopy, designed for time-resolved experiments with complex molecules. Utilizing a microfluidic HHG device with hollow core channels, we achieve high photon flux and effective phase matching for harmonics up to 200 eV and beyond [2]. The HHG source integrates with a beamline for Transient Absorption (TA) and reflectivity measurements, featuring a high-resolution EUV/soft-X spectrometer and a polarimeter.

In the TA setup, ultrashort UV, visible, and mid-IR pulses photoexcite samples, probing electronic dynamics by monitoring EUV absorbance changes. We have developed delivery systems to study biomolecules in high-vacuum conditions for gas and liquid phases. This microfluidic approach allows precise control of HHG beam properties, enabling structured EUV and soft-X light for ultrafast spectroscopy and high-resolution imaging. Furthermore, this integrated microfluidic approach allows controlling the spatial properties of the HHG beam through the direct manipulation of the waveguide's modal characteristics, making it possible to generate high-brightness attosecond XUV and soft-X ray pulses with unique polarization properties.

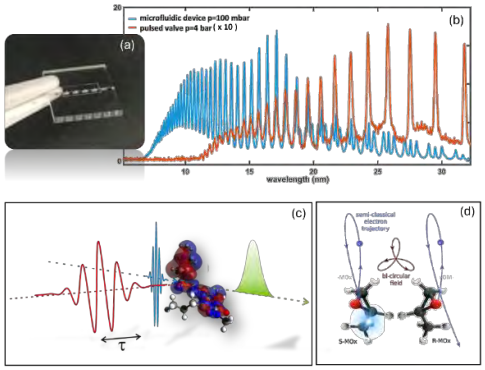


Figure 1. a microfluidic device used for HHG. **b** HHG spectrum generated with the microfluidic approach (blue) compared with the spectrum generated by gas-jet valve (red). **c** and **d** provide a concept-frame of TA and HHG spectroscopy in complex molecules.

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High Harmonic Generation with Two Non-Collinear Drivers: A Unique Gateway to Extreme Nonlinear Phenomena

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Synopsis High-order harmonic generation (HHG) is traditionally understood as a semi-classical interaction between matter, treated with a quantum formalism, and an intense laser field, incorporated classically. Recently, HHG was revisited as a versatile platform for extreme quantum optics. We have bridged the gap between these two approaches, by evidencing the multiple photon pathways adding up to form the standard HHG process. Our findings rely on the analysis of the yields and profiles of the harmonic beamlets issued from HHG driven by two non-collinear beams through three consistent observations.

High-harmonic generation (HHG) is typically described as a strong-field process, which accounts for key features such as an effective nonlinear order, the characteristic plateau and cutoff in the spectrum, and the temporal coherence of the harmonics. However, when HHG is driven by two laser beams, the emergence of multiple harmonic beamlets suggests an alternative interpretation in terms of photon pathways [1, 2]. This framework recovers fundamental conservation laws, including those governing energy, spin and orbital angular momenta, as established over the past decade. Yet, a fully photon-based interpretation of the harmonic beamlet yield and spatial profile has remained elusive.

Here, we present experimental evidence that HHG can indeed be described as a superposition of simultaneous photonic processes, adding up coherently [3]. Beyond the minimum number of photons required to produce a given harmonic, each harmonic channel involves one or more additional photon pairs, corresponding to stimulated absorption and emission events. A simple theoretical model enumerating these pathways and their probability given the number of involved photons aligns with experimental data, explaining that the yields of the beamlets rise and fall in succession. Additional signatures appear in the spatial profiles [4] and phases [5] of the beamlets, which were also observed experimentally. We anticipate that this remarkably simple picture of HHG could serve as a foundation for extreme quantum optics or fundamental optics [6].

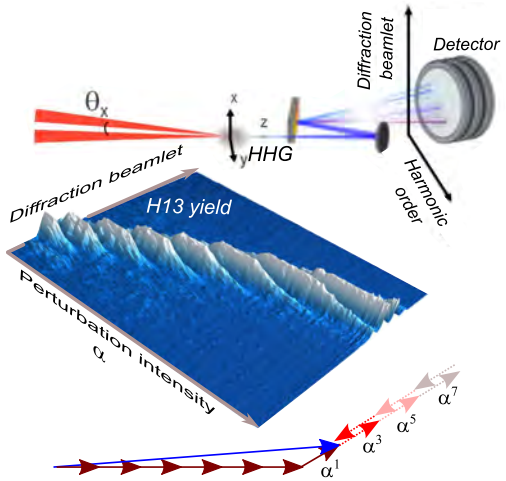


Figure 1. (Top) Two non-collinear beams intersect in the HHG gas medium to yield harmonics split in a series of beamlets (Middle) Yields of the beamlets of harmonic 13 against the perturbing intensity, showing successive bell-shaped yield curves reminiscent of interferences of several photon pathways. (Bottom) Selection of interfering photon pathways contributing to the order of diffraction 1 of harmonic 7.

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High-order harmonic up-conversion of spatiotemporal and spatioespectral optical vortices into the extreme ultraviolet

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Synopsis Spatiotemporal optical vortices (STOV) are structured light fields with distinctive topologies that intertwine the spatial and temporal domains. Their generation have been limited to the infrared/visible, limiting their potential applications. We demonstrate theoretically and experimentally the nonlinear up-conversion of infrared STOVs to the extreme-ultraviolet via high harmonic generation. These high-frequency STOVs are unique for probing ultrafast electronic dynamics with spatial and temporal coupling at attosecond and nanometer scales.

In recent years there has been a rapid growing interest in a new family of light vortices kwon as spatiotemporal optical vortices (STOV) or transverse vortices [1]. These unique light fields couple the spatial and temporal domains, carrying a twisted phase profile in the spatiotemporal domain. Unlike spatial or longitudinal vortices, whose transverse phase profile show a singularity aligned in the longitudinal direction, STOV beams are not propagation eigenmodes [2]. Thanks to their unique properties, as their nontrivial propagation dynamics or their associated transverse angular momentum, they are used in diverse applications, from information transmission and nanoparticle manipulation to the study of the electronic response on ultrafast timescales.

The low efficiency of optical elements beyond the ultraviolet limits the generation of high-frequency STOVs, constraining their applications to the infrared/visible regimes [3]. In this work we demonstrate the generation of extreme-ultraviolet (EUV) STOVs vis nonlinear up-conversion in high-order harmonic generation. We follow a similar strategy as that demonstrated to produce longitudinal EUV vortex beams carrying orbital angular momentum [4]. However, the propagation dynamics of EUV-STOVs is more complex as STOVs are not eigenmodes of propagation. With the help of advanced numerical simulations, we explore the correspondence between spatio-temporal and spatio-spectral optical vortices (SSOV) upon propagation.

Our understanding allows us to experimentally demonstrate the generation of EUV STOV/SSOVs, showing excellent agreement with our numerical simulations (see Fig. 1) [5]. Our work paves the route for potential applications of high-frequency STOVs/SSOVs with controlled topological charges at attosecond and nanometer scales.

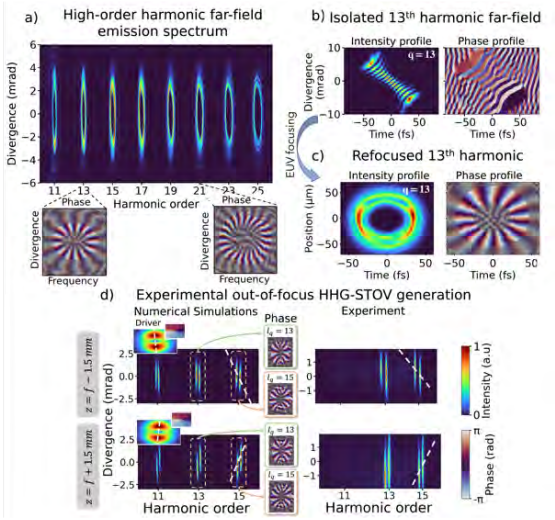


Figure 1. High-order harmonic generation results driven by STOVs. a) The simulated far-field spectrum shows elliptical SSOVs driven by a single charged elliptical STOV. b) Spatiotemporal intensity and phase distribution of the far-field 13th harmonic order and its refocusing (c), forming an elliptical STOV at the focus. d) On the left, numerical far-field spectra driven by a single charged inhomogeneous STOV before and after focus. On the right, the experimental far-field harmonic STOV spectra.

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Hollow Gaussian Beams for Low-Divergence High Harmonic Generation and Attosecond Pulse Optimization

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Synopsis We generate Hollow Gaussian Beams (HGBs), characterize their spatial properties, and investigate their impact on high-order harmonic generation (HHG) in argon. The harmonics generated with HGBs show a reduced divergence with increasing order compared to normal Gaussian-shaped beams. We predict theoretically how this effect can be used to enhance the attosecond pulse brightness. Our results suggest HGB-driven HHG as a promising approach for compact setups with high flux.

High-order harmonic generation (HHG) is an established method for producing coherent extreme-ultraviolet (XUV) radiation with femtosecond to attosecond pulse durations [1]. Controlling both the divergence and spot size of high-order harmonics is crucial for efficient focusing and optimizing the brightness. Driving pulses with Gaussian spatial profile usually generate harmonics with the smallest far-field divergence, when the gas target is placed before the geometrical focus, but the divergence increases with the gas target at focus [2]. Spatially shaped flat-top beams can reduce divergence of all the harmonics orders and improve XUV focusability by minimizing chromatic aberrations [3, 4]. Here, we present an alternative approach to control far-field divergence and focusability of the generated harmonics by using HGBs. A HGB is generated using a spatial light modulator and characterized before driving HHG in an argon gas jet. The far-field divergence of the generated harmonics is measured and compared to conventional Gaussian-driven HHG. As shown in Figure 1, harmonics generated with HGBs exhibit decreasing divergence with increasing harmonic order, in contrast to the increasing divergence typically observed at focus with Gaussian beams. This approach can be particularly advantageous for high pulse energy driving lasers, where peak intensity constraints require bulky setups. To interpret these results, we perform full quantum and macroscopic HHG simulations [5]. Theoretical modeling shows that the HGB's ring intensity profile and flat phase distribution suppress the influence of the harmonic dipole phase across all harmonic orders, leading to near-diffraction-limited harmonics. We show numerically that although the harmonics are generated on a ring in the gas target, they can be refocused to a single spot with minimal chromatic aberrations and very high intensity. We anticipate applications in both compact attosecond sources and high-power laser systems where managing peak intensity is critical.

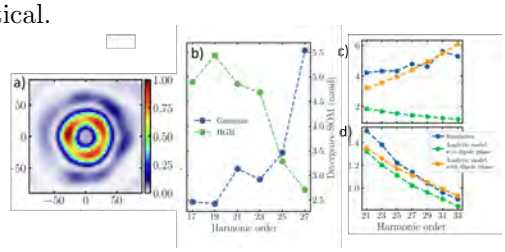


Figure 1. Harmonic generation with Gaussian and Hollow Gaussian Beams. (a) HGB intensity profile at focus. (b) Measured harmonic divergence. (c,d) Theoretical divergence for Gaussian- and HGB-driven HHG.

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Table-top all-attosecond transient absorption spectroscopy

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Synopsis Our first results on table-top all-attosecond transient absorption spectroscopy (AATAS) will be presented, showcasing examples of ultrafast dynamics studied in atoms, molecules and solids.

A longstanding goal in attosecond science is the ability to perform attosecond-pump attosecond-probe spectroscopy (APAPS). Pioneering experiments have been performed using HHG [1-3] and FEL [4,5] sources. All-attosecond transient absorption spectroscopy (AATAS) [4] is a particularly attractive technique, as it combines an extremely high temporal resolution in both the pump and the probe steps with an excellent spectral resolution.

Here I will present our first results on table-top AATAS in atoms, molecules and solids. Important milestones for achieving this goal will be discussed, which include XUV intensity scaling [6], repetition rate scaling [7] and the development of a terawatt-level, cascaded post-compression scheme [8].

In a first series of experiments, we studied electronic coherences in atoms [9]. Fig. 1**a** shows an AATAS map recorded in Ar, where the broadband blue features correspond to the depletion of neutral atoms. The narrowband white and red features are attributed to resonant absorption in the generated Ar^+ ions. Clear oscillating features with a period of 23 fs are observed (see also Fig. 1**b**), which are the result of generating these ions in a superposition of their ground-state and excited-state spin-orbit manifolds.

An attractive feature of APAPS is that no strong NIR laser fields are required. Nevertheless, it is important to understand the role of XUV strong-field effects. To this end, we have performed an AATAS experiment in He, showing that the lineshape of the 2p resonance can be modified with attosecond precision. A Fano-like lineshape is observed at high XUV intensities, demonstrating nonlinear frequency conversion induced by the intense attosecond pulses. Strategies to minimize strong-field effects in future AATAS experiments will be discussed.

Our results demonstrate that, owing to its broad bandwidth, the high stability and accessibility, HHG is a very attractive source for AATAS. The excellent signal-to-noise ratio suggests that it will be possible to extend table-top AATAS to higher photon energies in the future.

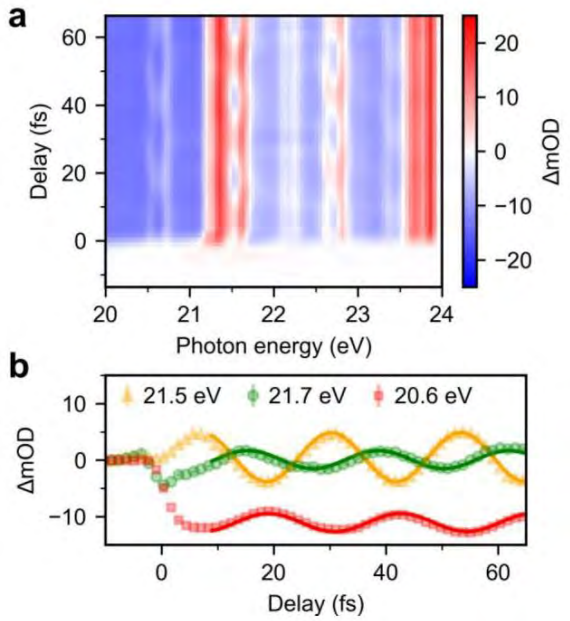


Figure 1. a, AATAS map recorded in Ar, showing oscillating features with a period of 23 fs, which are attributed to electronic coherences. **b**, Lineouts at three photon energies (data points) and corresponding fits (solid lines).

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Coincidence measurements of two-photon double ionization driven by a high-harmonic generation source

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Synopsis We present coincidence measurements of two-photon double-ionization (TPDI) of argon at 26.5 eV, below the second ionization potential. Using the results of atomic structure calculations, we interpret the measured photoelectron spectra with regard to three different double ionization mechanisms.

Recent advances in laser technology and high-harmonic generation (HHG) sources have made non-linear processes in the extreme ultraviolet accessible using table-top sources with kHz repetition rates [2]. One of the most fundamental non-linear process involving correlated electron dynamics is two-photon double-ionization (TPDI) of atoms. The process comes in two distinct variants: above the second ionization potential, ionization proceeds sequentially with the photoelectron energies given by the differences between the photon energy and the ionization potentials; below the second ionization potential, the excess energy is shared among the photoelectrons. The ratio of energy sharing is of fundamental interest to understanding the underlying electron-electron interaction [1].

Here, we present measurements of TPDI using quasi-monochromatic femtosecond pulses at 26.5 eV, obtained from the 11th harmonic of a frequency-doubled Yb:YAG fiber laser, which is operated at a repetition rate of 100 kHz. The XUV pulses are back-focused using a suitably coated mirror ($f = 75$ mm) into a cold jet of argon atoms, situated in the center of a reaction microscope. Ions and electrons are detected in coincidence with the total count rate kept well below one event per second, facilitating the measurement of clean ion-electron coincidences, as shown in figure 1. In the photoelectron energy distribution measured for Ar^{2+}, the prominent line arising from photoionization of Ar by the 11th harmonic is not absent. This indicates that double ionization does not proceed via the ground state of the Ar^+ cation but a more intricate mechanism than sequential TPDI is at play.

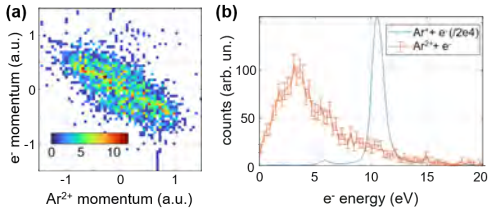


Figure 1. Photoeletron-photoion coincidence spectrum showing the correlations between the momentum components along the XUV polarization for photoelectron and Ar^{2+} ion. (b) Energy distributions for photoelectrons detected in coincidence with Ar^+ (blue) and Ar^{2+} ions (red).

In the talk, we will discuss the contributions of different TPDI mechanisms to the measured photoelectron spectra. While sequential double ionization is energetically not allowed . We will discuss We find that the predominant mechanism is a semi-sequential one, which involves the excitation of a window resonance. The ~ 8 fs lifetime of the intermediate autoionizing state increases the double ionization probability by a factor of $\approx$ 100 compared to non-sequential double ionization [3]. However, the results further indicate the contributions of conventional sequential and non-sequential double ionization.

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Using nanostructured dielectric surfaces for the generation of nano-focused XUV radiation

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Synopsis :We introduce an active XUV optical element that simultaneously generates and focuses isolated XUV light to the nanoscale. By illuminating an MgO nanostructured surface with an 800 nm femtosecond laser in transmission, we achieve coherent XUV generation and tight focusing (NA ≈ 0.35). However, IR contamination hampers applications. We suppress it by using p-polarized light at Brewster's angle and optimizing MgO nanofabrication. Our method overcomes XUV optics limits, aiming for <100 nm focal spots for high-resolution imaging, laser writing, and photoelectron applications.

High-resolution nanoscale imaging and metrology require precise generation and control of short wavelength light like extreme-ultraviolet (XUV) raditaion. A key challenge is the lack of suitable XUV optics. One approach to overcome the limitation is in-situ XUV generation via high-harmonic emission from a nanostructured MgO surface [1,2].

Previously, we demonstrated nanoscale XUV focusing using Fresnel zone plates (FZPs) patterned on MgO [2]. These 30 × 30 μm² FZPs achieve NAs of 0.18 (80 μm focal length) and 0.35 (40 μm focal length), focusing the 7th harmonic (114 nm) to a 150 nm waist radius with 18% efficiency. However, infrared contamination limits applications.

To suppress IR at the focus, we propose Brewster-angle (60°) illumination. Preliminary results show that 1D and 2D gratings illuminated at Brewster's angle generate and diffract XUV light (Figure 1), albeit with reduced diffraction efficiency and altered harmonic power. Further studies are underway. We are also developing a fabrication process for Brewster FZPs using electron-beam lithography and etching techniques.

Future advancements could enable direct laser writing, compact XUV sources, and nanoscale XUV microscopy.

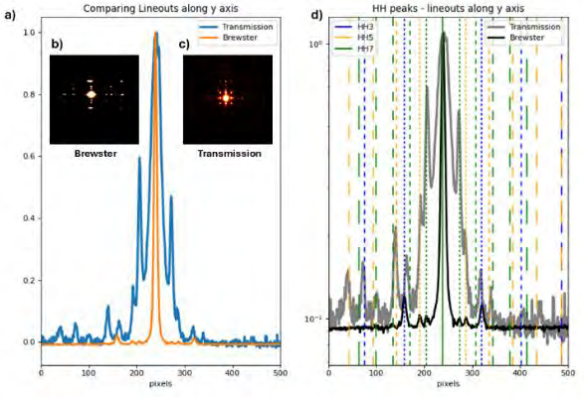


Figure 1. (a) Normalized y-axis lineouts for Brewster and Transmission geometries. (b,c) Diffracted XUV light from a 2D grating in both geometries. (d) A zoomed-in view of (a), highlighting diffraction peaks corresponding to different harmonic orders (HH). Vertical lines mark Brewster peak positions, with slight misalignment due to sample-to-MCP distance differences.

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Efficient water-window isolated attosecond pulse generation with tailored sub-cycle fields

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Synopsis Tailored sub-cycle optical fields driving high-harmonic generation, yield an eightfold efficiency increase in water-window isolated attosecond pulse generation with respect to a 1.5-cycle infrared field. Direct waveform measurement in the nonlinear gas medium uncovers the mechanisms behind the enhanced attosecond emission.

Isolated attosecond pulses (IAPs) spanning the water-window range (284–543 eV) enable site- and element-specific tracking of electronic dynamics in organic systems. However, when generated via high-harmonic generation (HHG), the low conversion efficiency limits their applicability. Tailoring the driving optical waveform is predicted to enhance this efficiency [1]. Here, we demonstrate such enhancement using sub-cycle fields centered at 1.4 μm, delivered by a Ti:Sa-based parametric waveform synthesizer (PWS) [2]. The PWS combines few-cycle, CEP-stable near-infrared (NIR, 0.8 μm) and infrared (IR, 1.8 μm) pulses. Waveforms are tailored by adjusting the relative delay (RD) between these pulses and applying a common CEP shift.

Water-window HHG is driven by the IR or synthesized (NIR+IR) field, both with similar energies (≈ 0.2 mJ). In either case, the beam is focused (f = 400 mm) into a 1-mm thick He gas cell. For synthesized fields, the NIR focused peak intensity is 37% of the IR (≈ 0.4 PW·cm^{-2}). Phase-matching is optimized only for IR-driven HHG by adjusting the backing pressure (6 bar) and gas cell position, and not for tailored drivers. The emitted soft X-rays are imaged onto a CCD grating spectrometer after metallic filters block <200-eV light. Driving waveforms are characterized at the He target using TREX [3], while HHG yields are measured with a calibrated photodiode. The efficiency (output energy/input energy) is derived from the HHG yield at the source and the input energy of the driving beam.

Figure 1 shows the conversion efficiency across the water-window range versus the RD (negative RD means the NIR precedes the IR). The blue horizontal line at 2.9×10^{-11} marks the IR-driven efficiency, optimized by tuning the IR field's CEP. At RD = 0.5 fs, the tailored waveform (inset) boosts the efficiency to 2.4×10^{-10}, an eightfold increase over the IR-only case.

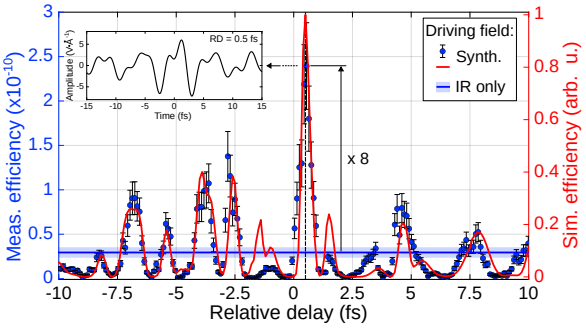


Figure 1. Water-window HHG efficiency vs. RD. Left axis: Measured for synthesized (blue circles) and IR-only fields (blue line, shading = error). Right axis (red): simulated with synthesized fields. Inset: Measured tailored field at RD = 0.5 fs.

Macroscopic HHG simulations using the measured driving waveforms (IR-only or synthesized) attribute this enhancement mainly to a higher ionization rate by the tailored field and a larger short-trajectory contribution. They also indicate water-window IAP generation with both drivers. This is supported by a strong π-periodic, CEP-dependence in measured spectra.

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Optimizing polarization gated generation of isolated attosecond pulses without CEP stabilization

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Synopsis We study polarization gating with 7 fs pulses, 800 nm pulses operating at 5 kHz without CEP (Carrier envelop phase) stabilization. With a polarization gate of duration $\tau_g \sim 0.7$ fs, the emission is so confined that the XUV generation is only significant for those CEP where isolated attosecond pulses are predominant and much lower otherwise. The observed continuous XUV spectra are compatible with the emission of isolated attosecond pulses and we observe that the XUV flux can be increased by using an asymmetric polarization gate that locates the gate in the falling front of the pulse.

Polarization gating [1-3] relies on the strong sensitivity of high order harmonic generation (HHG) in gases on the ellipticity of the driving pulse. Modulating the polarization of the driver during the pulse confines the XUV emission inside a polarization gate where the ellipticity is near zero. The gate duration, τ_g, can be controlled down to the sub half optical cycle of the fundamental, $T_0/2$. Isolated pulses can be generated with CEP stabilized laser when τ_g gets smaller than the spacing between two attosecond pulses $\sim T_0/2$. The single atom harmonic central frequency changes with CEP is smaller than the harmonic separation when the gate is located near the maximum of the pulse. Therefore, for strong confinement, the XUV spectra are strongly related to the temporal confinement when propagation effects can be neglected [2]. With random CEP and short polarization gate, continuous spectra cannot be due to harmonic shift as opposed to other confinement approach such as intensity gating [4]. With $\tau_g < T_0/2$, the number of attosecond pulses emitted in the gate and their amplitude is controlled by the CEP [4]. For $\tau_g < T_0/4$ the efficiency gets very dependent on the CEP. It is maximum for those CEP that leads to isolated attosecond pulse generation and strongly reduced when two pulses are generated. With random CEP, XUV emission is only efficient for those CEP where isolated attosecond pulses are generated.

We investigated this situation with $\tau_g = 0.7$ fs $\sim T_0/4$ for a laser centered near 800 nm, post-compressed down to 7 fs. Without gate, the harmonics generated in Ar cell are broad and discrete. With $\tau_g = 0.7$ fs polarization gate, we observed a reduced signal and continuous spectra (Figure 1) compatible with the generation of isolated attosecond pulses. Spectral changes were moderate with the gas pressure and did not show any significant harmonic shift. The spectra remained continuous in the cutoff for all pressures implying that the continuum is mainly due to the strong temporal confinement. We observed that the number of XUV photons is not maximum when the gate is centered. It can be increased by moving the polarization gate [5] in the falling front of the pulse.

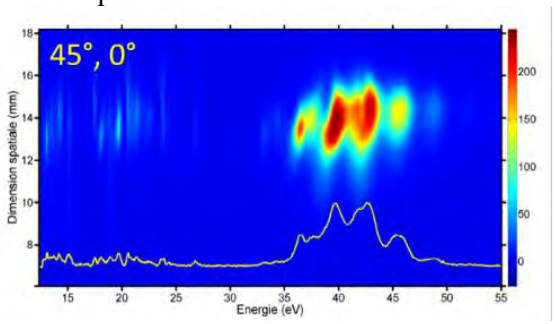


Figure 1. Spatially resolved XUV spectra obtained with a polarization gate with duration $\tau_g = 0.7$ fs..

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Direct measurement of the dipole phase in solid-state high-harmonic generation via XUV attosecond interferometry

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Synopsis We directly measure the intensity-dependent dipole phase of high-harmonic emission from solid MgO via ultrastable XUV interferometry. We reveal a linear relationship between the dipole phase and intensity. The results are supported by theoretical calculations (analytical, 2-band SBEs, and full band-structure SBEs in Wannier basis), enabling signal decomposition into inter/intraband and orbital-resolved currents. Our findings provide the first rigorous description of the intensity-dependent phases essential for phase matching as well as for divergence and focusing properties of solid-state HHG.

Solid-state high-harmonic generation (HHG) is rapidly emerging as a promising route to compact, coherent extreme ultraviolet (XUV) sources, and as a probe of nanostructures and attosecond material dynamics [1]. Efficient HHG and optimized XUV beam focusability depend critically on the spatial coherence and the wavefront of the emitted harmonics, which is fundamentally governed by the dipole phase. While well-established in gas-phase HHG [2, 3] and identified in solid-state HHG [4], the quantification of the dipole phase in solids has not been fully carried out. Solids, influenced by their electronic band structure, exhibit complex laser-induced intra/interband dynamics and multi-band contributions [5], impacting the dipole phase in a multifaceted way.

We utilized an ultrastable, birefringent common-path interferometer driven by an 800 nm laser to study the dipole phase in a magnesium oxide (MgO) crystal. Our approach allows us to directly measure the intensity and frequency-dependent dipole phase in MgO for several harmonic energies (Figure 1), isolate nonlinear effects, and validate the physical origins of the harmonic phase shifts. We interpret the results with an analytical semi-classical model, numerical simulations based on 2-band semiconductor Bloch equations, and a full band-structure framework in Wannier basis. This allows us to identify a dominant contribution from the 1st valence and 1st conduction band to the dipole phase of MgO in the XUV range, which coincides with the independently verified O: 2p and Mg: 3s orbitals as main real-space contributors. This is fully reproduced by an analytical theory which allows incorporating the effects of the dipole phase in e.g. phase matching or focusing simulations.

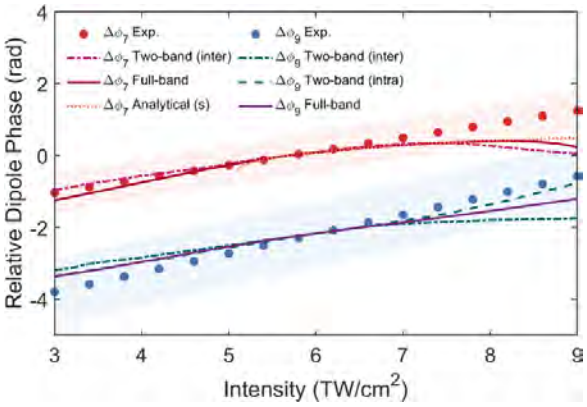


Figure 1. Intensity-induced relative dipole phase results for harmonics 7 ($\Delta\phi_7$) and 9 ($\Delta\phi_9$) in MgO in comparison with the analytical semi-classical, numerical 2-band, and full band-structure theoretical calculations.

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Attosecond chemistry: tutorial and theoretical perspectives

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Synopsis This presentation will be a tutorial about attosecond chemistry, a field born of the application of attosecond domain pulses to molecules. I will present the possibilities offered by the enhanced time resolution and large bandwidth of such pulses onto the study of polyatomic molecules and chemical reactions. I will also discuss how attosecond science could revolutionize photochemistry, and allow a control of the outcome of photochemical reactions, which is one of the main current challenges in attochemistry.

In 2001, the first attosecond domain (1 as = 10^{-18} s) pulses were generated [1]. Their application to molecules has given birth to the field of attosecond chemistry, also called "attochemistry" [2,3]. Thanks to the enhanced time resolution, attosecond domain pulses allowed for the first time the observation of electrons on their intrinsic dynamics time scale during various fundamental processes such as ionization or chemical reactions [3].

Because of the time-energy uncertainty principle, attosecond and subfemtosecond pulses have a large spectral width and therefore populate several electronic excited states of a molecule in a simultaneous and coherent manner; this is called an "electronic wavepacket". An electronic wavepacket is a non-stationary state. Interference between the electronic states in such a superposition alternates between constructive and destructive and leads to an oscillating motion of the electronic density, often referred to as "charge migration" (see Figure 1). The coupling between electrons and nuclei leads to electronic decoherence and damping of the oscillating motion in the electronic density.

From a chemical point of view, the absorption of light excites the electrons of a molecule, electrons being the particles that form the chemical bonds. As a result, the distribution of electrons and thus the chemical reactivity of the molecule in these electronically excited states differ significantly from the ones in the ground state. Thanks to this conceptually simple yet complex process, photochemistry has considerably broadened the spectrum of possible reactions, as compared to thermal chemistry. Yet, photochemical reactions are limited by the nature and finite number of electronic excited states in molecules.

By interference, the electronic distribution of an electronic wavepacket excited by an attosecond pulse is not the simple average of the individual electronic distributions of the different states: an electronic wavepacket is a new type of initial electronic state, with a new nature. What would be the reactivity of a molecule in this new electronic state? With the technological advances of attoscience, the concept of attochemical control, also called "charge-directed reactivity" has been demonstrated experimentally and theoretically mostly on diatomic. Although highly challenging, extending this to chemical reactions in polyatomic molecules is one of the current ambitious goals of attochemistry [4].

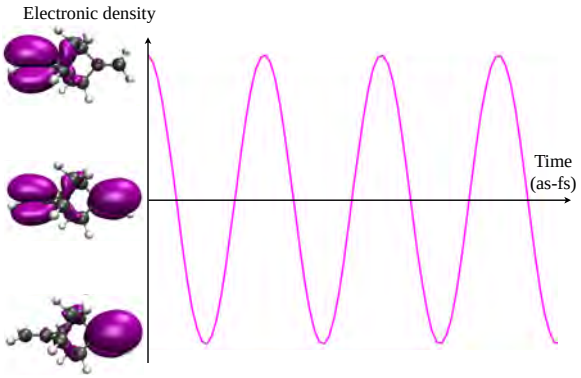


Figure 1. Oscillating motion in the electronic density of a molecule excited to an electronic wavepacket by an attosecond domain pulse.

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Advances in ultrafast ultraviolet spectroscopy of molecules

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Synopsis Recent advances enabled by our broadband UV light sources will be discussed, spanning applications from the gas to the liquid phase. Particular emphasis will be placed on our investigation of coherent electronic responses in photoexcited chiral molecules. We have observed the rapid dynamics of Rydberg wavepackets leading to the formation of transient chiral currents. Preliminary experimental results will be presented and suggest that these currents can contribute to chiral-sensitive directional fragmentation, pointing to a route for enantioselective photochemical control in neutral systems.

The development of few-femtosecond (fs) deep-to-vacuum ultraviolet (UV) light sources is opening up a new frontier in ultrafast spectroscopy, granting access to electronic and nuclear dynamics in neutral matter with high temporal resolution. These laser-driven sources allow for perturbative excitation below the ionization threshold, making it possible to initiate and track ultrafast processes in a regime that was previously difficult to access. In this context, we have synchronized our broadband UV technologies [1,2] with near-infrared (NIR) and attosecond extreme ultraviolet (XUV) pulses [3]. This versatile configuration enables a series of pump-probe experiments to investigate UV-induced dynamics with femtosecond-to-attosecond time resolution.

I will discuss our recent studies on photoinduced electron dynamics in chiral molecules and their implications for ultrafast enantioselective molecular control [4]. Photoelectron circular dichroism measurements in methyl lactate molecules revealed a rapid and periodic inversion of the chiroptical response within 10 femtoseconds after prompt UV-excitation, which is attributed to the formation of coherent electronic superpositions of Rydberg states. These results demonstrate the sensitivity of our chiral observable to electronic coherence, highlighting the role of electron dynamics in shaping the photoinduced molecular chiral response. Simulations indicate that the emerging chiral currents can drive the fragmentation direction during dissociative ionization in a manner that depends on the molecular handedness. I will present preliminary experimental results that support the feasibility of this approach for achieving enantioselective charge-directed reactivity.

In addition to these chiral-sensitive studies, I will briefly discuss recent experiments that mapped the electronic and vibrational dynamics of both gas and liquid samples. This includes a study of dissociative dynamics in iodomethane [5].

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Probing iodine chemical environment with ionization delays

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Synopsis Ionization delays have been measured in a set of iodine-containing molecules following photoemission from the iodine 4d core levels. We evidence the influence of the iodine chemical environment on the ionization delays by measuring significant relative delays between iodomethane (CH_3I), iodoethane (C_2H_5I), iodopropane (C_3H_7I) and iodobenzene (C_6H_5I). Strikingly, a delay difference of up to 131 ± 12 as is found between the two structural isomers of iodopropane. The localization of the initial orbital on a single atom thus reveals subtle differences in the molecular potential.

Photoionization delays result from the scattering of the outgoing electron on the atomic/molecular potential and thus provide a fine probe of the potential landscape. While ionization delays studies have been performed in readily accessible valence molecular shells [1, 2], one can now reach core shells of high-Z atoms, such as iodine 4d levels, with XUV attosecond pulses generated by High Harmonic Generation (HHG). These core levels are of particular interest as they act as localized electron sources on one particular atom in the molecular frame.

Molecular core shell ionization delays have been recently measured using the streaking technique with both HHG [3, 4] and XFEL [5] sources. Our approach is based on RABBIT (Reconstruction of Attosecond Beating By Interference of two-photon Transitions) spectroscopy, allowing to explore a wide energy range with high spectral resolution. In order to measure reliably the expected small delay differences and to avoid spectral congestion, we used two electron spectrometers in a two-foci configuration [6], in which two RABBIT spectrograms are measured simultaneously in parallel, one probing the molecules of interest, the other acting as an absolute timing and XUV spectral phase (attochirp) measurement in the neon reference gas.

We were able to measure significant relative delays between different iodine-containing molecules such as CH_3I, C_2H_5I, C_3H_7I and C_6H_5I. The present results reproduce previous measurements on iodoethane [4]. However, the theoretical prediction in [4] of an increase of the delay with the size of the carbon chain is observed only between iodomethane and iodoethane, and breaks completely for the larger carbon chains. A most remarkable effect is observed on the two structural isomers of iodopropane (1- and 2-iodopropane) with a delay difference reaching up to 131 ± 12 **as**. Such large difference was unexpected given their very small chemical shift difference, demonstrating the great sensitivity of our observable on the chemical environments.

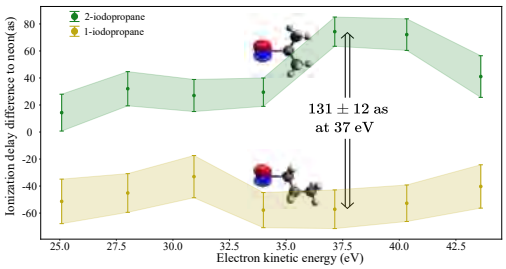


Figure 1. Ionization delays of the 4d electrons for the two iodopropane isomers with respect to neon.

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Probing the ultrafast vibronic wavepacket dynamics of ethylene with sub-4fs VUV pulses

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Synopsis The ultrafast relaxation dynamics of ethylene and its deuterated isotopologue following photoexcitation at 158 nm and 175 nm were investigated using time-resolved photoelectron spectroscopy with sub-4fs VUV pulses generated via resonant dispersive wave generation in a hollow-core fiber. Our measurements, supported by ab initio calculations, reveal a significant population transfer from the ππ* to the σπ* state within the first 20 fs after pump laser excitation. This study opens new avenues for exploring ultrafast molecular dynamics in the VUV spectral range with unprecedented time resolution.

Ethylene is the smallest unsaturated hydrocarbon molecule, featuring a carbon-carbon double bond. The ultrafast relaxation dynamics of ethylene, following vacuum ultraviolet (VUV) absorption (140 nm-175 nm) to its ππ* first valence electronic excited state, has been extensively studied both theoretically [1] and experimentally [2]. It is generally understood that within the first 25 fs after photoexcitation, ethylene undergoes ultrafast C=C stretching and torsional motion around the carbon double bond, eventually reaching a twisted-orthogonal structure where the two CH_2 groups are rotated by 90°. The molecule can then relax back to the ground electronic state through various conical intersections. Several studies have also highlighted that in the vicinity of the ππ*state, the π3s Rydberg state could influence the relaxation dynamics.

In previous experiments, VUV pulses with durations exceeding 10 fs were used, which prevented a complete characterization of the vibronic wavepacket dynamics occurring in the coupled ππ*-π3s potential energy surfaces of the molecule. In our study, we employed a newly developed VUV source that provides tunable, sub-4 fs pulses in the wavelength range from 150 nm to 200 nm. These pulses were generated through resonant dispersive wave generation inside a hollow-core fiber [3,4]. This allowed us to revisit the ultrafast photoexcitation dynamics of ethylene (C_2H_4) and its deuterated isotopologue (C_2D_4) using time-resolved photoelectron spectroscopy. Experiments were conducted at central wavelengths of 175 nm and 158 nm.

Our experiment, supported by high-level simulations, enabled us to track the entire vibronic wavepacket dynamics within the first 50 femtoseconds following laser excitation (see Fig. 1). In particular, we show that the early timescale dynamics are influenced by a strong coupling between the initially populated ππ* excited state and the σπ* state along the torsional motion. Notably, the σπ* state has not been previously considered in the photoexcitation dynamics of this molecule, yet our investigation reveals that it plays a critical role. Specifically, our study shows that the σπ* state actively participates in the torsional motion of the molecule as it approaches the conical intersection with the ground state.

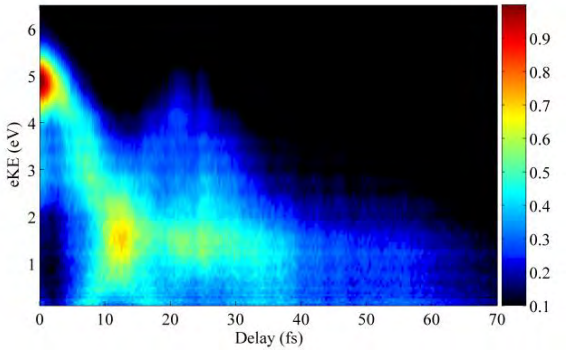


Figure 1. Time-resolved photoelectron spectrum recorded in ethylene using sub-4fs VUV pulse at a central wavelength of 158 nm .

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Strong electron-electron-nuclei correlations in multiphoton single and double ionization

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Synopsis This talk presents recent theoretical developments to describe multiphoton single and double ionization processes. A top-to-bottom approach is taken to first present recent applications exploring photionization time delays on small biomolecules, and our recent progresses for an accurate description of two-photon double ionization of H_2, where we have found novel intereferences arising from to strong electron-electron-nuclei interactions.

Electron dynamics in matter can be traced with attosecond resolution by using available coherent light sources. Most attosecond experiments to date have used attosecond pulses produced via high-harmonic generation (HHG). For many years, the low repetition rates of these sources limited the observation of non-linear phenomena in matter, although efforts are focused to overcome this limitation. Alternatively, free electron laser (FEL) facilities generate high brilliance and high intense pulses with a high frequency tunability, and, more recently, even producing coherent attosecond pulses. The combination of these intense XUV sources with advanced detection devices that enable coincident measurements of all charged fragments enables a complete dynamical characterization of non-linear phenomena in the XUV and X-Ray regimes that remained experimentally inaccessible until now [1]. This technological progress thus calls for accurate theoretical methods able to predict and interpret these XUV-induced non-linear phenomena and unravel the role of electron correlation in the excitation and ionization process [2]. Recent works on XUV-IR schemes on biomolecules and small molecules (e.g. N_2O, C_2H_2) will be discussed [3]. This talk will also present our new implementation for a full dimensional solution for the two-photon double ionization of the H_2 molecule. Very few theoretical works addressed this problem due to the difficulty and computational cost of achieving an accurate evaluation of the strong correlation between all fragments in the four-body Coulomb breakup, and only frozen-nuclei approaches were employed until now [4]. In contrast with our previous methods, which were very successful for H_2 single ionization problems including autoionization [5], but unable to accurately describe double ionization, in this work, we have developed from scratch a new tool to describe the multiphoton double ionization of H_2 including electronic and nuclear degrees of freedom on an equal footing, i.e., working beyond the Born-Oppenheimer approximation. We employ a numerical representation of the molecular wave function in a basis set of FEM-DVR (finite elements combined with a discrete variable representation) and apply an exterior complex scaling procedure to impose the appropriate many-body Coulomb boundary conditions [6,7]. Accurate angle and energy differential two-photon double ionization yields show significant energy displacements in the photoelectron spectra with respect to frozen nuclei approaches. More interestingly, counterintuitive angularly resolved double ionization yields with respect to its atomic analog are found, due to novel interferences that arise from sequential two-photon absorption paths through different cationic states [7].

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Attosecond Coherent Control and Measurement of the Photoionization Dynamics of Chiral Molecules

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Synopsis Here we present the generation, characterization, and application to chiral molecules of circularly polarized attosecond extreme-ultraviolet pulses created by non-collinear high-harmonic generation and detected by ion-electron coincidence spectroscopy. We measured the forward-backward time-delay asymmetry of chiral molecules, which is modulated by the coherent states of the light pulses.

We introduce chiroptical spectroscopy with attosecond pulses and demonstrate attosecond (as) coherent control over photolelectron circular dichroism (PECD), as proposed in [1, 2], as well as the measurement of chiral asymmetries in the photoionization delays of chiral molecules. We have developed a compact plug-in apparatus for circular non-collinear high-harmonic generation [3], and we have introduced a method named "Stokes-parameter and attosecond-resolved reconstruction of optical waveforms" (SPARROW) in order to characterize the ellipticity of the generated extreme-ultraviolet (XUV) attosecond pulse trains [4]. These techniques enable us to conduct the reconstruction of attosecond beating by interference of two-photon transitions (RABBIT) experiment using circular XUV pulses with a circularly or linearly polarized infrared (IR) dressing field. We measured the phase-resolved PECD of the *R*- and *S*-methyl oxirane (propylene oxide, C_3H_6O). Our results show that co-rotating attosecond and IR pulses can nearly double the PECD compared to single-photon ionization. Our experiments further reveal a surprisingly large forward-backward chiral asymmetry of the photoionization delays ranging up to 240 as, which contain an asymmetry of ∼60 as originating from the chirality of the continuum-continuum transitions, as shown in Fig. 1. Attosecond chiroptical spectroscopy opens the door to understanding and controlling the dynamics of chiral molecules on the electronic time scale.

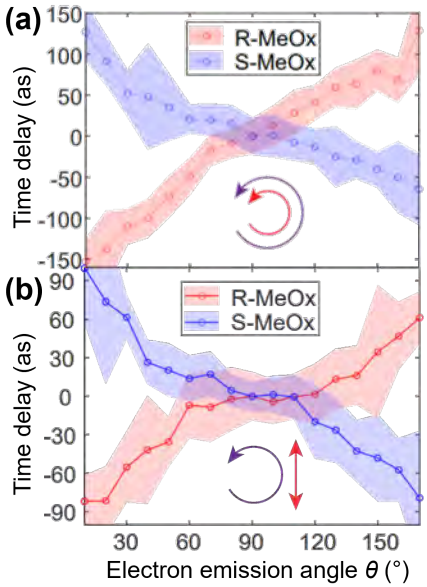


Figure 1. Angle-resolved photoionization time delays or *R*− (red) and *S*− (blue) methyl oxirane (MeOx) referenced to $\theta = 90°$, where θ is the angle between electron emission and light propagation. The polarization of the dressing IR is co-rotating with the XUV in (a) and linear in (b).

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Control of Photo-Ion Circular Dichroism Using Orthogonal Laser Beams

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Synopsis We present the results of photo-ion circular dichroism measurements of the chiral molecule fenchone using orthogonally propagating laser pulses of different colours. This configuration allowed us to show how differential ionization from opposite enantiomers can be influenced by controlling the direction of the linear polarization in one of the beams. We interpret this as a rare demonstration of the orientation dependence in the circular dichroism of isolated molecules.

Absorption circular dichroism (CD) has been studied since the 19th century but remains a key tool for studying chiral molecules. The more recently explored but related chiroptical technique of photoion circular dichroism (PICD) is manifested in the differences in photon yields from left and right circularly polarized laser pulses.[1] As PICD arises from the transition moments beyond electric dipole contributions, the asymmetry in the ion yields is typically low (≈ 0.1%). If these could be significantly enhanced, it would open up the possibility of using all-optical methods for processing chiral molecules such as enantiomeric purification.

In the present study, we have used orthogonal laser pulses of different wavelengths (260 and 520 nm) to ionize fenchone enantiomers with the resulting ions being analyzed in a time-of-flight mass spectrometer (Fig. 1). The yields of the parent and major fragment ions were measured for different polarizations configurations of the two pulses. Our results show that the difference in yields obtained when the 260 nm pulse is switched between left and right circularly polarized, is influenced by the presence of a linearly polarized 520 nm pulse and controlled by its direction (parallel or perpendicular to the plane of the 260 nm polarization).

This indicated that the 520 nm pulse was selectively exciting certain orientations of the fenchone molecules which have their transition moment aligned with the polarization direction. Subsequent absorption by the circularly polarized 260 nm pulse demonstrated the orientation dependence of the CD of these transitions, thus boosting the asymmetry.[2]

This chiroptical control of photoionization, could be further enhanced by using more exotic polarization states. There is great interest in creating locally chiral laser electric fields using multiple non-collinear beams which in principle would generate chiroptical effects for integrated yields within the dipole approximation.[3] This experimental setup provides a possible pathway for realizing these ambitious goals.

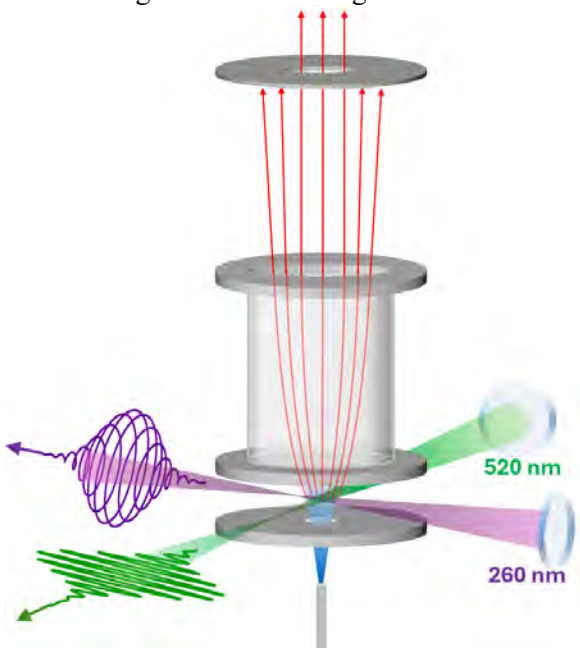


Figure 1. Schematic of the experimental apparatus used for the measurements. An effusive gas jet of fenchone is directed into a time-of-flight mass spectrometer where it is intersected by spatially and temporally overlapped 300 fs laser pulses at wavelengths of 260 nm and 520 nm.

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All-optical coherent control of chiral electronic transitions for highly enantioselective photochemistry using femtosecond lasers

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Synopsis We introduce an approach to drive highly enantioselective electronic population transfer in randomly oriented samples using a ~20 fs IR laser pulse and two harmonics, without intermediate resonances. This approach addresses electronic transitions, is relevant in liquids, and across interaction regions spanning many laser wavelengths, even with dispersion. Our ab-initio simulations in carvone yield ~30% enantioselectivity (10^3-fold enhancement over circularly polarised light) in a state leading to isomerization. This brings all-optical highly enantioselective photochemistry within reach.

Circularly polarized light (CPL) induces different populations in left- and righ-handed versions of randomly oriented chiral molecules. Such differences lead to differences in the product yields of photochemical reactions. Thus, CPL triggers all-optical enantioselective photochemistry. But the difference is usually below 0.1%, rendering CPL impractical for photochemical applications. Therefore, photochemistry still invariably relies on chemical (instead of all-optical) methods to control enantioselectivity. A practical all-optical alternative would allow unprecedented temporal and spatial control over the enantioselectivity of photochemical reactions. Beyond applications like enantiomeric enrichment, such control would also enable new technologies, like enantiomeric switches [1], which are currently hindered by CPL's low selectivity.

Here we introduce two schemes (Fig. 1) to enantioselectively control the excited state population of electronic states in chiral molecules. Our proposal [2] can be realized using readily available fs technology [3], requires neither all-resonant transitions nor long electronic coherence times, and yields enantioselectivities close to 30%. Our findings are supported by analytical theory revealing the role of light's polarization in both schemes, and by ab-initio simulations.

In the 1- vs 2-photon scheme, we find that the enantioselectivity oscillates on the scale of the fundamental wavelength. Thus, maintining enantioselectivity requires contraining the interaction region accordingly, e.g. using appropriately oriented thin flat liquid microjets. We find that the oscillation of the enantioselectivity across the interaction region can be avoided by using the 2- vs 3-photon scheme in Fig. 1b. Including liquid water dispersion yields oscillations on the scale of 50 μm, which can be easily addressed.

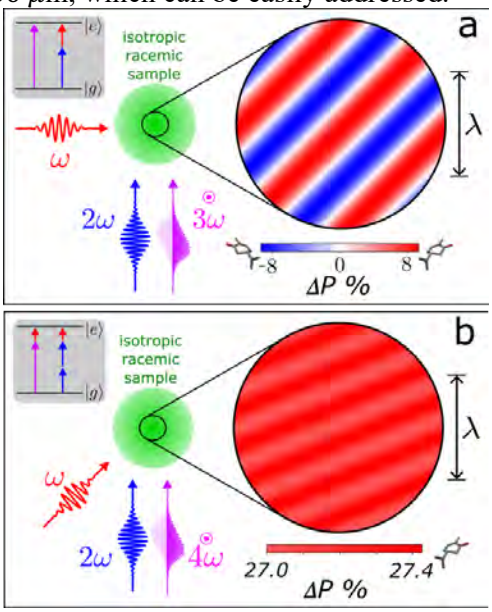


Figure 1. Excitation schemes for highly enantioselective population transfer. Color plots show the difference in the population of the first excited electronic state of S and R enantiomers of carvone across the interaction region as obtained by numerical integration of the time-dependent Schrödinger equation. $\lambda = 2\pi c/\omega = 956$ nm and 1.56 μm in a and b, respectively.

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Attosecond time-resolved electronic and vibrational dynamics in furan

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Synopsis We report on our joint experimental-theoretical work on attosecond carbon K-edge spectroscopy to trace the ultrafast non-adiabatic relaxation dynamics of furan induced by an intense light field. The high temporal resolution in combination with the broad coherent energy spectrum allowed tracking transitions along several conical interactions and identifying nuclear and electronic wavepacket dynamics. We here present our multi-physics approach to simulate these very different dynamics and calculate corresponding spectroscopic observables.

Ultrafast non-adiabatic dynamics lies at the heart of photochemical reactions in many biologically and chemically important systems. Investigation of these dynamics necessitates spectroscopic tools able to resolve the interplay of electron and nuclear degrees of freedom on these ultrafast timescales. Attosecond time-resolved x-ray absorption spectroscopy (as-XAS) meets these challenges. We demonstrate this on the prototypical furan molecule [1].

Experimentally, furan was strong-field excited by a 17-fs pump with a photon energy of 0.7 eV, followed by a time-delayed as-probe pulse with photon energies spanning the carbon K-edge region (near 285 eV).

For the theoretical description, we simulated the strong-field excitation by means of real-time TDDFT. The subsequent non-adiabatic relaxation dynamics was described by on-the-fly surface hopping dynamics. X-ray absorption spectra for specific nuclear and electronic configurations were calculated on the frozen-core equation-of-motion model on the coupled cluster singles and doubles level of theory.

This combined experimental-theoretical approach showed that initially, furan is excited from the ground state (S_0) into the $\pi\pi^*$ state (S_2). The now singly occupied molecular π orbital (SOMO) can be clearly identified as a new peak in the pre-edge region of the XAS spectra. Subsequently, ultrafast relaxation proceeds via a conical intersection into the S_1 state. The corresponding coherent electronic wavepacket dynamics manifests itself as quantum beats in the XAS spectra, which were theoretically predicted [2] but have never been experimentally observed before. Subsequent passages through conical intersections and the change in the character of the excited electronic state into a dissociative $\pi\sigma^*$ state leads to C-O bond breaking. The accompanying dephasing of the electronic wavepacket, the C-O stretching dynamics and the resulting symmetry breaking can be clearly identified in the XAS spectra, as the SOMO peak splits into four peaks for the now four symmetry-nonequivalent carbon atoms in the molecule. Finally, the system relaxes back into the electronic ground state on a longer timescale.

Our work shows that attosecond x-ray absorption spectroscopy is a powerful tool to trace the complex light-induced electronic and nuclear dynamics in complex molecular systems.

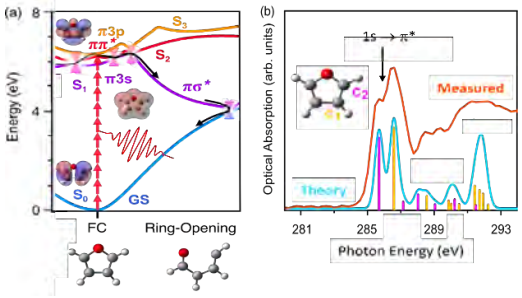


Figure 1. a) Potential energy surfaces. (b) Static (nonexcited) XAS of furan around the carbon edge.

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Attosecond dynamics in molecules: probing ultrafast charge motion with EUV/IR and UV/EUV spectroscopy

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Synopsis The investigation of ultrafast processes initiated in molecules by light absorption is crucial across molecular physics, materials science, chemistry, and biology. They are at the heart of emerging technological applications, where photo-induced electron transfer and charge transfer play a key role. Despite advances in ultrafast science, the interplay between electronic and nuclear motion after prompt excitation remains poorly understood. This study explores ultrafast electronic dynamics in molecular structures using attosecond-pump/IR-probe and few-cycle UV-pump/attosecond-probe spectroscopy.

Photoinduced charge transfer (CT) is a fundamental process that governs a wide range of natural phenomena and technological applications, such as photosynthesis, photooxidation, molecular damage, and electronic transport in semiconducting polymers, molecular wires, junctions, and photovoltaic devices. Despite significant progress in ultrafast science, a complete understanding of the initial stages of CT remains elusive. Specifically, the complex coupling between electronic and nuclear motion immediately after prompt excitation has yet to be fully characterized [1]. Gaining deeper insights into these ultrafast dynamics is crucial, not only for advancing our understanding of charge transport in complex systems but also for driving innovation in energy conversion, optoelectronics, and molecular-scale electronics.

Ultrafast charge dynamics can be initiated by extreme ultraviolet (EUV) attosecond pulses or by ultrashort pulses in the ultraviolet (UV) spectral region. In the former case, the molecular system undergoes ionization, enabling attosecond techniques to directly probe the electronic dynamics of molecular cations. In contrast, excitation with UV pulses allows the study of coupled electronic and nuclear dynamics in neutral molecules. To achieve high temporal resolution in this context, UV pulses must consist of only a few optical cycles.

This study investigates both experimental approaches. Attosecond EUV-IR photoion spectroscopy reveals the interplay between structural and electronic dynamics in a donor-acceptor molecule, providing a unified framework for interpreting experimental data [2]. Our results show that CT does not occur as a simple, continuous displacement of charge between molecular sites. Instead, it begins with an ultrafast redistribution of charge density, displaying quasi-periodic behavior driven by structural modulations within the molecule. This complex interaction between electronic and structural dynamics offers crucial insights into the early stages of CT and furthers our understanding of its mechanisms.

A novel beamline for photoelectron spectroscopy will also be introduced, employing tunable UV pump pulses with durations below 3 fs and attosecond probe pulses. To accurately measure the temporal resolution of the beamline, an *in-situ* characterization of the instrument's response function was carried out using a two-color photoelectron cross-correlation measurement in argon. The extreme temporal resolution of the beamline has enabled the investigation of ultrafast electronic dynamics in a few molecules, which serve as benchmark systems for studying nonadiabatic dynamics, ultrafast internal conversion, and vibronic coupling. This high-resolution capability allows for detailed observation of rapid electron motion and precise tracking of electronic state transitions, providing a deeper understanding of the complex interactions between electronic and nuclear motions in molecular systems.

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Time-resolved x-ray absorption spectroscopy in thiophene covering sulfur L and carbon K Edges

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Synopsis Ultrafast dynamics in thiophene are revealed with unprecedented temporal resolution using time-resolved X-ray absorption spectroscopy. Two setups capture attosecond-resolved dynamics within 20 fs and dynamics up to 500 fs with few-femtosecond precision. Oscillations at 285 eV and 286.5 eV indicate rapid charge redistribution, while a 40 fs delay in signal buildup at carbon and sulfur suggests fast ring-opening. This dual-apparatus XAS study provides novel insights into thiophene's electronic and structural relaxation upon strong-field ionization.

Thiophene shows ultrafast excited-state dynamics upon UV excitation, which has been studied with 190-fs UV pulses [1]. Understanding the behaviour of thiophene monomers is a cornerstone for a comprehensive understanding of dynamics in polymers, which are promising candidates for energy-storing systems [2].

In this work, we studied the dynamics of thiophene induced by strong-field ionization in thiophene monomers in the gas phase by utilizing two different experimental endstations dedicated to time-resolved X-ray absorption spectroscopy in the soft X-ray (SXR) regime using table-top high harmonic generation (HHG) source [3]. One endstation enables studying early dynamics in a 20 fs time window with attosecond time resolution. Subsequent dynamics within the first 500 fs are revealed by the other endstation with few-fs resolution. The transient absorption (TA) spectra obtained from these experiments, Figure 1 A, unveil new insights into thiophene's ultrafast behaviour post-ionization. Notably, the averaged lineout features in Figure 1 B at 285 eV (orange) and 286.5 eV (blue) exhibit out-of-phase oscillations within the initial 10 fs. Fourier analysis of these oscillations revealed periods of approximately 2.3 fs and 3.2 fs, corresponding to energy separations of 1.8 eV and 1.3 eV, respectively. Furthermore, a delayed buildup of the TA signal at 282 eV, occurring 40 fs after the 286.5 eV feature, indicates the initiation of a ring-opening process in the thiophene molecule. This structural transformation underscores the intricate interplay between electronic excitation and nuclear motion. Additionally, transient features observed between 161 to 165 eV, which change within the first tens of femtoseconds, are identified as transitions involving sulfur $2p_{1/2}$ and $2p_{3/2}$ electrons. These features could help to obtain a sulfur-based local probe of the electronic and structural dynamics.

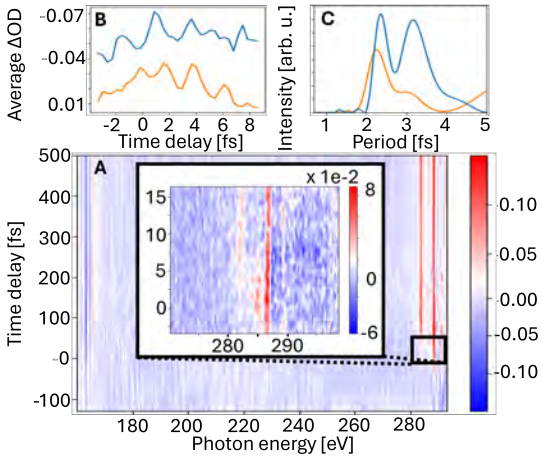


Figure 1. **A**: Temporal evolution of thiophene TA spectrum. Inset shows 20 fs delay range, with attosecond resolution. **B**: Average of the 285 eV (orange) and 286.5 eV (blue) line. **C**: Fourier transforms of averaged lines in **A** with same color code.

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Time-resolving molecular dissociation with FEL-pump and HHG-probe pulses

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Synopsis The coupling of electronic and nuclear motion on ultrafast time scales plays an important role in the understanding of photochemical reactions, in particular when tunneling events and electronic non-adiabatic couplings occur. Here we present the first FEL-pump-HHG-probe experiment to address such challenges in the dissociation of O_2 molecules, combining energy-release spectra of ionic fragments and XUV absorption spectroscopy.

We present an experimental scheme for investigating molecular dissociation dynamics based on the first-time combination of XUV-FEL-pump and XUV-HHG-probe pulses at the FL26 beamline at FLASH2 (DESY) [1]. In addition, the novel combination of transient absorption spectroscopy (TAS) with a reaction microscope (REMI) enables sensitivity to neutral fragments, electronic-state configurations, ionic kinetic-energy releases (KERs) and ionic coincidences at the same time [2].

The potential of this multidimensional all-XUV setup is demonstrated by studying the fragmentation of molecular oxygen from a specific excited state of the O_2^+ ion (Fig. 1A). The coupling of electronic and nuclear degrees of freedom, including a tunneling event, complicates the dissociation mechanism (Fig. 1B), but can be time-resolved with our novel scheme utilizing broadband HHG-probe pulses (Fig. 1C) on a 300 picosecond time scale [1]. Furthermore, the combination of independent, energy-tunable FEL-pump and broadband HHG-probe pulses, allows us to disentangle faster dissociation processes from different electronic and vibrational states. This will be illustrated by identifying the by-orders-of-magnitude different tunneling times of the two vibrational states of the O_2^+ ($c\ ^4\Sigma_u^-$) state. These findings are supported by the simultaneous ionic KER measurements with the REMI. The KER measurements of the O^+ fragments as a function of FEL-photon energy agree with previous findings for one-FEL-photon absorption processes [3], and further enable two-photon-induced dissociation mechanisms, as shown in [4].

In summary, our scheme allows the investigation of inner-shell electron dynamics and resulting molecular break-ups on a hundred-femtosecond time scale. In the future, the full potential of XUV-driven photochemistry is expected to be unlocked by tranfering our scheme to hetero-nuclear systems and to the few femto- and attosecond time scale by the use of advanced time tools.

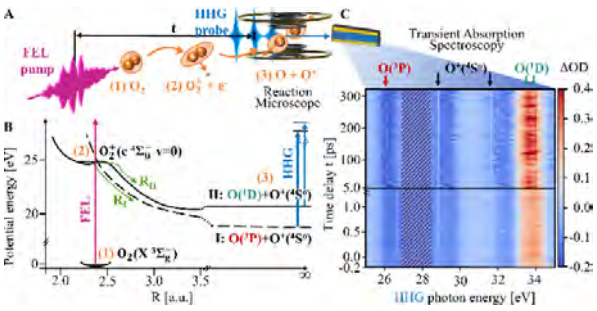


Figure 1. A Experimental scheme. **B** Potential-energy curves. **C** Time-resolved spectra. Figure adapted from [1].

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Observation of the Berry phase via attosecond interferometry

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Synopsis High harmonic generation (HHG) spectroscopy reveals fundamental light driven phenomena in condensed matter systems. In my talk I will focus on a novel manifestation of the Berry phase accumulated during the subcycle, field driven dynamics. By engineering an internal interferometer, we map this phase onto high harmonics. We further resolve the complex Berry phase accumulated during the tunneling process, opening new directions for strong-field metrology and topological physics.

HHG spectroscopy provides a unique probe of fundamental strong-field phenomena in condensed matter systems, resolving their evolution with subcycle accuracy. A key feature of HHG lies in its coherent nature: the coherent properties of the field-driven electronic wavefunction are mapped onto the optical properties of the emitted harmonics. Resolving this coherence is essential for reconstructing the internal dynamics, being one of the primary challenges in HHG spectroscopy.

In my talk, I will focus on how HHG can be applied to probe quantum geometric properties in condensed matter systems, focusing on the well-established Berry phase [1]. Since its discovery, the Berry phase has played a key role in a wide range of quantum phenomena. I will describe how the Berry phase can be accumulated through the subcycle strong field interaction [2]. By engineering an internal interferometer within a single optical cycle, we directly map this phase onto the high harmonic emission.

Building on this approach, we then extend our study to resolve the complex Berry phase accumulated during tunneling processes [3]. By precisely manipulating the tunneling barrier with attosecond precision, we directly observe the imaginary component of the Berry phase, accumulated as the electron tunnels during a fraction of the optical cycle. This work unifies strong-field light-matter interactions with geometric phase physics, expanding the frontier of strong-field metrology and providing a new platform for exploring geometric and topological quantum effects in condensed matter systems.

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Strongfield physics inside of (light-dressed) graphene and at the surface of metal needle tips

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Synopsis We will show (1) that strongly driven electrons inside of graphene allow us to measure band structure properties based on recording the electron's dynamical phase, and (2) that we can light-dress graphene and measure the anomalous Hall effect in a Floquet topological insulator state. (3) In photoemission from metal needle tips, we will show electron spectra resulting from intense quantum light and (4) demonstrate an optical nearfield effect leading to a strong CEP dependence of direct electrons.

The first part of the talk will focus on strongly driven electrons inside of graphene. When driven with intense NIR fields, electrons undergo coherent intraband motion and coupled interband transitions [1, 2]. The resulting dynamics can be understood in terms of Landau-Zener-Stückelberg-Majorana interferometry, where two coherent subsequent Landau-Zener transistions may lead to a closed interferometer [3]. Recently, we could demonstrate that we can use the dynamical phase an electron accumulates in the beam-split state to measure the slope of the bands [4].
The second part of the talk will show that we can light-dress graphene with a circular 1550nm light field to generate a Floquet Topological Insulator (FTI) state transiently. FTI states are highly interesting because of their topological properties and the associated protected transport properties. With a phase-locked second harmonic we could stroboscopically probe the FTI state. Most intriguingly, we could demonstrate the anomalous Hall effect, where the Berry curvature of the FTI state takes over the role of the magnetic field in the regular Hall effect. We could also show circular dichroism [5] and will report on the current state of the experiment.
The third part of the talk will deal with strong field physics at nanometric needle tips. We will show what kind of electrons spectra result when we focus intense quantum light in the form of bright squeezed vacuum (BSV) on these needle tips. The mean optical field of BSV is zero, yet its intensity can be so high that we can expect strong field physics to arise. Indeed, we observe high energy electrons, but no tell-tale recollision plateau. The plateau and a clearly visible 10 Up cut-off emerges only when we correlate the electron spectra with the photon number of each BSV's light pulse, i.e., when we postselect on the photon number [6].
Last but not least and now again with coherent few-cycle 1550nm laser pulses, we will show a strong CEP dependence in the *direct* electrons, an effect we call Nearfield-Induced Low Energy Stripes (NILES). It arises due to the electrons propagating inside of the spatially highly non-uniform optical nearfield in front of the needle tip.

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Subcycle band-structure videography of lightwave-driven graphene

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Synopsis We demonstrate for the first time subcycle band-structure videography directly accessing carrier dynamics over the entire momentum space with few-femtosecond temporal resolution. By combining momentum microscopy with strong phase-stable few-cycle mid-infrared and sub-10-fs extreme-ultraviolet pulses, we study lightwave-driven currents in graphene and their timing with the pump field. The 2D carrier distribution reveals the role of carriers and phonons in scattering dynamics, highlighting limits to fully coherent light-matter interaction.

Harnessing the carrier wave of light to drive electron dynamics in novel materials is the foundation of lightwave electronics [1,2]. To fully understand these phenomena on their ultimate timescales, timing these dynamics against the light transient over the entire momentum space is crucial. Subcycle angle-resolved photoemission allowed observing intraband currents [3] and light-driven bands [4], but only along specific directions and close to the origin of momentum space, missing out on key dynamics.

This work presents the next generation of subcycle band-structure videography. The unique combination of photoemission momentum microscopy with strong phase-stable few-cycle mid-infrared (MIR) transients and sub-10-fs extreme-ultraviolet (XUV, 21.7 eV) pulses guarantees access to field-driven dynamics over the entire Brillouin zone. In this work, we drive a graphene monolayer [5] with 27-THz pulses and probe the process with subcycle time resolution.

Focusing on a specific Dirac point, Fig. 1a shows subcycle currents driven by the pump field. Before the excitation (t = -70 fs), we observe the linearly dispersing Dirac cone. Closer to the peak field (t = -14 and 6 fs), the electron distribution changes: charges are accelerated away from the Fermi level and shifted along the Dirac cone. The instantaneous asymmetric occupation generates a field-driven current [3], while momentum streaking allows its direct comparison to the MIR waveform (Fig. 1b). Their relative subcycle delay (4.85 ± 0.21 fs) testifies to the ultrafast scattering processes dictating the timescales relevant for lightwave electronics,

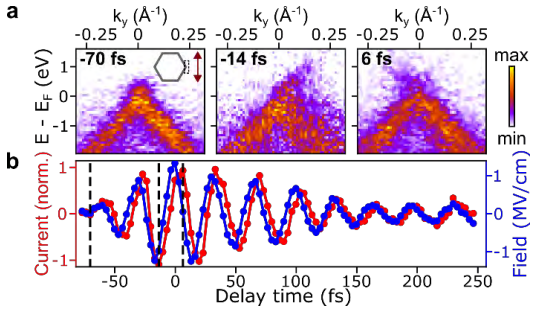


Figure 1. a, Band structure of graphene at the Dirac point before (t = -70 fs) and during (-14 and 6 fs) the interaction with the MIR field. The inset shows the Dirac point and the arrow marks the MIR polarization. **b**, Extracted current (red points) and MIR waveform (blue points). Black dashed lines mark the time instants shown in a.

while the transient 2D charge distribution elucidates the carrier and phonon contributions. Γ-LO and TO phonons lead to an isotropic redistribution, while the pseudospin favours collinear scattering towards the Dirac point. Subcycle access to 2D momentum space with attosecond precision turns out decisive in elucidating these dynamics prior to relaxation and thermalization.

Our approach unravels the key conditions for all-coherent field-driven interactions and may shed new light on intriguing strong-field physics ranging from inter- and intraband dynamics, via Landau-Zener tunneling, and dynamical Bloch oscillations, to Floquet-Bloch states.

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Discovery of a Hybrid Exciton State by Attosecond Spectroscopy

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Synopsis We investigate the ultrafast optical response of lithium fluoride to a strong infrared electric field using attosecond transient reflection spectroscopy. Comparing the experiment with real-time simulations accounting for different degrees of electronic correlations, we discovered an unexpected hidden hybrid excitonic resonance, shedding new light onto field-driven dynamics governing the coherent response of wide band-gap insulators.

Understanding the ultrafast dynamics that drive coherent light-matter interaction is crucial for advancing next-generation optoelectronics devices. In this regime, the optical response of dielectrics is governed by the intricate interplay between exciton, virtual, and real charge dynamics [1, 2], which are not fully understood.

Here, we used attosecond transient reflection spectroscopy (ATRS) [3] to study the ultrafast response of a lithium fluoride monocrystal driven by a few-cycle infrared (IR) pulse. Fig. 1a shows the static reflectivity of LiF measured with an attosecond extreme-ultraviolet (XUV) pulse impinging onto the crystal at 72° with respect to the surface normal (red). In agreement with literature data (blue), no sharp feature is detected [4]. The differential reflectivity $\Delta R/R$, defined as the relative change between the pumped and static, is reported in Fig. 1b. It reveals several transient features, mostly oscillating at twice the IR frequency ν_{IR}. The signals above 32 eV originate from IR-induced virtual carriers dynamics [3]. Instead, the sharp feature at ~29 eV refers to an excitonic resonance, hidden in the static data. Theoretical calculations revealed it originates from the interaction of a hole (h) in the top valence bands with an electron (e) into the high-lying conduction bands. Furthermore, this new excitonic resonance undergoes hybridization with the nearby e-h continuum. Dressing the crystal, the IR modifies the coupling with the continuum, affecting its ultrafast optical properties and making it visible in the experiment.

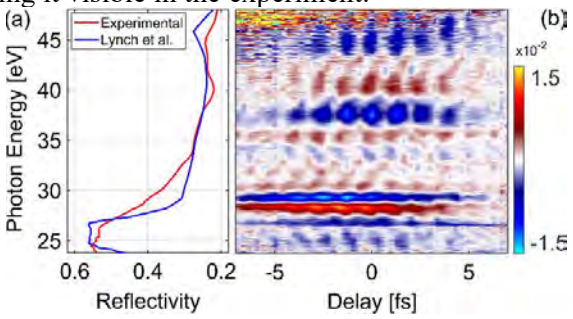


Figure 1. (a) Static reflectivity of LiF at 72° of incidence angle (red) compared to data from Ref. 4 (blue). **(b)** Differential reflectivity trace obtained using a 6-fs IR pump pulse with peak intensity of 20 TW/cm^2.

Our results demonstrate that ATRS is a powerful tool for revealing hidden excitonic features that would otherwise remain undetectable. Moreover, they provide novel insights into the ultrafast optical response of wide-bandgap insulators and our ability to control it with ultrashort light pulses - key aspects for advancing petahertz electronics and our comprehension of fundamental condensed matter physics.

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Correlations drive the attosecond response of strongly-correlated insulators.

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Synopsis Attosecond transient spectroscopy has advanced our understanding of light-driven coherent electron dynamics. However, focus has so far been on weakly-correlated insulators, leaving strongly correlated systems largely unexplored on sub- to few-femtosecond timescales, the natural timescale of electron dynamics. Here we experimentally compare the attosecond response of NiO, a strongly correlated insulator, to a weakly-correlated insulator, uncovering distinct behaviors. Supported by advanced ab initio calculations, we find that the on-site electronic interaction changes in a few femtoseconds.

Strongly correlated materials present peculiar behavior of both technological and fundamental interest, due to repulsive electron-electron interactions. Yet, their response to light excitation remains unclear. The challenge is that purely electronic dynamics requires attosecond temporal resolution. At this timescale, do correlations enhance, screen or have no effect on the sub-optical-cycle response to an external laser field? We address this question via attosecond transient reflectivity on NiO, a prototypical strongly correlated insulator. We first benchmark our setup with MgO [1], a weakly-correlated insulator. It exhibits oscillations at twice the pump laser frequency, a signature of the well-studied dynamical Franz-Keldysh effect by which the field coherently distorts the bands of the solid [2]. Strikingly, NiO does not show any sub-optical-cycle dynamics. Instead, it presents a much slower and non-reversible electronic response that arises in a few femtoseconds. To understand this temporal response we conduct state-of-the-art calculations in the framework of time-dependent density-functional plus self-consistent U theory, which describes ultrafast dynamics in strongly correlated systems. Our results [3], which reproduce the experiment quantitatively, explain this behavior as a light-induced renormalization of the correlation parameter U in the Hubbard model, confirming predictions made in prior theoretical work [4]. With this interpretation, the data yields direct access to the temporal evolution of the effective Hubbard U for the first time. It is found to respond in 7.0 ± 1.5 fs, a system-specific timescale characterizing the dynamics of electronic screening after light excitation. The ability to measure and control the Hubbard U at its true timescale could be foundational for accessing and harnessing new nonequilibrium material states.

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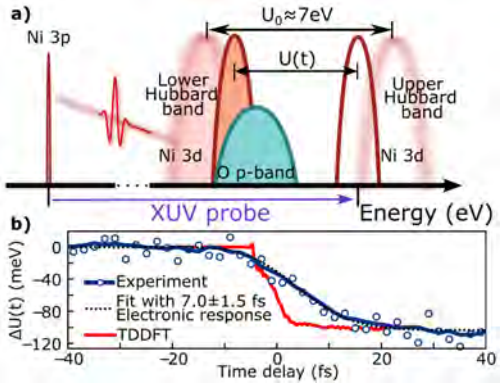


Figure 1. **a)**The XUV pulse probes the energy of the conduction band which is the Upper Hubbard Band, providing a direct measurement of the Hubbard U renormalization. **b)** Comparison of the measured $\Delta U(t)$ and our numerical results.

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Excitonic high harmonics and attosecond time delays in GaN

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Synopsis Quantitative theory-experiment analysis shows a set of excitonic resonances and the consequent attosecond time delays in high harmonics generated by a GaN crystal due to the ponderomotive detuning and many-body effects.

The role of many-body effects in high-harmonic generation (HHG) in solids remains an intriguing question [1]. Here we present a new route to access exciton effects and enhancements in semiconductor HHG. When an n-photon transition energy $n\hbar\omega_0$ matches an excitonic state energy, HHG is significantly enhanced. However, achieving this resonance experimentally requires precise photon energy tuning. To address this challenge, we introduce ponderomotive detuning, where electron-hole pairs acquire a ponderomotive energy $E_{\mathrm{PD}} \propto E_{\mathrm{peak}}^2$, blueshifting the transition energy. This allows excitonic resonances to be reached by adjusting the peak field strength, providing a more flexible approach. While ponderomotively detuned resonances has been observed in atomic HHG [2], its semiconductor counterpart has remained elusive.

To demonstrate ponderomotive detuning and excitonic resonances, we scan the intensity of a 2000-nm femtosecond laser driving HHG from a GaN crystal. Additionally, we use quantum dynamic cluster expansion (QDCE) [3] to theoretically predict HHG many-body effects. Figure 1(a) presents our experimental and theoretical results which agree quantitatively. We observe how ponderomotive detuning affects the 7th HH intensity, I_{H7}, as function of E_{peak}. As expected, the 7th HH produces a clear resonant enhancement at 24 MV/cm corresponding to ponderomotively detunes the 7-photon transition to become resonant with E_{1s}. Surprisingly, we also notice another resonance at 17MV/cm in the 7th HH which corresponds to detuning a 6-photon transition to become resonant with the 2p exciton.

To experimentally resolve attosecond-scale excitonic dynamics in HHG, we use a two-color excitation scheme. The fundamental pulse is combined with a weak second harmonic pulse with a delay Δt. We detect HH intensity as a function of Δt (Fig. 2(b)). When plotting the delays maximizing HHG as a function of field strength (Fig. 2(c)), we find the maxima to be associated with a strong participation of p-like exciton states, while the minima match s-like states. This provides a clear signature of attosecond-scale, few-level transitions between different excitonic states.

In summary, we have studied dynamical ponderomotive resonances in semiconductor HHG, using a quantitative theory–experiment comparison. Two-color spectroscopy enables studies of attosecond dynamics of exitonic states, offering a simple and robust pathway to explore ultrafast many-body effects and few-level transitions in condensed matter.

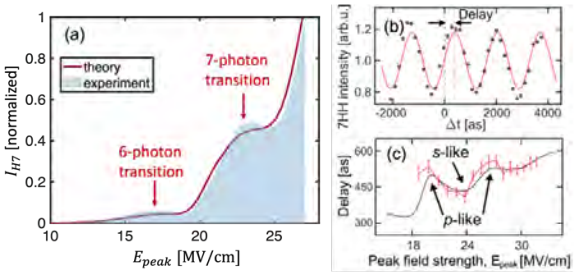


Figure 1. (a) Ponderomotive resonances with excitonic states in field strength scaling of H7 in GaN. (b) 7th harmonic intensity as a function of two-color delay Δt. (c) Attosecond delay of the 7th harmonic as a function of peak field strength.

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Probing ultra-fast electron dynamics in liquids and solutions using high-harmonic spectroscopy

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Synopsis We establish high-harmonic generation (HHG) in liquids as a powerful ultrafast probe for tracking spatial and temporal electron dynamics. Beyond the traditional gas-like picture, we uncover new nonlinear features such as multi-plateau structures and bandgap modification. These findings open pathways to attosecond-scale exploration of solvation dynamics, light–matter interactions, and electronic structure in complex media.

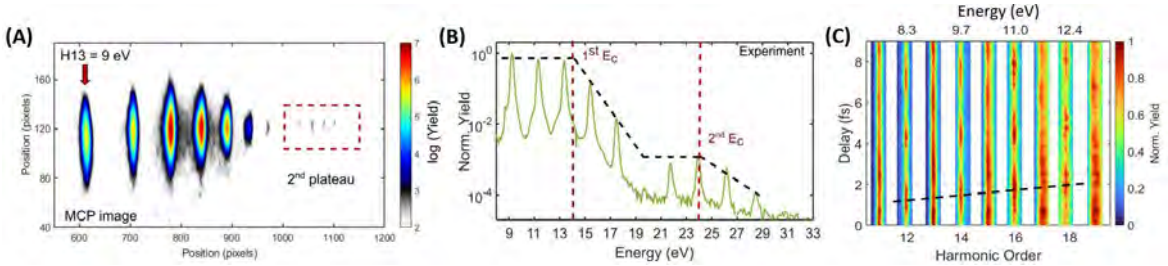


Figure 1. (A) Raw MCP image of high-harmonic spectrum of liquid D_2O measured at ~ 4.8 $\times 10^{13}$ W/cm^2 for 1800 nm wavelength, showing the presence of a second plateau feature. (B) Experimentally observed high-harmonic spectrum from D_2O liquid extracted from (A). (C) Intensity of high harmonics generated in liquid ethanol with 1800/900-nm two-color laser pulses focused to a peak intensity of ~ 4 $\times 10^{13}$ W/cm^2.

High-harmonic generation (HHG) in bulk liquids has been recently explained by the "scattering-limited three-step model" [1]. In liquids such as H_2O, D_2O, and alcohols, the harmonic cut-off energy remains fixed and independent of laser wavelength, intensity, and pulse duration [1, 2]- an outcome of strong electron scattering and dominant on-site recombination. However, this gas-like picture fails to account for the emergence of higher-order nonlinearities.

We report the discovery of a second plateau in HHG spectra (Fig. 1(A) and (B)) of multiple liquids, signaling a new regime of electron dynamics enabled by hole delocalization and recombination with neighboring molecules-especially from the second solvation shell [3, 4]. This plateau shows weak cut-off scaling and distinct ellipticity dependence, supported by experiments, ab-initio simulations, and semi-classical models. Additionally, two-color interferometric measurements [5, 6] provide attosecond-resolved access to the recollision process, revealing a large linear atto-chirp (Fig. 1(C)) and field-induced reduction of the electronic band gap by several electron volts. Aqueous salt solutions exhibit spectral minima whose positions and depths are sensitive to anion type and concentration. These features are well-described by a two-emitter interference model and reflect modulations in the relative phase and band structure induced by chemical environment and laser field.

Together, these findings position HHG in liquids as a powerful probe for ultrafast phenomena capturing both recollision dynamics and spatial recombination pathways-paving the way to explore complex light-matter interactions and solvation dynamics on attosecond time scales.

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Attomicroscopy imaging and control of electron motion in solid-state

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Synopsis Ultrafast electron diffraction and microscopy have enabled real-time imaging of atomic motion. However, challenges in electron pulse compression have constrained temporal resolution to around a hundred femtoseconds. This study demonstrates attosecond temporal resolution in transmission electron microscopy using optical gating, establishing the "attomicroscope." This tool captures electron motion dynamics in graphene, correlating electron density in reciprocal space with real-space movement. Additionally, we present light-induced quantum tunneling currents in graphene phototransistors, enabling current switching at 630 attoseconds and advancing ultrafast optoelectronics to the petahertz regime.

Ultrafast Electron Diffraction and Microscopy imaging have emerged as pivotal tools for capturing atomic motion in real-time and space [1,2]. The generation of electron pulses lasting just a few hundred femtoseconds has enabled the recording of molecular and atomic motion [3]. However, technical challenges in electron pulse compression have constrained the temporal resolution of electron imaging experiments to the hundred-femtosecond range.

Here, we demonstrate attosecond temporal resolution in a transmission electron microscope using optical gating [4], introducing what we term the "attomicroscope" [5]. We further utilize the attomicroscope to visualize electron motion dynamics in graphene. In the presence of a strong field, electrons traverse reciprocal space, following the waveform of the driving field. Attosecond electron diffraction experiments have allowed us to map electron density distribution in reciprocal space at different time instants and correlate it with real-space electron motion. This breakthrough in attomicroscopy paves the way for studying electron motion in neutral matter and holds promise for novel applications in physics, chemistry, and biochemistry [5].

Furthermore, we report the generation of light-induced quantum tunneling currents in graphene phototransistors under ambient conditions. This phenomenon enables precise measurement and control of field-driven currents, demonstrating current switching at an unprecedented 630 attoseconds (~1.6 petahertz) [6]. By modulating the density of photoexcited charge carriers via variable pump laser powers, we enhanced graphene phototransistor conductivity and achieved various logic gate operations. These findings open new avenues for optical switches, lightwave electronics, and optical quantum computing [7,8].

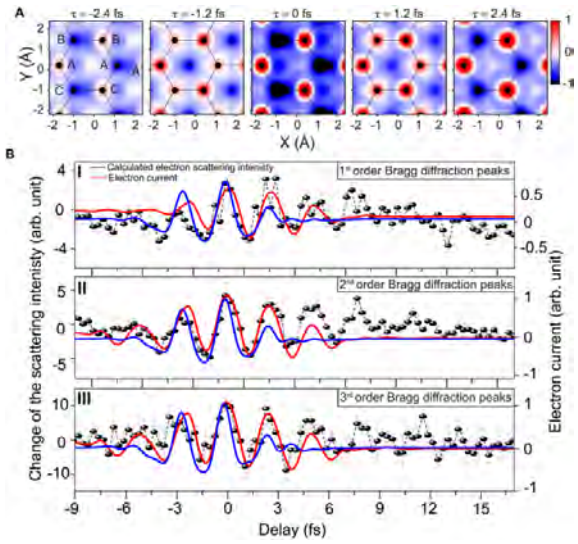


Figure 1. Attomicrocopy imaging of the electron motion in graphene. (A) the light-induced electron motion dynamics between the carbon atoms in graphene. (B) the time-resolved attosecond electron diffraction measurement for the first, second, and third order diffraction peaks of graphene.

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Attosecond spectroscopy of virtual charge dynamics

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Synopsis We report attosecond-scale electron dynamics in crystalline diamond triggered by few-femtosecond optical pulses. Backed by numerical simulations, our findings distinguish for the first time light-induced intra- and inter-band processes in virtual carrier dynamics that underlie the material field-driven response.

The interaction of intense, few-femtosecond infrared (IR) pulses with solids can trigger light-field–driven phenomena that reversibly alter their electro-optical properties on attosecond timescales, offering a pathway to ultrafast optoelectronics and petahertz devices [1]. However, exploiting these coherent light–matter states for future applications requires a deep knowledge of the physical processes unfolding during the interaction between a strong field and a solid. Among them, little is known about the interplay between real and virtual carrier dynamics [2], that could, in principle, limit the material response time, especially in wide-gap dielectrics [3].

In this work, we employ attosecond transient reflection spectroscopy to investigate ultrafast virtual electron dynamics initiated by an intense few-femtosecond IR pulse in monocrystalline diamond over an unprecedented broad photon energy range (Fig. 1). An absolute pump-probe delay calibration enables a direct, parameter-free comparison with theoretical simulations based on time-dependent density functional theory (TDDFT), revealing that virtual inter-band transitions (VITs) — often neglected in favor of intra-band motion [4] — play a significant role in determining the timing and adiabaticity of the overall nonlinear response. Being virtual, this kind of transitions manifest as soon as the light field dresses the material. As such, they can affect the system bandwidth and response time, well before real vertical transitions occur. A deeper analysis based on independent-particle picture and a simplified three-band model shows that VITs substantially influence the attosecond optical response over a wide range of pump intensities. Therefore, they represent a ubiquitous phenomenon in field-driven light–matter interactions that must be accounted for to extract precise spectroscopic information.

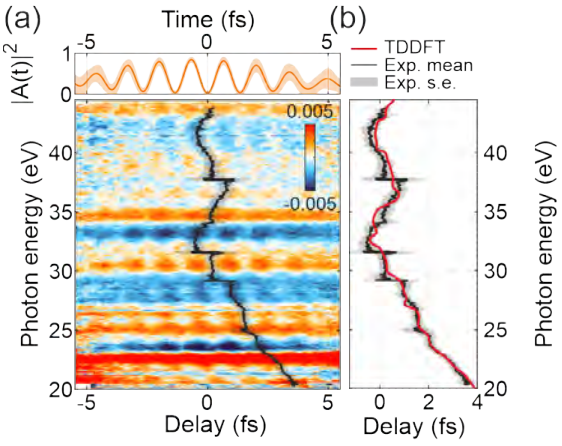


Figure 1. **(a)** Measured IR vector potential (upper panel) and diamond differential reflectivity trace (main panel). **(b)** Experimental (black) and calculated (red) absolute phase delay of the oscillations.

By elucidating the origin and impact of VITs, our work paves the way for exploiting these effects in next-generation optoelectronic devices, ultimately enhancing our ability to control material properties on sub-femtosecond time scales.

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Two-color control of attosecond scanning tunneling microscopy

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Synopsis Attosecond observations of coherent electron dynamics in molecules and nanostructures can be realized by integrating scanning tunneling microscopy (STM) with sub-cycle sculpted femtosecond laser pulses. By precisely controlling the time delay between a 1.8 μm femtosecond laser and its second harmonic, we achieve attosecond control of the laser-induced current. For the first time, we demonstrate that the waveforms can directly dictate attosecond tunneling through an STM junction, paving the way for ultrafast studies at the nanoscale.

Attosecond scanning tunneling microscopy (STM) opens the door to simultaneous attosecond-nanometer imaging of electron dynamics. Compared to conventional STM, which operates only under a static bias, attosecond STM is coupled with a sub-cycle sculpted near-infrared laser field, generating ultrafast current bursts [1]. According to a recent theoretical study from our group, photoemission occurring in the STM junction can be classified into the multi-photon regime and the laser-induced tunneling regime [2]. Specifically, in the latter regime, the attosecond current burst follows a novel three-step process: (1) the electron is emitted from the STM tip into the gap, (2) it propagates within the laser field inside the gap, and (3) it enters the sample, and vice versa.

However, the experimental observation of attosecond current and directional control remains challenging. One major problem is that the laser-induced current is mixed with the static current, as STM operates under a static bias. Here, we show for the first time that we can control the direction and magnitude of attosecond tunneling currents solely by controlling the waveform of the driving laser pulses. By shifting the time delay between a 1.8-μm femtosecond laser beam and its second harmonic, we successfully achieve attosecond control of the laser-induced current (see Fig. 1(a)). To eliminate static currents, the tip is temporarily frozen in place, and the static bias is instantly switched off, leaving only the laser-induced current. We successfully reverse the direction and modulate the magnitude of the attosecond current (see Fig. 1(b)).

Based on our approach, we systematically study the physics of the attosecond current bursts. The observation that the magnitude of the current is only weakly correlated with the STM gap width hints strongly at the strong-field regime (Fig. 1(c)), consistent with our theoretical predictions [2]. Furthermore, polarization modulation reveals that the attosecond current is highly nonlinear and confined to polarization angles where the laser field aligns with the STM tip, confirming that the current is generated solely by the laser (see Fig. 1(d)).

Our experiment achieves coherent control in STM with attosecond precision, enabling the study of ultrafast electron dynamics at the atomic and nanoscale levels.

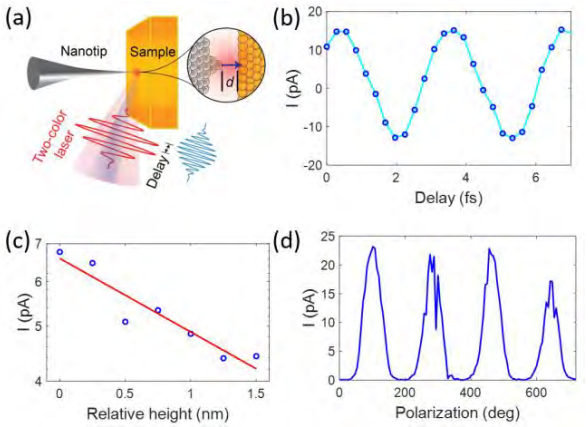


Figure 1. (a) Schematic of the two-color setup. (b) Modulation of the current direction by varying the time delay. (c) Current as a function of the relative tip height, the y axis is in logarithmic scale. (d) Current as a function of polarization.

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Attosecond electron pulses with chiral mass and charge

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Synopsis We show that a free electron can be laser-structured into a left- or right-handed coil with a chiral mass and charge while maintaining a flat de-Broglie wave front. Measurements by attosecond gating reconstructs their three-dimensional profiles. Our tailored chiral electron waves can be used for applications in chiral sensing and nanometric field control.

Chirality is a property with broad significance across particle physics, materials science, optics, chemistry, and biology. Chirality is an inherent signature for most of elementary particles with spin, but chiral phenomena can also appear in geometry through a proper composition of multiple non-chiral particles, such as human hands, chiral crystal lattices, and skyrmions. Elementary particles, such as electrons, atoms, neutrons and molecules can be imprinted with chirality by modifying its phase with spiral phase plates, magnetic needle tips, holographic gratings or photon-induced near-field interactions. Here, we propose that an elementary particle, e.g., an electron, can possess chirality by directly shaping its mass and charge in space and time, while maintaining a flat de-Broglie wave front [1].

In our experiment, we use an ultrafast transmission electron microscope with a Schottky field-emitter source [2]. We create femtosecond electron pulses by photoionizing a field-emitter tip and modulate the electron pulses with a beam of femtosecond optical vortex. In their interaction region, we place a silicon nitride membrane as a modulation element. The incoming electrons gain or lose forward momentum as a function of arrival time. The local electric field oscillations at a given position in the profile of optical vortex therefore directly modulate the electron matter wave at the corresponding position in the electron beam profile. After 18-cm free propagation the electron is shaped into an attosecond electron coil wave.

Then, the electron coil is probed by an attosecond streaking gate. Depending on the arrival time of the electron coil, its incoming parts are either accelerated or decelerated. It allows us to measure the attosecond-gated spatial shape of the incoming electron density of mass and charge.

Figure 1 shows three-dimensional reconstructions of the measured electron densities for different winding numbers ℓ of the optical vortex. For $\ell = 1$, we see a single spiral wave packet at a diameter of $\sim$ 200 nm and a pitch of 700 nm in form of a left-handed coil. For $\ell = -2$, we see a right-handed double-helix shape, where the two coils are intersected with each other. Chirality appears here without the need for spin or orbital angular momentum.

The size of the our electron coils is only limited by the de Broglie wavelength and can therefore reach picometer dimensions for applications in atomic or molecular spectroscopy, for example, to induce or probe the ring currents in atomic orbitals or magnetic materials.

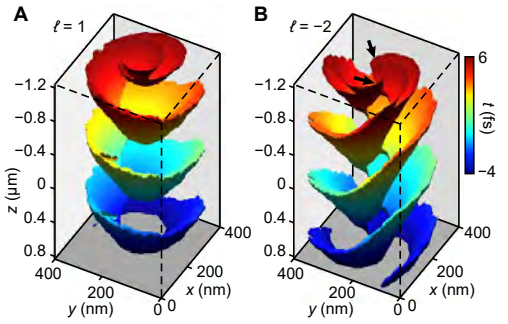


Figure 1. Reconstructed expectation values of the charge and mass of the electron coils. Free-electron modulation with an optical vortex of (A) $\ell = 1$ and (B) $\ell = -2$.

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High-harmonic spectroscopy probes lattice dynamics: extension to bandgap dynamics and structural phase transitions

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Synopsis We demonstrate a novel way to probe coherent phonon dynamics in solids using high-harmonic spectroscopy. Our technique additionally reveals bandgap dynamics and structural phase transitions.

Since the first demonstration of high-order harmonic generation (HHG) from solids in 2010 [1], there have been many works reported HHG from condensed matter and its spectroscopic applications. Here, we show that high-harmonic spectroscopy (HHS) can be used to probe lattice dynamics [2], obtaining both absolute electron-phonon coupling as well as anharmonic phonon-phonon coupling. Additionally, our technique allows observation of the bandgap dynamics [3] as well as structural phase transitions [4].

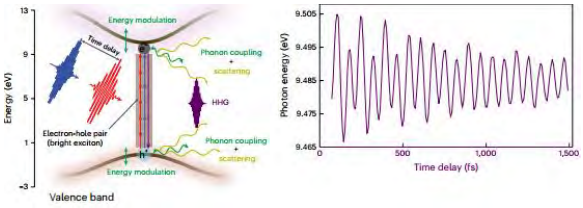


Figure 1. Physical mechanism (lelf panel) and coherent phonons signature (right panel). [2]

Our technique to probe lattice dynamics is background-free and has extreme sensitivity directly in the energy domain.

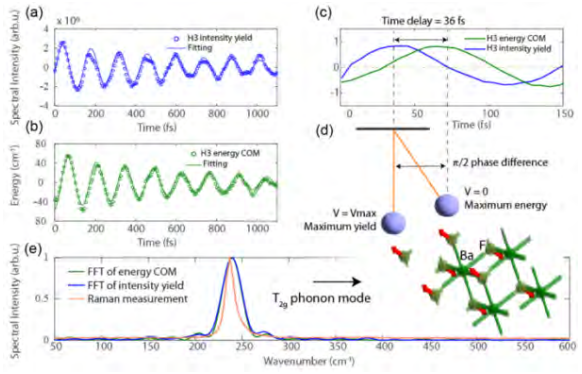


Figure 2. 2D observation of coherent phonons in solids (a,b). Relative phase (c) and physical analogue (d). Frequency representation (e). [3]

All-optical manipulation of bandgap dynamics via coherent phonons is shown in Fig. 2. The tr-HHG spectrum perturbed by a triply degenerate phonon mode T2g exhibits simultaneously a remarkable two-dimensional (2D) sensitivity, i.e., in both intensity and energy domains.

Probing photoinduced structural phase transitions via nonlinear spectroscopy is shown below

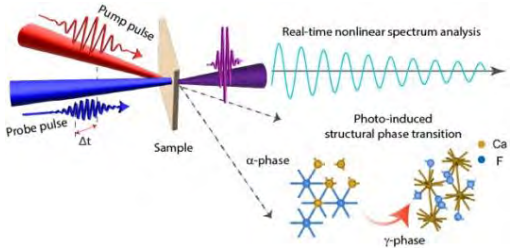


Figure 3. Observing structure phase transitions via their transient signature of coherent phonons. [4]

The coherent phonon mode initially exhibits a softening effect. Subsequently, the transition from α-CaF2 to γ-CaF2 occurs at higher carrier density, corresponding to a switch from the high-symmetry $Fm\bar{3}m$ to the low-symmetry *Pnma* space group.

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Ultrafast control of valley physics in 2D materials

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Synopsis We demonstrate ultrafast control of valley physics in 2D materials using tailored light fields. A trefoil field realizes a Haldane-like topological model, while phase-locked, polarization-controlled laser pulses achieve room-temperature valley switching.

2D materials exhibit intriguing features like non-trivial topology and valley physics, governed by time-reversal and spatial inversion symmetries. Valley physics emerges when the latter is broken in the crystal, as in hexagonal monolayers, leading to two energy-degenerate minima (K and K' valleys) in the conduction band. Circularly polarized light can selectively excite these valleys, enabling valleytronics—using valley states for data storage [1]. Effective valley control requires ultrafast methods due to rapid decoherence (~100fs at 100K). Time-reversal symmetry breaking, such as via next-nearest neighbor coupling, lifts valley degeneracy and can induce a topological phase transition, as shown in the Haldane model [2].

Here, I show how these fundamental symmetries can be manipulated on ultrafast timescales using tailored light fields, enabling coherent control of quantum properties. I will show that a trefoil-shaped field can controllably break time-reversal symmetry and transform a semiconducting monolayer into a laser-analogue of the topological Haldane model, with the band topology controlled on few-femtosecond timescales [3,4]. Application of these fields to multilayer systems extends valleytronics to inversion symmetric materials [5]. Finally, I will show the first realization of real-time valley switching at room-temperature in a WS_2 monolayer by employing a coherent control setup made of a sequence of few-cycle pulses with alternating polarizations [6,7].

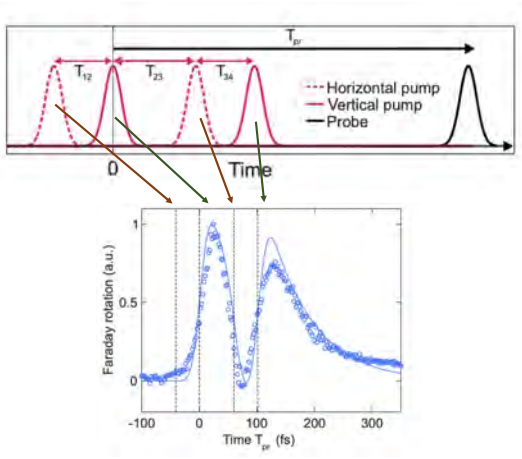


Figure 1. Four linear pulses with alternating orthogonal polarizations turn on-off-on the valley state, controlled by the attosecond delay between the pulses. Experiments are marked by circles and theory by the solid line.

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High harmonic generation from solutions

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Synopsis We study high harmonic generation (HHG) in halobenzene-methanol (PhX-MeOH) solutions. Our experimental results show evidence of local order in the solvation of PhF in MeOH. This manifests as a near-complete suppression of a single harmonic in the solution, which is absent in each of the pure liquids and absent in other PhX-MeOH solutions. We interpret the results in terms of a solvation shell that is formed in the PhF-MeOH solution and acts like a local barrier in the HHG process.

High harmonic generation (HHG) can be an exquisite probe of both structure and dynamics in quantum systems, and has been widely studied in gas-phase atoms and molecules as well as in condensed-phase periodic crystals. Only few studies of HHG in liquids exist and have documented low cutoff energies that are relatively insensitive to the driving laser parameters [1,2], and appear to be dominated by the reduced mean-free path of the continuum electrons in the liquid as compared to gas phase [3,4]. That is, the lack of short- or long-range order of the liquid phase has appeared to play an important role.

Here we report a joint experimental and theoretical study of HHG in solutions, consisting of a low concentration of halobenzene (PhX, around 10% by mole) solvated in methanol (MeOH) [5]. In their pure liquid form, the HHG yield of PhX is higher than that of MeOH by 1-2 orders of magnitude, with cutoff energies consistent with previous work [2,3], as shown in Fig. 1. In most of the PhX-MeOH mixtures, the solution-HHG yield is consistent with a simple sum of the yields from each liquid. However, as shown by the blue curve in Fig. 1, the PhF-MeOH solution exhibits an overall lower harmonic yield than either of the two pure liquids, and a near-complete suppression of the 17th harmonic near 12 eV.

We interpret this result as evidence of local structure in the PhF-MeOH solution. Our molecular dynamics simulations show that in the PhF-MeOH mixture, a new solvation shell is formed around the F atoms that interrupts the MeOH hydrogen-bonding network. This solvation shell is not present in the other PhX-MeOH solutions. Using a simple model solution of the time-dependent Schrödinger equation, we find that a small, localized structure near the ion core can lead to both an overall suppression of the harmonic yield, and a strong suppression of a single harmonic.

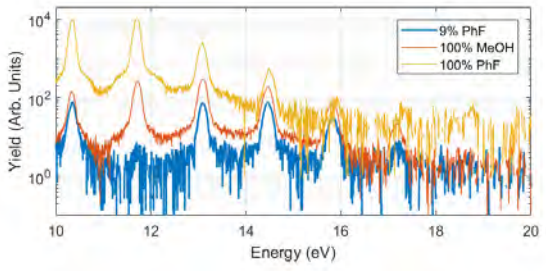


Figure 1. Harmonic spectra for pure MeOH (orange), pure PhF (yellow), and a solution of 9% PhF and 91% MeOH (blue) for a driving laser wavelength of λ = 1800 nm and I = 30 TW/cm^2. In the solution, the 17th harmonic near 12 eV is strongly suppressed.

This work shows that HHG in solution can be sensitive to unique features of the solute-solvent interaction, and thus to the local liquid environment, and increases our understanding of attosecond dynamics in chemically and biologically relevant liquid-phase systems.

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Enhancement of harmonic generation in liquid water jet by resonant excitation in the mid-infared pulses

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Synopsis Harmonic generation in the visible spectral range in a liquid flat jet of H_2O is studied using vibrationally resonant intense 3 μm pulses. With a weak prepulse at 3 μm, we observe an enhancement of the 5th, 7th, and 9th harmonics that appears about 2 ps after the excitation and persists for more than 120 ps. Such an enhancement is not observed in the case of non-resonant D_2O. We attribute this enhancement to ultrafast heating of the liquid water, which changes the macroscopic optical properties of liquid water.

High harmonic generation (HHG) in flat liquid jets has opened a new direction in strong-field and attosecond science, where intense light-matter interactions occur in high-density ensembles of molecules [1-3]. The pioneering work with resonant MIR excitation clearly demonstrates the influence of strong resonant absorption of water [1]. With intense ultrashort pulse in the SWIR, the spectral range of water HHG is extended to the extreme ultraviolet (EUV) region, where density effects reduce the higher order harmonics near the cutoff [2]. Similar experiments with isopropanol show the universal features of liquid HHG in EUV [3], which can be basically understood by the three-step model.

In our experiment [4], we generate harmonics with an intense MIR pulse (~30 μJ, 70 fs at 3100 nm) in a flat liquid jet of H_2O with a thickness of ~8 μm that is longer than the absorption length (~1 μm) at resonant wavelength of 2940 nm. When the focused intensity exceeds ~10^{14} W/cm^2, the intensity scaling of the 5th, 7th, and 9th harmonics starts to deviate from the perturbative scaling. Under this condition, we introduce a weak prepulse at the same wavelength to the main pulse. The intensity of the prepulse is set at ~10^{13} W/cm^2 where no harmonics are observed. Figure 1(a) shows the intensity change of the 5th harmonic as a function of the prepulse delay time. A slow increase in the harmonic yield is observed about 2 ps after the pre-excitation, regardless of the polarization configuration. This enhancement persists for more than 120 ps. Figure 1(b) shows the normalized harmonic enhancement with different MIR wavelengths. The enhancement becomes more pronounced as the MIR wavelength gets closer to the resonance, while the time scale to reach the persistent enhancement remains the same. We attribute this enhancement to the ultrafast heating of the liquid water to ~4200 K in our experimental condition. At this high temperature, it is likely that the hydrogen bonding is completely lost and the macroscopic optical properties of the water jet, in particular the absorption length, may have changed.

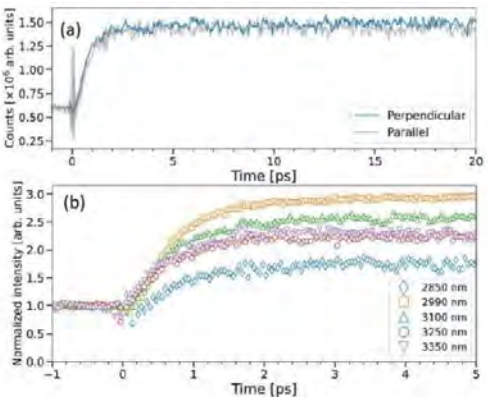


Figure 1. (a) Intensity change of the 5th harmonic as a function of the prepulse delay time for perpendicular or parallel polarization. The wavelength of the MIR pulse is set at 3100 nm. (b) Wavelength dependent enhancement of the 5th harmonic.

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Nonlinear science with multi-harmonic FEL at sub-femtosecond resolution

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Synopsis I will report on experimental results on nonlinear photoionization of atoms (He, Ne) with trains of attosecond pulses generated as a coherent superposition of commensurable harmonics with the Free Electron Laser FERMI. The relative phase between harmonics is an easily accessible machine parameter, and has been used to control the outcome of the processes under study.

The technical ability to generate phase-locked high harmonics of an optical laser by the HHG technique, and to monitor them with the RABBIT scheme opened the way to sub-femtosecond pulses [1, 2], and to the investigation of electronic motion in molecules on its natural timescale, decoupled from nuclear motion [3]. The advent of seeded Free Electron Lasers in the XUV range [4], and in particular the realization that they could be operated as a phase-locked multi-harmonic source [5], added valuable flexibility in the synthesis of more complex waveforms [6], as well as greater pulse energy. Let us note that all the experimental schemes implemented so far have been based on the mixing of two electric fields, *i.e.*, they are intrinsically non linear, and that one of the fields is that of an external, typically IR, laser.

I will present here recent results obtained with a FEL-only scheme, thus involving at least two FEL photons. In one experiment, autoionizing doubly-excited states of neutral helium [7] were excited with two FEL photons of combined energy of $\approx 60\,\mathrm{eV}$ in a three- and four-harmonic configuration. A similar experiment was performed in neutral argon, with two-photon ionization in the continuum via an intermediate autoionizing $(3s\,3p^6\,5p)$ resonance. In the latter experiment, non-resonant two-photon ionization of Ar^+ to Ar^{2+} was also observed, implying the presence of processes involving the absorption of three or more photons. In all cases, the experiments showed a clear dependence on the relative phase of the harmonics.

The results presented originate from the joint effort of many international laboratories and of a large number of researchers, whose work is gratefully acknowledged.

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Dephasing in attosecond science and the collective nature of structural quantum fluctuations

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Synopsis I will first discuss the important and useful role that subfemtosecond electronic dephasing played in the first all-x-ray attosecond pump-probe experiment on a condensed-phase system. I will then present a method that allowed recovering the collective properties of structural quantum fluctuations in a high-dimensional nuclear configuration space. Such quantum fluctuations can lead to rapid electronic decoherence and are therefore relevant to attosecond science.

There has been a long-standing debate regarding the interpretation of the splitting of the $1b_1$ peak in the x-ray emission spectrum of liquid water. One view is that this splitting reflects the presence of two discernible structural motifs in ambient liquid water [1]. This perspective is motivated by the need to find a microscopic explanation for the highly anomalous properties of water. Another view is that the splitting is caused by nuclear dynamics during the lifetime of the inner-shell-excited states that must be populated in order to trigger x-ray emission [2, 3]. A recent attosecond-pump/attosecond-probe experiment using an x-ray free-electron laser (XFEL) has brought clarity [4]: By eliminating hydrogen motion on the time scale of the inner-shell hole lifetime of about 4 fs, the $1b_1$ splitting could be shown to disappear.

Freezing the hydrogen motion in this all-x-ray attosecond-transient-absorption-spectroscopy (AX-ATAS) experiment relied not exclusively on the attosecond duration and delay of the x-ray pulses employed. By building on the theory [5] developed in the context of one of the first ATAS experiments [6], we demonstrated that the AX-ATAS response in liquid water is confined to the subfemtosecond time scale. This is a consequence of rapid dephasing of the electronic polarization induced by the probe pulse, which is caused by the large inhomogeneous broadening characteristic of a disordered condensed-phase system such as liquid water. Hence, in this case, the ultrafast decay of electronic coherence was key to the success of the experiment.

In other situations in attosecond science, when the primary goal is the production of long-lived electronic coherence, decoherence mechanisms can be a serious obstacle. For example, initial-state quantum fluctuations of the atomic positions and momenta in polyatomic systems can lead to electronic decoherence on the time scale of 1 fs [7, 8]. Characterizing the properties of these quantum fluctuations is therefore not only of general interest, but is also of immediate relevance to attosecond science. Capturing such collective phenomena requires multi-coincidence methods such as Coulomb Explosion Imaging [9]. Exploiting the high degree of ionization that may be induced using XFEL pulses, which creates optimal conditions for CEI [10, 11], we recently succeeded in recovering the 33-dimensional final-momentum distribution obtained in XFEL-based CEI of an 11-atom molecule. We showed that the correlated fluctuations present in the final momenta directly reflect the collective initial-state quantum fluctuations.

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Coherent control with two photons at a seeded free-electron laser

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Synopsis Ionization control in resonant two-photon absorption of atoms has been realized with chirped circularly-polarized laser pulses at a seeded free-electron laser. These measurements are a crucial step towards coherent-control experiments making use of spectral-phase shaping in the XUV range of the electromagnetic spectrum.

Rabi oscillations are an ubiquitous phenomenon in resonant driving of (any kind of) two-level systems. In a dressed-state picture this driving corresponds to a level splitting, which is usually referred to as Autler-Townes doublet. It has been known for a long time, that these two states in laser-driven systems can be selectively addressed by tailored pulses, the simplest ones being chirped pulses. To demonstrate this, ionization from those states have been measured with photo-electron spectra.

A few years ago we have shown [1] that the ionization step may strongly depend on the direction of the chirp. This is surprising, since it is very different from the process known as rapid adiabatic passage, whereby nearly perfect population inversion (100% population of the upper state in the two-level system) can be reached for both directions of the chirp. Most importantly, this dependence offers a new handle on controlling the ionization process on an absolute scale. We have predicted, that simply the chirp allows to switch between none and full ionization of a Helium gas, and explained the underlying mechanism [1].

This idea has been taken up by experimentalists from the Albert-Ludwigs-Universität Freiburg in order to perform one of the first quantum-control experiment at a seeded free-electron laser with shaped pulses. The shorter wavelengths at these large-scale facilities (as compared to the optical wavelengths in a typical laser lab) provide access to shorter time and length scales. In the experiment [2] at FERMI in Trieste/Italy the transition to the 1st excited state of Helium was chosen in order to have a strong coupling and a clean situation with neighboring states being far away in terms of the pulse bandwidth. Besides, the pulses were circularly polarized, which allowed for rather high intensities and, as a nice side effect, enabled a transparent theoretical explanation.

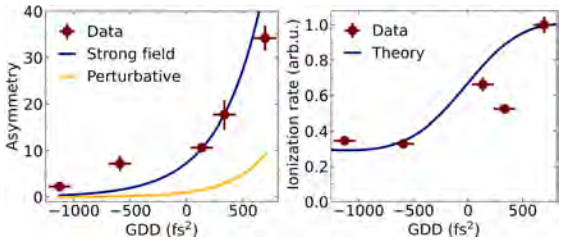


Figure 1. Left: Amplitude ratio between upper/lower photoelectron bands evaluated at the $1s^2 \rightarrow 1s2p$ resonance. Experimental data (red), TDSE-SAE model (blue), TDSE-SAE with the continuum treated perturbatively (yellow). Right: Dependence of the He ionization rate on the spectral phase of the driving field. Data (red) and TDSE-SAE model (blue) are compared. Figure from the experimental paper [2].

By propagating the time-dependent Schrödinger equation (TDSE) for a single-active electron (SAE) model of Helium we could fully interpret the measured data, cf. Fig. 1, and thereby confirm that controlling ionization on an absolute scale with the chirp works.

The experimental collaboration [2] was led by L Bruder (Albert-Ludwigs-Universität Freiburg).

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Complex attosecond waveform synthesis using regular- and irregularly spaced harmonic combs at seeded free-electron laser FERMI

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Synopsis This work presents the generation, characterization, and synthesis of complex attosecond waveforms using the temporal beating of phase-locked harmonics, spaced regularly and irregularly, produced at the FEL FERMI. Correlation based reconstruction of attosecond pulses (CoBRA), a two-colour photoionization scheme inspired by RABBIT, is a unique shot-to-shot correlation analysis that not only allows the characterization of the generated attosecond pulse trains but also the reconstruction of the fluctuating delay between the extreme ultraviolet and the near-infrared fields on a shot-to-shot basis.

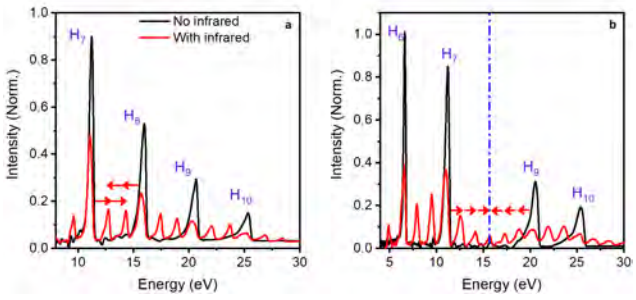


Figure 1, Harmonic comb with regularly (a) and irregularly (b) spaced harmonics. In panels (a) and (b), the curves in black and red are the average photoelectron spectrum acquired without and with the infrared probe, respectively [1,2]. The arrows indicate the absorption/emission of the additional infrared photons.

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Magnetization Dynamics probed with Helicoidal Dichroism

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Synopsis Optical probing of magnetization dynamics has traditionally relied on analyzing the polarization of plain Gaussian beams. Recently, we introduced a novel approach – Magnetic Helicoidal Dichroism – based on analyzing the reflectivity of beams with twisted wavefronts, i.e., beams carrying orbital angular momentum. Here, we demonstrate the power of this technique by identifying remarkably fast and unexpected variations in the topology of a magnetic vortex pumped by an infrared femtosecond laser.

While the spin angular momentum of light is widely used in dichroic studies — such as X-ray magnetic circular dichroism (XMCD) or Magneto-Optical Kerr Effect (MOKE)— the orbital angular momentum (OAM) degree of freedom has been far less explored. Recently, some of us demonstrated that OAM can enhance ptychographic imaging resolution [3]. Besides, by controlling the sign of the incoming OAM, we also showed that it can reveal the orientation of magnetic textures, such as the curling sense of a magnetic vortex [1, 2], an effect termed Magnetic Helicoidal Dichroism (MHD).

We have now leveraged this technique to track the ultrafast dynamics of an oriented inhomogeneous magnetic texture. In particular, we identified a transient inversion of the curling sense of a magnetic vortex during its recovery following partial demagnetization initiated by a femtosecond laser pulse [4]. A key advantage of Time-Resolved MHD is its ability to operate without a real space imaging of the spin texture, enabling rapid measurements of a meaningful parameter with just a few intensity patterns. These results hold great promise not only for monitoring the dynamics of complex magnetic textures but also for uncovering new forms of optical spin-orbit interactions mediated by magnetization [5].

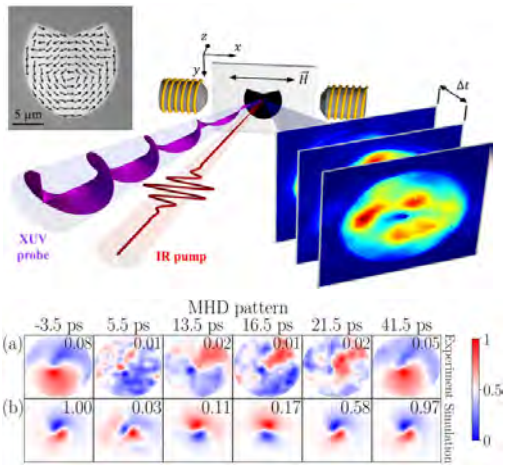


Figure 1. (top) Experimental sketch. A magnetic vortex texture is demagnetized by a Gaussian IR pump, P-Polarized; while a delayed XUV pulse with OAM is reflected off the surface and images are recorded. (bottom) Experimental (a) and simulated (b) MHD maps as a function of time .

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Ultrafast Electron Motion Probed with Attosecond X-ray Free Electron Lasers

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Synopsis Attosecond X-ray Free Electron Lasers (XFELs) provide the brightest source of attosecond radiation. The sources provide unique opportunities to probe the ultrafast motion of electrons in quantum systems.

X-ray Free Electron Lasers (XFELs) provide the brightest source of ultrafast x-ray pulses. With recent innovations in XFEL technology these sources are now able to produce attosecond-scale pulses [1, 2]. In my talk I will highlight our recent developments in probing electronic motion in small molecular systems. We have employed sub-femtosecond pulses from the XFEL to study ultrafast charge dynamics in both core-excited and low-lying cationic systems. Exploiting the interaction between strong-laser fields and the Auger-Meitner emission from core-excited systems we can probe electronic coherence [3, 4, 5]. I will also show our first results on attosecond pump/probe [6] experiments of ultrafast charge dynamics following valence ionization by attosecond x-ray pulses. I will also discuss our recent developments in developing brighter [7] or tailored [8] attosecond pulses from the XFEL.

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Characterization of polarized attosecond soft X-ray pulses at EuXFEL

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Synopsis Polarization-controlled FELs provide access to nonlinear and time-resolved investigations of the transient state of chiral matter. By combining multiple technological capabilities, such as attosecond pulses and undulator-based polarization control of SASE FELs, we enable the observation of ultrafast and nonlinear electron dynamics in chiral systems from specific observer sites. European XFEL, has recently demonstrated the availability of circularly polarized attosecond X-ray pulses for which we will discuss the characterization as well as the scientific prospects.

Polarization-controlled FELs provide unique access to nonlinear and time-resolving investigations of transient and otherwise inaccessible states of matter. They have demonstrated rich potential for magnetization studies at various facilities but also open new avenues for gas-phase science. The emerging field of site-specific investigations of dynamics in (transient) chiral matter, i.e., matter that possesses a handedness, is an essential example in this regard. Chirality has a profound relevance for a multitude of reasons. In fact, it influences our everyday life and is discussed to shape the fabric of our existence to a great extent. Gas-phase studies of chiral molecules at FELs can unprecedentedly add new perspectives on their (re-)formation dynamics, their ultrafast response to light of different energies in general, their ionization dynamics and ionic functionality, and the relevance of a stereocenter site for an effective choice of methodology. The rapid evolution of technological advances at (X)FELs has leveraged the accessibility of this new topic, among others, by enabling multi-color schemes for FEL-pump FEL-probe methodology, isolated attosecond pulses, high repetition rates, and finally, undulator-based polarization control. Only by combining these technological capabilities will we ultimately enable a new depth of observing and steering ultrafast dynamics in chiral systems. However, already, the metrology of such complex pulse properties, particularly at SASE-FELs, remains a challenge for itself, yet being a prerequisite for this young field of science. The status of advanced metrology schemes for variably polarized, high-power (X)FEL pulses at the attosecond frontier, the latest results from our angular-streaking campaign at the European XFEL, and the advent of related scientific prospects that are about to emerge at a variety of facilities worldwide will be presented. Emphasis will be set on the recent demonstration of single and two-color X-ray pulses with a few hundred attoseconds duration, several hundreds of GW power, and full polarization control at the SQS instrument of the European XFEL.

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Redirection and reshaping of intense extreme-ultraviolet radiation

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Synopsis We present an extreme-ultraviolet (XUV) spatio-spectral phase modulator based on an intense XUV laser beam propagating through an optically thick resonant target, introducing dispersion profile variations around the resonance both perpendicular to and along the laser propagation direction. As an experimental demonstration, the intense-XUV-induced double-peak off-axis structure in the far-field spectrum obtained at the Free-Electron Laser in Hamburg (FLASH) shows indications of the underlying XUV-driven Rabi dynamics and resonant pulse propagation effects.

The goal to control short-wavelength radiation for the investigation and manipulation of ultrafast dynamics in quantum systems coevolves with the growing availability of extreme-ultraviolet (XUV) and x-ray sources from high-harmonic generation and free-electron lasers. In the weak-field regime, the spatial redirection of XUV beam has been recently demonstrated by virtue of a spatially offset auxiliary infrared beam [1] or a gas jet with a transverse density gradient [2].

In this work, we demonstrate the spatial redirection and spectral reshaping of intense XUV radiation by passing solely a single XUV beam through a resonant medium. This is achieved by the self-induced change of the dispersion profile in the vicinity of an atomic resonance through the interplay of nonlinear XUV light-matter interaction and resonant pulse propagation in an optically thick medium [see Figs. 1(a)-1(c)]. As a proof-of-principle experimental demonstration, the characteristic off-axis two emission peaks measured at FLASH indicate the presence of XUV-driven Rabi dynamics and macroscopic pulse propagation effects [see Figs. 1(d) and 1(e)] [3]. The described phenomenon is universal and can be exploited as gas-based spatio-spectral phase modulators, here also proven to work in the intense-field regime at short wavelengths, where achieving pulse shaping is challenging but promising.

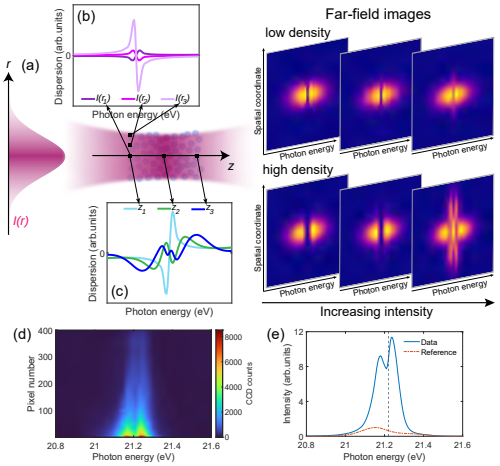


Figure 1. (a)-(c) Concept of the XUV spatio-spectral phase modulator. (d), (e) Experimentally recorded far off-axis XUV spectrum, showing the characteristic intense-FEL-induced double-peak structure.

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Impulsive stimulated X-ray Raman scattering in the condensed phase with attosecond XFEL pulses

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Synopsis We present the first evidence of Impulsive Stimulated X-ray Raman Scattering (ISXRS) in the condensed phase via observation of the photonic signal by use of an attosecond XFEL. We also show accompanying calculations modeling the polarization response of water molecules in these conditions, offering insight into the resulting wavepackets. As the ISXRS process results in superpositions of valence-excited states, it can allow for the creation (and in principle detection) of initially localised excitations in matter, enabling for the characterization of charge migration (CM) in neutral systems.

Electronic charge dynamics within molecular systems, or charge migration (CM), occurs on subfemtosecond timescales. CM can influence critical chemical processes unfolding over longer timescales, such as isomerization and bond cleavage. The effects that follow have spurred interest in the development of both experimental and theoretical methodologies to characterize CM across various systems in all phases of matter.

Impulsive stimulated X-ray Raman scattering (ISXRS) is anticipated to permit the creation and detection of localized valence-excited wavepackets in a site-selective manner, enabling the observation of CM between specific sites in heteroatomic systems. We present evidence for the first observation of ISXRS in the condensed phase by directing attosecond soft X-ray pulses at the chemRIXS endstation at the Linac Coherent Light Source.[1] This follows work done in the gas-phase.[2] Power densities of 10^{17} Wcm^{-2} were focused onto a flat sheet liquid jet.

Our observable is encoded in the scattered modes of the X-ray field, corresponding to stimulated emission into the valence-excited states of the water molecules. (See figure 1).

We show calculations for the polarization response of a water dimer following its interaction with the X-ray field, which facilitates the modeling of H-bonding interactions in a bulk solvation environment. This approach yields agreement with experimental results and offers valuable insights into the dynamic populations of core and valence-excited states throughout the process.

We anticipate this result will form a foundational element for more complex time-resolved ISXRS pump-probe schemes, thereby enabling CM characterization in neutral condensed phase systems.[3] Such schemes may involve multiple time-separated attosecond XFEL pulses, allowing for the creation and detection of valence-excited wavepackets in systems via inelastic and superelastic scattering processes respectively.

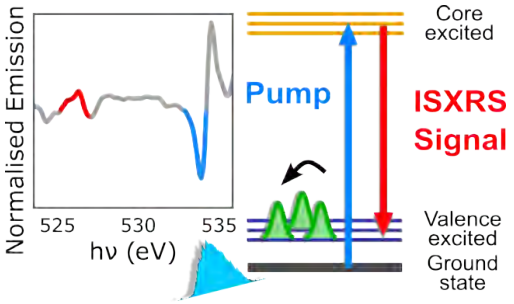


Figure 1. Level scheme of the ISXRS process. The measured ISXRS signal is shown in red.

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Single-shot measurements soft x-ray attosecond pulses at kilohertz repetition rate

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Synopsis We report the first time-domain characterization of attosecond soft x-ray pulses at the LCLS-II superconducting accelerator. Using an angle-resolving electron time-of-flight spectrometer, we perform angular streaking measurements at several kilohertz repetition rates, which we use to reconstruct the temporal profiles of 400 eV x-ray pulses. These measurements showcase LCLS-II's attosecond science capabilities at high repetition rates, laying the groundwork for nonlinear x-ray spectroscopy and x-ray pulse shaping to investigate ultrafast dynamics in complex systems.

The ultrafast motion of electrons in small quantum systems occurs on attosecond timescales. These motions underpin fundamental processes such as photosynthesis, catalysis, and chemical bond transformations. Understanding these phenomena requires tools with both high temporal resolution. Previous experiments at LCLS have demonstrated attosecond capabilities at 120 Hz ([1], [2]). Here, we report the first single-shot measurements of attosecond soft x-rays at LCLS-II at 8 kilohertz repetition rate.

Using an angle-resolving electron time-of-flight spectrometer [3], we perform angular streaking measurements to characterize the x-ray pulses. The streaking measurement enables us to characterize the spectral phase of the x-ray pulse and thus perform a time-domain measurement. The momentum distribution of the laser-dressed photoelectrons and spectral intensity measurement of the x-ray pulse are combined in a reconstruction algorithm [4] to recover the time-domain profile of the x-ray pulses. An example of a single reconstruction is shown in Figure 1. From the reconstruction, we obtain measurements of sub-femtosecond pulse durations.

These measurements demonstrate the attosecond science capabilities of LCLS-II at kilohertz repetition rates. The single-shot characterization of the pulses' temporal profiles lay the foundation for controlling and shaping x-ray pulses to study and control ultrafast dynamics in complex systems with precision.

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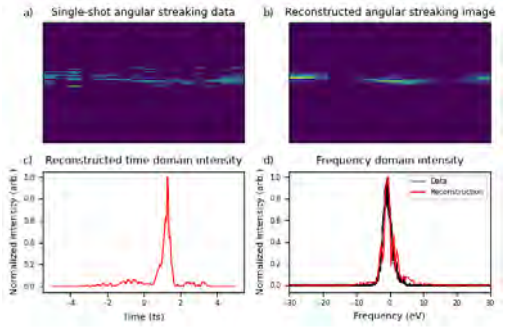


Figure 1. Single-shot x-ray pulse reconstruction from angular streaking measurement. a) Experimental measurement of angular streaking of krypton gas with a single shot of 400 eV x-ray pulse. b) Reconstructed momentum distribution from the nonlinear fitting procedure. c) Time-domain intensity profile of the reconstructed x-ray pulse. d) Experimental measurement of single-shot x-ray spectrum and spectrum of the reconstructed pulse.

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Abstracts for Posters:

Characterizing and controlling lifetimes of doubly-excited states directly in the time domain

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Synopsis Attosecond noncollinear four-wave-mixing (FWM) spectroscopy can characterize lifetimes in a quantum-state specific manner directly in the time domain. Here, we measure the natural lifetimes of doubly-excited states in helium around 60 eV with table-top extreme-ultraviolet/near-infrared (NIR) FWM spectroscopy. States that are accessible by both, one-photon ($^1P^o$ 2s*n*p Rydberg series) and two-photon transitions ($^1S^e$ $2p^2$ state) are addressed. Furthermore we demonstrate how the extracted lifetimes are controlled via Rabi cycling induced by intense NIR pulses.

We present the systematic experimental lifetime characterization of the $^1P^o$ 2s*n*p Rydberg series and the $^1S^e$ $2p^2$ doubly-excited state of helium directly in the time domain using noncollinear extreme-ultraviolet/near-infrared four-wave-mixing (XUV/NIR-FWM) spectroscopy [1]. This all-optical, table-top technique directly probes the population decay in a state-specific manner. Due to the nonlinear $\chi^{(3)}$ optical pumping of the FWM scheme, states that are not accessible from the ground state via a one-photon transition—so-called *dark states* like the $2p^2$ state—can be targeted. The extracted lifetimes are in excellent agreement with theoretically predicted values and spectral linewidth measurements [2]. While the static absorbance spectra of these autoionizing resonances exhibit Fano line shapes, the XUV/NIR-FWM signatures are exclusively of Lorentzian character due to the background-free FWM signal. Using few-cycle NIR laser pulses in combination with attosecond XUV pulses provides the required few-femtosecond temporal resolution, FWM signal strength, and access to natural lifetimes [see Fig. 1(a)] as long as the NIR intensity does not induce state couplings exceeding 0.3 Rabi cycles.

For higher NIR intensities, population is coherently transferred between bright and dark states, which results in a modified FWM lifetime signature due to Rabi cycling. To gain deeper insights into the underlying quantum dynamics, the measured FWM traces are compared to simulations based on the time-dependent Schrödinger equation. One key finding is the NIR-mediated temporal elongation of the 2s2p decay in Fig. 1(b): Here, the intense NIR pulses transfer population from the longer-lived 2s3p state via the $2p^2$ dark state to the 2s2p state to repopulate the already decayed state.

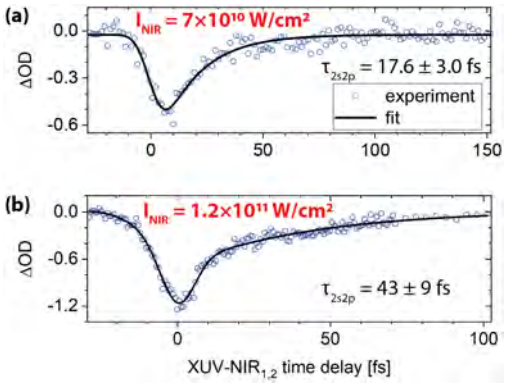


Figure 1. Measured lifetimes of the 2s2p doubly-excited state in helium by attosecond FWM spectroscopy given as differential absorbance ΔOD. (a) Natural lifetime and (b) elongated lifetime for higher NIR intensities due to Rabi cycling.

These experiments constitute an important step towards the direct characterization of core-level excited-state lifetimes in atoms and molecules. Furthermore, the strong-field approach adds to the toolbox of all-optical coherent population control techniques with potential applications in charge migration and attochemistry.

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RABBITT-HOP: RABBITT with Higher-Order Processes

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Synopsis The RABBITT technique enables precise extraction of amplitude and phase information from photoelectron spectra but is traditionally limited to weak IR fields and two-photon pathways. We present RABBITT-HOP, demonstrating how **H**igher-**O**rder **P**rocesses can be identified even at moderate IR intensities. We target the $1s4p$ and $1s5p$ states in He, and show how these resonances imprint in multiple, higher-lying sidebands and mainbands.

The RABBITT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) technique is a foundational tool in attosecond science, enabling the precise extraction of amplitude and phase information from photoelectron spectra. Traditional RABBITT is limited to weak infrared (IR) fields and two-photon pathways. We have generalised the framework through the RABBITT with Higher Order Processes (RABBITT-HOP) formalism. In this work we show how RABBITT-HOP can explain results from experiments and simulations with helium, and how, even at moderate IR intensities, the signatures of higher-order processes can be extracted from photoelectron spectra.

An attosecond pulse train is constructed to target the $1s4p$ and $1s5p$ states in He, such that the resonant pathways imprint the characteristic 'antiresonance' (See Fig. 1) into the phase extracted from the neighbouring sideband [1]. RABBITT-HOP then predicts that the same resonant pathways will contribute to higher-lying sidebands/mainbands via four-photon transitions, and that the antiresonance should manifest in the phase of higher frequency oscillations via four photon transitions. We verify these predictions with R-matrix with time-dependence [2] simulations of He, and identify the resonant signature in photoelectron signals in multiple sidebands and mainbands.

Moreover, via analysis of photo-electron angular distributions (PAD) we provide further evidence of these higher-order processes [3]. In particular, we use a heterodyne analysis approach, based on the delay-resolved anisotropy parameters to reveal the role played by the g_0 partial waves (reached by four-photon transitions) to the spectral phase shift in the PAD. Even though the amplitude of the g_0 wave is tiny compared to the s and d_0 waves, its *interference* is detectable. This provides an efficient method to resolve isolated partial wave contributions beyond the perturbative regime.

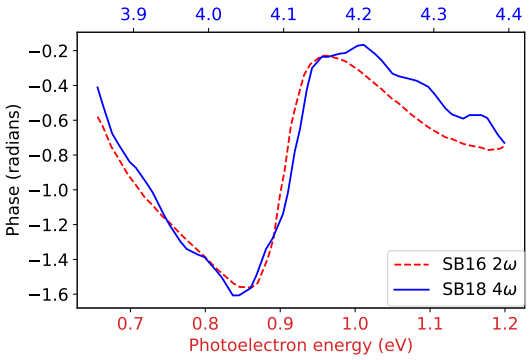


Figure 1. The sideband phase of the $2\omega_{\mathrm{IR}}$ oscillation in SB16 (red, dashed) and $4\omega_{\mathrm{IR}}$ oscillation in SB18 (blue, solid). The SB18 phase is shifted down by π radians to aid comparison. The characteristic 'antiresonance' profile is indicative of the role of pathways through the $1s4p$ and $1s5p$ states. The fact that the same phase profile is visible in the higher order, 4ω oscillation of the higher lying sideband is confirmation of the multiphoton mechanisms we report.

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Ultrathin liquid flat jet system for attosecond spectroscopy of solvated molecules

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Synopsis Water serves as the native physiological environment for a multitude of biochemical reactions, yet probing molecular dynamics in solutions at the attosecond time-scale remains challenging. Flat liquid jets with a sub-micrometer thickness make it possible to preserve the duration of few-femtosecond pulses while remaining below the penetration depth of extreme ultraviolet and soft X-ray wavelengths. We present the development of a table-top soft X-ray attosecond beamline integrated with a liquid flat jet system, enabling the study of electronic and vibrational dynamics in solvated molecules.

Liquid microjets offer optimal conditions for spectroscopic investigations at short timescales, with tunable thicknesses down to the sub-micrometer level [1, 2]. Their short interaction lengths minimize dispersion and absorption of ultrashort light pulses, preserving the temporal resolution necessary for attosecond spectroscopy in solution.

Recent advancements allow the generation of flat liquid sheets using gas-dynamic nozzles [3]. However, operating these systems in vacuum introduces significant technical challenges as high solvent evaporation rates, freezing, and nozzle clogging demand a careful engineering of the liquid jet chamber to ensure stable and uninterrupted experiments.

Here, we present an innovative liquid jet endstation aiming at simplifying the delivery of the liquid sample. This includes a quick venting system that separates low and high vacuum regions, allowing efficient solvent vapor removal, and a recycling system that permits the study of precious molecular samples in solutions available in limited quantities, enhancing the versatility of the setup.

The liquid jet chamber is positioned downstream of a high-order harmonic generation (HHG) light source, which produces ultrashort pulses via a home-built optical parametric amplifier (OPA) system operating at 1.5 μm. Our setup achieves cutoff energies within the so-called water window, spanning from the carbon K-edge (284 eV) to the oxygen K-edge (533 eV) [4, 5]. In this spectral range, water exhibits low absorption, enabling element- and site-specific spectroscopy [6, 7] and providing ideal conditions for investigating biologically relevant samples.

Potential applications of this system include the exploration of coherent electron dynamics and charge transfer processes in solvated molecules, as well as the detection of transient chiral responses in solution with few-femtosecond resolution.

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Conditions for Mollow Triplets in Ultrafast Absorption and Dynamical Fluorescence

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Synopsis The existence of Mollow-like triplets in the transient absorption spectra of atoms in strong resonant fields is predicted and their conditions are determined. The triplet depends on the shape of the driving pulse, being attainable by idealised flattops, but not commonly used Gaussian pulses. We propose the use of super-Gaussian pulses that can support the absorption triplet on ultrafast timescales. These absorption triplets may be measured in the parameter space enabled by seeded free electron lasers.

During the last decade, transient absorption spectroscopy has been used to measure coherent electronic dynamics with unprecedented resolution down to the attosecond time scale [1]. The Mollow triplet is well-known to exist in resonance fluorescence [2, 3, 4], however, Mollow-like absorption triplets have so far not been reported. This may be due to inadequate laser conditions, which are now changing with the rapid development of seeded free electron lasers, making the existence of Mollow-like absorption spectra a timely question. We employ strong-field transient absorption theory [5] for resonant XUV-FEL pulses in hydrogen to elucidate Mollow-like absorption triplets, with striking similar to the fluorescence spectra, Fig. 1a (flattop fields). We present general conditions for such triplets [6],

$$\mathcal{A}_n < \tau/T_{\mathrm{Rabi}} < \mathcal{B}_n.$$

To support the absorption triplet the atom must undergo at least one Rabi cycle and the pulse bandwidth must exceed the Rabi frequency. The Gaussian has a small time-bandwidth product and can not support the triplet. In contrast, the flattop's modulations support the triplet for all pulse durations. The super-Gaussian is an intermediate case and depends on the pulse area, see Fig. 1b. This is compared to the characteristic Fluorescence wings, calculated using the quantum optics model [3], as seen in Fig. 1c. In the high-intensity limit (not shown), the central absorption peak becomes asymmetric, which we attribute to atomic non-linear photoionization. In summary, conditions for Mollow-like triplets in absorption spectroscopy were presented, which may find applications in future FEL experiments.

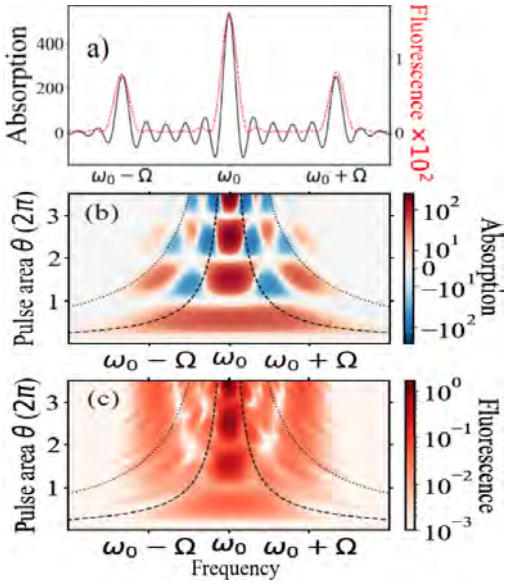


Figure 1. Dynamical Mollow triplet buildup. (a) Absorption and fluorescence spectra for a flattop field. Pulse-area (duration) resolved absorption (b) and fluorescence (c) spectra for a super-Gaussian.

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Absolute photoemssion timing and asymmetry parameter of the I4*d* giant resonance of the Iodoalkanes

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Synopsis We investigated the I4*d* → *f* photoionization with various different ligands attached to the iodine atom. The main tool was the attosecond streaking technique, performed at our beamline at the Technical University of Munich, to infer the photoemission delay relative to a helium reference. Additionally we measured the asymmetry parameter for some of the molecules at the ELETTRA synchrotron. All measurements were taken in the gas phase with unoriented molecules.

1. Our goal was to find a relation between photoemission delay and length of the carbon chain, building on previous work[1][2]. We found a dependence on the molecule species, however the picture is not as clear as predicted, longer chain does not lead to a larger delay.

2. The correlation we found is based on isomerism, those molecules with conformational isomers show a higher delay than those without. Those with isomers have a value close to the atomic calculation for Iodine[3].

3. The most obvious physical effect for the species dependent behavior is intramolecular scattering, the outgoing photoelectron wave packet scatters at the ligand before being exposed to the infrared probe pulse.

4. To tackle intramolecular scattering further, we investigated the asymmetry parameter, as it has been shown to be influenced by molecular surroundings and intramolecular scattering[4]. Our measurements were performed at the ELETTRA synchrotron, due to limited beam time only for Iodomethane, Iodoethane, 1-Iodopropane and 2-Iodopropane.

5. While the Iodomethane value was reproduced according to previous studies and 2-Iodopropane yielded very similar values, Iodoethane and 1-Iodopropane showed a significant deviation. Both photoemission delay and photoemission asymmetry of I4*d* show a dependence on the molecular environment, however not in the same way, so the underlying effect evades an intuitive explanation.

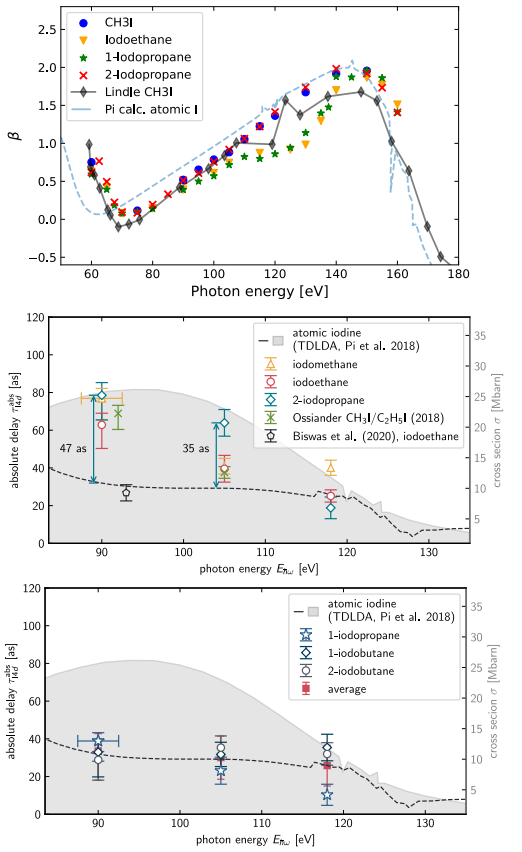


Figure 1. Top: Asymmetry parameter measurements. Mid: molecules without conf. iosmers. Bottom: molecules with conf. isomers.

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Attosecond resonant Auger spectroscopy

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Synopsis Ultrashort pulses are essential for achieving high temporal resolution in x-ray pump-probe spectroscopy. However, as x-ray probe pulses approach the attosecond regime, conventional x-ray photoelectron and x-ray absorption techniques suffer from severe spectral broadening, limiting their resolution [1]. In this work, we explore attosecond resonant Auger spectroscopy (RAS) as a probe in several pump-probe scenarios, achieving simultaneous high temporal and spectral resolution [2-4] thus providing an alternative to leverage sub-femtosecond x-ray probe pulses.

We present theoretical simulations for the UV-pump x-ray probe scenario with ultrashort pulses where RAS provides insight into core-to-valence electronic transition and impresses a rich fingerprint of the electronic structure and nuclear configuration at the time-initiating RAS process. Specifically, we propose using sub-femtosecond x-ray pulses to trigger RAS in a distorted molecule, which is generated from the nuclear evolution on a valence excited state pumped by a femtosecond ultraviolet pulse. With the time delay varied, the amount of molecular distortion can be controlled and the RAS measurements imprint both their electronic structures and changing geometries.

We demonstrate this approach in H_2O prepared in a dissociative valence state, where molecular and fragment lines appear in RAS spectra as signatures of ultrafast dissociation (Figure 1) [2]. While RAS can recover the vibrational structures not hidden by broadband excitation, the full reconstruction of the wavepacket is a standing challenge. Using CO as a case study, we show that time-resolved RAS enables retrieval of complete nuclear wavepacket information, including vibrational state populations and relative phases [3]. Furthermore, we illustrate how the attosecond RAS probe technique can detect ultrafast nonadiabatic electron-nuclear dynamics through conical intersections triggered by an ultrashort UV pump pulse [4].

A similar spectral resolution enhancement is observed theoretically in normal Auger decay process, where a core-shell electron is ionized by coherent ultrashort x-ray pulse leading to a formation of a cationic nuclear wave packet, studied via coincidence detection of photoelectrons, Auger electrons, and ion fragments [5].

Given its broad applicability to various molecular systems, attosecond RAS offers a powerful alternative pump-probe technique for mapping core and valence dynamics with ultrashort x-ray pulses.

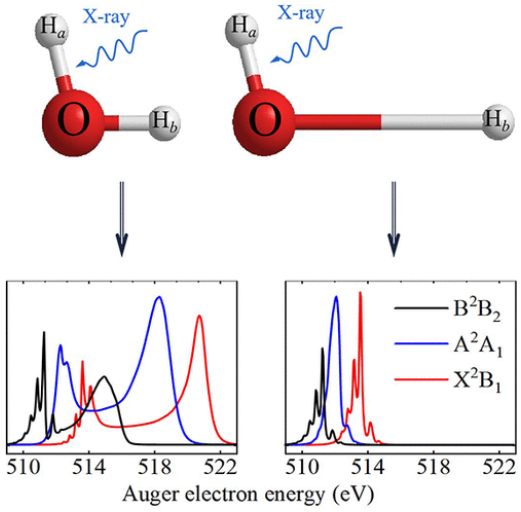


Figure 1. UV-pump x-ray probe of ultrafast fragmentation of water molecule with attosecond RAS.

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Attosecond partial wave meter and its application in resolving the photoemission time delays in atoms and molecules

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Synopsis Attosecond chronoscopy is central to understanding the ultrafast electron dynamics in matter with attosecond temporal resolution. We developed a polarization-controlled attosecond photoelectron spectroscopy serving as a partial wave meter to reveal the role of each partial wave during the photoionization process. The time delays within the quantum interference between the degenerate two-photon ionization pathways in neon atoms, and the angle-resolved continuum-continuum time delays in molecular shape resonance were experimentally and theoretically explored.

Photoionization is a process taking place on attosecond time scales. How its properties evolve from isolated particles to the condensed phase is an open question of both fundamental and practical relevance. Attosecond light pulses permit real-time observation and precise manipulation of ultrafast electron dynamics[1-4]. One of interesting questions in such attosecond physics is the photoelectron emission time-delays which are attributed to the energy dependence of the scattering phase shift during electron transitions in a short-range potential, Coulomb potential, and the laser-Coulomb-coupling effect in the continuum.

Based on an advanced attosecond coincidence metrology, we employed a polarization control scheme, where the polarization axes of the XUV-APT and NIR laser pulses are rotated from a parallel to a perpendicular orientation, to serve as a partial wave meter[5,6]. This polarizationskewed scheme allows us to monitor the photoelectron emission dynamics in time and space simultaneously by steering the different partial waves in the final sideband electrons.

We firstly used this metrology to deduce the pathway interference within the quantum 'black box" for the two-photon ionization of neon atoms. Based on the angle dependent and attosecond time-resolved photoelectron spectra, we resolved the quantum interference between the degenerate p → d → p and p → s → p two-photon ionization pathways. We also extend this metod to investigate the angular dependence of the continuum-continuum delays in NO and CH_3F molecules. A clear angle dependent two-photon ionization time delay differene was observed between parallel and perpendicular situation. Our experimental and theoretical study establishes a connection between the experimental observable and the underlying partial wave characters and reveals the interference effect of two-photon transition pathways.

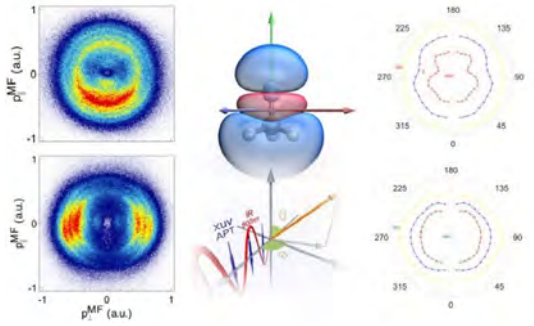


Figure 1. Pathway- and angle-resolved photoemission time delay in CH_3F molecules.

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Nonlocal resonance theory for electron–nuclear dynamics in molecules

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Synopsis We extend the nonlocal resonance theory from field-free electron–molecule collisions to external fields to study the effect of field-driven nuclear dynamics and electronic couplings in photoionization of neutral molecules and photodetachment of molecular ions. The basic theory of one-photon processes is demonstrated and the extension to two-photon processes is derived and illustrated on simple models of molecules.

We study nuclear dynamics in photoionization by extending the nonlocal resonance theory, commonly used to study inelastic electron–molecule collisions [1], to light–matter interactions. The nonlocal resonance theory describes coupled electronic–nuclear dynamics by Feshbach partitioning of the Hilbert space to resonant and background parts. This approach enables accurate treatment of nuclear dynamics in light–matter processes even in multi-electron systems, which has previously been limited to a few small systems [2]. The nonlocal, complex, and energy-dependent effective nuclear potential that is central includes all the electronic effects fully and is necessary to properly describe e.g. threshold effects.

In our upcoming work we present the theory and apply it to one-photon detachment of diatomic molecular anion models [3]. In Figure 1 we show a cross section from one-photon detachment of LiH^- obtained using the nonlocal resonance theory. The photodetachment cross sections show contributions from three separate processes: direct detachment, autodetachment, and direct detachment followed by reattachment and autodetachment. The model is constructed to qualitatively resemble LiH^- and is determined completely by the potential curves of the ground anion state, the neutral molecule, a resonant excited anion state (commonly discrete-state-in-continuum), together with the electronic coupling between the discrete state and the continuum, and the dipole transition elements between the ground anion and the discrete state and the continuum, respectively. Quantitative models of more involved systems may be given by fitting of eigenphase sums to fixed-nuclei calculations, or by direct implementation of the Feshbach projector in the outer region R-matrix calculations.

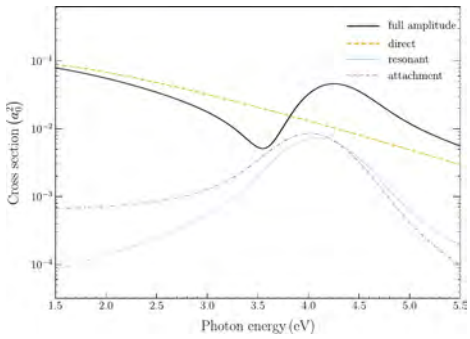


Figure 1. One-photon detachment cross section of LiH^- anion model using nonlocal resonance theory.

Furthermore, we have extended the nonlocal resonance theory to two-photon processes and derived the equations for the two-photon dipole matrix elements. The two-photon theory contains additionally the full nuclear dynamics associated with photon absorptions in the ionization continuum and will be used in combination with the one-photon theory to study the interplay between nuclear and electronic dynamics in RABBIT of non-polar and laser-dressed polar molecules [4, 5].

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Ultrafast Time-varying Nonlinearity at Epsilon-near-zero

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Synopsis Our study explores the ultrafast time-varying nonlinear response of a 310 nm Indium Tin Oxide (ITO) thin film using a time-resolved pump-probe setup, identifying the intensity-dependent optical amplitude and phase modulation. We observe distinct rise and relaxation rates, as well as power-dependent bulk plasma frequency shifts. These findings enhance our understanding of Epsilon-near-zero (ENZ) materials and contribute to advancements in ultrafast optics and time-varying nanophotonics.

Epsilon-near-zero (ENZ) materials, particularly transparent conductive oxides (TCOs), exhibit extraordinary nonlinear optical properties that enable time-varying modulations of light-matter interactions, offering promising avenues for ultrafast optical applications, such as all-optical signal control [1] and novel time-varying optical designs [2]. Among them, Indium Tin Oxide (ITO) has drawn significant attention due to its ability to undergo refractive index changes of order 1 upon optical excitation. This study presents a comprehensive investigation of the ultrafast nonlinear response of a 310 nm ITO thin film using a time-resolved pump-probe spectroscopy setup. A near-infrared pump (795 nm, 44 fs) excites the electron dynamics, while a 50 fs probe at the ENZ resonance wavelength 1250 nm records the optical modulation over a range of intensities up to 9.2 TW/cm^2. By varying the pump intensity, we identify three distinct nonlinear regimes of the time-varying modulations: (i) a low-intensity regime (0–2 TW/cm^2) where the modulations are proportional to the pump power, (ii) a saturation regime (2–6.6 TW/cm^2) where reflectivity and transmissivity exhibit a plateau, and (iii) a high-intensity regime (6.6–9.2 TW/cm^2) with even stronger nonlinear responses. Additionally, we observe different relaxation rates in the transient nonlinear response, linked to distinct thermal electrons and lattice cooling dynamics. Both the rise and decay rates show interesting dependence on the pump power. We further analyze the time-dependent wavelength shifts in both reflection and transmission, showing a transition from pure redshift at low intensities, to a combination of red- and blueshift at higher intensities, which is attributed to the decrease and increase of the bulk plasma frequency, respectively. Modeling results are also presented to support the experimental data. Our findings provide crucial insights into the ultrafast time-varying response of ENZ materials and will bridge the ultrafast laser and time-varying physics communities.

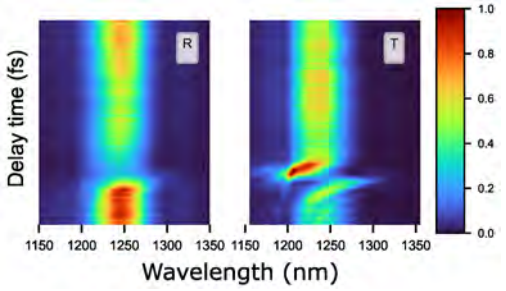


Figure 1. The time-varying probe spectrum of the reflected (left) and transmitted (right). Strong amplitude and wavelength modulations can be observed due to the nonlinear response of the ITO.

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Symmetries, dominance and quantum interference in below-threshold non-sequential double ionization with linearly polarized two-color fields

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Synopsis We investigate laser-induced nonsequential double ionization with linearly polarized bichromatic fields, focusing on recollision-excitation with subsequent ionization. Using the strong-field approximation, we analyze how field symmetries influence dominant events. By tuning frequencies and relative phase**,** we show how to control correlated electron-momentum distributions. Analytic quantum interference conditions are extended to arbitrary fields, showing how specific field features can confine distributions and interference patterns to specific momentum regions

Tailored fields are powerful tools for controlling laser induced processes like non-sequential double ionization. A systematic way to understand the imprint of the field and the type of the target on the resulting photoelectron momentum distributions is provided by symmetry. We investigate the recollision-excitation with subsequent ionization (RESI) mechanism with the (ω,2ω) and (ω,3ω) driving fields. Using the strong-field approximation, we assess how the symmetries of the field influence the dominant events. By manipulating the field parameters such as the field frequencies and relative phase between the two driving waves, we show how one can influence the correlated electron-momentum distribution [1].

In addition, quantum interference persists in RESI despite focal averaging and transverse momentum integration [2], thus challenging the notion that is is a classical type of electron-electron correlation. In this work, we perform a systematic analysis of quantum interference by elaborating several analytic interference conditions for RESI in arbitrary driving fields, with a focus on interference arising from the specific symmetries of bichromatic fields. For example, for waves of comparable strengths, multiple solutions per half cycle for the second electron must be included. This generalizes previous work, in which a single event per half cycle was considered [3].

We disentangle superimposed interference fringes originating from phase differences related to symmetrization, temporal shifts and a combination of exchange and event interference, revealing a rich assortment of patterns, such as 'spine lines', hyperbolae, chequerboards and even 'feathers'. We are able to predict such patterns from the constituent building blocks of each type of interference [4]. Such predictive power may be useful in controlling or shaping RESI interference patterns in specific momentum regions.

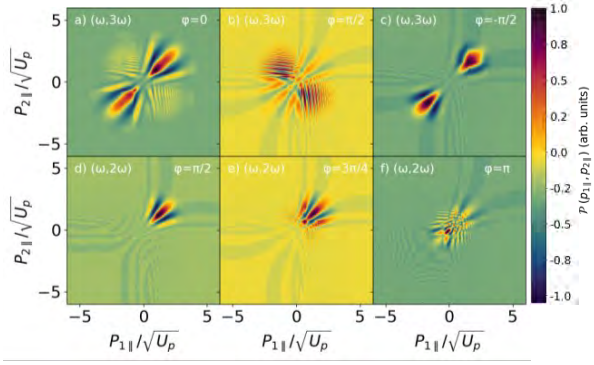


Figure 1. Interference patterns associated with symmetrization, temporal shifts and combined temporal-exchange shifts for relative phases indicated in the panels for the (ω,3ω) [top row] and (ω,2ω) [bottom row] driving fields for the 3s→ 3p transition in Argon. The ratio of the field amplitudes is ξ = 0.8. The intensity of the ω field component is 6×10^{13} W/cm^2 and the fundamental wavelength is 800 nm.

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Attosecond Fourier Transform Spectroscopy

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Synopsis The interaction of attosecond pulses with a quantum system captures its sub-cycle temporal evolution. However, their broad spectral bandwidth, spanning tens of eV, excites a wide range of quantum states that contribute to the optical signal, hindering the identification of their individual dynamics. We integrate the enhanced spectral resolution and signal-to-noise ratio of Fourier transform spectroscopy with the sub-cycle temporal resolution of attosecond metrology to follow the spectro-temporal evolution of individual meV-spaced light-induced states with attosecond resolution.

Attosecond science has revolutionized our ability to observe electron dynamics on their natural timescales by employing attosecond pulses to capture the evolution of quantum states with sub-cycle accuracy [1,2]. However, the broad spectral bandwidth of attosecond pulses -- spanning tens of eV -- excites a wide range of quantum states that are coherently imprinted onto the optical signal. The limited spectral resolution of existing extreme-ultraviolet (XUV) spectrometers hinders the ability to distinguish the dynamics of individual quantum states, presenting an important challenge for attosecond metrology.

We address this challenge by integrating the basic principles of Fourier transform spectroscopy [3] (FTS) with attosecond science. FTS is one of the most powerful techniques in optical metrology, offering enhanced spectral resolution and improved signal-to-noise ratio. Here, we extend this concept to the attosecond domain, achieving meV spectral and sub-cycle temporal resolution.

We establish Two-Attosecond Pulse Interferometrically Resolved Spectroscopy (TAPIRS), generating an XUV interferogram by resolving the transmission of an interferometrically stable attosecond pulse pair through a helium sample, together with a driving IR laser (Figure 1a). Scanning τ_{XUV} while maintaining a fixed arrival time of the IR and second attosecond pulse allows to simultaneously control both XUV-XUV and XUV-IR delay in a 1D approach.

The Fourier analysis of the measurement with respect to τ_{XUV} reveals the underlying linear (XUV only) and nonlinear (XUV+IR) resonant excitations in a background-free manner (Figure 1b). This two-dimensional frequency map identifies the complex amplitude of the laser-induced electronic coherences within the attosecond pulse's bandwidth [4]. The high resolution and contrast provided by the FTS measurement allows us to clearly distinguish the contribution of individual meV spaced states, while following their modification during the ultrafast light-matter interaction. Attosecond Fourier transform spectroscopy opens the door to resolving rich quantum dynamics in field-driven systems with unprecedented spectro-temporal precision.

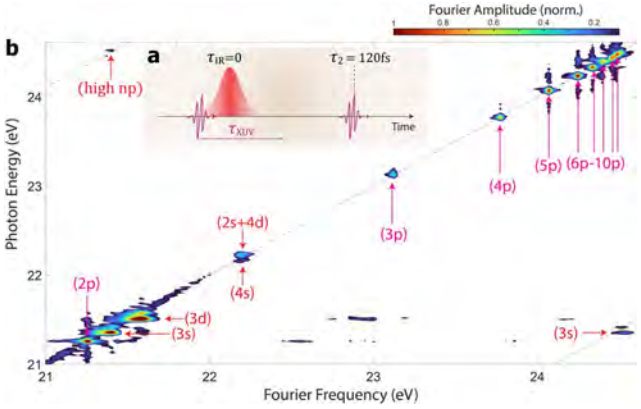


Figure 1. a, TAPIRS triple pulse sequence. **b,** Fourier analysis of the spectrally resolved interferometric measurement with respect to τ_{XUV}, revealing the underlying quantum states, separating between linear (XUV only, magenta) and nonlinear (XUV+IR, red) excitations within the helium *1snl* series.

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Electronic-vibrational dynamics in x-ray transient absorption of N_2^+ induced by strong-field ionization

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Synopsis Attosecond transient absorption spectroscopy (ATAS) has become a critical tool for investigating ultrafast dynamics in attochemistry, while the interpretation of measurements requires full consideration of couplings among various freedoms of motion. We develop a theoretical framework to explore coupled vibronic dynamics induced by strong-field ionization. Our simulations accurately reproduce experimental measurements and enable clear identification of contributions from each electronic state, which is essential for resolving the debate on nitrogen air lasing.

Strong-field ionization (SFI) of molecules creates extremely non-stationary ionic states that evolve coherently under the remaining laser field. The resulting electronic, vibrational and rotational excitation, as well as the quantum coherence make the molecular ions unique platform for investigating and manipulating the chemical reactions and extraordinary radiations such as air lasing and supercontinuum generation. However, unraveling the intricate multiple-freedom coupled sub-cycle ionic dynamics remains a significant theoretical and experimental challenge.

In this study, we develop the ionization-coupling model that incorporates transient absorption to explore the coupled electronic-vibrational dynamics induced by SFI [1,2]. By probing the ionic population and coherence using ATAS via tran- sitions from the nitrogen K-edge, we reproduce the measured absorption spectra of N_2^+ reported in [3]. Our results reveal that laser-induced electronic-vibrational coupling generates coherent vibrational wave packets with a broad distribution of vibrational levels, leading to spectral overlap in the K-edge absorbance. By identifying the contributions of each electronic state, we verify a remarkably high population of the excited A state, highlighting the importance of electronic-vibrational coupling in strong laser fields. Furthermore, we discover novel absorption features arising from forbidden transitions occurring during the laser pulse and demonstrate quantum beating in absorption induced by vibronic coherence.

This work establishes a theoretical framework for investigating the strong-field-induced intricated molecular dynamics that governs ionic radiation and absorption dynamics, charge migration and chemical reactions, thereby advancing attochemistry studies on photoionization-initiated dynamics and coherence.

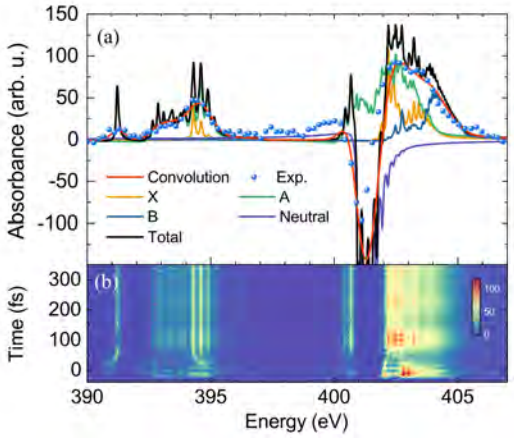


Figure 1. (a) The calculated time-integrated XAS of N_2^+ (red line) is compared with the experimental results from Ref. [3] (blue dots). (b) The transient XAS of N_2 molecules exposed to strong laser pulses.

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Photoemission Timing of Surface-Oriented Iodoalkanes on Pt(111)

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Synopsis We measured the photoemission delay of the I4d state of iodomethane and iodoethane adsorbed on Pt(111) via attosecond streaking with an excitation energy of 105 eV. The self-assembly process of these molecules allows for control of their orientation to the surface. This enabled the investigation of the delay of the I4d state with respect to the Pt valence band under a fixed angle. We show that there is a significant difference in delay between vertical and horizontal molecular orientation for iodomethane and iodoethane.

Delays in photoemission have been found in atoms in the gas phase and in condensed matter (c.f. e.g. [1, 2]) with attosecond streaking. Then, streaking was also used to investigate the photoemission delays of molecules in the gas phase (e.g. [3]). However, these gas phase measurements lack information about the molecular orientation's effect on the photoemission delay. Hence, the measured delay is only the average of an angle-dependent delay over the whole solid angle.

This study overcomes the problem of lacking molecular orientation by advancing attosecond streaking measurements of molecules to solids. For that, iodomethane and iodoethane were adsorbed on a Pt(111) surface, as their self-assembly process on Pt(111) has been studied exhaustively [4, 5]. It has been shown that both molecules align vertically or horizontally to the surface, depending on surface coverage. Therefore, varying the surface coverage enables photoemission delay measurements of these iodoalkanes under a fixed angle.

The iodoalkane\Pt(111) samples were excited with 105 eV XUV pulses with a bandwidth of 5 eV. This allowed us to extract the photoemission delay between the I4d state and the Pt valence band state. In the case of vertical molecular alignment, the I4d photoelectrons first have to traverse the molecular chain before reaching the continuum, while when having horizontal alignment, the I4d photoelectrons won't traverse the full molecular potential environment. This results in a significant jump in the photoemission delay at the coverage where the molecular orientation changes. Figure 1 exemplarily shows the coverage-dependent photoemission delay $\tau_{\mathrm{I4d}} - \tau_{\mathrm{PtVB}}$ for iodomethane on Pt(111).

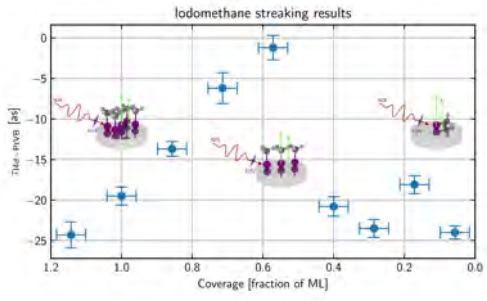


Figure 1. Coverage dependent photoemission delay of I4d and Pt valence band for ICH_3\Pt(111) at 105 eV excitation energy. We explain the first relative delay decreasing towards lower coverages with potential density effects on the surface. The jump in delay occurring at 0.5 ML is attributed to the change in molecular surface orientation.

Similar behavior in delay has also been found for iodoethane. Finally, to make the results more comparable to the gas phase, the photoemission from the Pt(111) surface was timed absolutely by using atomic iodine as a chronoscope.

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Attosecond ionization delay arising from Feshbach-resonance of N_2

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Synopsis We experimentally show that electron correlation can induce ionization delays in nonresonant channels within the Feshbach resonance region. Using a high-resolution attosecond interferometer, we captured the ionization dynamics of N_2 molecules. These observations reveal the intricate interactions between electrons and vibrations during molecular ionization.

Attosecond photoelectron spectroscopy has become a vital tool for probing electron dynamics in matter. With advancements in attosecond spectroscopy, researchers are now able to investigate the lifetimes of electron states and explore their dynamic behavior in greater detail. Shape and Feshbach resonances have attracted significant attention due to their strong molecular structure dependence, which directly influences ionization dynamics.It has been shown that single-channel ionization can significantly influence other nonresonant ionization channels. Further studies have revealed that ionization delays are highly sensitive to emission angles and bond lengths [1-3], and that shape resonances can introduce delays into other channels via interchannel coupling [4]. However, the impact of Feshbach resonances on vibrational-state dependence, particularly how electron-correlation effects mediate photoionization delays, is still not fully understood.

In this work, we extract the temporal information of the electron emission using the RABBIT (Reconstruction of Attosecond Beating by Interference of Two-Photon Transitions) technique [5]. This method leverages the interference between two-photon transitions to accurately measure attosecond-level time delays. We utilized extreme ultraviolet (XUV) radiation, composed of high-order harmonics, to ionize N_2 molecules and study the vibrationally dependent ionization dynamics in the Feshbach and shape resonance regions.We observe a variation of the photoionization delays of approximately 247 as across the resolved vibrational states($\nu = 0$ to 6) of the $A^2\Pi_u^+$ state of N_2^+. These results indicate that Feshbach resonances induce significant delays between vibrational states through electron correlation.

This study provides experimental evidence for understanding how Feshbach resonances influence molecular photoionization processes, with particular emphasis on the role of electron-correlation effects in vibrational-state dependencies. The attosecond time-resolved approach we used allows for a deeper exploration of the intricate interactions between electrons and vibrations in molecules. It offers an important experimental platform and theoretical basis for future investigations of similar molecular systems.

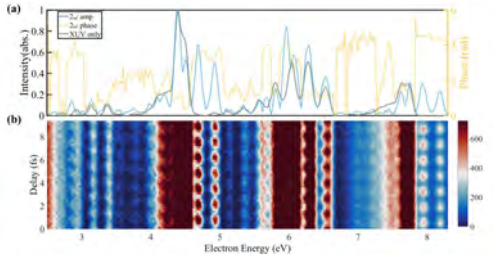


Figure 1. photoelectron spectra.(a) The 2ω amplitude (blue),2ω phase (orange), XUV-only spectrum (gray).(b)Photoelectron spectrum of N_2.

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Quantifying Centrifugal-Potential Effects: Channel-Resolved Attosecond Time Delays in Nondipole Photoionization

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Synopsis While advances in attosecond technology enable unprecedented temporal resolution, attosecond-resolved nondipole effects in nonlinear photoionization remain unexplored due to weak signals obscured by the dominant dipole background. Using angle-resolved two-photon interferometry, we characterize both dipole and nondipole electronic partial waves' complex amplitudes via *channel-resolved analysis*. Notably, we reveal a relative group delay **∼10 as** of **nondipole** waves than dipole ones. This quantifies how *centrifugal potentials* influence nondipole bound-continuum and continuum-continuum transitions.

Recent advances in attosecond spectroscopy, e.g., the RABBIT technique, have enabled precise measurement of photoionization time delays arising from dipole-allowed transitions. In the RABBIT scheme, the measured photoionization time delays are related to the electronic phase of two-step quantum transitions, the bound-continuum (BC) transition and the continuum-continuum (CC) transition. Recently, the photoionization time delay of individual electronic partial waves is disentangled in the angular-resolved RABBIT measurements [1, 2], elucidating the role of the short-range, Coulomb and centrifugal potentials on the dipole-allowed electronic dynamics.

In light-matter interaction, nondipole effect (e.g., electric-quadrupole and magnetic-dipole transitions, photon momentum transfer) becomes prominent for high photon energies and laser intensities, especially in the circumstances of vanishingly small dipole-allowed dynamics. Hopefully, the study of nondipole interaction prospers novel manipulation of both light and matter. While nondipole effects are known for a characteristic shift of photoelectron distribution, the associated phase information (or time delay) of nondipole quantum transitions is inaccessible from the angular distribution of photoelectrons in single-photon ionization. Recently, the two-photon pump-probe experiment has revealed the effect of the electric-quadrupole transition on the photoionization time delay of helium [3, 4], wherein all partial waves are mixed. Here, by using circularly polarized XUV APT and IR pulses, we disentangle both modulus and phase of individual dipole and nondipole partial waves and thus provide a complete characterization of nondipole photoionization. Intriguingly, we report on a notable photoionization time delay of around 10 attoseconds between the nondipole and dipole partial waves in helium, as shown in Fig. 1. This quantifies how *centrifugal potentials* influence nondipole BC and CC transitions [5].

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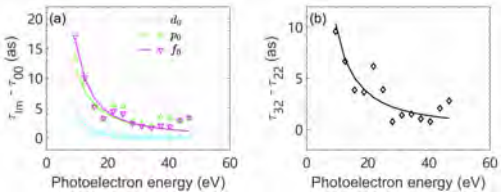


Figure 1. Relative group delays of different partial waves.

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Beyond-SFA Phase Measurements in Non-perturbative Single-color HHG spectroscopy

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Synopsis We introduce a new measurement scheme in HHG spectroscopy based on a non-perturbative single-color modulations of the recollision mechanism and XUV-XUV interferometry. Our measurement resolves clear phase deviations between Argon and N_2, despite their similar ionization potential, revealing the rich underlying dynamics that go beyond the strong field approximation picture.

High harmonic generation (HHG) spectroscopy encodes electron dynamics into the amplitude and phase of the emitted XUV spectrum. The widely successful Strong-Field Approximation (SFA) describes HHG using a recollision model, where the emitted phase and intensity are dictated primarily by the ponderomotive energy U_p and ionization potential I_p, capturing the light driven electronic dynamics [1], while neglecting rich underlying effects such as multielectron interactions and bound-state dynamics. Previous all optical perturbative measurements have confirmed this universal picture, demonstrating an excellent agreement between SFA predictions and measured phase [2]. While advanced measurements have resolved dynamics beyond the SFA [3], its identification remained hidden in many HHG experiments.

We introduce a new measurement scheme which combines two time-delayed IR fields, while resolving the complex HHG signal via XUV-XUV interferometry [4]. By interfering two single-color IR beams, we systematically modulate the driving field intensity over a broad dynamical range, in a non-perturbative, periodic manner. Performing XUV-XUV interferometry resolves the complex response of the HHG process – identifying both amplitude as well as phase modulations. This approach enables a direct comparison between atomic Argon and molecular nitrogen (N_2), two systems with similar I_p, yet distinct electronic and ionic structures.

Figure 1 presents the experimentally resolved phase of harmonic 15, measured in N_2 and in Ar, together with the SFA prediction. This measurement resolves high-frequency phase modulations that are not captured by the SFA. This new metrology scheme captures higher-order corrections to the semi-classical trajectory picture, allowing the observation of complex attosecond scale electron dynamics, so far hidden in many experimental observations.

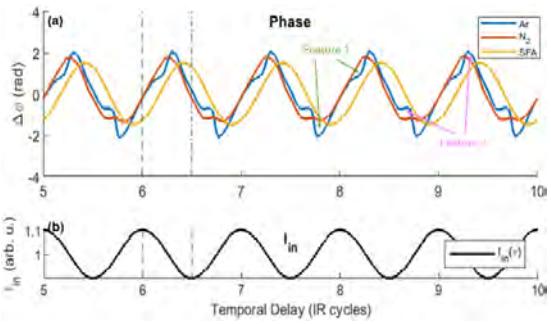


Figure 1. (a) Spectral phase modulation for Harmonic 15 as a function of temporal delay. Results are shown for Argon (blue), $\boldsymbol{N_2}$ (red), and SFA predictions (yellow). **(b)** Estimated driving field intensity. Vertical dashed and dot-dashed lines mark the delays corresponding to maximal and minimal input intensity, respectively. Key deviations from the SFA for a given input field are highlighted with arrows. Feature 1 (green) marks a 0.8 radian deviation between Argon and $\boldsymbol{N_2}$.

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Analytical Expression for Continuum-continuum Transition Amplitude of Hydrogen-like Atoms

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Synopsis We report an analytical expression for the continuum-continuum (CC) transition delays, which quantitatively agrees with TDSE including in the low-kinetic-energy region, confirmed by the angle-resolved RABBIT experiments. We also predict a Cooper-like minimum in CC transitions due to the centrifugal potential.

Attosecond chronoscopy typically utilizes interfering two-photon transitions to access the phase information, e.g. attosecond beating by interference of two-photon transitions (RABBIT). Simulating these two-photon transitions is challenging due to the continuum-continuum (CC) transition term. The hydrogenic approximation within second-order perturbation theory has been widely used but only (partially) asymptotic formulae have been available so far, which fail to correctly describe the low-kinetic-energy behavior, especially for high angular-momentum states. Here [1], we report an analytical expression based on the Appell's F_1 function [2] that overcomes these limitations. We show that the formula quantitatively agrees with the numerical simulations using the time-dependent Schrödinger equation (TDSE) as well as the experimental angle-resolved RABBIT measurements [3, 4, 5], which improves the accuracy compared to the other analytical approaches that were previously reported. Furthermore, for states with high angular momenta, the analytical formula predicts that the absorption pathway of $\ell \to \ell - 1$ has a Cooper-like minimum, which affects its CC time delay, manifesting that the centrifugal potential can lead to non-trivial structures in the above-threshold transitions.

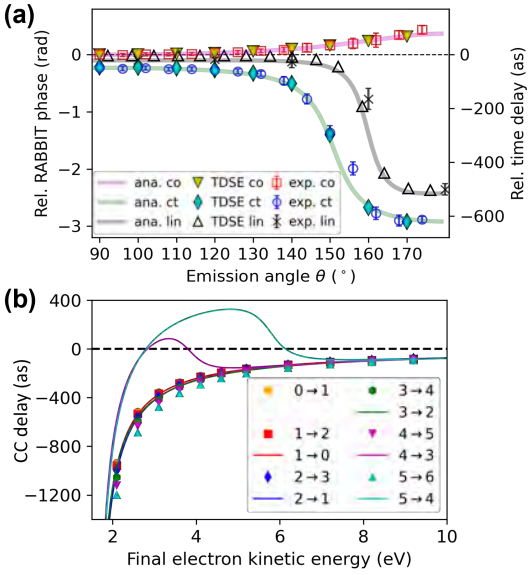


Figure 1. (a) Angle-dependent RABBIT phases and corresponding relative time delays of helium for co-rotating (co), counter-rotating (ct) and linear-linear (lin) XUV and dressing IR (800 nm) with final-state kinetic energy of 3.3 eV (sideband 18), referenced to the emission angle of 90°. The results based on one-photon transition amplitude calculated by `ePolyScat` combined with the analytical CC formula are indicated by the solid lines, while the TDSE results [3, 4] and the experimental values [3, 5] are shown as the symbols and the symbols with error bars, respectively. (b) The CC time delay defined as $\tau_{\mathrm{CC}} = \hbar(\phi^{\mathrm{emi}} - \phi^{\mathrm{abs}})/(2E_{\mathrm{CC}})$ with 800 nm dressing IR ($E_{\mathrm{CC}} = 1.55$ eV) for various ℓ-dependent channels, labeled as "intermediate $\ell \to$ final ℓ" and indicated by symbols for $\ell \to \ell + 1$ and lines for $\ell \to \ell - 1$, respectively.

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HHG-based all-attosecond transient absorption spectroscopy

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Synopsis We demonstrate time-resolved all-attosecond transient absorption spectroscopy (AATAS) using a table-top high-harmonic generation (HHG) source. This enables the observation of electronic coherences in Xe, where oscillating valence electron motion on a 3-fs timescale is observed. Our investigation is extended to Kr, Ar, and Ne, showcasing AATAS as a powerful and accessible tool for ultrafast spectroscopy.

Attosecond transient absorption spectroscopy (ATAS) has emerged as a powerful tool for probing electron dynamics, combining extremely high temporal resolution with excellent spatial resolution [1]. Traditional ATAS using a NIR pump or probe pulses is, however, not able to access timescales ranging from a few hundred attoseconds to a few femtoseconds. This can be achieved by performing all-attosecond transient absorption spectroscopy (AATAS). The first demonstration of AATAS in water was recently reported from LCLS [2].

This work demonstrates table-top AATAS using an HHG source [3]. Although the pulse energies obtained from HHG are orders of magnitude lower when compared to free-electron lasers, XUV intensity scaling using an 18-m-long beamline has allowed us to approach the saturation intensities of ionization [4]. While the transient signal changes in our experiments are comparable to the LCLS results [2], HHG benefits from the high accessibility and stability. By implementing a cascaded post-compression scheme [5], low fluctuations of both the NIR (0.17 % r.m.s.) and the XUV (0.67 % r.m.s.) pulse energies were obtained. Moreover, unlike previous realizations of all-attosecond pump-probe spectroscopy, our experiments were performed at 1 kHz [5,6], resulting in improved statistics.

AATAS results obtained in Xe are presented in Fig. 1. The attosecond pump pulse ionizes Xe, thereby creating Xe^+ in a superposition of spin-orbit states (Fig. 1(a)), which initiates coherent electron wavepacket motion. An oscillating signal with a period of 3.1 fs is observed in the AATAS map (Fig 1(b),(c)), which is in agreement with the 1.31 eV energy splitting between the $5p_{3/2}^{-1}$ and $5p_{1/2}^{-1}$ states. The high stability and robustness of our source have enabled systematic investigations of electronic coherences in Kr, Ar, and Ne.

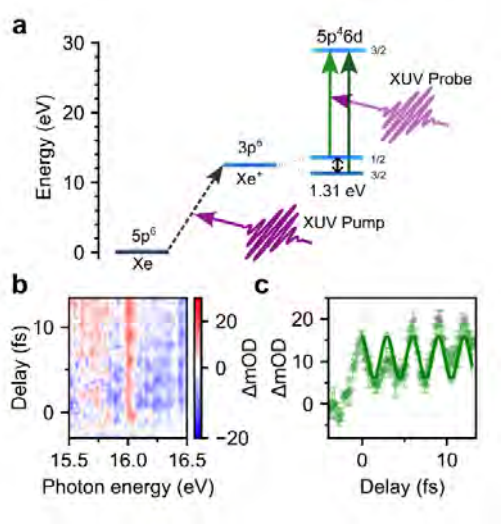


Figure 1. (**a**) An attosecond pump pulse ionizes Xe, creating a superposition of spin-orbit states. Resonant absorption is probed by a second, delayed attosecond pulse. (**b**) AATAS map, showing transient optical density (OD) changes vs. photon energy and delay. (**c**) OD oscillations at 16 eV fitted to a sinusoid, revealing a 3.1 fs period from the 1.31 eV spin-orbit splitting in Xe^+.

In the future, table-top AATAS will be applied to investigate extremely fast electron dynamics in more complex systems including molecules and solids. AATAS may further be realized in a very compact setup [6], enabling widespread application of this technique.

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Hole alignment in noble gas atoms using RTDCIS method

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Synopsis Using the relativistic time-dependent configuration-interaction singles (RTDCIS) method, we investigate ultrafast hole alignment in noble gas atoms. Hole alignment is defined as the population ratio of states with different magnetic quantum numbers, m_j, in the photoionized atoms. Our study explores how spectral features, such as Fano resonances and Cooper minima, affect hole alignment in neon, argon, and xenon when interacting with ultrashort pulses.

Hole alignment describes the selective population of specific magnetic quantum states (m_j) in ionized atoms [1]. It can play an important role in attosecond physics since the alignment of "holes" affects charge migration in molecules. This study presents a theoretical investigation of hole alignment in noble gas atoms subjected to femtosecond and attosecond pulses. Using the RTDCIS method [2], we compute the population ratios of different m_j states with hole a in the generated ion based on the properties of the photoelectrons emitted from the atomic system [3] as $A_a^{(m_j,m_j')} = P_a(m_j)/P_a(m_j')$. It is known that in neon, the ratio is affected by Fano resonances using femtosecond pulses [1]. However, attosecond pulses cannot efficiently enhance alignment due to the narrow spectral width of Fano profiles.

Cooper minima (C) and giant resonances (G) have broader frequency widths, see Fig 1 (a), which makes them alternative candidates to study hole alignment with shorter pulses. At the Cooper minimum of xenon, $a = 4d_{5/2}$, photoelectrons to f-waves are strongly suppressed, which leads to a high alignment ratio exceeding 200 : 1 for $m_j = 1/2$ and $m_j' = 5/2$ for a 300 as pulse, see Fig.1 (b). We have also studied the effect of pulse duration on hole alignment ratios in argon and xenon. We achieve a remarkable ratio of up to 1500 : 1 for a 2 fs pulse in the $4d$ Cooper minimum of xenon. This insight may be relevant for controlling charge migration in molecular systems and state preparation in quantum experiments.

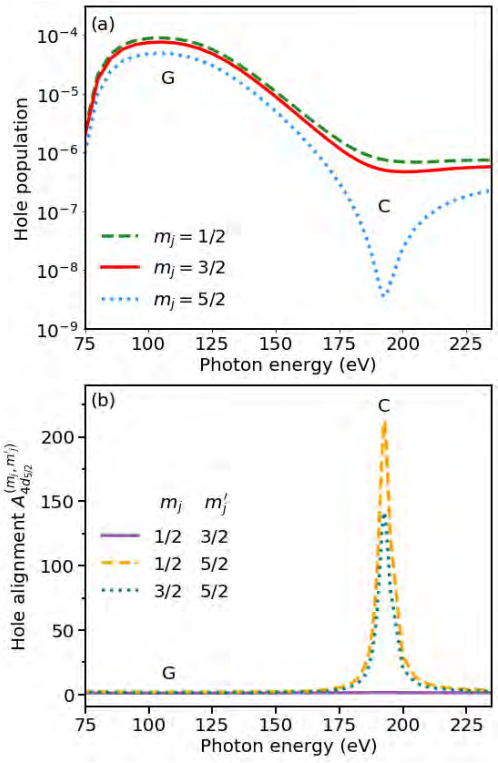


Figure 1. (a) Populations of ion hole $4d$ with $j = 5/2$ for different magnetic quantum numbers in xenon versus photon energy. (b) Hole alignment ratios of the populations in (a).

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Modelling sequential ionisation within time-dependent R-Matrix Theory

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Synopsis We combine R-Matrix Theory with Time Dependence with density matrix theory to model sequential ionisation processes, using the ionisation of Ne^+ to Ne^{3+} through XFEL as proof of concept. Variation in the time delay between the two pulses manifests itself through changes in the angular distribution of the photoelectron from the Ne^{2+} photoionization.

We present a new approach to model sequential ionization processes, employing a series of R-matrix with time-dependence (RMT) calculations to model the behaviour of the residual ion stages, and a density matrix to describe the coherences between the residual ion states. This combination allows us to describe correlated dynamics through multiple ionization stages.

RMT is a technique that can study the dynamics present in laser-driven, multielectron system on ultrafast timescales [1]. RMT partitions the system into two distinct regions. In the 'inner' region (close to the nucleus), the correlation between electrons is described fully. In the 'outer' region, electron exchange is neglected, and an ionized electron moves under the influence of the laser and the long-range potential of the ion.

Density matrix theory has been applied to understand the attosecond-scale dynamics (see [2] for an example studying correlations between photoelectrons in sequential photoionization). The density matrix can summarise the ionic quantum state left after ionization. The population of the residual ion states is contained in the diagonal elements, while the off-diagonal elements contain the correlation coefficients between these states.

As a demonstration, we model pump-probe ionisation of Ne^+ to Ne^{3+} using an XFEL with photon energies of 95 eV [3]. We start with Ne^+ in $^2P^o$. Separate calculations are run for Ne^+ m=0 and Ne^+ m=1. An initial XUV photon ionizes Ne^+ to Ne^{2+}. After a time delay (up to 1.2 fs), a second XUV pulse photoionises Ne^{2+} to Ne^{3+}. The main observable of interest is the momentum-resolved angular distribution of the photoelectrons. Separate angular distributions are obtained for each residual ion state, including orbital and spin magnetic quantum numbers.

Figure 1 shows how the angular distribution of the outgoing electron is affected by changes in delay time. For a delay time of 0.39 fs, the distribution is more aligned along the polarization axis compared to a delay time of 0.94 fs.

This change in the angular distribution is caused by the coherent superposition of the Ne^{2+} ion after ionisation by the pump pulse. This superposition is seen for the m=0 levels of the ^1S and ^1D states, as these two states are built from the same independent electron basis states.

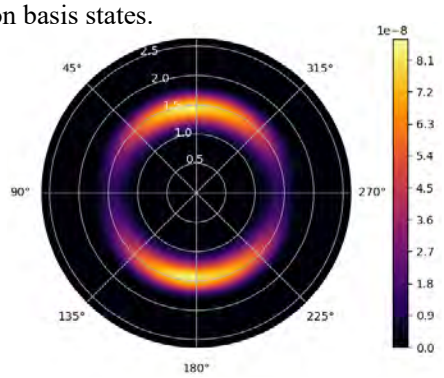


Figure 1. The angular distribution of the photoelectron for photoionization of Ne^{2+} for two different delay times. Both calculations used two 95 eV, 8-cycle pulses. The left half shows the angular distribution following a delay time of 0.39 fs, while the right half of the graph shows the angular distribution following a delay time of 0.94 fs.

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SoftMeter: Ultrafast molecular imaging driven by a tunable two-color field

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Real-time investigation of ultrafast electron dynamics requires high-spatiotemporal-resolution laser spectroscopy. We propose a two-color scheme that revises the prototypical strong-field photointeraction by splitting the three-step dynamics. This approach is promising to enhance the capabilities of laser-induced electron diffraction and high-order harmonic generation at moderate intensities, especially for applications in systems such as large molecules or interfaces.

Time-resolved laser spectroscopy has proven invaluable in observing and manipulating ultrafast photo-induced electron dynamics. However, real-time imaging demands a few-femtosecond to attosecond temporal resolution and picometric spatial accuracy, posing significant challenges [1,2].

We propose a two-color method that enhances the prototypical strong-field interaction [3]. In this approach, single-to-few-photon ionization is triggered by a weak, tunable DUV/VUV pulse with a few-fs pulse duration produced through resonant dispersive wave [4], while the subsequent acceleration and recollision of the photoelectron are driven by an IR field at moderate or weak intensity depending on the wavelength. In this scheme, electron self-diffraction and high-order harmonics are produced (see Fig. 1a).

Simulations based on time-dependent density functional theory were performed to support the main concept of our two-color spectroscopy approach. In particular, we computationally demonstrate efficient driving of photoelectron rescattering at laser intensities on the order of 10^{12} W/cm^2, two orders of magnitude lower than typically used in conventional strong-field studies (see Fig. 1b). This aspect, in conjunction with the tunability of the two-color scheme, poses promising perspectives to develop a novel platform for electron self-diffraction and high-harmonic generation. The experimental setup is currently under development.

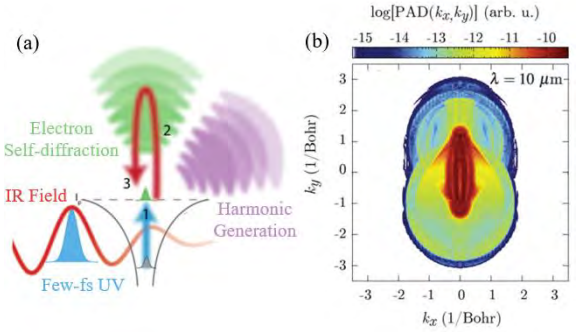


Figure 1. (a) Schematic of the soft-field method. Ionization is triggered by a weak few-fs DUV pulse, followed by photoelectron acceleration and recollision via an oscillating IR field. Resulting in soft electron self-diffraction (green wave) and soft-field harmonic generation (purple wave). (b) Angular photoelectron momentum distribution for an IR field at a wavelength of 10 μm.

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Core ionization of positronium chloride: a numerical study

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Synopsis The ability of some chemical elements to bind a positron (the antimatter counterpart of an electron) or a positronium atom (the bound state of a positron with an electron) is a long-standing prediction which is very challenging to confirm experimentally. We report on calculations of positronium chloride (PsCl) core-ionization. Our results suggest new ways of proving the existence of PsCl and a path to study attosecond dynamics in electronic clouds perturbed by a light positive particle. We propose an experimental setup to observe the products of the core ionization of PsCl.

The attosecond timescale of electron (e^-) dynamics in atoms and molecules can be related to their binding energy, determined by their mass. With identical mass but opposite electrical charge, positrons (e^+) should also undergo attosecond processes for the same reasons. For example, a positron takes approximately 150 attoseconds to orbit the antiproton in an antihydrogen atom in the Bohr model, just as the e^- around the proton in hydrogen. More intriguing is the case of e^+ dynamics in positron binding ordinary matter systems. The existence of these systems has long been predicted [1] and is still actively studied [2]. So far, little experimental evidence revealing their existence and properties has been collected [3, 4]. These systems are difficult to study because of their nanosecond annihilation lifetime and low production rates.

Here, we study the possibility to use core ionization as a diagnostics for the existence of positronium (Ps) binding atoms and as a potential gate way to e^+ attosecond dynamics in matter-antimatter exotic systems. We focus on the case of positronium chloride (PsCl) which production time can be controlled with nanosecond precision by inducing collisions between pulsed Ps clouds and chloride anions [5]. We compute the cross-section of the different reactions happening in $PsCl^+$ when a core e^- is removed from the 2s or 2p shell, including the dissociation into Cl and e^+ and the emission of e^-, e^+ or Ps via Auger-Meitner relaxation processes. We find that the energy of the Auger-Meitner e^- is shifted compared with calculations in Cl^-, reflecting the sensitivity of Auger-Meitner processes to the chemical environment of the core-ionized atom. We also find that the cross-section of the emission of e^+ via an Auger-Meitner relaxation process is sufficiently large to be observed experimentally. This makes it an ideal candidate for background-free direct observation of PsCl.

Finally, we propose that this experiment could be realized with a setup similar to the one used in [6] where short X-ray pulses with photon energy up to 400 eV were used to induce Auger-Meitner processes in Argon. However, in the case of PsCl, time-of-flight spectrometer would collect positrons instead of electrons. The annihilation lifetime of the studied system should not prohibit the feasibility of this experiment since it is orders of magnitude larger than the pulse duration. Last, a two color setup should eventually allow to observe attosecond dynamics in PsCl [7].

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Attosecond transient absorption spectroscopy of laser-dressed dense helium: Spectral lineshape modification and weak transitions brought to light

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Synopsis We show that by combining strong-field dressing and resonant propagation of extreme-ultraviolet (XUV) pulses, the transition of absorption lines from a domain of primarily asymmetric and rapidly changing with propagation, to a domain of Lorentzian-like near-symmetric profiles, *in the limit of optically thick samples*. In addition, by exploiting a strong laser-coupled pathway, we report the first time-resolved observation of the weakly XUV-populated *2p3d* and $sp_{2,4}$- doubly excited states in helium.

Attosecond transient absorption spectroscopy (ATAS) has been established as a powerful tool for tracking ultrafast electronic dynamics in gaseous media. Here, we integrate microscopic ultrafast laser control of atoms and macroscopic pulse reshaping up to the high-density limit. The extreme case of a large optical thickness is considered, leading to the transition of absorption lines from a domain of laser-controlled spectral shapes to a domain of Lorentzian-like near-symmetric profiles. This unexpected spectral evolution results from the significant temporal reshaping of the driving pulse, which gets temporally prolonged and delayed, masking the dressing effects imposed by the external laser field and effectively shifting the ultrafast nonlinear interaction to later times. We first demonstrate this concept in numerical calculations, both using a model system and through a fully coupled solution of the time-dependent Schrödinger equation and the Maxwell wave equation. For an experimental demonstration, we conduct ATAS measurements in helium with a widely varying gas pressure, showing the evolution of the *1snp* spectral profiles as the pulse propagates [1].

In addition, we employ ATAS to verify a general method to enhance the spectroscopic signal of weak transitions, whose small cross sections scaling with the square of the transition matrix elements generally challenge experimental detection. The presented method breaks this scaling law by exploiting a stronger laser-coupled pathway. Experimentally, we report the first time-resolved observation of the weakly XUV-coupled *2p3d* and $sp_{2,4}$- doubly excited states in helium (see Fig. 1). As confirmed by large-scale numerical calculations, we observe an order of magnitude enhancement of the cross sections of these two resonances [2]. Enhancing single-photon-suppressed transitions can find widespread applicability from spectral diagnostics of complex molecules to precision spectroscopy of weak transitions.

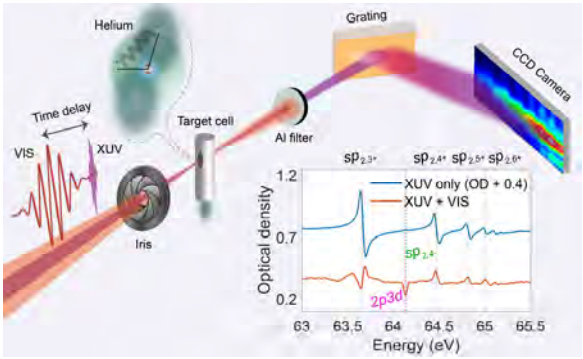


Figure 1. Illustration of the experimental ATAS setup. Inset: Absorption profile at 1.5-fs time delay (orange line) along with the result in the absence of the visible (VIS) pulse (blue line), showing the enhanced spectral visibility of *2p3d* and $sp_{2,4}$- states at around 64.1 eV.

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Observation of Bloch Oscillation phenomena via High Harmonic Generation Spectroscopy

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We study Bloch oscillations by solid-state HHG spectroscopy by attempting to observe the negative ionization branch where electrons recombine via band periodicity at high fields. Using semi-classical Bloch equations and Gabor analysis, we show its dominance at high intensities. Two-color perturbation techniques enable real-time observation, providing direct insight into Bloch oscillations.

In solid-state systems, the interplay between an intense laser field and a periodic lattice potential can drive Bloch oscillations: electrons accelerated to the Brillouin zone (BZ) edge are reflected by the band structure's inherent periodicity [1]. Although Bloch oscillations have been measured in superlattices [2], no direct observation in bulk solids has yet been achieved, largely due to ultrafast scattering processes that obscure the dynamics. From the perspective of high-order harmonic generation (HHG), a negative ionization time branch emerges, wherein electrons ionized before the field peak can still recombine via the band structure's periodicity at sufficiently high laser intensities[3-4]. We present a new manifestation of these sub-cycle dynamics and aim to measure this negative ionization branch directly.

By applying semi-classical Bloch equations and Gabor transform analysis, we isolate the temporal evolution of these trajectories. In [Fig 1] the blue region highlights a gate over the negative ionization times, showing that this branch dominates the HHG signal at higher field amplitudes. To observe and analyze this branch, we applied two-color perturbation techniques[5] acting as a subcycle gate, probing the signal in real time. This approach allows us to resolve the distinct temporal characteristics of the negative ionization branch, thus offering a direct window into Bloch oscillation dynamics in solid-state HHG.

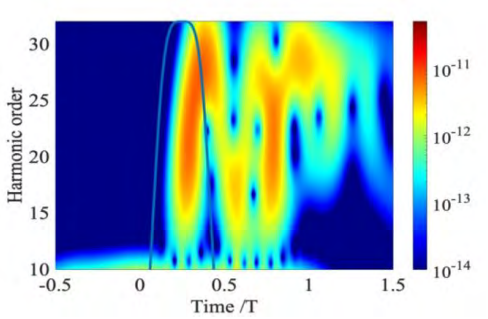


Figure 1. Gabor transform analysis of the semi-classical Bloch equations for field amplitude 0.01 (atomic units). Represents the intensity of the emitted photonic energy vs the time of the optical cycle, presenting the contribution of each branch to the signal. (This calculation has been done by Zhaopin Chen, from Michael Kruger's group at the Technion).

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Electron recollisional excitation on dissociative ionization of OCS in phase-locked two-color intense laser fields

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Synopsis We investigate dissociative ionization of OCS in phase-locked two-color intense laser fields. The electron momentum distribution shows asymmetry along the laser polarization direction, reflecting the asymmetric shape of the laser electric fields. In OCS^+ and S^+ channels, the electron asymmetry flips at electron kinetic energies of 8.2 eV and 4.2 eV, respectively, reflecting the excitation energy from the X state to the A state of OCS^+. Classical trajectory Monte Carlo simulations clarify that electron recollisional excitation occurs to form fragment ions in intense laser fields.

Tunneling ionization and subsequent electron recollision processes in intense laser fields [1] induce ultrafast molecular and electronic dynamics such as high-order harmonic generation, laser induced electron diffraction, and non-sequential double ionization. Since the tunneling electron returns to and collides with the parent ion, the electron recollision is a powerful tool to investigate electron scattering by ions. In this study, dissociative ionization of OCS, $OCS \rightarrow OCS^+ + e^- \rightarrow OC + S^+ + e^-$, in phase-locked two-color intense laser fields has been investigated by using a photoelectron-photoion coincidence momentum imaging technique to clarify the contribution of electron recollisional excitation on molecular dissociation in intense laser fields [2].

The output of a Ti:Sapphire chirped pulse amplifier (800 nm) is introduced into a β-barium borate crystal to generate the second harmonic pulses (400 nm). The fundamental and second harmonic pulses are separated to tune the time-delay between the pulses. Two laser pulses are co-linearly combined again to obtain the phase-locked two-color laser fields, $F(t) = F_\omega \cos(\omega t) + F_{2\omega} \cos(2\omega t + \Delta\phi)$, where F_ω and $F_{2\omega}$ are electric field amplitudes of the fundamental and second harmonic fields, and $\Delta\phi$ is the phase difference between the pulses. The three-dimensional momentum vectors of the generated ions and electrons $\mathbf{p} = (p_x, p_y, p_z)$ are obtained by measuring the position on the detector (x, y) and the arrival time t in a single-shot acquisition mode. A classical trajectory Monte Carlo (CTMC) simulation [3] with a potential calculated using density functional theory has been performed to clarify excitation mechanisms.

To evaluate the electron ejection direction quantitatively, asymmetry parameter is defined as $A = (Y_+ - Y_-)/(Y_+ + Y_-)$, where Y_+ and Y_- are the yields of electrons with positive and negative momenta along the laser polarization direction with the acceptance angle of 30°, respectively. The asymmetry parameter depends on $\Delta\phi$, and flips at the electron kinetic energy E_{flip} of 8.2 eV and 4.2 eV in the OCS^+ and S^+ channels, respectively, as shown in Fig. 1 as solid lines. The CTMC simulations clarify that the electrons with lower and higher energy than E_{flip} are assigned to the forward- and backward-scattered electrons and that the difference in E_{flip} corresponds to the excitation energy from the X state to the A state of OCS^+. This indicates that the electron recollisional excitation takes place to form the electronically excited ions leading to molecular dissociation in intense laser fields.

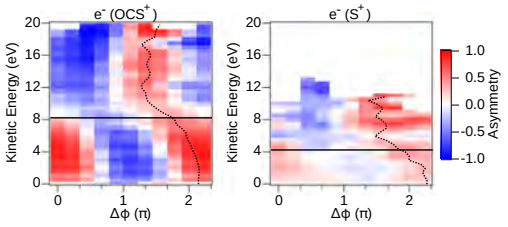


Figure 1. Two-dimensional plots of asymmetry parameter A of the OCS^+ and S^+ channels. Dotted lines indicate $\Delta\phi$ at which the asymmetry has a maximum value at each electron kinetic energy. Solid horizontal lines indicate E_{flip}.

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Ro-vibronic coupling in vibrational resolved attosecond photoelectron spectroscopy of acetylene

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Synopsis Photoioization delay of four cation states, $\widetilde{X}^2\Pi_u$, $\widetilde{A}^2\Sigma_g^+$, $\widetilde{B}^2\Sigma_u^+$, $\widetilde{C}^2\Sigma_g^+$, from C_2H_2 molecules is measured with high energy resolution RABBIT spectroscopy. The vibrational-dependent ionization delay time from give a hint on the ro-vibronic coupling of the $\widetilde{X}^2\Pi_u$ state after photoionization by absorbing an XUV photon.

Attosecond pulses provide unprecedented time resolution for real-time observation and manipulation of electron dynamics. However, such ultrashort pulses often have a broad spectral distribution, resulting in limited energy resolution when detecting molecular ionization dynamics, and it is difficult to accurately analyze the contributions of different electronic states or vibrational energy levels. This contradiction can be solved by using attosecond pulse train generated by narrow-band high-order harmonics[1]. The formed attosecond pulse train not only maintains the time resolution, its narrow-band characteristics also provide excellent energy resolution. This light source with unique time-frequency characteristics provides a powerful tool for the study of electron correlation[2], and nuclear-electron coupling dynamics[3] in the molecular ionization.

Under extreme ultraviolet light, the photoionization of acetylene not only triggers the independent contribution of the outermost valence electron orbital ($1\pi_u$) and a series of well-spaced photoelectron energy bands generated by deeper molecular orbitals ($3\sigma_g$, $2\sigma_u$, $2\sigma_g$). In the RABBIT spectrum, we observed photoelectron from four electronic states($\widetilde{X}^2\Pi_u$, $\widetilde{A}^2\Sigma_g^+$, $\widetilde{B}^2\Sigma_u^+$, $\widetilde{C}^2\Sigma_g^+$). Moreover, the vibrational levels of the $\widetilde{X}^2\Pi_u$ state can also be resolved clearly. By measuring the attosecond photoionization delay between vibrational levels from $\widetilde{X}^2\Pi_u$ state, such as the relative ionization delay time of $\tau_{v_1-v_0}$ and $\tau_{v_2-v_0}$ at SB20, which are 44 as and 110 as respectively, we might captures the ro-vibronic coupling of the linear C_2H_2 molecule after photoionization by absorbing an XUV photon. The results offer new insights by resolving the coupled electron and structural dynamics simultaneously.

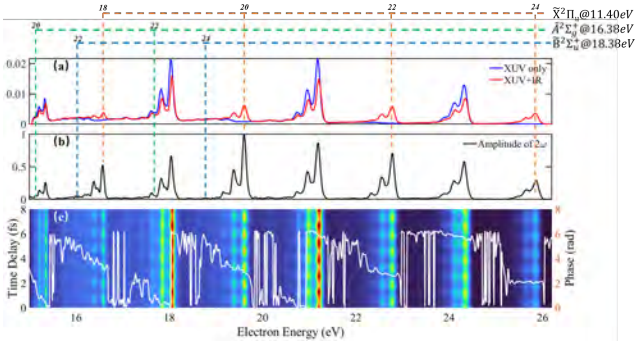


Figure 1. Channel-resolved C_2H_2 RABBIT spectra. (a) Identification the SB channels of three cation states($\widetilde{X}^2\Pi_u$, $\widetilde{A}^2\Sigma_g^+$, $\widetilde{B}^2\Sigma_u^+$) with the XUV-only and XUV+IR spectrum. (b) the amplitude spectrum. (c) Full XUV-IR spectrum as a function of the delay with the phase spectrum.

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Attosecond dynamics of direct electrons in optical near-fields

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Synopsis We show measurements of carrier-envelope phase (CEP) resolved strong-field energy spectra of electrons driven with two-cycle light pulses, photoemitted from a nanometer-sharp metal needle tip. The strongly inhomogeneous optical near-field around the tip apex leads to a new CEP-dependent spectral feature in the *direct* electrons.

High harmonic generation upon electron recollision as well as backscattering of photoemitted electrons are hallmarks of strong-field physics and rely on the non-perturbative driving of electrons using strong and ultrashort light fields. Combined with the control of the carrier-envelope phase (CEP) of few-cycle light pulses, these processes have served as door openers for probing the attosecond dynamics of electrons emitted from atoms, molecules, clusters and needle tips [1].

Similar dynamics as known from atoms are also found for electrons emitted from ultrasharp metal needle tips. A key difference compared to the photoemission from atoms is the presence of the localized optical near field at the tip apex. This near field decays on the scale of the tip radius (typically a few to a few tens or hundreds of nanometers) and can substantially influence the cut-off energy of the plateau electrons [2].

Here, we drive electrons using a two-cycle light pulse (central wavelength 1550 nm) in the optical near-field of a metal needle tip with a radius of curvature of 10 nm. Fig. 1 shows a map of energy spectra for an optical near-field intensity of $2.0 \times 10^{13}\mathrm{W/cm^2}$ as a function of the CEP and the CEP-averaged spectrum (brown curve). The spectra comprise the typical features with direct electrons (below 10 eV) and the plateau of rescattered electrons. Besides the well-known CEP-dependence of the high-energy electrons at the cut-off of the plateau [3], we find a strong modulation of the direct electrons below 5 eV, which manifests as stripe-like features. We explain this modulation by a sub-cycle-dependent drift motion of electrons within the optical near field. This feature allows us to track the emission time of direct electrons on the attosecond time scale.

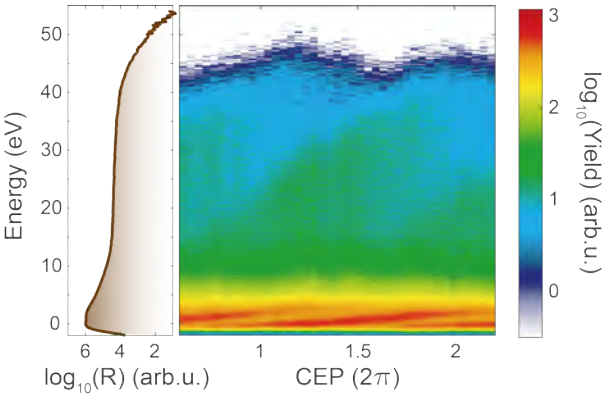


Figure 1. Measured electron energy spectra as a function of the CEP on a logarithmic scale (right) and the CEP-averaged spectrum (left). The low-energy electrons show a new CEP-dependent energy feature, visible as stripes in the red and yellow region.

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Influence of catastrophes and hidden dynamical symmetries on ultrafast photoelectron physics

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Synopsis The catastrophes which appear in a variety of ultrafast photoelectron scenarios, ranging from hard and soft rescattering [1] to strong field tunnel ionization, are investigated. On the way, hidden dynamical symmetries of the combined atomic and laser field system are illuminated and semiclassical trajectories are disentangled for a more general understanding of photoelectron dynamics.

Catastrophic phenomena are present across physics and all of the sciences and can be used to explain systems where small incremental changes can be followed by sudden rapid changes in behavior. In general, the study of any system which responds in a non-linear fashion to changes in a parameter space can benefit from the catastrophe theory perspective.

In strong field physics, it has long been understood that the high-energy above threshold ionization cutoff is due to a fold-type catastrophe, which necessitates the use of a uniform approximation [2] to accurately calculate the photoelectron momentum distribution (PMD) using orbit-based methods. The comparable strengths of the Coulomb force due to an atomic potential and the forces on electrons due to a strong laser field breeds highly non-linear dynamics which are very complicated and can be more simply understood through a catastrophe lens. Due to their combined importance, we will use a theory which puts the Coulomb potential and laser field on an equal footing, the recently implemented hybrid forward-boundary CQSFA (H-CQSFA) [3].

We discuss the effect on PMDs, of using potentials which deviate from the Coulomb potential, by introducing different degrees of softening. This influences the ridges observed in the PMDs associated with backscattered electron trajectories. In the limit of a hard-core Coulomb interaction, the re-scattering ridges close along the polarization axis, while for a soft-core potential, they are interrupted at ridge-specific angles. We analyze the momentum mapping of the different orbits leading to the ridges, which are shown in Figure 1. For the hard-core potential, there exist two types of saddle-point solutions that coalesce at the ridge. By increasing the softening, we show that two additional solutions emerge as the result of breaking a hidden dynamical symmetry associated exclusively with the Coulomb potential. Further signatures of this symmetry breaking are encountered in subsets of momentum-space trajectories. This implies that, in the presence of residual binding potentials in the electron's continuum propagation, the distinction between purely kinematic and dynamic caustics becomes blurred.

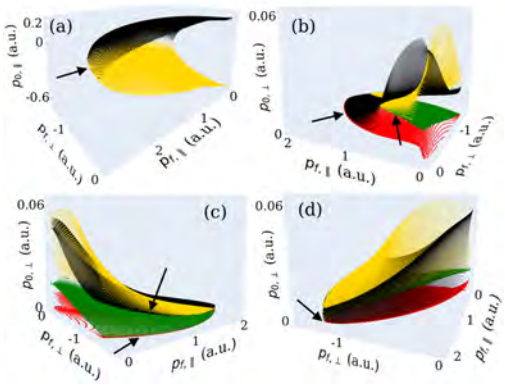


Figure 1. Initial to final momentum mapping for a hard (a) and soft (b), (c) and (d) atomic potential. The different color points represent trajectories with different qualitative features.

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Interference of direct and resonant two-photon ionization in helium

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Synopsis We study two-photon resonant ionization of the 1s3p Rydberg state in helium using high spectral resolution photoelectron interferometry. By measuring the interferogram over a pump-probe delay range of up to 80 fs we observe a delay dependent splitting of the resonant sideband. Using model calculations, we show that this splitting can be attributed to the destructive interference between the direct two-photon ionization and the sequential resonant ionization from the 1s3p state.

Two-photon resonant ionization of Rydberg states in helium has been studied extensively using pump-probe photoelectron interferometry. Previous measurements of the 1s3p state have shown a double peak structure [1, 2], however no clear interpretation has been given. We perform RABBIT (Reconstruction of Attosecond Beatings By Interference of Two-photon transitions) measurements over a long pump-probe delay range (up to 80 fs), with high spectral resolution. The long delay scan allows us to observe the effect of the build-up of the 1s3p state population.

Figure 1(a) shows the measured RABBIT interferogram around the resonant sideband 16. At negative delays, corresponding to the peak of the probe pulse arriving first, a clear splitting is observed. When the delay is increased, corresponding to the pump pulse arriving first, only one peak is seen.

We attribute this double peak structure to the interference between the direct (non-resonant) two-photon ionization and the sequential resonant ionization from the 1s3p state. For positive delays, the resonant path becomes dominant due to the large population in the 1s3p state, and the double peak structure is suppressed. Figure 1(b) shows an interferogram obtained from model calculations, including the resonant and direct two-photon absorption paths [3].

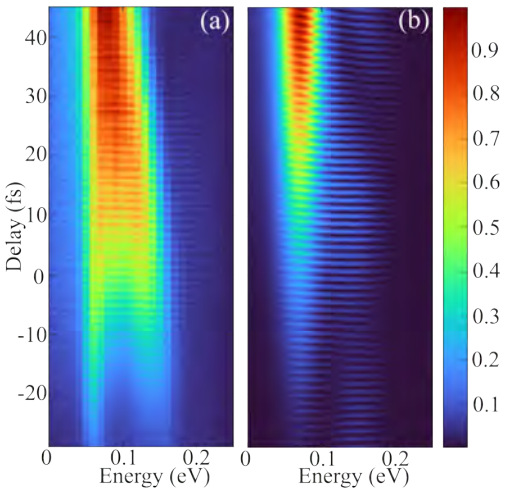


Figure 1. Measured (a) and modelled (b) RABBIT interferogram of the resonant sideband 16.

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Estimation of ZnO thickness from high harmonic spectrum using machine learning algorithms

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Synopsis The physical and chemical properties of 2D material may be sensitive for change in their thickness. When applying them in industry, it is important to produce nearly identical sample and to monitor their thickness without causing any damage in the samples. Here we propose and demonstrate a non-destructive and economic method to estimate the thickness of thin ZnO layers relying on high harmonic spectra generated with mid-infrared laser and exploiting the capability of machine learning algorithms to identify complex patterns.

Two-dimensional materials have an atomic-scale thickness, with intra-layer atoms bonded covalently, while inter-layer interactions are governed by weak van der Waals forces. Accurate, rapid, and non-destructive techniques for thickness characterization are crucial for understanding how material properties depend on thickness. Methods such as atomic force microscopy, optical imaging, Raman spectroscopy and angle-resolved photoemission spectroscopy have been employed.

High harmonic generation (HHG) in bulk crystals has proven to be a powerful tool for extracting structural information. Recently, research has shifted toward thin-target materials, leading to the development of models explaining the dependence of harmonic yield on material thickness. In [1, 2] it was shown that both current density effects created by the laser and propagation effects are affecting the HHG yield in very thin films. By the increase of the thickness propagation effect becomes predominant.

Here we demonstrate that HHG spectra from ZnO thin layers given to a pre -trained machine learning (ML) algorithm could be an alternative way to estimate and monitor the layer thickness. Spectroscopic ellipsometry was used to obtain the "true" layer thicknesses and along with corresponding HHG spectra driven with a MIR laser, was given as an input to the ML algorithm. After training the model, new harmonic spectra was fed into it and the efficiency of its performance is plotted in Figure 1 as a confusion matrix. The high number of cases lying on the diagonal shows the precision of the method. The overall correct classification is 78%. We have to remark, that the model was mistaken in cases where the thicknesses were close to each other.

This method could serve as an important, economic and non-destructive tool in thin layer thickness monitoring for industrial purposes.

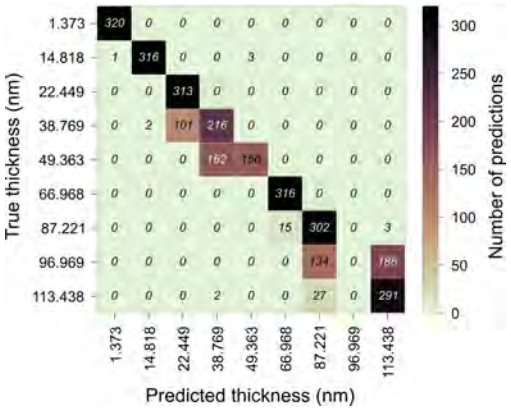


Figure 1. Confusion matrix presenting the efficiency of the ML algorithm in classifying the sample thicknesses.

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Anisotropy Induced Electronic Coherence Signal in Attosecond Transient Absorption Spectroscopy

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Synopsis Attosecond transient absorption spectroscopy (ATAS) is a key method for probing electronic coherence in molecules. However, the role of anisotropy created in molecules by the intense pump pulse remains unclear. Using a recently developed density matrix approach, we demonstrate that coherent ATAS signal in N_2^+ ions critically depends on the anisotropy from strong-field ionization. We suggest combining our approach with quantum chemistry methods to model the evolution of electronic coherence in molecules and call for future ATAS experiments to test our predictions.

Probing electronic coherence in molecules is essential for controlling chemical reactivity. Although ATAS has been a prominent method for probing electronic coherence, the strong-field pump process of the molecule is not well understood due to the computational challenges in first-principle approaches. The current theory of ATAS for molecules [1] employs the sudden ionization approximation, where the population and coherence of the molecular ion are isotropic. Therefore, the role of anisotropy created by the strong-field pump pulse on the ATAS signals is unclear.

Using a density matrix approach recently developed by our group [2, 3], we simulated the pumping process of N_2 by an intense few-cycle mid-infrared laser pulse. Accounting for the anisotropy in the ATAS theory [4], we demonstrated that the emergence of coherent ATAS signals in nitrogen molecular ions critically depends on the anisotropy induced by strong-field ionization, while the incoherent ATAS signals are also moderately influenced by the anisotropy (see Fig. 1).

We emphasize that the density matrix approach can be integrated with more accurate quantum chemistry methods to describe the creation, evolution, and detection of electronic coherence in molecules. It also motivates further development in first-principles approaches to obtain orientation-dependent density matrices for the electronic states of molecular ions created by strong laser pulses. Lastly, we call for future ATAS experiments to validate our findings.

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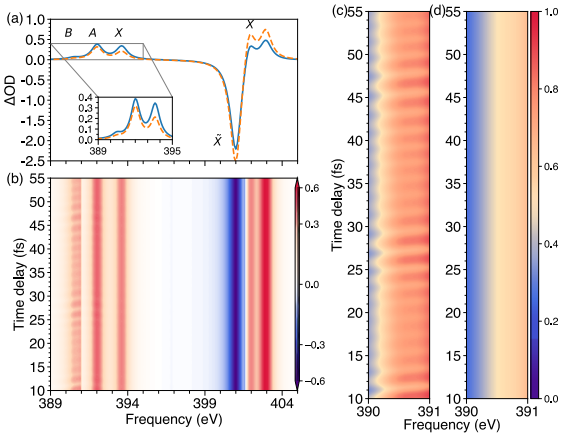


Figure 1. (a) The pump-probe optical density (ΔOD) at 10 fs time delay for the $\tilde{X}$ state of N_2 and the X, A, and B states of N_2^+. The spectra are calculated using the anisotropic (blue solid line) and isotropic (orange dashed line) density matrix, respectively. (b) ΔOD calculated using the anisotropic density matrix at different time delays. (c-d) Comparison of ΔOD between 390 and 391 eV calculated using the anisotropic (c) or isotropic (d) density matrix.

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Probing the attosecond dynamics of high-energy-radiation-induced collisional ionization cascades in condensed-phase samples

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Synopsis We study inelastic electron scattering in liquid water using a new ab initio approach developed to accurately describe both low and high transferred energy regimes. This allows us to fully simulate the dynamics of the collisional cascade in liquid water, and to provide a complete molecular picture of the collision-induced damages. Observing such processes recently became possible thanks to the emergence of new x-ray pump/probe spectroscopies with attosecond/sub-femtosecond time resolution.

Absorption of high-energy radiation leads to the ejection of fast electrons. In condensed phases, their kinetic energy is deposited into the medium through collision-induced excitation and ionization. Thus, a single electron triggers the formation of several excited and ionized molecules and the release of further electrons, leading to additional radiation damage.

In liquid water, the collisional cascade occurs within a few femtoseconds. Emerging electron spectroscopy with attosecond (as) resolution provides the opportunity to give novel insights into these fundamental processes. Recently, the first x-ray pump/x-ray probe experiment with as resolution was achieved on water at the LCLS XFEL, displaying clear signatures of the collisional dynamics [1].

We developed an ab initio procedure to address electron inelastic scattering in bulk water. By using a plane-wave description of the projectile and a pseudo-continuum approach, our method yields accurate cross sections for both excitation and ionization regimes, thus going beyond the state-of-the-art. Kaufmann basis functions [2] were used to facilitate the pseudo-continuum description and the use of the Stieltjes imaging procedure.

By employing our theoretical cross sections as a basis for a Monte Carlo simulation, we extract quantitative information about the inelastic electron scattering cascade dynamics, and we provide a complete molecular picture of the state of the damaged surrounding medium.

Importance of an inelastic electron scattering cascade in liquid water is shown in Figure 1. An electron projectile initially carrying 500 eV of kinetic energy leads to the release of more than 15 secondary electrons through collisional ionization. Additionally, more than 20 water molecules are expected to be excited or ionized as a result of the collisional cascade. We show that total dissipation of the projectile energy is achieved in about 2 fs, while signatures of collision-induced damage are expected to appear as early as 150 as after release of the initial projectile. Knowledge of these properties is key to model time-resolved x-ray absorption spectroscopies of condensed phase systems subjected to high-energy ionizing radiations.

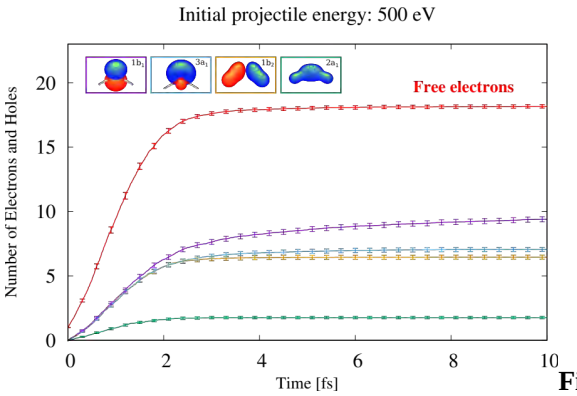


Figure 1. Evolution of the number of free electrons and molecular hole states in the liquid water medium during the inelastic scattering cascade of an electron projectile with an initial kinetic energy of 500 eV.

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Modeling angularly resolved phase-jumps in heavy atoms

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Synopsis RABITT has for a long time been a central tool for the full reconstruction of attosecond electron dynamics. The present communication employs RRPAE to accurately model RABBIT near autoionizing resonances occurring between the first ($np_{3/2}$) and second ($np_{1/2}$) ionization thresholds in radon and xenon. Further, simple analytical models have been employed to predict and explain the behavior of the phase.

The ubiquitous technique for studying photoionization on an attosecond timescale is the Reconstruction of Attosecond Beatings by Interference of Two-Photon Transitions (RABBIT) [1]. RABBIT is an interferometric approach that employs an attosecond XUV pulse train to ionize an atom, while simultaneously applying a weak, IR probe field. Varying the pump-probe delay, allows for probing not only the amplitude, but also the phase of the released wave packet, which is equivalent to the photoionization time delay.

The atomic phase η_A measured in RABBIT can be divided into two distinct contributions [2]: The first, η_W, corresponds to the Eisenbud-Wigner-Smith-like delay associated with the one-photon ionization process. The second term, η_{cc}, accounts for the continuum–continuum contribution. Due to interference between ionisation channels of different symmetries, the profile of the phase varies with emission angle. Although also true in featureless regions, it is even more relevant near resonances, where the shape of the quickly varying phase can change dramatically with emission angle.

The present study uses Relativistic Random Phase Approximation with Exchange (RRPAE) [3] to calculate the angularly resolved phase in the vicinity of autoionizing resonances between the first ($np_{3/2}$) and the second ($np_{1/2}$) ionization thresholds in radon and xenon. Individual components of the phase are disentangled and their dependence on the emission angle θ relative to the direction of the linearly polarized XUV light is investigated. Further, analytical models are employed to explain the shape and the energy position of the resonant phase jumps.

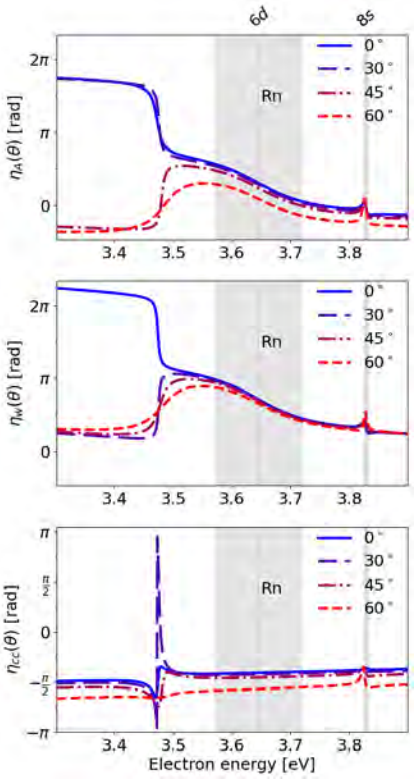


Figure 1. Angular dependence of the atomic phase and its components for a photoelectron ionized from the $6p_{3/2}$ orbital in radon. The top panel shows the atomic phase, the middle panel shows the Wigner phase, and the bottom panel shows the continuum-continuum phase. The positions and widths of the $6p_{1/2}^{-1}6d$ and $6p_{1/2}^{-1}8s$ resonances are shown in gray.

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Neural Network Assisted Denoising in Attosecond Transient Spectroscopy Beyond Correlated Source Noise

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Synopsis We demonstrate an approach to denoise attosecond transient absorption measurements based on a predictive neural network. Strong correlations between the near-infrared and the corresponding high-harmonic radiation allow for a more than threefold noise reduction. This method is widely applicable and capable of reducing noise beyond correlated source noise.

Attosecond transient spectroscopy (ATS) enables uncovering ultrafast dynamics in gases, liquids, and solids. This work demonstrates an easily applicable, neural-network based approach that achieves a more than threefold noise reduction, without solely relying on suppressing correlated noise nor requiring a modified setup [1].

Commonly in ATS a few-femtosecond, NIR pulse is split into two: a pump beam that induces dynamics in a sample, and a beam that drives high-harmonic generation (HHG) for the extreme ultraviolet (XUV) probe. Typically a referencing scheme is employed which alternates between acquiring the XUV transmission or reflection of the excited sample and the equilibrated sample. A significant contributing factor to noise in ATS are the spectral intensity fluctuations of the XUV probe. Our approach reduces the impact of these fluctuations. Recording the NIR and corresponding XUV spectra reveals complex spectral correlations between the generating and generated radiation (**Fig. 1**, left). A second-harmonic (SH) spectrum is additionally generated from the NIR pulse as a sensitive indicator for varying pulse durations. These highly correlated spectral intensities enable the prediction of a HHG spectrum solely from the corresponding NIR+SH spectra. This is achieved by training a predictive convolutional neural network (CNN). The CNNs predictions are then used to simultaneously compare excited (signal) and equilibrated (reference) spectra, with the latter being obtained from the NIR+SH spectra which can be recorded at the same time as the signal spectrum. This simultaneous referencing reduces source noise contributions and halves the measurement time.

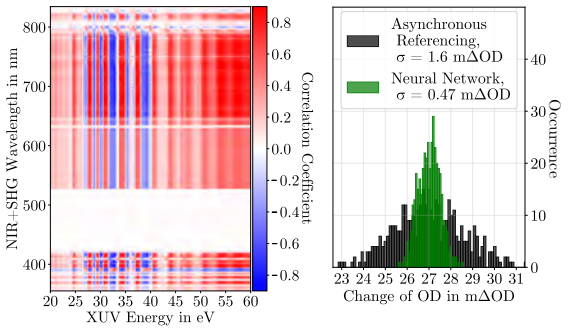


Figure 1. **Left**: Pixel-wise correlation coefficients between a NIR pulse, its SH and the HHG spectrum. **Right**: Measurement histogram of a single energy and delay pixel for conventional referencing (black) and neural network approach (green) with identical measurement parameters.

The improvement offered by the aforementioned scheme, compared to conventional referencing, is demonstrated by looking at the statistics for a singular (select photon energy and delay time) pixel from a transmission ATS measurement. This pixel results from averaging 400 ΔOD values and the underlying statistics are compared for both approaches (**Fig. 1**, right). The neural network approach achieves a more than threefold improvement in its standard deviation.

This easily applicable technique can be used to reduce noise in NIR pump - XUV probe referencing schemes, beyond merely suppressing correlated source noise, and allows for improved sensitivity and thereby to discern more subtle dynamics.

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Multi-photon and strong-field ionization with a twist

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Synopsis We investigate the combined influence of the spin and orbital angular momentum of light in the photoionization of chiral and non-chiral targets. We observe a helical dichroism from $\sim 1\%$ to $\sim 10\%$ depending on the ionization regime, reflecting different influence of Rydberg states and continuum-continuum transitions.

While the influence of the polarization state, i.e. the Spin Angular Momentum (SAM) of light, on the angular distribution of photoelectrons has been experimentally studied for decades, the role of the Orbital Angular Momentum (OAM) has only recently been considered. De Ninno *et al.* showed that when helium atoms are ionized by a circularly polarized XUV pulse in the presence of a circularly polarized IR field carrying OAM, the angular distribution of the photoelectrons resulting from two-photon two-color absorption is sensitive to the sign of OAM in the IR beam [1]. The resulting Helical Dichroism (HD) effect is large, in the 10% range. This was explained by two elements: (i) the favorable geometry of the experiment, in which the XUV beam was overlapped with the central part of the OAM-carrying IR beam, in the vicinity of the phase singularity; (ii) the delocalized nature of the intermediate continuum state, populated by absorption of the XUV photon, which makes the continuum-continuum transition more sensitive to non-dipole effects.

We performed experiments to investigate the influence of OAM in photoionization in different regimes. First, we used 515 nm pulses to excite Rydberg states of ethanol by absorption of 4 circularly polarized photons (SAM $\sigma_{515} = \pm 1$), and 1030 nm pulses carrying OAM ($m_{1030} = \pm 1$) to ionize the excited molecules by absorption of 1 to 5 circularly polarized photons (SAM $\sigma_{1030} = \pm 1$). We measured the HD defined as :

$$\mathrm{HD} = \mathrm{PAD}^{(\sigma_{515},\sigma_{1030},m_{1030})} - \mathrm{PAD}^{(\sigma_{515},\sigma_{1030},-m_{1030})} \quad (1)$$

and observed a $\sim 10\%$ helical dichroic signal (Fig (a)), reflecting the non-dipolar character of the ionization from Rydberg states[2]. Second, we increased the intensity of the 515 nm pulses to produce above-threshold ionization (ATI), and used weak 1030 nm pulses to induce continuum-continuum transitions, creating sidebands. We observed a $\sim 3\%$ HD at the sideband location, with opposite sign compared to the Rydberg ionization case (Fig. (b)). Last, we used strong 1030 nm elliptical pulses carrying OAM to produce ATI in chiral fenchone molecules. Remarkably, at low laser ellipticity the HD reveals interference structures in the low momentum area (Fig. (c)). The modeling of these results is ongoing, as well as measurements to extract chirosensitive signals as observed in absorption experiments [3].

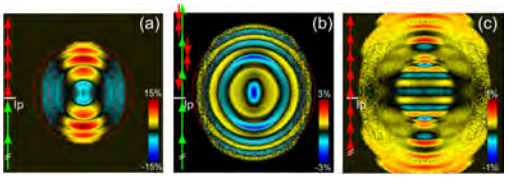


Figure 1. Normalized HDs measured with 1030 and 515 circularly polarized pulses in ethanol (a-b) and 1030 nm elliptically polarized pulses in fenchone (c)

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From megabarns to attoseconds: How to relate the cross-section with the time delay

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Synopsis Photoemission is delayed relative to absorption of ionizing radiation. We demonstrate that this delay is related to the photoionization cross-section by a simple Hilbert transform connecting the modulus and phase of the ionization amplitude. Several examples including resonant ionization processes and Cooper minima are used as illustrations. Extension from single-photon to two-photon XUV+IR ionization is also demonstrated.

The Kramers-Kronig (KK) relation connects the real and imaginary parts of an analytic function by way of the Hilbert transform [1]:

$$\mathcal{H}\left\{\mathrm{Re}\ g(E)\right\} \equiv \frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{\mathrm{Re}\ g(x)dx}{x-E} = -\mathrm{Im}\ g(E)$$

We apply the KK relation to the logarithmic derivative of the ionization amplitude $f(E)$ as a function of the photoelectron energy E

$$g(E) = \frac{f'(E)}{f(E)} = \frac{1}{2}\frac{\sigma'(E)}{\sigma(E)} + i\tau(E)\ ,\ \sigma(E) \propto |f(E)|^2$$

Such a logarithmic Hilbert transform (LHT) relates the photoemission time delay, also known as the Wigner time delay [2], with the photoionization cross-section:

$$\tau(E) = \frac{\partial \arg f(E)}{\partial E} = \mathcal{H}\left\{\frac{1}{2}\frac{\sigma'(E)}{\sigma(E)}\right\}$$

Application of the LHT requires the knowledge of $f(E)$ in the infinite limits of the photoelectron energy. This can be bypassed in the case where $f'(E)$ dies out quickly beyond a distinct feature such as a resonance or a sharp (Cooper) minimum in the photoionization cross-section. It is these features that are marked by a rapid variation of the photoelectron phase and a large magnitude of the time delay.

The LHT can be used to relate the cross-section and time delay in a large number of single-photon resonant ionization processes. It provides a good estimate of time delay near atomic and molecular shape resonances [3], confinement resonances in the endohedrally trapped Xe@C_{60} and the Cooper minima in the valence shells of noble gas atoms [4]. Fano resonances can also be treated in the similar way. Particularly interesting is application of the LHT to two-photon XUV+IR ionization where the Fano resonance appears in one of the two interfering channels of the RABBITT process [5]. Such a case is illustrated in the Figure.

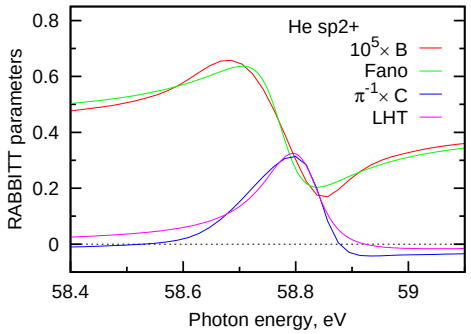


Figure 1. RABBITT magnitude B and phase C parameters near the $sp2+$ resonance in He. The numerically calculated parameters agree closely with the Fano parameterization for the magnitude and the LHT for the phase.

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Quantum trajectory selector: a novel method to clock attosecond dynamics within recollision physics

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Synopsis We introduce Quantum Trajectory Selector (QTS), a novel measurement scheme capable of resolving individual quantum orbits responsible for strong-field phenomena in real time. By combining an extreme ultraviolet (XUV) attosecond pulse train with a phase-locked, strong infrared (IR) field the time of ionization and thus the launch of an electron along a specific quantum trajectory can be chosen. We show that there is an intensity-dependent shift in the ionization time associated with double ionization in argon, and we clock this shift as it varies by 250 as.

Understanding electron dynamics on the femto- and attosecond time scales in atoms and molecules is not only of great general fundamental research interest to the scientific community [1] but also has the potential of many future applications: once we understand them, we might also be able to control them.

We introduce a novel, self-referencing method for clocking individual quantum trajectories on the attosecond time-scale to investigate recollison physics, called Quantum Trajectory Selector (QTS) [2], based on a previously introduced theoretical model [3]. In Figure 1, the principle is illustrated in the picture of the semi-classical three-step model [4, 5]. In contrast to tunnel ionization with a strong infrared (IR) field only, where a multitude of different electron trajectories is enabled (top), the QTS scheme combines the IR with an attosecond XUV pulse. The time window for ionization is reduced drastically as photo-ionization due to the XUV, creating a near-zero kinetic energy electron in the continuum, dominates over the tunnel ionization induced by the IR field. Using this method, ambiguities that arise from large number of quantum trajectories resulting in identical observables are limited to a minimum. In a recent publication we use the QTS scheme on an argon target to clock intensity dependent shifts in ionization time for double ionization and rescattered electrons [2]. The QTS technique will be extended to different parameter regimes to perform measurements including targeting core-level dynamics in atoms and molecules.

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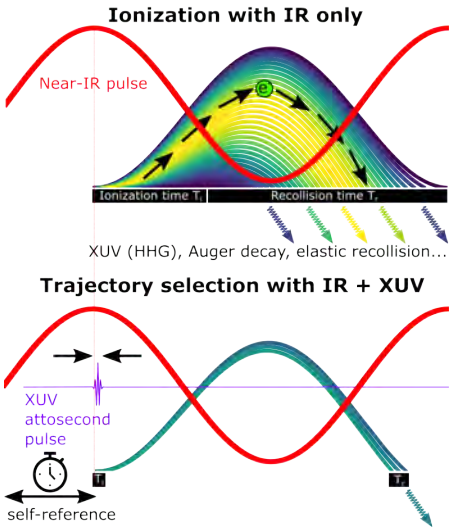


Figure 1. Comparison of ionization with an IR field only, enabling a multitude of possible quantum trajectories (top) with the QTS scheme where the time window for ionization, and thus recollision is shortened sufficiently (bottom).

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Orientation-dependent production of electron spirals from multi-orbital heteronuclear diatomic molecules

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Synopsis The discovery of symmetric Archimedean spirals from atomic ionization by a sequence of broadband oppositely circularly polarized pulses revealed their potential of discerning orbital symmetry in atoms. Here, this method is extended to hetero-nuclear diatomic aligned molecules where multi-orbital ionization is possible. Although broadband pulses create multiplexed signals, hiding thus any molecular orbital information; we show how molecular orientation can allow selective ionization control by revealing asymmetries in the HOMO.

The discovery [1] and measurements [2] of symmetric normal Archimedean spirals from atomic ionization by a pair of time-delayed broadband counter-rotating circularly polarized (CRCP) pulses revealed their potential of discerning orbital symmetry in atoms. Transferring this tool to molecules increases theoretical and experimental challenges. Not only calculations are nontrivial due to the involvement of many degrees of freedom, molecules need to be aligned. The available ultrashort pulses have larger spectral width ΔE, expected to dramatically affect the initial, intermediate, and final states of ionization processes in molecules. Indeed, ionization from multiple orbitals is possible if ΔE is larger than the energy gap between the outer and inner orbitals. The congestion of electronic intermediate states is possible in multiphoton ionization, leading to dissociative ionization among other channels. Finally, the photoelectron momentum distributions (PMDs) from multi-orbital ionization are expected to be complex.

In this talk, to bypass the congestion associated with intermediate states we consider the Einstein's photoelectric effect of the closed-shell HF and CO molecules, with the aim of using the pattern of the PMD as a probe for the asymmetric property of their highest-occupied molecular orbital (HOMO) [3]. Here, the pulse bandwidth is broad such that an electron from the HOMO-1 can also be ionized leaving HF^+ or CO^+ in an excited state. For CRCP pulses propagating along the molecular axis, the resulting PMD shown in the left panel of Fig. 1 for CO is found to be intricate, hiding any information about the asymmetry of the HOMO. However, for light propagating perpendicularly to the molecular axis the right panel of Fig. 1 exhibits an asymmetric spiral, reflecting the HOMO asymmetry. Although disentangling contributions of electrons emitted from different orbitals is far from being straightforward, our results demonstrate how this goal can be achieved by molecular orientation with respect to the light propagation direction. First-order perturbation theory is used to analyze our R-matrix with time dependence (RMT) [4] calculations.

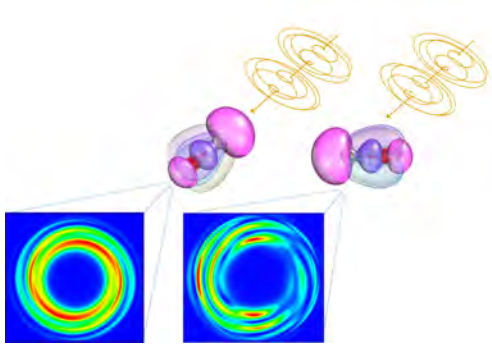


Figure 1. PMDs for photoionization of aligned CO molecule when the time-delayed CRCP pulses propagate parallel (left) and perpendicular (right) to the molecular axis. Laser parameters are: intensity $I = 1$ TW/cm^2, carrier frequency $\omega = 20$ eV, and pulse duration $T = 1.2$ fs, and time delay $\tau = 1.5$ fs.

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Guiding synthetic chiral light

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Synopsis We show that synthetic chiral light arises naturally during the propagation of multicolor fields in hollow dielectric waveguides and can be guided under nearly phase-matched conditions. We demonstrate efficient enantio-sensitive sum-frequency generation in gas phase chiral molecules. Our results establish natural way of generating locally and globally chiral light and pave the way to a new generation of on-chip photonic devices for extremely efficient chiral sensing.

An object is called chiral if it can not be superimposed with its mirror image. Ever increasing understanding of the importance of molecular chirality [1] constantly shifts the accuracy standard for detecting enantiomeric excess to the combination of higher precision and speed, where attosecond electronic response to light gives clear advantage to optical methods. However, standard optical methods rely on ineffective interactions with the magnetic field component of light and are thus severely limited in accuracy by the ratio of the molecular size to light's wavelength, making enantiosenstive signals very small. The fast and accurate detection of enantiomeric excess of medium-sized chiral molecules in dilute samples is therefore an outstanding challenge. Synthetic chiral light [2] is an appealing concept, where local polarization of phase-locked two- or multi-color laser beams can be shaped within a single cycle, leading to control over light's chirality in time, locally in space, in full 3D, enabling all-optical enantio-sensitive detection of chiral molecules with unprecedented sensitivity [2]. However, creating 3D polarizations in free space has proved to be challenging. Here we show that using hollow core photonic waveguides one can take advantage of the longitudinal electric fields naturally arising due to spatial confinement, leading to 3D chiral polarization of such fields and enabling many millimeters long phase matched guiding of enantio-sensitive signal. We solve coupled Maxwell-Schroedinger equations to investigate the enantio-sensitive response generated by the co-propagating two-color field with controlled two-color delay $\phi_{\omega,2\omega}$ at the input of the fiber (70 μm radius). Our implementation involves coupling the driving fields into carefully chosen propagation modes of the gas filled (Carvone, 10 $mbar$) hollow fiber. The IR(800 nm, $5\times10^{12}W/cm^2$) driver is coupled into the Transverse Magnetic mode TM_0 while the phase locked VI driver(400 nm, $5\times10^{11}W/cm^2$) is propagating in the Transverse Electric mode TE_1. Fig 1 shows phase matched generation of enantiosensitive UV(266 nm) signal via interfering chiral and achiral pathways with chiral dichroism (CD) reaching about 10%. We gratefully acknowledge ERC-2021-AdG project ULISSES, grant agreement No.101054696.

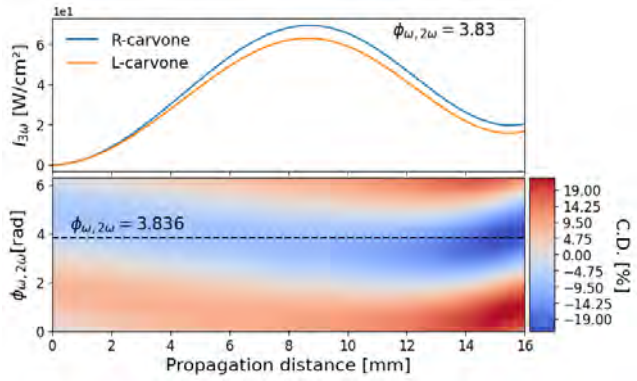


Figure 1. Chiral sum-frequency generation in a fiber filled with carvone gas. (a) Sum-frequency signal vs propagation length for $\phi_{\omega,2\omega} = 3.83$ rad for right (blue) and left (orange) carvone. (b) Chiral dichroism as a function of $\phi_{\omega,2\omega}$ and the propagation length.

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Attosecond transient absorption in the water-window regime driven by self-compressed light-transient pulses

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Synopsis We showcase the first results from an attosecond transient absorption spectroscopy (ATAS) beamline operating in the water window, driven by light transient mid-IR pulses (2.9-5.2 fs) generated via soliton self-compression in a gas-filled hollow-core fiber. These pulses drive soft X-ray HHG and, through a split-toroidal mirror, provide pump-probe measurements with ≈ 9 as delay stability. The first field-resolved measurements of these pulses provide sub-fs information of the pump pulse at the sample site, enabling attosecond-resolution studies of electronic coherence with atomic site specificity.

A long-standing goal in attosecond science is the ability to track electronic coherences in matter. High-harmonic generation (HHG) enabled attosecond pulses, and its extension to core-edge transitions has opened new avenues for probing ultrafast dynamics with atomic resolution [1]. However, realizing the full potential of attosecond pump-probe spectroscopy also demands advances in the driving pulses. Soliton self-compression offers a promising route to few-cycle, high-energy pulses [2], yet its potential as the ideal pump has remained unexplored so far.

Here, we present the first results from a beamline optimized for attosecond transient absorption spectroscopy (ATAS) exploiting soliton self-compressed light transients. The setup utilizes 2.4 mJ, 35 fs, carrier-envelope-phase (CEP) stable pulses at 1.8 μm to drive soliton self-compression in a 2.6 m stretchable hollow-core fiber (HCF). The resulting single-cycle mid-IR pulses directly drive soft X-ray (SXR) HHG and are simultaneously recycled as the pump via a split-toroidal mirror setup. This approach enables ATAS in the water window, reaching energies beyond the nitrogen K-edge, while ensuring unprecedented monolithic delay stability (≈ 9 as). We implemented the CRIME technique [3] for high-precision in situ pulse characterization, achieving the first field-resolved measurements of soliton self-compressed pulses at the sample position with attosecond accuracy. This enables tracking and modeling the light-electron interaction on its natural timescale.

Light transient mid-IR pulses (2.9 fs FWHM), Fig. 1a, have been attained with 530-μm HCF filled with Ne at the expense of poor SXR radiation due to excessive blue-shifting. In contrast, a 450-μm He-filled HCF at 3.5 bar, with an additional 1 mm fused silica plate, yields quasi-single-cycle pulses (5.2 fs FWHM) and enough SXR flux, Fig. 1b, to measure ATAS, illustrated through measurements in argon, Fig 1c, and organic molecules.

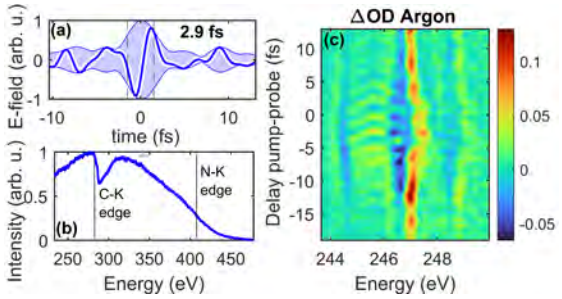


Figure 1. **(a)** CRIME reconstruction of the shortest measured light transient. **(b)** HHG spectrum with 5.15-fs pulse and its correlated **(c)** Ar-ATAS.

With these light transients, strong-field ionization is confined to a single half-cycle, enabling sub-femtosecond resolution in water-window ATAS. This breakthrough will open new possibilities for attosecond spectroscopy of coherent electronic motion, charge migration at multiple core-edges, and strong-field pump-driven ultrafast dynamics in atoms and molecules.

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Tuning the Fano resonance line shape in hundred zeptosecond timescale by using two attosecond pulses

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Synopsis We use two extreme ultraviolet (XUV) attosecond pulses as pump pulses and a femtosecond infrared (IR) pulse as a probe pulse to control the dynamics of helium 2s2p autoionization state and the profiles of attosecond transient absorption (ATA) spectra. Our results show ATA spectra are highly sensitive to variations in the time delay between the two attosecond pulses. Even within a range of several hundred zeptoseconds, the line shape undergoes significant changes.

The asymmetric Fano line shape is generally generated in the course of excitation of discrete quantum states embedded in and coupled to a continuum state [1]. The evolution of the atomic Fano line shape can be reconstructed [2] and regulated [3] through the XUV pump - IR probe scheme.

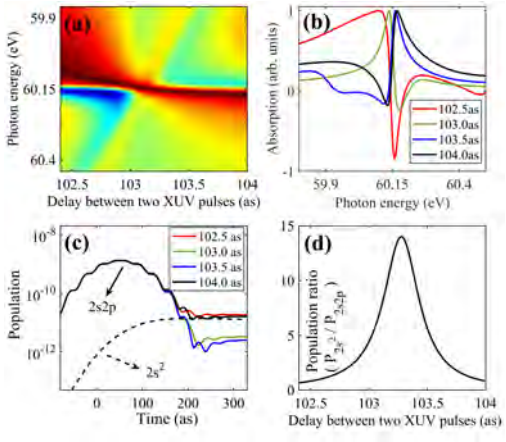


Figure 1. (a) The ATA spectra with time delay between two XUV pulses. (b) The absorption lines from (a) for some selected delay. (c) The evolution of the population of bright state $2s2p$ and dark state $2s^2$ over time during the pump process. (d) When both XUV pulses end, the population ratio of the dark state $2s^2$ to the bright state 2s2p varies with the time delay between the two XUV pulses.

In this theoretical work, we use two attosecond XUV pulses as pump pulses to excite the helium atom, with a time delay of τ_x. And use a femtosecond IR pulse as the control pulse, with a time delay of τ from the first XUV pulse. For the generation of two XUV pulses, we simulated the method used for autocorrelation, which is to completely divide one beam of XUV into two parts and control their time delay, so that these two beams of XUV have a completely time shift relationship in the time domain. Therefore, the phase difference between these two XUV pulses at frequency ω is $\Delta\phi = \omega\tau_x$. By changing τ_x, the phase difference of two XUV pulse at a frequency can be altered, resulting in different evolution of the population of excited states during the pumping process as shown in Fig.1 (c), and then causing changes in the ATA spectra.

As shown in Figure 1 (a) and (b), when the time delay of the two attosecond pulses is around 103 as, the ATA spectra undergoes a drastic change with the variation of the time delay of the two attosecond pulses, although the time delay variation is only 0.5 as, the corresponding modulated Fano lines are also very different. The reason for this, as shown in Fig. 1 (d), is that at the end of XUV, the population ratio of dark and bright states is very sensitive to the time delay, resulting in significant differences in the subsequent autoionization dynamics and fano lines.

This work provide a scheme to control the motion of electrons and resonance lines within the few-hundred zeptosecond time scale.

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Heterodyne analysis of high-order partial waves in attosecond photoionization of Helium

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Synopsis We propose a heterodyne analysis approach, based on the delay-resolved anisotropy parameters to reveal the role played by high-order partial waves during photoionization. This extends the RABBITT technique into the few-photon regime. Even for moderate ($\sim 1TW/cm^2$) intensities, near-infrared-assisted photoionization of helium through Rydberg states results in a tiny contribution from the g_0 wave, which has a significant impact on the PADs via interference with the s- and d_0-waves. This modulation also causes a substantial deviation in the angular distribution of the recovered spectral phase shift.

Attosecond chronoscopy is central to the understanding of ultrafast electron dynamics in matter from gas to condensed phase with attosecond temporal resolution. One of the most commonly used methods to determine ionization time delays is the RABBITT. Only partial waves that follow the two-photon transition rules contribute to the observed side-bands in general RABBITT, but at higher NIR intensities, higher-order partial waves arise through multiphoton transitions. It has, however, not yet been possible to determine the effects of higher-order partial waves, and steering their contribution has been a substantial challenge.

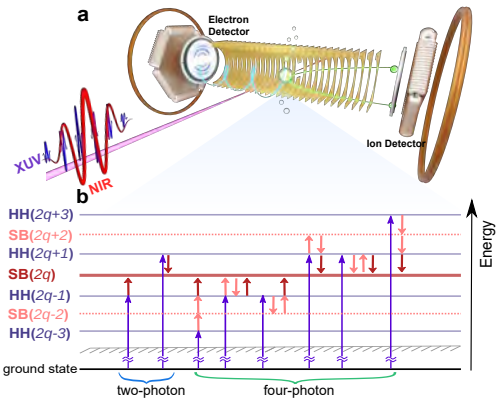


Figure 1. Schematic of the attosecond coincidence interferometer.

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In this work, we outline a technique akin to heterodyne detection, using the interference between partial waves to characterize the high-order contribution to RABBITT in helium atoms, as illustrated in Fig. 1. We get the attosecond delay-resolved asymmetric parameter β_n from the experimental and RMT simulated angular distribution of photoelectrons, and then use Fourier transform to extract the oscillation components. The significant β_6 parameter is extracted from the $4\omega_{\mathrm{NIR}}$ oscillating components. It comes from the interference between d_0 and d_0 waves, and d_0 and g_0 waves, and thus directly proves the population of higher order partial waves, g_0 waves. Further analysis of angle-resolved phase shift distributions in Rydberg state resonance ionization channels shows that, although the contribution of g_0 wave is small when the intensity of NIR detection light increases to TW magnitude, its interference with s and d_0 waves can have an important effect on the photoelectron ionization delay [1]. We provides an efficient method to resolve isolated partial wave contributions beyond the perturbative regime and paves the way towards understanding resonance-enhancement of partial waves.

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Exciting autoionizing states with multiple photons

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Synopsis Fano lineshapes arising from autoionization of states excited by multiple photons are studied numerically. We find significant changes in the lineshapes depending on the order of the multiphoton process. These variations can be traced to the phases of the underlying multiphoton transition matrix elements. The computations were enabled by the tRecX-haCC package developed by the authors.

Intense coherent extreme ultraviolet (XUV) pulses that are capable of inducing multiphoton processes in atoms and molecules are being increasingly available. At certain wavelengths, these pulses can excite atoms into doubly excited states (DES) that autoionize, in addition to direct photoionization. The autoionization results in asymmetric Fano line-shapes in photoelectron spectra. The width of the line-shape is determined by the lifetime of the DES and the asymmetry is determined by the relative phases between the direct ionization pathway and the pathway involving the decay from the DES.

Fano line-shapes following single photon ionization have been widely reported for various systems in literature. However, studies involving multiphoton processes are rare. We present here our recent theoretical studies on multiphoton Fano lines for atoms (He and Ne) [1] and nitrogen molecule [2]. For a given DES, we find that the asymmetry in the line-shape depends on the number of photons absorbed. In Fig. 1, this is demonstrated using the example of one- and three-photon ionization of neon. These variations can be traced to the phases of the underlying multiphoton transition matrix elements.

The numerical studies were performed using the tRecX-haCC package developed by the authors [3]. We solve the time dependent Schrödinger equation in the presence of an external field using a hybrid anti-symmetrized coupled channels basis for the multielectron wavefunction and compute the photoelectron spectra using the infinite time extension of the time dependent surface flux method (iSurff). Resolving the sharp Fano lines using iSurff requires solving many large systems of linear equations which is efficiently achieved using generalized minimal residual method with a flexible shift-and-invert preconditioner.

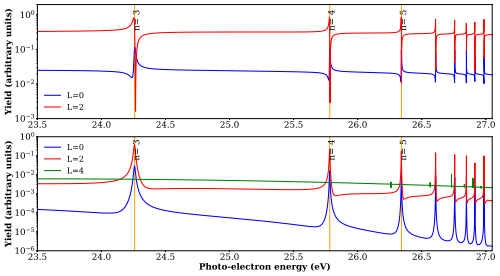


Figure 1. Partial wave analysis of the photoelectron spectra for Neon atom ionized by 2-cycle pulses with $\cos^8$ pulse envelope and peak intensity of 10^{14} W/cm^2 . Upper panel: 27nm (1 photon process), Lower panel: 81nm (3 photon process)

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Imaging ultrafast structural rearrangement in ammonia molecule with high harmonic spectroscopy

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Synopsis We demonstrated a machine learning (ML)-assisted high harmonic spectroscopy (HHS) approach to study the ultrafast structural rearrangement in ammonia molecule after ionization. By leveraging the sensitivity of molecular recombination dipole moment to molecular structures, the temporal evolution of the nuclear geometry of ammonia was directly reconstructed with attosecond resolution from the angle-resolved single-molecule harmonic dipoles retrieved by the ML algorithm. Our approach provides a general and robust route towards imaging ultrafast nuclear dynamics in polyatomic molecules.

Dynamic imaging of structural rearrangement within single molecules is crucial to the fundamental understanding of complex chemical reactions or biological functions. Achieving this fascinating goal requires imaging techniques with combined atomic spatial and femtosecond to attosecond temporal resolutions. In the last two decades, advances in the studies of interaction between intense lasers and molecules have promoted the development of molecular imaging approaches, such as laser-induced electron diffraction (LIED) [1] and high harmonic spectroscopy (HHS) [2], to study molecular dynamics with femtosecond to attosecond time resolutions.

Specially, it has been reported that nuclear motion during the ionization and recombination steps plays an important role in molecular HHG. By incorporating the nuclear motion into the standard Lewenstein model, Lein [3] theoretically predicted that laser-driven nuclear motion will introduce an amplitude modulation in the harmonic spectrum via the nuclear autocorrelation function, and the information about nuclear dynamics can be reconstructed from the harmonic ratios of two isotopic molecules with sub-femtosecond time resolution. With this approach, the probe of proton motion in hydrogen molecule has been demonstrated in experiment [4]. Later, this method was also employed to study the nuclear dynamics in polyatomic molecules, e.g., CH_4 [5] and NH_3 [5-7], both theoretically and experimentally. However, in these polyatomic molecules, the nuclear dynamics can only be indirectly deduced from the measured nuclear autocorrelation function. A quantitative measurement of the molecular structure is still a challenge due to the increased degree of freedom of molecular geometry.

Here, we show that direct imaging of nuclear dynamics in polyatomic molecules can be achieved by using a machine-learning (ML) based HHS method. Different from Lein's method, our scheme relies on the sensitive dependenc of molecular recombination dipole moment on the molecular structure. With the help of the ML restruction algorithm [8], we are able to retrieved the angle-resolved single-molecule harmonic dipole moments from the HHG signals measured from aligned NH_3 molecules. The ultrafast structural rearrangement of NH_3 is then successfully reconstructed from the retrieved harmonic dipoles based on a modified HHG model. We have demonstrated our scheme in the 800 nm and 1300 nm experiments, and also in the isotopic molecule ND_3. All the reconstruction results are in good agreement with the quantum chemistry calculations.

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New 1D atomic model potential for more accurate HHG spectra

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Synopsis Combining two soft-core Coulomb model potentials, we introduced an upgraded 1D Gaussian-windowed soft-core Coulomb model potential for simulating a single active electron atom in a strong, linearly polarized laser field. This potential enhances accuracy and computational efficiency of quantum simulations for high-order harmonic generation (HHG) spectra.

Building on the favorable properties of previously used one-dimensional (1D) atomic model potentials, we introduced a new 1D Gaussian-windowed atomic model potential in [1] for simulating the quantum dynamics of a single active electron atom driven by a strong, linearly polarized near-infrared laser pulse. This new model potential upgraded the accuracy of numerical simulations for single-atom HHG spectra under commonly used driving laser pulse parameters.

We utilize the conventional soft-core Coulomb (SC) potential:

$$V_{SC}^{1D}(z) = -Z \cdot (z^2 + \alpha^2)^{-1/2},$$

where α is a smoothing parameter. We also make use of the improved soft-core Coulomb potential with modified parameters (MSC) [2], which is based on the 3D ground state density:

$$V_{MSC}^{1D}(z) = -Z^* \cdot (z^2 + \beta^2)^{-1/2},$$

where for single elenctron atoms $Z^* = Z/2$, $\beta^2 = 1/(4 \cdot Z^2)$. By combining the two we define the Gaussian windowed soft-core Coulomb (GSC) potential (with $a = 2.551$ and $b = 2$) as

$$V_{GSC}^{1D}(z) = V_{SC}^{1D}(z) \cdot e^{-\left(\frac{z}{a}\right)^2} + V_{MSC}^{1D}(z) \cdot \left(1 - e^{-\left(\frac{z}{b}\right)^2}\right).$$

The GSC potential offers an outstanding balance of accuracy and computational efficiency, enabling TDSE simulations that generate reliable HHG spectra within minutes in the 1.26–6.88×10^{14} W/cm^2 peak intensity range. Our research suggests that the GSC potential performs best when the Keldysh parameter is within $0.45 \leq \gamma \leq 1$ and the 1D ground state population loss simulated with the GSC potential, is not greater than 0.6 at the end of the laser pulse, which is well in line with most GHHG methods.

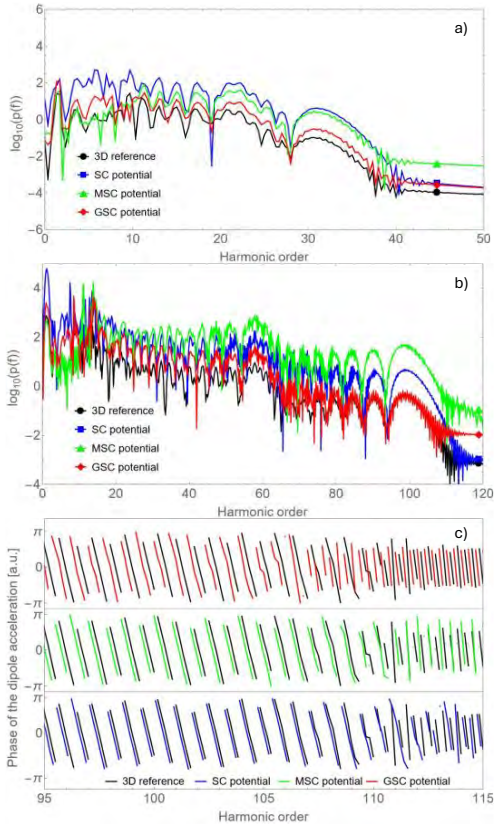


Figure 1. Assuming a laser pulse of 800 nm central wavelength and 3.1 fs pulse length, simulated (a) HHG spectra for Ar at 1.26×10^{14} W/cm^2; (b) HHG spectra and (c) spectral phase for Ne at 6.88×10^{14} W/cm^2 peak intensity. Results with a 3D reference simulation (black), SC (blue), MSC (green) and GSC (red) potentials.

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Attosecond electron dynamics in two-color multiphoton processes in helium

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Synopsis We look into electron dynamics in helium induced by different two-color multiphoton schemes. We study double ionization processes via two XUV pulses, demonstrating high degree of control of the total and energy-differential yields, and on the angular electron emission. Then, we examine IR-induced interferences in the photoelectron spectra upon ionization with phase-controlled specific harmonics. Our theoretical approach solves the time-dependent Schrödinger equation in full dimensionality, gaining insights in to electron correlations, and providing reliable data for comparison with experiment.

The widespread adoption of free electron lasers worldwide, capable of generating short and high-energy pulses, has greatly advanced the exploration of nonlinear phenomena in atoms. One such phenomenon, two-photon ionization, has revealed intriguing aspects of light-induced physical processes like asymmetric photoelectron emission [1, 2], providing insights into electron correlation. In this theoretical investigation, we explore the electron dynamics induced by multiphoton processes, with a focus on both single and double ionization. Our goal is to uncover deeper insights into the complexities of electron dynamics within the helium atom.

We first investigate double ionization processes in helium induced by two-color schemes. We analyze how the manipulation of double ionization yields is achieved by adjusting the time-delay between pulses of different colors. Furthermore, we showcase how this time-delay enables control and manipulation of angular electron emission, demonstrated through the variation of triple differential cross sections (TDCS) for different time-delays. Additionally, we demonstrate that strongly correlated two-electron states can result in both electrons being emitted in the same direction at specific energies. In the second scenario, we explore the interference of multiphoton transitions occurring between odd triple harmonics of an IR pulse. Our theoretical investigation focuses on retrieving all the relevant relative and absolute phases induced by both the atomic potential and those introduced by the electromagnetic pulses, in order to design new strategies to access phases of different wave packets.

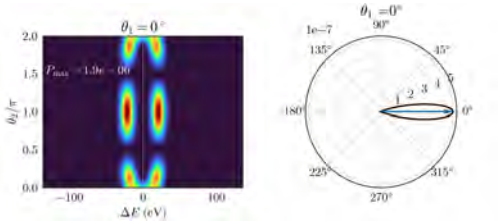


Figure 1. Energy-angular differential distribution and triple differential cross section of the two-photon double ionization of helium induced by a two-color scheme.

In both scenarios, we solve the time-dependent Schrödinger equation (TDSE) in full dimensionality, employing a numerical method that describes the wave function using a Finite Element Method and a Discrete Variable Representation (FEDVR). [3, 4, 5] Time evolution of the TDSE is achieved using a Lanczos propagator, while the scattering function is extracted by extending the propagation to infinite time via Exterior Complex Scaling (ECS) of the electronic coordinates.

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Retrieving the density matrix form experimental KRAKEN data

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Synopsis Recent research on electron-parent ion entanglement has lead to a new method for reconstructing the one-photon density matrix, namely KRAKEN. Using interferometric measurement of the two-photon electron distribution, one can fully reconstruct the single-harmonic one-photon density matrix. This communication uses well established numerical methods to retrieve density matrices from the experimental data.

The density matrix, ρ, of the photo-electron can be represented as,

$$\rho = \sum_I \iint d\epsilon_1 d\epsilon_2 c_I(\epsilon_1) c_I^*(\epsilon_2) \big|\epsilon_1\big\rangle \langle\epsilon_2| \,,$$

where the parent ion states, I, can be traced over, assuming the system is fully decoherent. If the system is coherent, cross terms between neighboring ground states are required to represent the density matrix.

KvanttillståndstomogRafi av AttoseKundsElektroNvågpaket (KRAKEN) [1, 2] is a recently developed attosecond Quantum State Tomography pump-probe method which uses a multicolour probe in order to fully reconstruct the single-harmonic one-photon density matrix. Due to experimental limitations, only a finite number of off-diagonals are directly measured.

The present communication proposes an algorithm for retrieving the full density matrix $\rho_{ij}^{(r)}$ (retrieved density matrix) from incomplete experimental data $\rho_{ij}^{(t)}$ (target density matrix), by parameterizing $c_I(\varepsilon)$ and minimizing the error

$$Err = \frac{1}{N^2} \sum_{i,j} \left| \rho_{ij}^{(r)} - \rho_{ij}^{(t)} \right|^2 .$$

The model does not assume decoherence, but quantifies the degree of coherence in the system.

Density matrices obtained by Relativistic Random Phase Approximation with Exchange[3] (RRPAE), were preprocessed to mimic the experimental data. Using the aforementioned method, density matrices were retrieved with high fidelity.

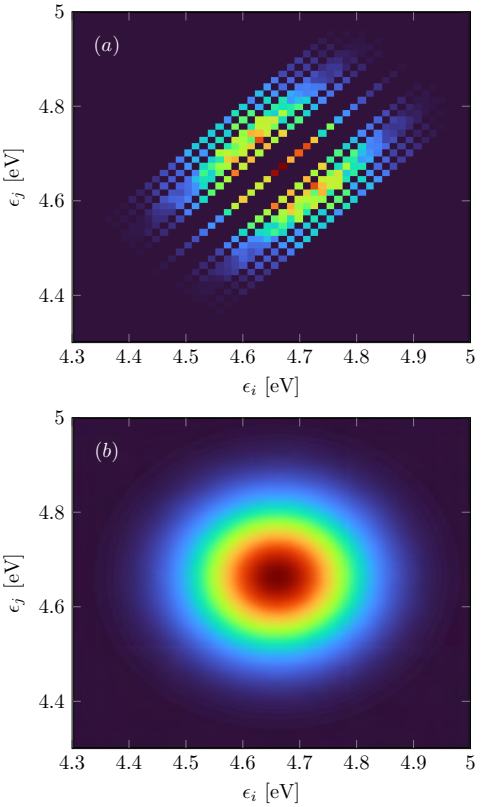


Figure 1. The amplitude of the density matrix (a) as obtained in experiment, with noise added and only certain diagonals present; and (b) retrieved from (a) by the algorithm (Fidelity $F = 0.999959$).

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Partial-wave transitions in attosecond time delays

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Synopsis Traditional methods interpret the RABITT delay as a sum of the Wigner delay and the continuum-continuum delay. This is unreliable at low photoelectron kinetic energies and does not account for angular dependency. Instead, we propose an alternative analytical approach that accurately predicts correction terms down to the threshold and incorporates the angular dependence of the continuum-continuum delay. This method is computationally straightforward and we present its validation against ab initio calculations and comparison with experimental data obtained from a pure two-harmonic RABITT setup.

In our recent work [1] we tackle the intricate challenge of accurately interpreting photoionization delays in atoms and molecules measured using multiphoton interference techniques, with a particular focus on the Reconstruction of Attosecond Beating by Interference of Two-photon Transitions (RABITT). Specifically, we investigate implications of a "two-harmonic RABITT" experiment performed in argon, featuring a single central sideband, two mainbands and two outer sidebands, each alone well usable for extraction of compatible timing information.

Current methods, which rely on separation of the measurable delays into Wigner-like delay and continuum-continuum delay τ_{CC}, often fall short at low photoelectron kinetic energies and fail to capture the correct angular dependencies. To address this, we have developed an innovative analytical approach that not only predicts correction terms accurately even at very low energies but also incorporates the angular dependence of the continuum-continuum delay. Our method is computationally efficient and has been rigorously validated against state-of-the-art ab initio calculations using the stationary multiphoton R-matrix method [2] and the R-matrix with time-dependence (RMT) [3].

By introducing a partial-wave-resolved continuum-continuum contribution, we achieve a precise modeling of the infrared field's influence on different photoelectron partial waves. This advancement allows for a clearer interpretation of both angularly resolved and angle-integrated RABITT delays, demonstrating excellent agreement with the full above-threshold perturbation theory. Our experimental setup, utilizing a simplified RABITT configuration, effectively isolates higher-order multiphoton pathways, offering a promising method for analyzing complex molecular photoionization spectra.

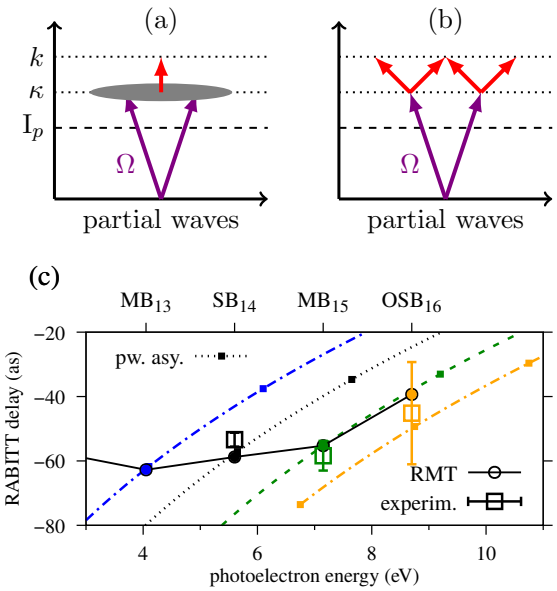


Figure 1. (a) Universal τ_{cc}. (b) Partial-wave resolution of amplitudes. (c) Emission-integrated 2H-RABITT delays in argon.

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Wigner vs. Smith: Time delays in anisotropic systems

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Synopsis Scattering properties and time delays for general, i. e. symmetric and non-symmetric, potentials in terms of the respective S-matrices are discussed paradigmatically in one dimension (1D). Only in the symmetric case so-called Wigner and Smith time delays coincide.

Time delays related to scattering phases have been discussed for a long time [1] in rather different contexts ranging from transport problems over acoustics and electro-magnetics to photo-ionization by ultra-short laser pulses. Experimentally, photo-ionization time delays have been extracted from streaking of electrons released by a short XUV pulse with an NIR field or so-called RABBIT measurements. The link of the photo-ionization time delay from streaking and the time delay from scattering theory has been questioned [2].

Recent experimental advances have made it possible to measure time delays originating in photo-ionizing molecules, that is from anisotropic potentials. This success motivates to ask for the theoretical foundation of time delays and their formulation for general interactions, since almost always time delay and S-matrices are discussed in the context of spherically symmetric potentials.

Here, we elucidate basic properties of time delays in the simplest setting [3], which is general enough to be sensitive to the properties of the (anisotropic and isotropic) potentials, namely scattering in 1D. We show, that contrary to the common usage of Wigner-Smith time delays, there are actually Wigner [1a] or partial time delays and Smith [1b] or proper time delays, which do not agree in general. In 1D the S-matrix and related quantities can be described by 3 parameters (α, β, γ) and the resulting time delays read [3] with $\xi' \equiv \frac{\mathrm{d}}{\mathrm{d}E}\xi$

$$\widetilde{\tau}_{1,2} = s'_{1,2} = \beta' \mp \frac{\cos\alpha\,\cos\gamma\,\alpha' - \sin\alpha\,\sin\gamma\,\gamma'}{\sqrt{1-\cos^2\gamma\,\sin^2\alpha}} \qquad (1a)$$

$$\overline{\tau}_{1,2} = q_{1,2} = \beta' \pm \sqrt{\alpha'^2 + \sin^2\alpha\,\gamma'^2}. \qquad (1b)$$

The partial time delays $\widetilde{\tau}_j$ are obtained from the eigenphases s_j of the S-matrix, the proper time delays $\overline{\tau}_j$ from the eigenvalues q_j of Smith' Q-matrix. Apparently they differ for $\gamma \neq 0$, cf. Fig. 1.

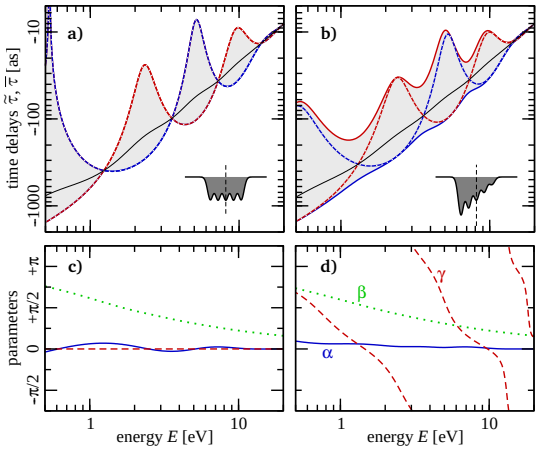


Figure 1. Upper raw: Time delays for a symmetric and an asymmetric potential as a function of energy E, according to Eq. (1). Partial time delays $\widetilde{\tau}_j$ (dashed lines) are compared to proper time delays $\overline{\tau}_j$ (solid lines). The lightgray-shaded areas between the proper time delays are the possible dwell times τ. Blue and red refer to the 1st and 2nd channel, respectively. Bottom raw: The corresponding parameters α (solid-blue line), β (green-dotted), γ (red-dashed) as a function of energy E. Figure from the 1D paper [3].

We furthermore show, that time delays depend on the location of the potential, a feature that is unexpected at a first glance.

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Extreme-Ultraviolet Optical Response of Atomically Thin MoS_2 Crystals

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Synopsis We report the first optical characterization of mono- and bilayer MoS_2 crystals in the extreme-ultraviolet energy range with multi-angle reflectivity measurements using synchrotron radiation. Unlike the visible-infrared region, their response does not present sharp excitonic signatures, as a result of strong electron correlations.

Transition metal dichalcogenides (TMDs) offer unique opportunities for the realization of ultrafast and ultrathin optoelectronic devices. Their optical response can be manipulated exploiting the strong interaction between ultrashort pulses, charge carriers, and excitons, unfolding on the sub-femtosecond time scale. Attoscience now allows for the real-time investigation of these extremely fast processes with pump-probe spectroscopy in the extreme-ultraviolet (XUV) energy range [1]. However, extending this technique to atomically thin crystals remains a formidable task relying on the optical characterization of few-layer TMDs in this photon energy region.

By measuring multi-angle reflectivity at the BEAR beamline of Elettra synchrotron light source [2], we characterized the XUV optical response of mono- and bilayer MoS_2 crystals for the first time. A nonlinear fit based on two different models for the Fresnel coefficient (thin slab, 2D conductivity [3]) retrieves the complex refractive index, in agreement with the X-ray optics (XOP) database (see Fig.1a).

To interpret the experimental results, we resort to numerical simulations based on ab initio Bethe-Salpeter equation and density matrix formalism (see Fig.1b). The broad resonance in the imaginary part corresponds to dipole-allowed transitions from the $4p_{3/2}$ (N_3 edge, 35.5 eV) and $4p_{1/2}$ (N_2 edge, 37.6 eV) semi-core levels of Mo to unoccupied electronic states in conduction band. Surprisingly, we observe no sharp excitonic feature in this energy range, which we ascribe to the effect of strong electron correlations.

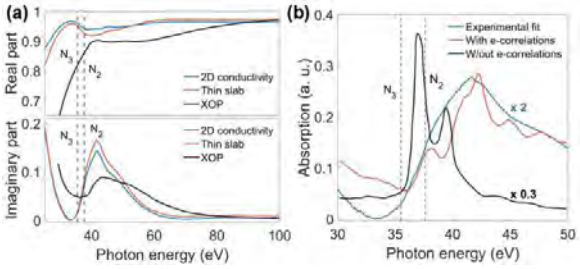


Figure 1. (a) Extracted refractive index of MoS_2 (blue for 2D conductivity, orange for thin slab model) compared to XOP data (black). (b) Comparison between fit (blue) and calculations with (orange) or without (black) electron correlations. Dashed lines indicate Mo N_3 and N_2 edges.

In conclusion, we report the first optical characterization of atomically thin MoS_2 in the XUV energy range, paving the way to pump-probe investigations of these materials with attosecond resolution.

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Effect of attosecond electronic coherences on conical-intersection dynamics in core-excited states

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Synopsis
Using fully quantum dynamical simulations, we investigate the creation and evolution of electronic coherences through conical intersections between core-excited states. We show that the electronic coherences created by conical intersections play a key role in the system's evolution. Our study demonstrates that controlling the initial coherent superposition opens new perspectives for steering chemical dynamics on electronic time scales.

Conical intersections (CI), often referred to as photochemical funnels, are regions where strong nonadiabatic effects enable ultrafast interstate crossings and generate electronic coherences. Recent advances in X-ray free-electron facilities [1] have made it possible to study such processes in core-excited or core-ionized states, opening new and unexplored territories. Here, we aim at studying ultrafast molecular dynamics of core-excited 1,1-difluoroethylene (CH_2CF_2), a system that exhibits CIs in its carbon K-edge potential energy surfaces (see Fig. 1).

Attosecond X-ray pulses can initiate core-excited dynamics by creating coherent superpositions of carbon (1s)-excited resonances. By carefully controlling the pump pulse's photon energy and selecting its linear polarization in conjunction with aligned molecules, one can tailor the initial superposition and thus influence the electronic coherences that emerge at the conical intersection. This strategy allows us to investigate how such coherences affect the branching ratios of electronic dynamics.

Although conical-intersection dynamics have been studied across various time scales, the impact of electronic coherences and the potential for coherent electronic control have remained largely unexplored in core-excited states. To address these challenges, we employ high level *ab-initio* calculations alongside a full quantum treatment of nonadiabatic dynamics using the multiconfigurational time dependent Hartree (MCTDH) method [2]. This complete quantum treatment of the nuclei is essential for accurately capturing the evolution of electronic coherences [3, 4].

In this study, we demonstrate how the initial coherent superposition affects the creation, evolution as well as the destruction of coherences through CIs. Together, this integrated approach opens new avenues for steering chemical dynamics on electronic time scales.

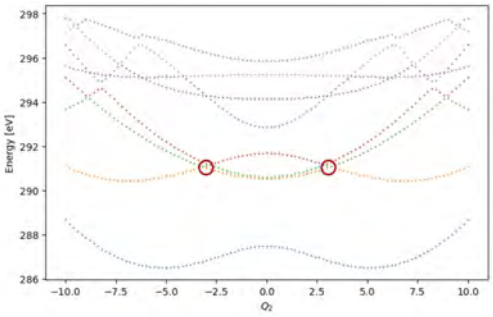


Figure 1. Potential energy surface along the F–C–C out-of-plane bending coordinate for the first eight carbon (1s) resonances (dots) and the corresponding conical intersections (red circles) in 1,1-difluoroethylene.

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Cross-Process Interference in Strong-Field Ionization: Insights from CEP-Dependent Photoelectron Spectra

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Synopsis Strong-field photoemission spectra are governed by cut-off features and interference patterns from direct and backscattered electrons. The spectral modulations due to inter- and intracycle quantum interference are commonly dominated by superposition of multiple quantum pathways belonging to the same process (self-interference). Here, we expose the opposite, i.e. cross-process interference (CPI), using single-cycle pulses and metallic nanotips. Quantum simulations predict distinct CEP-dependent CPI features, whose mechanisms are identified by an extended trajectory model.

Photoelectron spectra from strong-field ionization highlight classical and quantum effects in atoms, molecules, and nanostructures [1]. In a classical picture, direct and backscattered electron trajectories explain the well-known 2 U_p and 10 U_p cutoffs. Quantum trajectories, in turn, describe above-threshold ionization (ATI), where photoelectron peaks are spaced by the photon energy of the laser field. Additional modulations in ATI spectra arise from intracycle interference, caused by quantum pathways within a single optical half-cycle [2].

Typically, intra- and intercycle interference features result from self-interference of direct or backscattered electrons. However, cross-process interference (CPI) between these pathways can also shape photoelectron spectra, providing valuable insights. To isolate CPI, self-interference effects must be suppressed. This is achieved using intense, single-cycle optical pulses [3] that confine electron liberation to a single period. Additionally, sharp metallic nanotips restrict electron motion to one half-space, ensuring strong backscattering and suppressing revisits. Strong-field photoemission from nanotips closely resembles atomic ionization [4], with trajectory-based models successfully explaining sub-cycle tunneling [5] and attosecond electron emission [6].

To study CPI between direct and backscattered electrons, we analyze nanotip photoemission under CEP-controlled single-cycle pulses. Quantum simulations solving the time-dependent Schrödinger equation predict strongly CEP-dependent spectra with distinct interference features. Comparison with an extended trajectory model confirms their origin in CPI and explains their strong CEP dependence, offering new insights into the interplay of direct and backscattered electron pathways.

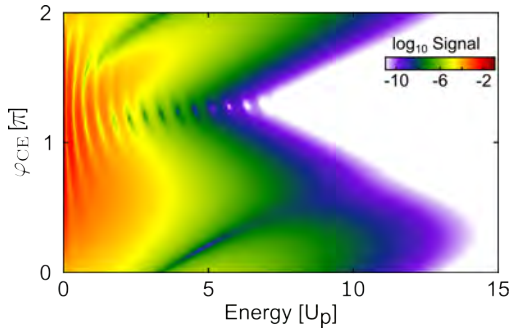


Figure 1. Energy spectra as function of the CEP for single-cycle driven strong-field electron emission from a nanometric metal tip extracted from quantum simulations.

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All-attosecond transient absorption spectroscopy of gas-phase molecules

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Synopsis We report on the first demonstration of all-attosecond transient absorption spectroscopy (AATAS) in gas-phase molecules, allowing us to investigate phase- and state-resolved vibrational wavepacket motion. Following ionization of O_2 by an attosecond pump pulse, bound-bound transitions in the generated cation were probed by a second attosecond pulse. These experiments benefit from the ability to specifically excite molecules in both the pump and the probe steps in combination with the extremely high temporal and excellent spectral resolution.

An important advantage of attosecond-pump attosecond-probe spectroscopy (APAPS) is that no strong laser fields are required, which are typically used in attosecond experiments and may change or mask the dynamics of interest. Furthermore, APAPS enables the investigation of dynamics evolving on timescales from the attosecond to the (few-)femtosecond range, which is difficult to achieve in traditional XUV-NIR pump-probe experiments. All-attosecond transient absorption spectroscopy (AATAS) is a particularly appealing technique, as it combines an extremely high temporal resolution with an excellent spectral resolution. Recently, first AATAS results were obtained at the free-electron laser LCLS and using HHG [2].

Here we report on the first application of AATAS to gas-phase molecules. To this end, attosecond pulses with a spectrum covering 16-35 eV and with an intensity on the order of 10^{14} W/cm^2 were generated using an 18-m-long HHG beamline [3,4]. The pump and probe pulses were selected using a split-and-delay unit, and they were focused in a noncollinear geometry, enabling separate detection of the pump and the probe spectra.

The attosecond pump pulse was used to ionize O_2, and the evolving dynamics were tracked using an attosecond probe pulse. Figure 1 shows changes in the optical density (OD) as a function of the photon energy and the time delay. The broad-band blue features are attributed to the depletion of O_2 molecules following ionization by the pump pulse. Furthermore, clear oscillating features with a period of 18 fs are visible, which are assigned to the dynamic motion of the vibrational wavepacket of the $X^2\Pi_g$ state of O_2^+. The narrow vertical structures between 21 and 22 eV are attributed to the vibrational progression structure of the $X^2\Pi_g$, v=1-8 states. Intriguingly, the individual vibrational states exhibit energy shifts as a function of the time delay. Details of the phase- and state-resolved coherent vibrational wavepacket motion will be discussed.

Our results open new avenues for the investigation of coupled nuclear and electron dynamics in molecules. The high stability and robustness of our attosecond source [4] enable systematic investigations and application of AATAS to more complex molecules. AATAS may be used, e.g., to study coherent electronic wavepacket motion and conical intersection dynamics.

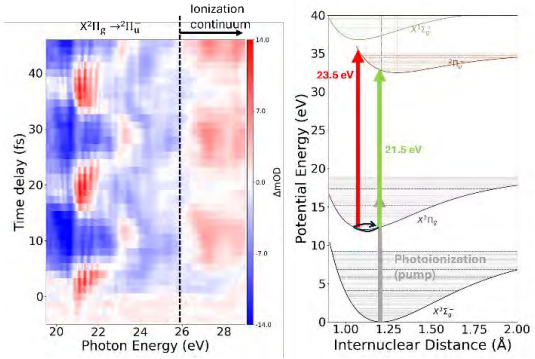


Figure 1. (left) AATAS map of O_2 in the 19-29 eV region, showing state- and phase-resolved vibrational wavepacket motion. (right) Potential energy curves of O_2. The green arrow represents the vertical transition after photoionization. The red arrow refers to the phototransition at the inner wall of the potential curve.

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Attosecond Raman time-delay in resonant transitions of ions

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Synopsis We report the probing of the transient absorption of Kr vacancy during its creation in near-infrared (NIR) strong laser field. By tracing the real-time evolution of the ionic states, we have observed ~400 attoseconds time delay between two resonant absorption channels. It is shown that coupling of the ions with the remaining field leads to suppression of ionic coherence, and coherent Raman coupling contributes to the attosecond time-delay. This discovery not only offers valuable insights into laser-ion interactions but also enriches the field of attosecond chronoscopy.

The generation of coherent extreme-ultraviolet (XUV) sources with pulse duration down to attoseconds empowers the exploration of ultrafast electron dynamics, which extends far beyond chemical reactions, delving deep into the atomic core across various phases of matter with attosecond-scale resolution. The burgeoning field of attosecond science has been demonstrated to have a profound impact on multiple disciplines, including condensed matter physics, materials science, chemistry and biology.

In this study, we employ isolated attosecond pulses (IAP) to investigate the transient absorption dynamics of krypton vacancies generated during strong-field ionization in intense near-infrared (NIR) laser field. Upon removal of a 4p electron via strong-field ionization, the resulting ionic system forms two spin-orbit split states that evolve as a spin-orbit wave packet within the residual laser pulse. As the detached electron escapes, the ion is statistically distributed among excited states with partial loss of the coherence. Since quantum coherence encodes spatiotemporal correlations in dynamical processes, the temporal characteristics of ionization are inherently linked to the coherent evolution of the ionic wave packet. Through precise control of the NIR-IAP time delay, we observe distinct modulation frequencies in two resonant absorption channels $4p_{3/2}^{-1} \rightarrow 3d_{5/2}^{-1}$ and $4p_{1/2}^{-1} \rightarrow 3d_{3/2}^{-1}$ as shown in Fig. 1, despite their mere 0.6 eV energy separation in the field-free regime [1].

By developing a comprehensive theory [2], we identified the crucial roles of coherence of ionization and the resulted ionic coherence. The continued interaction of the ion with the remaining laser field modifies the population distribution and their coherence among the vacancy states. It is shown the coherent Raman process leads to the observed time-delay. The concept of Raman-delay provides insight into laser-dressed bound-bound transitions as well as transient ionization injection.

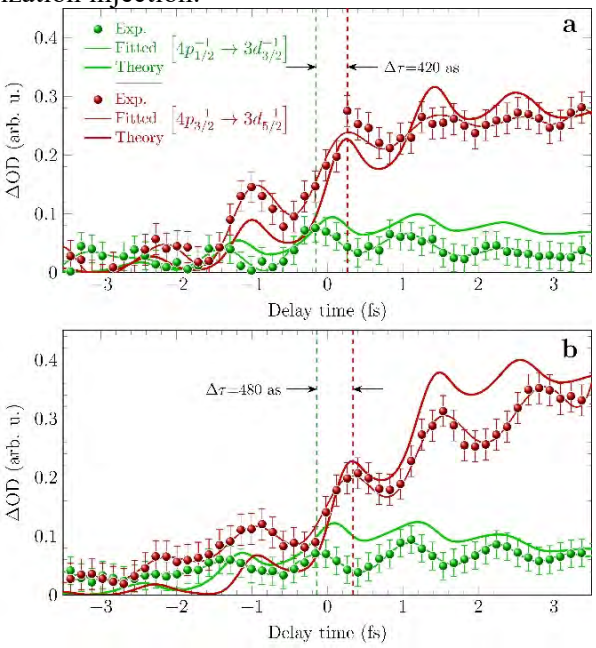


Figure 1. Delay-dependent ΔOD for transitions $4p_{3/2}^{-1} \rightarrow 3d_{5/2}^{-1}$ and $4p_{1/2}^{-1} \rightarrow 3d_{3/2}^{-1}$ show 420 as and 480 as time delay for two different laser intensities.

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Imaging in the water window with coherence tomography using high-harmonic generation

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Synopsis High harmonic generation (HHG) sources enable high-resolution imaging due to their spatial coherence and temporal precision. While widely used in the extreme ultraviolet (EUV) range, no imaging had been achieved in the soft X-ray water window (WW) due to low photon flux. This work presents WW imaging using a high-flux WW-HHG source and the flux-efficient coherence tomography method. Axial resolutions below 20 nm were achieved, enabling material studies beyond silicon, particularly oxides in semiconductor technology. The results were validated by SEM and TEM measurements.

Imaging with high harmonic generation (HHG) sources offers great potential due to their spatial coherence, high temporal resolution, and compact setup. This has driven the development of various extreme ultraviolet (EUV) imaging techniques, with lensless methods playing a key role. However, in the water window (WW) between 2.3 nm and 4.4 nm, HHG-based imaging has not yet been demonstrated due to limited photon flux and efficiency losses from monochromatization.

Here, we present WW imaging using a high-flux WW-HHG source combined with our flux-efficient imaging technique EUV and soft X-ray coherence Tomoggraphy (XCT) [1,2]. The source, driven by a 2.1 µm OPCPA system delivering 2.7 mJ pulses at 10 kHz, covers the full WW range up to 530 eV [3].

XCT, inspired by optical coherence tomography (OCT), is a Fourier-based imaging method using broad spectra and short coherence length for high axial resolution. By scanning the sample, cross-sectional and 3D imaging become possible. Previously, HHG-based XCT was limited to EUV applications. Extending it to the WW enables the study of more materials, particularly oxides, which are crucial as insulators in semiconductor technology. The sample analyzed consists of a Pt/Al_2O_3 layer on a ZnO substrate, achieving an axial resolution below 20 nm. The structure was validated using SEM and TEM.

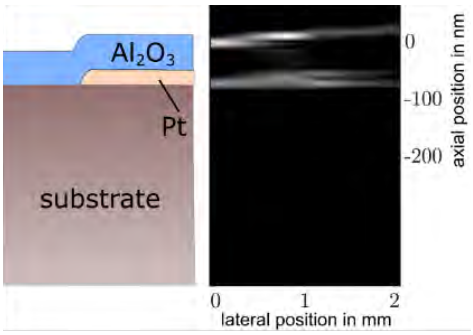


Figure 1. Cross-sectional image generated with OCT in the WW range using HHG. The structure and the rising of the Pt-layer can be resolved with <20nm axial resolution.

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Attosecond Optical Orientation

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Synopsis Circularly-polarized extreme ultra-violet (XUV) pulses of attosecond duration bring novel perspectives to ultrafast spectroscopy. Circular dichroism allows to study non-inversion symmetry dependent phenomena in matter such as orientation, chirality and topology. Here we introduce circular dichroic attosecond transient absorption spectroscopy to characterize the XUV polarization all-optically, measure orbital orientation of excited states in rare gas atoms and core-excitons of ionic crystals, and measure spin-polarization in semiconductors with attosecond temporal resolution.

In recent years, technical advancement has allowed for the generation of broadband extreme ultra-violet pulses of attosecond duration with circular polarization. This advancement has, for example, allowed for the probing of ultrafast magnetization dynamics using X-Ray magnetic circular dichroism [1,2]. However, the possibilities of employing circularly polarized attosecond pulses extend far beyond magnetization dynamics and include electron-spin dynamics and effects of broken inversion symmetry in matter such as chirality, topology and orientation.

We introduce circular dichroic attosecond transient absorption spectroscopy with co- and counter-rotating two-color XUV+NIR fields [3]. The technique is exemplified on the experimental attosecond optical orientation of He Rydberg states, which shows selectivity over magnetic quantum number specific coupling pathways (optical orientation).

The results are accompanied by time-dependent Schrödinger equation, as well as perturbative calculations. The results additionally show, how the technique can in turn be used to characterize the XUV polarization state in an all-optical geometry in a typical gas-phase XUV transient absorption spectroscopy experiment by varying the ellipticity of the NIR pulse. We furthermore present results of optical orientation with circular dichroic attosecond transient absorption of Ar and Ne autoionizing resonances, as well as core-exciton states of LiF, demonstrating broad applicability of the technique. Our results show how attosecond optical orientation can measure the orientation of excited state orbital contributions as well as, through spin-orbit coupling, spin-dynamics.

The latter is further shown in an experiment of circular dichroic transient absorption spectroscopy of photo-excitation of carriers in Ge which reveals an ultrafast decay of spin-polarization of holes deep in the Ge valence bands.

Our technique opens the door to combine the excellent temporal and spectral resolution of optical spectroscopy methods using attosecond pulses with effects of broken inversion symmetry, such as orientation and chirality and spin-dynamics in non-magnetic materials, including spin transfer dynamics across junctions. Sensitivity of the circular dichroic excitation of states to external perturbation could allow for example the development of ultrafast magnetometers [4] and bring quantum sensing to the attosecond domain.

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Simulation of transient-grating high-harmonic spectroscopy from dissiociating molecules

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Synopsis Transient grating created by irradiating two XUV pulses into a gaseous sample allows extracting dynamic information of the molecule through non- and first-order-diffracted harmonic signals. A model has been suggested and successfully applied for different cases, but it still has room for improving theory to match the timing and tendency of experimental measurement. In this study, we suggested a low-dimensional model for high harmonic from XUV transient grating. The results show that our model can improve the simulation with the minima's depth and tendency of the asymptotic region.

HHG from gaseous molecule-based transient grating has been measured and explained by using a simple model [1]. However, the simulated pump-probe-delayed signals are still different from the experimental data, particularly, the minima's depth and tendency of asymptotic region. This may come from the simplicity of the theoretical model, only the propagation phase of electron within one sub-cycle, the second one in the three-step model, is included to calculate the single-response HHG.

In this work, we construct a low-dimensional single-active-electron potential model for Br_2 molecule, by adjusting the potential parameters to mimic potential energy curves from *ab intio* calculation [2], including two lowest singlet states of neutral and the ground state of the cation. With this model, the time-dependent Schrödinger equation for the interaction of molecule with the external field can be solved numerically; thus the HHG from the single-molecule response is improved.

With this model, the simulated pump-probe-delayed signals consist better with the experimental data, including timing, the minima's depth near the zero time delay, and the tendency of the asymptotic region. Remarkably, we found that around zero time delay, where the pump and probe pulses overlap, the phase arising from electron propagation is perturbed by the XUV pulses. Because the phase difference between two adjacent half cycles is significant, up to π, the sub-cycle motion of the electron plays a crucial role in the presence of the minima.

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Probing of ultrafast nuclear vibrational and rotational dynamics with X-ray

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Synopsis We proposed several x-ray spectroscopy methods allowing for direct probing of the recoil-induced ultrafast nuclear dynamics, including recoil-induced fragmentation of the molecule, recoil-induced ultrafast molecular rotation probed by dynamical rotational Doppler effect, and time-resolved all X-ray pump–probe spectroscopy of recoil-induced rotation.

Photons and photoelectrons can transfer significant linear momentum as well as angular momentum in the processes of x-ray absorption, scattering, and ionization. The related recoil-induced vibrational and rotational excitations have received significant attention in x-ray spectroscopies. However, due to the limited resolution of conventional X-ray spectrometers, the rotational structure cannot be resolved directly through x-ray spectroscopies.

We proposed here several x-ray spectroscopy methods allowing for direct probing of the recoil-induced ultrafast nuclear dynamics, including recoil-induced fragmentation of the molecule, recoil-induced ultrafast molecular rotation probed by dynamical rotational Doppler effect, and time-resolved all X-ray pump–probe spectroscopy of recoil-induced rotation.

It was found that the large recoil energy delivered to vibrations and rotations can not only changes the vibrational progression structure but can also breaks the chemical bond [1]. The recoil-induced rotation can be probed by subsequent Auger electron emission shown as asymmetrical features in the Auger electron spectra[2] or probed by a time-delayed short probe X-ray pulse visualized as the revival structures in the x-ray absorption spectra (Figure 1) [3-4]. Furthermore, The amplitude of the recoil-induced revival structures for the individual ionization decreases monotonously with a decrease in the photoelectron energy, whereas the amplitude of the Cohen–Fano interference contribution remains sufficient even at the photoelectron kinetic energy below 1 eV. The profile and intensity of the interference depend on the phase difference between the individual ionization channels related to the parity of the molecular orbital emitting the photoelectron. This phenomenon provides a sensitive tool for the symmetry analysis of molecular orbitals.

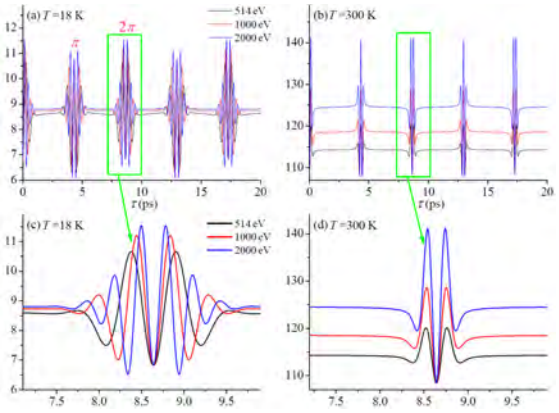


Figure 1. Revival structure of the probe O1s→5σ x-ray absorption for the CO molecule due to the recoil rotation after the ionization of the 5σ electron.

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Unifying two manifestations of multielectron polarization in high harmonic generation: peak's frequency shift and odd-even intensity modulation

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Synopsis Recently, we demonstrated the multielectron polarization (MEP) in the odd-even harmonic intensity of CO molecule interacting with multicycle laser pulses. This report reveals MEP effect on another important aspect of high harmonic generation. We found that MEP induces significant shifts in harmonic peak frequency compared to ignoring MEP when a polar molecule interacts with a few-cycle pulse. Furthermore, we reveal that these two MEP-induced effects share the same underlying: the MEP-induced distortion of the phase difference between two adjacent emission bursts in the time domain.

The interaction of ultrashort intense lasers with matter is commonly understood using the single-active electron (SAE) approximation. However, recent experiments have underscored the significance of multielectron effects, particularly multielectron polarization (MEP), which can alter nonlinear spectra. MEP affects both the orientation-dependent intensity of high-order harmonic generation (HHG) and the shape and intensity of the emitted HHG from molecules. While these changes can be subtle and challenging to measure, a significant MEP effect on the symmetric properties of HHG from the CO molecule has recently been identified and confirmed by existing experimental data. Specifically, MEP notably impacts the even-to-odd harmonic ratio, which is the intensity ratio between the even-order harmonics and the neighboring odd-order harmonics [1].

The odd-even harmonics mentioned above can only be detected using multicycle laser pulses. In contrast, harmonic peaks generated by few-cycle laser pulses may not be arranged in integer orders. Our recent findings indicate that the modulation of odd-even intensities and the frequency shifts of harmonic peaks can serve as indicators of laser-target asymmetry breaking in HHG when utilizing multicycle and few-cycle pulses, respectively [2]. This leads us to question how MEP affects the frequency of harmonic peaks generated by few-cycle laser pulses, particularly given its significant effect on the even-to-odd intensity ratio observed in CO molecules when exposed to a multicycle pulse.

In this report, we first highlight the significant harmonic frequency shift induced by the MEP effect in HHG emitted from CO molecules when exposed to a few-cycle laser pulse [3]. Notably, we observe MEP-induced frequency shifts of up to 0.8 orders (1.24 eV) for harmonics in the cutoff region during the interaction between the polar CO molecule and the few-cycle laser pulse. We provide an analytical explanation for these frequency shifts, which is further validated by numerically solving the time-dependent Schrödinger equation.

Importantly, we have identified a close relationship between the MEP-induced harmonic frequency shift discovered in this study and the previously reported MEP-induced intensity modulation observed in odd-even HHG [3]. We propose that these phenomena are two manifestations of the MEP effect on HHG, both arising from the distortion of the phase difference between two adjacent attosecond bursts. Understanding the unified diverse MEP manifestations in high harmonic generation is crucial for gaining insight into the physical nature of core electrons.

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State-resolved femtosecond phase control in dense-gas laser-atom interaction enabled by attosecond XUV interferometry

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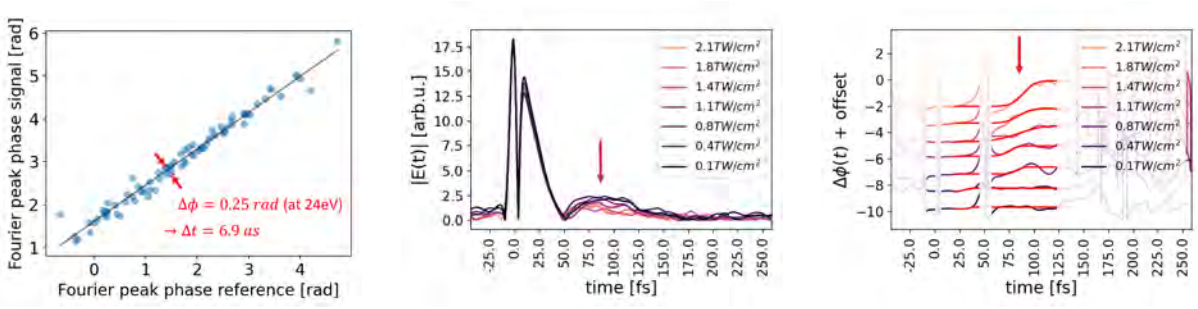
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Synopsis We demonstrate the interferometric measurement of extreme ultraviolet (XUV) pulses passing through a dense gas–phase target exhibiting narrow resonances in the presence of an intense femtosecond infrared laser pulse. Using the stable phase relation between two spectral regions allows us to reach an effective interferometer stability of 7 as. We benchmark this measurement approach with the $1s4p$ state of helium to reconstruct and control the temporal structure of the transmitted XUV pulse beyond the single–atom response.

The interferometric measurement of an extreme ultraviolet (XUV) attosecond pulse train passing through a dense gas–phase target in the presence of an intense near–infrared (NIR) laser pulse allows the reconstruction of the complex reshaped transmitted field $E(t) = |E(t)| \exp[i\phi(t)]$ in both amplitude and phase. To achieve the attosecond stability of the two optical paths required for interferometry in the XUV, we exploit the stable phase relation between two spectral regions of the same pulse train. In our case, two neighboring harmonics HH15 (signal) and HH17 (reference) are used to correct for instabilities during post-processing which results in an effective interferometer stability of 7 as (Fig. 1a).
With this method at hand, we analyze the resonant light–matter interaction with the $1s4p$ state of helium gas. Our approach allows not only to reconstruct the transmitted XUV pulse's amplitude with the subpulse structure characteristic of resonant pulse propagation (Fig. 1b), but also to demonstrate the femtosecond phase control of the interaction by means of a time–delayed intense NIR pulse (Fig. 1c). An NIR–intensity–controlled phase step between 0 and 2 rad is measured on the transmitted XUV pulse, originating from laser-induced transient Stark shifts. Thus, XUV spectral interferometry analysis enables the full reconstruction of the ultrafast dynamics beyond the single–atom response as imprinted on the transmitted XUV pulse.

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(a) (b) (c)

Figure 1: (a) Stable phase relation between HH15 and HH17. (b) Reconstructed XUV pulse's amplitude. (c) Reconstructed XUV pulse's phase difference with respect to the phase at lowest NIR intensity. Regions with low amplitude, where the phase is ill defined, are masked with transparent overlay. The NIR delay is ∼ 80 fs for both (b) and (c), its position is marked with the red arrow.

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Controlling the Orbital Angular Momentum of Electron in Neutral Atom

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Synopsis We demonstrate the precise control of orbital angular momentum in neutral atoms by integrating attosecond transient absorption spectroscopy with the Zeeman effect. Using density matrix simulations, we show that the orbital magnetic quantum number can be manipulated via external fields. This advancement paves the way for applications in chemical reaction control, magnetic property tuning, and the development of novel circularly polarized laser sources.

Angular momentum consistently plays a central role in quantum mechanics. The manipulation of these quantities remains a subject of profound interest for the scientific community. For example, a research in 2016 [1] reported achievement of *spin* polarization of electrons *ionized* from Xenon atoms. However, the majority of applications should be performed in their natural and stable states: neutral atoms or molecules. Moreover, orbital angular momentum has its importance over spin angular momentum as it directly governs the spatial distribution of electrons.

In this work, we numerically demonstrate the control of the orbital angular momentum of electrons in neutral atoms by integrating the attosecond transient absorption spectroscopy [2, 3] with the Zeeman effect [4]. The concept is that a static magnetic field facilitates the Zeeman splitting of orbitals with distinct orbital magnetic quantum numbers. Subsequently, XUV and IR pulses are employed to excite the ground-state atoms to the splitting states via electric dipole transitions. By manipulating the time delay between XUV and IR fields and the strength of the static magnetic field, the absorption probability of states with different angular momentum can be controlled.

Specifically, taking singlet He as an example, the asymmetrical factor defined in Equation (1), calculated from two orbitals with $m_j = 1$ and -1 of the $1s2p(^1P_1)$ state can be precisely controlled within a wide range, as shown in Figure 1. The underlying physics is the modification of the population of the involved states by the applied electric field and magnetic field.

$$\text{Asym. factor} = \frac{S_{m_j=-1} - S_{m_j=1}}{S_{m_i=-1} + S_{m_i=1}} \quad (1)$$

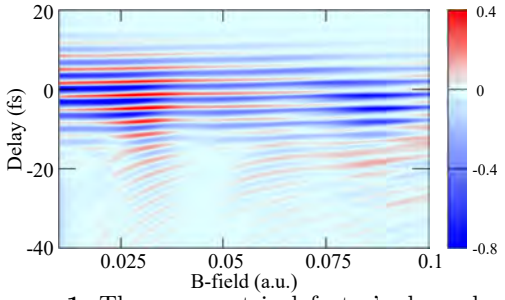


Figure 1. The asymmetrical factor's dependence on the magnetic field strength and time delay.

The development of this method would benefit for many significant advancements. For instance, the manipulation of chemical reactions, which is heavily influenced by the spatial distribution of electron orbitals, could be achieved. Additionally, modification of the magnetic properties of atoms and molecules, which are determined by the orbital magnetic quantum number, could be realized. Furthermore, the invention of a novel type of laser emitting circularly polarized light could be accomplished, as the selection rule for this type of laser requires the modification of the orbital-magnetic quantum number.

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Chiral Optical Tweezers – efficient enantioseparation of molecules

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Synopsis We present an efficient, all-electric dipole, approach to trap chiral molecules in an enantioselective fashion. Our theory leverages a form of synthetic chiral light – tricolour chiral field – which induces a chiral Stark shift, trapping the chosen enantiomer in the laser beam, while the other one is shaken away. We optimise the parameters of the field to maximise the chiral Stark shift and to minimise the achiral one, based on the specific molecule. We propose an experimentally realisable configuration aiming at enantiopure separation of small molecules.

Optical tweezers have proven to be an extremely powerful and versatile tool for trapping and manipulating matter from atoms to micron-scale objects. One long-lasting dream, which has yet to be realised, is the enantiosensitive trapping of chiral objects in an optical tweezer: an optical trap which will keep, say, the right-handed version of a molecule, but will reject (and, indeed, eject) its left-handed mirror version, providing an optical solution to an essential task of the chemical and pharmaceutical industries over the past 200 years.

A chiral optical trap, however, has remained elusive to standard methods, as linear chiroptical phenomena rely on magnetic-dipole and electric-quadrupole interactions, and these are dominated by the much stronger electric-dipole contribution.

Here we leverage recent developments in chiral nonlinear optics, direct brainchildren of attosecond and ultrafast science, which permit pure electric-dipole interactions in the ultrafast regime, built around synthetic chiral light (SCL) [1] as the main tool: a multicolour light field, which couples to a multi-photon chiral molecular pseudoscalar [2], that exhibits its chirality over time. We construct a tightly-focused tricolour chiral (TRICC) beam, designed to be globally chiral [3]. We demonstrate how this leads to the highly efficient enantioseparation of small molecules using only electric-dipole transitions that give rise to a nonlinear (three-photon) chiral Stark shift.

This chiral Stark shift, the product of a molecular and a light-field pseudoscalar, which is tuned through the relative phases and frequencies of the TRICC field, permits the enantiosensitive optical trapping of a specific enantiomer – i.e., a "Chiral Optical Tweezer" – in the same way that the the linear achiral Stark shift enables standard optical tweezers.

We analyse the nonlinear susceptibility of the molecule of choice to optimise the TRICC-beam frequencies, and we show that results in a realistic trap in the presence of a cold buffer gas.

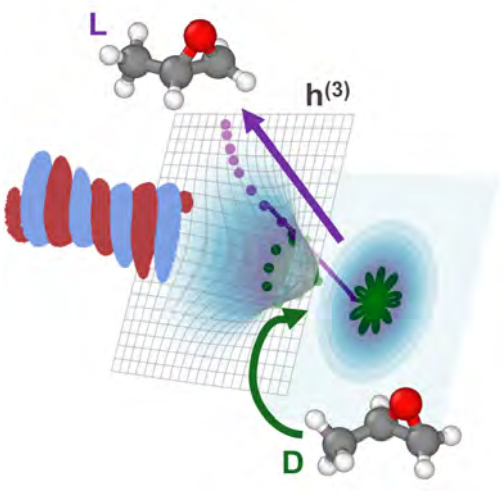


Figure 1. Chiral optical trap created by a tightly-focussed TRICC beam simultaneously trapping the D enantiomer and discarding the L enantiomer shown by their corresponding trajectories.

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Time Delays in Photodetachment of Atomic and Molecular Anions at Low Energies

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Synopsis In contrast to photoionization, time delays from photodetachment of anions are not obscured by the Coulomb interaction at low energies and can be dramatically affected by low-lying shape resonances. Here, we present kinetic energy streaking calculations that address the measurement problems with extracting the time delays at low photoelectron energies.

Photoionization time delays have attracted a significant amount of theoretical and experimental attention in the last decade since the delays contain information about the electronic structure and dynamics of the atomic and molecular systems. However, in the case of photoionization of neutral species, the Coulomb interaction between the outgoing photoelectron and residual cation dominates the dynamics and obscures the electron correlation and polarization effects at low energies (below 5 eV) [1, 2].

In the case of the photodetachment of anions, the outgoing photoelectron interacts with a neutral atom or molecule and the correlation and polarization dominate the dynamics at low energies. Due to the high polarizability of lithium, the time delay goes to 1.7 fs and 0.5 fs at the threshold for the valence and core photodetachment of lithium anion, respectively. The dynamics is even more interesting in the case of molecular anions that have low-lying shape resonances. Recently, we have studied the core photodetachment of C_2^- and CN^-, where we calculated large time delays (up to 4 fs) with dramatic asymmetries in the molecular frame [2].

To our knowledge there have been almost no experimental results in the case of photodetachment due to the challenges connected with the preparation of the anions and measuring time delays at low photoelectron energies. In this theoretical contribution, we address the measurement problems associated with the low-energy region to stimulate the experimental effort. We will discuss our preliminary results of time-dependent photodetachment calculations using the R-matrix method [3] adopting the pump-probe scheme of the conventional and angular kinetic energy streaking [4, 5] with realistic XUV and IR pulses.

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Ultrafast atomic physics between the perturbative and strong-field regimes.

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Synopsis We present *ab initio* ATAS calculations of Neon autoionising states, with laser intensities that are neither in the perturbative nor strong-field regime. We show that these results require models that solve the full Schrödinger equation and explore the mechanisms behind them.

To allow the use of theoretical models to aid our understanding, ultrafast atomic physics has often focused on laser-atom interactions that are in one of two regimes. In the perturbative regime the lasers have low intensities and the laser-atom system can be modeled accurately by perturbation theory, whereas in the strong-field regime the lasers have high intensities and the strong-field approximation can be applied. However, there is a plethora of interesting phenomena that take place between these regimes that has yet to be extensively studied. The *R-Matrix with Time-Dependence* (RMT) [1] code avoids the need for these approximations and can allow us to explore these mid-intensity phenomena.

RMT is able to solve the full multi-electron Schrödinger equation by using different numerical techniques to solve for the regions close to and far from the atom, allowing for efficient solutions and massive parallelism. RMT has been successfully used in the past to reproduce and explain results in both the perturbative [2] and strong-field [3] regimes. More recently, however, RMT has been used to show that results of RABBITT experiments previously thought to have been conducted in the perturbative regime can't be adequately explained by perturbative models [4].

Another area where these approximations do not hold is in truncated autoionisation. If an autoionisation process is stopped by an intense laser pulse, the resonance in absorption does not show the expected Fano lineshape (see figure). At high intensities this is well described by a strong-field model [5], but at lower intensities this model can't reproduce the observed lineshapes. We will present calculations showing the truncated autoionisation process at lower intensities, and show how RMT can be used to understand the mechanisms behind it.

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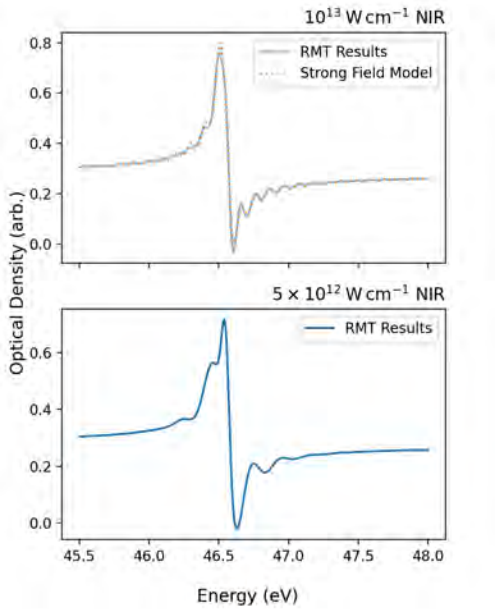


Figure 1. Absorption spectrum of the Neon $2s2p^63p\ ^1P^o$ autoionising resonance, truncated after 40fs by a near-infrared laser pulse. At high intensities the effect of the truncating pulse can be modeled with strong-field assumptions, but at lower intensities these assumptions break down.

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Multidimensional spectroscopy protocols with attosecond pulses

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Synopsis There is a growing interest in measuring the density matrix of photoelectron wavepackets. In multidimensional spectroscopy, additional dimensions of a same variable are added by means of the Fourier transform of a signal along a time delay between pulse pairs. Judiciously chosen pulse shapes can be used to measure the coherence of a wavepacket along a second dimension. In this work, we present a multidimensional protocol that can faithfully reconstruct the density matrix in simple systems. We discuss its limitations and outlook on more complex systems.

There are many ways in which decoherence can appear in atomic and molecular systems. It can arise from the partial measurement of entangled degrees of freedom or from interaction with a stochastic environment. The presence of these processes makes it essential to measure the density matrix of photoelectron wavepackets to fully understand their quantum state.

Such methods must measure the coherence between states of a wavepacket in addition to their populations. One approach is to do an interferometric detection of pairs of states, although this requires scanning over all pairs of states and adding the result [1, 2]. Another approach is through multidimensional spectroscopy, where an additional dimension introduced using time delays between pulse pairs is used to encode the coherence between all pairs of states in a single time delay scan.

In this work, we present a multidimensional protocol using one extreme ultraviolet, one broadband infrared and one narrowband infrared pulse to reconstruct the density matrix. Simulations on helium (Figure 1) and argon show excellent fidelities for the reconstruction [3]. Such methods pave the way for characterizing photoionization in increasingly complex systems.

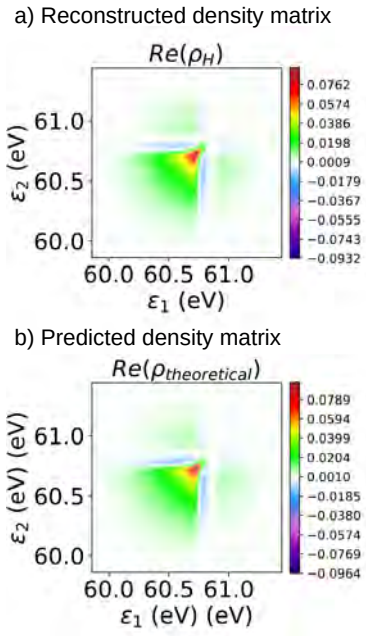


Figure 1. a) Reconstructed and b) predicted density matrix for He (only real part is shown).

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Enhancing Chiral Recognition in Gas-Phase Molecules Using Chiral Topological Light

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Synopsis Chiral discrimination in gas-phase molecules is limited by the low sensitivity of traditional techniques. We address this challenge by generating chiral topological light via a circularly polarized 800 nm, 30 fs Gaussian beam that passes through a q-plate to acquire orbital angular momentum (OAM) and form an optical vortex, which is then modified by displacing its singularity or introducing a slit. Preliminary COLTRIMS measurements suggest potential for improved enantiosensitive responses.

Chiral discrimination in gas-phase molecules remains a significant challenge due to the inherent limitations of traditional techniques. Photoelectron circular dichroism methods achieve only percent-scale contrast, while high-contrast Coulomb explosion techniques [1] are difficult to extend to molecules with more than a few atoms. Recent theoretical advancements suggest that structured and synthetic chiral light can enhance chiral discrimination [2-5]; however, these approaches have yet to be experimentally verified in the gas phase. This work aims to address this gap by employing structured chiral light—a novel form of light designed to achieve robust enantiosensitive measurements in gas-phase chiral molecules.

In this work, we generate chiral topological light by passing a circularly polarized 800 nm, 30 fs Gaussian beam through a q-plate with topological charge $\boldsymbol{q}$ imparting orbital angular momentum (OAM) $\ell=\pm 2q$ and creating a phase singularity with a central null intensity region. The handedness of the incident polarization determines the wavefront's helical rotation.

We further modified the singularity in two distinct ways—(i) by introducing a slit and (ii) by displacing the singularity—to break the cylindrical symmetry and modify the phase topology. This approach enhanced the beam's longitudinal components and allowed precise control over its phase and intensity distribution. The resulting light then interacted with gas-phase chiral molecules, and we measured the resulting photoelectron angular distributions (PADs) using Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS), which enables precise reconstruction of momentum vectors of charged fragments.

This approach embeds topological properties into the enantiosensitive optical response, thereby enhancing both the efficiency and robustness of chiral discrimination. By measuring photoelectron angular distributions (PADs), we assess the enantiosensitive response induced by topological chiral light in chiral molecules such as propylene oxide and achiral gases such as Argon.

Preliminary measurements indicate that both the introduction of a slit and the controlled displacement of the vortex singularity can modulate the chiral asymmetries observed in the PADs. These findings underscore the promise of our approach in enhancing the sensitivity and robustness of enantiosensitive measurements. By embedding topological modifications into the optical field, our method provides improved control over light–matter interactions, paving the way for advancements in chiral spectroscopy, molecular physics, and ultrafast laser-driven enantioselective studies for gas phase molecules.

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Chiral topological light for detection of robust enantiosensitive observables

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Synopsis We create chiral topological light – light that is chiral locally and is described globally by a chiral topological charge – by combining polychromatic tightly focused structured light beams. This enables the ultrafast, highly-sensitive, detection of percent-level enantiomeric excess in chiral molecular mixtures that is robust against common experimental noise.

In synthetic chiral light [1] (SCL) the chirality of the electric field is encoded in its Lissajous curve over one optical cycle, leading to strong enantiosensitive signals and enabling ultrafast and highly sensitive detection of molecular chirality. We combine the strong chirality of SCL with the topological properties of structured light fields, creating chiral topological light (CTL) – light that displays chirality locally with topological global properties. Specifically, we consider $\omega - 2\omega$ beams carrying spin (σ) and orbital (ℓ) angular momentum in a collinear setup with tight focusing, inducing strong longitudinal field components. Within the Rayleigh range of the beams, the transverse spatial distribution of chirality is described by an azimuthal phase that is quantized in terms of a topological charge $C = 2(\sigma_\omega + \ell_\omega) - (\sigma_{2\omega} + \ell_{2\omega})$, offering high tunability via control of SAM and OAM. The topological structure is directly imprinted onto the high-harmonic generation (HHG) response of an ensemble of randomly oriented chiral molecules, leading to an enantiosensitive C-fold azimuthal intensity modulation of the HHG response that can be detected in the spatially resolved far-field signal, enabling the detection of few-percent level of enantiomeric excess. Our simulations show that the chiral signals are robust against intensity fluctuations and imperfect SAM/OAM content of the driving beams thanks to their topological properties. We further extend the concept of CTL to vector-vortex beams, which offer the advantage of higher degrees of local chirality when compared to scalar vortex beams, while retaining the topological properties of CTL.

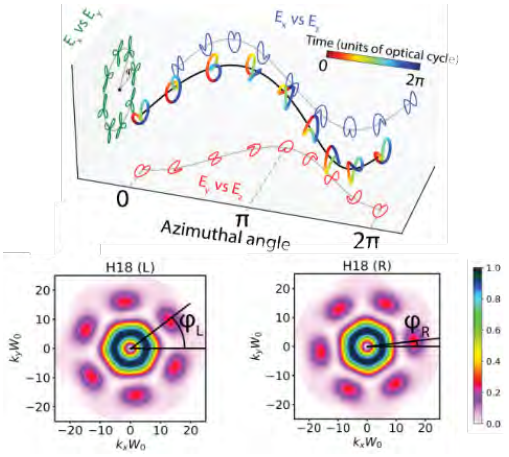


Figure 1. Evolution of the chiral Lissajous curve of CTL at the focus along the azimuthal angle θ for $C = 6$ (top). Far-field harmonic spatial intensity profile for left (bottom left) and right (bottom right) handed fenchone.

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Complete retrieval of photoelectron dynamics from partially-coherent measurements in entangled photoemission

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Synopsis We propose a way to access complete photoemission dynamics articulated around a time-energy analysis of the electron's final reduced density matrix, in situations where its energy is entangled to unresolved degrees of freedom. We illustrate this method with simulations on a model molecule displaying a significant entanglement of the photoelectron energy and the ion's vibration. We assess its validity by confronting the reconstructed dynamics to the simulated ones. Our work demonstrates the possibility to fully access decoherence dynamics in matter at the attosecond time scale.

Attosecond spectroscopies rely to a large extent on interferometric measurements in the spectral domain, combined to time-domain treatments based on first principles of quantum wavemechanics [1]. In this context, any cause of decoherence in the pump/probe sources, in the investigated processes or in the detection stages, constitutes at first glance a severe limitation jeopardizing the time domain access by interferometry.

Here, we address photoemission processes where decoherence results from (i) the entanglement of the photoelectron energy ε with other degrees of freedom and (ii) a partial characterization of the final photoelectron/photoion products. Considering that only ε is resolved, a comprehensive description of the final quantum state is given by the asymptotic reduced density matrix (RDM) of the photoelectron, $\rho(\varepsilon,\varepsilon')$, which recently became a measurable quantity [2, 3].

Assuming that $\rho(\varepsilon,\varepsilon')$ is available, the time domain can be recovered by adapting to its eigenmodes an *ad hoc* Fourier analysis initially developed for coherent spectral amplitudes [4]. This gives access to the complete photoelectron dynamics by reconstructing the time evolution of the RDM all through the process, from the photoelectron birth to its asymptotic detection, regardless of the unresolved degrees of freedom [5].

We illustrate our general approach with numerical simulations on a model diatomic molecule ionized by broadband XUV pulses designed to induce significant entanglement between the photoelectron energy and the photoion vibrational state [6]. We first verified that $\rho(\varepsilon,\varepsilon')$ is measurable by simulating a mixed-FROG [2] experiment, and assessed the validity of our time-domain analysis by confronting the reconstructed dynamics to the numerically exact ones.

With this work, we expand attosecond time-resolved interferometry to partially coherent measurements, and demonstrate the possibility to fully access entanglement dynamics in matter at a most fundamental level.

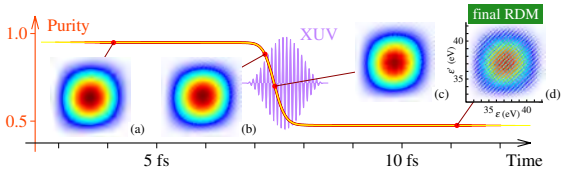


Figure 1. Photoelectron dynamics in entangled molecular ionization triggered by a double XUV pulse (figure centered on the second pulse). The time-dependent RDM, here shown at 3 illustrative times (a-c), is reconstructed out of the final RDM (d). It gives access, e.g., to the purity evolution (orange curve).

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Photoelectron momentum distributions using crossed linearly polarized attosecond pulses

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Synopsis We plan to study the dynamics of photoionisation with attosecond light fields of different polarisation states. To characterize the photoelectron momentum states by their orbital angular momenta, we developed simulations based on the strong field approximation together with retrieval methods using a basis of spherical harmonics.

The three-dimensional momentum distribution of photoelectrons is determined by conservation laws embodied in the selection rules. Different polarisation states result in different distributions of the photoelectron momentum, and by using multiple light pulses with multiple polarizations, quantum interference of photoelectron wave packets should be observed, even, if the light fields themselves might not show interference [1, 2].

Simulations based on the strong field approximation using light fields of different polarization states are developed to study photoelectron wavepacket interferences. More specifically, the pulse configuration, encompassing the number of pulses, their polarization, carrier-to-envelope phase, and the delay between them, can be specified, whereupon the photoelectron momentum distribution is found from the transition amplitude. An example of the results is shown in Figure 1. The key factors influencing the interferences are given by the amplitude of the electric fields, the number of pulses and their relative delay and -phase. These simulations clarify the relationship between the light field configuration and the photoelectron momentum, and show how and under which conditions quantum interference appears.

To characterize the three-dimensional angular distributions of the photoelectrons arising from complicated light fields, we project the distributions onto a basis of spherical harmonics and retrieve orbital angular momenta quantum numbers. This is done using Gaunt coefficients, which linearises the product of two spherical harmonics [3], and can thus be used for expressing the coherent sum of spherical harmonics in an orthogonal basis. This approach is robust to determine both the amplitude and phase of the ionization when only a few angular momenta are involved. With this tool, we expect to better be able to extract information about field configurations from the photoelectron momentum distributions.

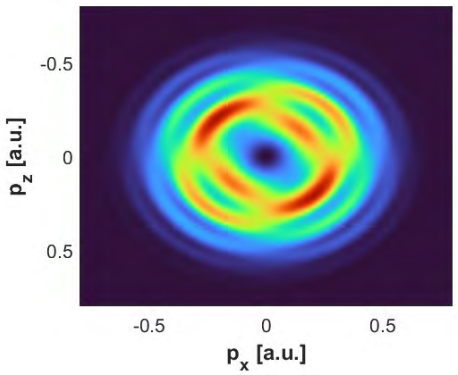


Figure 1. An example of the two-dimensional projection of the photoelectron momentum distribution following from two cross-polarised light pulses.

Finally, we plan to present preliminary results of experiments, where the light fields are arising from High Harmonic Generation in gas [4, 5]. This is expected to enable a new configuration for time-slit experiments in the attosecond time-domain [6], giving the prospect of probing decoherence in this regime.

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Ultrafast photoionization dynamics in the vicinity of CO autoionzing states

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Synopsis We employed the ASTRA suite of codes to study the carbon monoxide $^1\Sigma^+$ autoionizing states using two complementary observables: the attosecond transient absorption and the photoelectron spectra. We reconstruct the time-resolved Auger decay of selected autoionizing states and determine the channel-resolved photoemission delays for resonances above the $B\,^2\Sigma^+$ threshold.

Attosecond science allows us to resolve ultrafast photoelectron emission processes using complementary optical and photoelectronic techniques such as attosecond transient absorption spectroscopy (ATAS) [1, 2] and reconstruction of attosecond beating from interference of two-photon transitions (RABBITT) [3, 4]. In this work we reconstruct the Auger decay of autoionizing states (AIS) in the CO molecule, alongside their position, width, and branching ratios, from the spectra simulated for both the ATAS and RABBITT spectroscopies.

We use ASTRA [5], a new implementation of the multi-channel close-coupling method for molecular ionization, to study the carbon monoxide $^1\Sigma^+$ AIS series up to $D\,^2\Pi$ ionization threshold. For the AIS above the $B\,^2\Sigma^+$ cationic state, we study the resonant contribution to the photoemission delay to the available ionization channels. Since the molecule does not have inversion symmetry, using linearly polarized light along the internuclear axis allows us to observe the interference between $2n$- and $(2n+1)$-photon ionization amplitudes even in optical spectroscopy [6]. To study isolated AIS below the $A\,^2\Pi$ threshold, therefore, we use a pump-probe excitation scheme to induce the interference between a direct one-photon ionization amplitude to a structureless continuum and a two-photon transition amplitude. The pump pulse excites the autoionizing state, whereas the probe pulse records the resonant transition phase and amplitude into a neighboring structureless portion of the spectrum. These measurements allow us to both reconstruct the time-resolved decay of the resonance and extract its original position and width.

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The combination of optical and photoelectron methods enables a comprehensive characterization of AIS, including their branching ratios and channel-resolved photoemission delays. The resonant parameters extracted from the simulated attosecond pump-probe spectra are compared with reference values computed by diagonalizing the molecular field-free Hamiltonian in the presence of a complex absorbing potential (CAP), finding excellent agreement.

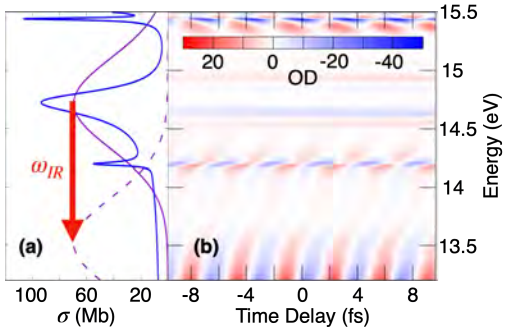


Figure 1. **(a)** Pump-probe scheme and cross section. **(b)** Corresponding ATAS.

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Time-resolved Auger decay of an O_2 intruder-state.

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Synopsis We simulate the attosecond transient absorption spectrum (ATAS) of the oxygen molecule to reconstruct the Auger decay of a bright intruder-state below the $a\ ^4\Pi_u$ threshold using ASTRA, a new molecular ionization code. To do so, we measure the interference between a direct one-photon ionization amplitude to a structureless continuum, serving as holographic reference, and a three-photon transition amplitude in which the target resonance is an essential intermediate state.

Many relevant physical processes ranging from photochemistry in planetary atmospheres to radiation damage in biological systems [1] and dissociative recombination of molecular ions [2], occur on the attosecond time scale. Extreme ultraviolet (XUV) attosecond pulses enabled by advances in laser technology [3] allow real-time observation of such excitations.

Simulating Feschbach autoionizing states (AIS), which play an important role in molecular ionization processes, requires wave-function-based *ab initio* methods. To that end, we use ASTRA, a new implementation of the close-coupling method designed to describe multi-channel molecular ionization processes and suited for studying ultrafast electronic dynamics [4]. We identified a set of $^3\Pi_u$ intruder-states converging to the $A\,^2\Pi_u$ oxygen threshold and designed a pump-probe excitation scheme that allowed us to reconstruct the resonant profile of the AIS with $A\,^2\Pi_u \otimes 4s$ dominant configuration. The pulses were tailored to observe the interference between a single-photon ionization amplitude to a structureless portion of the continuum and a three-photon transition amplitude whereby the resonant absorption of an XUV photon to the $A\,^2\Pi_u \otimes 4s$ AIS is followed by the stimulated emission of two IR photons. We employed the Breit-Wigner resonant-amplitude model to extract the AIS parameters, under the assumption that the phase of the non-resonant one-photon ionization amplitude does not depend on the XUV-photon energy [5]. The parameters of the resonant profile in the ATAS spectrum compare well with those obtained through direct diagonalization of the Hamiltonian with complex absorbers.

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These results demonstrate the possibility of reconstructing the Auger decay of an intruder-state using optical attosecond pump-probe spectroscopy. We also studied more excited autoionizing state series above the $a\ ^4\Pi_u$ threshold, for which we reported branching ratios and channel-resolved photoelectron angular distributions.

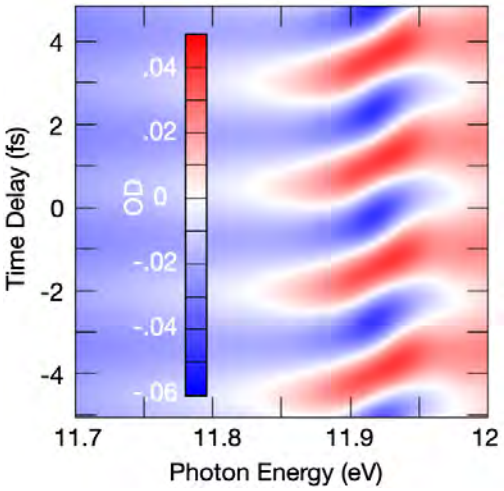


Figure 1. ATAS beating due to the interference between one-photon and three-photon ionization amplitudes, highlighting the phase and amplitude of the excitation profile of the $A\,^2\Pi_u \otimes 4s$ resonance.

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Probing attosecond dynamics of chiral bound wave packets with an attoclock

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Synopsis We use the attoclock with a few-cycle circularly polarized light to image the temporal evolution of atomic chiral bound wave packets on the attosecond time scale. Using the time-to-angle relation of the attoclock, we obtained time-resolved PECD spectrum, which reflects ultrafast changes in atomic chirality. From time-resolved PECD, we reconstruct the evolution of the chiral bound wave packet. In the reconstructed results, the chirality flip of the atomic chiral bound wave packet is clearly visible.

The coherent superposition of two current-carrying electronic states can form a chiral state in an atom [1]. We study strong-field ionization of such a chiral electronic state in a few-cycle carrier-envelope-phase-stabilized circularly polarized laser field with an attoclock configuration by numerically solving three-dimensional time-dependent Schrödinger equation. We find that the photoelectron momentum along the laser propagation direction reveals a high sensitivity to the photoelectron emission angle in the laser polarization plane. The time-to-angle mapping relation of the attoclock allows us to obtain time-resolved photoelectron circular dichroism spectrum on the attosecond timescale. We reconstruct the temporal evolution of the chiral bound wave packet from the time-resolved photoelectron circular dichroism spectrum, which shows the phenomenon of the electronic chirality flips occurring on the attosecond timescale.

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Extended RPAE method solves the Argon delay-puzzle

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Synopsis We present an extended RPAE-type method for photoionization cross sections and delays and show that correlation beyond RPAE has a profound effect on the latter in the Ar $3s$ Cooper-minimum region. A matrix formulation of the RPAE equations is used, and selected correlations effects beyond traditional RPAE are systematically included and investigated.

The photoionization delay in neon is an attosecond physics success story. The experiment by Isinger *et al* [1] was able to isolate the pure $2s$ signal from that of nearby shake-up satellites, and several theoretical descriptions could then reproduce the delay of electrons ionized from that orbital, as measured relative to those from the $2p$ one. Although argon is just a slightly more complicated system, its photoionization delay has proven to be a much harder problem. In the vicinity of the $3s$ Cooper minimum even the sign of the delay differs between different approximations. As discussed in Ref. [2], the difficulty to discriminate against shake-up channels at the $3s$ minimum has also for quite some time prevented a firm conclusion from experiments. Very recently, however, new experiments have been able to give a definite answer, showing that the $3s-3p$ delay difference is indeed negative [3].

The method *random phase approximation with exchange*, (RPAE), was originally developed for photoionization cross-section calculations, but can be extended to describe delays as measured in laser-assisted photoionization experiments [4, 5], such as RABBIT. RPAE accounts for channel mixing and ground-state correlation. It is manifestly gauge independent, and is known to produce good overall cross sections, while still being computationally rather inexpensive. Still lacking is higher order correlation, such as the admixture of two-hole-two-particle excitations. This prevents the description of shake-up satellites, and limits the obtainable accuracy of resonance parameters.

To investigate the role of higher order correlation we have developed a matrix formulation of RPAE where selected two-hole-two-particle contributions can be added. While the cross section around the Ar $3s$ Cooper minimum is only slightly affected, the effect on the delay is dramatic, i.e. the phase of the outgoing wave packet is strongly modified, although its amplitude remains more or less the same. A model explaining the physics behind this behaviour will also be presented in the contribution.

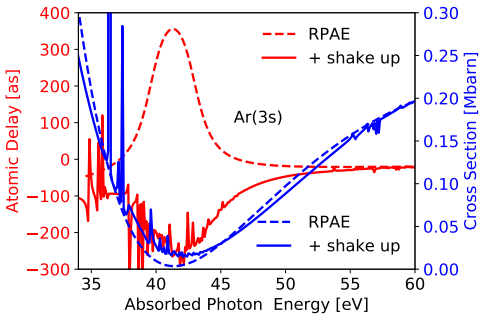


Figure 1. Argon $3s$ in the Cooper-minimum region: cross sections in blue, and atomic delay in red. Dashed curves show the results with RPAE, while solid curves also include selected shake-up configurations. The delay changes dramatically, while the cross section is just slightly affected. The effect on the Wigner delay (not shown) is similar to that on the shown atomic delay.

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Signature of nuclear wavepacket interference in high-order harmonic generation during HCN/HNC isomerization process.

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Synopsis High-order harmonic generation (HHG) from hydrogen cyanide (HCN) during the HCN/HNC isomerization process, pumped by a bending-resonant laser, is investigated. By changing the pump-probe time delay, the interference of two nuclear wavepackets moving oppositely on the molecular potential energy surface is observed in the HHG spectrum. This effect is significant for a critical HHG intensity.

High-order harmonic generation (HHG) is an effective tool for probing molecular structural and dynamic information [1]. Many studies showed the possibility of monitoring molecular dynamics such as vibration and dissociation using HHG spectra. Hydrogen cyanide (HCN) isomerization dynamics has also been investigated [2, 3]. Following this trend, the present study investigates the interference effect of nuclear wave packages in the HHG during the HCN/HNC isomerization.

In our theoretical study, the hydrogen cyanide is pumped by a bending-resonant laser from the lowest vibrational state, leading to the HCN/HNC isomerization. This process is described by the motion of H around the CN's center of mass [3], and the motion is simulated by the evolution of the ground state vibrational state of HCN on the potential energy surface [4]. An intense infrared pulse, a probe pulse, is used to excite the emission of HHG signals to monitor the isomerization process.

Our simulation shows that when the pump gets around its maximum intensity, a portion of the nuclear wave function moves from the HCN well to the HNC well and returns to the HCN for each half cycle. This motion leads to the interference of the nuclear wavepacket from two adjacent half cycles (Fig. 1 (a) and (b)). Remarkably, this effect connects with a significant rise of HHG intensity in and beyond the cutoff region of the molecule without the pump (Fig. 1 (c) and (d)). The project is still ongoing to explain the underlying physics of this interesting effect.

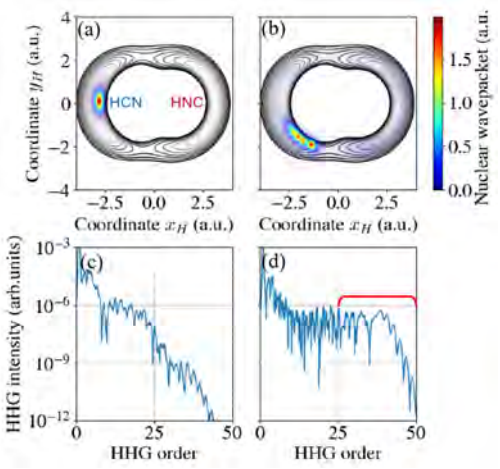


Figure 1. Simulation for the motion of H around CN's center of mass: (a) Lowest vibrational density and (b) nuclear wavepacket when interference happens, after being pumped by a bending-resonant laser. Panels (c) and (d) show the HHG spectra from the molecule corresponding to two situations (a) and (b).

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Quantum path interference in high harmonic generation driven by highly bichromatic fields

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Synopsis We observe a novel quantum path interference (QPI) signature in high harmonic generation (HHG) driven by an orthogonally polarised bichromatic field with comparable intensities. In this highly bichromatic regime, QPI manifests as harmonic intensity modulations over the relative phase of the two colour component. These modulations appear to be more complex than reported ones in the weak second colour regime. Using the strong-field approximation and the saddle-point method, we disentangled quantum orbit contributions to QPI, which inherits the dynamic symmetry of the driving field.

XUV sources based on HHG in the gas phase can be empowered by highly bichromatic field drivers, with significant flexibility coming from the control handles they provide. The second colour field enables a fine control over the electronic wave packet dynamic, i.e breaking of the interaction symmetry and modulating the high harmonic spectral intensity with the relative phase between the two colour components [1], thereby facilitating control over the quantum paths involved [2].

We generated high harmonics in argon using orthogonally polarised highly bichromatic fields ($2\omega/\omega$ intensity ratio at 12%) and observed a new type of QPI. The interference results in distinct intensity modulation patterns for the odd and even harmonics, monomodal and bimodal, respectively, over a scan of the two-colour relative phase (Figure 1). In contrast, previous observations of QPI are typically revealed over an intensity scan of the driver [3]. SFA calculations shows that the bimodal nature of the even harmonic intensity modulations can be attributed to the interference between the y components of the trajectories' dipoles (x being the polarization direction of the ω field). This is explicitly revealed in the spectral domain since the symmetry of the field requires even harmonics to be polarised in the same direction as the 2ω driver. Our results demonstrate this new type of QPI, and give a comprehensive picture of HHG driven by highly bichromatic laser fields.

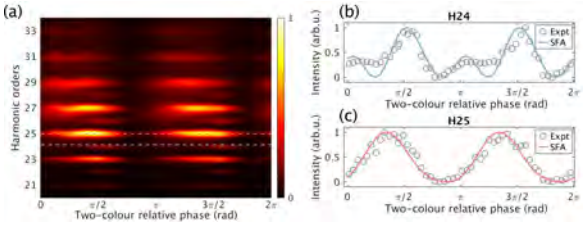


Figure 1. (a) Total high harmonic intensity as a function of the two-colour relative phase; (b) and (c) Lineouts of high harmonic 24 and 25 showing the bimodal and monomodal oscillations(experimental data in circles and SFA simulation in solid lines).

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Decoherence phenomena of electron dynamics in liquid water

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Synopsis We performed RABBIT measurements in both gas and liquid phase water over the photon energy range of 37–74 eV. The analysis of the sidebands oscillation contrasts reveals that RABBIT could address decoherence induced by the solvation environment, providing insights into electron dynamics in liquid water. Indeed, we observed that the contrast in liquid phase water was much lower compared to its gas phase counterpart, with a significantly different behaviour.

The dynamics of electron scattering in liquid samples is of prime importance due to its fundamental implication in a wide range of fields, including radiobiology as well as ultrafast chemistry and physics. It presents stringent experimental challenges as it occurs starting from the attosecond timescale [1] and the effective mean free path of electrons in water is limited to only 8 Å [2]. In order to look into the dephasing dynamics of photoelectrons we performed Reconstruction of Attosecond Beating By Interference of two-photon Transitions (RABBIT) experiments [3] in gas and liquid phase water over the photon range from 37 to 74 eV. The RABBIT technique is based on an interferometric principle and the oscillation contrast can be used as an observable of the decoherence phenomena [4] occurring during liquid water experiments. The experimental setup used for this work combines a liquid micro jet device [5] with a two-foci geometry and two magnetic bottle electron spectrometers enabling us to have a simultaneous atomic reference in the measurement [6].

We measure a striking difference between the contrast in liquid water that increases from 0.2 to 0.3 and that in the neon reference gas that decreases from 0.7 to 0.6 over the investigated spectral range. Decoherence may originate from physical processes such as electron scattering or from measurements biases (e.g. temporal jitter, focusing aberrations) and spectral overlap in complex systems. The influence of the instrumental decoherence is ruled out by the measument in the neon reference in exactly same conditions. The effect of spectral overlap is gauged by measuring the contrast in gaseous water, which shows a monotonic decrease of contrast from 0.7 to 0.6, similarly to the atomic reference. This large difference with liquid water indicates that RABBIT can be used to assess solvation-environment decoherence in liquid jets. Our simulations treat the electron as an open quantum system in interaction with its surrounding will shed light on electron's scattering dynamics in liquid water [7].

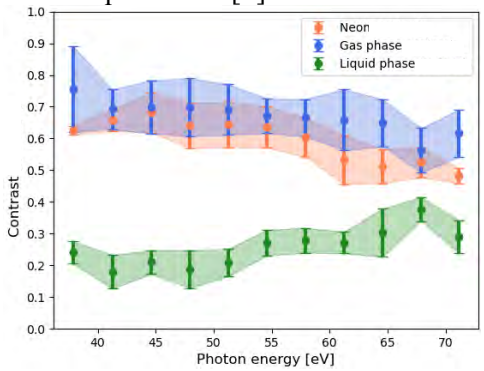


Figure 1. Evolution of the contrast extracted from sidebands oscillations in the atomic reference (orange), gas phase (blue) and liquid phase water (green).

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A general approximator for strong-field ionization rates

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Synopsis We demonstrate that the sub-optical-cycle dynamics of nonlinear photoionization can be retrieved from ionization probabilities obtained for a set of few-cycle pulses. The core of our proposed retrieval procedure is a General Approximator for Strong-Field Ionization Rates (GASFIR), which is non-adiabatic by design and provides accurate rates of multiphoton ionization for gases, semiconductors, and dielectrics.

Whether to generate attosecond pulses via high harmonic generation or to sample electric fields with petahertz bandwidth [1, 2], understanding and accurately modeling strong-field ionization dynamics is important for advancing attosecond technologies. This requires precise knowledge of when and how electrons are freed during a laser pulse—that is, an understanding ionization rates. However, no experimental method has so far demonstrated direct access to multiphoton ionization rates, and they are also not easily accessible in *ab initio* calculations. Quantum mechanics allows for a unique and rigorous definition of an ionization probability only in the absence of a strong electric field. Given an electron's wavefunction during a laser pulse, it is possible to *estimate* which part of the wavefunction populates unbound states, but the energy-time uncertainty is a fundamental obstacle to knowing it exactly.

To circumvent this fundamental problem, we investigate whether a model for predicting ionization rates $\Gamma(t)$ can be retrieved solely from ionization probabilities $\int_{-\infty}^{\infty} \exp[-\Gamma(t)]\, dt$, which are unambiguous quantum-mechanical observables. Because strong-field ionization is inherently non-adiabatic (that is, diabatic) [3], $\Gamma(t)$ depends on the ionizing field within a certain time interval. In general, $\Gamma(t) = \int_{-\infty}^{\infty} K(t,T)\, dT$, where the integral kernel $K(t,T)$ depends on electric field in the time interval between $t-T$ and $t+T$. Without any prior knowledge, $K(t,T)$ cannot be retrieved from ionization probabilities. We show that the general properties of strong-field ionization make such a retrieval possible by restricting the set of viable kernels. We propose a General Approximator for Strong-Field Ionization Rates (GASFIR), which encodes these properties. With a few adjustable parameters, GASFIR describes multiphoton and tunneling ionization not only in atomic gases, but also in solids (we tested it for Si, SiO_2, and diamond).

The detailed and accurate ionization rates provided by GASFIR enable the development of advanced carrier generation strategies tailored to attosecond applications.

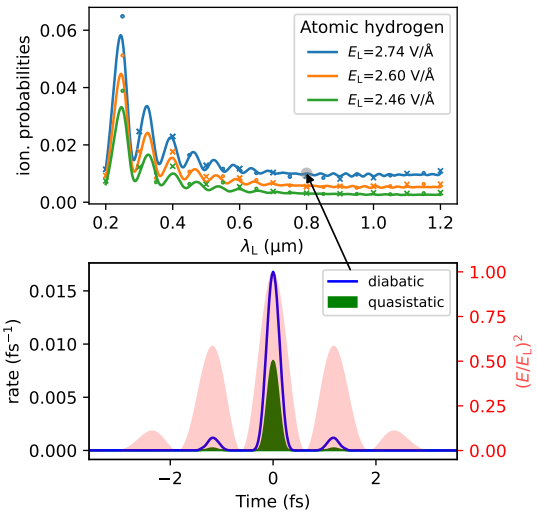


Figure 1. Multiphoton ionization of a hydrogen atom by a single-cycle pulse. Top: GASFIR predictions (solid lines) obtained from the training data (ionization probabilities marked by crosses) match the validation data (dots). Bottom: Comparison of diabatic (blue) and quasistatic (green) ionization rates for $\lambda_L = 0.8$ μm and $E_L = 2.74$ V/Å.

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State- and Phase-Resolved Vibrational Wavepacket Dynamics in N_2 Studied using All-Attosecond Transient Absorption Spectroscopy

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Synopsis Vibrational wavepacket motion in N_2^+ was induced following ionization of N_2 using an attosecond pump pulse. An attosecond probe pulse excited the generated cation to higher-lying bound states and allowed for state- and phase-resolved detection of the evolving vibrational wavepacket.

Attosecond-pump attosecond-probe spectroscopy (APAPS) holds great promise for investigating ultrafast electron dynamics in various materials. We recently demonstrated HHG-based All-Attosecond Transient Absorption Spectroscopy (AATAS) to systematically probe coherent electron wavepacket dynamics in atoms [1].

Here we apply AATAS to study state- and phase-resolved coherent vibrational wavepacket motion in N_2.

The experiments were performed using an 18 m long HHG beamline optimized for high XUV intensity [2]. Cascaded post-compression of the NIR driving pulses ensured high NIR and XUV pulse stability [3], crucial for AATAS. The attosecond pump and probe pulses were selected using a split-and-delay unit and their spectra were individually recorded using an XUV spectrometer.

Fig. 1(a) shows an AATAS map of optical density (OD) changes vs. photon energy and time delay. The Fourier map in Fig. 1(b) exhibits a dominant oscillation peak at ≈ 0.23 eV, corresponding to an oscillation period of 17.80 ± 0.16 fs. This oscillation period can be assigned to vibrational levels of the $A^2\Pi_{ui}$ state of N_2^+ [4]. The narrow-band features in the AATAS map are attributed to the population of several vibrational progressions in higher-lying bound states. Compared to our recent AATAS experiment in O_2 [5], the vibrations are phase-shifted by π. The underlying reasons will be discussed.

Our results demonstrate the feasibility of tabletop AATAS for probing ultrafast molecular dynamics with high temporal resolution. Future AATAS studies will explore coherent electron dynamics and the coupling with nuclear degrees of freedom.

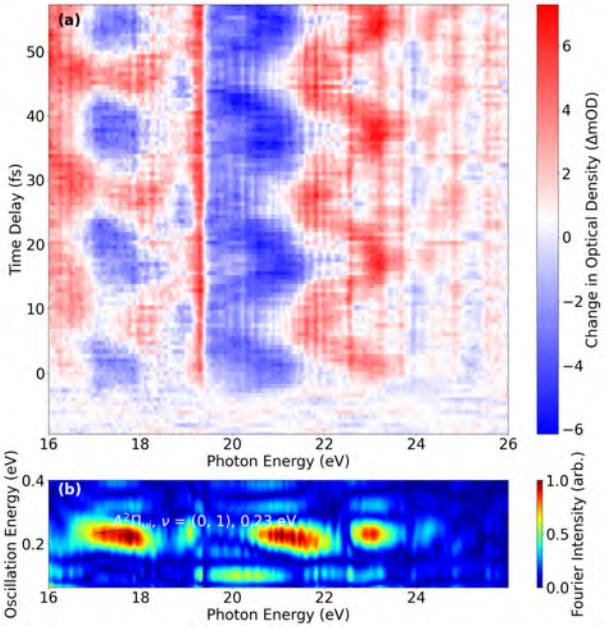


Figure 1. a) AATAS map showing the change in optical density (ΔOD) as a function of the photon energy and the time delay.
b) Corresponding Fourier map. The horizontal line marks the vibrational energy difference between the $A^2\Pi_{ui}, \nu = 0, 1$ states of N_2^+.

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Chiral Measures make Chiral Moments

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Synopsis Chirality is a ubiquitous feature of nature, earning a key place in attoscience through chiral photoelectron spectroscopy and synthetic chiral light, with a remaining need to reliably quantify chirality. We develop a family of stable and versatile chirality measures. Our approach allows for chirality measures to be expressed as a factorised triple product of tensorial moments of a given distribution – capturing the most characteristic pseudoscalar in nature. We demonstrate these chirality measures for the photoelectron distribution resulting from the ionisation of hydrogen by chiral light.

Chirality has solidly taken a key spot within attoscience: starting with photoelectron chiral dichroism (PECD) [1] as an avant-garde atto chiral spectroscopy, and developing into the all-dipole revolution [2], which allows chiral light-matter interactions without relying on magnetic responses, by switching from chirality structured in space to a dynamical chirality structured in time – synthetic chiral light (SCL).

Nevertheless, any spectroscopy is limited by the characterisation methods for the setup and for the observables, setting the demand for solid and reliable ways to quantify chirality. For SCL this has been done with the introduction of nonlinear correlation functions [2], capturing the temporal chirality of multicolour light and well-suited for describing multiphoton processes. For photoelectron spectra (PES), however, this has not yet been done in a consistent manner, with a solid link to the chirality of the driving light.

We develop a fully general approach to treat chiral distributions and introduce a family of universal, solid and reliable chirality measures [3]. Our chiral family is built on tools coming from cosmology [4] and molecular science [5], based on spherical harmonics and multipolar moments with angular momentum ℓ. We show that all such measures $\chi_{\ell_1\ell_2\ell_3}$ can be presented as factorised triple products of tensorial multipolar moments, making them a chiral moment of the given distribution:

$$\chi_{\ell_1\ell_2\ell_3} = \left(\mathbf{M}^{(\ell_1)} \times \mathbf{M}^{(\ell_2)}\right) \bullet \mathbf{M}^{(\ell_3)}. \qquad (1)$$

We showcase these moments in action for resonantly-enhanced multiphoton ionisation (REMPI) of hydrogen driven by a three-colour chiral field (see figure). The distribution has a characteristic spiral shape, easily captured by our chiral moment with minimal order χ_{234}.

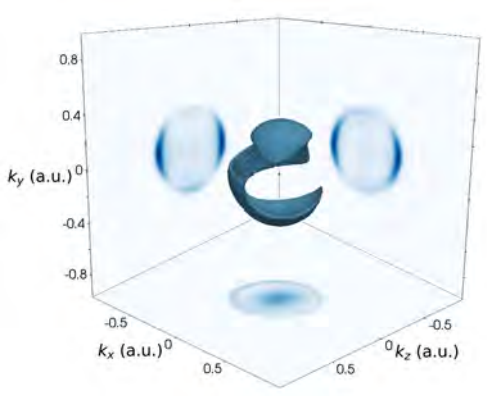


Figure 1. TDSE calculation of PES of hydrogen ionised by SCL composed of both elliptically-polarised ω and 2ω, and perpendicular linearly-polarised 3ω, with the fundamental at 120 nm.

Our chiral moments show a great potential to describe the chirality of various nonlinear-optical processes, from canonical nonlinear optics to the nonperturbative ultrafast one.

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Observing the Jahn-Teller dynamics of the propadiene cation using interferometric strong-field ionization

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Synopsis The ultrafast relaxation dynamics of propadiene (C_3H_4) after strong-field ionization were investigated in an interferometric pump-probe scheme with attosecond resolution. Molecular dynamics on the order of tens of femtoseconds have been observed that might give insight into the interplay between two competing Jahn-Teller (JT) distortion pathways.

It is well known that the Born-Oppenheimer approximation breaks down when nuclear and electronic motion are coupled. This can readily be studied in systems such as valence-excited propadiene. Here, there are two competing JT-active modes for relaxation, connected by a conical intersection. One corresponds to the C=C=C asymmetric stretching mode and the other to the torsional mode [1, 2]. See Figure 1a) and b).

The experimental method uses a few-cycle pump to strong-field ionize the target (4.5 fs, 10^{14} W/cm^2) and a delayed, much weaker pulse (5.3 fs, 10^{11} W/cm^2) to probe the dynamics. The delay-dependent photo-ion spectrum was measured mass-spectrometrically. The waveforms of both pump and probe pulses were reconstructed from an atomic target [3, 4], while for a molecular target the probe interrogates the evolution of the wavepacket through linear excitations on the cationic manifold. This can be revealed in the delay-dependent branching ratios.

The emerging molecular response, seen for the $C_3H_3^+$ ion in Figure 1c), shows two main features. Firstly, fast oscillations, originating from the interferometric superposition of the trailing edge of the pump with the central part of the probe. Secondly, a large change in amplitude, with the first maximum being 10 fs after the arrival of the pump, showing when the wavepacket appears in the region accessible to the probe. The modulation recurs, as parts of the wavepacket return to the probe region.

Time-frequency analysis, shown in Figure 1d), shows that the Fourier spectrum of the molecular signal changes throughout the delay scan. For $C_3H_3^+$, there are two main components observed: a frequency corresponding to 1.85 eV, peaking at 10 fs as well as a slightly less pronounced feature at 1.7 eV, peaking at 72 fs. The latter appears to be created from the merging of two frequency components, separated by 0.3 eV, that are observed between delays of 40 and 60 fs and manifest as a clear beating in the time domain. These modulations have preliminarily been assigned to the energy differences between the potential energy surfaces being coupled. Thus, this method shows applicability to the study of coupled electronic-nuclear dynamics and JT-active systems.

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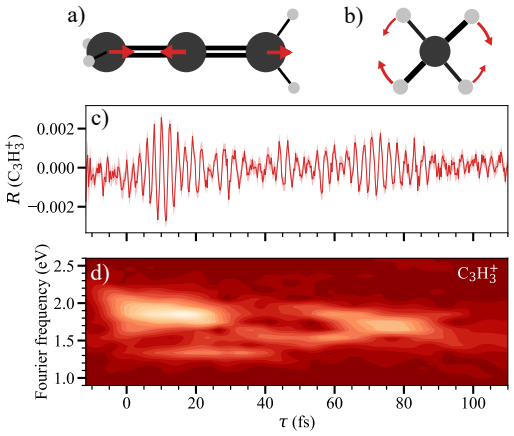


Figure 1. The asymmetric stretching (a) and torsional (b) modes of propadiene, the change in the branching ratio of $C_3H_3^+$ (c) and its time-frequency analysis (d).

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Photon Momentum Transfer and Partitioning: From One to Many

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Synopsis We present a general and consistent framework for photon momentum transfer covering an arbitrary number of absorbed photons. In the multi-photon limit, we show that with each additional photon absorbed above the ionization threshold, the photoelectron acquires on average twice the momentum of the absorbed photon.

The transfer of photon momentum is indispensable in initiating and directing light-matter interactions, which underpins a plethora of fundamental physical processes from laser cooling to laser particle acceleration. The transferred photon momentum is distributed between the photoelectron and the residual ion upon ionization. Our study presents a general and consistent framework for photon momentum transfer covering an arbitrary number of absorbed photons. Our results firstly bridge the gap between the previously considered limiting cases of single-photon and multi-photon strong-field ionization and suggest revising the current consensus for the multi-photon limit by demonstrating that with each additional photon absorbed above the ionization threshold, the photoelectron acquires on average twice the momentum of the absorbed photon (see Figure 1). Our work paves the pathway towards a comprehensive understanding of the fundamental processes of photon momentum transfer in light-matter interactions, with implications for both theoretical physics and practical applications that harness the transfer of photon momentum [1].

References

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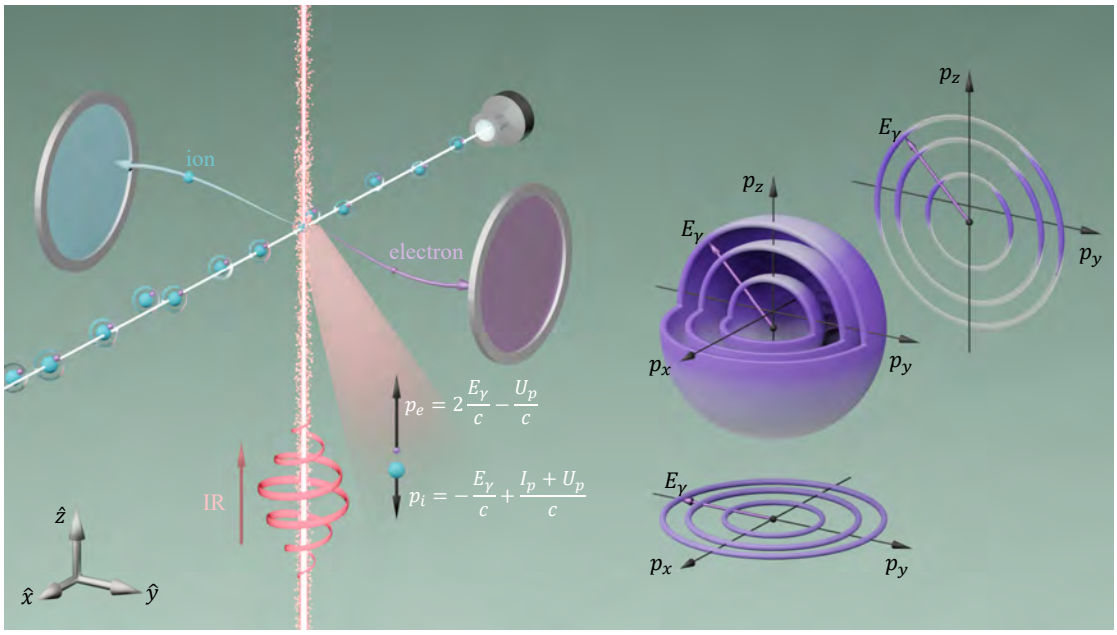


Figure 1. Photon momentum transfer and partitioning

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Subcycle Conservation Law in Strong-Field Ionization

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Synopsis A new conservation law between the angular momentum and energy in strong-field ionization applicable at the subcycle level is established. Based on it, we find that the under-barrier tunneling is driven by the electric field, while the subsequent continuum motion is governed by the vector potential. We further establish a magnetic quantum number m-resolved saddle-point approximation for atomic p orbitals.

Exploiting the infinite-order continuous dynamical rotational symmetry of circularly or elliptically polarized classical light pulses (see Figure 1), we establish the conservation law between the angular momentum L_z and energy E at the tunnel exit in strong-field ionization for circular pulse that is applicable at the subcycle level:

$$E = \omega L_z - I_p, \tag{1}$$

where I_p is the ionization potential and ω is the angular frequency. In the asymptotic region, the conservation law becomes:

$$E = \omega L_z - I_p - U_p, \tag{2}$$

where U_p is the ponderomotive potential. We illustrate the conservation law through the correlated spectrum of angular momentum and energy of photoelectrons, both at the tunnel exit and in the asymptotic region. Moreover, we propose a protocol based

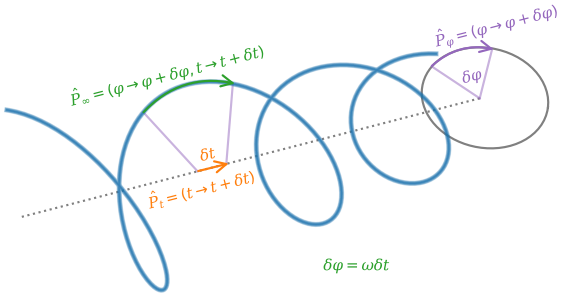


Figure 1. Dynamic symmetry in circular pulse

Light can be described either by its field or potential, which are often interchangeable. We theoretically investigate the distinct roles of the electric field and the vector potential of an ultrashort laser pulse in strong-field tunneling ionization, based on their distinct central frequencies. Employing the conservation law between the angular momentum and energy, we demonstrate that quantum tunneling dynamics in driven by the electric field, while the subsequent classical motion in the continuum is governed by the vector potential [2].

The saddle-point approximation (SPA) within the framework of strong-field approximation is extensively applied in strong-field physics as it offers a clear physical insight into intense light-matter interactions. With the help of the conservation law, we introduce a magnetic quantum number m-resolved saddle-point approximation (m-SPA) to analyze the ionization dynamics of atoms initially in p orbitals (or d orbitals...) subjected an intense laser field. Our results reveal that m influences not only the prefactor of the ionization rate but also the saddle-point equation itself, an effect overlooked in prior studies. The m-resolved saddle-point equation can be expressed as:

$$\frac{1}{2}\left[\boldsymbol{p} + \boldsymbol{A}(t_s)\right]^2 + I_p + m\varepsilon\omega = 0, \tag{3}$$

where ε and ω are the ellipticity and angular frequency of the laser field, respectively. The accuracy of the m-SPA method is validated through comparisons with the backpropagation method and the strong-field approximation, showing its superiority over the conventional SPA approach [3].

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Ultrafast Spin Migration in the Argon Dimer Cation

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Synopsis We introduce the concept of *spin migration*, i.e. time-dependent spin density, and illustrate the concept by *ab initio* calculations of the coupled vibronic motion of the electrons and the nuclei in the argon dimer cation. We find that the spin density varies on multiple time scales, given by the natural time scales of the electronic and nuclear wavepackets.

Upon sudden removal of an electron from a neutral molecule, the remaining electrons do not have time to relax, and a non-stationary electronic wavepacket is formed, resulting in a quasi-periodic oscillatory motion commonly referred to as *charge migration* [1, 2]. In the presence of spin–orbit interaction, the electronic motion must necessarily also involve spatio-temporal dependence of the spin density; correspondingly, we term this phenomenon *spin migration*.

Of vital importance to the electronic coherence, is the correlated motion of the nuclei. Unless there is spatial overlap of the nuclear wavefunctions correlated with different electronic states, coherence between the electronic states does not lead to observable interference patterns [3]. On the other hand, the motion of the nuclear wavepacket, can be also be used to drive the electronic coherences, and hence the spin migration.

As model system, we choose to study the argon dimer cation, where the separation of vibronic states are comparable to the spin–orbit splitting in the argon atoms at infinite separation. Hence, even a simple linear combination of a few vibronic eigenstates can be expected to induce non-trivial spatio–temporal dependence of the spin density, on multiple time scales simultaneously. In Figure 1, we show a nuclear wavepacket consisting of a linear combination of four vibronic eigenstates, associated with two electronic states. The total nuclear wavepacket is also shown for two different times, along with the resulting electronic spin densities. The latter shows an intricate pattern, with many features, varying at different time scales.

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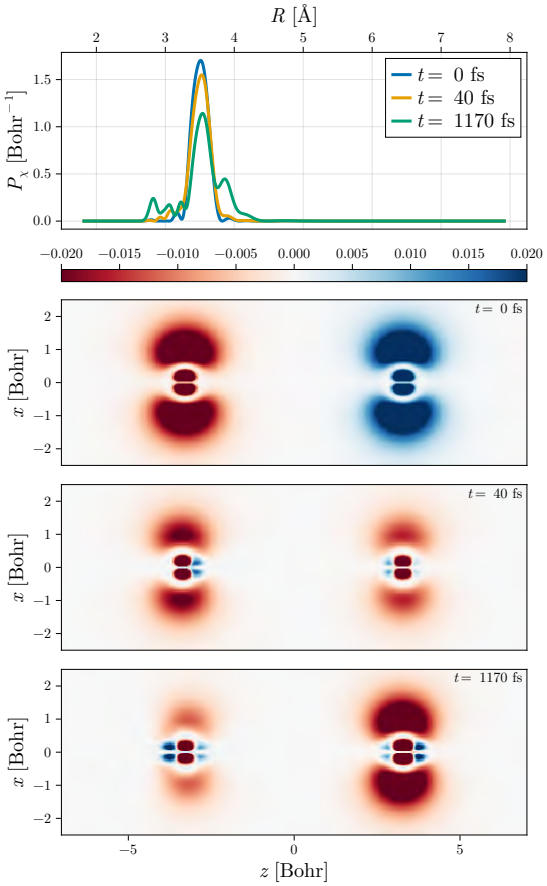


Figure 1. *Top panel*: Time dependent nuclear wavepacket, for three different times; *Lower panels*: corresponding electronic spin density, with a mirror average of the initial distribution subtracted to emphasize changes.

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Attosecond coincidence spectroscopy in small molecules

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Synopsis We present a study of photoionization dynamics in CH_4/CD_4 and CO_2 using attosecond photoelectron interferometry. We investigate nuclear motion effects and field-induced ionization channel coupling using two-color photoionization and coincidence spectroscopy. Additionally, by introducing a complex parametrization of the angle-resolved two-color signal, we effectively describe the angle dependence of photoionization time delays.

Attosecond photoelectron interferometry is commonly utilized to investigate photoionization dynamics in molecular systems by measuring photoelectron spectra generated in a two-color field, consisting of an extreme ultraviolet (XUV) comb and a synchronized infrared (IR) field. These cross-correlation techniques provide valuable insights into electron correlation effects and coupled electronic-nuclear dynamics [1].

In this study, we investigate photoionization dynamics in an isotopic gas mixture of methane CH_4 and deuteromethane CD_4 and in carbon dioxide CO_2, utilizing photoelectron-photoion coincidence spectroscopy. The absorption of an XUV photon from an attosecond pulse train excites a manifold of cationic states, which couple to the evolving photoelectron wave packet. An additional absorption or emission of an IR photon results in a two-color photoionization spectrogram, allowing for detailed tracking of time-resolved ionization dynamics.

A collinear configuration is implemented to control the synchronization between the XUV and IR fields with IR sub-cycle precision. Both fields co-propagate and are focused by a toroidal mirror into the reaction microscope, where they interact with the gas target.

In the case of CH_4/CD_4, attosecond interferometry provides access to the nuclear response of the two isotopologues, since their electronic structures remain largely unaffected by the two-color field. By analyzing the angle-integrated amplitude and contrast of oscillations of the photoelectron peaks, we obtain information regarding the nuclear motion upon photoionization, as well as its effect on the coherent properties of the two-color signal [2]. Additionally, by introducing a complex parametrization of the angle-resolved two-color signal, we describe the oscillatory components and phases, providing insights into the angle dependence of photoionization time delays [3], [4].

Furthermore, we investigate CO_2 photoionization time delays, considering the impact of field-induced coupling between ionization channels [5]. Finally, by measuring ejected electrons in coincidence with O^+ and CO^+ dissociation fragments, we determine the attosecond time delays associated with the different photodissociation channels for randomly oriented molecules.

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Investigating Photoionization Delays Using an Attosecond Source Synchronized with an Infrared Non-Collinear Optical Parametric Amplifier

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Synopsis Photoionization delays play a crucial role in understanding ultrafast electron dynamics. In this work, we are aiming at extending the RABBIT technique using an attosecond pulse train in the extreme ultraviolet generated by an 800-nm driving field and a synchronized pulse delivered by an optical parametric amplifier operating at 1200 nm. Our experimental setup will enable precise phase analysis of the two sidebands formed between photoelectron peaks. This approach is expected to enhance our understanding of photoionization on attosecond timescales.

The precise measurement of photoionization delays on attosecond timescales is critical to understand ultrafast electron dynamics in atoms and molecules. When an atom absorbs a photon, the resulting ionization process is not instantaneous but occurs on the attosecond timescale due to the complex interaction between the emitted electron and the remaining ionic potential. Measuring photoionization delays allows us to probe these interactions in detail, providing insight into how electrons propagate in the continuum after being ionized. One of the most effective techniques for measuring these delays is the Reconstruction of Attosecond Beating by Interference of Two-Photon Transitions (RABBIT) [1]**.** This method relies on the formation of photoelectron sidebands caused by two-photon transitions: an initial ionization event triggered by an attosecond extreme ultraviolet (XUV) pulse, followed by an additional interaction with a phase-locked infrared (IR) pulse. The relative phase shifts between these sidebands encode crucial timing information, related to the phase of harmonics and the atomic phase consisting of Wigner phase and the phase of the continuum-continuum transitions [2].

In this work, we present the development of an experimental setup designed to extend the RABBIT scheme to a multisideband configuration. The setup employs an 800 nm driving laser to generate attosecond XUV pulses through high-order harmonic generation (HHG). These XUV pulses are then overlapped with a synchronized 1200 nm IR pulse generated by a non-collinear optical parametric amplifier (NOPA). The IR pulse interacts with the photoelectrons, leading to the formation of two sidebands between photoelectron peaks caused by the absorption of single XUV photons. In this scheme, we expect to observe two sidebands (instead of a single one) between two consecutive harmonics, thus reproducing the conditions recently achieved in the experiments performed at the free electron laser FERMI [3]. We will discuss the status of the experimental setup, focusing on the development of a two-stage carrier-envelope phase stable NOPA, and we will also present preliminary results about the stabilization scheme we are going to implement to secure sub-cycle synchronization between the attosecond pulse train and the IR field.

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Nonlinear spectroscopic studies using intense Gas High-order Harmonics at the SYLOS COMPACT beam line, EL-ALPS

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Synopsis The SYLOS COMPACT beam line at the ELI-ALPS Research Institute investigates electron dynamics in atoms and molecules on the femtosecond to attosecond time scale. It generates adequate XUV pulse energies (~μJ) at suitable repetition rates (10 Hz, 50 Hz, and 1 kHz) using Gas High-Order Harmonic Generation (GHHG) to trigger non-linear processes in the XUV domain. We introduce the beamline's capabilities as a user facility, highlighting new results from recent user campaigns, with an emphasis on XUV nonlinear ionization of atoms.

Ultrafast spectroscopic studies of light-matter interactions on the attosecond time scale provide essential insights into the evolution of different electronic quantum matter phases. Specifically, XUV radiation produced through the High-order Harmonic Generation in Gases offers excellent intrinsic temporal and spatial coherence properties across various atoms and molecules. Only a limited number of laboratories worldwide are capable of generating the XUV pulse energies required for pump-probe experiments that probe the intrinsic timescale of electronic motion in different phases of matter. This remains a challenge, as pulse energy, repetition rate, and XUV photon energy often involve trade-offs in current HHG technology, which limits the available possibilities in this domain.

In this communication we present the implementation of the recently commissioned Sylos COMPACT GHHG beam line [1] at ELI-ALPS based on loose focusing (~several meters) geometry. At the heart of the beamline is the SYLOS laser (EKSPLA Ltd. [2]) providing more than 2.75 TW with ~12 fs CEP-stable pulses (~850 nm) at 1kHz and SEA lasers delivering 4.5 TW with 12 fs pulses at 10 Hz/50 Hz centered around 850nm. The beam line has the capability to deliver XUV pulses in the μJ range as well as high photon energies reaching up to 150 eV generating both from (multiple) gas jets or from gas cells. Our experiments showcase the attosecond duration of pulses (~340 as) in a few femtosecond pulse train using the RABBITT technique [3]. We discuss the design features of the COMPACT beam line that enable it to handle high average power, improve XUV conversion efficiency through strategic target selection [4], and enable comprehensive XUV pulse diagnostics. We also present recent performance metrics and explore the opportunities for future conducting experiments here. A key highlight is our first results on the two-photon double ionization of neon [5], achieved during the inaugural user campaign. These findings emphasize the ELI compact beam line's ability to capture non-linear phenomena in the XUV spectral range, paving the way for exploration of electronic correlations in quantum phases of matter at relevant time scales.

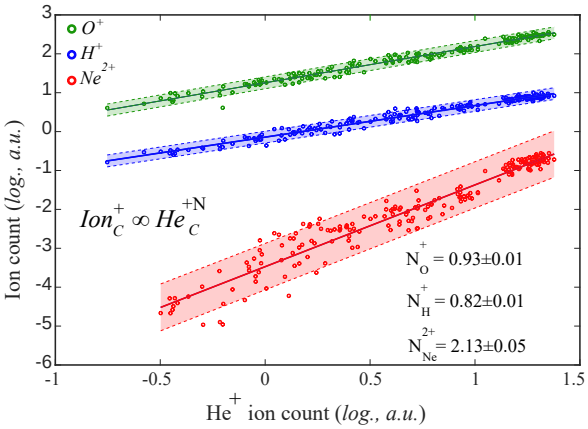


Figure 1. Intensity dependence of various ion signals w.r.t He^+ ion signal as a result of XUV two-photon double ionisation (N is degree of nonlinearity, soild lines are fit to the data).

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Rotational Pseudo-Doppler Effect in Solid-State High Harmonic Generation

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Synopsis Here, we report a rotational pseudo-Doppler effect in high harmonic generation from a non-rotating crystal driven by a waveform-shaped femtosecond laser pulse. The magnitude of the frequency shifts in the high harmonic spectra increases linearly with harmonic order, which is consistent with the prediction of rotational Doppler effect in nonlinear optics [1]. Our work adds an additional component to the precision measurement of the rotational microscopy with ultrahigh sensitivity by probing the electron motion in the high-order nonlinear process.

The traditional Doppler effect was first proposed by C. Doppler in 1842 to describe the frequency shift of a sound wave moving towards or backward an observer. Its counterpart, the rotational Doppler effect, is associated with the angular momentum of waves. The rotational Doppler effect has been extended from rotating macroscopic medium to rotating microscopic molecules [2, 3], with rotational frequencies ranging from Hz to THz.

We highlight that the ultrafast motion of electrons on the attosecond time scale can be excited and steered by a waveform-shaped femtosecond laser pulse, offering an ideal platform to investigate the rotational Doppler effect. High-harmonic spectroscopy, in particular, provides an exceptionally sensitive probe for observing subcycle electron dynamics. We demonstrate a rotational pseudo-Doppler effect in high harmonic generation from a non-rotating crystal, driven by a waveform-shaped femtosecond laser pulse, whose rotational frequency is on the PHz scale. By controlling the waveform of the driving laser pulse, the frequency shifts exhibit red or blue shifts. Our work demonstrates a novel rotational pseudo-Doppler effect in high-order nonlinear processes, provides insight into the fundamental mechanisms of the attosecond rotational Doppler effect in high harmonic generation, and inspires potential applications for precision measurements.

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Quantum beating and cyclic structures in the phase-space dynamics of the Kramers-Henneberger atom

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Synopsis We investigate the phase-space dynamics of the Kramers-Henneberger (KH) atom solving the time-dependent Schrödinger equation for reduced-dimensionality models and using Wigner quasiprobability distributions in the KH frame for a short-range potential. We find that, for both the time-averaged and the full time dependent potential, coherent superpositions of eigenstate trigger a cyclic motion in phase space. This is similar to the cyclic behavior present for a molecule. A key difference is the lack of spatial confinement in the KH atom.

Light-induced potentials have been realized in many research areas, such as Bose-Einstein condensates, quantum simulations, and conical intersections [1]. In strong-field physics, a quintessential example is the KH potential. The KH potential is a light-induced dichotomous potential that has been extensively studied in the context of stabilization. Stabilization means that, in specific intensity and frequency regimes, ionization will be suppressed. The electronic wave packet is trapped in this dichotomous potential, whose shape has often been linked to that of a molecule.

Population trapping also happens in a molecule and can be identified by the cyclic motion observed in phase space [2]. Our studies show that the same cyclic behavior occurs for the KH atom if the stabilization regime has been reached. This is the case for the time-averaged KH potential and even if the full time dependent dynamics of the KH atom are taken into consideration. However, there are time delays for the full potential, and some tail-shaped spilling of the quasiprobability flow towards higher momentum regions. These tails are signatures of ionization, indicating that, for the potential studied, a small momentum spread is associated with stabilization. A comparison of the quasiprobability flow with classical phase-space constraints shows that, for the KH atom, the momentum must be bounded from above. This is a major difference from a molecule, for which the quasiprobability flow is confined in position space for small internuclear separation [3]. Furthermore, we study the stability of the KH atom for different initial conditions and time propagation.

Our studies present a new viewpoint for understanding the formation of the KH atom and its coherent dynamics. An interesting question is whether the level of suppression of ionization and localization that we have observed is good enough for a theoretical model of a qubit. This will form the basis of future investigations.

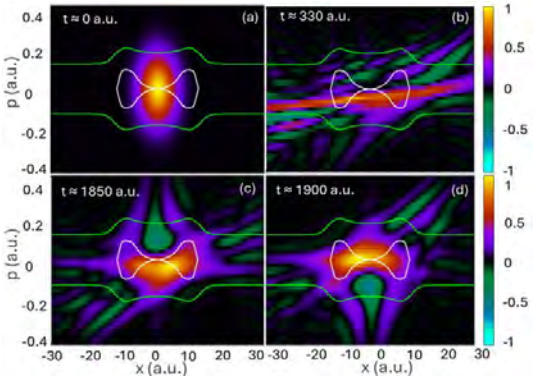


Figure 1. Wigner distribution for a short-range potential in a field of intensity $I = 5.7 \times 10^{13}$W/cm^2 and frequency $\omega = 0.0628$ a.u. [3], using the atomic ground state as the initial condition for the full expression of the potential. (a) shows the Wigner distribution at t=0 a.u. (b) shows the Wigner function at t = 330 a.u. when stabilization has not yet set in and (c) and (d) show the cyclic behavior of the trapped wave packet at t = 1850 a.u. and t = 1900 a.u., respectively, which is after the onset of stabilization.

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Transient extreme ultraviolet scatterometry for nanostructure morphology reconstruction and probing material dynamics

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Synopsis We use 0th order scatterometry to reconstruct the morphology of periodic Si nanostructures that resemble semiconductor metrology targets. The excellent match between experimental data and simulations allows us to quantitatively reconstruct the nanomorphology parameters of the target, namely groove height and critical dimension. In the future we will use this technique to elucidate the ultrafast dynamics both of bulk and atomically thin nanostructured materials.

Future integrated-circuit design will require functional metrology that may combine nanostructure retrieval with measuring carrier dynamics between materials that are characteristic of electronic functionality [1].

An optical method for reconstructing the surface morphology of a periodic structure with accuracies below the diffraction limit is 0th order scatterometry, where the reflectivity of a structured surface is experimentally recorded and then compared to simulations [2]. In our case, we use High Harmonics generated in Ne as our Extreme UltraViolet (XUV) source driven by 800 nm, 35 fs pulses. The XUV beam illuminates the samples at a 78.3° angle of incidence (AOI).

Our targets consist of Si gratings with (111) crystal orientation, where we studied a set of samples with varying critical dimension (CD) and groove height. All of the gratings were created in pairs, with perpendicular line geometries, as for the case where the lines are perpendicular to the beam propagation plain (Lamellar diffraction), the diffraction efficiency is no longer dependent on the CD and therefore decorrelates this parameter from the reflectivity of the other. The reflectivity is then compared to simulations based on Rigorous Coupled Waveform Analysis.

As seen in Fig. 1, the simulation and experimental data are in good agreement with minor discrepancies, which we currently link to a thin oxide layer on top of the Si samples. The absolute values of the parameters have been verified by a Scanning Electron Microscope.

So far we have measured targets with groove heights that span from 24 to 140 nm and CDs from 300 to 700 nm in the 40 – 120 eV photon energies. The ultimate goal is to perform femto- and attosecond resolved transient scatterometry near the Si L_{23} absorption edge with the aim of studying the evolution of electron-hole dynamics linked to the nanostructure of the material. The static measurements on nanoscale gratings have revealed sensitivity to (sub-)nm changes in CD or groove-height via shifts of spectral minima. This gives an exciting promise for ultrafast sensing of small sub-nm lattice expansions.

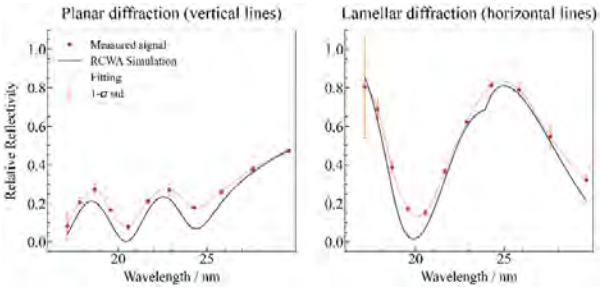


Figure 1. Experimental and simulated relative reflectivity of a Si grating with 300 nm CD and 122 nm groove height.

A benefit of the technique is that atomically thin to bulk materials can be studied by changing parameters like the AOI and probe beam spectral range. Recording the ultrafast optical response of layered and flat surfaces (like atomically thin dichalcogenides deposited on Si or SiO_2) is an intriguing field of investigation, as those may be integrated into future atomically-thin integrated circuits.

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Attosecond Ionic Photoionization Spectroscopy

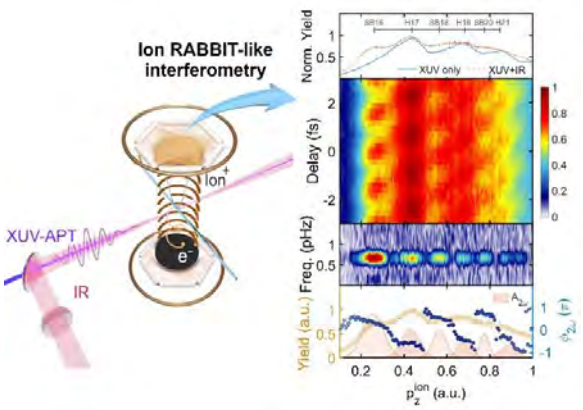
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Synopsis We experimentally illustrate the attosecond ion reconstruction of attosecond beating by interference of two-photon transition (RABBIT-like) interferometry through the development of high-resolution ion momentum detection in atomic photoionization processes. Our ion interferometry presents identical momentum- and time-dependent scattering phase shift, as we observed in photoelectron spectroscopy, which, thus, is proved to be a possible alternative attosecond approach to resolve the photoionization process, without the electron homogeneity limitation.

As one of the most commonly used methods to determine attosecond ionization time delays, the reconstruction of attosecond beating by interference of two-photon transitions (RABBIT) has been widely applied to investigate the photoionization process at the attosecond time scale [1-3]. Since the existing studies directly focus on the main actor in the ionization process, i.e., the photoelectron, these measurements are based on the direct recording of photoelectron momenta or kinetic energy. However, it is equally imperative to investigate the ions, which also manifest periodic distributions in response to pump–probe time delays as a manifestation of momentum conservation during the half-scattering process. Formal investigations suggest that, in existing implementations of attosecond electron–ion coincidence measurements, the detected ions are expected to be more effectively utilized, especially via the powerful attosecond chronoscopy.

In this work, by taking the advanced attosecond ion–electron spectroscopy combined with the XUV-APT and IR laser field, we report the energy- and time-resolved ion emission phase shifts in atomic helium. The ionic momentum distributions exhibit periodic oscillations with the pump–probe delay, allowing us to extract time information encoded in interfering wave packet phases. The ion momentum-resolved phase shift recapitulates the properties of photoelectron measurement, as well as the resolution. Our measurement demonstrates the feasibility of ion attosecond interferometry and effectively simplifies the experimental conditions by subtracting the previously necessary electron–ion coincidence measurement [4].



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Interplay of locally chiral rotational and electronic currents in chiral molecules

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Synopsis We report the effects of chiral molecular rotations on photoionization and photoexcitation dynamics in chiral molecules. First, we show that one can create chiral rotational trajectories even in achiral spherical top molecules. Second, we explore the control of ultrafast enantio-sensitive electronic response in rotating chiral molecules, leading to new, efficient enantio-sensitive observables, such as enantio-sensitive transient absorption and photoelectron circular dichroism of chiral rotors.

Chiral molecules are crucial in pharmaceuticals, as enantiomers (mirror-image forms of the same molecule) can interact differently with biological systems. Chirality also influences materials science and agrochemicals, affecting properties like reactivity, taste, and toxicity.

Differentiating between enantiomers is a complex challenge that drives the study of enantio-sensitive effects in light-matter interactions. A new frontier is nonlinear methods based on the electric dipole interaction, which could enhance chiroptical sensitivity by orders of magnitude and introduce concepts like local chirality [1]. Though chirality is typically a property of spatially extended object (that depends on the mutual orientations of its parts), local chirality arises from a vector tracing a chiral trajectory in time. For instance, locally chiral Lissajous figures of light can generate strong enantio-sensitive signals in HHG [2].

Here, we examine the interplay of two locally chiral structures: the induced polarization vector of chiral electron currents and the rotational trajectory of a molecule, both driven by locally chiral fields. We formally investigate how controlled molecular rotation affects ultrafast photoionization and photoexcitation. In a pump-probe setup, the pump pulse excites the rotational motion of a spherical top with chiral symmetry, generating a chiral rotational wave packet. This motion can be visualized as the molecular axis tracing a chiral trajectory, or more precisely, a 3D Lissajous curve in space, drawing an analogy to synthetic chiral light over one full rotation period. The locally chiral probe pulse then induces ionization or excitation, with the time delay between the pump and probe pulses serving as one of the parameters controlling the interplay of two locally chiral objects . We then analyze the enantio-sensitivity of the time-dependent ionization or absorption yield as a function of the pump-probe delay, incorporating rotational resolution to identify signatures of the interferences between two chiral motions evolving on vastly different time-scales. This analysis has been conducted for both an achiral molecule and a chiral molecule using chiral hydrogenic electronic states [3] as a model of chiral electronic density.[†]

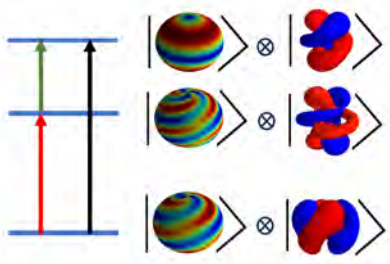


Figure 1. Excitation scheme induced by the probe pulse, acting on molecular state as product of chiral rotational wavepacket (left ket in the product) and chiral electronic state (right ket in the product).

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Ultrafast TACOS

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Synopsis We introduce TACOS – Terahertz-Assisted Chiro-Optical Spectroscopy – for ultrafast and highly efficient chiral recognition. TACOS relies on driving ultrafast nonlinear electronic dynamics in chiral molecules using terahertz and few-cycle optical pulses, without requiring optical CEP stability.

Chiral molecules exist in pairs of enantiomers: non-superimposable mirror images that behave identically unless they interact with another chiral object. Enantio-discrimination is vital, but also challenging. Traditional spectroscopy relies on weak magnetic interactions and yields tiny enantiosensitivity (<0.1%). One can overcome this limitation with locally chiral light, which can drive ultrafast and highly enantiosensitive chiral electronic dynamics via purely electric-dipole interactions. Such light can be created in a multi-color noncollinear setup. However, ensuring that the field's handedness is maintained globally in space requires exquisite control over the two-colour phase delays [1]. This is not a requirement for chiral microwave fields [2] thanks to their long wavelengths. However, microwaves are not well suited for ultrafast electronic or vibronic dynamics.

Can we combine both approaches to get the "best of both worlds"? That is, can we drive enantiosensitive electron dynamics without needing to worry about the spatially dependent phase delays associated with optical fields?

We provide positive answer to this question by introducing ultrafast TACOS – Terahertz-Assisted Chiro-Optical Spectroscopy. TACOS combines a long-wavelength THz field with an ultrashort optical pulse, see Fig. 1a. The linear response of the molecules to the THz field induces a transient orientation of the electronic density, breaking the isotropy of the randomly oriented ensemble. This allows us to probe the molecular handedness with an ultrashort linearly polarized pulse.

TACOS relies the on interference between an achiral 3-photon and a chiral 4-photon pathways, see Fig. 1b, which can spectrally overlap thanks to the low-frequency of the THz radiation and the broad bandwidth of the optical pulse. As a result, the polarization of the emitted light encodes the medium's handedness with high enantiosensitivity.

TACOS can be implemented using available optical instrumentation and it does not require working in vacuum. Furthermore, because both multiphoton pathways involve absorption of 3 optical photons, optical CEP stability is not needed. This simplifies the experimental realization of our proposal, creating a new route for ultrafast and highly enantiosensitive imaging and manipulation of molecular chirality on ultrafast timescales.

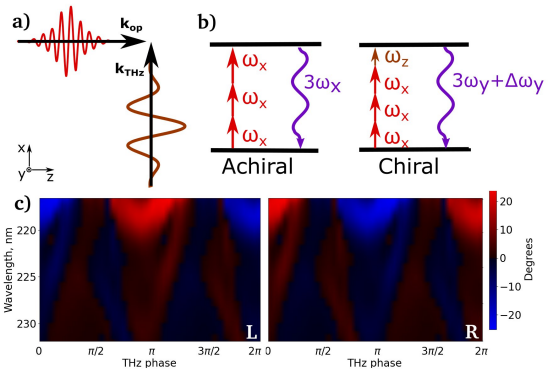


Figure 1. a) Cross-propagating optical (λ_{opt}=730nm) and THz (λ_{THz}=80μm) pulses. b) Relevant multi-photon pathways. c) Enantiosensitive polarization angle of the radiation emitted by randomly oriented carvone as a function of the output wavelength and the CEP of the THz pulse.

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Strong-field theory for attosecond scanning tunneling microscopy

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Synopsis Coherent electron dynamics in molecules and nanostructures can be resolved with Ångström-attosecond precision by integrating scanning tunneling microscopy (STM) with few-cycle femtosecond laser pulses. However, a robust strong-field theory with a time-dependent perspective has remained elusive. Here, we present a comprehensive model for attosecond STM, valid across all regimes, revealing a striking analogy to the standard STM model. Additionally, we reveal how the intuitive three-step model and energy cutoff apply to attosecond STM.

Attosecond science provides access to the intrinsic time scale of electronic dynamics in atoms and molecules. Capturing these ultrafast processes demands innovative approaches that combine attosecond temporal resolution with sub-nanometer spatial precision. Conventional scanning tunneling microscopy (STM), which relies on DC tunneling, is constrained by electronic limitations. To overcome this, recent advances propose integrating STM with few-cycle femtosecond laser pulses to achieve attosecond time resolution [1, 2]. Using this technique, STM paves the way for simultaneous attosecond-nanometer imaging of electron dynamics.

However, despite significant technological advances, a comprehensive strong-field theory that describes all photoemission regimes in attosecond STM remains elusive. A classical electron with the Fermi velocity of gold can traverse a 1 nm junction in just 754 as, a timescale comparable to the optical cycle of near-infrared light (2.8 fs for 830 nm). The interplay between traversal time, driving duration, and strong-field photoemission highlights the necessity of a time-dependent strong-field theory.

Here, we introduce our theoretical work on attosecond STM. Unlike the commonly used strong-field approximation (SFA) model, our approach explicitly incorporates both the Coulomb binding potential and the image-charge potential. The theory is directly derived from the time-dependent Schrödinger equation (TDSE) in an ultrathin nanojunction, without approximations, ensuring that key quantum properties are naturally retained. We have formulated a time-dependent extension of Bardeen's tunneling matrix, which is the standard theory of conventional STM. By deriving the saddle-point equations, we systematically classify the various types of photoemission. We identify a novel energy cutoff law that applies across all regimes (Fig. 1(a)). Furthermore, we find a novel three-step model (Fig. 1(b)), where the electron is emitted into the gap between the metals (step 1), propagates in the laser field (step 2) and then enters the substrate (step 3) [2].

In the STM junction, a distinct photoemission process can occur, where electrons absorb only a few photons and tunnel through the junction with sub-threshold energies. To characterize this from strong-field ionization, we introduce a new parameter, ζ. We show that strong-field photoemission occurs only when $\zeta < 1$.

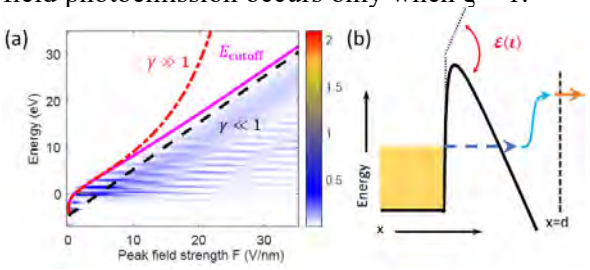


Figure 1. (a) Comparison of the tunneling spectrum from TDSE (color mapping) with the theoretical cutoff. Approximations in different regimes are also plotted. (b) The three-step model for laser-assisted STM: Step 1: adiabatic tunneling, Step 2: field-driven acceleration, and Step 3: transmission.

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Ultrafast and topological chiral sensing with vector beams

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Synopsis We show how to realize a nonlinear interferometer for chiroptical sensing by tilting light's polarization in a vector beam. Our approach relies on purely electric-dipole interactions, thus enabling higher chiral sensitivity. The topological properties of our tilted vector beam are imprinted into the ultrafast optical response of randomly oriented chiral molecules, leading to a separation of the macroscopic emission from left- and right-handed molecules into two distinct rings, creating exciting opportunities for ultrafast, all-optical, efficient and topologically protected chiral observables.

Attosecond science unlocks exciting possibilities for *ultrafast* and *efficient* chiroptical sensing and manipulation. The key to *efficient* is to rely solely on purely electric-dipole interactions, rather than on the interplay with magnetic effects [1]. Along this route, one can create *locally* chiral light, where the electric-field vector draws a chiral Lissajous figure in time [2]. Endowing locally chiral light with topological properties enables *robust* chiroptical measures [3]. But, is local chirality a necessary requirement?

We have recently shown [4] that tight focusing can turn an elliptically polarized wave into an efficient chiroptical tool. The strong longitudinal field upon tight focusing tilts the polarization plane towards the propagation direction, in a way that it is no longer orthogonal to it. As a result, the nonlinear response of chiral molecules becomes strongly sensitive to its handedness. Importantly, this effect arises within the electric-dipole approximation and it does not require locally chiral light.

Here we show how to realize a nonlinear interferometer for efficient chiroptical sensing via tilting light's polarization in an elliptically-polarized vector beam. Since locally, at each point in space, our field is elliptically polarized, it is *not* locally chiral. The topological nature of the vector beam rotates the polarization ellipse as we move azimuthally in the transverse plane, in a way that the major polarization component is oriented along the radial direction, see Fig. 1a. The intensity gradients across the radial direction lead to strong longitudinal components that tilt the field polarization in a topological manner, already at moderate tight focusing conditions.

Our numerical simulations and analytical modeling reveal that the topological properties of our vector field lead to a radial separation of the nonlinear optical response of randomly oriented chiral molecules. That is, the left-handed molecules can be forced to emit harmonic light as a ring with smaller radius, whereas the right-handed molecules emit a ring with bigger radius. Which of the two enantiomers emits where remains under our complete control.

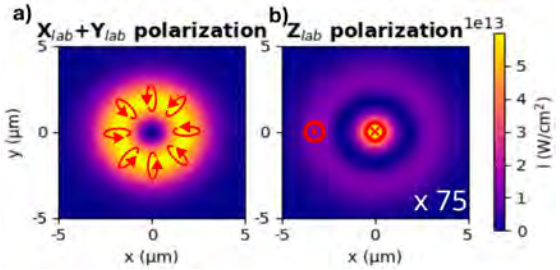


Figure 1. a,b) Intensity of a tightly-focused elliptically-polarized vector beam in the laboratory frame. The red arrows correspond to a schematic representation of the elliptical beam polarization.

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Theoretical study of photoemission time delay for two-photon transition via Multiple Scattering approach

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Synopsis We derived an expression for two-photon transition from valence orbitals using XUV attosecond pulsed lasers to study the angular dependence of photoemission time delay for molecules. Our theory is based on Multiple Scattering theory, which divides the system into regions having the center at the center of gravity of the potentials in most cases, using Green's function. We applied our theory to diatomic molecules and discussed the angular dependence with the geometrical coordination for the incident light.

Two-photon transitions from orbitals using XUV attosecond pulsed lasers dressed by IR femtosecond pulsed lasers can be utilized to measure photoemission time delay, leading to interesting ultrafast dynamics. Theoretical calculations for the continuum-continuum (cc) delay for those measurements are done by atomic calculations. [1] In our previous studies [2, 3], we theoretically confirmed that photoemission time delay from core states of hetero and homo nuclear molecules exhibit strong anisotropy about angle distributions. Therefore more rigorous treatment for cc transition is desired. In this study, we employ Multiple Scattering approach for the continuum states to describe cc-transitions. [4]

The two-photon transitions amplitude is expressed as

$$M_{2\mathrm{ph}}(\mathbf{k}, E_\kappa) = \langle \psi^{(-)}_{\mathbf{k}}(\mathbf{r}')| \, \Delta_{\mathrm{IR}} \; G^{(+)}(E_\kappa, \mathbf{r}', \mathbf{r}) \; \Delta_{\mathrm{XUV}} \, |\phi(\mathbf{r})\rangle \, ,$$

where $\psi^{(-)}_{\mathbf{k}}$ is continuum state of photoelectrons with incoming boundary condition with momentum vector $\mathbf{k}$, Δ_{IR} and Δ_{XUV} are light operators for the XUV and dressing IR fields, respectively. $G^{(+)}(E_\kappa, \mathbf{r}', \mathbf{r})$ is Green's function within outgoing boundary condition with intermediate state energy E_κ, ϕ is initial molecular orbitals. In the Multiple Scattering theory, we divide a targeting system such as molecules into atomic sites (Ω_{I}), interstitial region (Ω_{II}) and outside of the outer sphere (Ω_{III}). (Fig.1) The Green's function is expanded by the local solutions with the origin of each center and connected at the boundaries by the so-called multiple scattering matrix. [4] In Ω_{III} the cc-transition can be exactly estimated by Coulomb wave functions. [5]

In the presentation, we will discuss the details of the theory and calculations, including its application to diatomic molecules.

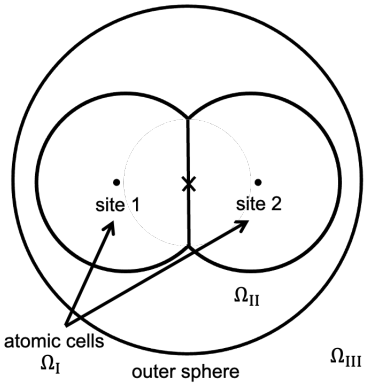


Figure 1. Partitioning of the space of the target molecule for Multiple Scattering approach. Atomic sites (Ω_{I}), interstitial region (Ω_{II}) and outside of the outer sphere (Ω_{III}).

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Ultrafast dissociation dynamics of alkyl iodides induced by few-fs UV pulses

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Synopsis We studied dissociation dynamics and photoprotection schemes in neutral alkyl iodides photoexcited by few-femtosecond UV pulses. In CH_3I, the experimental results have been used to benchmark a theoretical model that provides insights into the nonadiabatic nature of the relaxation dynamics, including the wavepacket's arrival time at the conical intersection. In C_2H_5I, UV–XUV pump-probe measurements capture fragmentation patterns of only a few femtoseconds.

Alkyl iodides serve as key model systems for studying ultrafast nonadiabatic dynamics. UV radiation excites the A-band, with an absorption spectrum centered ~ 260 nm [1], leading to a rapid C–I bond cleavage. This neutral fragmentation is intrinsically governed by a conical intersection (CI) [2]. However, the limited temporal resolution of previous experiments has hindered direct observation of the early relaxation dynamics in the absence of the UV excitation pulse [3]. Here, we present measurements using few-femtosecond (fs) UV pulses [4,5] with a duration corresponding to <7% only of the vibrational period in methyl and ethyl iodide (CH_3I and C_2H_5I).

In CH_3I, the dynamics is probed by few-cycle near-infrared (NIR) pulses through multiphoton ionization. Trajectory surface hopping calculations are performed to identify the dissociation channels involved, relating to the number of absorbed NIR photons. Fig. 1 shows the comparison between the experimental ion signals and the fit based on the calculated contributions from different ionization channels. The time resolution achieved with 4-fs UV pulses allowed us to benchmark our theoretical modelling, which can precisely determine the arrival to the CI of the neutral system. Furthermore, the results demonstrate that NIR ionization within a 5-fs window after UV excitation can prevent the otherwise inevitable cleavage of the C–I bond [6].

We also present UV pump–extreme ultraviolet (XUV) probe measurements of ethyl iodide [7]. Fitting of the time-dependent yields of the fragment ions ($C_2H_2^+$ and $C_2H_5^+$) reveals a relative time delay of 6.7 fs between these two fragments, indicating the presence of a few-fs relaxation channel following the UV-photoexcitation.

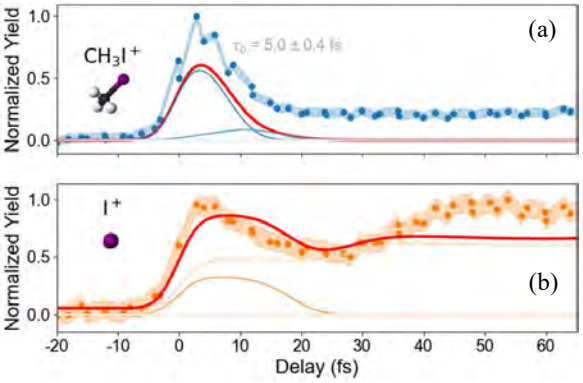


Figure 1. The experimental time-dependent yield of the parent (a) and iodine (b) ions. The solid lines represent (a) 3- and 4-photon channels and (b) 5- and 6-photon channels calculated with the theory model. The red line shows the fit obtained with the best linear combination of these contributions. Adapted from [6].

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Reconstruction of the ionization and recombination times from the spectral phase of the emitted HHG radiation.

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Synopsis In this work, we show that the ionization and recombination times in high-harmonic generation (HHG) can be determined from the measurement of the spectral phase of attosecond pulse trains generated by a two-color driving field ($\omega + 2\omega$). By performing a comprehensive analysis of the spectral phase as the temporal profile of the driving field is varied, we have unraveled a specific feature for the phase that carries information about the ionization times, allowing for the retrieval of the tunneling time, the born time, and the recombination time.

The recent development of extreme ultraviolet light sources with attosecond duration has opened up new possibilities for observing and controlling quantum dynamics in matter down to the natural timescale of electron motion. Among the various forms of attosecond pulses currently available in the laboratory, attosecond pulse trains (APTs) made of odd and even harmonics have proved to be very efficient in resolving and manipulating quantum orbits in the HHG process, for example [1, 2, 3].

Such pulses are generally synthesized with a two-color femtosecond driving field composed of a fundamental field and its second harmonic. The literature is rich with experimental and theoretical studies reporting the dependence of both the spectrum and the polarization of the resulting APTs on the temporal profile of the femtosecond driving field [4, 5].On the contrary, the dependence of the APTs' spectral phases on the driving field waveform shape still remains elusive, though.

In this work, we have performed a comprehensive analysis of the spectral phases of APTs generated by intense, collinear, two-color femtosecond fields ($\omega + 2\omega$) as the temporal profile of the HHG driving field is varied. We have found that the relative spectral phases between consecutive odd and even harmonics strongly depend on both the ratio r and relative phase ϕ between the two colors of the driving field. Intriguingly, a maximum for the relative spectral phase is observed at a given angle $\phi \sim 50°$ for the whole range of ratios r considered in this study (0.05 - 5%). Within some approximations, we show that the times associated to the ionization and recombination steps in high harmonic generation (namely the tunneling, the born, and the recombination times), can be retrieved from this specific feature. Our method is supported by quantum-orbit calculations demonstrating the feasibilty of the retrieval procedure.

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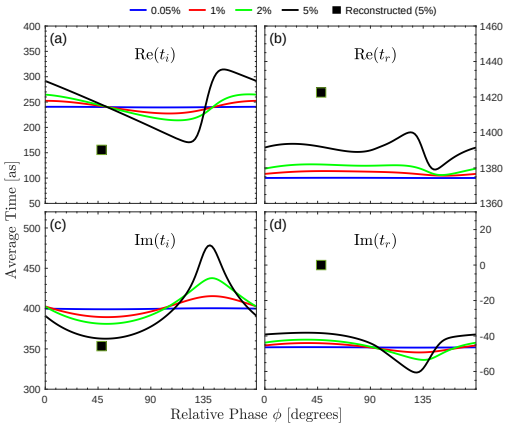


Figure 1. Real (Re) and imaginary (Im) parts of the ionization (t_i) and recombination (t_r) times in high harmonic generation (solid lines: QO theory calculations for various ratio r between the two color of the driving field, square dot: reconstructed from the spectral phase of the HHG radiation).

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Spin-Orbit Delays in Argon: Tracking Ionization Delays Across an Autoionizing Rydberg Resonance

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Synopsis We report high-resolution RABBIT measurements with a 400 nm driver in argon, tracking ionization delays between the spin-orbit components of Ar^+ across the autoionizing $3s^1 3p^6 5p^1$ Rydberg resonance. Using a self-built second-harmonic bandwidth compressor, we resolve the small spin-orbit splitting while scanning photon energies across the resonance. Our results reveal a narrow resonance-induced peak in the delays.

Previous studies on ionization delays between the $^2P_{1/2}$ and $^2P_{3/2}$ spin-orbit components of the first ionization threshold in noble gases using the reconstruction of attosecond beating by interference of two-photon transitions (RABBIT) technique have focused on krypton and xenon, due to their relatively large spin-orbit splittings. One study [1] reported spin-orbit delays in xenon of up to 33 as at a photon energy of 33.4 eV, which was attributed to autoionizing Rydberg resonances. Despite its fundamental interest, spin-orbit delays in argon have remained unexplored due to its small spin-orbit splitting of 0.18 eV [2], which is challenging to resolve with conventional RABBIT schemes. We present a detailed study of spin-orbit delays in argon, tracking them across the autoionizing $3s^1 3p^6 5p^1$ Rydberg resonance at 28.0 eV [2].

To overcome the challenge of scanning photon energies across the resonance while simultaneously resolving the small spin-orbit splitting, we employed a self-built second-harmonic bandwidth compressor (SHBC) [3]. This specialized sum-frequency generation (SFG) scheme generates tunable narrowband 400 nm second-harmonic light with a bandwidth of 0.92 nm from the output of a Ti:sapphire amplifier. This narrowband light drives high-harmonic generation (HHG), yielding extreme ultraviolet (XUV) radiation with an approximate bandwidth of 20 meV. The tunability of the SHBC allows us to scan the ninth harmonic photon energies between 27.4 eV and 28.2 eV, covering the 28.0 eV Rydberg resonance. We use harmonics 7 and 9 in combination with the narrowband second-harmonic light to generate sideband 8 and extract the ionization delays between the $^2P_{1/2}$ and $^2P_{3/2}$ spin-orbit components.

As shown in Fig. 1, the extracted delays exhibit a distinct peak of approx. 10 as at the resonance, decreasing rapidly to near 0 as away from it. This strongly supports the claim that the resonance is the cause of the non-zero spin-orbit delays. Additionally, an asymmetry in the extracted delays towards lower energies from the singlet resonance at 28.0 eV suggests contributions from the lower-lying triplet resonance. This singlet-triplet mixing is attributed to spin-orbit coupling, necessitating advanced theoretical models for an accurate description of the spin-orbit delays.

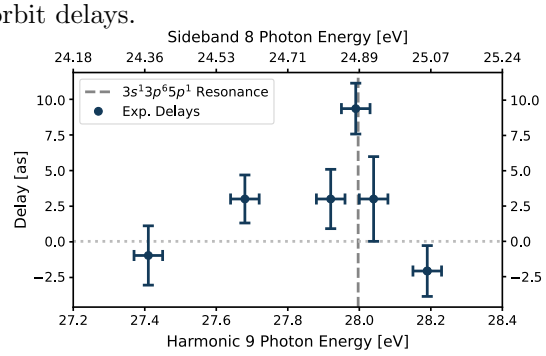


Figure 1. Experimental delays between the $(3p)^{-1}$ $^2P_{1/2}$ and $^2P_{3/2}$ spin-orbit components of argon as a function of photon energy of harmonic 9 (bottom axis) and the measured, corresponding sideband 8 (top axis).

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A highly accurate semiclassical method for atoms and molecules in ultrafast intense laser pulses

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Synopsis We extend our recently developed semiclassical strong-field Herman-Kluk (SFHK) method to calculate high-order harmonic generation (HHG) from atoms and molecules in intense lasers. We further demonstrate our method for photoelectron momentum spectra from molecules.

We show that our recently developed semiclassical strong-field Herman-Kluk (SFHK) method [1], based on a combination of the Herman-Kluk propagator and the strong-field approximation, can provide highly accurate for both high-order harmonic generation (HHG) spectrum and phase, comparable to those from the exact numerical solutions of the time-dependent Schrodinger equation. We provide detailed analyses of our method and its applications to the HHG process, particularly the analyses of the recombination time. We also demonstrate our method for calculating the photoelectron momentum spectra from molecules.

Within the SFHK, the main computational task is to solve the classical Newton equations for the active electron in the combined atomic (or molecular) potential and laser-electron interaction. The motion of the centers of the electron wave packets, modeled by coherent states, is governed by independent classical trajectories so that the computation can therefore be parallelized very efficiently. The method can be extended to polyatomic molecules in a straightforward manner.

As an example, we compare in Fig.1 HHG spectra and dipole phase for hydrogen atom in intense laser field. The laser is of 1,600 nm wavelength with an intensiy of 10^{14} W/cm2.

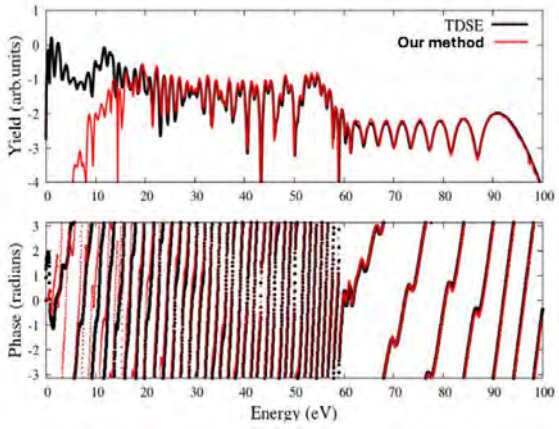


Figure 1. (a) Comparison of the HHG yield vs emitted photon energy from our SFHK method with exact numerical solution of the TDSE for hydrogen atom. The yields are plotted in logarithm scale. (b) Same as in (a) but for the HHG dipole phase.

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Laser-driven high-flux source of coherent quasi-monochromatic XUV radiation for coincidence spectroscopy

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Synopsis We have built a quasi-monochromatic coherent XUV source with a flux of more than 10^{12} photons per second at 26.5 eV. The source is based on high-harmonic generation in argon, driven by a frequency-doubled 100 kHz repetition rate fiber laser providing 30 fs pulses centered at 515 nm. As shown with argon, the XUV flux is sufficient to pump atomic and molecular dynamics in a single photon process. The advantage over multiphoton processes is the exactly known energy input. In addition, we report two-XUV-photon double ionization in argon.

Coincidence spectroscopy proved to be a powerful tool to study the dynamics of molecules, atoms and electrons. So far, our experiments have relied on intense laser pulses in the visible or near-infrared regime to pump and probe molecular and atomic dynamics [1, 2]. As these are multiphoton processes, the exact amount of absorbed energy is not well known and several reaction pathways are possible, which complicates the data interpretation.

With the goal to gain more control over the excitation, we investigate the usage of single XUV photons with well known energy to either pump or probe the system. This requires a very high flux of ideally monochromatic XUV radiation.

Here, we use high-harmonic generation in argon with 30 fs 515 nm pulses from a frequency doubled 100 kHz repetition rate fiber laser [3]. In order to maximize the HHG flux, we conducted scans of several parameters and related them to numerical simulations of the phase matching conditions [4]. The position of the plasma relative to the focus has proven to be the most interesting parameter and best phase matching is achieved a few millimeters before the focus.

In order to effectively monochromatize the generated XUV light, we spectrally filter the 11th harmonic (26.5 eV) with a specially coated mirror and focus it into a COLTRIMS setup, where we have performed first successful coincidence measurements with argon. Remarkably, The XUV flux is even sufficient for two-photon double ionization of argon. Furthermore, our setup allows for overlapping XUV and intense laser pulses at 515 nm to realize pump-probe experiments, and measure the XUV pulse duration.

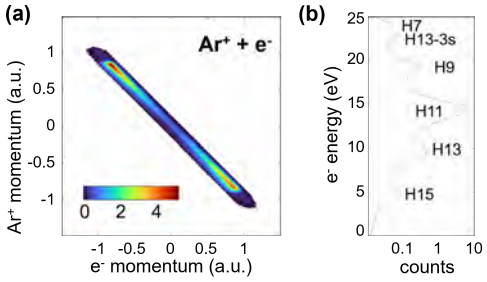


Figure 1. (a) Momentum of Ar^+ and e^- dissociated by XUV radiation and measured in coincidence. (b) Kinetic energy of the electrons showing the that 90% of the signal arises from the absorption of H11 at 26.5 eV.

In conclusion, we demonstrate coincidence spectroscopy using quasi-monchromatic XUV radiation from a high flux HHG source. Our observation of non-linear photoionization suggests that lab-based XUV-pump XUV-probe coincidence spectroscopy is within reach.

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Ionization-induced blue-shift of the driving pulse leads to increased harmonic yield and single attosecond pulse

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Synopsis In a strongly overdriven configuration for HHG we show the possibility to obtain an increased high-harmonic flux accompanied with the possibility to generate SAP. The targeted spectral domains for the XUV radiation are around 20 eV and 35 eV, respectively. Our main motivation is to facilitate the attosecond pump – attosecond probe experiments designed at MBI Berlin. We explain the mechanism of the driving pulse's blue shifting during propagation, investigate the influence of the initial chirp of the pulse, and we show the robustness of this configuration against CEP variation.

In the paper *Compact realization of all-attosecond pump-probe spectroscopy* [1] the feasibility of the attosecond pump attosecond probe spectroscopy (APAPS) experiment is demonstrated, using a commercial 1 kHz Ti:Sa laser in an out-of-focus geometrical configuration. This regime is called the strongly overdriven regime [2], which is a compact geometrical arrangement; in addition it exploits the possibilities given by the high-density noble gas medium and high initial intensity of driving laser up to 10^{16} W/cm^2. It has the advantage that it can be implemented in regular research labs.

Here we explore the physical mechanisms leading to increased harmonic yield and single attosecond pulse (SAP) around 20 eV and 33 eV as was targeted in the APAPS experiments. We reveal at optical cycle temporal resolution level the occurrence of the driving laser pulse's blue-shift. Then we investigate the consequences of the driver's blue-shift on the spatial, temporal, spectral properties of the generated harmonics. We demonstrate the feasibility of the SAP in the spectral domain covering the 18-40 eV domain, ideal for the APAPS experiments. We examine the robustness of the proposed configuration against CEP variation. The main reason for this is to relax the requirements for the experimental setup.

In Figure 1(a) we show the time-frequency distribution of the propagated driving pulse, emphasizing that a strong blue-shift occurs in the leading edge. Further we prove that in the most blue-shifted time-window the generated harmonic emission resists over propagation and results in one strong SAP (Fig. 1(b)).

The presented results have been obtained through simulation using a 3D non-adiabatic model for pulse propagation in Ar medium and HHG under high ionization conditions.

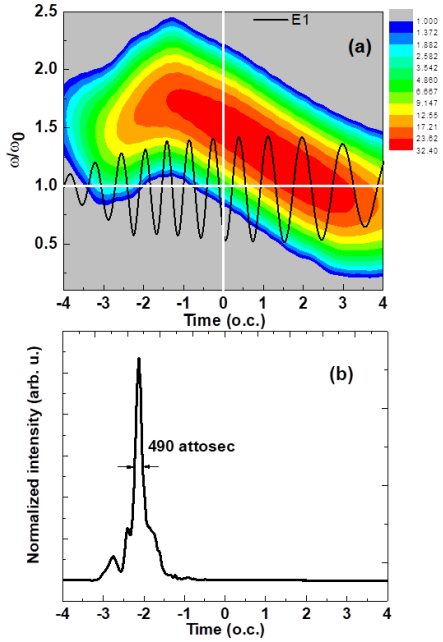


Figure 1. (a) Time-frequency map of the propagated driving pulse. The full electric field (E_1) is also shown as reference. (b) SAP is obtained after refocusing in the far-field.

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Polarization anisotropy of high harmonic generation in silicon controlled by coherent combination of ω-3ω fields

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Synopsis In this contribution we experimentally and numerically investigate high harmonic generation in crystalline silicon induced by synthesized two-color ω-3ω waveform. We observe modulation of amplitude and phase of the generated high energy photons as a function of both the relative phase of the ω-3ω fields and the direction of linear polarization with respect to crystallographic axes of silicon. This approach allows us to separate the individual processes contributing to high harmonic generation in this material.

The nonresonant optical interaction of ultrashort high-intensity laser pulses with a solid-state system induces coherent electron dynamics, leading to the emission of high harmonic photons in the form of subfemtosecond pulses, which represent the basis of attosecond physics [1]. Due to the nonlinear nature of this interaction, the properties of the generated high harmonic photons depend strongly on the waveform of the excitation field.

We use a precise waveform tailoring of the driving field by a coherent combination of a strong mid-infrared pulse with its third harmonic frequency. Changing the relative phase between the ω-3ω fields leads to time modulation of the driving field waveform, which allows us to control the time window of electron tunneling to the conduction band with attosecond precision [2]. By changing the direction of the driving field linear polarization, we are able to control electron trajectories in both reciprocal and real space. Consequently, the observed amplitude and phase modulation of high harmonic photons generated in a silicon crystal depend on the polarization direction of the field. In case of linearly polarized driving field oriented along [100] direction of silicon, we observe high harmonic photons with the energy of up to 9.5 eV corresponding to the 15th harmonic frequency [2] with clear modulation of the spectra as a function of the relative phase between the ω-3ω fields. However, with the linear polarization along [110] direction, higher photon energies are generated with the same driving field [3], but the modulation in the highest 17th harmonic order practically vanishes. This behavior is qualitatively reproduced by numerical simulations performed using time-dependent density functional theory (Fig. 1). Because the time window of electron tunnel excitation and the tunneling probability within each half-cycle of the fundamental wave change with the relative ω-3ω phase [2], this behavior can be explained by competing processes causing the modulation of the 17th harmonic order, which are out of phase and cancel each other.

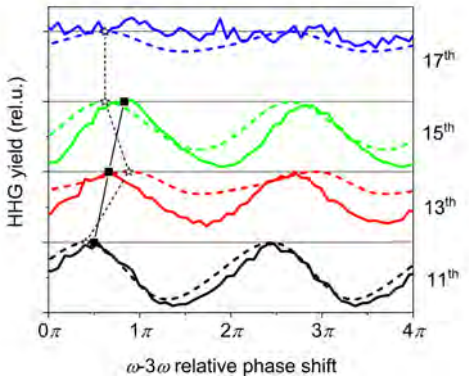


Figure 1. Measured (full curves) and calculated (dashed curves) modulation of high harmonic yield (from 11th to 17th harmonic order) for driving pulse linear polarization along [110] direction.

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Complete polarization measurements on broadband high-harmonic radiation

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Synopsis We demonstrate a comparative study on complete polarization measurements of a broadband high-harmonic source. The results of an optical method using a double-stage, reflection-based extreme-ultraviolet polarimeter are compared with those of an in-situ, non-optical measurement technique based on the investigation of photoelectron momentum distributions recorded by a reaction microscope.

Information on the polarization state of extreme-ultraviolet (XUV) radiation is essential in the research of chiral materials, and holds remarkable potential for revealing the geometrical and magnetization properties of matter [1]. One of the methods to unequivocally describe the complete polarization state of any electromagnetic radiation is by determining the related Stokes parameters using transmissive polarization retarders [2]. However, such measurements in the XUV wavelength range face serious challenges due to the short attenuation length.

In this work, the full polarization characterization of broadband XUV radiation produced via high-harmonic generation (HHG) is conducted simultaneously with two complementary measurement techniques. The Stokes parameters measurements with a double-stage XUV polarimeter [3] are compared with the outcome of a non-optical, in-situ method based on the investigation of the photoelectron momentum distributions (PMDs) of electrons resulting from the ionization of atomic targets recorded by a reaction microscope [4].

The experiments were performed on the HR GHHG Gas beamline at the Extreme Light Infrastructure Attosecond Light Pulse Source (ELI ALPS) Facility [5]. High-harmonics were generated in the 15–40 eV spectral region in a loose focusing geometry by using a 10 kHz repetition rate, near infrared laser source equipped with two post compression multi-pass stages to produce few-cycle pulses for HHG.

Figure 1 presents the recorded datasets ((a) and (c)) and the retrieved Stokes parameters ((b) and (d)), measured with the non-optical technique, and with the reflection-based, double-stage XUV polarimeter, respectively.

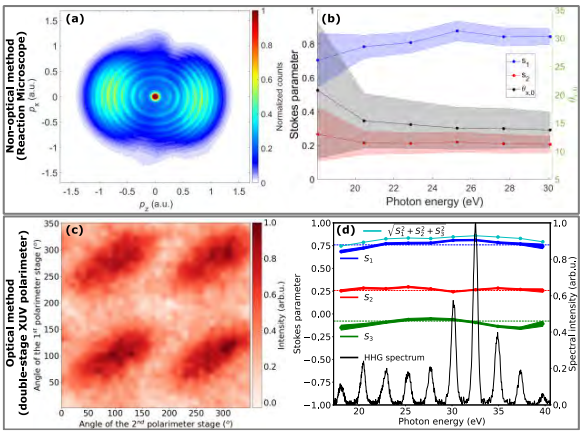


Figure 1. Measured datasets: the PMD (a) and the so-called Malus-surface (c) recorded with the non-optical (top) and the optical (bottom) measurement techniques. The corresponding retrieved Stokes parameters are shown in subfigure (b) and (d), respectively, representing the measurement error as well.

Besides revealing a photon energy-dependent change of the polarization state of broadband high harmonic radiation in successful comparative measurements, these results also present a high-harmonic source offering with variable polarization for the investigation of chiral and magnetic phenomena with attosecond resolution.

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Beamline development optimized for STARPES : a highly tunable extreme ultraviolet lightsource

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Synopsis We present the upgrade of an attosecond beamline, which was previously using a Ti:Sa laser source [1]. We increase the repetition rate from 10 to 100 kHz by using an industrial-grade Yb-doped laser system. Based on the High Harmonic Generation (HHG) process, the beamline is optimized for Spin-, Time-, and Angular-Resolved Photoemsission Spectroscopy (STARPES) in solids and gases. Its design allows the tuning of the pulse duration from 300 fs to 25 fs as well as switching between the full extreme ultraviolet (XUV) spectrum or monochromatized harmonics down to 200 meV bandwidth.

HHG-based STARPES allows the investigation of the momentum-resolved dynamics of electronic spin polarization upon light excitation [2]. Up to now, it has mostly been performed using Ti:Sa sources, which are limited in their repetition rate.

On the other hand, the advent of Yb-based sources has now allowed for much larger repetition rates. In the context of STARPES, this solves two major limitations: (1) Reduce the acquisition time required by increasing the counts of photoemitted electrons. (2) Mitigate the space charge occurring whenever more than one photoelectron is emitted in vacuum. This modifies both the spectral and angular distributions of photoelectrons leading to a loss of resolution in STARPES experiments.

Here we present our implementation of an Yb-doped laser system for STARPES. The laser provides 800 µJ energy per pulse at 100 kHz centered at $\lambda_0 = 1030$ nm with a pulse duration of 350 fs. The pulses are post-compressed using self-phase modulation (SPM) within a gas-filled multi-pass cell (MPC) to spectrally broaden the pulses and subsequently compensate for the group delay dispersion (GDD) using chirped mirrors. Simulations have guided the development of an Argon-filled Herriot-type MPC, optimized for a 495 mm cavity length and 250 mm radius of curvature spherical mirrors. Experimental results demonstrate the compression of the 350 fs input pulses down to 24 fs.

The pulses are then used in high-harmonic generation, either directly at 1030 nm, yielding the $q = 17^{th}$ to $q = 49^{th}$ harmonic, or with the frequency-doubled beam. Because of the favorable wavelength scaling law [3], the 515 nm driver results in significantly increased XUV flux, at the cost of lower cutoff energy ($q = 9$ to $q = 13$).

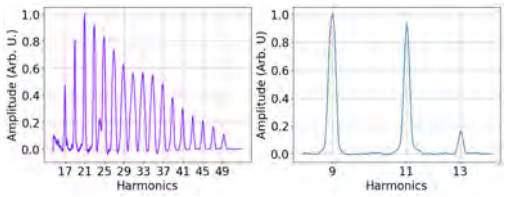


Figure 1. Harmonics of the 1030 nm (left) and 515 nm (right) drivers.

We will then present ARPES measurements performed in this configuration, as well as a complete characterization of the XUV stability, demonstrating the advantage of this new laser source.

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Enhancement of vacuum-ultraviolet laser generation through near-threshold harmonics driven by a few-cycle two-color laser field

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Synopsis Vacuum-ultraviolet (VUV) lasers are key in scientific and industrial use. Traditional generation methods using large facilities like free-electron lasers are costly. Recent advancements reveal that near-threshold harmonic (NTH) generation offers a compact alternative. However, low pulse energy limits its use. We used a two-color femtosecond laser field to enhance NTH generation, significantly boosting VUV laser intensity by two orders of magnitude over previous methods. This results in high-intensity, adjustable VUV lasers, marking significant progress in the field.

Recent advancements in the generation of vacuum-ultraviolet (VUV) lasers have sparked considerable interest due to their critical applications in scientific research and various industrial processes. For example, a nuclear clock was recently achieved by utilizing a femtosecond VUV laser frequency comb to drive the 229mTh nuclear isomeric transition, significantly advancing precision measurement techniques[1]. Traditional methods for producing VUV lasers involve large and costly setups such as free-electron lasers and synchrotron sources. Near-threshold harmonics (NTH) generation provides a significant method to obtain compact, table-top sources of femtosecond coherent VUV lasers[2, 3, 4, 5]. However, how to further improve the single pulse energy of the VUV laser remains a challenge, which is crucial for strong field and atomic and molecular physics. Our study presents a breakthrough in generating and enhancing VUV lasers through near-threshold harmonics (NTH) generation, using a novel few-cycle, two-color femtosecond laser field approach. In our research, we have successfully demonstrated a significant increase in the intensity of the VUV laser—by two orders of magnitude—compared to previous methods employing longer-duration two-color lasers. This enhancement was achieved by optimizing the excitation of low Rydberg states through precise control of the laser field parameters. Additionally, we showed that adjusting the driving laser's ellipticity and the type of target atom allows for the modification of the VUV laser's ellipticity and wavelength, thus providing a versatile tool for various applications.

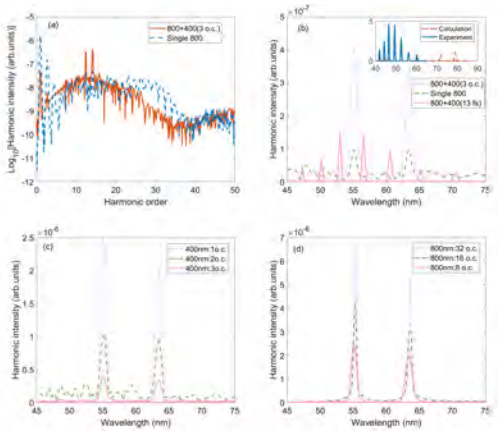


Figure 1. Harmonic spectra generated by a single 800 nm laser field compared to those produced from a two-color laser field interacting with Ne. The inset is a comparison between the pink NTH spectrum and the experiment results in reference [4].

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High-power and high-energy post-pulse-compression of a thin-disk laser

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Synopsis The recent breakthroughs in nonlinear spectral broadening have unlocked the potential for extending Yb-doped lasers to generate high-flux and high-repetition-rate extreme ultraviolet (EUV) attosecond pulses. We reported our recent progress in developing high-power and high-energy ultrafast light sources by implanting two main post-pulse-compression techniques of multiple thin plates (MTP) and multipass cells (MPC) in Yb-based thin-disk laser systems.

Over the past two decades, attosecond science has emerged as a novel research field. Significant achievements in attosecond optics often rely on advanced laser source technologies, demanding further enhancements in pulse intensity, average power, and pulse duration. The advanced ultrafast pulse techniques have boosted attosecond pulse generation and attosecond physics development. However, up to now, in most novel applications in attosecond science, such as attosecond coincidence spectroscopy, attosecond water-window transient spectro-scopy, and EUV coherent nano-imaging, a high-flux and high repetition EUV light source is demanding. Due to the limitation in the high cost of pump sources, substantial quantum efficiency losses, and severe thermal effects, Ti: Sapphire-based laser systems came across the neck-bottle in the new age of attosecond science. During the last decades, Yb-, Ho-, Tm-doped high energy and high repetition rate lasers obtained an efficient development. Here, we reported our recent significant development of two main post-pulse compres-sion techniques of MTP and MPC in Yb-doped high energy and high power laser systems.

To reduce the high-order dispersion affect, we employed a two-stage MTP technique to generate a high-quality compressed laser pulse with a pulse duration of 70fs in the first stage and 22 fs in the second state [2]. As shown in Fig.1, a high temporal contrast pulse was achived. To scaling the high pulse energy situation, we modified the MPC structure by employing seperated high refleticity concave mirrors to spatially reproduce the beam focus region. The first stage is designed to maintain a pulse compression from 1.3ps to 300fs. A broadband spectrum covering 960nm to 1100nm in the second stage supports a sub-40fs laser pulse with a final efficiency above 80%. By seperating the pulse compression into two stages, the huge chirp management versus the broadband spectrum could be well controlled. The rare gas atoms were choosen as the nonlinear interaction media in both stages. Depending on the final application requirement of attosecond temporal spectroscopy or EUV imaging spectroscopy, the pulse duration from the second stage can be adjusted between 80fs and sub-30fs.

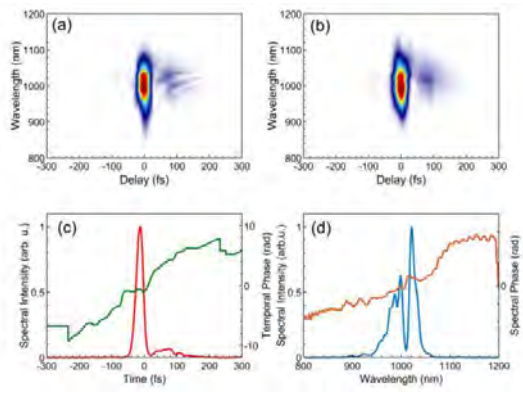


Figure 1. Experimentally measured and theoretically reconstructed FROG trace in MTP.

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Generating VIS-UV sub-femtosecond pulses in a single fiber and sampling their fields

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Synopsis We generate sub-fs VIS-UV light pulses by self-compression of the output of a commercial Ti:Sapph CPA in a hollow capillary. We finally sample the field waveform using self-gating.

Compressed few-fs VIS-NIR light pulses have been the workhorse of attosecond experiments since the birth of the field. Extending the same tools to the UV is desirable in order to cover low energy electronic excitations in molecules and solid state systems. However, generating and field-sampling single-cycle pulses in the UV region has posed significant challenges: first of all, manifacturing UV optics becomes increasingly difficult at short wavelengths, or even impossible (as in the case of broadband chirped mirrors between $\lambda = 60$ nm and $\lambda = 200$ nm); secondly, the conversion efficiency from the VIS to the VUV is not easy to reconcile with good temporal compression; finally, the field sampling requires a gating event shorter than half of the period of the UV radiation.

Soliton dynamics in a hollow core fiber are a promising approach to overcome these limitations and generate intense compressed pulses down to the VUV [1]. Resolving the electric field of these transients with nonlinear photoconductive sampling (NPS [2]) establishes their subfemtosecond nature [3].

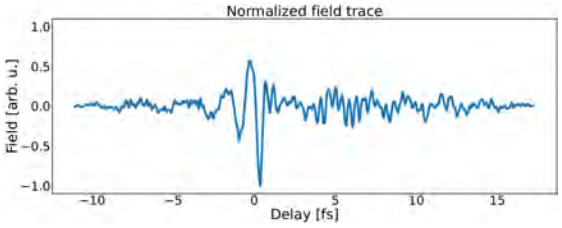


Figure 1. Electric field of the soliton and resonant dispersive wave as a function of time (delay of the gating event).

In this work we demonstrate that we can access these soliton dynamics directly with a 800 nm, 26 fs pulse, obtaining a sub-femtosecond soliton centered around 400 nm along with a resonant dispersive wave centered at 130 nm. We sample the complete waveform of the field through NPS (Fig. 1). In particular the gate event is provided by the soliton itself, eliminating the need for an external gating pulse.

In this configuration we investigate the change in the waveform when we increase the gas pressure in the fiber. We observe the effect of dispersion on the waveform in the time domain, as well as the gradual energy transfer from the soliton to the resonant dispersive wave.

The setup described here simplifies significantly the generation process and field sampling of sub-fs pulses, additionally providing spectral coverage down to 120 nm. These technical capabilities will help investigating both linear and nonlinear behaviour of solid-state, molecular and gaseous systems in the UV region, by enabling for example polarization sampling and UV pump-UV probe spectroscopy. Ultimately, this proof of principle experiment is heading toward closing the spectral gap between the near-UV (>200 nm) and the extreme-UV (<60 nm).

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200-kHz laser system delivering few-cycle near-infrared pulses for attosecond strong-field experiments

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Synopsis By focusing few-cycle pulses with a long central wavelength into a gas jet, high-order harmonics with a high cutoff-energy can be generated. We report on a laser system that provides driving pulses for high-order harmonic generation, centered around 2 µm and with a pulse duration of less than 20 fs at a repetition rate of 200 kHz. The XUV light generated in a home-built argon gas target covers photon energies up to 150 eV. It can be used for subsequent photoemission experiments from the surface of solid samples.

The nonlinear process of high-order harmonic generation (HHG) is a method for generating ultrashort pulses in the extreme ultraviolet (XUV) spectral range. The generated broad spectrum corresponds to a pulse train with pulse durations on the attosecond time scale and can thus be used to probe electron dynamics on surfaces on their natural time scale [1]. To perform strong field experiments on nanostructures with attosecond resolution, a high repetition rate is required due to the Coulomb repulsion of the photoelectrons, and a long wavelength to reach the strong field regime is favourable. Few-cycle laser systems meeting these parameters are not commercially available and only a few have been demonstrated in the literature [2, 3].

We report on a compact home-built laser system that provides driving pulses for such strong-field experiments, centered around 2 µm, with a repetition rate of 200 kHz and a pulse energy of up to 30 µJ. Good passive carrier-envelope phase (CEP) stability with an RMS value of the CEP noise of 108 mrad measured over 10 minutes, averaged over 200 pulses, is found due to the difference frequency generation (DFG) process used to generate the near-infrared (NIR) pulses. The temporal structure of the NIR pulses is investigated using the dispersion-scan (d-scan) technique, which reveals a pulse duration of less than 20 fs (Fig. 1 (a)) [4]. By tightly focusing the intense, few-cycle NIR pulses, intensities of up to $5 \cdot 10^{14}$ W/cm^2 are achieved, making HHG in a home-built argon gas target possible. The resulting XUV spectrum covers photon energies up to 150 eV, measured with a home-built XUV spectrometer (Fig. 1 (b)).

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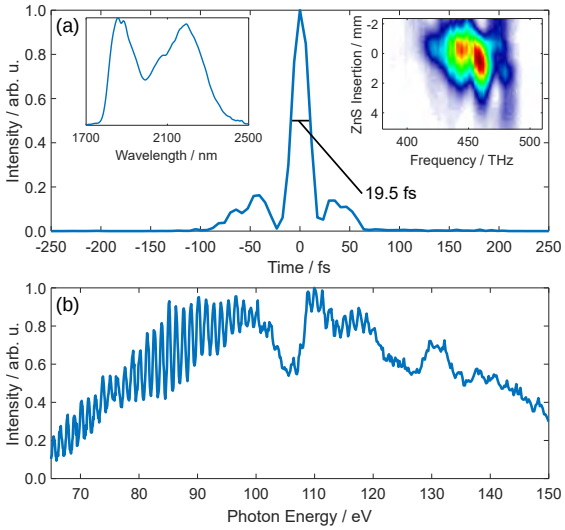


Figure 1. (a) NIR pulse characterization via d-scan (b) Resultant HHG spectrum when the NIR pulses are focused into an argon gas jet.

In combination with the NIR pulses, the generated XUV pulses can be used to conduct attosecond interferometry experiments on the surface of solid samples [5]. As an outlook, time-resolved measurements on a zinc oxide surface are performed, where photoelectrons emitted from the well-defined Zn-3d and O-2p core level states are detected with a photoelectron spectrometer [6].

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Intensity Control of Few-Cycle Pulses

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Synopsis We present a simple and cost-effective method for continuous intensity control of few-cycle pulses using a non-ultrabroadband half-waveplate placed before a gas-filled hollow fiber, followed by a thin-film broadband polariser. This method enables the pulse energy to be controlled by a factor of ~ 25 while preserving a pulse duration of 5.4 ± 0.2 fs. We demonstrate the efficacy of this technique in high harmonic generation (HHG) experiments in Kr, covering an intensity range of 30−190 TW/cm^2, enabling precise control of harmonic yield in few-cycle-driven HHG.

Precise intensity control of few-cycle laser pulses is essential for strong-field physics and attosecond science [1], enabling the study of ultrafast electron dynamics and isolated attosecond pulse generation. Conventional intensity control methods, such as neutral density filters, suffer from spectral limitations, unwanted dispersion, or discrete attenuation steps. Additionally, placing a non-ultrabroadband half-waveplate after the fibre leads to undesired stretching of the laser pulse.

We present a simple, cost-effective solution by placing a non-ultrabroadband half-waveplate *before* a gas-filled hollow-core fiber (HCF) and a broadband thin-film polariser after compression. This approach allows for smooth and continuous tuning of the output pulse energy by a factor of $\sim$25 while preserving carrier-envelope phase (CEP) stability and maintaining a nearly constant pulse duration of 5.4 ± 0.2 fs using an argon-filled HCF.

We use our scheme with a commercially-available Ti:Sapphire chirped pulse amplifier (Femtopower HE CEP) which delivers 0.60 mJ, 30 fs pulses into a 1-m-long, 350-μm diameter argon-filled HCF at 1 bar. The spectrally broadened pulse is then temporally compressed using chirped mirrors and a wedge pair before being injected into the interaction region for high harmonic generation (HHG) in a krypton gas jet. The driving pulse duration was retrieved using the dispersion-scan technique [2] (Figure 1 (a)) from a pick off after the gas jet, ensuring that the same beam path was used throughout.

To validate our approach, we measure the harmonic yield for harmonics 11-17 (H11-H17) for an intensity range from 30–190 TW/cm^2 (Figure 1 (b)). This work demonstrates that precise intensity control can be achieved without altering the pulse duration, eliminating the need for costly, achromatic optics. Additionally, our intensity control method can be easily integrated into existing HCF setups.

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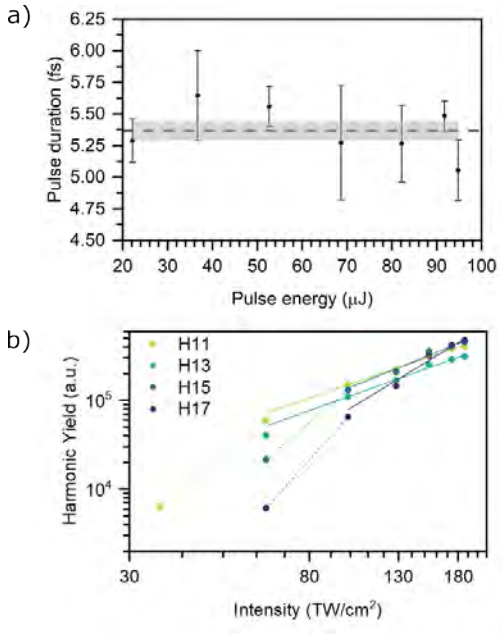


Figure 1. (a) Retrieved pulse duration as a function of energy. Error bars of the data correspond to the standard error, the dashed line and grey bar correspond to the mean and standard error of all of the data. (b) Log-log plot of the harmonic yield as a function of driving intensity for harmonics 11−17. The solid and dashed lines show a linear fit.

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Spectral modulation and double attosecond pulse emission from relativistic laser-plasma interactions

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Synopsis We demonstrate the coexistence of the Relativistic Oscillating Mirror mode and the Coherent Synchrotron Emission mode in the process of high-order harmonic generation resulting from relativistic laser-plasma interactions. Spectral modulations arise when these two mechanisms coexist, leading to the emission of harmonics with a sub-femtosecond time delay between consecutive orders, offering a novel approach to introduce controlled subfemtosecond delays between two attosecond pulses. It has potential applications in XUV-pump, XUV-probe spectroscopy experiments and for studying nanobunch dynamics.

Bright ultrafast pulses of radiation open the door to investigate a wealth of new regimes, the high-order harmonics generations (HHG) from laser-plasma interaction (LPI) becomes a promising method to generate intense attosecond pulses [1].

There are two dominant mechanisms to generate harmonics [2,3]: (1) Relativistic Oscillating Mirror (ROM) and (2) Coherent Synchrotron Emission (CSE). As displayed in Fig. 1, with given specified laser parameters, the HHG spectral amplitute oscillates as the harmonic order increases, which is called “modulation". We prove this modulation is due to the transition of the HHG mechanism from ROM to CSE model, during which both mechanisms coexist and emit HHG in sequence with attosecond time delay. The period of the modulation depends on both the laser parameters and plasma parameters. It is due to the transition between the ROM and CSE mechanisms, which can also provides insight into the physics behind the nanobunch generation and thus may be utilized to generate extremely intense attosecond pulses via CSE HHG. The steepness of the density gradient at the plasma-vacuum interface is examined to see how this parameter affects the high-order harmonic generations and isolated-attosecond pulse creation.

The subfemtosecond time delay between the ROM and CSE attosecond pulses can be varied by controlling the experimental conditions, and their high peak intensities could be exploited to perform novel XUV-pump, XUV-probe spectroscopy experiments.

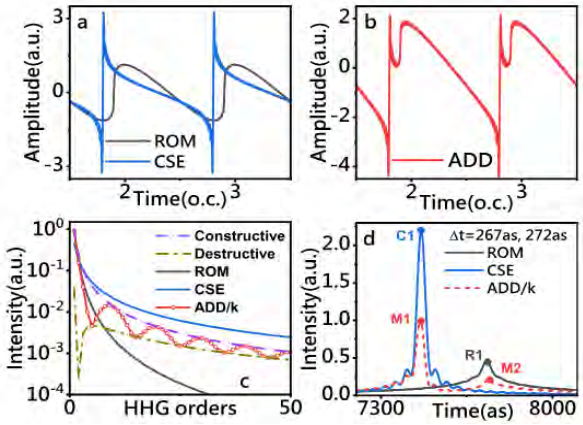


Figure 1. (a,b)Temporal profiles of the electric field; (c) the power spectrum and (d) separated attosecond pulses of ROM and CSE. The laser parameters are: ω=0.057 a.u., I_0= 5.3×10^{20}W/cm^2, and the plasma density Ne/Nc = 20.

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High-intensity attosecond beamline for XUV pump – XUV probe investigations

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Synopsis The realization of attosecond-time-resolved XUV pump – XUV probe experiments is one of the big challenges of attosecond physics. Over the last decade, a few groups used this technique to measure electron dynamics in various interactions. However, those experiments were limited to photon energies below 50 eV. Here, we present a new intense source of XUV attosecond pulses with up to ~140 eV.

The XUV (Extreme Ultraviolet) pump-XUV probe technique is an advanced ultrafast spectroscopy method crucial for enhancing our understanding of ultrafast electronic processes in materials, molecules, and quantum systems at the attosecond time scale. A few measurements were performed with low photon energy, specifically below 50 eV [1-3], and around 100 eV [4]. It quickly becomes evident that achieving this goal is challenging due to the need for high XUV intensity and photon numbers.

In this project, we have developed an intense XUV beamline of up to 140 eV photon energy by scaling up the high harmonic generation (HHG) process [5]. To this end, a homemade wave-form-controlled optical parametric synthesizer was used, which provides a peak power of ~ 100 TW, covering the NIR-VIS spectrum and producing pulses with a duration of less than 4.5 fs. By focusing the laser pulses with a 22 m focal length mirror into a neon gas cell high harmonics are generated. After generation, the various properties of XUV radiation are measured (spectrum, energy and beam profile) and optimized. Under optimal conditions, a broad XUV continuum is produced (Fig. 1).

For the planned applications, the XUV beam is focused with an ellipsoidal mirror to a focal point of about 4-6 micrometres. The XUV focus can be monitored in real-time using an adjusted CMOS camera during the experiment.

Different detectors are implemented in the beamline including ion microscope and velocity map imaging electron spectrometers to realize the XUV pump XUV probe experiment.

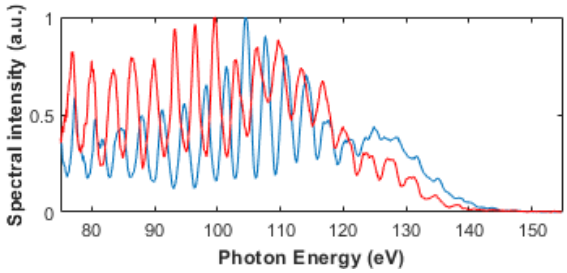


Figure 1. XUV spectral intensity for two different laser CEP values

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High-harmonic spectroscopy probes the directional charge-density-wave-strengths of $TiSe_2$

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Synopsis With high harmonic generation as a sensitive probe for electronic and structural properties we investigate the charge density wave of TiSe2 through its phase transition down to 14 K. Asymmetry in the non-linear response between previously thought symmetric directions is found and identified as subtle changes in the CDW strength by our mean-field model.

Strongly correlated phases enable quantum effects to appear on a large scale, as seen in superconductors and superfluids [1]. At the core of these phenomena is the formation and interaction of quasiparticles during the Bose-condensation process. A closely related phase is the charge-density wave (CDW), which in $TiSe_2$ forms a 2×2×2 supercell structure and transitions into a superconductor when intercalated with Cu atoms. The origin of the CDW phase remains uncertain, potentially arising from either electron-phonon or excitonic interactions. Recent studies suggest the presence of a new phase of matter in $TiSe_2$: the exciton condensate [2]. To investigate $TiSe_2$ across its phase transition at low temperatures, we use high-harmonic spectroscopy (HHS), a novel method for quantum phase characterization [3]. The emitted harmonics, driven by charge carrier motion within the solid, provide insights into excitonic couplings and structural changes through polarimetry. In Fig. 1(c), the intensity of the 5th harmonic (H5) aligns with the hexagonal lattice of the material. As the system enters the CDW phase below 200 K, an anisotropic response emerges, showing a significant enhancement at temperatures far below Tc, particularly along one of the previously symmetric Γ-Mi directions. Using a mean-field model, we systematically eliminate alternative explanations for this asymmetry, identifying the CDW strength (ΔQi) as the primary factor. Even small directional differences within the crystal uniquely generate the strong asymmetric signal observed experimentally (Fig. 1(c), solid lines).

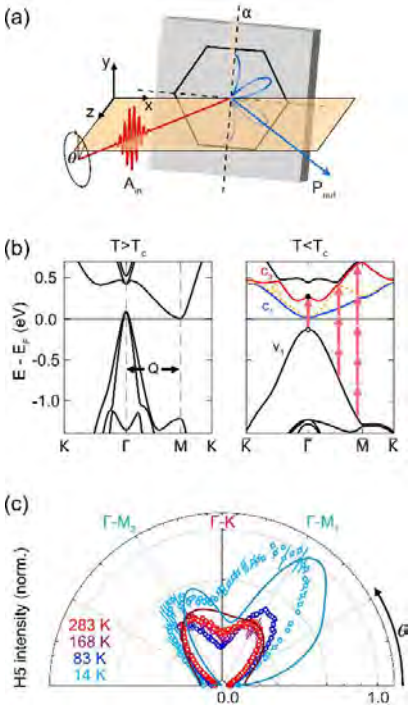


Figure 1. (a) Setup with driving field incident at 45° and generated harmonics in reflection geometry. (b) Bandstructure in the normal & CDW phase with nearly resonant driving field and harmonics H3 & H5. (c) H5 as function of driving field polarization and temperature, maximizing for G-M alignment and anisotropic signal below T_c.

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XUV yield optimization of two-color high-order harmonic generation in gases

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Synopsis We perform an experimental two-color (1030 nm and 515 nm) high-order harmonic generation study in argon. The intensity of the second harmonic and its phase are varied while keeping the total intensity constant. We extract the optimum values for the relative phase and ratio of the two colors which lead to a maximum yield enhancement for each harmonic order in the XUV spectrum. An analysis of the curve mapping time to energy within the three-step model predicts the required two-color phase and ratio for a given harmonic order, total laser intensity, fundamental wavelength, and ionization potential.

High-order harmonic generation (HHG) is an established method for producing highly coherent radiation in the extreme ultraviolet (XUV) spectral region with femtosecond to attosecond pulse duration [1]. Due to the highly nonlinear nature of HHG the efficiency of generating XUV is very low. For industrial applications, enhancing the XUV flux at specific energies is of great interest [2]. We present an experimental HHG study with a two-color laser driver ($\omega/2\omega$, 1030 nm and 515 nm) in argon. In the experimental setup, the intensity ratio R between the second harmonic 2ω and the total intensity I_{tot} can be varied while keeping I_{tot} constant in the gas target, in addition to relative phase (ϕ) control with a 2π range [3]. For each (R, ϕ) combination the XUV yield for each harmonic order H is measured and the optimum values $(R_{\mathrm{opt}}, \phi_{\mathrm{opt}})$ for the maximum yield enhancement compared to $R = 0$ (single color HHG with only the fundamental ω) are extracted, showing enhancement factors > 20. An intuitive analysis based on the three-step model [4] at $(R_{\mathrm{opt}}, \phi_{\mathrm{opt}})$ reveals that this enhancement occurs due to a flattening of the time-energy curve [5], which we have validated experimentally and generalized with a predictive equation [6]. This equation relates the yield of the emitted harmonics E_H of corresponding order, to the intensity ratio R:

$$E_H = I_p + \frac{1}{2}U_p\left(\sqrt{2\cdot 3.17\cdot(1-R)} - \sqrt{R}\right)^2, \quad (1)$$

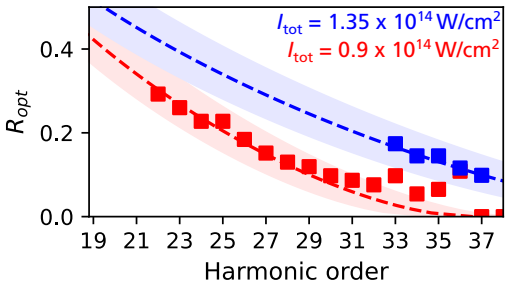


Figure 1. Comparison between Eq. (1) (dashed lines) and experimental results (squares). The shaded areas represent the effect of a 10 % change of I_{tot} and R in Eq. (1). Figure adapted from [6].

where U_p is the ponderomotive potential and I_p the ionization potential. Figure 1 shows excellent agreement between Eq. (1) (dashed lines) and the experimentally found R optima for each harmonic order (squares) and different values for I_{tot}. Current work extends this study to a fundamental-third harmonic ($\omega/3\omega$) configuration, address phase matching, and explores the interplay between ionization and trajectory selection to further optimize XUV yield.

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Tracing electronic coherences at conical intersections with heterodyned attosecond four-wave-mixing spectroscopy

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Synopsis Conical intersections are omnipresent in photochemistry. We propose a novel experimental table-top approach that is directly sensitive to electronic coherences at conical intersections: heterodyned attosecond four-wave-mixing spectroscopy (Hd-FWM). This scheme is theoretically applied to photoexcited CH_3I, where it can resolve small coherence magnitudes, $< 1\%$. Surprisingly, about 1/3 of the maximum coherence magnitude is still present in the atomic iodine fragments after photodissociation. The full complex coherence is time-resolvable by applying phase cycling to Hd-FWM.

Electronic coherences at conical intersections (CIs) have sparked considerable interest, e.g. with the TRUECARS approach [1], but remain experimentally challenging. To address this, we theoretically introduce heterodyned attosecond four-wave-mixing spectroscopy (Hd-FWM) as a table-top technique. Hd-FWM combines the temporal resolution of isolated attosecond pulses with the direct sensitivity to small coherence magnitudes by heterodyned detection. The noncollinear FWM scheme disentangles populations from coherences via phase-matching considerations [see Fig. 1(b)]. We theoretically test the Hd-FWM scheme on the nonadiabatic dynamics of gas-phase, UV-excited CH_3I. Here, the potential energy surfaces $^3Q_0$ and $^1Q_1$ result in $j = \{1/2, 3/2\}$ iodine fragments after dissociation [see Fig. 1(a)]. Surprisingly, about 1/3 of the maximum coherence magnitude induced at the CI is preserved in the atomic limit. The Hd-FWM approach is not only sensitive to these small ($< 1\%$) coherences but also temporally resolves the 3.8 fs oscillation periods [see Fig. 1(c)]. Furthermore, the complete complex coherence can be reconstructed by cycling the relative carrier-envelope phase of two laser pulses. Preparing coherences via CIs and transferring them to long-lived atomic coherences is of interest for ultrafast quantum information technology.

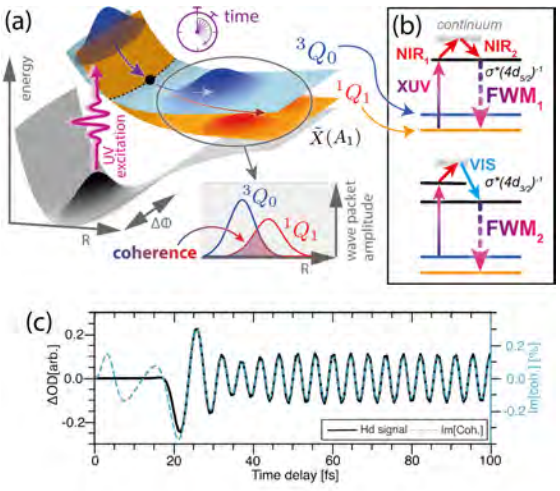


Figure 1. Electronic coherence at a CI in CH_3I. (a) Potential energy surfaces and wave-packet dynamics. (b) Hd-FWM pathways. (c) Imaginary part of the electronic coherence (blue dashed line) mapped by Hd-FWM (black solid line).

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High power sub-20-fs laser based on gas multi-pass cell

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Synopsis We demonstrated high power of 244 W at 500 kHz with pulse duration of sub-20-fs using a nonlinear pulse compression scheme based on a gas multi-pass cell. This laser source, characterized by high average power (>200 W) at 500 kHz with pulse duration of <20 fs, will provide a critical driving light source for the development of HHG-based ultrafast Tr-ARPES systems.

By integrating high harmonic generation (HHG) [1] sources with angle-resolved photoemission spectroscopy (ARPES) [2], which provides momentum- and energy-resolved detection capabilities, the resulting time-resolved ARPES (Tr-ARPES) system has emerged as an ideal experimental platform for investigating ultrafast processes such as electron dynamics, owing to its broadband spectral tunability and ultrafast temporal resolution. However, achieving efficient HHG and its application in Tr-ARPES necessitates high-power, high-repetition-rate, shorter femtosecond driving sources. A promising approach to generate such high-power few-cycle pulses is post-compression techniques, including hollow-core fibers, thin-solid-plates, and multi-pass cells (MPCs) [3]. Among these, the MPC-based nonlinear pulse compression scheme stands out due to its advantages in spectral broadening uniformity, high transmission efficiency, low cost and complexity, and excellent flexibility, making it one of the main solutions for high power few-cycle femtosecond pulse generation.

This study proposes and experimentally validates a nonlinear pulse compression scheme based on a gas-filled multi-pass cell. Utilizing argon as the nonlinear medium, spectral broadening is achieved via the self-phase modulation (SPM) effect. Experimental results demonstrate that under initial pulse parameters of 73.8 fs/145 W/500 kHz and 1.3 bar argon，nonlinear spectral broadening exceeding 200 nm was realized. Subsequent dispersion compensation using chirped mirrors yields a compressed pulse duration of 19.8 fs with a system efficiency of 82.7%. When the input power is increased to 260 W under 0.5 bar argon pressure, the output power reaches 244 W (system efficiency exceeding 93.8%) while maintaining an effective spectral bandwidth >200 nm.

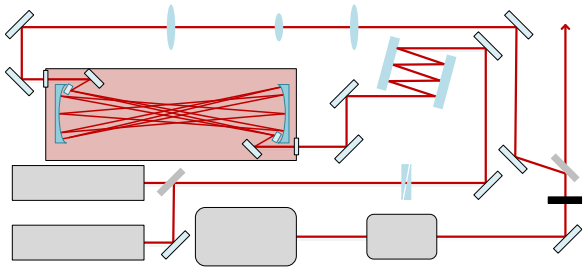


Figure 1. Experimental setup.

This high power laser developed, which has power of > 200 W at 500 kHz with pulse duration of sub-20-fs, provides critical driver for developing HHG-based ultrafast Tr-ARPES systems, with significant applications in strong-field physics and ultrafast science.

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Filamentation-Assisted Isolated Attosecond Pulse Generation

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Synopsis Isolated attosecond pulses (IAPs) are essential for capturing ultrafast dynamics, yet achieving high stability and contrast remains challenging. We demonstrate that filamentation in a semi-infinite gas cell enables infrared driver self-compression, leading to high-contrast IAPs. In argon, Yb-based pulses self-compress from 4.7 fs to 3.5 fs, simultaneously producing 200-as IAPs at 70 eV. Filamentation in neon and helium further shortens IAPs to 69 as at 100 eV and 65 as at 135 eV. This approach advances both post-compression techniques and attosecond science.

Ultrafast pulses generating high-order harmonics (HHG) are essential for attosecond light, enabling ultrafast dynamics observation. IAPs are typically produced using few-cycle CEP-stabilized pulses in short gas cells (SGC). However, the semi-infinite gas cell (SIGC) has emerged as a promising alternative, utilizing filamentation for HHG, balancing Kerr self-focusing and plasma defocusing for pulse self-guidance and self-compression.

Here, we experimentally demonstrate IAP generation via filamentation using Yb-based laser pulses. By focusing <5 fs pulses at 1030 nm into Ar-, Ne-, and He-filled SIGCs, we achieve high temporal IAP contrast [1]. Simulations attribute this to self-compression and self-guidance in SIGC. Specifically, 4.7 fs pulses in Ar undergo 25% self-compression (to 3.5 fs) with improved spatial mode. Compared to SGCs, SIGC-generated IAPs show higher temporal contrast and brightness. Streaking confirms stable filamentation in Ar, producing 200-as IAPs at 70 eV, with similar results in Ne (69 as, 100 eV) and He (65 as, 135 eV).

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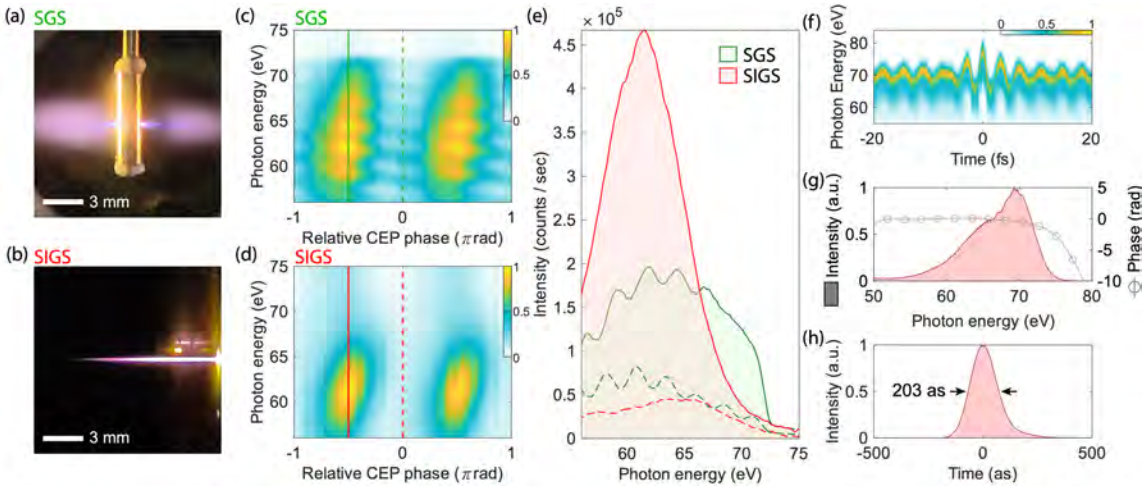


Figure 1. HHG in (a) SGC versus (b) SIGC. (c) and (d) show the normalized CEP-dependent HH spectra, driven by post-compressed <5 fs pulses from a Yb laser, in SGC and SIGC, respectively. The solid line indicates the optimized CEP (relative CEP phase = 0) for maximum HHG yield. A relative change of 0.5π rad in CEP (dashed lines) leads to significant variations in the high harmonic yield. (e) presents a direct comparison of these CEP-dependent HH yields. (f) Attosecond streaking spectrograms were obtained from self-compressed IR pulses and the resulting IAPs generated from filaments in Ar. (g) The retrieved spectrum and spectral phase, as well as (h) the pulse duration of 203 as, were extracted using PROOF [2].

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Attosecond X-ray Core-level Chronoscopy of Aromatic Molecules

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Synopsis We performed angular streaking of the core-shell ionization using the attosecond XLEAP mode of LCLS, determining the relative delays between N-1s and C-1s photoionization for pyridine, pyrimidine, and 1,3,5-triazine on a dense energy grid.

We conducted attosecond angular streaking on the K-shell photoionization of a series of aromatic azabenzenes using attosecond X-ray pulses from the Linac Coherent Light Source (LCLS) at SLAC National Accelerator Laboratory [1]. The combination of the linearly polarized X-ray (400∼450 eV) pulses and a co-propagating circularly polarized IR (1300 nm) beam allows the mapping of the photoionzation time delay into the momentum modulation in the polarization plane, which is recorded by the co-axial velocity map-imaging (c-VMI) spectrometer perpendicular to the light-propagation direction [2, 3]. The atom-like core-level spectra avoid spectral congestion and convey information about the scattering dynamics from point-like sources.

The relative photoionization delays between N-1s and C-1s have been measured at photon energies covering 400∼450 eV, where the C-1s delays are negligibly small because of the high kinetic energy of the electrons. The measured ionization time delay shows two maxima in this range for all of the three azabenzenes, as plotted in Fig. 1, which are assigned to shape resonances [4]. We find that the delays systematically increase with the number of nitrogen atoms in the molecule and feature multiple resonances, which is supported by calculations using the Schwinger variational principle with a correlation-polarization potential in combination with the analytical expression for the Coulomb-laser-coupling (CLC) delay that quantitatively agrees with the time-dependent Schrödinger equation (TDSE) [5]. We identify two previously unknown mechanisms regulating the associated attosecond dynamics, namely the enhanced confinement of the trapped wavefunction with increasing electronegativity of the atoms and the decrease of the coupling strength among the photoemitted partial waves with increasing symmetry.

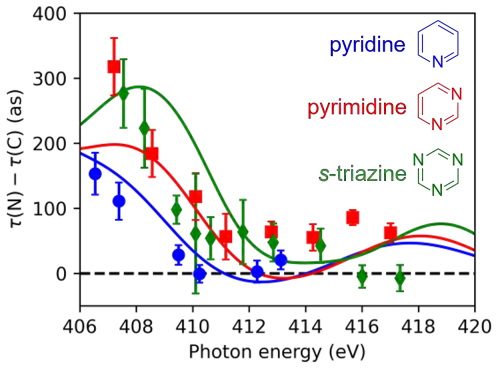


Figure 1. Measured relative photoionization time delays (markers with error bars indicating the 95% confidence intervals) compared to calculated N1s photoionization time delays (lines) including CLC delays for pyridine (blue circles), pyrimidine (red squares), and s-triazine (green diamonds).

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Double-blind holography of ultrashort free-electron laser pulses

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Synopsis Single-shot characterization of free electron laser (FEL) pulses is essential for studying the dynamics of matter at extreme time scales. In this study, conducted at the FL26 beamline of FLASH2, we present an all-optical scheme based on double-blind holography to reconstruct the temporal profile of ultrashort single-mode self-amplified stimulated emission (SASE) FEL pulses.

Ultrashort light pulses provide the temporal resolution necessary to track and control electron dynamics in real time. Recent experiments have demonstrated the generation of few-femtosecond or attosecond pulses at free-electron laser (FEL) facilities [1, 2]. Conducting time-resolved experiments with such ultrashort pulses requires precise characterization of shot-to-shot spectral content, coherence, and pulse duration, along with accurate delay tagging.

In this work, we introduce an all-optical scheme for single-shot temporal characterization with few-femtosecond resolution delay tagging of FEL pulses, based on the concept of double-blind holography (DBH) [3,4]. The experiment was conducted at the FL26 beamline of FLASH2, using a self-amplified spontaneous emission (SASE) FEL source producing pulses of about 10 fs. This source was synchronized with a table-top high-order harmonic generation (HHG) setup [5]. The energy-dispersed FEL and HHG beams were partially overlapped in a quasi-collinear geometry (0.5 mrad offset) on a two-dimensional (2D) camera, where their single-shot interferogram was recorded. A 2D reconstruction algorithm was employed to retrieve the spectral phase of both pulses, enabling their full temporal characterization. Figure 1 illustrates a typical reconstructed profile. The interference patterns were also analyzed in terms of single-shot delay tagging. Our benchmarking of the FL26 beamline's timing tool demonstrated an accuracy better than 6 fs root mean square. The method presented here is suitable for both source diagnostic and spectroscopic applications at the atto-to-femtosecond time scales.

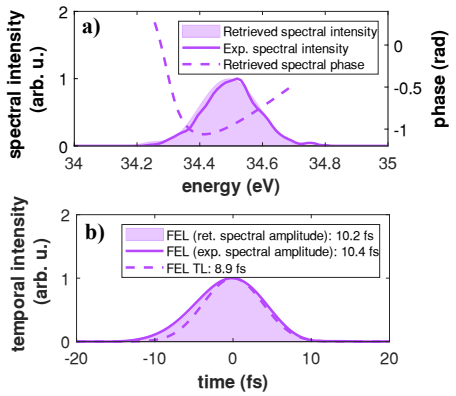


Figure 1. (a) Single-shot FEL pulses: measured spectrum (purple full line), retrieved spectrum from the 2D reconstruction (shaded area), and reconstructed spectral phase (dashed line). **(b)** Reconstructed FEL temporal profile, using either the measured (full line) or the retrieved spectra (shaded area), compared with the transform-limited pulse profile (dashed line).

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Attosecond plasma lens

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Synopsis We introduce the first lens enabling the focusing of attosecond pulses, which is achieved by utilizing refraction due to free electrons in a hydrogen plasma. The concept is demonstrated for attosecond pulses with central photon energies of 20 eV and 80 eV, showing the tunability of wavelength and focal length. Low absorption losses make this concept interesting for photon-demanding applications.

Focusing ultrashort laser pulses using refractive lenses is challenging due to their broad spectral bandwidths. Using refractive elements for attosecond pulses in the extreme-ultraviolet (XUV) range is further impeded by the fact that all materials in this spectral range are highly absorbing. Curved mirrors are commonly employed for focusing attosecond pulses, yet they present notable drawbacks such as low reflectivities and susceptibility to degradation.

Here we propose and demonstrate a refractive plasma lens for focusing attosecond pulses. The refractive index n of a plasma is given by:

$$n = \sqrt{1 - \omega_p^2/\omega^2},$$

where ω_p and ω are the plasma and light field frequencies, respectively. ω_p depends on the free-electron density, n_e. For $\omega_p < \omega$, the refractive index has real values below unity. In this case, a concave-shaped plasma can be used to focus XUV light.

Plasma was generated from hydrogen molecules using a capillary discharge source [1] consisting of a 5 cm long sapphire block with a 300 µm diameter channel. After ignition, the plasma temperature becomes inhomogeneous due to energy exchange with the relatively cooler capillary walls, leading to the formation of a parabolic plasma density profile [2]. Notably, this lens has a tunable focal length, achieved by varying the hydrogen pressure inside the capillary.

We experimentally demonstrate that the plasma lens focuses attosecond pulses centered at 20 eV from an initial $1/e^2$ diameter of 193 µm down to 81 µm (see Fig. 1(a)). It is further shown that an attosecond pulse centered at a higher photon energy of 80 eV with a 25 eV bandwidth can be focused, see Fig. 1(b). In this case, the transmission of the lens was 84 %. Numerical simulations showed that moderate temporal stretching from 103 as to 126 as was induced by the lens.

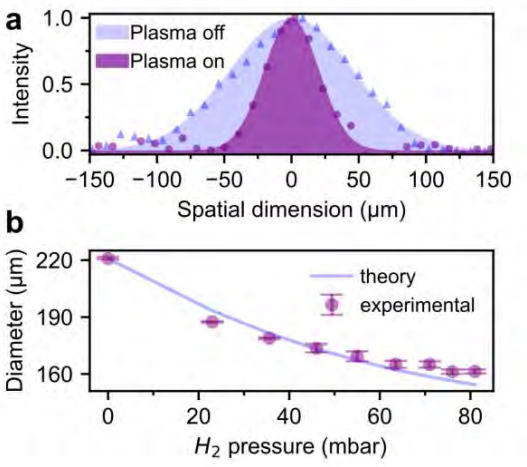


Figure 1. (**a**) Beam profiles of attosecond pulses centered at 20 eV, as measured at 17 cm after the lens using an H_2 pressure of 73 mbar (circles) and an evacuated capillary (triangles). The solid curves show Gaussian fits. (**b**) Diameter of a beam at 80 eV (circles) as a function of the hydrogen pressure inside the lens compared with simulated beam diameters (solid curve).

When used in combination with HHG, the attosecond plasma lens defocuses the co-propagating NIR light, reducing the on-target intensity to 10^8 W/cm^2. This eliminates the need for thin metal filters and further enhances the transmission of HHG beamlines, providing novel opportunities for photon-demanding applications.

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Resolving attosecond time-delay in strong-field ultrafast process of valence-shell electrons of argon

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Synopsis We propose a novel approach to accurately determine the time either photoionization or rescattering processes of valence subshell electrons of atom. This approach is applied to understand and explore the dynamics of wave-packet motion of valence subshell $3s^23p^6$ electrons in argon following strong-field ionization and rescattering.

Real-time probing of electron dynamics attract substantial attention in ultrafast science and technology. We combine an *ab initio* time-dependent density-functional theory (TDDFT) [1] with Bohmian mechanics (BM) [2] to accurately determine the time either photoionization or rescattering processes of valence subshell electrons of many-electron atomic systems in intense laser fields.

Based on the TDDFT-BM scheme, the dynamics of wave-packet motion of valence subshell $3s^23p^6$ electrons resolved by orbital angular momentum orientation in argon following strong-field ionization and rescattering are clarified. The HHG of valence subshell electrons can be calculated accurately and efficiently by solving the *self-interaction-free* TDDFT approach. The TDDFT provides an alternative approach for nonperturbative treatment of multiphoton processes of many-electron systems in intense laser fields. In Figs. 1(a), we present the HHG of valence subshell electrons for individual $3s$, $3p_0$, $3p_1$, and $3p_{-1}$ orbital orientations of Ar atom, respectively. In Figs. 1(b), we perform the calculation of time profiles of the harmonic 5 (H5). The time profile for a specific harmonic can be obtained from a cross section of the time-frequency spectra at a given frequency. The time profile of each harmonic at the peak intensities suggests the emission times of such harmonic during the interaction of the atom with the laser field. It is seen that the time of the maximum peaks of dominant emission of H5 is different for individual $3s$, $3p_0$, $3p_1$ and $3p_{-1}$ orbital electrons. This result indicates that time delay may occurs in laser-driven rescattering processes for individual subshell electrons.

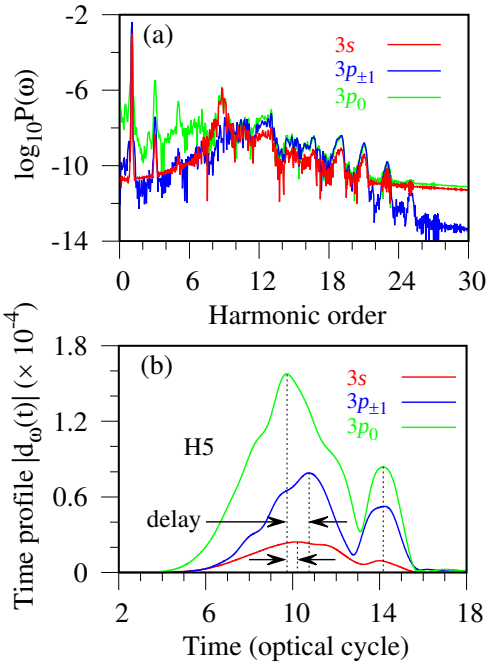


Figure 1. (a) HHG and (b) Time profiles of the harmonic 5 (H5) of valence subshell electrons for individual $3s$, $3p_0$, and $3p_{\pm1}$ orbital orientations of Ar atom, respectively. The dotted lines indicate the maximum peaks of the time profiles of the H5 as shown in (b).

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Exploring the effects of pulse duration on high-order harmonic generation with tunable pulse post-compression

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Synopsis We investigate how the driving laser pulse duration and intensity affect high-order harmonic generation (HHG) in an argon gas jet. Using a bulk multi-pass cell post-compression setup, we deliver tunable pulse durations without changing other generation parameters. Simulations of HHG conversion efficiency (CE) closely match experimental results. Short pulses yield higher CE at high intensities, while long pulses optimize CE at low intensity. These findings show that CE depends on the interplay between the single-atom response and phase-matching.

Coherent extreme ultraviolet (XUV) radiation can be produced via high-order harmonic generation in gases, an inherently inefficient process. Improving conversion efficiency (CE) is essential for high-flux XUV applications such as coincidence spectroscopy [1] and coherent imaging [2]. Previous models suggest that short driving pulses enhance the CE [3]. Experimentally investigating this requires varying the Fourier-transform-limited pulse duration independently of other parameters, such as the focusing geometry.

To address this challenge, we employ a pulse post-compression scheme based on a bulk multi-pass cell [4] to tune the pulse duration of an ytterbium laser. We then generate high-order harmonics in an argon gas jet. Figure 1a) presents the experimentally measured yield of the 25th harmonic for pulse durations 45 fs, 57 fs, and 180 fs as function of laser intensity. At low intensities, longer pulses lead to higher yield but require higher pulse energy. Shorter pulses reach higher intensities, where we observe yield saturation, and thus maximum CE. This maximum is indicated by the red cross in Figure 1b).

We perform simulations of HHG by solving the time-dependent Schrödinger equation and propagation equations [5] and observe good agreement with experiments. Figure 1b) shows the pulse duration at which the simulated CE of harmonic 25 is optimized for different intensities. These results emphasize the CE's dependence on the interplay between the intensity-dependent single-atom response and phase-matching.

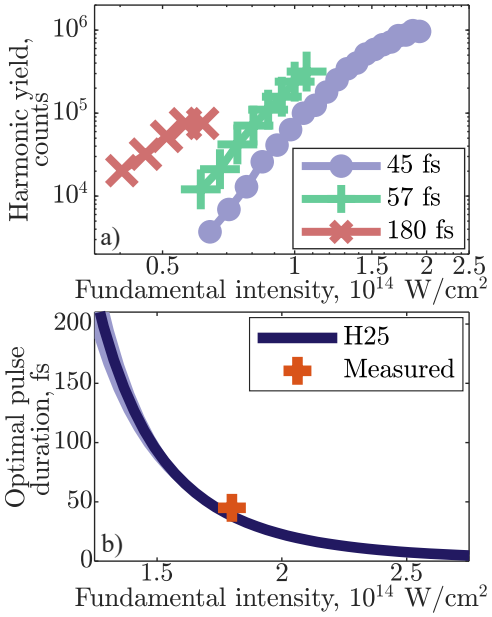


Figure 1. a) Experimental yield of harmonic 25 as function of laser intensity for 45 fs, 57 fs and 180 fs driving pulses. b) Duration at which the simulated CE of harmonic 25 is maximized for different laser intensities. The red cross is the experimentally obtained optimum using a 45 fs driver.

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Ultrafast bright non-Gaussian state of light

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Synopsis We generate bright squeezed vacuum via high-gain parametric down-conversion accompanied by nonlinear Kerr effect. Using single-shot spectral interferometry, we characterize the resulting state of light showing its non-Gaussianity.

Non-Gaussian states of light are of great demand for quantum technologies as they allow universal quantum computation [1] and, lately, they are being explored in attosecond science [2]. Conventional approaches demonstrate the conditional preparation of non-Gaussian states with small mean photon numbers limiting access to nonlinear light-matter interaction. However, we can reach strong light-matter coupling by confining the quantum state of light on the femtosecond timescale. Recently, we generated 25-fs single-mode bright squeezed vacuum (BSV) through an optical parametric amplification (OPA) of vacuum fluctuations [3]. The Wigner function of BSV (Fig. 1a) is centered at zero and features very strong fluctuations of the antisqueezed quadrature, covering extremely large electric-field amplitudes, up to 1V/Å. These large amplitudes of BSV electric field experience strong nonlinear Kerr phase shift. As a result, the BSV Wigner function bends, taking on an S shape and becoming non-Gaussian [4] (Fig. 1b).

Here, we demonstrate the resulting non-Gaussian state. Figure 1c shows the preparation of single-mode BSV at 1600 nm using two nonlinear crystals (OPA I and OPA II). A dichroic mirror (DM) separates the BSV from the pump and reflects it to the second harmonic generation (SHG) crystal. The second harmonic of BSV and the residual 1% of the pump form a nonlinear interferometer. Figure 1d shows the spectral-domain interference fringes, where the single-pulse spectra are sorted according to their integral energy. The position of spectral fringes shifts linearly at energy, which is a clear signature of a nonlinear phase shift appearing in OPA II. As a result, the bright quantum state produced by an unseeded OPA strongly pumped by ultrashort pulses is not only nonclassical, but also non-Gaussian.

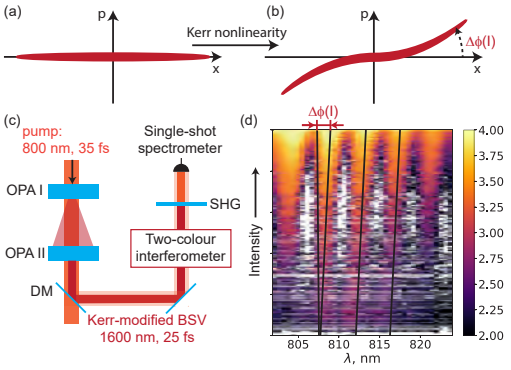


Figure 1. Wigner function of BSV without (a) and with (b) Kerr nonlinearity, respectively. (c) Experimental setup. (d) Post-selected single-shot spectra at different levels of BSV amplitude.

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Enhancement of extreme ultraviolet pulse generation by orthogonally polarized laser-controlled spontaneous emission of Ar atoms

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Synopsis We present a theoretical study of extreme ultraviolet emission generation through the control of spontaneous emission from an excited atomic state. This control is achieved by applying a two-color orthogonally polarized laser field.

We theoretically explore the enhancement of extreme ultraviolet (XUV) generation by orthogonally polarized laser-controlled spontaneous emission of Ar atoms. The three-dimensional time-dependent Schrödinger equation is solved numerically using the generalized pseudospectral method. When applying a strong third harmonic field from a 1030 nm laser, a series of spectral lines emerge in the harmonic spectrum ranging from 13.5 eV to 15.8 eV. These lines relate to transitions without external fields from the ground state to the $3s^2 3p^5 nd$ $(n = 3, 4, 5, \cdots)$ excited states of argon. The emissions arise from the optical free induction decay (FID) of coherent excited states in argon[1, 2]. When a weak multicycle fundamental laser pulse is introduced perpendicularly to the third harmonic field, narrowband XUV emission centered at 14.15 eV is produced, corresponding to the transition from the $3s^2 3p^5 3d$ excited state to the ground state. This configuration strongly suppresses other transitions, as the fundamental laser field modifies the absorption cross-section of other excited states, effectively closing the multiphoton channel.

Our findings demonstrate the potential of orthogonal two-color laser pulses in generating bright, narrowband XUV radiation. This research provides new insights for developing XUV and soft X-ray light sources, offering promising applications in various fields requiring precise and intense XUV radiation.

The advantage of this method is that it offers an effective approach to generate enhanced XUV light sources with specific spatial properties, as the radiation produced by FID maintains similar spatial characteristics to the excitation pulse. By introducing a time-delayed laser pulse (e.g., an infrared laser pulse), the XUV emission can be precisely controlled temporally and spatially. This method enables precise directional control of XUV radiation, as shown in Ref. [2].

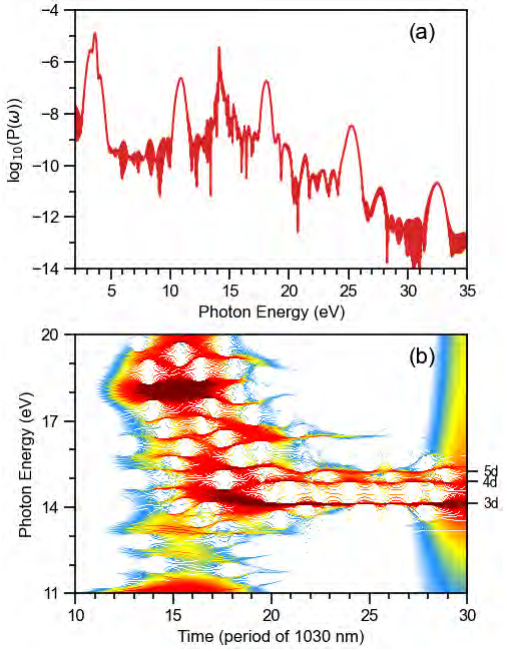


Figure 1. (a) Harmonic spectrum and (b) SST time-frequency spectrum along the direction of 3ω laser field in two-color orthogonally polarized field.

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High harmonic generation from a solid-gas dual-target configuration

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Synopsis We propose a novel dual-target configuration to investigate ultraviolet (UV)-assisted high harmonic generation (HHG). An obvious modulation of quantum paths was observed in the experiment, which has been proved by the theoretical simulations. This work provides a simplified and valid configuration for quantum path control in HHG.

High harmonic generation (HHG) has become a powerful extreme ultraviolet (XUV) source for probing ultrafast properties of matter. When an ultraviolet (UV) light has been added to the infrared (IR) laser field, the UV photon will modulate the ionization rate of high harmonic emission at different times, thereby realizing a selection rule for controlling the contributions of different quantum paths [1,2]. In the majority of UV-assisted gas-phase HHG experiments, a dual-target configuration is employed and both targets are filled with gaseous media. The process of adjusting and optimizing the gas target is a time-consuming one, which in turn increases the experimental complexity in this dual-target structure. Compared to HHG in gases, the experimental setups for solid-state HHG are relatively straightforward, and the requirement for peak laser intensity is less stringent, which facilitates the generation of a broad spectrum of UV light.

For these reasons, we introduce a novel dual-target configuration for HHG, employing a solid target for the generation of seed UV light, followed by a gas target to generate XUV light. By moving the solid target to change the intensity of solid-state harmonics, it was evident that as the solid target approached the focus, the intensity of 21th to 25th order harmonics underwent a decline, and their spectral center depression is deepened as Figure 1 shows. We have successfully observed this kind of spectral splitting in the theoretical simulations. Moreover, time-frequency analysis has revealed that this phenomenon is attributable to quantum path modulation between long and short trajectories emission.

In conclusion, we present a solid-gas dual-target configuration for the quantum path control of HHG in gases. With the addition of delay control elements, our work has the potential to become a promising configuration for precise enhancement of different quantum paths in HHG.

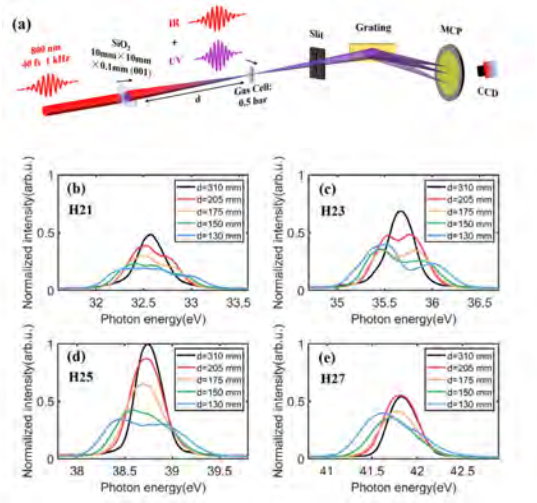


Figure 1. (a) Schematic of the experimental setup; (b-e) HHG spectra of 21th to 27th order harmonics at different dual-target distance *d*.

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Laser-Assisted X-ray Second Harmonic Generation

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Synopsis We theoretically demonstrate x-ray second harmonic generation combined with laser-driven high harmonic generation from core electrons in gas-phase atoms. Simulations confirm x-ray second harmonic generation via x-ray two-photon excitation of core electrons, followed by optical laser interaction. This technique may serve as a powerful tool for probing core-electron dynamics, generating attosecond x-ray pulses, and integrating laser-driven attosecond technology with nonlinear x-ray methodologies enabled by x-ray free-electron lasers.

Attosecond technology based on laser-driven high harmonic generation (HHG) is a powerful tool for ultrafast studies in atomic, molecular, and condensed matter physics [1], but its efficiency in the x-ray regime is limited. Meanwhile, x-ray free-electron lasers (XFELs) have enabled second harmonic generation (SHG) in the x-ray regime [2, 3]. However, achieving x-ray SHG in randomly oriented gas-phase systems remains challenging due to symmetry constraints. To address this, we propose HHG-XSHG, a method combining laser-driven HHG with x-ray SHG (Fig.1a). This approach leverages optical laser and x-ray pulses to drive nonlinear interactions in core electrons, generating HHG-XSHG. By enabling x-ray SHG in gas-phase systems, this approach expands the potential of attosecond spectroscopy and nonlinear x-ray methodologies.

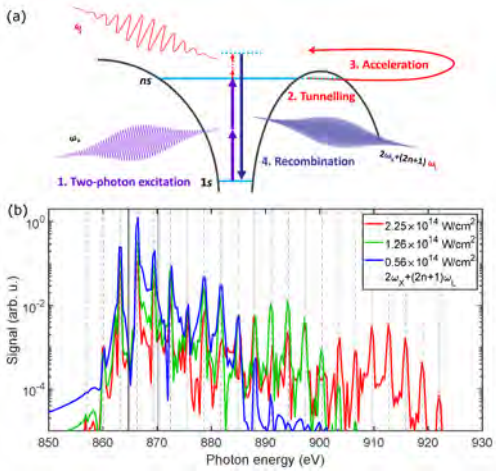


Figure 1. (a) Schematic of the HHG-XSHG process. (b) Simulated x-ray SHG spectra for three optical laser intensities with the x-ray at two-photon resonance.

We simulate HHG-XSHG by solving the time-dependent Schrödinger equation with a neon-like model using the single active core electron approximation. To enhance the signal, we exploit two-photon excitation resonance, which boosts HHG-XSHG and provides insights into core-electron dynamics. Fig.1b presents the simulated HHG-XSHG spectrum in the photon energy range between 850 and 930 eV for three near-infrared (800 nm) laser intensities with an x-ray photon energy of 433 eV at two-photon excitation resonance of neon atoms, revealing a broad multi-peak structure, an extended plateau, and a sharp cutoff—features reminiscent of HHG [4]. According to selection rules, only harmonics involving two x-ray photons and an odd number of optical laser photons contribute to the second harmonic signal.

In summary, we theoretically demonstrated the feasibility of HHG-XSHG through detailed simulations of gas-phase atoms. This mechanism integrates strong-field-driven HHG with x-ray SHG, bridging attosecond techniques from HHG with XFEL-driven nonlinear x-ray methods. Our approach enables the generation of element-specific attosecond x-ray pulses and facilitates the study of ultrafast core-electron dynamics and nonlinear interactions with high temporal and spectral resolution.

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High repetition rate, high average power XUV sources based on High Harmonic Generation

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Synopsis We present our recent work on power scaling of our high harmonic generation based XUV source using the Class 5 Black dwarf laser system at 17 W and 50 W average power as well as the next steps to scale our source to operate with 200 W and even 500 W average power of the femtosecond driver laser. The ultimate goal within our MEGA EUV collaboration project is to deliver up to 1 mW of XUV flux at the industrial relevant wavelength of 13.5 nm.

High harmonic generation (HHG) sources [1] provide fully coherent laser-like radiation in the Extreme-Ultraviolet (EUV/XUV) and soft X-ray spectral range with femto- or attosecond pulse duration. These sources are ideal metrology tools in scientific or industrial applications: The short wavelength allows very high spatial resolution in scattering or imaging of nanostructures created by state-of-the-art microchip EUV lithography. Their spectral bandwidth and ultrashort pulse durations are very useful in ultrafast spectroscopy to investigate the fast reactions occurring in solar cells, catalysis or to observe directly the carrier dynamics in superconductors or next generation semiconductors. For all these applications, HHG sources with high repetition rates and high average powers are required to minimize the acquisition times, leading to a fast turnaround in sample characterization. At the same time, a high robustness and stability of the source is needed for low-maintenance operation with minimal down times. Based on robust high-power femtosecond Ytterbium (Yb) laser systems together with the multipass cell pulse post compression technology [2], Class 5 Photonics is developing the Moonlander high harmonic generation source to fulfill these requirements.

In our contribution, we will present the current status of our HHG source driven by the Class 5 Black Dwarf laser, providing 17 W average power at 100 kHz repetition rate and a pulse duration of 29 fs. We measured an XUV output power of 1.4 µW for argon target gas and 2.8 µW for krypton target gas within the aluminum filter window (22-75 eV). The measured key parameters are shown in Figure 1.

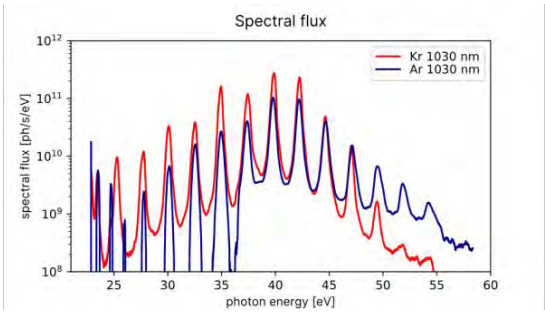


Figure 1. Measured XUV spectral photon flux using a Class 5 Black Dwarf with 17 W average power at 100 kHz repetition rate

We will also show first results with a driver at an average power of 50 W at much high repetition rate of 750 kHz. Our next steps are the demonstration of stable operation of our HHG source for driver lasers at 200 W and 500 W average power within the joint research project MEGA-EUV with DESY, University of Hamburg and Amphos. We will discuss the technical challenges and prospects in the power scaling of these XUV sources.

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Apparatus for attosecond spectroscopy driven by dual-color few-cycle lasers

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Synopsis We have established a versatile attosecond beamline at the Synergetic Extreme Condition User Facility (SECUF) [1]. Isolated attosecond pulses are generated via high harmonic generation (HHG) using polarization gating technique. The iterative retrieval of attosecond streaking spectrogram indicates a pulse duration down to 60 as.

Ultrashort laser pulses have made it possible to explore the intrinsic properties of materials governed by the dynamics of electrons that are usually evolve in femto- and attosecond time scales. HHG delivering attosecond pulse duration has been demonstrated as a robust table-top coherent light source.

In SECUF, we established an attosecond beamline to carry out attosecond metrology and spectroscopy[2-3]. The apparatus is equipped with an dual channel OPCPA laser system that outputs few-cycle and carrier-envelope phase stabilized pulses with central wavelengths at 900 nm (IR channel) and 2100 nm (MIR channel). Each channel outputs pulses with 1kHz repetition rate and 7 mJ energy per pulse. The pulse duration is 7 fs for IR channel and 25 fs for MIR channel. In simultaneous mode, both channels delivers about 3.5 mJ pulses with other parameters similar as in independent mode and potentially allow for coherent synchronization. The apparatus are equipped with a time-of-flight (TOF) spectrometer to characterize the photoelectron energy, and a high-resolution plane grating spectrometer to analyze the HHG spectrum.

The laser beam passes through a 0.18 mm thick quartz plate and a quarter-wave plate. It results in a near-linearly polarized pulse with a time window approaching a single optical cycle[]. The pulse is then focused by a concave mirror onto a 10-mm long gas cell and interact with Ne gas from a pulsed piezo valve synchoronized with the driving laser. The HHG is filtered by Zr foil and focused to the TOF spectrometer, recombined with a small fraction of the fundamental laser. The characterization of the temporal profile is done using FROG-CRAB (Frequency Resolved Optical Gating for complete reconstruction of attosecond bursts) technique. The retrieved pulse duration is 59 as.

The attosecond apparatus driven by dual-color OPCPA laser act as an advanced platform open for global users to study the fundamental dynamic processes using photoelectron or transient absorption spectroscopy, especially promising in extending these studes to short wavelength such as in the "water window" region.

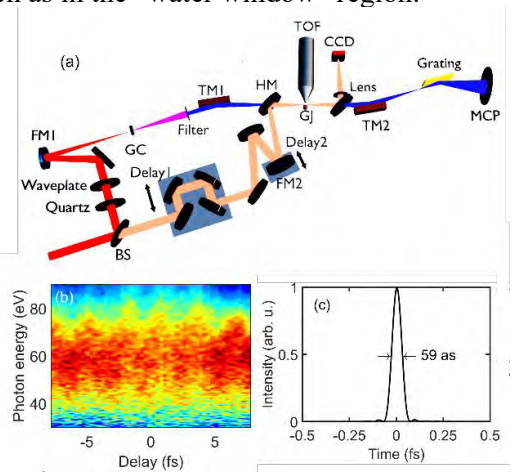


Figure 1. (a) Experimental setup. (b) Measured attosecond streaking spectrogram. (c) Retrieved attosecond pulse profile.

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Stimulated Raman transitions below and above the ionization threshold of neon using a free-electron laser

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Synopsis We investigate stimulated Raman transitions in neon, by measuring metastable excited states. These excited states are prepared by exciting through an intermediate state and stimulated emission to a lower-lying state, using a coherent combination of photon frequencies, generated by the free-electron laser at FERMI. The intermediate states are found both below and above the ionization threshold.

Theoretically predicted Raman transitions via continuum states [1] have recently become experimentally accessible at research facilities such as FERMI, which offers two-colour XUV pulses.

Figure 1 shows the experimental scheme; using a combination of harmonics of the scannable free-electron laser seed $\hbar\omega$, we are able to drive Raman transitions (resonantly and non-resonantly) in neon. The neon atom is excited from its ground state $|0\rangle$ to an intermediate state $|1\rangle$ using $H_i = i\hbar\omega$, and subsequently stimulated to deexcite to $|2\rangle$ using $H_j = j\hbar\omega$; $E_2 - E_0 \approx (i-j)\hbar\omega$. From thence, it decays to metastable states, which are measured by our detector [2]. We focus on the combinations H_5H_1 and H_9H_5, and explore these and other possible excitation pathways using *ab initio* calculations [3].

The analysis is complicated by the presence of low-level background harmonics, which may lead to excitation pathways competing with the desired ones: a Raman transition induced by H_5H_1, may be masked by direct transitions driven by H_4.

Figure 1 also shows the recorded spectra. The peaks labelled $A_{1,2}$ are tentatively assigned to one-photon excitation of $2p^{-1}$ 3d states by H_4; those labelled $B_{1,2}$ to Raman transitions to $2p^{-1}$ 4p using H_5H_1; that labelled C intermediate excitation of $2s^{-1}$ 3p via H_9. The last peaks coincides with, and possibly hides, the Raman transition driven by H_9H_5. Further analysis is necessary to establish these assignments conclusively.

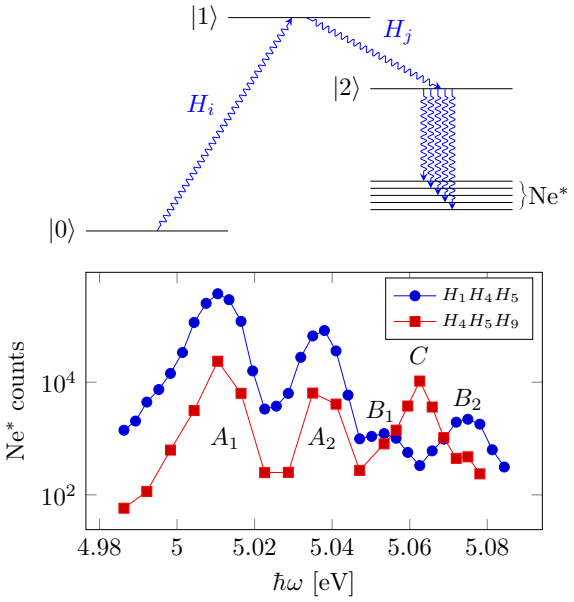


Figure 1. *Top*: Experimental scheme; see main text. *Bottom*: Yield of metastable Ne*, as a function of the seed laser photon energy $\hbar\omega$, for various combinations of harmonics.

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Attosecond pulses produced by a few-cycle laser

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Synopsis High-order Harmonic Generation (HHG) in noble gases with few-cycle carrier-to-envelope phase (CEP) controlled near-infrared (NIR) pulses exhibits temporal properties that cannot be explained solely by the well-established-"three-step" model. This study establishes the connection between the experimental characterization of the attosecond pulse train (APT) properties and the theoretical modelling of dynamical phase-matching.

High-order Harmonic Generation (HHG) in noble gases is a coherent source of attosecond (10^{-18}s) light pulses in the eXtreme Ultra-Violet (XUV) whose spectral properties can be measured by a cross-correlation technique (RABBIT, Reconstruction of Attosecond Beatings By Interference of Two-photon transitions). This technique relies on the interference of electron wave packets (EWPs) emitted when atoms absorb APTs in the presence of a near-infrared (NIR) laser field [1]. When the APTs are generated by a laser containing only a few optical cycles, the interference pattern strongly depends on the temporal characteristics of the attosecond pulses [2]. Here, we investigate the effect of phase-matching of high-order harmonics on the temporal properties of the APTs and its consequences on photoionization in a dressing field.

We performed experiments with femtosecond (~6 fs) near-infrared pulses of stable and controllable carrier-to-envelope phase (CEP). The CEP is manipulated to generate APTs with either two or three attosecond pulses, which are sent to an experimental chamber, where helium is ionized. The EWPs are imaged with a three-dimensional momentum imaging spectrometer. RABBIT spectrograms, recorded by scanning the delay between the XUV and the NIR exhibit complex interference patterns, which change with the CEP as depicted in Figure 1.

Many of the spectral features can be explained solely by accounting for the microscopic description of HHG, often referred to as the three-step model [3], but not all. We propose an interpretation of the experimental results using a one-dimensional analytical model describing the macroscopic dynamics of HHG driven by few-cycle lasers. We show that in some cases, the generation of high harmonic orders can be restricted temporally to the falling/rising edge of the pulse while the generation of low orders takes place over a longer time. This leads to different interference patterns in the corresponding RABBIT interferograms, at high energies. The results are currently confronted with a complete, three-dimensional model including laser reshaping effects in the gas.

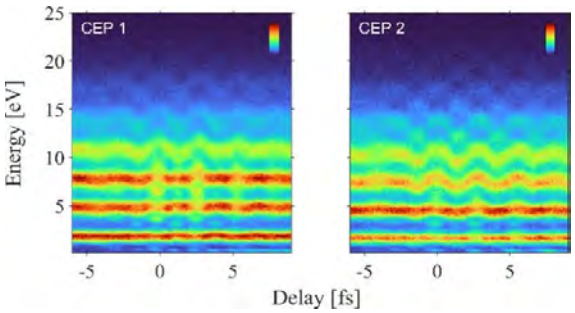


Figure 1. Photoelectron counts as a function of the kinetic energy and of the XUV-NIR delay for two different CEP values. Only electrons photoemitted upwards by the comb of harmonics are shown.

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High harmonic generation from a high-repetition-rate liquid plasma mirror

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Synopsis High-harmonic radiation can be generated when an ultra-intense laser pulse is reflected on the surface of an over-dense plasma. Here we demonstrate high-hramonic generation (HHG) from a flat liquid-sheet plasma mirror at kHz repetition rates, accesing both the coherent wake emission (CWE) and relativistic oscillating mirror (ROM) regimes. This advancement enables high-repetition-rate attosecond light sources and opens new avenues in ultrafast laer-matter interaction research.

High-harmonic generation (HHG) from plasma mirrors occurs when an ultra-intense laser pulse is reflected from a dense plasma surface. This process can produce attosecond extreme ultraviolet (XUV) radiation with relatively high conversion efficiency. However, solid targets are destroyed at each laser shot, preventing long data acquisition.

Here we employ a liquid sheet plasma mirror made of propylene glycol and propylene carbonate, which is self refreshed by itself. It provides an optically flat surface at high repetition rates. By thightly focusing ultrashort pulses onto the liquid jet at 1 kHz repetition rate, we generate harmonic spectra in both the coherent wake emission (CWE) and relativistic oscillating mirror (ROM) regimes [1]. In the CWE regime, we observes harmonic orders up to the plasma frecuency cutoff.

By increasing the laser intensity to relativistic regime and optimizing prepulse for scale-length control, the harmonic spectrum up to 51.1 eV, confirming that the relativistic oscillating mirror mechanism has been accessed. These experiments show that a properly timed prepulse control over the plasma gradient, so ROM happens.

Our results indicate that a high-repetition-rate liquid target can sustain intense laser-plasma interactions continuously. This capability paves the way for bright attosecond pulse sources ideally suited to strong field physics and time-resolved spectroscopy.

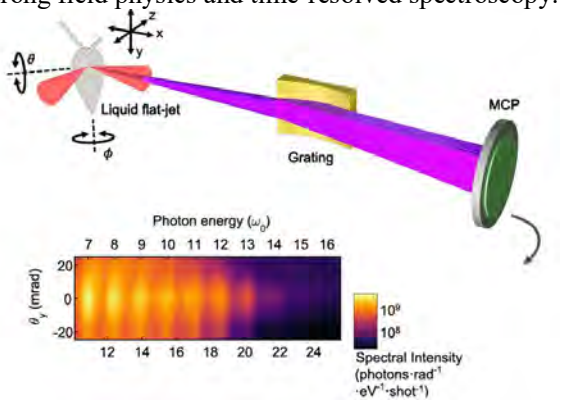


Figure 1. Schematic diagram of the experimental setup and a CWE HHG spectrum generated from a liquid target.

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Carrier-envelope phase-stabilized sub-10-fs multiple-plate compression for MHz isolated attosecond pulses

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Synopsis We demonstrate MHz-repetition-rate operation of a CEP-stabilized 2.7-cycle multiple-plate compressor, which delivers an average power of 47 W. Short-term CEP fluctuations were measured to be 233 and 262 mrad for uncompressed and compressed pulses, respectively. This few-cycle driver source will enable generation of much brighter isolated attosecond pulses.

Increasing the repetition rate and average power of few-cycle CEP-stabilized intense pulses is a direct way to generate brighter isolated attosecond pulses. A promising approach is post-compressed Yb laser pulses. A few studies have recently reported few-cycle CEP-stabilized intense pulses with gas-based hollow-core fiber (HCF) [1] or multi-pass cell (MPC) [2] compressors at 100 kHz. Solid-based technique, multiple-plate compression (MpPC) [3], offers another powerful alternative that is simpler, more compact, and lower-cost with the capability of a mono-cycle duration [4]. However, to the best of our knowledge, the preservation and impact of an MpPC on CEP stability have not been reported.

Here, we demonstrate CEP-stabilized sub-10fs pulses compressed using a two-stage MpPC at a MHz repetition rate. The CEP-stabilized pulses from Yb:KGW regenerative amplifier (80 μJ, 1 MHz, 80 W) were post-compressed from 190 to 9.4 fs. The first compressor shortened the duration from 190 to 27 fs with 96% throughput. The second compressor achieved 47-W, 9.4-fs pulses with 66% throughput after a spatial filter. This duration corresponds to 2.7 optical cycles. The power fluctuation over 3 hours remains at only 0.2% without optical or thermal damage. This high stability suppresses the intensity-CEP crosstalk [5].

The CEP stability was characterized with an *f*-2*f* interferometer with 3-shot averaging. To stabilize the CEP, 5% of the initial pulses were used for an in-loop *f*-2*f* interferometer after the amplifier exit. Figure 1(a) compares the long-term CEP stability of uncompressed and the 2.7-cycle pulses with an out-of-loop *f*-2*f* interferometer over 2 hours. No fatal phase jump was observed, but the root mean square (RMS) increased from 220 to 404 mrad. As this increase is likely attributed to slow drift, we measured short-term stability (Fig. 1(b)). The short-term RMS of the 2.7-cycle pulses achieved 262 mrad, which exhibits less degradation compared to the 100-kHz operation of few-cycle HCF [1] or MPC [2]. Our results mark a significant milestone toward highly stable isolated attosecond pulses at a MHz repetition rate.

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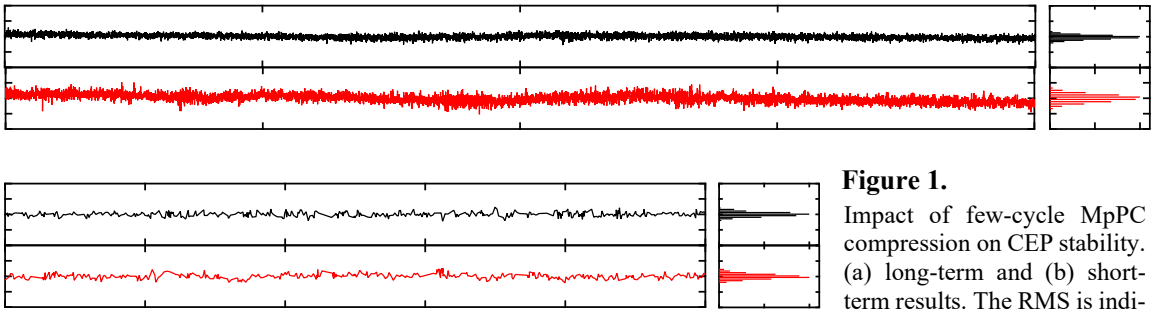


Figure 1.
Impact of few-cycle MpPC compression on CEP stability. (a) long-term and (b) short-term results. The RMS is indicated in the histograms.

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Improving absorption measurements in attosecond XFEL experiments using machine learning

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Synopsis We present the first implementation of machine learning to improve absorption measurements in attosecond XFEL experiments. We have developed an artificial neural network (ANN) that accurately predicts the spectral shape of attosecond X-ray pulses and can be applied in real experiments to enhance results. This approach provides an efficient solution to overcome the limitations of XFELs caused by their stochastic nature.

X-ray free electron lasers (XFELs) generate bright, coherent, tunable, and ultrashort X-ray pulses but with inherent stochastic fluctuations. Even for attosecond pulses generated by the enhanced SASE methods there remain shot-wise fluctuations in intensity and spectrum. These fluctuations make X-ray absorption spectroscopy measurements challenging. One solution is full characterization of each X-ray pulse, retaining only those with desired properties. However, this approach is costly, time-consuming, and not always practical. Machine learning methods such as ANNs offer a fast, low-cost alternative by leveraging incidentally recorded parameters to predict pulse properties. Several attempts have already been made, predicting the central pulse energy [1] and the shape of the spectrum [2]. To our knowledge, ANNs are not systematically used in any beamline at XFEL facilities. Here, we demonstrate a method that uses ANNs to improve the measurements of absorption with attosecond X-ray pulses.

We measured the absorption of biaxially-oriented polyethylene terephthalate (BoPET, Mylar) near the O K edge using attosecond pulses generated in the XLEAP mode of LCLS [3] where the finite bandwidth makes it a challenge to measure accurately the absorption in the wings of the spectrum. We then predicted the incident spectra when measuring transmission and used the predictions to calculate the absorbance on Mylar (see Figure 1). Before the first peak (photon energy < 529 eV), we expect the absorption to be approximately flat. The machine learning method corrects the shape of the spectrum that matches the synchrotron data, while accurately preserving peak positions. The synchrotron data has a wider peak because the spectrometer has a lower resolution.

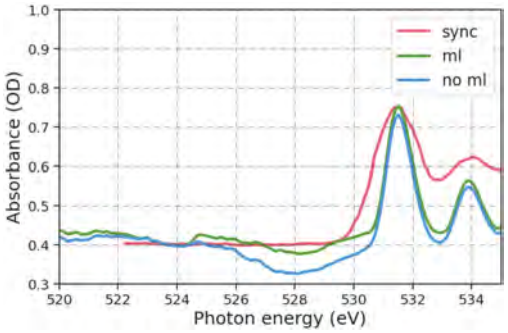


Figure 1. Implementation of machine learning method on the calculation of absorbance of Mylar. The red line shows the data measured by the synchrotron [4]; the green and blue lines represent the Mylar absorbance calculated using the machine learning and the traditional method respectively.

Our model achieves accurate predictions even when trained on a small subset of experimental data, highlighting its potential to perform efficient X-ray characterization in high repetition-rate XFELs and can be extended to complex experiments such as time-resolved spectroscopy.

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Using AttosecondRayTracing to design a beam transport setup for attosecond pulses from surface plasma relativistic HHG

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Synopsis We present the first public release of the AttosecondRayTracing code; an open source simulation tool for preliminary design and characterization of attosecond transport beamlines. We also present an example of using the code to design our own refocusing system for relativistic plasma mirror HHG.

When transporting sub-femtosecond pulses, complex spatio-temporal couplings can quickly arise and ruin the temporal pulse profile. While those same problems exist for traditional ultra-short lasers, they are considerably more severe in the cases of attosecond pulses.

Such couplings can be inherent to the type of optics used in the beamline, for instance toroidal mirrors will always generate some degree of spatio-temporal couplings. They can also arise from slight misalignments of optics. Quantifying those couplings is a necessary step in designing an attosecond transport beamline. This can be done using ray-tracing simulations [1,2], where such couplings will manifest as varying delays of rays arriving at the detector. While conventional tools such as Zemax and Code V work well for normal ray-tracing tasks, they are not well suited for tracking the delay of each ray.

For that reason, we introduce AttosecondRayTracing (ART): an open source Python ray-tracing code to greatly simplify preliminary design, analysis and tolerancing of attosecond beam transport setups. Being open source lends the code to modifications and further improvements from the attosecond community.

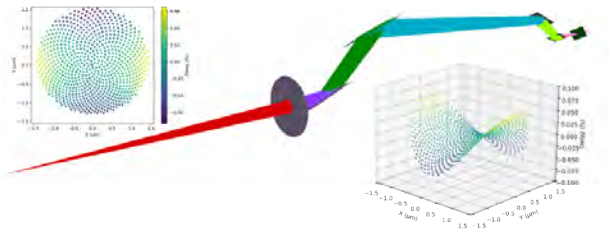


Figure 1. Analysis plots produced using ART for a variant of our refocusing setup.

With ART, designing a beamline is as simple as installing the Python package and creating a configuration file. ART then provides functions to characterize the performance of the setup.

As a practical example of using ART, I will present the various steps in designing our own refocusing beamline. We generate broadband pulses of light using relativistic plasma mirror surface high-order harmonic generation. The bandwidth of the pulses supports attosecond durations once refocused, and the source is inherently chirp-free [3].

The generation process is predicted [4] to be much more efficient than gas based HHG, opening the door to unprecedented intensities for a table-top attosecond beamline. However, the bandwidth and divergence of the source make the design challenging. We measure a 5 octave spectrum, from IR to 35 eV, which makes finding a suitable mirror coating for the beamline quite challenging. The beam is also strongly divergent, with a FWHM divergence of around 50 mrad

Our setup has been designed using ART to work well within our constraints. It will refocus around 60 mrad of the source beam, reaching projected pulse durations under 400 as and peak intensities above 10^{16} W/cm². I will present in detail the planned beamline as well as how ART was used for the design process.

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The effect of driving laser properties on the coherent extreme-ultraviolet wavefront from high harmonic generation

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Synopsis This study investigates how the properties of the driving infrared (IR) beam profile influence the generated coherent extreme-ultraviolet (XUV) beam in an argon gas cell. By analyzing the XUV wavefront and spatial distribution, significant dependencies between the IR and XUV beam characteristics are observed. The results help in optimizing harmonic wavefront, which is crucial for improving XUV beam quality and applications.

The XUV radiation, produced through high-order harmonic generation (HHG) has extensive applications in physics, chemistry and biology [1]. Coherent XUV can be generated from different media, such as gaseous atoms by intense IR beams. Among other experimental parameters, the driving laser characteristics largely determine the spatial and temporal properties of the XUV beam. Plach *et al.* showed that fine-tuning the driving wavefront is essential for optimal spatial and temporal quality of the harmonics [2].

The motivation behind this study was to identify the effect of the IR beam profiles on the harmonic beam in an argon gas cell, by examining the XUV wavefront and spatial profile. The conducted experiment took place on the SYLOS Long beamline at ELI-ALPS [3], using the SYLOS 2 laser system with long Rayleigh range (200 mm). Harmonics were generated in argon gas, in a 325 mm long cell, with a pressure of 1.05 mbar. Different aberrations, such as 0° or 45° astigmatism, horizontal and vertical coma, were systematically added to the driving IR beam by a deformable mirror. With the HASO EUV sensor, the XUV intensity profiles, phase profiles, and the Zernike polynomials were measured [4].

The spatial profile of the XUV exhibits discernible changes in spatial intensity and orientation depending on the alterations of the astigmatism of the driving IR beam (see Fig 1. a)). These effects were also supported by similar changes in the Zernike polynomials as well (see Fig 1. b)). In contrast to the observations in [2], our results show that the XUV beam profile shape follows the spatial profile of the most intense part of the laser focus. Overall, these findings contribute to understanding the link between the generating conditions of HHG and the wavefront of the harmonics that may lead to improvements in refocusing the harmonics helping to achieve nonlinear XUV-XUV pump-probe measurements.

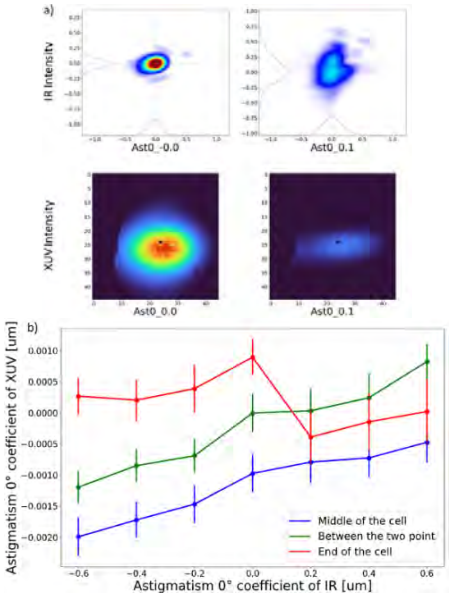


Figure 1. a) The intensity profiles for both IR (top) and XUV (bottom) beams with or without added 0° astigmatism. **b)** The XUV astigmatism 0° coefficient as the function of same coefficient of the IR beam in three different focus point position (with respect to the gas cell).

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Continuous Relativistic High-Harmonic Generation from a Liquid-Leaf Plasma Mirror at kHz Repetition Rate

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Synopsis We demonstrate continuous relativistic high-harmonic generation (RHHG) for the first time at 1 kHz repetition rate from a micron-scale liquid-leaf plasma mirror target. Precise control of the driving near-single-cycle laser waveform enables the generation of a highly stable XUV beam featuring a quasi-continuous spectrum, expected to correspond to a stable kHz train of energetic isolated attosecond pulses. These results establish liquid-leaf targets as a key technology for the development of high-power attosecond lasers.

Relativistic high-harmonic generation (RHHG) from plasma mirrors is a promising approach for producing intense attosecond pulses, offering efficient conversion of ultra-high peak power laser light into coherent extreme ultraviolet (XUV) and soft X-ray radiation [1, 2]. Until very recently, RHHG has been almost exclusively driven from solid disk targets offering too low shot numbers for practical applications.

The development of liquid-leaf target technology marks a major breakthrough by enabling continuous plasma mirror operation at kHz repetition rates [3, 4]. Liquid-leaf targets, with their optical surface quality and self-replenishing nature, have already been used to demonstrate high-harmonic generation at 1 kHz in the sub-relativistic regime and even RHHG in single-shot mode [5].

In this work, we present the first demonstration of continuous RHHG at 1 kHz from a liquid-leaf plasma mirror [6] driven by waveform-controlled relativistic-intensity near-single-cycle laser pulses. Using an ethylene glycol sheet with a controlled surface plasma density gradient, we manage to produce an XUV beam with highly reproducible spatio-spectral properties over extended periods of time.

As illustrated in Fig. 1, by carefully tuning the waveform of the 1.5-cycle driving light field [7], we can transition from a highly modulated RHHG spectrum to an XUV quasi-continuum, which are expected to correspond to the production of a stable kHz-train of energetic isolated attosecond pulses. These results highlight the critical role liquid-leaf target technology should play in the development of the next generation of high-power attosecond light sources for nonlinear applications.

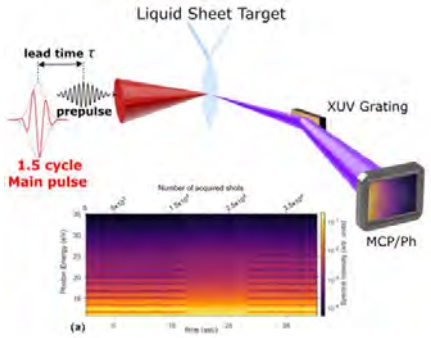


Figure 1. Spatio-spectral detection setup for RHHG from a liquid-leaf plasma mirror driven by relativistic-intensity 1.5-cycle laser pulses at 1 kHz. Inset (a) shows how the continuously acquired RHHG spectrogram can be reproducibly controlled by tuning the driving laser waveform.

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High Harmonic Polarization Shaping Around a Cooper Minimum

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Synopsis High harmonic yield decreases with increasing the driving IR ellipticity, but the polarization response of high harmonic generation remains unclear. For the first time, we resolve how the polarization of high-order harmonics scales with IR ellipticity, revealing significant changes in the tilt angle and ellipticity near the Cooper minimum, reflecting the angular momentum dynamics of the recombination dipole matrix element.

High-order harmonic (HHG) spectroscopy is a novel approach that utilizes the ionization, acceleration, and recombination of photoelectrons, leveraging the short de Broglie wavelength of the recombination electron to enable self-probing of the parent ion with unprecedented spatial and temporal resolution. However, HHG yield drops significantly with increasing ellipticity of the driving field, limiting the feasibility of polarization spectroscopy. This technique is essential for probing angular momentum dynamics, ultrafast electron motion, molecular structures, and chiral effects.

The Cooper minimum (CM) plays a crucial role in HHG and attosecond science [1]. It results from a sign change in the recombination dipole matrix element, causing a suppression in the recombination cross-section and a π phase shift. This additional recombination phase near the CM significantly alters the temporal profile of attosecond pulses [2].

In this work, we use a 76 W, 23 fs post-compressed Yb laser at 100 kHz to generate bright HHG, reaching tens of microwatts when driven by linearly polarized IR pulses [3]. This high-brightness HHG enables full polarization analysis—including helicity, ellipticity, and tilt angle—using an energy-resolved polarimeter [4], even with 1/10,000 transmission. Fig. 1(a) shows our experimental setup for HHG polarization spectroscopy in Ar driven by ε_{IR} = -0.22. We first observe that the helicity of the generated harmonics follows that of the driving IR field, meaning that reversing the IR helicity causes all resulting EUV harmonics to switch accordingly. However, ellipticity and tilt angle vary dramatically with photon energy. As shown in Fig. 1(b), IR ellipticity transfer to HHG is generally weak, but near the CM (~50 eV), angular momentum transfer reaches its maximum. Fig. 1(c) further reveals a sharp tilt angle reversal just before the CM (~48 eV). These findings indicate that the recombination cross-section and π phase shift around the CM significantly alter HHG polarization.

In summary, for the first time, we demonstrate that the CM plays a crucial role in shaping HHG polarization, inducing significant variations in ellipticity and tilt angle. This highlights the potential of high-harmonic polarization spectroscopy as a powerful tool for probing angular momentum dynamics and electronic structure in matter.

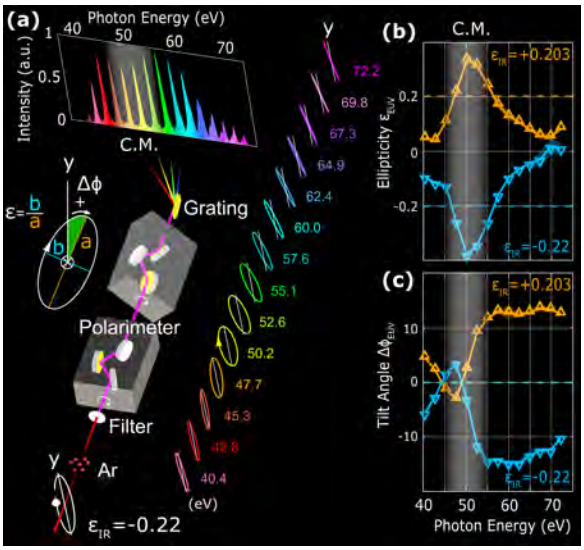


Figure 1. (a) Schematic of high-harmonic polarization spectroscopy, where the IR ellipticity (ε_{IR}) is varied (e.g., set to ε_{IR} = -0.22 and $\Delta\phi_{IR}$ = 0). The resulting HHG from Ar is analyzed using an energy-resolved polarimeter to measure ε_{EUV} and $\Delta\phi_{EUV}$. (b) and (c) show ε_{EUV} and $\Delta\phi_{EUV}$ as functions of photon energy, respectively. C.M. denotes the Cooper Minimum.

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Asymmetric polarization gating for spectral tuning and temporal confinement of high-order harmonics

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Synopsis We study, both experimentally and theoretically, the asymmetric polarization gating (APG) scheme that permits to localize a polarization gate anywhere within the driving pulse. We show [1] that it can be used to achieve simultaneous spectral tuning and temporal confinement of high-order harmonics together with optimizing the XUV flux. By means of SFA simulations, we give detailed insight into the APG scheme, and study in particular the role of short and long trajectories.

Polarization gating [2] is obtained by the combination of two delayed counter-rotating driving pulses with equal amplitudes. It results into a complex field which is linearly polarized only during a short time ("the gate"), where harmonic generation are efficiently produced. Using two pulses with different peak amplitudes [3], allows moving the gate and tuning the slope of the field envelope inside the gate. We delve in this scheme and demonstrate the controllability of the gate position while keeping its duration.

The frequency control is apparent from the qth-harmonic dipole approximated as

$$d_q(t) \propto \mathrm{e}^{-\mathbf{i}(q\omega_0 t + \alpha_q I(t))},$$

where ω_0, $I(t)$, and α_q are the fundamental's central frequency, the driver's intensity profile, and a trajectory-dependent microscopic phase coefficient, respectively [4]. This yields the instantaneous harmonic frequency

$$\omega_q(t) = q\omega_0 + \alpha_q \frac{\mathrm{d}I(t)}{\mathrm{d}t}.$$

The intensity evolution in the gate is approximately linear and results in a linear frequency shift. This shift is controlled through $\frac{\mathrm{d}I(t)}{\mathrm{d}t}$ by adjusting the delay and the ratio of the peak amplitudes of the two pulses.

Experimentally, we use 28fs-long 800nm laser pulses with peak intensity ~ $2 \times 10^{14}\mathrm{W/cm^2}$ for high order harmonic generation (HHG) in Ar gas cell. The polarization gate is created by two quartz plates (a 1mm thick plate splitting the pulse in two pulses delayed by ~ 30 fs and a zero order λ/4 plate making them circularly polarized) with a constant 45°-angle between their optical axes. Rotating them respective to the polarization axis by θ_{MO}, one controls the gate position. The resulting spectra are analyzed by a flat spectrometer (see [1] for details). The generation remains efficient in the plateau within $|\theta_{\mathrm{MO}}| < 20°$. These harmonics exhibit different frequency shifts for short and long trajectories (Fig. 1a), which is due to trajectory-dependent α_q's.

SFA simulations (Fig 1b) provide further insight and reveal, e.g., the contributions of different electron ejections within the gate. These findings, quantifying the spectral and temporal aspects, will be detailed in our presentation.

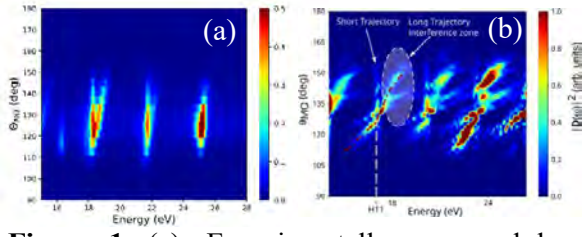


Figure 1. (a): Experimentally measured harmonic spectra as a function of the orientation of the plates (θ_{MO}). It shows the different frequency shifts of long and short trajectories. (b): SFA simulation in high intensity regime (defined by $4 \times 10^{14}\mathrm{W/cm^2}$).

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Isolation of bright soft x-ray attosecond pulses in a highly ionized medium using two-color synthesized driving laser

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Synopsis We propose a robust scheme for generation and isolation of broadband attosecond pulses with $\sim 10^{-7}$ conversion efficiency in 150-320 eV. First, a two-color driving field, synthesized by the fundamental laser and its third-order harmonic with an optimized relative phase, is applied for high-order harmonic generation in the overdriven regime. By optimizing the gas pressure to a high value, the nonadiabatic phase-matching condition is fulfilled. Finally, the ionization-induced wavefront rotation of attosecond pulses results in the full spatial separation in the far field.

The bright, coherent, and intense isolated attosecond pulses (IAPs) provide a powerful tool for investigating electron dynamics on an attosecond timescale. However, implementing all-attosecond pump-probe experiments based on high-harmonic generation (HHG), particularly in the soft x-ray region, remains challenging due to the low energy of IAPs. To enhance the pulse energy of attosecond pulses, three main approaches are typically considered: i) Enhancing harmonic conversion efficiency at the single-atom level. ii) Increasing the driving laser pulse energy or intensity. iii) Achieving the absorption limit under phase-matching conditions.

In this study, we employ a two-color synthesized waveform, comprising the fundamental laser and its third-order harmonic, to enhance short-trajectory harmonic emission at the single-atom level [1]. This two-color synthesized field (with parameters detailed in Table 1) is used to generate high-order harmonics in neon atoms in an overdriven regime, where the initial ionization level exceeds 10%. By optimizing the gas pressure to a high value, the nonadiabatic phase-matching condition for the short-trajectory harmonics is fulfilled. Meanwhile, ionization-induced wavefront rotation of attosecond pulses results in the full spatial separation of successive attosecond bursts [2]. After spatial filtering and integration, two isolated attosecond pulses are obtained. According to our estimated conversion efficiency of approximately 10^{-7}, the pulse energy of these IAPs is expected to exceed nJ under accessible experimental conditions.

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Table 1. Parameters of the synthesized laser field.

$\lambda(nm)$	I(10^{14} W/cm^2)	$\phi_{cep}(\pi)$	τ_{FWHM}(fs)
1600	4.0	0	26.7
533.3	0.2	1.5	26.7

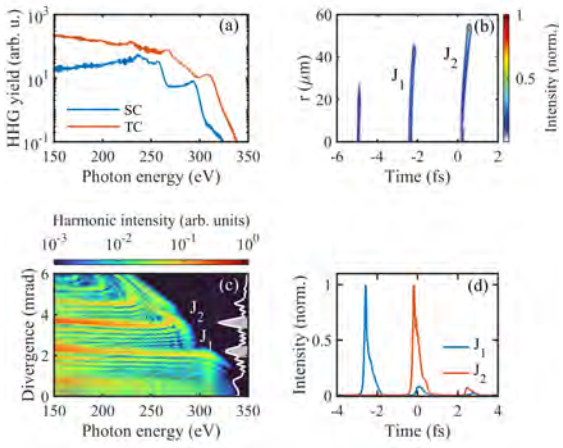


Figure 1. Soft x-ray isolated attosecond pulses generation. (a) High-order harmonics spectrum generated by 1600 nm laser (SC) and two-color synthesized field (TC). (b) Wavefront rotation of attosecond pulses in the near field after macroscopic propagation. (c) Spatial high-order harmonic spectrum in the far field. (d) Isolated attosecond pulses obtained by spatial filtering.

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The generation of photoelectron vortices by chirped attosecond pulses

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Synopsis The generation of photoelectron vortices is studied using a pair of chirped attosecond pulses. We find that a reversible vortex emerges in the absence of a time delay between the pulses, whereas no vortex appears in the chirp-free case. An analytical model is developed to explain and predict the vortex patterns induced by chirped pulses.

Vortices are a pervasive and fundamental phenomenon observed across various scales in nature, ranging from vast cosmic nebulae where galaxies take shape to microscopic quantum systems composed of atoms and molecules. Photoelectron vortices, a recently discovered phenomenon, arise from the interaction between ultrashort laser pulses and atomic or molecular structures. Many studies have shownthat photoelectron vortices with distinct interference characteristics can be generated by adjusting the time delay between a pair of counter-rotating circularly polarized attosecond pulses [1,2].

The advent of attosecond light pulses enables the combination of attosecond-level ultrahigh time resolution and atomic-scale ultrahigh spatial resolution, allowing for the control of extremely ultrafast processes in the atomic and subatomic levels. Chirp is an important parameter of pulsed lasers, and most attosecond pulses generated by current attosecond pulse schemes inherently possess a certain degree of chirp. By studying the interaction of attosecond pulse pairs with hydrogen atoms under different chirp parameter combinations and time delays, we find that photoelectron vortices can still form even at zero time delay. In this case, the electron vortices exhibit a reversible pattern. Furthermore, when the time delay is nonzero, completely reversed electron vortices cannot be obtained by simply exchanging the pulse sequence [3], as shown in Figure 1.

We have investigated these phenomena by extending the theoretical analytical model of electron vortices generated by chirped attosecond pulses and find that the time dependence of the chirped pulse frequency is responsible for these effects. These results demonstrate that the generation and characteristics of photoelectron vortices can be controlled by tuning the chirp of the laser.

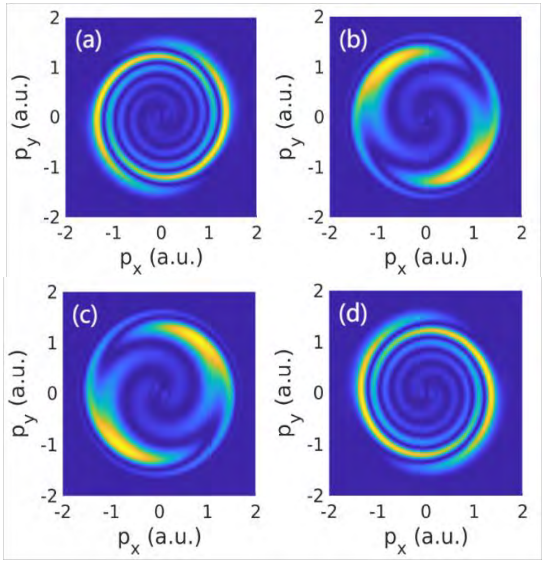


Figure 1. The generation of hydrogen atom photoelectron vortex under different combinations of laser chirp parameters. The laser parameters are as follow: $\omega_0 = 1.52$ a.u., $I_0 = 10^{14}$ W/cm^2, $\tau = 8$ o.c.. The time delay t_d and chirp parameters β for the first and second pulses are: (a) $t_d = 2$ o.c., $\beta = \pm 0.02$, (b) $t_d = 2$ o.c., $\beta = \mp 0.02$, (c) $t_d = -2$ o.c., $\beta = \pm 0.02$, (d) $t_d = -2$ o.c., $\beta = \mp 0.02$.

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Compact attosecond XUV source for the investigation of Raman-induced attosecond electronic coherences

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Abstract: Attosecond XUV pulses enable the observation of ultrafast electron dynamics in molecules with an extreme time resolution. Coherent electron dynamics in neutral molecules can be induced by core-shell impulsive Raman excitation but it requires to reach a peak intensity above 10^{15}W/cm^2. Here, we present first measurements performed using our newly developed attosecond XUV source based on the HHG process in the overdriven regime to generate very intense isolated attosecond laser pulses with pulse energy exceeding 70 nJ for the investigation of impulsive XUV Raman excitation in molecules.

Raman spectroscopy is a powerful method to investigate ultrafast molecular dynamics in the UV-Visible-IR spectral range thanks to the variety of laser systems and parametric amplifiers matching these wavelengths. With the discovery of High-order Harmonics Generation (HHG), coherent attosecond pulses in the XUV and X-ray range have become available [1]. If the attosecond pulse reaches a certain intensity threshold, an impulsive Raman process from a core-shell orbital of the molecule can be triggered leading to the formation of an electronic wavepacket and an ultrafast oscillation of the electronic charge distribution on attosecond to few-fs timescale. The keynote challenge to experimentally trigger such an XUV Raman process is the extremely high intensity requirement, exceeding the values of 10^{15} W/cm^2, which so far has only been achieved at large scale facilities, such as at a Free Electron Laser [2].

To generate very intense attosecond XUV pulses, we have developed a compact table-top XUV source based on the HHG process in the so-called overdriven regime [3]. Departing from a commercial Ti:Sapphire system (SpectraPhysics SpitFire, 800 nm, 12 mJ, 1 kHz, 35 fs), we generated 6 mJ, sub-4 fs pulses based on pulse broadening in a 3.75 m long stretched hollow-core fiber compressor [4]. At the output of the fiber, the spectrally-broadened pulses were then compressed by a set of chirped mirrors and a pair of fused silica wedges before to be refocused into a 10 cm long gas cell filled with argon. To allow for very efficient HHG [3] while keeping the source compact, the intensity of the driving pulse in the gas cell was adjusted to induce a dense plasma leading to a substantial spatio-temporal reshaping of the driving laser pulse while propagating in the gas cell. In this configuration, we demonstrate the generation of a quasi-continuous HHG spectrum extending to 50 eV with a pulse energy above 70 nJ measured after a 200 nm Aluminium filter (see Figure 1). First measurements of the multiphoton ionization of rare gas atoms obtained by refocusing tightly the attosecond XUV pulse into a velocity map imaging spectrometer will be shown.

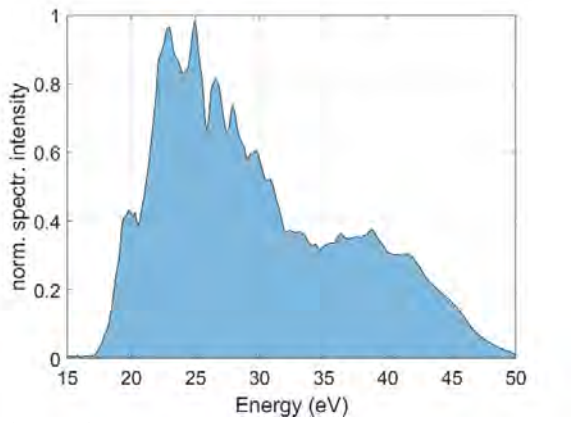


Figure 1. Continuum spectrum in a 10 cm long Argon gas cell.

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Single-stage multipass cell spectral broadening and compression of 90 fs, 300 µJ laser down to 8.5 fs

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Synopsis We report nonlinear broadening and pulse compression in a single multipass cell based on dielectric mirrors. The 90 fs and 300 µJ pulses from industrial-grade laser were compressed to 8.5 fs with 6 W average power and 80 % efficiency.

Spectral broadening and compression of ultrashort pulses is a widely employed technique to widen the optical bandwidth and shorten the pulse duration, which is required for the generation of few-cycle electric fields. Their applications range from ultrafast pump-probe spectroscopy [1] to the development of X-ray [2] and XUV [3] sources. A fundamentally new approach for spectral broadening and pulse compression using a quasi-waveguide system, namely the Herriott-type multi-pass cell (MPC), was introduced recently [4]. The technique was implemented with MPCs filled with bulk and gaseous media, thereby covering an impressive input peak power range from 9 MW up to 81 GW. So far, only a few demonstrations reached the sub-10 fs regime, mainly due to two challenges: The limited bandwidth of optics and the high demands on dispersion management inside the MPC. In this work, we report on our system breaking the 10 fs limit by employing custom-made dispersive mirrors covering the bandwidth from 850 to 1300 nm and Argon gas as a nonlinear medium.

The industrial-grade driving laser (PHAROS-UP by Light Conversion) delivered 90 fs and 300 µJ pulses at 25 kHz, resulting in 7.5 W average- and ~3.1 GW peak power. After coupling inside the multipass cell operating under absolute 3.6 bars of Argon, the output pulses were broadened to cover the 850 to 1300 nm range. The compression was performed with dispersive mirrors, providing an overall dispersion of approximately -550 fs2 of GDD and 170 fs³ TOD. The compression was verified with a commercial D-scan device from Sphere Photonics (Fig.1). The output pulse duration was measured to be 8.5 fs, with the main peak containing 86 % of energy as compared to the Fourier-transform limit. This corresponds to the peak power boost of 7.5 x and peak power of ~23 GW. The transmission of the cell and dispersive mirror compressor remained at 80 %. The long-term operation over 100 h shows excellent stability and no sign of degradation. The whole multipass compression unit (MIKS1_L) has a footprint of 1.5 x 0.24 m^2.

Our next step is further energy scaling to 400 µJ and pulse compression to a few cycles, sub-8 fs for attosecond pulse generation.

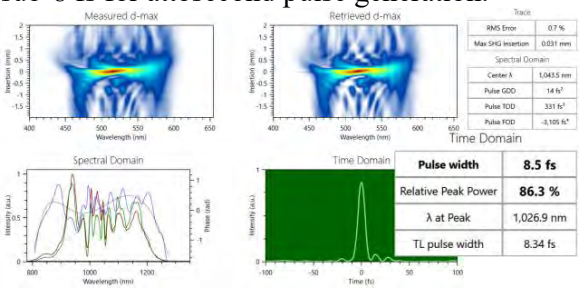


Figure 1. (Bottom-Left) Red: Measured linear spectrum. Green: Retrieved linear spectrum. Blue: Spectral phase. (Bottom-right) Temporal intensity of the pulse.

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Few-cycle Isolated Attosecond Pulses Generation with NIR Driving Fields

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Synopsis We demonstrate the generation of IAP with pulse duration of 51 attoseconds driven by few-cycle near-infrared (NIR) laser pulses during 1 fs linear polarization gate formed by generalized double optical gating (GDOG) technique. It is shown that IAP generation favors certain carrier envelope phases (CEP) in the narrow gate, which provides a way to eliminate the requirement of CEP stabilization. The demonstrated scheme for IAP generation in principle has much higher conversion efficiency than the long-wave driver scheme according to the wavelength scaling law of HHG.

The generation of ultrashort isolated attosecond pulse (IAP) has always been one of the core research aspects in the development of burgeoning attosecond science and technology. Long wavelength driving lasers are usually preferred for shortening the IAP temporal width. However, the conversion efficiency drops dramatically as the wavelength of driving laser increases, so that tradeoff has to be made between pulse duration and photon flux during IAP generation.

By employing GDOG technique, which preserves both plateau and cut-off region photons in high-harmonic generation, we achieve significant suppression of sample ionization caused by the leading edge of the driving laser field. This suppression enables operation at higher saturation intensities, thereby extending IAP cut-off energy [1]. Notably, GDOG eliminates the need for CEP stabilization in driving laser pulses, as experimentally established in prior studies. Through numerical simulations, we demonstrate that the narrow linear-polarization gate created by GDOG selectively enables efficient IAP emission only from specific laser waveforms with particular CEP values. This selectivity arises from the critical dependence of IAP generation on the carrier waveform of the driving laser, effectively enabling passive ultrabroadband IAP gating without requiring CEP stabilization. This mechanism contrasts fundamentally with alternative CEP-insensitive approaches that utilize phase-matching-assisted amplitude gating. While such methods preserve only cut-off spectral components at the expense of temporal resolution, our GDOG-based strategy maintains both spectral bandwidth and temporal confinement.

The generated IAPs were characterized using an attosecond streak camera and reconstructed through qPROOF algorithm [2], as illustrated in Fig. 1. Reconstruction results confirm the production of isolated attosecond pulses with durations as short as 51 attoseconds [1].

This work exhibits the feasibility of the generation of ultrashort attosecond pulses with sophisticated Ti:Sapphire driving lasers, and thereby holds out the prospect of high-flux and short-duration IAP generation for future attosecond pump-attosecond probe experiments.

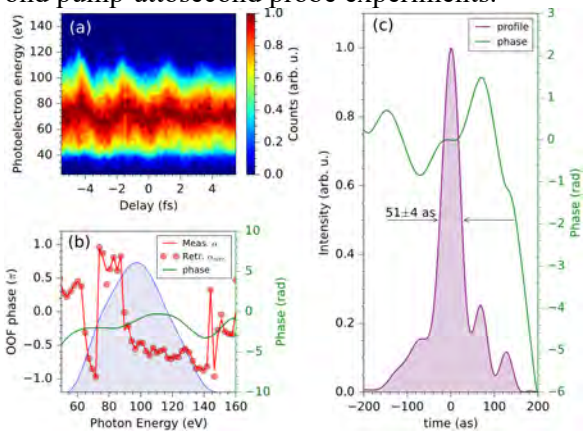


Figure 1. (a) Measured streaking trace; (b) the retrieved spectral phases with qPROOOF algorithm; (c) the reconstructed pulse profile with duration of 51 as.

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Few-femtosecond soft X-ray transient absorption spectroscopy with tuneable DUV-Vis pump pulses

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Synopsis This study presents an innovative system combining few-femtosecond deep-UV to visible tuneable pump pulses with an attosecond soft X-ray probe. This enables precise studies of ultrafast electron dynamics in a large range of photochemical reactions.

Achieving few-femtosecond resolution for a pump-probe experiment is crucial to measuring the fastest electron dynamics. As almost all molecules resonantly absorb radiation in the UV and/or visible, generating ultrashort pump pulses in this part of the optical spectrum has huge potential for probing excited state electronic dynamics while not ionizing the molecule or material. However, traditional UV-Vis pump pulses cannot achieve few-fs durations and usually operate at fixed wavelengths. Almost every material and even air begin to be absorbent and highly dispersive in this region, making the generation, transport, and characterization of ultrashort UV pulses extremely challenging. Technology to produce few-femtosecond tuneable UV pulses was recently realized by the generation of resonant dispersive wave (RDW) emission in gas-filled capillary fibers [1]. RDW emission is a revolutionary new way to pump photochemical systems that overcome many limitations of traditional pumping regimes.

Here, we present, to our knowledge, an unprecedented temporal resolution and pump tuneability for UV-Vis pumped soft X-ray transient absorption spectroscopy. We have combined few-fs deep-UV to visible tuneable pump pulses from resonant dispersive wave emission in hollow capillary fiber with attosecond soft X-ray probe pulses from high harmonic generation [2]. We achieve sub-5-fs time resolution, sub-fs interferometric stability, and continuous tuneability of the pump pulses from 230 to 700 nm. We demonstrate that the pump can initiate an ultrafast photochemical reaction and that the dynamics at different atomic sites can be resolved simultaneously. These capabilities will allow studies of the fastest electronic dynamics in a large range of photochemical, photobiological and photovoltaic reactions.

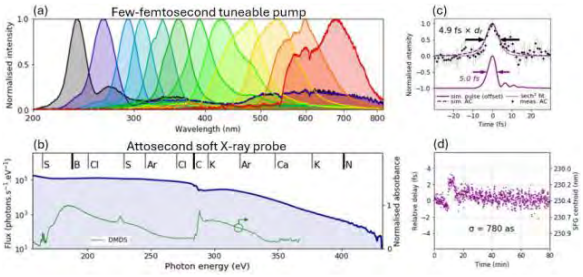


Figure 1. (a) Normalized measured spectra showing the continuous tuneability of the pump from 230 to 700 nm. (b) Typical soft X-ray probe spectrum generated with neon, extending in the water-window region (blue) and absorption spectrum of gas-phase dimethyl disulfide (DMDS, green). The sulfur L edges and carbon K edge are simultaneously resolved, and the energies of other atomic edges are labelled. (c) Measured intensity autocorrelation (AC) of the DUV pump pulse (270 nm) after propagation (with error bars and sech2 fit) and simulated AC from the simulated pulse temporal profile (offset). (d) Attosecond interferometric stability of the setup measured on the target.

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A high repetition HHG source of ultrafast XUV pulses for the SXP instrument at the European XFEL

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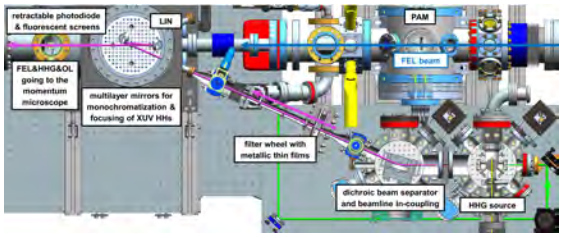
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Synopsis The Soft X-ray Port (SXP) at European XFEL combines a High-Harmonic Generation (HHG) source with its soft x-ray beamline, producing ultrafast XUV beams at MHz rep rates. This integration boosts XFEL's ability to perform advanced time-resolved spectroscopy and characterization of complex systems all year round.

At the European X-ray Free Electron Laser (XFEL) facility, a new instrument, the Soft X-ray Port (SXP), has recently commenced operation [1]. Designed primarily for time- and spin-resolved X-ray photoelectron spectroscopy (TR-XPES), SXP also supports investigations of complex chemical and bio-inorganic molecular systems using fluorescence spectroscopy, as well as studies on highly charged ions [2].

SXP is positioned downstream of the SASE 3 soft X-ray (SXR) undulator system, delivering ultrafast photon pulses with variable polarization in the energy range of 260 eV to 3000 eV at MHz repetition rates. Each pulse can contain up to 10^{12} photons, focused to a micrometer-scale spot, achieving intensities exceeding 10^{18} W/cm^2. These specifications enable the complete electronic, chemical and atomic characterization of solids, surfaces and interfaces using TR-XPES. To facilitate such studies, SXP is equipped with two powerful pump-probe lasers covering a broad spectral range from the mid-infrared to the ultraviolet. In this contribution, an HHG source is presented, which is designed to operate in conjunction with the SXP instrument as well as with the existing laser infrastructure, providing ultrafast XUV light in the range of 20 eV to 72 eV with MHz repetition rates.

The figure below shows a top view of the SXP instrument model, excluding the TR-XPES end station but displaying key permanent beamline components and the paths of the XFEL (blue) and HHG (pink) beams. The FEL beam enters the SXP instrument from the right, first passing through the Photon Arrival Time Monitor (PAM) and then into the Laser IN-coupling unit (LIN), further detailed in [1, 2, 3]. The beam continues toward a momentum microscope (not shown) on the left. The PAM monitors the relative arrival times of the SXR and synchronized optical laser (OL) pulses used for sample excitation, while the LIN enables co-linear coupling of the OL/HHG beams into the XFEL path.



The XUV beam (pink) originates in the center of the HHG source chamber. It is generated through a nonlinear interaction of a tightly focused femtosecond OL beam (green) with a noble gas that is injected via a 1/4" stainless steel pipe into the HHG chamber. Both beams propagate co-linearly into the adjacent separator chamber hosting a B_4C-coated fused silica mirror. From here, the XUV beam is reflected into the LIN where a pair of curved multilayer mirrors select and focus a single harmonic into the sample interaction zone, while the OL beam is rejected.

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Tuneable few-femtosecond far-ultraviolet laser pulses generated by Yb-laser-driven resonant dispersive wave emission

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Synopsis We present a three-stage setup for generating few-femtosecond resonant dispersive wave pulses in the far-ultraviolet (100–350 nm) at up to 100 kHz repetition rate using an Yb-based ultrafast laser. We characterise the far-ultraviolet pulses using in-vacuum transient-grating frequency-resolved optical gating.

Few-femtosecond far-ultraviolet (UV) laser pulses in the 100–300 nm range are an important tool for next-generation ultrafast science, because virtually all materials and molecular systems have strong absorption resonances in this wavelength region. Resonant dispersive wave (RDW) emission from higher-order solitons propagating in gas-filled hollow capillary fibres (HCFs) can enable the efficient and wavelength tuneable generation of bright and extremely short far UV pulses [1]. For high-sensitivity measurements, it is vital to increase the pulse repetition rate from the initial experiments based on titanium-doped sapphire amplifiers. We have previously demonstrated far UV generation above 200 nm at repetition rates up to 50 kHz and average power up to 360 mW with a compact, ytterbium-based drive laser [2]. However, RDW emission below 200 nm, further scaling to Watt-level average power, and the few-femtosecond duration of the pulses [3] have yet to be demonstrated with such a source.

Here we present a three-stage setup (Fig. 1a) to generate few-femtosecond RDW pulses at up to 100 kHz repetition rate. The first two stages of stretched HCF compress the pulses from an Yb-based ultrafast Laser (Light Conversion Carbide) from 330 fs to 11 fs. Far UV pulses are generated in the third-stage HCF, which is coupled to a vacuum chamber. So far, we have obtained far UV emission above 200 nm at up to 33 kHz. We have also obtained VUV emission down to 140 nm at lower repetition rate (Fig. 1b). The pulse compressor operates at up to 100 kHz (80 W input power) with an overall efficiency of 66% (Fig. 1c, d). All-in-vacuum transient-grating FROG measurements demonstrate that the source is capable of generating extremely short pulses (Fig. 1e–g).

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We are currently working on scaling the system to 100 kHz for few-femtosecond vacuum ultraviolet (VUV) generation.

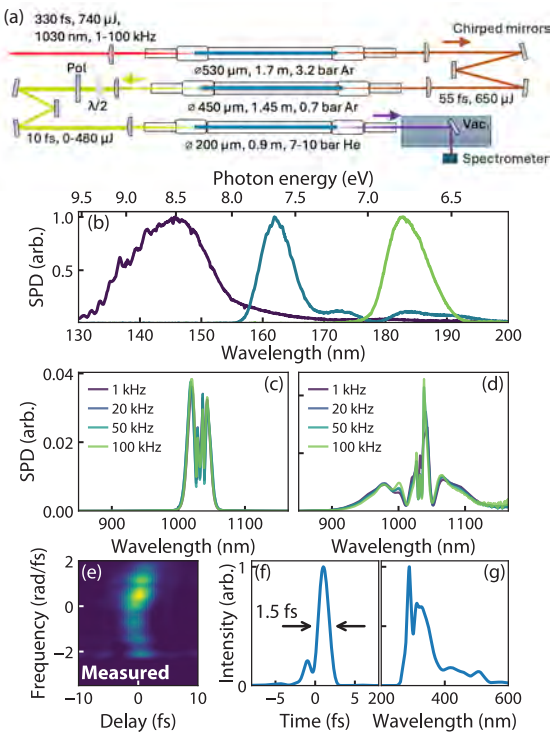


Figure 1. (a) Layout of the light source. (b) Spectral power density (SPD) of VUV RDWs down to 140 nm at 4 kHz. (c, d) Normalised SPD of the two compressor stages (e-g) TG-FROG measurement of a deep UV RDW at 33 kHz.

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Few-cycle optical vortices for strong field physics

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Synopsis We generate optical vortices with few-cycle pulse duration, delivering 500 µJ per pulse at a 1 kHz repetition rate. A 25 fs, 800 nm laser beam is shaped with a helical phase and coupled into a hollow-core fiber. The outgoing pulses, measured with a dispersion-scan setup, have a duration of 5.5 fs. We confirm that the pulse's orbital angular momentum (OAM) content is transferred to the newly generated frequencies using a spatially resolved Fourier-transform spectrometer. These few-cycle vortex beams exhibit a well-defined OAM, and sufficient energy to drive strong-field processes.

Laser beams carrying orbital angular momentum (OAM), such as optical vortices, have become a common tool in many areas of photonics [1], including particle manipulation, optical communications and strong-field physics. In this last domain however, one regime had yet to be achieved: the few-cycle optical vortex.

Generating a clean ultrashort vortex beam is challenging due to its intrinsic broad spectrum. Vortex beams are typically produced by imprinting an azimuthally varying phase onto a Gaussian beam, using either a spiral phase plate, a q-plate or a spatial light modulator. However, these devices are highly chromatic, leading to an inhomogeneous OAM content over a broad spectrum.

In this work [2], we overcame this challenge by shaping our pulse before post-compression, where the spectrum is still narrow. We then compressed the pulse using a hollow-core fiber filled with argon gas, achieving a pulse duration of 5.5 fs (≃2.1 optical cycles), as measured with a dispersion-scan device. Additionnaly, we analysed the beam's diffraction pattern through a triangular slit inserted in a spatially resolved Fourier-transform spectrometer. The results confirm that the vortex charge of the driving beam is transferred across the entire spectrum without any impurities. This is due to the guided propagation within the fiber, which constrains the OAM modes excited during propagation.

The overall conversion and transmission efficiency is 25%, yielding an output of 0.5 mJ – significantly lower than what can be achieved with a standard Gaussian beam. This discrepancy primarily arises from the difficulty of efficiently coupling the beam into the fiber. Nonetheless, given the short pulse duration, we can reach high irradiance values ($1.5\text{x}10^{14}$ W/cm^2 with a 1 m focal lens), sufficient to drive strong nonlinear processes. For instance, using this type of beam in high-harmonics generation could produce isolated attosecond vortices which, to our knowledge have never been observed. Such vortices would be valuable for probing the dynamics of recently discovered helicoidal dichroisms [3-4].

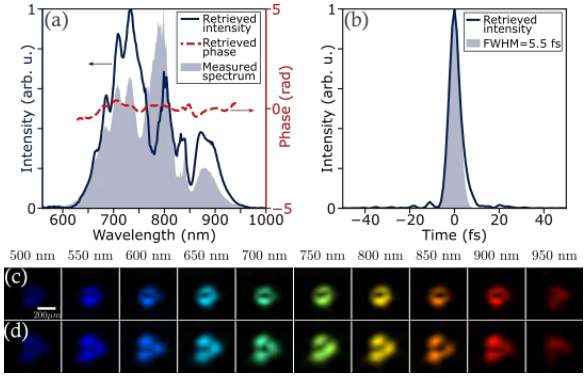


Figure 1. **(a)** Retrieved spectral intensity and phase. **(b)** Retrieved temporal pulse profile. **(c)** Focus and **(d)** diffraction pattern through a triangular slit for several wavelengths, retrieved by Fourier-transform spectroscopy. The three-dot diffraction pattern is characteristic of a beam with OAM dominated by $\boldsymbol{\ell = \pm 1}$.

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Towards a New HHG Driver Laser Platform: Bridging Q-Switched and Mode-Locked Laser Technology via Multi-Pass Cell Post-Compression

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Synopsis We present a bulk-rod Herriott-type multipass cell to compress a 0.5 ns pulse to 24 ps in a 10-cm fused silica rod. Bridging the parameter space of Q-switched and mode-locked lasers, our method is promising to realise a novel class of extreme-ultraviolet light sources driven by compact, robust and cost-efficient laser systems.

Pulsed laser generation relies on different technologies depending on the desired pulse duration, energy, and repetition rate. Typically, two different regimes are distinguished: Q-switched and mode locked lasers. Q-switching allows cost effective, robust and compact designs delivering high-energy pulses (~ mJ – J) with ns- to ps durations. However, their limitations in achievable peak power restrict the application of Q-switched lasers, for example, in high-precision material processing [1] or as direct drivers of higher order nonlinear optical processes such as high-harmonic generation (HHG) [2]. In comparison, mode-locked systems provide shorter pulse durations (ps- to fs-region), however at the expense of greater complexity, price and footprint.

The compression of Q-switched lasers has posed major challenges. Stimulated Brillouin scattering (SBS) [3] enabled to compress ns-lasers to ps durations, but intrinsic limitations, such as the Raman response of materials, have restricted compression efficiency and energy scaling. As a result, fully bridging Q-switched to mode-locked laser parameters in terms of pulse duration and peak power has not been achievable yet. This achievement could greatly impact the laser market and enable the realization of a new laser platform for ultrafast science applications.

In this work, we present a new technology driving self-phase modulation in a Herriott-type multi-pass cell (MPC) consisting of a 10 cm UVFS glass rod to spectrally broaden the pulses from an ultracompact Q-switched laser. Using a glas rod as nonlinear medium together with a compact MPC design allows us to extend MPC technology to the few-MW input peak power regime [4]. Before entering the MPC, the pulses have an energy of 0.94 mJ and a duration of 472 ps at a repetition rate of 100 Hz. The broadened pulses reach a transform-limited duration of 20 ps and are compressed in a transmission grating compressor. We measure a compressed pulse duration of 24.1 ps, corresponding to a compression factor of ~19.6 with respect to the input pulses. We

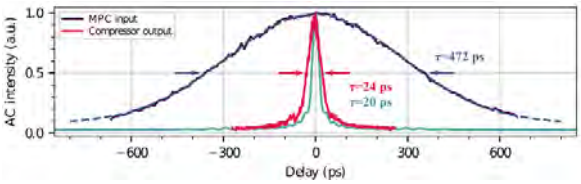


Figure 1. Autocorrelation measurements before (purple) and after (red) spectral broadening and compression, as well as the simulated fourier-limited pulse duration (blue).

support the experimental results with 3D simulations based on a symmetric split-step Fourier algorithm, indicating scalability to shorter pulse durations with compression factors close to 100, approaching GW-level peak powers. Importantly, our method is capable of suppressing the competing process of stimulated Raman scattering, using a spectral filtering scheme. We believe our approach offers great potential for multiple fields, providing a novel laser platform that is remarkably compact, robust and cost-effective. In particular, we envision our technology enabling Q-switched lasers as direct drivers for HHG sources.

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Batch Bayesian optimization of attosecond betatron pulses from laser wakefield acceleration

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Synopsis
Laser wakefield acceleration produces broadband X-ray femtosecond-scale betatron radiation from electrons accelerated by an intense laser pulse in plasma. Recent advancements in laser technology allow for extending these possibilities even further into attosecond scales. We optimize photon production through numerical simulations by changing the plasma density, utilizing batch Bayesian optimization. A multi-parametric scan identifies conditions that increase the peak of the betatron power spectrum up to more than 350% of the initial value, while maintaining the betatron duration around 100 as.

In laser wakefield acceleration (LWFA) [1], pC-class electron bunches can be accelerated to relativistic energies by a plasma wakefield wave driven by an ultrashort laser pulse. Intrinsically, the accelerated electron beam undergoes transverse oscillations, emitting broadband X-ray betatron radiation of high brightness. Our previous simulations showed that sub-femtosecond betatron pulses can be generated by 10-mJ fs-class laser pulses [2, 3]. Here, we modify the plasma profile by adding a short region of increased density, as shown in Fig 1. This increases the betatron oscillation amplitude, enhancing the betatron radiation while sustaining its duration short.

We alter the placement, length, and peak density of this density step in particle-in-cell simulations with the Smilei code [4]. The radiation gain is then calculated based on the electron trajectories from Smilei. The results are then optimized using batch Bayesian optimization, adjusting the step properties for the next batch of simulations. After 8 initial simulations, the peak of the betatron pulse power spectrum increased to 190% of the reference case without the density step, and to 370% after additional 24 simulations. While the pulse FWHM duration slightly increased from the original 81 as, it still remained on the order of tens of attoseconds, as shown in the inset of Fig. 1. Moreover, the critical photon energy was boosted above 1 keV compared to the reference case of 0.2 keV. In general, the emission is enhanced most efficiently when a short ($\lesssim$ 100 µm) high-density step (peak value $\sim 4\times$ the original density) is placed close to the end of the acceleration.

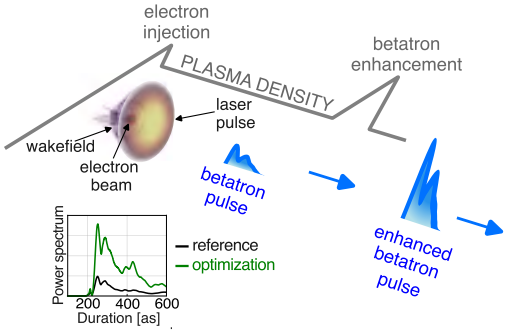


Figure 1. Set-up for the betatron enhancement and temporal profiles of the reference and optimized betatron pulses.

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High repetition rate deep ultraviolet and infrared beamline for time-resolved experiments on the few-femtosecond timescale

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Synopsis: Few-femtosecond (fs) UV pulses are key for studying electron dynamics in neutral molecules. We present a high-repetition-rate deep UV beamline based on an Yb-fibre laser amplifier, generating microjoule-level few-fs UV radiation up to 50 kHz for pump-probe spectroscopy. This setup provides high temporal resolution and improved collection statistics for electron-ion covariance measurements. Key components include a dispersion-engineered multipass cell (MPC) for pulse post-compression, a hollow-core fiber for UV emission, and an all-vacuum pump-probe setup with a covariance spectrometer.

Understanding ultrafast dynamics in molecular systems of increasing complexity requires advanced spectroscopic techniques providing high temporal resolution, statistics and signal-to-noise. Recent advances in table-top UV sources producing few-fs UV pulses have opened new possibilities for ultrafast molecular dynamics [1, 2]. Here, we present a high-repetition-rate deep UV beamline based on an Ytterbium fibre laser amplifier, operating up to 200 kHz repetition rate. Pulse post-compression down to sub-20fs duration is achieved using a single-stage dispersion-engineered multipass cell [3]. We demonstrate that our MPC can drive UV resonant dispersive wave (RDW) emission in a gas-filled capillary [4] up to 50 kHz. Figure 1 schematically illustrates the beamline and RDW-based setup at 50 kHz. The few-fs UV pulses are combined to near-infrared pulses in a pump-probe configuration that allows time-resolved measurements with few-fs time resolution. This beamline is intended to be used in combination with our double-sided spectrometer for covariance measurements with high-statistics. Applications of this unique setup include the investigation of ultrafast chiral dynamics via time-resolved photoelectron circular dichroism (tr-PECD).

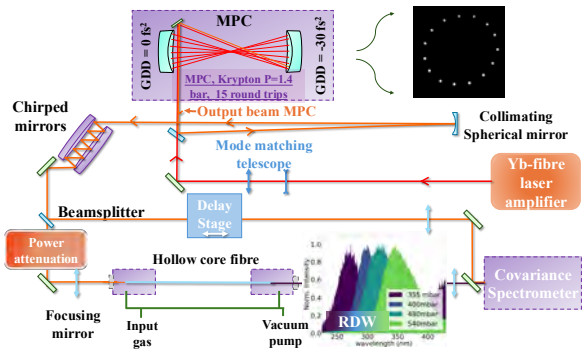


Figure 1. Schematic of the beamline, including the dispersion-engineered MPC compressor and the RDW capillary for UV generation. A typical RDW emission obtained at 50 kHz is also shown. The beamline includes an IR-UV pump-probe scheme and a double-sided covariance spectrometer for electron-ion measurements.

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Soft X-rays beyond 600 eV from a 45 W average-power, few-cycle, short-wave infrared driver

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Synopsis We present the generation of broadband soft X-rays extending beyond 600 eV driven by a high-average-power (45 W on-target), CEP-stable, few-cycle, short-wave infrared (2.1 µm central wavelength) optical parametric chirped pulse amplifer.

Attosecond pulses in the spectroscopically important soft X-ray (SXR) spectral region can be generated from carrier-envelope-phase (CEP) stable, few-optical-cycle, short-wave infrared (SWIR) driving pulses [1]. However, applications have been limited by the generally low photon fluxes in the SXR available in practice.

There is, therefore, a strong motivation to develop high-average-power SWIR sources to serve as SXR drivers. In this paper, we present a SXR broadband emission extending beyond 600 eV driven by a CEP stable, optical parametric chirped pulse amplifier (OPCPA) that delivers an on-target average power of up to 45 W. This represents the highest-average-power, phase-stable SXR driver to date.

The OPCPA system was recently reported in Ref. [2]. The pump source is 465 W from a Yb:YAG system that outputs 1.03 µm, 850 fs pulses at 52.6 kHz repetition rate (DIRA 400-50, upgraded to 500 W, Trump Scientific Lasers GmbH). The OPCPA is split into two parts; a commercial 'front-end' OPCPA (Starzz, FASTLITE) and a high-power 'booster' OPCPA. The 'front-end' outputs passively CEP-stable, ~100 µJ pulses with a spectrum spanning 1.8 and 2.5 µm (19 fs Fourier transform limit). The single-shot-resolved CEP stability was measured to be ~160 mrad.

In the high-power 'booster', the SWIR pulses are amplified to ~ 1 mJ (52 W average power) in two OPCPA stages based on MgO-doped lithium niobate crystals. After compression, the measured pulse duration is ~20 fs, corresponding to ~3 optical cycles. The beam exhibits an excellent spatial profile ($M^2 \leq 1.4$), which is vital for efficient SXR generation. Presently, the shot-resolved CEP stability of the amplified pulses is measured to be of the order 600 mrad. Further studies into the CEP instabilities arising in the on-target, amplified pulses are ongoing.

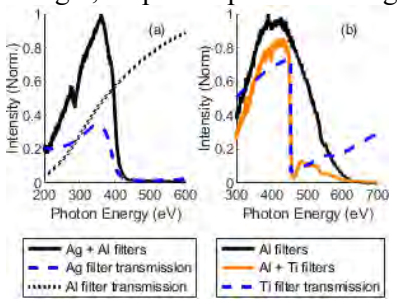


Figure 1. Measured soft X-ray spectra after transmission through (a) Ag and Al filters (b) (black curve) two Al filters and (orange curve) Al and Ti filters. Dashed lines illustrate filter transmissions obtained from Ref [3].

In a first experiment, the pulses were focused to a peak intensity of ~ 7 x 10^{14} W/cm^2 inside a 1.6 mm-long gas cell filled with helium (9 bar backing pressure). After spectral filtering with thin foils, SXRs were detected using a flat-field grating coupled to an X-ray CCD camera. The measured spectra extend beyond 600 eV (figure 1) in a low-divergence beam (2 mrad half-angle). This platform is thus set to expand the possibilities for attoscoend core-level spectroscopy.

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Shaping attosecond pulses using the minimum in high-order harmonic generation spectrum of argon atoms

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Synopsis The advent of attosecond light pulse technology has revolutionized scientific research by enabling unprecedented temporal resolution. This study demonstrates the shaping of attosecond pulses by utilizing minima in HHG spectra, such as the Cooper minimum in argon. These minima in HHG can split a single attosecond burst into dual peaks within each half optical cycle. This approach highlights the potential for advanced attosecond pulse manipulation, offering new opportunities for applications in ultrafast science and spectroscopy.

The emergence of attosecond light pulses technology has provided a brand-new perspective for scientific research in numerous fields and demonstrated its immense application potential. One of the most effective methods for generating attosecond pulses is through the synthesis of high-order harmonics generation (HHG) produced by the interaction between intense laser fields and matter such as atoms and molecules etc.. If a discrete spectrum of HHG is selected, it will result in the synthesis of attosecond pulse trains(APT), which manifest as a series of emission peaks, each with a duration on the attosecond scale. In contrast, if a continuous spectrum of HHG is synthesized, it will manifest in the time domain as isolated attosecond pulse (IAP) with well-defined pulse envelopes. Neither attosecond pulse trains nor isolated attosecond pulses can meet all application scenarios, and for some applications, it is necessary to shape the spectral structure of attosecond pulses.

In 2014, Schoun et al. [1] first used the Cooper minimum of argon HHG spectroscopy to experimentally shape the spectral structure of attosecond pulses, generating a split attosecond pulse train within a semi optical period. In theory, Jin et al. [2,3]. utilized the minimum value in the HHG spectrum of CO_2 molecules to achieve the splitting of attosecond pulse trains.

In this paper, we demonstrate the use of minimum in Ar HHG spectrum, such as Cooper minimum, or the minimum producing by the interference effect of two gas targets HHG signals to shape the ATPs, resulting in splitting of one attosecond burst into two emission peaks every half optical cycle. As shown in Figure 1, when there is no minimum in the HHG spectrum, the ATP spectrum exhibits an emission peak within every half optical cycle. However, when there is a minimum in the HHG spectrum, the original emission peak splits into two peaks.

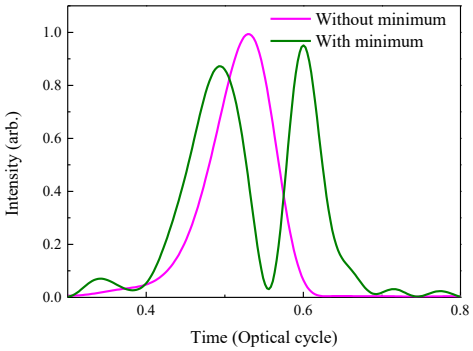


Figure 1. APT synthesized with H13–H30 using Ar HHG spectrum. The magenta curve is the HHG spectrum with no minimum, and the olive line is the HHG spectrum with a minimum.

This splitting ATPs obtained through the minimum of HHG spectrum has unique advantages in studying the Fano resonance phenomenon in atoms, molecules, and nanostructures [4].

Acknowledgment

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Nonlinear spin-orbit coupling of light in harmonic generation at solid surfaces

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Synopsis We experimentally demonstrate a novel mechanism of optical spin-orbit coupling in 2nd and 3rd harmonic generation by utilizing solid surfaces. Our findings reveal new pathways for spatio-temporal light control in extreme nonlinear optics effectively using characteristic properties of solids.

Nonlinear conversion of spatio-temporally structured light based on spin angular momentum (SAM) and orbital angular momentum (OAM) enables unique functionalities in spectroscopy, laser processing, micromanipulation, and optical communication [1]. Recent studies on gaseous media have demonstrated the control of angular momentum in extreme-ultraviolet high harmonic generation (HHG) [1,2]. However, due to the spherical symmetry of atomic gases, the conversion mechanism has relied solely on the independent conservation of SAM and OAM [2]. Here, we show that solid surfaces facilitate unique SAM-OAM coupling in harmonic generation by enhancing symmetry breaking in light-matter interactions.

We investigated second-harmonic (SHG) and third-harmonic generation (THG) at the surface of a 150 nm-thick indium tin oxide (ITO) thin film under epsilon-near-zero (ENZ) conditions, which enhance surface nonlinearities. As depicted in Figure 1(a), a right circularly polarized (RCP) driving field with a wavelength of 1300 nm—generated by an optical parametric amplifier operating at a 3 kHz repetition rate—was shaped into a Gaussian profile using an iris. This beam was then tightly focused onto the ITO film with a high-numerical-aperture lens (f = 3.1 mm). This wavelength corresponds to the ENZ region of ITO, enhancing the electric field component perpendicular to the surface plane. Spatial images of the reflected harmonics were captured using a CMOS camera.

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We observed that the spatial profiles of the SHG and THG beams for RCP components exhibited donut shapes, as shown in Figure 1(b). These donut shapes suggest nonzero OAM. To confirm this, we analyzed the dependence of focused profiles on the Gouy phase, altered using a tilted lens, and determined the OAM values of SHG and THG to be 1 and 2, respectively. These results indicate a novel nonlinear SOC of light, where nonzero OAM emerges from the electric field perpendicular to the surface (SAM = 0) of the driving laser at the focal position. This SOC arises from symmetry breaking in the perpendicular component, a phenomenon not observed in our recent study on HHG that preserved multiscale dynamical symmetry. Our findings introduce a new mechanism of SOC and expand the controllability of spatiotemporal optical structures in extreme nonlinear optics.

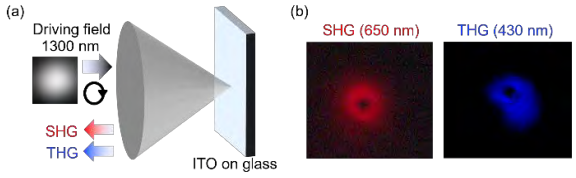


Figure 1. (a) Schematics of experimental setup. The inset image is spatial profile of driving field. (b) RCP spatial profiles of second and third harmonics.

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High-power, few-cycle, short-wave infrared source for high-flux, soft X-ray generation at 200 kHz repetition rate

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Synopsis We present the ongoing development of a high average power, ytterbium-based OPCPA, operating at 200 kHz. The system delivers CEP-stable, few-cycle pulses around 2 μm with an anticipated average power of 50 W. The building of a pump-probe beamline for high-flux, soft X-ray attosecond pulses is underway.

Recent years have seen a growing interest in pulsed soft X-ray (SXR) radiation, particularly in the water window [282-533 eV], where high-order harmonic generation (HHG) provides compact table-top sources. To achieve HHG in this region, femtosecond light sources with central wavelengths in the mid- and short-wave infrared (SWIR) are required. Advances in laser technology have enabled the development of such sources, which are becoming increasingly more common [1, 2, 3]. However, obtaining high-flux, coherent SXR emission remains challenging due to the unfavorable scaling of HHG conversion efficiency with longer wavelengths [4], which continues to motivate the development of high peak and average power infrared drivers.

In this work, we present the progress in the development of a high-power, high repetition rate (200 kHz) optical parametric chirped pulse amplification (OPCPA) source centered at 2 μm for driving HHG in gases. The system is based on an Ytterbium (Yb) CPA laser, including an Yb:YAG thin-disk multipass amplifier, delivering pulses with a duration of 1 ps at 1.03 μm and 500 W of average power, while preserving a high beam quality (M^2 <1.2). The seed pulses are obtained via white-light generation and noncollinear optical parametric amplification (NOPA) in the visible, followed by difference frequency generation (DFG). This yields a broadband SWIR spectrum (1.6-2.6 μm) with a Fourier transform limit of ∼15 fs and passive carrier-envelope-phase (CEP) stability. The SWIR pulses are then amplified in three consecutive NOPA stages, aimed to achieve 250 μJ of pulse energy (50 W average power). In the current development phase, the first stage is realized with 15 μJ of output pulse energy. Compression to the few-cycle regime is achieved using a combination of bulk materials with positive and negative dispersion (zinc sulfide and infrared-grade fused silica, respectively), along with chirped mirrors for third-order dispersion management. The pulse is characterized using third-harmonic generation (THG) dispersion scan (d-scan), showing a retrieved pulse duration of 21 fs.

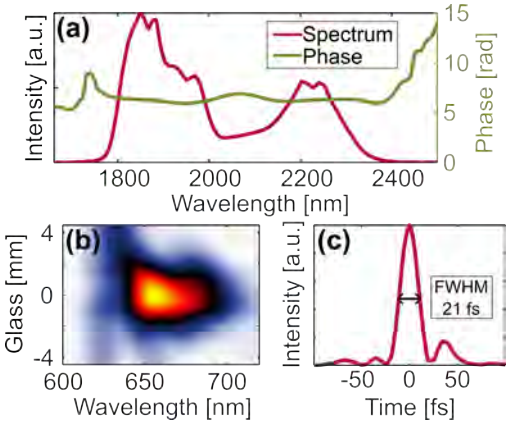


Figure 1. (a) Amplified SWIR spectrum (b) THG d-scan trace (c) Retrieved pulse intensity profile

The goal of the laser- and beamline developments is to achieve sufficiently high SXR flux towards the water window to enable attosecond pump-probe photoelectron spectroscopy in the gas phase.

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Dark Pulse Interferometry for Attosecond Pulse Characterization

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Synopsis We propose a novel technique for retrieving the spectro-temporal properties of attosecond pulses—specifically their time-frequency structure—using dark pulse interferometry. By interfering high harmonic radiation generated by a bright driving pulse with that produced by its corresponding dark pulse, we theoretically demonstrate a method to extract the spectro-temporal characteristics of high-order harmonic pulses. This approach offers a promising alternative for attosecond pulse characterization.

High-harmonic generation (HHG) is a highly nonlinear and non-perturbative process that enables the production of coherent extreme ultraviolet (XUV) and soft X-ray radiation. Its remarkable coherence allows translating properties of the driving field to the harmonics, making HHG a key tool in attosecond science. The temporal and spectral structure of attosecond pulses is crucial for applications such as ultrafast spectroscopy and strong-field physics, where phase information plays a fundamental role. The development of diagnostic tools to retrieve the harmonic phase, through challenging, opened the era of attosecond science [1, 2]. However, existing techniques require photoionization studies after HHG, and all-optical techniques remain elusive.

In this work, we propose the use of dark pulse interferometry as an alternative all-optical technique for attosecond pulse characterization. Our theoretical studies using advanced HHG simulations [3], demonstrate that the interference between the harmonic radiation produced by a bright pulse and its corresponding dark pulse [4], enables the retrieval of its spectro-temporal properties. In other words, we provide a proposal to experimentally retrieve the time-frequency structure of attosecond pulses.

To extract phase information of the high-order harmonic pulses, we conduct a systematic temporal scan of dark pulses—acting as a temporal gate [5]—across different background pulses, recording the corresponding harmonic spectra. The resulting spectrograms after a proper interference with the former bright pulse provide for a direct visualization of the temporal emission of individual harmonics, a crucial step for the full experimental characterization of attosecond pulse formation [See Figure 1].

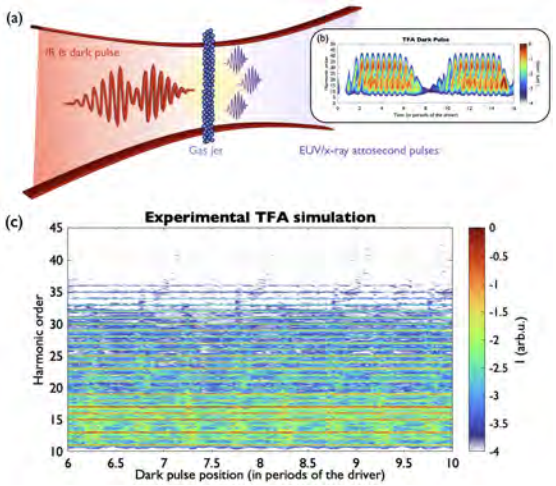


Figure 1. **(a)** Schematic view of a HHG experiment driven by dark pulse. **(b)** The corresponding time-frequency analysis (TFA) of its XUV/soft X-ray attosecond radiation. **(c)** Spectro-temporal characterization of high-order harmonic pulses via dark pulse interferometry.

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A Beamline with Efficient Isolated Attosecond Pulse Generation and Detection for Time Resolved Spectroscopy Driven by Synthesized Sub-Cycle Pulses

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Synopsis A novel beamline for attosecond transient absorption spectroscopy driven by synthesized optical waveforms is presented. It utilizes a central control system and a high-efficiency reflective-zone plate spectrometer and includes an in situ characterization method for the intricately shaped synthesized waveforms, allowing for sophisticated waveform controlled attosecond experiments.

The development of high-energy optical waveform synthesizers in recent years has created new possibilities for studying and controlling electronic motion on sub-cycle time scales[1]. The capability to shape the optical waveform on these time scales allows the generation of non-sinusoidal fields, enabling complete control over the formation of attosecond pulses through high-harmonic generation (HHG). Using our parametric waveform synthesizer[2], we showed that such pulses increase the efficiency of HHG and that their sub-cycle nature leads to a direct isolation of a single attosecond pulse.

We introduce our newly constructed attosecond beamline, tailored for studying ultrafast processes with synthesized multioctave spanning infrared pulses and broadband isolated attosecond pulses generated by these infrared waveforms. The system permits the usage of up to 10 bar of backing pressure for the HHG allowing the generation of water-window harmonics. To facilitate complex experimental procedures, the beamline makes use of the TANGO control system, which unifies the interaction of the more than 30 active components. Furthermore, the beamline utilizes a novel reflective-zone plate (RZP) XUV to soft X-ray spectrometer with improved diffraction efficiency, developed in cooperation with Nano Optics Berline and BESSY, making our system extremely flexible, by enabling the measurement of photon energies from 20 eV up to 600 eV.

Finally, we will present our newly developed characterization technique TREX[3], incorporated into our beamline, based on the third-order response of the high-harmonic gas target. This technique enables in situ characterization of the sub-cycle waveforms at the experimental interaction point, providing exceptionally accurate information for the study of ultrafast phenomena. In combination with the synthesizer control system and a tunable dispersion management scheme, the TREX technique allows the independent characterization and control of both pump and probe (HHG driver) optical waveforms.

These developments pave the way for advanced waveform-controlled attosecond experiments to study and control (electronic) motion on the few- to sub-femtosecond timescale.

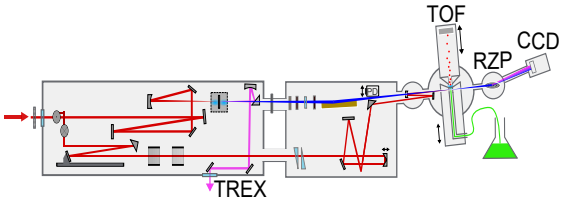


Figure 1. From left to right: HHG generation chamber; XUV optics and pump-probe recombination chamber; target chamber including sample-delivery and time-of-flight (TOF) spectrometer; RZP-spectrometer.

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Efficient High-order Harmonic Generation in the Overdriven Regime in a Sub-mm Glass Chip

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Synopsis In this work we present experimental and theoretical results on high-order harmonic generation (HHG) in the overdriven regime. Thanks to the optimal gas confinement offered by a micromachined glass chip, we obtained a remarkable cut-off extension up to 240 eV when drving the process with an 800-nm driver in Neon.

In recent years, the growing applications of table-top attosecond extreme ultraviolet (XUV) and soft X-ray sources [1] have driven the optimization of their photon flux, maximum reachable photon energy, and brightness. In this context, we introduce an efficient configuration for high-order harmonic generation (HHG) in the overdriven regime [2] using a micromachined glass cell allowing for gas confinement over a sub-mm length. The overdriven regime indeed requires high laser intensities at the interaction point (up to 10^{16} W/cm^2) and a short, well-confined gas medium.

We adapted the design of a laser-micromachined glass chip used for third harmonic generation (THG) [3] to suit the HHG geometry. The generation was driven by 800-nm pulses of 30-fs duration. Under optimized conditions—250 mbar Neon or Argon and $8{\cdot}10^{15}$ W/cm^2 at the focus—we observed a cut-off extension up to 240 eV in Neon (Fig. 1).

These results are explained in the framework of the overdriven regime, where radiation can be efficiently phase matched at higher photon energies compared to conventional phase matching over short propagation distances [4].

Further cutoff extension beyond the C K edge (~284 eV) was observed by driving the overdriven HHG process with the output of a two-stage, carrier-envelope phase stable optical parametric amplifier (OPA) [5], centered at 1500 nm (35-fs pulses, $3.5{\cdot}10^{15}$ W/cm^2 at the generation focus).

Insights into the experimental results are supported by real-space simulations of harmonic generation and propagation, offering deeper understanding of the mechanisms at play.

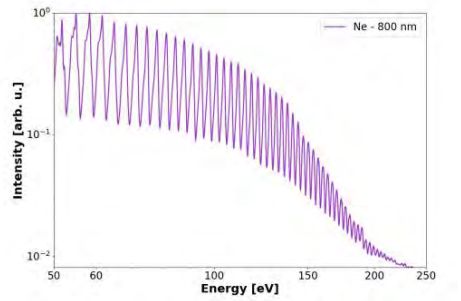


Figure 1. HHG spectra with 800-nm driving pulses (1.4 mJ, 30 fs) in Neon at 250 mbar.

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Hyper Spectral Ultrafast Source HYPUS

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Synopsis Post compression of a 2mJ, 80W Yb laser down to sub 7fs using two cascaded hollow-core fibers (HCFs) is achieved with ~70% efficiency and long term hands off operation. As a subsequent application of the broadband HCF continuum, we exploit its spectral content to derive new wavelengths. Employing various nonlinear conversion technics, we achieve a spectral extension ranging from the deep UV (200 nm) up to the far IR (12 um).

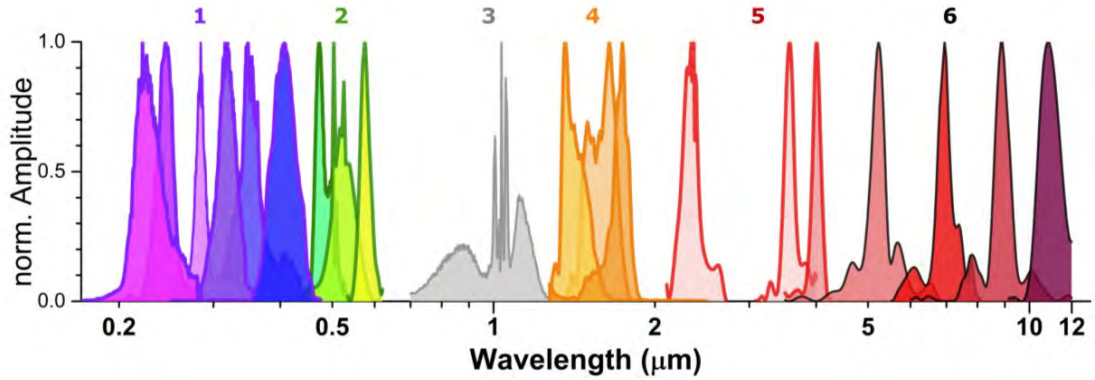


Figure 1. Six output channels labeled 1-6 of HYPUS that can be available simpultaneously. The independent tuning within each channel is depiced by exemplary spectra of same line color.

We will present single and double stage HCF compression systems ranging from 80 W – 120 W. A key aspect of this sources is the shot-to-shot as well as long term stability. For a single stage for instance, we achieved a single shot St. Dev. of 0.3% over close to 80 million individual shots (30 min at 40 kHz).

Spectrally broadening and compression of an 80 W laser down to sub 7 fs using two cascaded hollow-core fibers (HCFs) is achieved with up to 70% efficiency. The corresponding spectyrum is seen as channel 3 (grey curve) in Fig.1. As a subsequent application of the broadband continuum, we exploit its spectral content to derive new wavelengths. Employing sum frequency generation (channel 2) and difference frequency generation (channels 4 -6), respectively, we achieve a spectral extension covering the UV (200 nm) up to the far IR (12 um).

Noteworthy, the deep UV channel 1 is achieved vis the mechanism of resonsnt dispersive wave (RDW) emission and the repetition rate can be tuned up 100 kHz.

In contrast to OPAs, multiple spectral channels are available simultaneously. Our source offers up to five different channels (UV, VIS, NIR, IR, far IR) that are all derived from the same HCF output pulse. The IR and far IR channels are supposedly passively CEP stabilized.

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Atto-seconds electron beams for XFEL and MeV-UED

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Synopsis Inverse free electron laser (IFEL) is one of the most powewrful tools for laser manuplating electron beam. IFEL has been explored extensively for producing femto to atto seconds electron beams.The success of atto-sec XFEL is the latest example. Here we present a THz IFEL for atto-seconds MeV-UED. With atto-seconds MeV-UED, we will be alble to image all stages of pulse radiolysis in water from physical (as to fs) , physical-chemical (fs to ps) and to the chemical (μs) stages for the first time.

Laser has played critical role in electron beam generation and manipulation since 1980s. High-brightnee electron beam produced by the photo-cathode RF gun enabled X-ray Free Electron Lasers (XFEL) and MeV Ultrafast Electron Diffraction (MeV-UED) [1]. The combination of photocathode RF gun and electron beam bunch compressor led to femto-seconds electron beam for XFEL and MeV-UED [1]. It is the IFEL opens the frontier of femto-second [2] to atto-second electron generation [3]. In an IFEL, high power laser interacts with electrons in a periodic magnetic field, which lead to energy modulation (or energy chirp within the laser wavelength). The energy modulated electrons will convert to spatial modulation (microbunching) after a drift space. The micro-bunched electron has a pulse width shorter than a quarter of the laser wavelength, and an order of magnitude higher peak current (Figure 1).

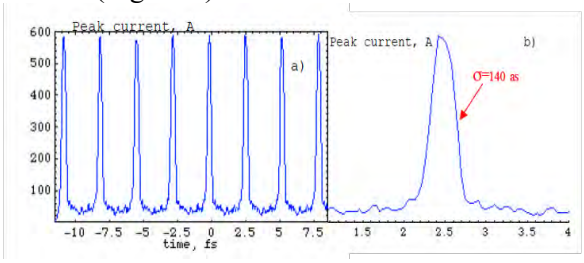


Figure 1. Microbunched electron pulse train generated by 0.8 μm IFELwith (a), atto-sec microbunch with 6 times higher peak current (b).

MeV-UED and XFEL are complementary approaches. With MeV electrons, the repelling Coulomb force is significantly mitigated, and MeV-UED offers stronger diffraction, less multiple scattering and higher sensitivity to structure dynamics compared to lower energy UED [4]. MeV-UED had a broad and transformative impact on ultrafast material science and chemical dyanmics, such as the first ultrafast structure dynamics of 2-D materials[5]; light-induced transient states of quantum materials[6], the first direct imaging of fundamental chemical processes [7-8] and hydrogen bond dynamics in liquid water [9].

To image all stages of pulse radiolysis in water using MeV-UED, atto-second electron pulse is needed. With THz IFEL, we not only abe able to produce atto-second electron pulse, but also preserve the transverse brightness of electron beam.

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Bright isolated XUV attopulse generation through high-order frequency mixing and ellipticity gating techniques

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Synopsis We explore the generation of bright isolated attosecond pulses using high-order frequency mixing and ellipticity gating technique. By adding the weak low-frequency driver to accompany the strong one, the XUV intensity is improved due to propagation, and a single attopulse is selected from the train by controlling the ellipticity of the driver. Our findings highlight the potential for a novel source of bright attosecond pulses, advancing their application in cutting-edge spectroscopic techniques.

The interaction of an intense laser pulse with a highly nonlinear medium generates attosecond XUV pulses through high-harmonic generation (HHG). However, increasing the laser intensity to achieve higher XUV pulse intensity enhances stronger ionisation, leading to a blue shift of the driver, disrupting harmonic phase matching, and ultimately weakening the pulse intensity.

This challenge is addressed by employing high-order frequency mixing (HFM) [1], which combines a strong IR laser with a weaker low-frequency laser field, as shown in Figure 1. By combining these two laser sources, HFM generates an additional frequency comb that is shifted with the respect to the HHG comb by one or two photons of the low-frequency field towards the red side of the spectrum, effectively cancelling the blue shift and enhancing phase matching.

We numerically model both HHG and HFM processes using Python-based software, that we develop, simulating a coupled microscopic response using the time-dependent Schrödinger equation (TDSE) and strong-field approximation (SFA), along with macroscopic propagation through the medium in one and two dimensions. By controlling the ellipticity of the driving IR laser pulse, we generate a single attosecond pulse, and its brightness is enhanced by switching from HHG to HFM through the addition of a weak low-frequency mid-infrared laser pulse.

Our results demonstrate the potential for a new source of bright isolated attosecond pulses using HFM and ellipticity-gating techniques [2], offering a significant advancement in optimising isolated attosecond pulse generation, which are essential for advanced spectroscopic applications, more precisely for attosecond pump-probe measurements.

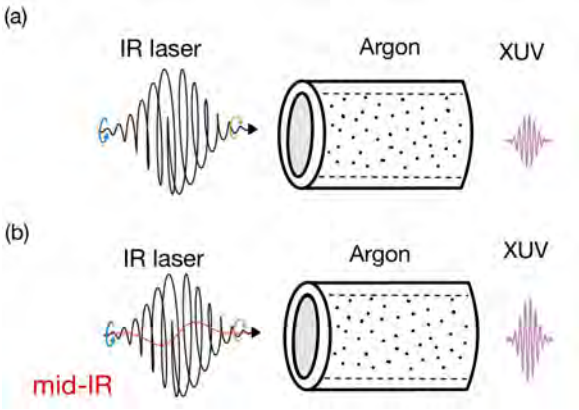


Figure 1. Schematics for the comparison of isolated attosecond pulse generation in argon gas for (a) the HHG process, where the signal is generated by a strong IR field alone, producing the characteristic harmonic spectrum, and (b) the HFM process, where the signal is generated by mixing a strong IR field with a weak mid-IR field, resulting in a significantly more intense XUV signal.

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Towards 300 mJ, 1 kHz, < 1 ps thin-disk regenerative amplifiers

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Synopsis We present the new Dira 300-1 system: a Ytterbium regenerative amplifier featuring two disks. The system delivers up to 400 mJ pulses before compression at 1 kHz. The system shows beam quality < 1.2 M^2 and pulse-to-pulse energy stability < 0.25% rms. The amplified spectrum supports < 700 fs pulse duration. Both turn-on and -off routines of the system are fully automated, making it very simple to operate.

Since its first demonstration over 30 years ago, the thin-disk laser technology has undergone significant advancements, driving advancements in both industry and science. In many industrial sectors, it has become indispensable for cutting, welding, and precision machining. At the same time, ultrafast Ytterbium thin-disk amplifiers play a crucial role in attosecond physics, where they are used to pump Optical Parametric Amplifiers. When employed for filamentation, they can even serve as lightning protection, as demonstrated in the Laser Lightning Rod project [1]. After nonlinear pulse compression, they can serve as driver source for laser-based particle acceleration, further demonstrating their broad application fields.

Here we present the new Dira 300-1 thin-disk regenerative amplifier developed at TRUMPF Scientific Lasers and built upon the Dira 200-1 regenerative amplifier series previously reported by S. Klingebiel *et al.* [2].

The Dira 300-1 system incorporates a ring amplifier with two Yb-doped thin-disk active mirrors. Both disks are diode pumped at a central wavelength of 940 nm with up to 900 W of average power per disk. When seeded with up to 50 µJ pulse energy from an industrial fiber-based ultrafast laser (TruMicro 2000, TRUMPF SE), the amplifier delivers up to 400 mJ pulse energy at 1 kHz. In Fig. 1 both optical-to-optical and slope efficiency of the amplifier are plotted. The system reaches up to 50% slope efficiency, only reduced due to saturation towards the high energy range. The amplified output exhibits excellent beam quality with a measured M^2 < 1.2 and pulse-to-pulse energy stability < 0.25% rms. The spectral bandwidth after amplification corresponds to Fourier-transform-limited pulse duration below 700 fs.

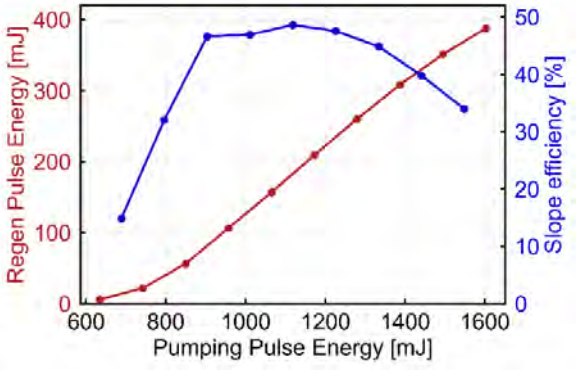


Figure 1. Optical-to-optical and slope efficiency of the Dira 300-1 amplifier at 1 kHz shown in red and blue, respectively.

The system is equipped with fully automated beam pointing, intracavity diagnostics, temperature and pulse energy control systems, ensuring outstanding long term stability and reliable operation.

In conclusion, we demonstrate a new generation of regenerative amplifiers, which combine an industrial grade reliability with an excellent beam quality and pulse energy stability suited for scientific applications. In case higher pulse energies or peak intensities are required, the output of the Dira 300-1 system can be further amplified in a multi-pass amplifier or spectrally broadened using nonlinear broadening technique, such as a Herz solution from TRUMPF Scientific Lasers [3].

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An in-line setup for the generation of a cross-polarized double attosecond pulses with controllable delay

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Synopsis In this work we present an in-line optical setup capable of producing a cross-polarized double attosecond pulse with controllable delay. A pair of birefringent wedges, one of which is mounted on a motorized translation stage, is used to create two orthogonally polarized delayed replicas of a 6 fs pulse. Each of them is then used to generate a single attosecond pulse through the technique of polarization gating in high-order harmonic generation (HHG). This new light source could be used for new fundamental studies of light-matter interaction.

Our goal is to have an in-line optical setup capable of generating a pair of isolated attosecond pulses (IAPs) with controllable delay between them. To achieve this, our idea is to combine the delay feature present in birefringent interferometers with the generation of an isolated attosecond pulse through polarization gating [1]. An sketch of our experimental setup is shown in figure 1.

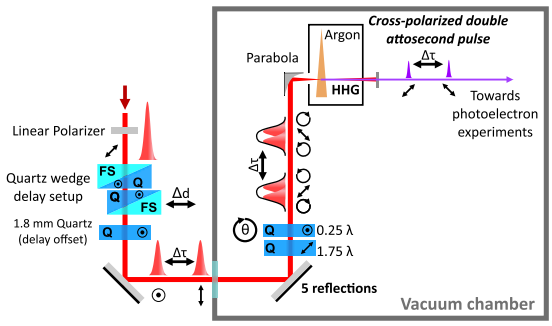


Figure 1. Diagram of the experimental setup.

We use a home-built OPCPA laser to produce 6 fs pulses, around a center wavelength of 830 nm, with a repetition rate of 200 kHz [2]. The laser is transmitted through a combination of quartz and fused silica wedges. Due to birefringence, the quartz wedges introduce a group delay between the S and P polarizations. One of the quartz wedges is mounted on a motorized translation stage to adjust the amount of quartz insertion, allowing us to change the delay over a range of 22 fs. The fused silica wedges are glued to the quartz wedges such that the total amount of glass remains constant regardless of the stage position, thus maintaining a constant pulse duration over the scanning range. Our polarization gating scheme employs a combination of two quarter-wave plates. The first waveplate (1.75λ) introduces a delay of 4.4 fs between the $-45°$ and $+45°$ polarizations, while the second waveplate converts the circular polarization to linear and vice versa. After the two waveplates, the input pulses S and P have acquired a time-dependent polarization with opposite linear polarization at the center of the pulse. High harmonic generation is achieved by tight focusing of the infrared beam into an argon gas jet with an off-axis parabola.

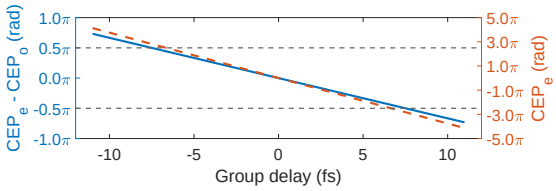


Figure 2. CEP as a function of group delay

Changing the quartz wedge position not only adds a delay between S and P polarizations but it also affects their respective carrier-to-envelope phase (CEP). In order to generate two IAPs simultaneously the difference in CEP between the two optical pulses needs to be a multiple of π. As we show in figure 2, in the scanning range of our delay setup, the difference in CEP evolves from -0.7π to $+0.7\pi$. Since we can externally control the CEP for one of the two pulses, there will be a maximum of 2 delay positions where the simultaneous generation of two IAPs is possible.

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Control of attosecond XUV pulses using an intermediate-field spatial filtering

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Synopsis Spatio-spectral control of XUV beams is demonstrated by combining divergence monitoring through generation geometry with intermediate-field spatial filtering. This control is nicely reproduced by simulations showing that spatial filtering results in an effective improvement of attosecond radial beam homogeneity.

XUV beams produced by high-order harmonic generation are commonly used as a table-top source of attosecond pulses. Controlling the focusing properties of the XUV beam remains challenging as harmonic divergence and foci positions may depend on the harmonic order [1,2]. We have demonstrated, based on an optics free scheme, the ability to control the XUV beam spatial properties and in particular its spatial chromaticity by varying the gas jet position relative to the driving laser focus [3]. As a consequence, using a pinhole in the path of the XUV beam located in the intermediate field, we can perform XUV spectral filtering with high contrast and selectivity [4]. We show that this filtering improves the spatial homogeneity of the attosecond pulses in the far field.

Broadband spectral tuning with a high contrast is experimentally demonstrated (Fig.1a) while changing the gas jet position. We do observe a subsequent change in the pinhole transmitted spectrum from high order harmonics (H29 to H47) generated in Neon. We simulated the observed effect by solving the Time-Dependent Schrödinger Equation [5] for the XUV source term generated in a thin gas medium and calculating the intermediate and far fields using a Green function describing exactly the Gaussian beam propagation. From this model, we calculate the transmittance of each harmonic beam (Fig.1b). The evolution of the transmittance curves with respect to gas jet position is in good agreement with experimental findings.

We then calculate the attosecond profile in the far field at different radial distances. Without spatial filtering (Fig. 1c) the attosecond temporal structure is not homogeneous and show distorsion and broadening with increasing radial distance as the spectral amplitudes and relative phases vary. In contrast, using a spatial filter tends to spatially homogenize the attosecond structure (Fig. 1d).

High throughput spatio-spectral filtering provides a straightforward way to significantly improve the radial homogeneity of the attoseond pulses associated with spectral tunability which is an important issue for attosecond metrology and applications.

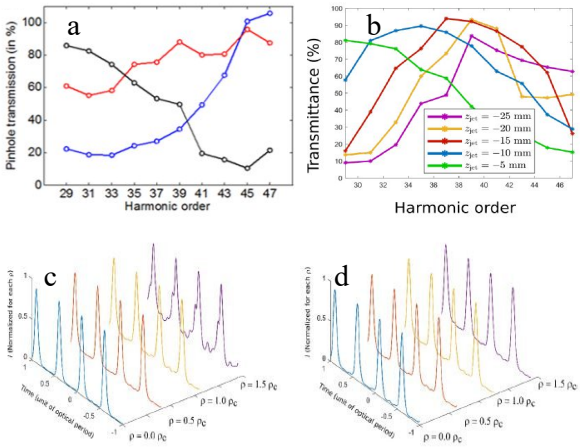


Figure 1. (a) Transmittance measured for all harmonics from H29 to H47 for jet position = -58 mm (blue), -36 mm (red) and -20 mm (black). (b) Simulated transmittance versus harmonic order for five gas jet positions. Temporal structures are depicted in (c) for free propagation and (d) with filtering. We define ρ_c as the radius that contains 50 % of the XUV beam power.

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Attosecond metrology of vacuum-ultraviolet high-order harmonics generated in semiconductors via laser-dressed photoionization of alkali metals

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Synopsis We present a novel scheme for the generation and temporal characterization of ultrashort pulses emitted by laser-driven semiconductors. Using a cross-correlation approach, in which we photoionize Cesium atoms, we demonstrate that high-order harmonics generated through the interaction of mid-IR pulses with a ZnO crystal produce attosecond pulses in the VUV spectral region.

In the last years, high-order harmonics produced by the interaction of semiconductor crystals with intense laser fields has gained significant attention. The generation process has been employed to investigate the dynamics of solids in the presence of strong laser fields, but its potential to be utilized as an attosecond light source for applications in attosecond science has remained experimentally unexploited.

The interaction between semiconductor materials and intense mid-IR laser pulses results harmonics with a cut-off in the few eV photon energy range. This is below the ionization threshold of noble gases preventing the use of conventional attosecond pulse characterization techniques [1]. To overcome this issue, we investigated the photoionization of Cesium atoms by a comb of VUV high harmonics and delayed mid-IR pulses. We observed that the number of electrons ejected along the laser polarization direction oscillates at the frequency of the laser electric field, and these photoelectrons originating from the different harmonics oscillate almost in phase. The experimental results match reasonably well with our theoretical calculations. Our findings open up the realm of VUV attosecond pulses from strongly laser-driven semiconductors.

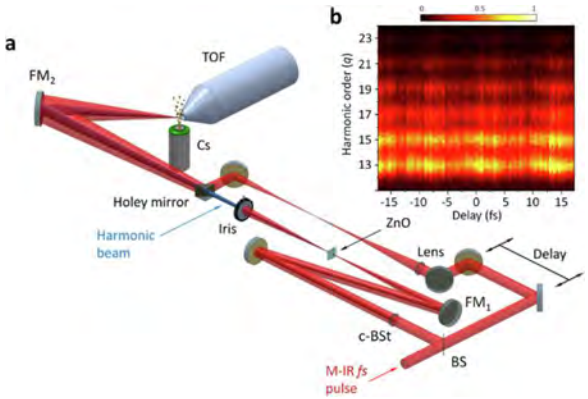


Figure 1. (**a**) Scheme of the experimental setup and (**b**) photoelectron spectrum as a function of the delay between the VUV and mid-IR fields [2].

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XUV-pump IR-probe resonant shake-up ionization of helium

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Synopsis We present an *ab initio* study of resonant shake-up ionization of helium doubly excited states (DES), selectively excited by an XUV free-electron laser pump pulse, in the presence of a strong dressing IR pulse, to reproduce recent measurement at FLASH, in Hamburg. The angularly- and energy-resolved resonant photoelectron distributions are shown to be extremely sensitive to the dynamic correlation in the DES, which we can finely tune in a dedicated helium code. This result opens the way to monitor in real time the evolution of dynamic correlation in coherently excited metastable wavepackets.

Helium is an ideal prototype to investigate electronic correlation in ionization processes with high experimental [1, 2] and theoretical accuracy [3]. Pump-probe ionization processes through doubly excited states (DES), in particular, open a window on studying the effect of correlation resolved in time [4]. In this work we present an *ab initio* study of the XUV-pump IR-probe ionization of DES made possible by the recent development of high-repetition-rate multparticle coincidence spectroscopy with intense XUV pulses at Free-Electron Lasers (FELs) [5,6]. The long duration and tunability of the XUV pulse allow us to isolate the multiphoton shake-up signal associated to a specific DES.

The time-dependent Schrödinger equation is integrated via a second-order split-exponential operator [3]. To reproduce ionization by long FEL pulses, we have implemented a periodic extraction of the external portion of the wavefunction, which allows to carry out simulations in comparatively small quantization boxes. In our close-coupling approach electron correlation can be tuned from a minimal static-exchange model to full convergence of the configuration-interaction (CI) space, which allows us to highlight the effect of dynamic correlation on the DESs above-threshold ionization spectra (ATI).

The simulated ATI spectra are in good agreement with recent measurements in T. Pfeifer's group [7]. The effect of dynamical correlation is qualitatively illustrated by the conditional electron-pair probability of the DESs obtained by diagonalizing the field-free Hamiltonian with a complex absorption potential (Fig. 1). This work is supported by the NSF grant No. PHY-1912507.

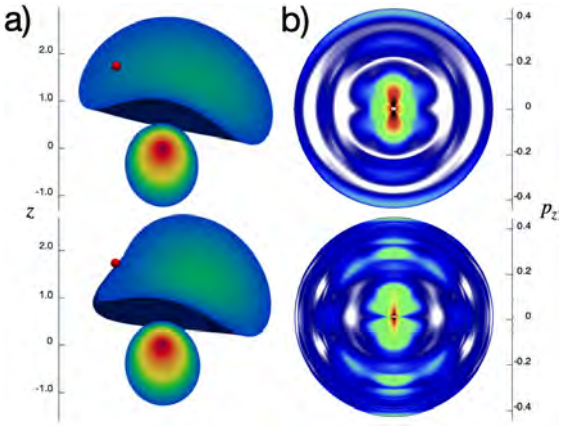


Figure 1. (a) $3s3p$ conditional probability distribution with one electron held fixed at $r = 2$ a.u. (red circle) and angled 30° from the z-axis. Top static-exchange basis. Bottom converged CI basis: notice the Coulomb hole due to dynamic correlation. (b) Corresponding differential photoelectron momentum distribution XUV centered at $3s3p$ with dressing IR then ionizing into the N=3 threshold.

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SNAIL:
A library for fast SFA calculation for HHG in gas targets

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Synopsis We present the open-source Python package SNAIL (SFA Numerical Accelerated Integration Library), which leverages Numpy's efficient matrix operations to swiftly numerically integrate HHG response in atoms with custom driving fields. It allows for the generation of large datasets, which can be used in landscape studies or ML applications.

High-harmonic generation (HHG) is hard to picture without the aid of SFA: decades after its introduction it is still one of the sturdiest theories to describe the interaction of particles with strong laser fields. In 1994, Maciej Lewenstein and others introduced one of the most powerful tools to calculate HHG in a landmark paper [1], remaining more robust than the saddle-point approximation, and much faster than the more accurate time-dependent Schrödinger equation (TDSE) solution for propagation simulations.

The main assumptions of the Lewenstein model are: a) The ground state does not deplete, b) The ground state is the only bound state that affects the evolution of the system, c) The free electron only feels the external field, ignoring the atomic potential. The consequence of these assumptions is the theory is low-order harmonics with energy below the atomic ionization potential are not accurately described.

This allows landscape studies, where simulations are run where many parameters are changed progressively, which would be very costly and time consuming with TDSE. Additionally, with the rising popularity and utility of machine learning (ML) in the field [2], training networks will require quickly generating large training datasets.

However, there is as of yet no standard user-friendly open-source package in Python which implements this. Based on Maximilian Högner's work [3], SNAIL provides a library which can calculate single-atom HHG spectra in less than half a second, as shown in Figure 1. Thanks to a parallelization module, it can also be used in high-performance computing (HPC) setups. Using the Lewenstein SFA, it numerically integrates over the excursion time of the electronic wavepacket; the driving field is fully customizable, allowing for simple beams, and more complex multi-coloured fields; and can be manually inputted as a Numpy array. It also seamlessly connects with current ML libraries, most of which are written in Python.

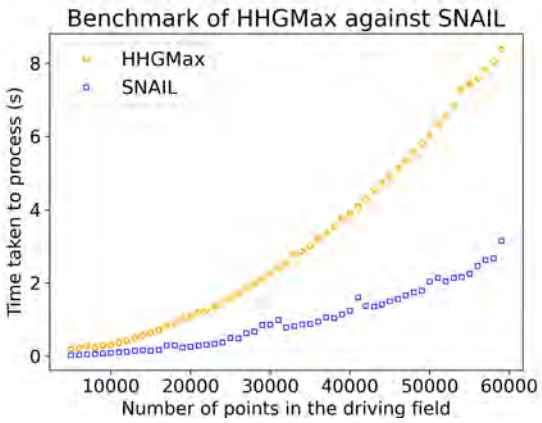


Figure 1. Comparison of processing time for HHGMax (current standard for SFA calculations), against SNAIL. Shows an improvement in performance, especially at higher precision.

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Xenon photoionization in the vicinity of 4*d* giant resonance and Cooper minimum using an XUV-NIR pump-probe experiment at FLASH

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Synopsis XUV-NIR pump-probe experiment was performed on Xe using the reaction microscope at FLASH with 90 eV and 160 eV XUV photon energy and 800 nm NIR wavelength. Transient enhancdement of Xe^{4+} and Xe^{5+} inaccecible with single-XUV-photon absorption are observed and attributed to the transient intermediate states.

The relaxation processes in atomic xenon following core ionization of the 4*d* and 4*p* subshells by extreme ultraviolet (XUV) pulses from a free-electron laser (FLASH: FL26 beamline) are investigated using ion time-of-flight spectroscopy (see [1] for detailed description of the experimental setup). We compare the dynamics following ionization and Auger-Meitner decays at 90-eV photon energy, i.e., near the giant resonance, with those at 160 eV, near the Cooper minimum, where cross sections for photoionization of the 4*d* and 4*p* subshells are similar. Final states with charges higher than 4 show signatures of sequential absorption of two XUV photons, followed by subsequent Auger-Meitner decay. The averaged lifetimes of some important excited states are measured in a two-color XUV-pump–near-infrared-probe experiment (XUV pulse duration is (20±10) fs and 800 nm NIR pulse duration is (15±3) fs). A transient enhancement in the ion yield of Xe^{5+} with an average lifetime of (49 ± 3) fs is obtained, attributed to transient intermediate states following the decay of 4*d* double-core-hole states [2]. This lifetime for Xe^{5+} is twice as short as observed earlier. A different decay channel of Xe^{6+} was observed, most likely arising from NIR-induced double ionization of doubly excited states of Xe^{4+}. This work demonstates that utilizing intense XUV and x-ray sources, such as FEL, enables the selective population of core-hole states and with NIR pulses allows studying decay dynamics in atoms and molecules.

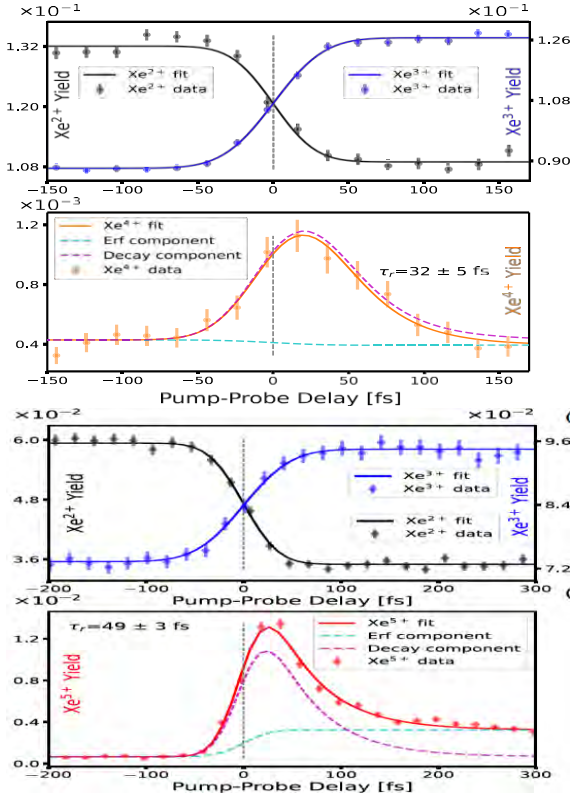


Figure 1. Time-dependent Xe ion yields with XUV photon energy 90 eV (two top panels) and 160 eV (two bottom panels).

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First principles simulation of attosecond XUV pump - XUV probe spectra for small organic molecules

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Synopsis We evaluate attosecond-resolved transient photoionization spectra that unravel the evolution of the ultrafast electron-nuclear dynamics generated by an attoseond XUV pulse in the phenol molecule.

Recently, intense sub-fs soft X-Ray (SXR) pulses became available at the LCLS X-Ray free electron laser (XFEL) [1]. This breakthrough paved the way for the first realizations of all-sub-fs pump-probe studies to unravel the evolution of electronic coherences in molecules with attosecond resolution. These frontier experiments are expected to provide key insights that can not only improve the understanding of molecular charge motion, but also may ultimately lead to its control.

To decipher the transient spectra obtained in these experiments, first-principle modeling of the observable is desirable. However, simulating the individual steps, i.e., (i) ionization by the sub-fs SXR pump, (ii) coupled electron-nuclear dynamics, and (iii) interaction with the sub-fs SXR probe are extremely challenging tasks on their own already. We present here a protocol that allows complete, (i)-(iii), simulations of sub-fs pump-probe spectra in the XUV to SXR photon energy range for small organic molecules. Therein, the molecular bound and continuum states are respectively described with the RASPT2 and B-spline RAS-DFT methods. This allows to simulate the interactions with the pump and probe pulses explicitly. The electron-nuclear dynamics after the pump is modeled with generalization of the the trajectory surface hopping method, launching an ensemble of trajectories in every coherently populated electronic state [2]. We demonstrate our approach by evaluating attosecond-resolved transient photoionization spectra that unravel the evolution of the ultrafast electron-nuclear dynamics in the phenol molecule, see Figure 1, after valence ionization with a sub-fs XUV pulse.

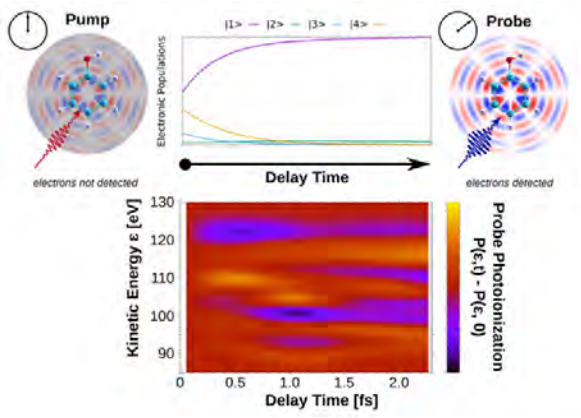


Figure 1. Top: attosecond XUV pump-induced electronic populations along a nuclear dynamics trajectoryin the phenol molecule. Bottom: Corresponding attosecond XUV pump-probe ionization spectrum.

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Optical Parametric MPC: A Hybrid Approach for High-Power Few-Cycle Laser Pulse Generation

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Synopsis We present a novel optical parametric multi-pass cell (OPMPC) scheme that integrates optical parametric amplification within an MPC. Using numerical simulations, we demonstrate few-cycle pulse generation with excellent spatial beam quality and pump-to-signal conversion efficiency exceeding 50%.

Multi-pass cell (MPC) post-compression enables large compression factors, high transmission efficiency, and excellent beam quality, making it key for generating high-power few-femtosecond laser pulses [1]. However, its spectral flexibility and temporal contrast are limited. Optical parametric chirped pulse amplification (OPCPA) offers broader spectral flexibility and higher contrast but suffers from low pump-to-signal conversion efficiency and limited beam quality [2]. Overcoming these limitations is crucial for applications requiring high average and peak power. We propose an optical parametric MPC (OPMPC) amplifier that combines MPCs and OPCPAs. We numerically demonstrated very high amplification efficiency ($> 50\%$), broad bandwidth, and excellent pulse quality..

We simulate the OPMPC in (3+1)D using the nonlinear pulse propagation tool chi3D. [3]. Collinear pump (1.03 µm) and signal (1.6 µm) pulses propagate through a 2.76 m long MPC, while the idler is filtered out every pass to suppress back-conversion. The setup features four 0.15 mm thick $KNbO_3$ [4] crystals spaced 25 mm apart. Complementary crystal orientations mitigate walk-off, while diffraction between crystals compensates for Kerr-lensing. The pump pulses have a 1 ps duration and 1 mJ energy, and the seed spectrum spans 1.3–2.1 µm.After 20 round-trips, the amplified signal reaches conversion efficiencies of more than 50%, approaching the quantum limit of 64%. The remaining offset is due to linear losses of ∼80%, which we have factored into our simulation. Pulse compression via 2nd and 3rd order phase compression yields a 20 fs (FWHM) compressed pulse, close to the 18 fs Fourier limit. The quasi-waveguiding properties of the MPC yield excellent spatial beam quality, highlighting the potential for high-efficiency ultrafast laser systems. As conclusion, we have shown through simulations how parametric amplification and MPCs can be synergistically combined to efficiently generate high quality, high power few-cycle laser pulses.

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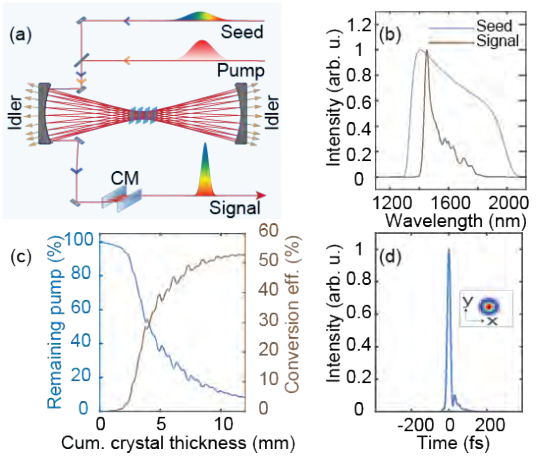


Figure 1. (a) OPMPC schematic. CM: Chirped mirrors. Simulated (b) input seed and output signal spectra, (c) pump depletion and conversion efficiency. (d) Compressed output pulse (20 fs FWHM, FTL: 18 fs). Inset: output signal beam profile.

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High-order Harmonic Generation driven by Vector Necklace Beams

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Synopsis We explore the degrees of freedom available in the design of high-frequency light when driving high-order harmonic generation using vector necklace beams, i.e., light fields with a linear combination of both spin and orbital angular momentum and a discrete symmetry in their transverse intensity and phase profiles.

High-order harmonic generation (HHG) is a highly non-linear optical and parametric process which has been shown to possess the great advantage of conserving and up-converting various properties of light such as energy, linear momentum, orbital angular momentum (OAM), spin angular momentum (SAM), and even the linear combination of OAM and SAM, i.e., generalised angular momentum (GAM) [1]. By adjusting the SAM and OAM of the driving field, it has been both theoretically [2] and experimentally shown [3] that we can tune the SAM and OAM of high-order harmonics via their combined conservation.

According to Noether's theorem, a conserved quantity appears as a result of a continuous symmetry within a given system. The conservation and up-conversion of GAM during the process of HHG therefore requires the driving GAM field to be invariant under a particular kind of symmetric transformation defined in [4] as a coordinated rotation along the propagation axis. This involves a rotation of the spatial dependence by an angle ϕ, followed by a rotation of the polarization vector by a fraction $\gamma\phi$ of that angle.

In the context of a continous symmetry, GAM light fields are invariant under coordinated rotations of any real-valued angle ϕ. However, in the case of a *discrete* symmetry, the specific values that the angle ϕ can take will be restricted. For instance, a necklace-shaped beam in both intensity (see Figure 1) and phase is only invariant under rotations that are multiples of $\frac{2\pi}{N}$ with N the number of necklace lobes. Generating high-order harmonics using beams with discrete symmetries was first attempted in [5] which demonstrated their ability to tune the line-spacing and divergence properties of the harmonics generated.

In this work, we explore the possibility of combining all of these degrees of freedom when designing the structure of harmonics fields and consequently XUV pulses, i.e., SAM, OAM, line-spacing and divergence, by driving HHG using vector necklace beams which possess both GAM and a discrete symmetry in their transverse intensity and phase profiles.

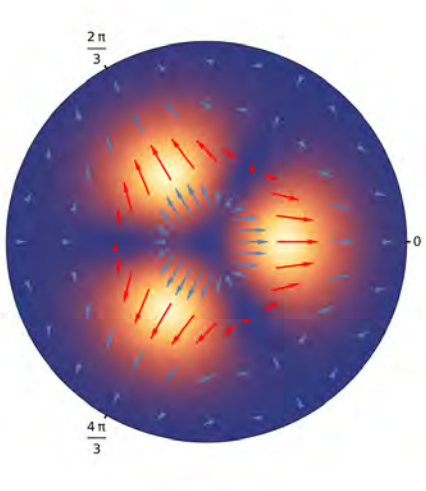


Figure 1. Example of the transverse intensity profile of a vector necklace beam with three lobes ($N = 3$) with varying linear polarisation orientations across the transverse plane arising from its GAM content.

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Direct Optical Characterisation of a Driving Field with a Time-Dependent Ellipticity for High Harmonic Generation

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Synopsis HHG sources can be greatly enhanced with complex single and multicolour fields, particularly with complex polarisations. However, to properly use these fields it is important to be able to characterise them using direct optical methods before the generation of high harmonics. Here, we discuss the use of the POLLIWOG method to measure ellipticity dependent laser fields for the generation of high harmonics and its potential extension to measure multicolour complex polarisations.

High Harmonic Generation (HHG) can be controlled using complex driving fields. Most simply, generating harmonics with a linear ω field and a linear co-polarised or orthogonal 2ω field allows for a breaking of the interaction symmetry, generating both odd and even harmonics [1]. By changing the polarisation across a single pulse of the driving field, further control can be achieved. Taking advantage of the fact that circular polarisations do not produce harmonics, the ellipticity gating technique [2] was proven to efficiently confine the harmonic emission to one half optical cycle in such a way that, rather than producing a train of attosecond pulses via HHG, an Isolated Attosecond Pulse (IAP) is produced [3]. Extending this into two colour fields, by overlapping two oppositely circularly polarised ω and 2ω fields, a field with 3 fold symmetry can be produced. This changes the selection rules of the produced harmonics [4], allowing for circularly polarised harmonics. Further control of the ellipticity of these two fields can provide control over these rules [5] providing an interesting route to tailoring the laser driver. These fields have been mostly indirectly characterised with the use of HHG as a highly nonlinear signal. However, from the perspective of tailoring the laser field to provide a versatile HHG source, it is necessary to develop a robust optical method to directly measure these complex laser fields that does not rely on using the produced XUV radiation.

Here we present efforts towards a direct optical measurement of these fields, inspired by the *POLLIWOG* method [6]. This method relies on the use of spectral interferometry, combined with a pre-characterised reference pulse, to obtain the phase difference between the reference and unknown pulses. By interfering a linearly polarised pulse at 45° to the horizontal and measuring two orthogonal components of the field, the full, time dependent spectral phase of the pulse can be retrieved and therefore the full form of the pulse. We will discuss how this method can be extended to two colour fields.

This work was supported by funding from the STFC XFEL Hub program.

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Attosecond pulse trains generation in hollow core waveguides

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Synopsis Macroscopic numerical simulation describes the formation of attosecond pulse trains in hollow core waveguides filled with He and irradiated with 25 fs pulses at 800 nm central wavelength. High order harmonics are selected in this study, and their time structure reveals the role of off-axis contributions generated in propagation along the waveguide. Valuable parameters for optimizing the generation conditions are derived, including the waveguide geometry and the irradiation conditions.

High order harmonic generation (HHG) in gas media provides the leading method to produce attosecond pulses, both isolated or in trains. The experimental configurations used to host the gas vary from static cells, gas (mono or multi-) jets and hollow core waveguides (HCW), which confine the gas density and the driving laser field intensity over long distances. The latter configuration was proven to significantly increase the HHG yield.

Modeling HHG in HCW complements the experimental data and helps their explanation. We use a dedicated 3D non-adiabatic model [1] specifically developed to reproduce the experimental generation configuration reported in Ref. [2]. We consider HHG by a 0.4 mJ pulse being injected into a HCW of 8 mm length and 130 μm diameter, filled with 850 Torr of He. The goal is to explore the formation of the attosecond pulse train (APT) in this macroscopic volume considering the spatial and temporal structure of the generated harmonics, which are dependent on the driving field intensity and phase along the waveguide.

The radial and axial dependence of the driving field inside HCW influences the intensity and phase of the single atom response and, therefore, the final harmonic yield. Moreover, the gas distribution along the waveguide affects the macroscopic polarization of the medium, and thus the overall HHG efficiency.

In Figure 1 we present the time structure of the total harmonic field filtered in the cut-off region (H65-H85). The time structure of the on-axis far field is also plotted and the comparison of the two quantities gives us an estimation of the off-axis contributions to the total field, which are shifted in time and are characterized by a lower intensity.

The present case assumes the best coupling condition where the beam focus is placed at the entrance plane and the beam waist w is $w=2a/\pi$, a is HCW radius.

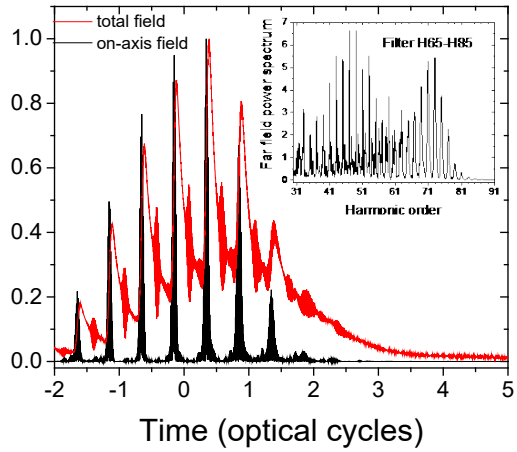


Figure 1. Attosecond pulse train generated as described in the text. On-axis and total far field is plotted, the far field power spectrum being shown in the inset.

However, the model allowed us to capture how the harmonic build-up during propagation is strongly influenced by the focus position with respect to the HCW entrance plane, leading to a substantial modulation of the time and frequency structure of the harmonic field. This parameter as well as the density profile proved to be important options in optimizing HHG efficiency and cut-off extension.

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XUV chromatic aberrations in high order harmonic generation controlled by flat-top spatial shaping of the laser driver

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Synopsis We study chromatic aberrations in high order harmonic beams generated in Argon with a intense Titanium Sapphire laser. We observe strong chromatic aberrations when harmonics are generated with a Gaussian shaped driving beam with significant longitudinal separation of the harmonic foci. We show that these aberrations can be significantly reduced when harmonics are generated with a driving beam shaped to a flat top spatial profile near the focus where the gas jet is located. Similar results were obtained with harmonics emitted in a gas jet or a gas cell.

XUV beams generated via high order harmonic generation (HHG) in gases have spatial properties evolving with the harmonic order. Chromatic aberrations manifest when the harmonics are focussed so that, near focus, the spectral content can change significantly during propagation [1 - 3].

We study the spatial properties of harmonics [4] generated with a Gaussian beam driver and focussed with grazing incidence XUV mirrors. We observe that the harmonics are not all focussed at the same position with a relative focus displacement along the longitudinal coordinate, z, comparable to the harmonic confocal parameter. Hence, spectral content, harmonic dephasing and attosecond temporal profile evolve during propagation. We compare these properties with those from harmonics generated with a spatially shaped driving beam having a flat top intensity profile. With flat top driver, the XUV beams are less divergent and the foci shift are less pronounced.

To quantify the impact of the chromatic aberrations in both configurations [4], we considered the evolution of the spectral width, relative harmonic dephasing and attosecond temporal profile on the beam axis (r = 0) vs Z. We observe that the spectral width near focus can be significantly smaller than the bandwidth estimated at infinity. To extract a temporal profile, we considered harmonic 11 to 19 with the amplitudes calculated at r = 0 and assumed that they have no atto chirp far from focus.

Figure 1 shows the evolution of the pulse temporal profile with Z. The difference between the attosecond temporal profiles obtained with the flat top beam or the Gaussian beam are striking. While the temporal profile is very regular with flat top driver, the pulse changes significantly during propagation when a Gaussian driver is used. Similar results were obtained with harmonics generated in a jet or in a 1 cm gas cell. Simulations also show that XUV chromatic aberrations can be significantly reduced by spatially shaping the fundamental driver [4].

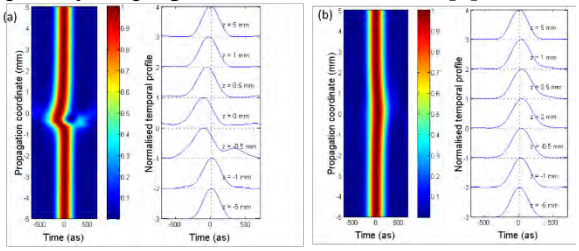


Figure 1. Evolution of normalized on-axis intensity profile during propagation when harmonics 11 to 19 are generated with (a) a Gaussian driver or (b) a spatially shaped flat top driver. The estimated temporal profile is shown near the XUV focus for z = -5, -1, -0.5, 0, + 0.5, +1 and + 5 mm assuming no attosecond chirp at large z..

This work is supported by Laserlab Europe, grant agreement 654148.

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Tailoring XUV Sources for Ultrafast Science: Advances at ELI Beamlines

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Synopsis High Harmonic Generation (HHG) enables compact, coherent XUV sources for ultrafast studies but faces challenges in photon flux and spectral matching with target applications. Advances in high-power OPCPA based lasers, such as ELI Beamlines' L1 (45 mJ, 15 fs, 1 kHz), enhance XUV yield and enable high XUV-intensity experiments. We present an advanced toolbox for tailoring XUV beam parameters to specific applications, including a high-repetition soft X-ray laser at 41.8 nm. Additionally, we highlight prospects for XUV-XUV pump-probe experiments.

High Harmonic Generation (HHG) is a key technique for generating compact, coherent extreme ultraviolet (XUV) radiation, essential for studying ultrafast atomic and molecular dynamics. However, limited XUV photon flux and spectral mismatches with target electronic structures present challenges for nonlinear XUV interactions.

Recent advancements in high-power Optical Parametric Chirped-Pulse Amplification (OPCPA) based lasers, such as the L1 laser at ELI Beamlines (45 mJ, 15 fs, 1 kHz), significantly enhance XUV photon yields [1]. Furthermore, a synchronized auxiliary beam with an electronically adjustable delay is in development, which will enable XUV-XUV pump-probe experiments. The independent nature of the two driving beams opens possibilities for using two separate XUV sources, expanding opportunities in nonlinear XUV physics.

To tailor XUV characteristics for specific applications, we introduce two HHG process modifications that achieve spectral control without the need for XUV optics, minimizing energy losses and pulse distortions. The first approach enables tunable single-frequency XUV generation by modulating the primary driving laser and its second harmonic, allowing 2 eV tunability within the 20.4–22.4 eV range [2]. The second approach enhances spectral selectivity through medium density modulation and a Bessel-Gauss driver beam [3].

Demonstrating the versatility of our HHG Beamline, we present a kHz soft X-ray laser (SXRL) at 41.8 nm, achieving the highest repetition rate reported for plasma-based SXRL sources [4]. Additionally, we have implemented a routine online XUV energy measurement method [5]. We present details of our system's design, methodologies, and recent results, from conceptual development to practical applications.

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High harmonic generation control by sub-cycle multi-color waveform synthesis of an ultraviolet to near-infrared driver

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Synopsis We aim to experimentally control high harmonic generation using a three-color synthesizer consisting of the fundamental (800 nm) with it's second and third harmonic. Current results for synthesized waveforms of the fundamental with the third harmonic show stable sub-cycle control over harmonic yield. By addition of the second harmonic, we predict to further increase the tunability of the ultraviolet to near-infrared waveform to enhance the control over the harmonic yield modulation.

High harmonic generation (HHG) provides a tabletop source for coherent extreme ultraviolet (XUV) to soft x-ray radiation, with applications in metrology and spectroscopy.

A major drawback of HHG is the low conversion efficiency (CE) of the driving laser into the generated radiation. Fully optimized HHG based XUV sources from gases can reach CE of 10^{-5} per harmonics up to 60 eV [1]. In solids the efficiency has been shown to be at least two order of magnitudes lower, but also that the CE can be increased by an appropriate combination of the driving wavelengths [2]. HHG CE benefits from higher intensities and shorter wavlengths of the driver. However, maintaining ideal phase matching between subsequent halfcycles limits the maximum intensity. Alternatively, shorter driving wavelengths reduces the maximum generated photon energy.

Altering the driving laser waveform by coherently combining multiple frequencies has been shown to increase harmonic yield while preserving phase matching and the cut off frequency by enhancing the electric field strength at ionization times of the short trajectories. Additionally, waveform synthesis can alter electron excursion times allowing spectroscopy of the parent ion at varying times [3]. Lastly, harmonic deactivation as function of relative phase between the multiple colors can enable below diffraction limit microscopy [4].

This research aims to realize a highly stable tunable three color waveform synthesizer of the fundamental (800 nm), second and third harmonic of a Ti:Sapphire laser, with pulse energies of 1.20 ,0.40 and 0.06 mJ respectively. Initial two color experiments shows stable relative phase control, within 50 as, between the fundamental and third harmonic. Harmonic modulation up to 60% was observed as function of phase delay and photon energy, see figure 1.

With the addition of the second harmonic the tunability of this ultraviolet to near-infrared waveform synthesized driver can be pushed beyond what, to our knowledge, has been experimentally realized before.

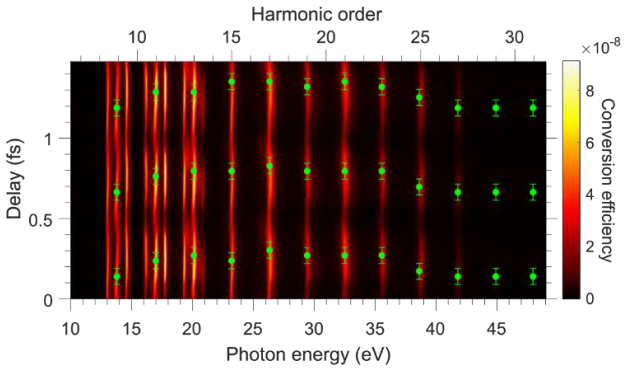


Figure 1. HHG CE of harmonics generation in Argon by a two color field of the fundamental and third harmonic. Green points show the maximum signal per harmonic within a half cycle of the third harmonic.

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Generation and characterisation of optical attosecond pulses with terawatt peak power

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Synopsis Optical attosecond pulses can be directly generated through soliton self-compression. Here we describe the generation and characterisation of TW-scale sub-femtosecond pulses using our high-energy XSOL (Extreme Soliton) beamline and an in-vacuum dispersion-free TIPTOE apparatus. We obtain 2.1 mJ pulses with an envelope full-width at half-maximum duration of 0.77 fs and a peak power of 1.36 TW.

Optical attosecond pulses – pulses with sub-femtosecond duration but with a spectrum in the near-ultraviolet to infrared spectral range – have only been generated through two techniques: light-field synthesis [1] and soliton self-compression [2]. Such pulses are both hard to create and hard to measure. So far, they have also only been achieved with relatively low peak power: sufficient for ultra-high time resolution measurements in the perturbative regime but insufficient for driving relativistic intensity strong-field physics.

In this work, we describe the generation and characterisation of sub-femtosecond pulses with terawatt peak power using our high-energy XSOL (Extreme Soltion) beamline and an in-vacuum dispersion-free TIPTOE (tunneling ionization with a perturbation for the time-domain observation of an electric field) apparatus [3, 4]. The full XSOL beamline is described in a separate submission and represents a ten-fold energy up-scaling compared to our previous soliton source [2]. The soliton-compression stage consists of a 4.7 m long, 530 µm core diameter hollow capillary fibre, pumped with 5.5 fs, $\sim$ 3 mJ pulses. The input and output of the fibre are in vacuum, while helium gas is fed into the centre of the fibre with the pressure tuned to obtain soliton self-compression at the output.

For temporal characterisation, we constructed a custom in-vacuum TIPTOE apparatus, similar to [3, 4]. The output of the fibre and the entire TIPTOE apparatus are contained within a single large vacuum system. The characterisation chambers are filled with a few mbar of helium to act as the gas medium for the TIPTOE measurement. Figure 1(a) shows the measured pulse envelope and the square of the electric field. We obtained a full-width at half maximum envelope duration of 0.77 fs for a 2.1 mJ self-compressed pulse energy, corresponding to 1.36 TW peak power (34× Ref. [2]). The squared electric field transient has a duration of just 313 attoseconds, which represents, to our knowledge, the shortest ever generated and characterised optical pulse. Figure 1(b) compares the spectrum of these pulses obtained from TIPTOE to that measured with an independent spectrometer, confirming the fidelity of our measurement.

Our next goal is to tightly focus these pulses and use them to drive relativistic nonlinear optics in the sub-cycle regime for the first time.

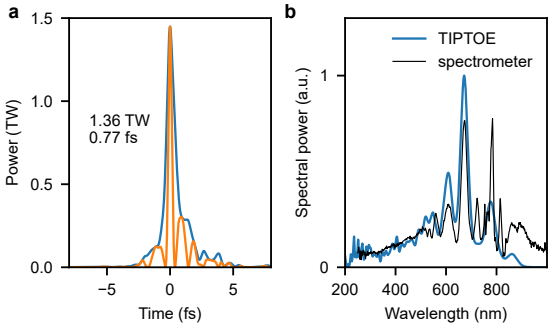


Figure 1. (a) Retrieved pulse envelope (blue) and absolute square of the electric field transient (orange). (b) Corresponding spectra.

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Generation of ultra-broadband millijoule-level radial vector beams

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Synopsis We demonstrate the generation of ultra-broadband radial vector beams. Through the use of soliton self-compression dynamics in gas-filled hollow-core fibres on the XSOL (Extreme Soliton) beamline, mJ-level pulses with multi-octave spectra reaching down to 140 nm are generated. Characterisation of the output polarization state shows that the radial polarization of the input pulses is transferred to the multi-octave spectrum.

If radial vector beams are tightly focused, the resulting longitudinal field in the propagation direction can be used to accelerate electrons [1]. In order to maximise the peak intensity at focus, either the pulse energy has to be increased or the pulse duration has to be decreased. When using segmented waveplates for the conversion of linearly polarised light to radial vector beams, the limited bandwidth of these optics sets a limit to the minimum pulse duration. While previous work has overcome this limit by spectrally broadening radially polarised pulses in gas-filled hollow-core fibres [2], the minimum pulse duration in that case is still at the few-cycle level, due to the limited bandwidth of chirped mirrors. Our work aims to overcome these limits by self-compressing radially polarised light in gas-filled hollow-core fibres using soliton dynamics [3].

The beamline used is called XSOL (Extreme Solitons), a three-stage hollow-core fibre setup that is capable of generating sub-cycle TW pulses that cover a multiple-octave spanning spectrum, reaching into the vacuum ultraviolet. In brief, it is built of three helium filled fibre stages. The first two stages compress 40 fs linear polarised pulses down to 5.5 fs with pulse energy up to 5 mJ. These pulses are sent into the third stage, where soliton dynamics cause not only spectral broadening but also temporal self-compression [3], making the generation of TW pulses with sub-femtosecond duration possible.

In the current work, before sending the pulses into the soliton stage, they are converted to radially polarised beams with a segmented achromatic waveplate. This input state preferentially couples to the TM_{01} mode of a 4 m long, 530 µm core diameter, hollow-core fibre that is filled with helium from a central pressure point. These pulses experience soliton dynamics, causing their spectral broadening and temporal self-compression. The wavelength of the generated ultraviolet light is dependent on the gas pressure. For 1.4 bar it reaches the deep ultraviolet, as shown in Figure 1. By reducing the pressure even further, the spectrum extends into the vacuum ultraviolet, reaching down to 140 nm so far.

This spectral bandwidth, together with an output pulse energy of 1.4 mJ, allows the generation of TW-scale, sub-femtosecond, radially polarised beams. While the full temporal characterisation for radial polarisation is still pending, this claim is supported by our recent characterisation of 0.77 fs, 1.36 TW self-compressed linearly polarised pulses generated on the same beamline.

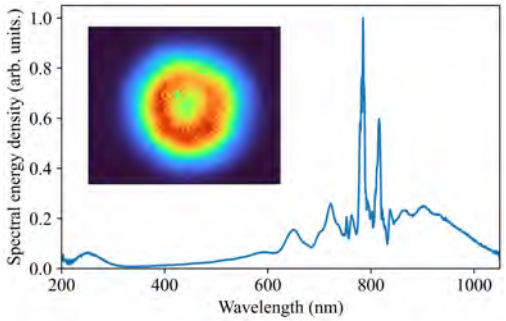


Figure 1. Spectrum of our radially polarised beam for a gas pressure of 1.4 bar, keeping the spectral extension within the deep ultraviolet. The inset shows the corresponding full output beam profile.

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Commercial table-top beamline for attosecond science

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Synopsis We present compact table-top commercial beamline, consisting of nonlinear post-comporession stages, high-harmonics generation chamber, pump-probe re-focusing and delay line with attosecond resolution and VUV-XUV spectrometer. The beamline is compatible with various lasers, based on Ti:Sapphire or high-repetition rate Yb-technology.

Since 2009, UltraFast Innovations (UFI®) has provided a wide range of specialized and customized optics and components, as well as instrumentation for ultrafast optical science. Continuous development and extensive collaboration with Professor Krausz's research group (Nobel Prize in Physics, 2023) have enabled UFI to bring to market a series of complete solutions for pump-probe spectroscopy, XUV imaging, pulse compression, and the generation of coherent secondary radiation[1],[2].

A prominent example of such a solution, encompassing most of the company's expertise, is an XUV beamline driven by a high-repetition-rate, industrial-grade laser. The beamline includes post-compression stages based on nonlinear spectral broadening and in-house-produced high-dispersive mirrors (SAVANNA), a beam stabilization system, a high-harmonics generation chamber (NEPAL), a programmable IR-XUV delay and refocusing stage (K2), an XUV polarization converter (AURORA), and a VUV-XUV spectrometer (EVEREST). The performance of each module is optimized and tailored for specific applications in ultrafast optical science.

In this presentation, we will introduce a compact tabletop XUV beamline and discuss the parametric space of the NEPAL and SAVANNA instruments, including the high-power version (HP), developed in collaboration with the Institute for Nanophotonics IFNANO in Göttingen. This new version of the post-compression module enables both peak and average power of the laser system to reach extreme values of 400 GW and >100 W simultaneously, with up to 30-fold spectral broadening[2]. In combination with in-house-developed ultra-broadband high-dispersive mirrors with third-order dispersion (TOD) compensation, it provides a platform for generating few-cycle pulses, as demonstrated, for example, in Ref. [3].

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High-power and ultrashort IR-driver platform for industrial coherent XUV metrology

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Synopsis We report on the development of a robust 100W and 35fs IR platform to efficiently generate coherent XUV radiations down to 140eV. The secondary photon flux is used to performed ptychographic imaging of nanometric structures on semiconductor-wafers.

Over the past few years, the emergence of extreme ultraviolet (XUV) lithography has allowed to drastically downsize the spatial scale of semiconductor structures. This has pushed Moore's Law further and allowed the emergence of plethora of new possibilities, such as the advent of artificial intelligence. While EUV lithography scanners are operated for high-volume manufacturing (HVM), traditional optical metrology platforms are not able anymore to resolve the printed nanometer-scale structures, preventing yield optimization and scaling. This is why XUV light sources become more and more relevant for novel at-resolution metrology concepts, including lithography mask inspection, device overlay and CD (crititcal dimension) metrologies, and various applications in lithography research. Amongst other, lensless coherent diffraction imaging of a XUV light source has appears as a versatile tool to characterize these structures.

We report here on the development of an integrated IR-driver designed to support coherent XUV generation through high-harmonics generation (HHG) around 13.5nm and wafer metrology at customer side. The IR sources allow to generate ready-to-use and reproductible high-flux XUV light for wafer' nanostructures characterization with typical move-acquire-measure times of 1-240 seconds [1].

The IR-source is based on an industrial, high-power and high repetition rate 100W, 200kHz, 500fs Yb-doped laser amplifier combined with a downstream gas-filled multipass cell [2] for efficient pulse shortening. The source also includes built-in metrology sensors and actuators to provide in-depth survey and remote control of crucial parameters, ensuring continuous operation with reduced downtime. The overall system has a high transmission efficiency of >95% transmission, corresponding to >95W output power supporting the generation of 20-40fs pulses with pristine output beam quality of M^2<1.2. When focused into a gas jet, it allows the generation of high-brightness, µ-Watts level, XUV spectra spanning around 8-20nm with good stability to perform wafer control measurements.

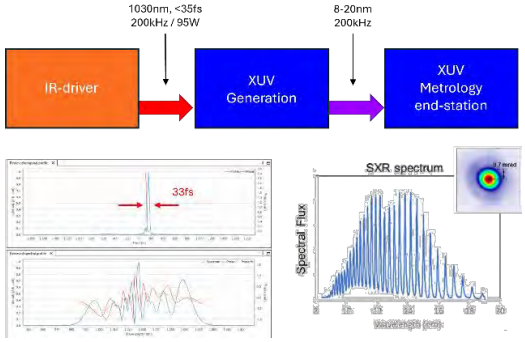


Figure 1. Sketch of the setup (top), and temporal and spatial measurement at the IR-driver output (bottom).

We will present the IR source design with emphasis on source parameters stabilities and reproducibility in an industrial environment. We will also show examples of nanostructures characterizations performed on customer side. Finally, we will and look at the power scaling capacities of such sources for HVM in-line metrology compatible with always increasing throughput.

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Post-compression of an Yb laser in two steps for attosecond transient absorption spectroscopy

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Enhancing laser repetition rates using Yb-based systems is an opportunity for attosecond science. This development is restrained by the long pulse duration provided by these systems and a priori ill suited to generate attosecond pulses. Efficient post-compression technics[1,2] based on self-phase modulation is an efficient possibility to make these sources suited for attosecond science. In this study, we employ a two-step system[3] to post-compress pulses at the millijoule level, reducing the pulse duration close to the single cycle regime, with an overall transmission efficiency exceeding 45%.

The experimental setup is shown in Fig. 1 (top panel). A carrier-envelope-phase (CEP)-stabilized Yb-doped laser system (Flint and Carbide, Light Conversion) is employed. This laser delivers pulses with an energy of 2 mJ, a pulse duration of 330 fs, and an average power up to 80 W. Post-compression is achieved through two successive stages. In the first stage, spectral broadening is induced via 14 round trips in a homemade argon-filled multipass cell (MPC) [4]. The MPC is 590 mm long and includes mirrors with a 300 mm radius of curvature and low group delay dispersion (GDD). Then, the laser pulse is compressed near its Fourier limit using chirped mirrors (-1720 fs²) and subsequently coupled into a 2.1 m long stretched hollow-core fiber with a helium pressure gradient. Pulse characterization at various stages is performed using a single-shot frequency-resolved optical gating (FROG) device, an imaging spectrometer commercially available (Femtoeasy) and a homemade TIPTOE [5].

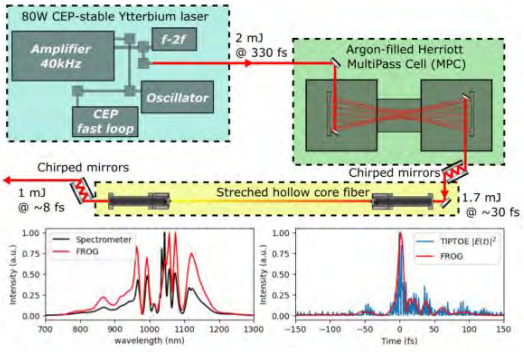


Figure 1. Top: layout of the experiment setup. Bottom: measured spectra and time trace at the hollow-core fiber output.

Significant spectral broadening is achieved at the MPC output by adjusting the argon gas pressure. Under optimal conditions, the spectral bandwidth reaches a maximum of 140 nm, allowing for the generation of 20 fs pulses, corresponding to a compression factor of K=16. The MPC also demonstrates a high transmission efficiency exceeding 90%.

In order to seed the second post-compression stage, the argon pressure in the MPC is tuned to produce 25 fs pulses with a well-defined temporal profile. The second stage reaches a transmission efficiency of 60% in the hollow-core fiber, delivering pulses with an energy of 960 µJ and a duration of 7 fs (Fig. 1 bottom right). By further increasing the gas pressure, we attain a maximum spectral bandwidth that supports Fourier-limited pulse durations as short as 4 fs.

This two-stage hybrid compression system achieves a total compression factor of 47, paving the way for the generation of isolated attosecond pulses. Moreover, the exceptional stability of this setup offers the potential to incorporate an additional nonlinear stage, such as a short hollow-core fiber, for the generation of tunable vacuum-ultraviolet (VUV) and deep-ultraviolet (DUV) [6] pulses.

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The Effect of Laser Pulse Duration On The Yield Of High Harmonic Generation

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Synopsis Since the emergence of high-order harmonic generation, one of the primary objectives has been to understand and enhance harmonic yield. Here, we present a long-overdue experimental study on the impact of the Fourier transform-limited driving laser pulse duration on harmonic yield. Our findings reveal that, at a constant laser intensity, the harmonic yield increases linearly with pulse duration.

With the advent of modern OPCPA-based lasers, shorter and more powerful laser systems have become available for high-order harmonic generation (HHG). To fully harness these high-energy laser pulses, experimental parameters such as pulse energy, focusing geometry, and gas pressure must be scaled accordingly to maintain optimal conditions for maximum yield across the spectral bandwidth. While several studies have explored different scaling approaches [1, 2], the effect of pulse duration on HHG yield has primarily been investigated for pulses longer than 20 fs [3, 4], leaving the behavior of ultrashort pulses largely unexplored. In this work, we experimentally and numerically investigate the dependence of HHG yield on laser pulse duration while maintaining constant laser intensity at the focal spot, as illustrated in Figure 1. We conducted a series of macroscopic HHG simulations using a three-dimensional (3D) non-adiabatic model [5, 6] across various pulse durations while keeping laser intensity fixed. To validate these simulations, we carried out experimental measurements using the SYLOS Experimental Alignment (SEA) laser at the ELI-ALPS research facility. The laser system delivers a peak performance of 50 mJ at 12 fs, longer pulses were created by limiting the bandwidth while ensuring they remained at their Fourier transform limit. High-order harmonics were produced in a gas cell under loose focusing conditions on the SYLOS LONG beamline, and the resulting spectra were recorded using a commercial imaging XUV spectrometer.

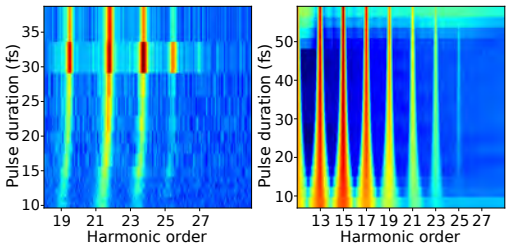


Figure 1. Experimental (left plot) and simulated (right plot) harmonic spectra as the function of laser pulse duration for the 19th to 27th orders generated at same laser intensity.

The plot clearly shows that longer pulses generate a higher harmonic yield at the same laser intensity while producing narrower spectra. This can be attributed to the multicycle nature of the laser, where each half-cycle contributes to the harmonic yield. As demonstrated in [4], shorter pulses require higher laser energy to reach ionization saturation of the medium, leading to lower yield under conditions optimized for longer pulses.

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Single-Shot Carrier-Envelope Phase Measurement at 586 kHz Using Optical Fourier-Transform Interferometry

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Synopsis In this work, we demonstrate single-shot and every-shot measurements of the carrier-envelope phase (CEP) of ultrashort pulses at a repetition rate of 586 kHz. Synchronous measurements using a commercial CEP detector, operating at a lower sampling rate (117 kHz), show excellent agreement, validating the accuracy and reliability of our approach.

In the few-cycle regime, the absolute phase of ultrashort light pulses, known as the carrier-envelope phase (CEP), plays a crucial role in controlling light-matter interactions, such as the generation of isolated attosecond pulses [1]. Precise, single-shot CEP detection is therefore essential, but particularly difficult at high repetition rate. We have recently demonstrated a novel, all-optical, single-shot (and every shot) method to measure the CEP of ultrashort laser pulses at a repetition rate of 200 kHz [2]. In the present work, we extend the measurement speed to a repetition rate of 586 kHz, which to our knowledge is the fastest reported CEP measurement in single-shot.

CEP is commonly encoded in the form of spectral fringes in f:2f spectra (Fig. 1a, left panel). Our technique employs optical Fourier transform, implemented via filtering the f:2f spectrum with optical elements that have a spectrally periodic transmission function (Fig. 1a, middle panel). The CEP is then simply determined as the arctan of two photodiode signals (Fig. 1a, right panel), a measurement that can in principle be performed at many MHz repetition rate up to the speed of common ultrafast oscillators. Our approach effectively circumvents bottlenecks limiting conventional CEP detectors, such as the need for high-speed spectrometers and the time required for numerical Fourier transform operations [2].

We also have performed synchronized measurements with a commercial, high-speed CEP detector (Sphere Ultrafast Photonics), based on a fast CMOS linear sensor for measuring the f:2f spectra and numerical FFT to calculate the phase of the fringes. A CEP-stable Pharos laser (Light Conversion), was measured at the full repetition rate of 586 kHz with the high speed detector (indicated as "analog" in Fig. 1b), while every fifth pulse (117 kHz) was sampled with the commercial detector (indicated as "digital" in Fig. 1b). Unlike previous correlation measurements [2], here two completely independent CEP detectors, each with its own white-light generation and f:2f stages, were used. Fig. 1b shows a time interval of 500 μs of the two synchronized detectors with an arbitrary CEP offset between them (f:2f is not sensitive to absolute phase). The agreement is excellent, indicating at least similar performance as compared to conventional CEP detectors.

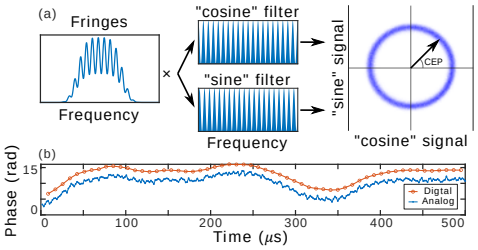


Figure 1. (a) Principle of obtaining the phase of f:2f spectral fringes via optical Fourier transform. (b) Comparison with the commercial CEP detector.

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Over-Barrier Ionization Dynamics Studied by Backpropagation

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Synopsis Tunneling ionization (TI) and over-barrier ionization (OBI) are the primary ionization mechanisms under intense lasers. While TI is extensively studied, OBI dynamics has remained elusive, since prevailing theories are derived from strong-field approximation assuming zero-range potentials that don't support occurrence of OBI. Our study extends the backpropagation method to include OBI, allowing us to distinguish TI and OBI contributions to PMD and ionization times, revealing that OBI depends on both field strength and initial transverse momentum.

Tunneling and over-barrier ionization are the primary processes of strong-field ionization of atoms and molecules. While the dynamics of tunneling ionization have been extensively studied, exploration of over-barrier ionization dynamics has remained a significant challenge. In this study, we investigate the dynamics of over-barrier ionization using the backpropagation method specifically adapted for this context. By analyzing the topology of the backpropagating trajectories, we differentiate the contributions of tunneling and over-barrier ionizations to the distributions of photoelectron momentum and ionization time. While the transition from tunneling to over-barrier ionization is known to depend on the field strength, our results reveal that it is also influenced by the initial transverse momentum of the outgoing electron (see Figure 1). When the field strength exceeds the threshold required for over-barrier ionization to occur, over-barrier ionization is observed at low initial transverse momenta. In contrast, at higher transverse momenta, tunneling ionization remains the dominant process. In addition, we clarify how ionization probabilities vary with intensity for each mechanism, highlighting a competitive relationship between them. We further find that accounting for the Stark shift is essential for accurately determining the threshold field strength for over-barrier ionization. Our work provides a detailed understanding of the dynamics of over-barrier ionization and lays the groundwork for exploring new mechanisms in intense laser-matter interactions [1].

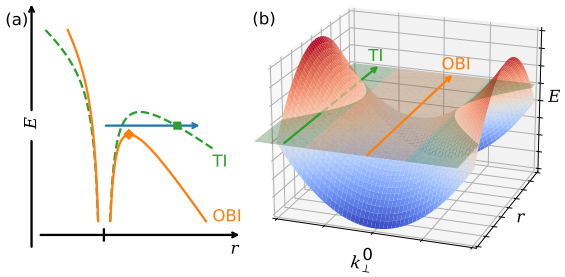


Figure 1. (a) Sketch of tunneling ionization (TI) and over-barrier ionization (OBI). (b) Effective potential barrier as a function of radial position r and initial transverse momentum $k_\perp$. Green and orange arrows represent TI and OBI, respectively.

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Optimizing Harmonic Generation Yield and Refocusing Conditions

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Synopsis
Controlling the wavefront curvature of the infrared (IR) driving laser pulse is essential for optimizing the properties of high-order harmonics. The approach in this work is to maximize the yield and improve the focusing conditions, important for reaching high intensity. We analyze the effect of the IR beam wavefront at the generation position on the XUV beam properties. Additionally, we calculate the conversion efficiency for different generation positions. Combining these approaches gives us insights about the XUV beam size and energy and thus about the accessible intensity.

Studying electron motion in molecular dynamics requires attosecond time resolution. In addition, high intensity is needed to induce nonlinear processes such as double photoionization, or to perform XUV-XUV pump-probe spectroscopy. Achieving such conditions necessitates precise control over the high-order harmonic generation (HHG) process, for example, by controlling the infrared driving beam profile.

At the Intense XUV Beamline in Lund, we control the IR wavefront to optimize XUV generation. Our analysis is based on a Gaussian optics model for the IR and XUV pulses, presented in [1] and based on the model introduced in [2]. This model evaluates the effects of the generation positions and aberrations [3,4] of the driving pulse on the XUV pulse properties. We adjust the IR beam wavefront to allow for ideal refocusability in the targeted system.

Figure 1 shows the beam waists of the harmonics as a function of the distance between the IR focus and the generation medium. The intensity at the generation position is kept constant at $I = 2 \cdot 10^{14}\,\mathrm{W/cm^2}$. In our calculations, we assume a fundamental phase transfer to the harmonics wavefronts and we also take into account the contribution of the dipole phase. The beam waists of the harmonics and their longitudinal source positions are then retrieved by the derivation of their radius of curvature.
It becomes evident here that by generating out of focus we can minimize the beam waist of the harmonics. In addition, the harmonics tend to be created all at the same position, minimizing also the chromatic aberrations.

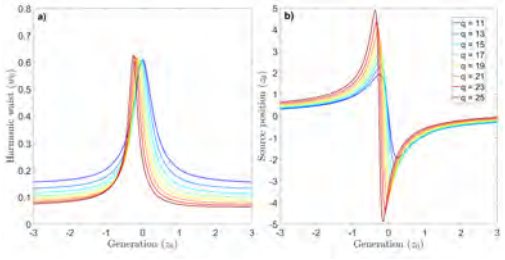


Figure 1. a) Beam waists of Harmonics 11 to 25 generated in Argon, in unit of the beam waist of the fundamental (w_0), and b) harmonics source positions, as a function of the generation position in unit of the Rayleigh length of the fundamental (z_0).

These generation conditions are also inspected via phase matching calculations, exploring the conversion efficiency (CE). The CE is proved to be maximal for precise regimes of pressures and gas lengths [5].

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KeV attosecond pulse generation driven by MIR lasers through waveform control and phase matching

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Synopsis We theoretically investigate generation of keV isolated attosecond pulses (IAPs) through high-order harmonic generation in a gas cell, utilizing coherent control of waveform and mid-infrared laser propagation. By combining a few-cycle, carrier-envelope-phase-controlled 4-μm driving laser with a single-cycle weak 400-nm laser, we achieve the generation of bright keV IAPs in a helium gas cell through the synergy of phase matching and two-color waveform control. In contrast, using only a single 4-μm driving laser makes IAP generation challenging even under favorable phase-matching conditions.

Generating isolated attosecond pulses (IAPs) in the keV X-ray regime through high-order harmonic generation (HHG) requires the use of mid-infrared (MIR) or long-wavelength infrared (LWIR) driving lasers, as the high harmonic cut-off energy scales with the square of the driving wavelength [1, 2]. However, due to the low HHG conversion efficiency associated with long-wavelength driving lasers [3], control techniques such as phase matching optimization and waveform control are essential. Recently, Chang [4] theoretically explored HHG in the keV X-ray regime driven by an LWIR laser. Single-atom simulations in a helium gas demonstrated that bright high-order harmonics with ultrashort pulse durations can be generated by coherently combining a strong LWIR laser (8 μm) with a weak, single-cycle, carrier-envelope-phase (CEP) stable near-infrared (NIR, 800 nm) laser. The enhancement of HHG is driven by controlling the ionization, acceleration, and recombination processes using a two-color laser field. However, research on generating keV IAPs with MIR and LWIR lasers through macroscopic propagation in gas media remains limited. In this study, we theoretically investigate the generation of keV IAPs through HHG in a helium gas cell by leveraging coherent control of waveform and propagation of few-cycle MIR laser pulses. We begin with single-atom simulations and then identify phase matching conditions for enhanced keV HHG, achieved by combining a few-cycle, CEP-controlled 4-μm driving laser pulse with a single-cycle weak 400-nm laser pulse. Next, we extend our simulations to HHG propagation in a short helium gas cell. Our calculations (Fig. 1) reveal that the combination of phase matching and two-color waveform control generates significantly brighter high harmonics (black curve) in the 0.8 to 1.25 keV range compared to those produced using only a single 4-μm driving laser (red curve). Furthermore, temporal intensity calculations (inset figure) indicate that IAPs are generated exclusively with the two-color laser configuration.

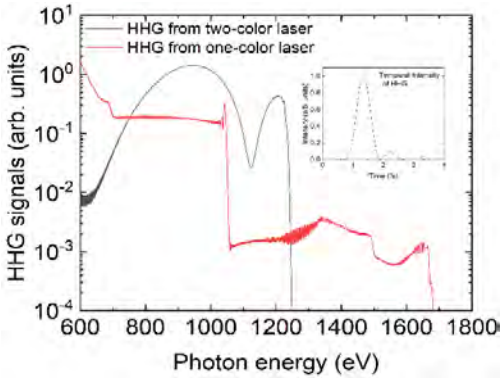


Figure 1. Far-field high-harmonic spectra from a He gas cell for waveform-controlled two-color laser (black curve) and one-color laser (red curve). The inset shows the temporal intensity of an IAP from the two-color laser.

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Spatial features of high-order harmonic generation from liquids

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Synopsis We measured the far-field spatial features of high-order harmonic generation (HHG) from liquids. The experimental results show that the spatial features depend sensitively on the laser intensity and focusing position. In particular, the spatial features exhibit a significant dependence on the focusing position under high laser intensity. This research may contribute to our understanding of fundamental physical mechanisms underlying HHG from liquids.

High-order harmonic generation (HHG) is a crucial approach for exploring attosecond science, generating extreme ultraviolet light, and facilitating real–time microscopic studies of electrons, which has drawn considerable attention in strong-field physics research. Over the past two decades, research on HHG from gases and solids has made substantial progress. In contrast, the study of HHG from liquids is relatively limited. In recent years, with the development of new technology for optically flat and thin sheets of liquid, extreme–ultraviolet HHG has been generated in the liquid phase[1]. Liquids have gradually emerged as a novel and promising medium for HHG, benefiting from the implementation of the liquid flat–microjet technology. This advance has extended HHG research to various liquids, as well as the exploration of electronic structure and ultrafast scattering processes[2].

In this presentation, we measured the far-field spatial features of HHG from liquids under different parameters of the driving laser. We observed that the far-field spatial features of HHG from liquids are sensitive to the focusing position and laser intensity. We employed a driving laser of 800 nm and ethanol as the target. By changing the focusing position or laser intensity, the variations in spatial features of 7th-order harmonic were measured, which are shown in Fig.1. Under the same laser intensity, the spatial features (e.g., the divergence) of HHG from liquids depend sensitively on the focusing position, especially under high laser intensity. These results may offer new possibilities for understanding the fundamental physical mechanisms underlying HHG from liquids.

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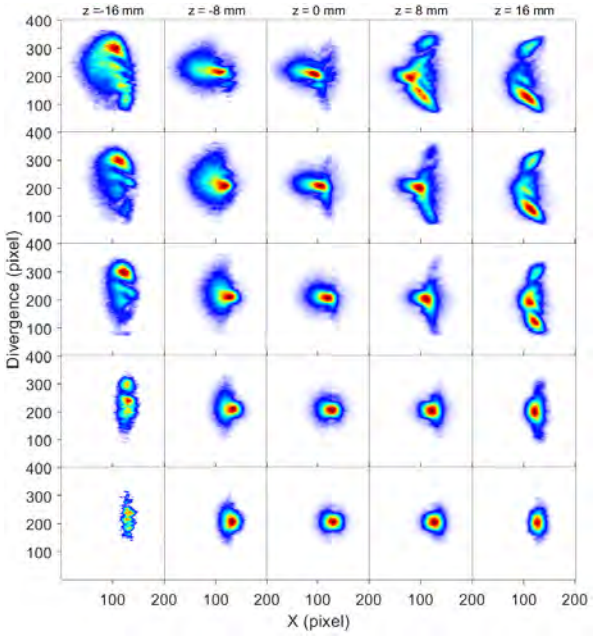


Figure 1. Spatial feature of 7th-order HHG from ethanol were measured under different focusing position and laser intensity. Each column corresponds to the focusing position z=-16, -8, 0, 8, 16 mm, negative values indicate that the liquid film is ahead of the focusing position while positive values indicate the opposite. Each row corresponds to the intensity 48, 35, 22, 10, 6 TW/cm^2 from top to bottom.

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Driving the high harmonic process using a multi-pass cell

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Synopsis The time-resolved investigation of Auger-Meitner decay in atoms needs XUV photons in the 100 eV range. By using a temporal compression scheme based on a gas-filled multi-pass cell, the necessary peak intensities for high-order harmonic generation are reached. Using neon as generating medium extends the cutoff to the desired range. The resulting attosecond pulse trains will be used in the attosecond beamline in Freiburg for the investigation of electronic dynamics in atoms driven by correlation effects.

Extreme ultraviolet (XUV) pulses in the soft X-ray and specifically in the range of 60-100 eV enable among others the investigation of electronic-correlation-driven processes such as the Auger-Meitner decay in krypton [1]. This process can be resolved in time, by using electron-ion coincidence (Reaction microscope or COLTRIMS [2]) in combination with attosecond pulse trains (APT) obtained by high-order harmonic generation (HHG).

Conducting such experiments poses two challenges. The first challenge is HHG at high repetition rates since measuring in the coincidence regime requires long acquisition times. Yb-based laser systems offer a sufficiently high repetition rate while also being commercially available and offering high average power.

To obtain the necessary peak intensity for HHG, the pulses from the Yb-based system have to be temporally compressed. For this purpose, a post-compression scheme based on a gas-filled multi-pass cell (MPC) [3] was developed. MPCs offer a high power efficiency and a low pointing sensitivity which is needed for long term stability. The temporal duration of the laser pulse measured with a frequency-resolved optical gating (FROG) setup before (a) and after (b) the post-compression is shown in figure 1.

The second challenge is to extend the cutoff of the HHG to the required photon energies while also maintaining a sufficient photon flux for photoionisation experiments. With the temporally compressed pulses the peak intensity is high enough to extend the cutoff. Using neon as generation gas, the cutoff is extended to above 100 eV, as shown in figure 1 (c).

The generated APT will be used for the time-resolved investigation of electron dynamics using a RABBIT scheme implementing an ultrastable delay line [4].

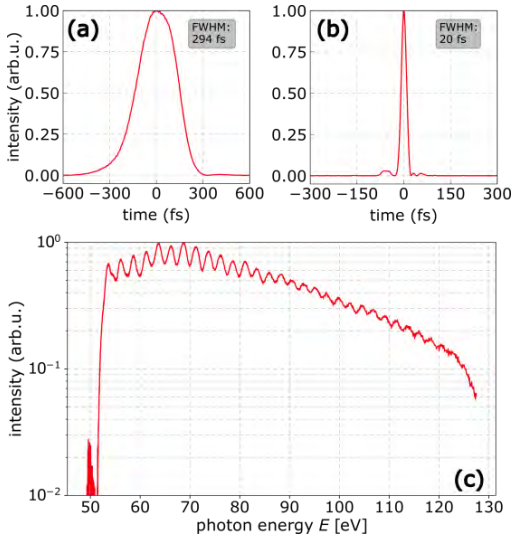


Figure 1. The pulse duration of the Yb-based laser before (a) and after (b) the temporal pulse compression. (c) XUV spectrum generated in neon (c).

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Short attosecond pulse trains for the time-slit experiments

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Synopsis Traditionally, high-order harmonic generation is used in two configurations to produce isolated attosecond pulses or attosecond pulse trains. In this work, we will report on an intermediate scenario where we generate very short attosecond pulse trains containing a few attosecond pulses (two or three) within the pulse train. We will show the possibility of controlling the relative amplitudes and the number of attosecond pulses using the carrier-envelope phase of the few femtosecond infrared driving pulses and discuss the possibility of performing Young's double slit experiment in the time domain.

High Harmonic generation (HHG) in atomic gas targets has been a reliable source of attosecond pulses with central frequencies routinely extending into the extreme ultraviolet (XUV) spectral region [1,2]. Depending on the duration of the driving laser pulses used for the HHG process, attosecond pulse trains (APTs) or isolated attosecond pulses (SAPs) are generated. When a driving laser with long pulses is implemented for the HHG process, the emission of an attosecond burst every half-cycle of the driving laser results in attosecond pulse trains [3,4]. However, if the emission of the attosecond burst could be confined to a single half-cycle, single attosecond pulses are generated. SAPs are routinely achieved by using single-cycle pulses or by implementing gating schemes [4].

In our setup, we generate APTs using a home-built OPCPA producing carrier-envelope phase (CEP) stable 6 fs pulses with a pulse energy of 10-15 uJ at a 200 kHz repetition rate. Due to the short duration of our driving pulses, only a few cycles contribute to generating the attosecond bursts [5]. Consequently, we have a few attosecond pulses (two or three) within the pulse train [6]. We use reconstruction of attosecond beating by a two-photon transitions (RABBIT) protocol for characterizing the generated APTs, and a typical spectrogram acquired using a 3D momentum spectrometer (ReMi or COLTRIMS) is shown in Fig.1 [7]. The spectrogram trace contains peculiar features corresponding to streaking and RABBIT in high and low photoelectron energy regions, respectively. We attribute these features to the differing number of attosecond pulses within the pulse train. By controlling the CEP of the driving pulses, we showcase the ability to control the relative amplitudes of the attosecond pulses and also the number of pulses within the pulse train. The changes made are reflected in the RABBIT spectrograms acquired for different CEP values. Furthermore, this possibility allows us to perform the Young's double slit experiment in time-domain and motivate investigations of the time-slit expeiments with varying delay between the attosecond pulses.

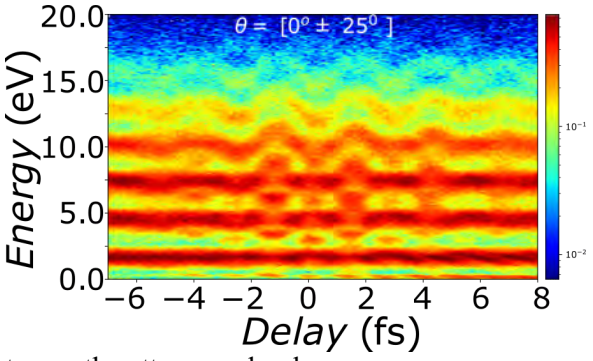


Figure 1. RABBIT spectrogram acquired with the 3D momentum sspectrometer with the short attosecond pulse trains, the signal is integrated over a cone of 25 degrees.

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Dispersion-scan (re)meets neural networks

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Synopsis
In this work, we implement neural networks (NN), capable of retrieving the spectrum and phase of an ultrashort pulse from a single-shot dispersion-scan trace, without requiring a separate measurement of the fundamental spectrum. We also explore NNs that can be trained in a short time duration, as low as 30 minutes

Novel laser technologies provide high peak and average power and high repetition rate, with applications ranging from high-harmonic generation to terahertz generation [1], many of which call for single-shot pulse characterization capabilities. Single-shot measurement of ultrashort pulses is additionally appealing, as it delivers shot-to-shot information of laser dynamics and enables the correlation of measurements to specific laser pulses in experiments. Neural networks (NNs), for retrieving the pulse characteristics from a d-scan or FROG trace, are known to be fast (on the order of milliseconds), and make promising candidates to be used in pulse characterization at high repetition rates.

Here, we present a NN based on an implementation of DenseNet which has earlier been used for d-scan retrieval, but only for the spectral phase and with the requirement for a separate measurement of the fundamental spectrum [2]. In situations where it is impossible or inconvenient to obtain a measured spectrum, it is necessary to develop NNs that can leverage the data redundancy present in d-scan traces to retrieve simultaneously both the spectrum and the spectral phase. As the electric field of a pulse is commonly represented in complex form (amplitude and phase), a NN for retrieval should give complex-valued output as well, ideally with all internal weights and biases in complex form. A direct complex-valued NN approach faces convergence issues in training due the high variety of spectra and phases. Instead, taking into account that the electric field of an ultrashort pulse can be sparsely represented in the time domain, e.g. a central peak followed with a few pre- and post pulses, a real-valued NN was used, but with an Inverse Fast Fourier transform (IFFT) before the linear layers of DenseNet. This makes the training data more predictable for the neural network, and hence, enables the retrieval of both desired quantities.

While working well for simulated data, the inference time rose from 20 to 130 ms for experimental data. To reduce the inference time, when retrieving measured data, we developed a simpler real-valued DenseNet that outputs the real and imaginary parts of the electric field. The inference time on measured data was brought down to ≈20 ms, which seems to suggest a correlation between network complexity and inference time which renders NNs into a bespoke solution that depends on the laser source to be measured. A comparison with conventional iterative methods will be shown.

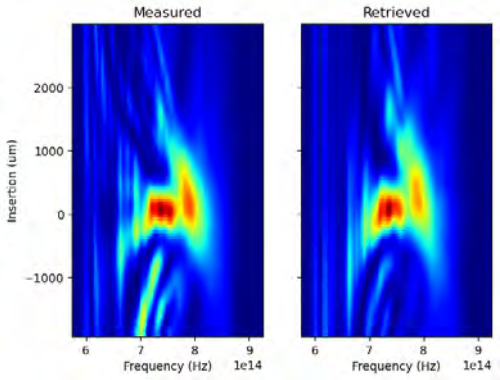


Figure 1. Left) Measured trace. Right) Retrieved trace using NNs.

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Angular Asymmetry of Double Ionization of Helium by Ultrashort Extreme Ultraviolet Laser Pulses

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Synopsis We investigated the double ionization of helium atoms in the presence of ultrashort Extreme Ultraviolet (XUV) laser pulses by solving the full dimensional time-dependent Schrödinger equation with applying the finite-element discrete variable representation method. The analysis on the joint angle distributions and their asymmetry parameters of the two photoelectrons show that the asymmetry parameter effectively indicates the strength of electron correlation in the double ionization process.

In recent years, X-ray free-electron lasers (FELs) have shown great potential in atomic and molecular research due to their ultrahigh brightness, ultrashort pulse duration. Using ultrashort and intense XUV pulses, researchers have extensively studied the few-photon double ionization (DI) dynamics of atoms and molecules theoretically [1–3]. Further investigation of this system will deepen our understanding of electron correlation effects in atomic, molecular and condensed physics.

We numerically solve the full dimensional time-dependent Schrödinger equation to simulate the two-photon non-sequential (photon energy $\hbar\omega = 45$ eV) and sequential ($\hbar\omega = 60$ eV) double ionization processes of a helium atom induced by XUV laser pulses. The joint angular distributions of the photoelectrons are been analyzed with their asymmetry parameters, as shown in the Fig. 1. The energy sharing integrated joint angular distributions of two-photon double ionization for (a) $\hbar\omega_{XUV} = 45$ eV or (b) $\hbar\omega_{XUV} = 60$ eV, which corresponding to non-sequential and sequential DI, are similar in their structure, both are back-to-back emission dominant. However, they are different in the symmetrical emission ($\theta_1 + \theta_2 = 2\pi$). The asymmetry parameter is defined to describe such difference.

In the bottom row of Fig. 1, the asymmetry parameters are presented for both (c) non-sequential and (d) sequential two-photon DI. The obvious difference in asymmetry parameter for non-sequential and sequential DI reveals an effective indicator of the strength of electron correlation, quantifying its role in double ionization. Our further investigations reveal such indicator can also be applied to the DI processes with more photon involved, such as the three-photon DI.

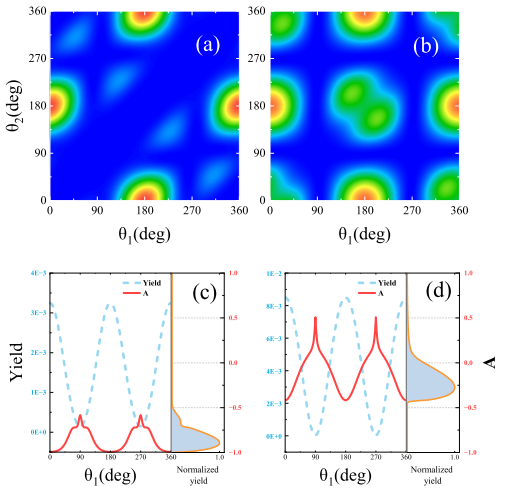


Figure 1. Calculated joint angular distributions (top row) and the distributions of asymmetry parameter (bottom row) for the two-photon DI of He in the presence of XUV pulses with (a,c) $\hbar\omega_{XUV} = 45$ eV or (b,d) $\hbar\omega_{XUV} = 60$ eV. The XUV pulse has a peak intensity of 5×10^{14} W/cm^2 and pulse duration of 1 fs.

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Cumulative effect on extreme ultraviolet high harmonics generated by high repetition rate Yb-based amplifier

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Synopsis We developed a high-harmonic generation system using a high-repetition-rate Yb-based laser amplifier and systematically investigated cumulative effects on the gas target. Our results highlight the critical role of these effects in optimizing harmonic generation efficiency at high repetition rates.

Bright high-harmonic sources with MHz-level repetition rate are in high demand for applications such as photoelectron spectroscopy, coherent diffraction imaging, and electron-ion coincidence detection. Recent advancements in Yb-based laser technology and optical parametric chirped pulse amplifiers provide high-repetition-rate, high-average-power drivers for high-harmonic generation (HHG) at hundreds of kilohertz to megahertz [1]. Although cumulative effects on HHG when increasing the repetition rate has empirically been known [2], the quantitative studies of their direct impact on HHG has been overlooked.

In this study, we systematically investigated HHG dependence on repetition rate by using nonlinear post-compressed Yb laser amplifier with repetition rates ranging from 5 kHz to 100 kHz. The Yb-based laser amplifier, delivering 500 μJ pulses with 500-fs duration, was compressed to 50 fs using a multi-pass cell and focused into an argon gas cell in a vacuum chamber with focal lengths of 175 mm and 250 mm. The estimated focused intensities were 1.35×10^{14} and 6.50×10^{13} W/cm², respectively. High harmonics were delivered into grating spectrometer equipped with micro channel plate (MCP) and phosphor screen.

Our results clearly show that HHG efficiency decreases with increasing repetition rate. Figure 1(a) shows a typical high-harmonic spectrum at a 20 kHz repetition rate and a backing pressure of 9.0×10^{3} Pa. Figure 1(b) plots the normalized intensity per pulse of the 33rd-order harmonic as a function of repetition rate (5–100 kHz). This result shows a monotonic decrease for both focal lengths. Notably, the normalized intensity drops by a factor of 0.2 at 100 kHz compared with 5 kHz. This decrease suggests a cumulative effect on gas target at the focal position. The most probable cause is local gas density reduction due to the heating of gas from the focal region, with a relaxation timescale on the millisecond scale [3]. The estimated impact of gas depletion from a previous study [4] agrees well with our observations. These findings provide crucial insights for optimizing HHG sources at high repetition rates enabled by emerging Yb-based laser technologies.

This work was partially supported by JSPS KAKENHI 20H05670.

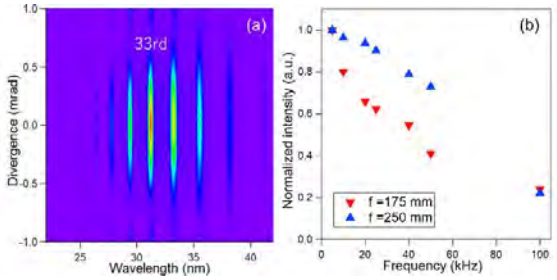


Figure 1. (a) Harmonic spectrum recorded on phosphor screen. (b) Dependence of normalized intensity of the 33rd harmonics per pulse as a function of repetition rate.

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Highly Efficient Enantiosensitive Low-Order Harmonic Generation*

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Synopsis We demonstrate control over highly enantiosensitive low-order harmonic generation in gas-phase chiral molecules driven by synthetic chiral light with intense fundamental and second harmonic components. Our approach enables ultrafast chiral discrimination in the ultraviolet to vacuum ultraviolet range. We achieve chiral dichroism up to two orders of magnitude higher than conventional methods relying on circularly polarized light, opening new paths for non-invasive ultrafast chiral imaging and spectroscopy.

Chirality is a fundamental property of nature, playing a crucial role in molecular biology, chemistry, and pharmacology. The ability to distinguish and manipulate left- and right-handed enantiomers is essential for applications ranging from disease diagnostics to drug development. Traditional methods for chiral discrimination, such as circular dichroism spectroscopy and vibrational circular dichroism, rely on weak interactions beyond the electric-dipole approximation, limiting their sensitivity and efficiency. Synthetic chiral light, which is locally chiral within the dipole approximation, has emerged as a powerful alternative [1].

Here we investigate ultrafast interactions of synthetic chiral light with randomly oriented chiral molecules to demonstrate generation of highly enantiosensitive low-order harmonics ranging from $682 - 170$nm. In contrast to [1], we use a large opening angle of 45° between the two laser beams of which one carries a second-harmonic component. The resulting chiral electric field maintains elliptically polarized in-plane and carries an intense out-of-plane 2ω component to enable strong enantiosensitive interferences. Unlike previous approaches relying on high harmonic generation (HHG) [1, 2, 3], our method exploits bound-state dynamics at high laser intensities, enabling simultaneous excitation of multiple achiral and chiral multiphoton channels. This results in novel interference effects amplifying enantiosensitivity, which can be controlled by tuning the phase delay between the light field's two colors. Compared to conventional methods relying on circularly polarized light, our results show that locally chiral light achieves up to two orders of magnitude higher chiral dichroism, even in non-resonant 2nd and 3rd harmonics. Hence, chiral discrimination can be realized using low-order harmonics, paving the way for new, non-invasive ultrafast imaging and spectroscopic techniques.

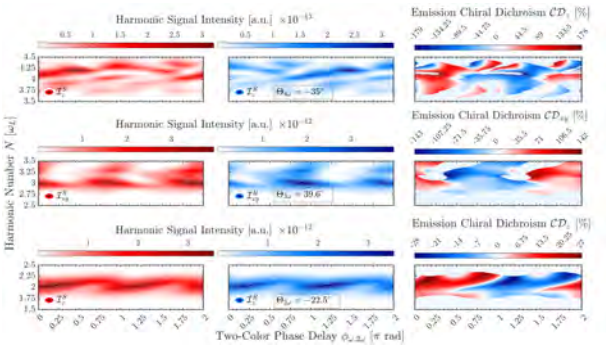


Figure 1. Far-field harmonic signal intensity of (S)- and (R)-Methyloxirane (left, middle) and chiral dichroism (right) in harmonics $N = 2, 3, 4$ (row 1,2,3) as a function of the two-color phase delay.

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Time-Resolved X-ray Absorption Spectroscopy of Strong-Field Ionized Water

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Synopsis We apply X-ray Absorption Spectroscopy (XAS) to study dissociation pathways induced by strong-field ionization of isolated water molecules. Combining XAS with spectral-domain ghost imaging, we observe electronic dynamics within the water cation with sub 30-fs resolution. This measurement benchmarks the time-resolution capabilities of instruments at the high repetition rate XFEL facility, LCLS-II.

Dissociation pathways of isolated water molecules following strong-field ionization have been temporally resolved through observation of momenta of dissociated ion fragments following strong-field ionization (e.g. [1]). These experiments reveal time-resolved dynamics in nuclear structure for the multiple dissociation pathways for water molecules following ionization. Observation of the electronic structure using X-ray Absorption Spectroscopy (XAS) performed using next-generation x-ray free electron laser (XFEL) light sources offers more direct insight into the electronic dynamics that drive these nuclear motions.

We apply XAS to study the dissociation pathways induced by strong-field ionization of isolated water molecules. We employ an 800 nm pump pulse (intensity ~10^{14} W/cm^2) to drive strong-field dynamics, followed by a tunable x-ray pulse to probe the core to valence excitation in water molecules ionized by the laser pulse. We then apply spectral-domain ghost imaging to the resultant electron spectrum to obtain precise insight into the electronic structure of the water molecule [See figure 1]. We observe a 'turn on' in absorption in both the water cation and dication following laser ionization with sub-30 fs resolution.

This measurement benchmarks the time-resolution capabilities of instruments at the high repetition rate XFEL facility, LCLS-II.

This work was supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences (BES), Chemical Sciences, Geosciences, and Biosciences Division (CSGB).

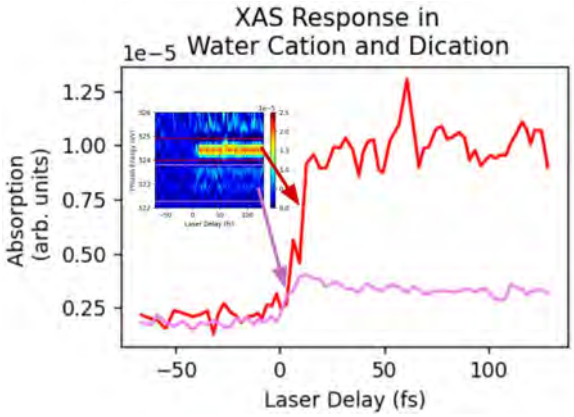


Figure 1: Water cation and dication electron yield after onset of ionization. Yield of features corresponding to cation and dication production in 2-D maps generated through spectral-domain ghost imaging (inset). Positive delays imply laser pulses arrived before x-ray pulses. Onset of electron dynamics in both cation (red) and dication (purple) appear as sharp increases in yield just after 0 fs delay.

Use of the Linac Coherent Light Source (LCLS), SLAC National Accelerator Laboratory, is supported by the US DOE, Office of Science, BES, under Contract DE-AC02-76SF00515.

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Synthesis of Tesla-Scale Attosecond Magnetic Pulses with Ring-Current Gating

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Synopsis We theoretically propose a method to synthesize the first attosecond magnetic pulses with tunable waveform. Our *ab initio* calculations predict a magnetic pulse with a duration of 787 as and a high flux density of ~1T at a few hundred nanometers from the source, paving the way for attosecond control and measurement of magnetization and chiral dynamics.

The conventional generation of strong magnetic fields in conducting or superconducting materials arranged in coils or solenoids is effective for studying long-term magnetic responses. However, these approaches do not allow for the exploration of attosecond magnetization dynamics[1, 2]. In recent years, femtosecond laser pulses have been employed to induce ring currents[3] associated with strong magnetic fields[4-7]. Still, high-resolution control over the temporal and spectral content of the magnetic field was not previously addressed. We also aim to reach the attosecond domain required for probing predicted attosecond magnetism[1].

In this work, we introduce current gating schemes as a method to generate a strong magnetic field confined to attosecond duration [8]. By considering trains of nonresonant femtosecond pulses with counter-rotating circular polarization, we switch on and off the generation of ring currents in Ne atoms. As shown in Fig. 1, the time delay between the gating pulses can be tuned to control the duration of a unipolar magnetic field or the spectral content of the magnetic pulse (see panels (a) and (b) with two and three gating pulses respectively). This procedure enables a temporal shaping of the magnetic field and brings options to isolate the generated magnetic field from the driving electromagnetic field, which is highly desirable.

Our *ab initio* calculations use the TDDFT and Maxwell frameworks of the Octopus software. By combining this with a macroscopic model of elementary ring currents, we predict magnetic fields with a high flux density of ~1T at a few hundred nanometers from the source with a pulse duration of 787 as. We foresee that tunable attosecond magnetic fields will be an additional tool for advancing attosecond spectroscopy within magnetic and chiral-systems, opening unprecedented pathways for understanding and manipulating quantum matter.

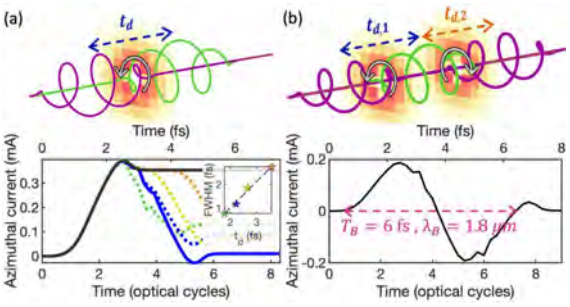


Figure 1. (a) Two time-delayed counter-rotating circularly-polarized laser pulses temporally confine the ring current and the generated magnetic field. The inset shows the linear relation of the full-width at half-maximum (FWHM) intensity of the magnetic field with the time delay. By adding a third gating pulse in (b), we show the synthesis of a single-cycle magnetic pulse, whose wavelength λ_B can be tuned with the driving time delay.

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Probing Many-Body Effects in Strongly Correlated Materials via Reflectivity Using Infrared Laser Fields

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Synopsis
We simulate the interaction of strongly correlated materials with strong few-cycle infrared pulses and determine how the pulse drives changes in material reflectivity on a sub-cycle timescale. We use a 2D square lattice Hubbard model as a prototypical strongly correlated system using non-equilibrium dynamical mean field theory to simulate the interaction with external laser fields and calculate the expected change in reflectivity through the induced current response.

We report on a theoretical investigation motivated by recent experimental results on how a strong infrared field can affect the reflectivity of strongly-correlated materials on sub-cycle timescales. We do this by simulating the interaction of a two-dimensional square Hubbard lattice modelled with a strong few-cycle infrared pulse using non-equilibrium dynamical mean field theory (NE-DMFT) [1, 2, 3].

We use a 2D square Hubbard model serves as a prototypical representation of strongly correlated materials. The NE-DMFT formalism is employed to capture local correlation effects in a non-perturbative manner, making it ideally suited for exploring the transient dynamics driven by intense infrared fields. The system is excited by a strong few-cycle laser pulse that drives it far from equilibrium, leading to a rapid reorganisation of the electronic states. This reorganisation is directly reflected in the time-dependent dielectric function and, hence, in the optical reflectivity of the material.

We simulate the interaction of our system with a strong pump pulse and calculate the induced change in reflectivity through the current induced by a weaker probe field, from which the optical conductivity and reflectivity can be obtained [4]. Analysis of the reflectivity reveals that the strong field induces a transient restructuring of the density of states reflecting a clear Mott metal-insulator phase transition [3]. Most notably, we observe clear field-driven changes in the reflectivity on sub-cycle timescales—on the order of hundreds of attoseconds, resulting from many-body dynamics of the system.

The emergence of attosecond-scale reflectivity changes indicates that the electronic system responds almost instantaneously to the strong field, with the electron correlations dynamically modulating the optical properties within each cycle of the laser pulse. This sub-cycle timescale suggests that strong-field excitation can be used to control and probe the ultrafast optical properties of strongly correlated materials, offering a route to manipulate light-matter interactions on previously inaccessible time scales.

Our work provides new insights into the transient optical behavior of strongly correlated systems and highlights the critical role of many-body effects in shaping ultrafast reflectivity. These results pave the way for future experimental investigations using time-resolved reflectivity measurements, where attosecond resolution may uncover rich dynamics in correlated materials under extreme conditions.

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Orbital Angular Momentum Control in Attosecond Pulses of Light

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Synopsis We theoretically and experimentally demonstrate the generation of attosecond vortex pulse trains, i.e., a succession of attosecond pulses with a helical wavefront, resulting from the coherent superposition of a comb of extreme-ultraviolet high-order harmonics carrying the same orbital angular momentum (OAM). The control of spin and OAM degrees of freedom in attosecond pulses brings new capabilities in the spectroscopy and imaging applications to chiral and magnetic systems.

High-order harmonic generation (HHG) has been paramount for developing extreme ultraviolet (EUV) structured beams[1-4]. In previous schemes, the up-conversion of orbital angular momentum (OAM) follows a linear scaling with the harmonic order and the OAM of the driver[5,6]. This selection rule has precluded the emission of vortex beams with attosecond pulse duration. Indeed, the synthesis of several harmonic orders presenting a linear scaling of the OAM results in an attosecond light spring whose pulse duration is similar to the femtosecond driving pulse[5].

In this work, we demonstrate theoretically and experimentally a novel setup of HHG that produces harmonic vortices with the same topological charge as the driver[7]. The noncollinear superposition of two circularly polarized vortex beams with opposite spin and OAM helicity creates a forked polarization tilt-angle grating at the generation plane. Then, the simultaneous conservation of linear momentum, spin, and OAM yields two spatially separated high-harmonic beams (Fig. 1a-c) with order-independent OAM (Fig. 1d). According to our simulations, the synthesis of a comb of high-order harmonics with the same OAM results in attosecond vortex pulse trains, wherein the electric field describes spatiotemporal helicoids evolving over hundreds of attoseconds (Fig. 1e-g). Further characterization techniques are required for measuring the helical wavefront of attosecond vortex pulses. The control of the spin and OAM of light at the attosecond timescale provides an additional degree of freedom for attosecond spectroscopy in chiral and magnetic systems.

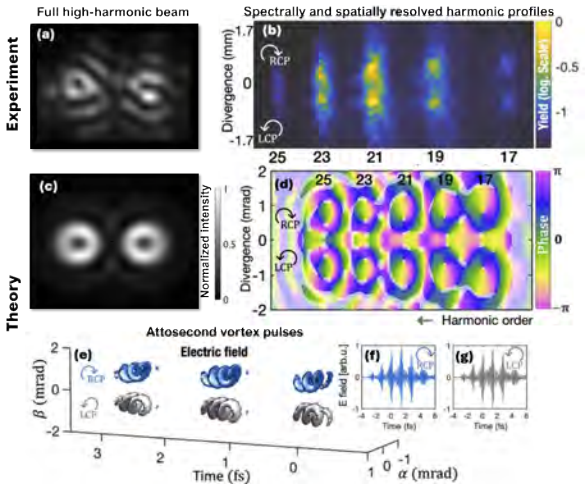


Figure 1. a), c) Far-field full-beam intensity profile (comprising 17th–25th harmonics) in experiment and theory. b) Spatially and spectrally dispersed experimental HHG, showing the spatial overlap of 17th–25th harmonics. d) Computed phase profiles in HHG demonstrating order-independent OAM. e) Far-field electric field obtained from simulations, yielding two separated attosecond vortex pulse trains with opposite handedness in spin and OAM. f), g) Local electric field of the RCP and LCP attosecond pulse trains at the maximum intensity point.

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Combining tunable few cycle UV and soft x-ray pulses at the European XFEL

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Synopsis We present ongoing developments to produce high energy burst-mode few cycle UV pulses using resonant dispersive wave (RDW) emission in a gas-filled hollow core fiber at 1.1 MHz intra-burst repetition rate and combine them with attosecond duration soft x-ray pulses at the SQS instrument at the European XFEL.

Many chemical systems have photoexcitations in the UV spectral range involving valence shell electronic and nuclear dynamics [1]. To investigate fast electron dynamics, pulses with sufficient energy and durations on the shortest time scale are necessary to both excite and probe at high repetition rate. Soft x-rays are an excellent probe and by resonantly exciting and ionizing core electrons give site-specific information in molecular targets. At the European XFEL, subfemtosecond pulse durations from the Free Electron Laser (FEL) have already been demonstrated [2,3] on the terawatt scale and at megahertz intra-burst repetition rates. However, the corresponding generation of sufficiently short laser-based pulses in the UV for excitation remains a challenge. One encouraging method is resonant dispersive wave (RDW) emission produced during soliton fission in gas-filled hollow core fibres, which has been used successfully to efficiently produce tunable pulses down to deep UV wavelengths [4,5]. This promising approach can produce UV spectra with a Fourier transform limit (FTL) on the order of 1 fs and was demonstrated to be feasible at high repetition rates with small core diameter fibres [6], a regime where recombination heating can affect the process [7].

We have implemented the technique in larger diameter capillary fibres using the European XFEL in-house laser system [8] in burst mode at an intra-burst repetition rate of up to 1.1MHz. This system produces 1030nm pulses with 850fs duration in 10Hz bursts, an energy of 3mJ at 1.1 MHz intra-burst rep. rate and can be synchronized to the FEL. Multiple compression stages are used to decrease the pulse duration, ultimately reaching 10fs duration with up to 0.5mJ energy. These pulses are coupled into a 2m long, 250um diameter hollow core fiber, filled with 415 mbar of Argon gas in a negative gradient configuration. We show that while the RDW energy can degrade at high repetition rates, conditions exist in which it is stable over our burst. The measured RDW spectrum has a ca. 5fs FTL and an energy in the few microjoule range.

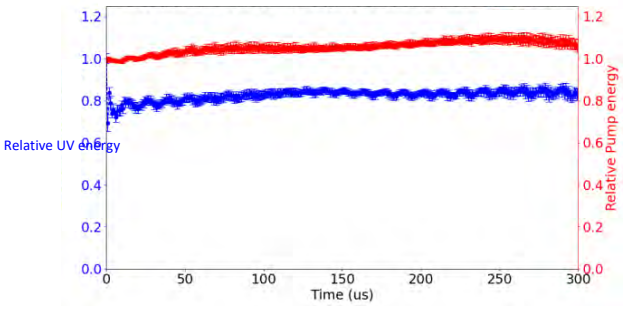


Figure 1. Pump energy into the RDW fibre (red) and of the 330nm RDW energy (blue) for a 300us 1.1MHz burst.

We discuss integration of the source to the SQS instrument as well as the prospects and challenges for reaching even shorter pulse durations in the future.

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In-situ Waveform Sampling in a Reaction Microscope

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Synopsis We introduce a novel *in-situ* method that utilizes momentum microscopy via single-atom ionization to capture the electric field of a femtosecond laser pulse.

Studying ultrafast, strong field-driven phenomena in small quantum systems such as atoms, molecules and clusters, is at the forefront of attosecond science [1]. Photoionization with ultrashort laser pulses provides direct access to the time scale of the electron dynamics, bond formation, and dissociative processes. Reaction Microscopy or Cold Target Recoil-Ion Momentum Spectroscopy (COLTRIMS) [2] is a powerful tool for unraveling ultrafast atomic and molecular phenomena by measuring the 3-dimensional photoelectron and photoion momentum distributions in coincidence. Since many field-driven processes evolve on a sub-cycle time scale, the complete characterization of the ionizing pulse is a critical aspect of many ultrafast photoionization experiments.

Near infrared femtosecond laser technology has been the main driver of ultrafast photoionization science for the past two decades. However, most currently available near-infrared pulse characterization techniques that rely on nonlinear optical effects (e.g., FROG, SPIDER, D-scan [3-5]) are primarily sensitive to the intensity envelope and do not provide information about the sub-cycle temporal evolution of the pulse. Moreover, they rely on sophisticated reconstruction algorithms. In contrast, techniques that are based on strong-field ionization (e.g., [6–8]) can provide direct access to the electric field waveform, although they often require complex measurement setups—such as dedicated vacuum systems—or the use of a few-cycle visible sampling pulse. All of the above techniques have in common, that they are measured outside the COLTRIMS, rather than in the interaction region, and thereby are *ex-situ* techniques.

Recently, Tunneling Ionization with a Perturbation for the Time-domain Observation of an Electric Field (TIPTOE) has been utilized to measure arbitrary light waveforms via sub-cycle tunneling ionization in gases [9], or tunnel excitation in solids [10]. Here, we extend this technique to the single-atom ionization regime, thereby enabling *in-situ* waveform measurements in a reaction microscope.

In our setup an intense, near-infrared femtosecond laser pulse is split into an intense, ionizing pulse and a weak, but otherwise identical, sampling pulse. The delay-dependent modulation of the ionization yield directly maps to the electric field of the sampling pulse. In addition, the photoelectron momentum distribution also provides direct access to the vector potential of the laser pulse.

In summary we present a novel *in-situ* technique that combines COLTRIMS and TIPTOE to sample electric field waveforms via momentum microscopy using single-atom ionization. This approach offers a new level of control for studies of strong-field dynamics using photoionization coincidence momentum spectroscopy.

We acknowledge support by the Joint Centre for Extreme Photonics. SGM and PBC acknowledge support by Army Research Office.

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Attohallen – A New Attosecond Science Facility in Sweden

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Synopsis We present the new high-power laser facility Attohallen in Gothenburg, focusing on its first high-order harmonic generation beamline. Key features include the femtosecond drive laser, XUV generation up to ~120 eV with diagnostics, a homebuilt Mach-Zehnder grazing incidence interferometer, and a versatile end-station for high-resolution attosecond timing experiments. First commissioning results demonstrate the beamline's capabilities.

We present the new high-power laser facility *Attohallen* located in Gothenburg, with focus on its first high-harmonic generation (HHG) beamline of *Attohallen*, which is developed for photon-demanding, attosecond time-resolved experiments in the energy range up to ~120 eV. This beamline employs loose focusing conditions as outlined in Refs. [1] and [2], enabling efficient XUV generation at 100 Hz repetition rate.

Key aspects of the facility include the design and operation of its femtosecond OPCPA drive laser system, the HHG source with diagnostics, a home-built Mach-Zehnder grazing incidence interferometer, and a versatile magnetic bottle electron spectrometry end-station of unique spectral resolving power. The beam line and spectrometry system is tailored for low-density targets such as neutral and ionized atoms and molecules, offering high spectral and temporal resolution.

Fig. 1 provides a first example of the comissioning results obtained, reflecting the generation of photoelectron sidebands in Xenon as the ionization target species using XUV + IR provided by our grazing incidence interferometer. Photoelectron peaks corresponding to the harmonic energies (H11-H23) are observed for all delays, whereas sidebands (S12-S22) are generated only when the two pulses overlap not only spatially, but also in time.

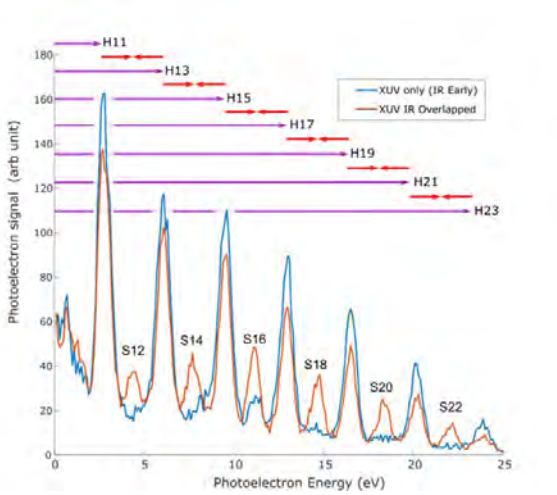


Figure 1. Generation of photoelectron sidebands in Xenon as the ionization target species using XUV + IR provided by our grazing incidence interferometer.

With its state-of-the-art capabilities, *Attohallen* is positioned to serve as a powerful facility for exploring quantum dynamics and aims to become a valuable resource for the broader ultrafast science community.

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Rydberg states generation driven by quantum light

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Synopsis We study the strong-field excitation driven by quantum lights. Developing a quantum-optical trajectory Monte Carlo method, we simulate the Rydberg states distribution with squeezed state light, which manifests as notably different interference structures from that with coherent-state (classical) light. We analyze the electron recapture dynamics and reveal that the photon statistics of squeezed-state light fields endows the tunneling electron wave packets with a time-varying phase uncertainty and modulates the photoelectron intracycle and intercycle interferences.

Rydberg atoms are important building blocks for quantum technologies, exploited to new applications in quantum computing, quantum communication and quantum sensing due to their unique tunable quantum properties. Besides the widely-used few-photon resonant excitation for the specific Rydberg state, multiple Rydberg states can be populated coherently and efficiently through the frustrated tunneling ionization (FTI) or the Coulomb potential recapture effect in strong laser field[1].

Nonclassical states of light are defined as states whose properties cannot be described by classical electromagnetism, and therefore require the framework of quantum optics. These states have become crucial for the development of photonic quantum technologies, providing versatile and resilient tools for quantum information science applications[2,3].

In this work, we study the strong-field excitation of hydrogen atoms with quantum lights. We develop a quantum-optical trajectory Monte Carlo (QOTMC) method, where the light is described by the generalized Glauber P representation. Employing QOTMC, we show novel structures of Rydberg states distribution generated by squeezed-state light. Based on the QRS theory, we reveal that the quantum fluctuation of light endows the recapture electrons with a time-varying phase uncertainty. The calculated time-dependence of recapture rate is negatively related to the laser envelope and the residual laser interaction time, which is termed as envelope effect. Combined with the carrier-wave effect, an analytic formula is proposed to calculate the Rydberg states population in strong quantum optical fields.

We illustrate the modulation behavior of Rydberg state population with laser intensity. By precisely tuning the CEP of laser pulses, the optimal laser parameters for the generation of specific Rydberg state are identified, which opens new avenues for the symbiosis of attosecond physics and quantum information.

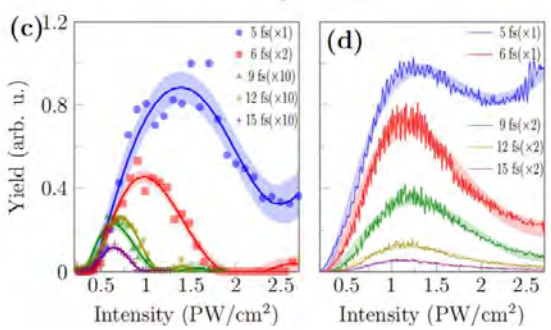


Figure 1. The QOTMC calculated results of Rydberg states population under the same laser parameters

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Probing coherent phonon in BaF_2 through time-resolved high harmonic generation spectroscopy

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Synopsis: High harmonic generation in solid has significant implications in fundamental science and practical applications. We demonstrate coherent phonons and their dynamics can be probed using time-resolved HHG spectroscopy. Band structure influences how electrons are driven and recombined, affecting the efficiency and spectrum of HHG. The excited phonons after the pump modify the band gap and show a 2D sensitivity of modulation in intensity and energy domains of HHG. Manipulation of band structure dynamics through coherent phonons can be performed by controlling pump pulse polarization and intensity.

High harmonic generation (HHG) is an extreme nonlinear optical process in which new electromagnetic radiations from the visible region to the soft-X-ray region are generated from the interaction of strong field laser pulses with maters. The field of high harmonic generation from solids has grown tremendously since the first observation of high-order harmonic from zinc oxide [1]. Numerous reports of HHG from various materials, as well as their potential applications in spectroscopy and industrial markets, have been made. Recently, high harmonic spectroscopy has shown the ability to probe lattice dynamics in alpha-quartz crystal [2]. Here we demonstrate that coherent phonons in BaF2 crystal can be excited and detected by time-resolved high harmonic generation (TR-HHG) spectroscopy. TR-HHG spectrum, affected by T2g phonon mode, illustrates a remarkable two-dimensional (2D) sensitivity on both intensity and frequency domains. The modulations in harmonic intensity and central frequency show a phase shift value of $\pi/2$. By employing a quantum model including electron-phonon interaction, we successfully complement our experimental observation. Our work laid solid groundwork for engineering the electronic structure through coherent phonons.

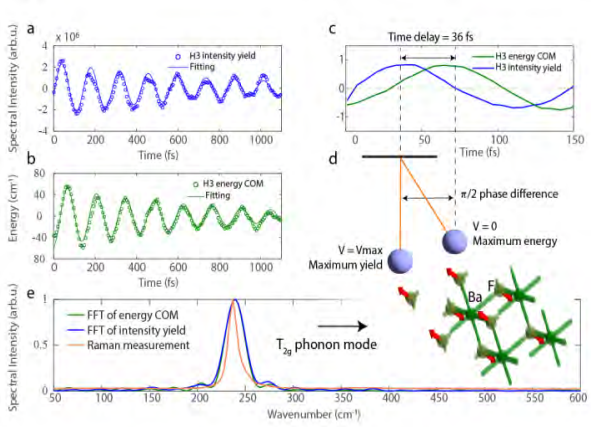


Figure 1. Coherent phonon oscillation dynamics probed with tr-HHG spectroscopy. [a] Time-resolved intensity yield of H3 (blue circles) and its fitting (blue curve). [b] Center of mass of H3 photon energy (dark green circles) and its fitting (dark green curve), subtracted by the average value. [c] Same as a and b but with a shorter time window. [d] The simple pendulum model indicates the initial phase difference, where the maximum and minimum velocities of the pendulum correspond to the maximum harmonic yield and maximum harmonic energy modulations, respectively. [e] Phonon spectra were measured using different techniques: FFT of the center COM (dark green curve), FFT of the intensity yield (blue curve), and Raman measurement (light orange curve). The intensities of all traces are normalized to their maximum.

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Observation of an evanescent field-induced harmonic generation in pre-ionized liquid water

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Synopsis We demonstrate an evanescent field-induced harmonic generation in pre-ionized water above the critical plasma density. Observed narrow spectral width of the harmonics clearly indicates that the high harmonics are generated by the penetrating evanescent field. The intense evanescent field has a potential application to study strong field physics beyond the electric dipole approximation due to its nonuniform electric field.

High harmonic generation (HHG) has opened a new frontier to study intense light-matter interactions on the unprecedented ultrafast time scale. In the past decade, almost all investigations of HHG have been extensively studied within the electric dipole approximation. Very recently, using a plasmonic device, it has been reported that a nonuniform near field induced-HHG exhibits dipole-forbidden emission due to the quadrupole contribution of the near field [1]. However, the applicable electric field strength is limited by optical field-induced damage.

In this work, we demonstrate an intense evanescent field-induced harmonic generation in pre-ionized water above the critical plasma density. Our approach enables us to generate a more intense nonuniform field with no intensity limit. We use an intense MIR source (3100 nm, 100 µJ, 160 fs) consisting of a KTA-based optical parametric amplifier (OPA) [2]. The MIR pulse is focused into a flat liquid water jet to generate harmonics of the MIR pulse. In addition, an NIR pulse (800 nm, 50 fs) is used as a pump pulse to generate many electrons in the liquid water by tunneling ionization. As a result, the pre-ionized liquid water above the critical plasma density of the MIR pulse is prepared. In such pre-ionized water, the MIR probe pulse cannot propagate, and the electric field becomes the rapidly decaying evanescent field.

Figure 1 shows the fifth harmonic of the MIR pulse as a function of the time delay between the NIR and MIR pulses. We found that the harmonic yield drops after the NIR pump. Moreover, the spectral broadening of the harmonics occurs before the NIR pump, but the spectral width remains narrower after the pump. By solving the nonlinear propagation equation, we show that the MIR field in pre-ionized liquid water decays rapidly due to the plasma shutter effect. In such a condition, the evanescent field exists only around the interface, resulting in an extremely short propagation length and decrease of the harmonic yields. In addition, the spectral broadening of the fundamental light is suppressed, because the real part of the refractive index becomes always zero, which means that the phase of the MIR probe pulse remains constant. These results are consistent with the observed behavior of harmonics. The intense evanescent field has a potential application to study strong field physics beyond the electric dipole approximation.

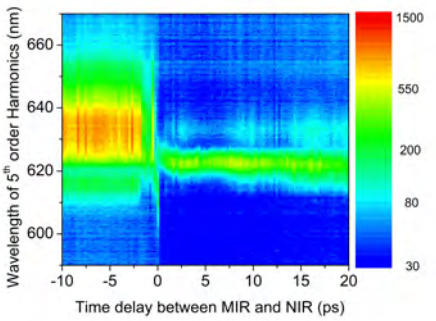


Figure 1. The fifth harmonic of the MIR pulse as a function of the time delay. The intensities for both pump and probe pulses are 2×10^{14} W/cm^2.

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Tracking Bandgap Dynamics in Solids with Extreme Ultraviolet High Harmonic Interferometry

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Synopsis We investigate excitation-induced bandgap dynamics in bulk solids using spectral interferometry of phase-locked extreme ultraviolet (XUV) pulse pairs. High-harmonic phase shifts of opposite sign are observed in amorphous SiO_2 and crystalline MgO, correlated with transient modifications of the electronic structure. Numerical simulations support the experimental findings, revealing potential for all-optical control and characterization of band structure changes on sub-cycle timescales.

Optical interferometry has been a foundational tool in ultrafast physics, providing insights into light-matter interactions across various domains. The advent of attosecond science and the pioneering demonstration of high-order harmonic generation (HHG) in solids[1] have further opened new avenues for probing ultrafast electronic dynamics and band structure modifications in condensed matter systems.

In this work, we employ spectral interferometry with phase-locked extreme ultraviolet (XUV) pulse pairs, generated through HHG in solid targets, to investigate excitation-induced bandgap dynamics in bulk materials. Specifically, we study amorphous SiO_2 and crystalline MgO, materials previously explored in HHG experiments. Unlike earlier studies focusing on intensity-dependent harmonic phase shifts[2], our approach examines excitation-induced phase variations between the two collinearly propagating XUV pulses.

Figure 1 illustrates the spectral interference patterns of harmonics 5, 7, and 9 obtained in SiO_2 at various intensities of the few-cycle infrared driving fields. The shift in the interference fringes reflects phase shifts of the high harmonics, which provide direct insight into the excitation-induced electronic dynamics within the material.

Our experimental observations reveal high-harmonic phase shifts of opposite sign in the two materials, which we attribute to transient modifications of their electronic structures. These findings are supported by numerical simulations based on the semiconductor Bloch equations, providing a consistent theoretical framework. This technique introduces a new pathway for all-optical control and characterization of band structure dynamics on sub-cycle timescales.

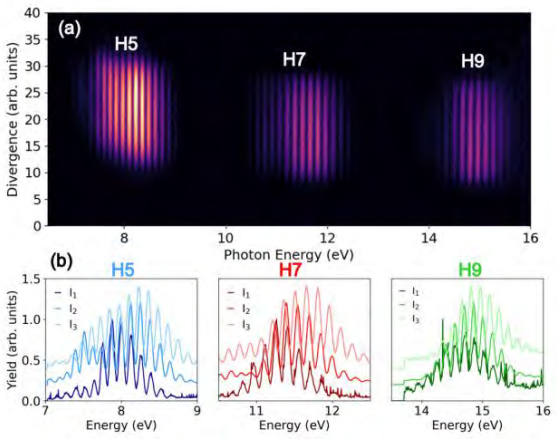


Figure 1. XUV interferometry in bulk SiO_2. (a) Interferometric pattern of harmonics 5,7 and 9 at an XUV-XUV delay of 30fs. (b) Spatially integrated harmonic signal of the individual odd harmonics obtained at different NIR peak intensities projected onto the energy axis. The spectra are offset vertically for visibility.

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Decoding The Ring Currents In Organic Molecules Using tr-ARPES

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Synopsis Using the NIR pump–XUV probe technique, we initiate and observe electron current dynamics in organic molecules. We capture these dynamics through time- and angle-resolved photoelectron spectroscopy (tr-ARPES). The calculated angle-resolved photoelectron distributions help us identify the direction of ring currents following photoexcitation, providing crucial insights into the molecule's reactivity.

Recent experimental and theoretical advancements in attosecond science have enabled us to revisit and verify the chemical properties of materials with unprecedented precision. In this study, we employ advanced quantum chemistry calculations to investigate ring currents in organic molecules[1, 2]. Utilizing the NIR pump–XUV probe technique, we initiate and observe these dynamics, generating time- and angle-resolved Photoelectron spectroscopy (tr-ARPES). The calculated angle resolved photoelectron distribution allow us to identify the direction of ring currents in the organic molecule after photoexcitation. This information is important to expand our understanding of the chemical nature of the materials and its reactive properties.

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Phase evolution of strong-field ionization

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Synopsis We investigate dipole phase evolution during strong-field ionization of xenon using attosecond transient absorption spectroscopy. Combining experiments with R-Matrix with Time-dependence calculations, we identify three key processes: ionization accumulation, transient excitation, and reversible population of excited states. The complex dipole response reveals interference effects between ionization and excitation, providing insight into attosecond-scale quantum dynamics and phase behavior under moderate NIR intensities.

Strong-field ionization (SFI) underpins several pivotal phenomena in attosecond science, such as high harmonic generation. We investigate the time-dependent dipole phase evolution during SFI of xenon using attosecond transient absorption spectroscopy (ATAS). This work provides new insights into the interplay between ionization dynamics and transient state populations in the presence of overlapping near-infrared (NIR) and extreme ultraviolet (XUV) pulses.

The experimental setup[1] employs a few-cycle, 760 nm NIR pump pulse to drive SFI and a time-delayed 250 as XUV probe to interrogate the dipole response associated with the $4d \rightarrow 5p$ transition in Xe. Using R-Matrix with Time-dependence (RMT[2]) calculations, we analyze the absorption spectra's dependence on time delay, revealing complex interference in the dipole response.

Three distinct processes contribute to the observed dynamics:

1. **Accumulation of Ionization** driven by the NIR pulse.
2. **Transient Population** of high-lying Rydberg states.
3. **Reversible Population Exchange:** Polarization-induced retrapping of excited states, modulating the dipole response.

These dynamics are quantified by analyzing the dipole phase behavior in the complex plane, where interference effects manifest as characteristic 'loop' structures (Fig. 1). At moderate NIR intensities ($< 2 \times 10^{14}\mathrm{Wcm}^{-2}$), interference between ionization and excitation pathways dominates. However, at higher intensities, direct ionization suppresses these effects, simplifying the dipole response.

Our findings demonstrate excellent agreement between experimental ATAS results and RMT simulations, validating the mechanistic understanding of phase evolution during SFI. This study highlights the potential of phase-resolved techniques to measure and control real-time electron dynamics at attosecond timescales.

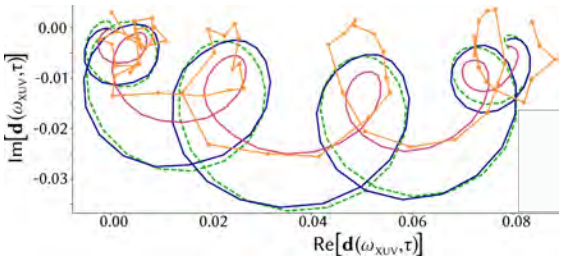


Figure 1. The complex dipole response for the $4d \rightarrow 5p$ transition following SFI of Xe from various RMT calculations (blue, green, red) and experiment (orange).

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Control of the o-Hydroxybenzaldehyde Enol-Keto Tautomerization via Selective Photoexcitation of Electronic Coherences

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Synopsis The control of photoinduced proton transfer during the tautomerization of OHBA via photoexcitation of excited electronic coherences is investigated. Our aim is to control the nuclear dynamics and ultimately augment the yield of proton transfer to the detriment of competing rotamerization of both tautomers. To do so, a multireference SCF method is coupled to semi-classical surface hopping for on-the-fly electronic structure calculation of trajectories, that are analyzed to identify the nuclear coordinates involved in the dynamics.

Electronic coherences of excited states can be used to steer the nuclear dynamics at play via selective photoexcitation [1, 2, 3] of neutral molecules, opening new avenues for controlling chemical reactivity. We apply this approach to proton transfer mechanisms that are at the heart of many chemical reactions and biochemical processes.

We investigate the excited state intramolecular proton transfer that induces the enol-keto tautomerization of o-hydroxybenzaldehyde (OHBA) [4, 5, 6] and the competition with the rotamerization of both tautomers. [7] The tautomerization process is induced by an ultrashort UV few fs pulse. The dynamics in the excited states is modeled by the semi-classical ab initio surface hopping method.

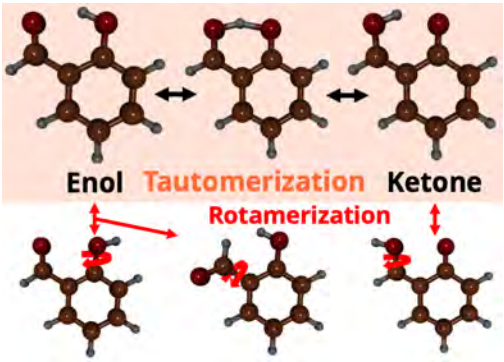


Figure 1. Competition between tautomerization of OHBA and rotamerization of both tautomers.

The electronic structure of the excited states is computed at state-averaged complete active space SCF method and compared with TD-DFT and experimental results. [8] The lowest excited states S_1 and S_2 are a dark ($n\pi^*$) and a bright state ($\pi\pi^*$) that are close in energy in the Franck-Condon (FC) region. On the ground state (S_0), the enol conformer is more stable than its tautomer while it is the contrary on S_1, thus allowing stabilization of the molecule through relaxation on the ground state after tautomerization on the first excited state. 100 trajectories were initiated in the FC region on states S_0 to S_3, and conical intersections (CIs) were optimized near hopping geometries. Trajectories were then analyzed with a singular value decomposition (SVD) technique [9] to identify the relevant nuclear coordinates describing the competing tautomerization and rotamerization and their respective CIs and design control schemes.

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Predicting ultrastrong coupling effects in extreme ultraviolet fields with combined Floquet and effective Hamiltonian theory

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Synopsis We propose a theoretical framework to describe attosecond effects beyond the rotating-wave approximation by combining Floquet theory and effective Hamiltonian theory. Giant counter-rotating oscillations of the 1s3d population in two-photon Rabi cycling helium atoms are predicted. Our theory is tested against *ab initio* simulations with pulses attainable from seeded free-electron lasers.

Intense XUV light from seeded free-electron lasers enables the exploration of novel regimes in light-matter interaction, such as Rabi oscillations in helium [1]. The experimental conditions in Ref. [1] fell within the strong coupling regime, where the rotating-wave approximation (RWA) is valid and photoionization is negligible. How to reach the *ultrastrong coupling* regime with XUV light is an open question. There, we expect the breakdown of the RWA and significant counter-rotating (CR) effects.

In two-photon Rabi cycling helium atoms, we predict that the 1s3d state can display CR oscillations on attosecond timescales that are of similar size as the Rabi oscillations themselves [2]. In contrast, such effects are absent in the ground state $1s^2$, see Figure 1. We confirm that the effect can be generated with realistic pulses, and for a range of one- and two-photon transitions in helium.

CR effects can be predicted accurately by simulating the time-dependent Schrödinger equation, e.g. with the time-dependent configuration-interaction singles (TDCIS) method [3]. However, such *ab initio* methods are inconvenient to explore the required parameter space over both intensity and wavelength of the field. We desire an approach that (a) is computationally efficient, (b) makes quantitative predictions, and (c) gives qualitative insights.

To this end, we have developed a theoretical treatment of CR effects using Floquet theory within an essential states approach. In this way, we describe both *average effects* (changes in the resonance condition) through calculation of an effective Hamiltonian H_{eff} [4], and *sub-cycle effects* of the atomic populations. It is computationally cheap, quantitatively accurate, and yields analytical expressions for the time evolution.

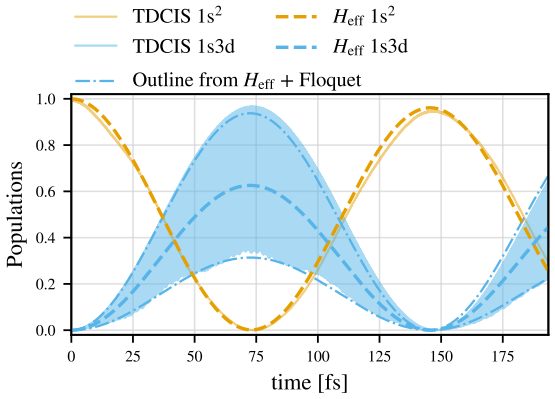


Figure 1. Two-photon Rabi oscillations of $1s^2$ and 1s3d, predicted by TDCIS and H_{eff}, with outlines of CR oscillations being predicted by combination of H_{eff}- and Floquet theory.

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Linear Dichroism of Core Excitons in LiF

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Synopsis Orbital alignment-dependent effects of bright and dark states have been the subject of countless studies in atomic systems, but these investigations have yet to be extended to the condensed phase. We investigated the polarization dependence of core exciton dynamics in LiF using attosecond transient absorption spectroscopy and theoretical analysis. By switching from parallel to orthogonal XUV and NIR polarizations, transient features associated with couplings between the 1s2p bright exciton and a previously unidentified higher-lying s-like dark state are suppressed by up to 90%.

The prospect of an experimental probe of the orbital composition of solid-state materials is highly exciting, given the complexity and high density of states of many systems. Recent attosecond transient absorption studies in He [1] revealed that NIR-induced couplings between bright np and dark ns Rydberg states that are allowed when using parallel XUV and NIR polarizations are eliminated by using crossed beam polarizations, while np to nd couplings are unaffected by the change. To determine if such sensitivity can be used to identify the character of states in the condensed phase, we investigate the polarization dependence of core exciton dynamics in LiF thin films at the Li K edge using attosecond transient absorption spectroscopy.

Linear XUV pump pulses excite electrons from the Li 1s core level into Li 2p-like regions of the conduction band, forming electron-core hole pairs (core excitons) aligned along the XUV polarization axis. A NIR probe arriving at variable time delays then perturbs the XUV-excited states, mapping the coherence decay of the core exciton. Similar to other time-resolved core exciton studies [2, 3, 4], the observed 2.4 ± 0.4 fs coherence time is attributed to a phonon-mediated dephasing mechanism [5]. The differential absorption features are shown to be highly sensitive to the relative polarization of the XUV and NIR beams. The parallel NIR probe induces couplings between the p-like bright exciton and previously unidentified resonant s-like dark excitons. When crossed pump and probe polarizations are used, the coupling between the bright and dark states is suppressed by approximately 90%. This interpretation of the orbital character of the participating states, namely the identification of the high-energy s-like dark state, is supported by simulation of a few-level model system, along with DFT calculations of the band structure.

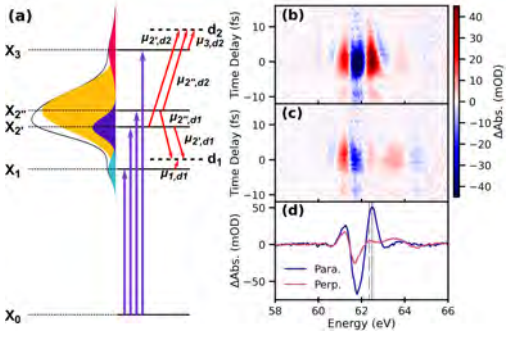


Figure 1. (a) Few-level model of LiF. The transient absorption spectrum when using parallel (b) and perpendicular (c) XUV and NIR polarizations. Panel (d) gives the differential absorption for each polarization taken at $\tau = 0$ fs.

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Rise and demise of entanglement in strongly coupled photoionization via quantum relay to spontaneous emission

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Synopsis The recent development of the Free-Electron Laser (FEL) opens up a new short-wavelength and high-intensity parameter space, enabling predictions of new ultrafast strong-coupling phenomena. Here generation of entanglement in photoionization is investigated, unveiling how it is generated, how it is altered by harnessing time symmetry, and how the electron-ion entanglement is relayed into electron-photon number entanglement through fluorescence.

In 2022, Rabi oscillations were observed at extreme ultraviolet (XUV) wavelengths using the seeded FEL at FERMI [1]. The FEL parameter space allows for investigating quantum entanglement in the photoelectric effect, generated between a photoelectron and a light-dressed atomic ion [2]. The atom, g, is ionized into the ionic ground state, α, and the field dresses the ion, inducing Rabi oscillations to state β, see Fig. 1(b). The entanglement is manifested in the photoelectron doublet, in agreement with the seminal work of Grobe and Eberly [3] and more recent works [4, 5], see Fig. 1(a). We show how this entanglement is fundamentally altered by harnessing time symmetry [6]. Using odd (zero-area) pulses changes the photoelectron distribution, forming non-overlapping photoelectron peaks fig. 1c and delayed entanglement generation. This allows for detecting entanglement without phase measurements and constitutes the first usage of the parity of time symmetry in strong-field interactions.

Including spontaneous emission [7], $\beta \rightarrow \gamma$, gives the characteristic the Mollow triplet during the pulse, Fig. 1d, and a narrow resonant peak after, Fig. 1f. The shape of the photoelectron spectra is conserved, Figs. 1(c,e), with the population being transferred $\beta \rightarrow \gamma$. The population is presented in Fig. 1g, showing how α and β increase during the pulse, and the subsequent decay $\beta \rightarrow \gamma$ on the timescale of spontaneous emission. The entanglement entropy, Fig. 1(h), between electron and ion, $\alpha\beta$, is built up during the interaction with the pulse to the maximum value of 1 (for qubit system). After the pulse, this is *quantum relayed* to fully entangle the electron with the emitted photon, $\alpha\gamma \rightarrow 1$. During the spontaneous decay, the full system entanglement (comprising: ion, electron and emitted photon) $\alpha\beta\gamma\delta$ exceeds the maximum for a qubit system.

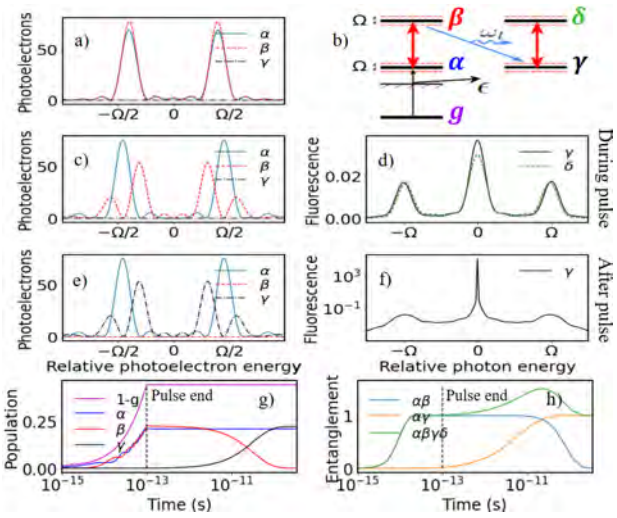


Figure 1. Photoelectron spectra (a,c,e). Schematic (b). Fluorescence spectra (d,f). Time-resolved populations (g) and entanglement (h).

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Non-classical bright states of light generated via light-matter entanglement in strong laser fields

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Synopsis Quantum optics and strong-field physics are joining forces in bringing a revolution to attosecond physics, particularly to high harmonic generation. Here we show that squeezed and massively entangled bright states of harmonic light can be obtained through a control of non-adiabatic excitations of matter driven by classical laser fields. Our results reveal significant opportunities at the interface of attosecond physics and quantum optics, with implications for quantum information science.

Traditionally, the quantum description of light has been predominantly applied in the weak-field regime, while the remarkable phenomena using intense laser, such as high harmonic generation (HHG) and above-threshold ionization (ATI), have been described in the context of classical treatment of the driving light fields. However, recent observation[1] of Schrödinger cat states in strong-field regime has upended this tradition and has opened up an exciting possibility of using intense light-matter interactions to generate bright quantum states of light.

Here we demonstrate a novel route to generating harmonics in non-classical quantum states carrying massive amount of photons. Starting with a strong laser pulse in a coherent state at a fundamental frequency, ω, and a quantum matter in an uncorrelated ground state (e.g., atomic system), the excitation of the matter by the generated harmonic light at a particular harmonic, $n_0\omega$, leads to a variety of non-classical effects, from squeezing of every harmonic to entanglement between harmonics. Crucially, the key properties of these non-trivial quantum states of light are sensitive to the shape and amplitude of the driving laser pulse due to the intensity-dependent resonances arising when matter is "dressed" by the driving pulse. The quantum effects in the generated nonlinear-optical emission emerge already in a sodium atom at the driving field intensities as modest as 10^{11}W/cm^2, as shown in Fig. 1 [2].

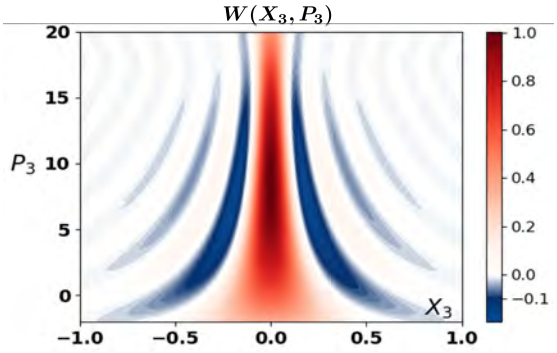


Figure 1. Wigner function for the 3rd harmonic in a sodium atom driven by fundamental radiation at 1770 nm with intensity 10^{11}W/cm^2. Here, the 3rd harmonic is resonant with the 600 nm transition between the ground and the first excited state of the soidum atom.

Building on these results, we will describe the cases involving different quantum matter dynamics, including solid-state systems driven by two-color fields, paving the way to the generation of non-classical bright states of light in solid-state devices.

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Semiconductor Wannier equations: a real-time, real-space approach to the nonlinear optical response in crystals

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Synopsis We derive the equations of motion for the electronic reduced density matrix in a crystal using a real-space basis, the semiconductor Wannier equations, as an alternative to the semiconductor Bloch equations. SBEs present several drawbacks: the need of a smooth Bloch gauge along the k-space and the introduction of extremely ultrafast dephasing times. With SWEs, we overcome these difficulties. Dephasing is introduced in real-space and the need for smooth gauge is solved. The real-space formulation of SWEs provide a bridge to strong-field intuition developed for atoms and molecules.

In the context of strong field physics in condensed matter, the semiconductor Bloch equations (SBEs) were widely as a numerical tool to provide further insight. SBEs are usually presented in the length gauge. When comparing length and velocity gauge, length gauge is preferable for two reasons: the physical interpretation in terms of band populations and the inclusion of phenomenological relaxation models [1]. However, a major disadvantage of working with the SBEs in length gauge is the necessity of derivatives in reciprocal space, and therefore, a smooth gauge choice for the eigenstates in the Brillouin zone.

The use of maximally localized Wannier functions is a way to circumvent this problem. In a previous work [2], the construction of Bloch functions from MLWFs allowed for the construction of a smooth gauge in the reciprocal space, suitable for the numerical propagation of the SBEs. In this work, we go one step further. Rather then working in a basis of Bloch functions, we directly propagate the density matrix in the basis of localized Wannier orbitals and derive the semiconductor Wannier equations (SWEs) [4]. We show that in this formulation, the inclusion of a real-space dephasing, recently introduced [3], is straightfoward to implement. The derivatives present in the SBEs are no longer present in the SWEs. The resulting equations of motion in the SWEs only involve transition dipole moments between localized states and result into a more familiar formulation able to bridge with the numerical and theoretical frameworks developed in the context of atomic and molecular attoscience. Furthermore, in the SWEs, the inclusion of electron-electron interactions at the mean-field level is simpler and physically more intuitive than in the SBEs.

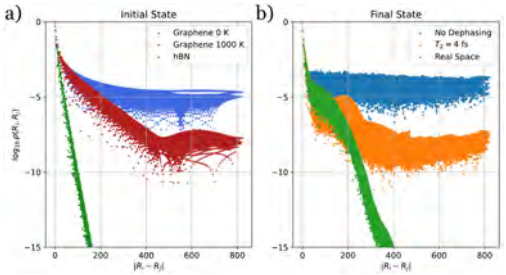


Figure 1. **a)** Scatter plot in logarithmic scale of $\rho_{\alpha\beta}(\vec{R})$ for 3 different systems: graphene at 0K, graphene at 1000K and hBN. **b)** Final state after interaction with mid-IR for three different cases: no dephasing, dephasing and real-space dephasing.

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Ultrafast Imaging Below the Diffraction Limit with High Harmonic Deactivation Microscopy

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Synopsis Combining super-resolution and label-free microscopy techniques promises new insights into ultrafast physics. We modify a pump-probe scheme so that the pump has a donut-shaped intensity profile. The donut controls non-linear emission in space while both beams are close to temporal overlap. This technique called harmonic deactivation microscopy (HADES) can resolve nanometer-sized objects at a (sub-)femtosecond temporal resolution.

Nonlinear optical microscopy provides elegant means for label-free imaging of biological samples and condensed matter systems. The widespread areas of application could even be increased if resolution was improved, which the famous Abbe diffraction limit now restrains. Super-resolution techniques can break the diffraction limit, but most rely on fluorescent labeling. This makes them incompatible with (sub)femtosecond temporal resolution and applications that demand the absence of labeling. In a recent study, we introduce harmonic deactivation microscopy (HADES) for breaking the diffraction limit in nonfluorescent samples [1].

This technique generates high harmonics in a sample (Fig. 1A) by tightly focusing near-infrared light. A donut-shaped beam with radially varying fluence deactivates the harmonics spatially (Fig. 1B). HADES reduces the harmonic point-spread function (PSF) noticeably. For third harmonic generation (THG) microscopy, a fourfold resolution improvement compared to the Abbe diffraction limit can potentially be achieved. Experiments showed that deactivation scales with harmonic order (Fig 1B inset), leading to increased resolution gains for higher-order HADES [2]. For fifth harmonic generation microscopy, we can thus expect sub-100 nm resolution.

We deactivate the High Harmonic Generation (HHG) by overlapping two incommensurate electric fields. In the material, decreased HHG efficiency in these two-color fields and increased non-coherent scattering lead to an overall amplitude suppression [2,3]. HADES utilizes these effects and increases the resolution of an existing third harmonic generation (THG) microscope visibly (Fig. 1C,D). In the next step, we develop a higher harmonic order microscope (Fig. 1E,F) capturing up to 5th-order non-linearities. Compared to THG microscopy, fifth harmonic microscopy gives a sharper image, which is in line with power-scaling laws for HHG yield with harmonic order.

Combining HADES with higher harmonic orders enables nanoscale resolutions. The general nature of harmonic deactivation allows to translate the results to a variety of materials subject to current research, e.g. 2D materials, topological insulators [2]. The intrinsic attosecond nature of HHG yields the potential of unveiling sub-femtosecond processes on a nanoscale [4].

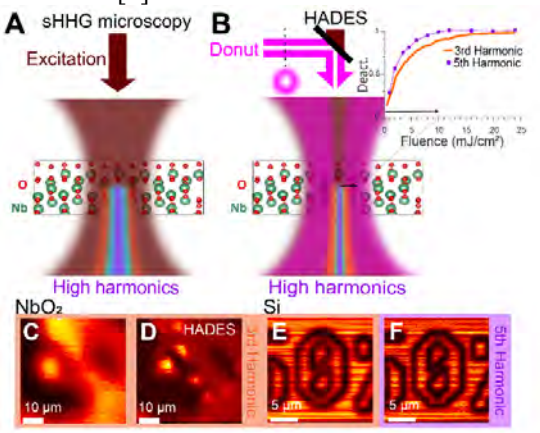


Figure 1. A: Tightly focused NIR beam excites HHG in a sample for imaging. B: A donut-shaped beam reduces the spatial extent of HHG. The deactivation for higher orders (inset, red) is faster than for lower harmonics (blue), increasing the resolution with harmonic order. C-D: Harmonic deactivation microscopy (HADES) improves the resolution of a THG microscope visibly in an NbO2 sample. Further comparison of a THG image (E) in Silicon to a fifth harmonic image (F), shows increased sharpness for higher harmonics.

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Origin of sub-optical-cycle dephasing in high-order harmonic generation in crystalline solids

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Synopsis High-order harmonic generation in solids is critically sensitive to the dephasing of the coherent electron-hole motion. The exact physical mechanisms, defining the dephasing, have remained unclear. Using detailed experimental measurements and rigorous numerical simulations, we show that electron-phonon scattering is the predominant ultrafast dephasing mechanism, suggesting a novel concept of the momentum-dependent dephasing time and resolving the contradiction between the ultrafast dephasing, commonly used for HHG modeling, and narrow line widths observed for low-power excitations (excitons).

Experimentally, we measure the wavelength dependence of HHG spectra at fixed laser intensity, following the prediction that the harmonic yield drops exponentially with increasing the laser wavelength, and the decrement depends on the dephasing time [1]. The experiments are carried out with WS_2 and MoS_2 monolayers and a polycrystalline 140 nm thick CdSe film, deposited on a sapphire substrate. The harmonics were generated by femtosecond mid-IR pulses tuned within the 3-5 µm spectral range. The wavelength dependence in the harmonic yield, defined as as the integrated spectral intensity within the photon energy range 2-5.5 eV, is shown in Fig.1. The dependence shows a power-law decay with the power index of 8.9 ± 1 and 8.2 ± 0.9 along the zigzag and armchair directions correspondingly. The power index is only weakly dependent on the laser intensity (Fig.1c,d).

Simulations that intrinsically lack dephasing processes, such as ab-initio real-space and real-time time-dependent density functional theory (rt-TDDFT), failed to reproduce the experimental results. The physical origin of the dephasing is linked to the major scattering processes in solids, namely electron-electron and electron-phonon scattering. The weak dependence of the slope on the laser intensity excludes the first as the major contribution to the dephasing. To theoretically investigate the contribution of the electron-phonon scattering, we used DFT methods to calculate the *k*-dependent, band-specific electron-phonon scattering rates. The rates demonstrate a sharp dependence on the electron lattice momentum, changing from $5 \cdot 10^{-3}$ fs^{-1} at the K-points to ≈0.5 fs^{-1} in the vicinity of the Γ-point. These rates are used to calculate *k*-dependent dephasing times, which are utilized for the interband polarization in our multi-band, two-dimensional semiconductor Bloch equations (SBE). The calculated wavelength dependence of the harmonic yield shows very good agreement with the experimental results.

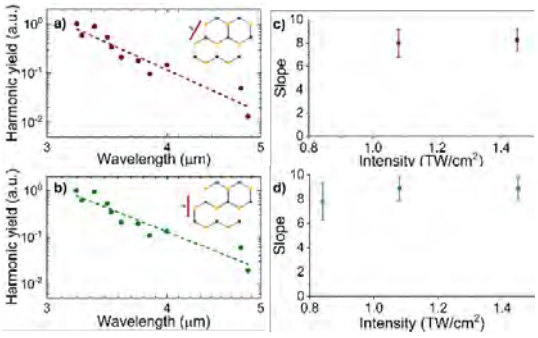


Figure 1. Wavelength dependence in the harmonic yield in WS_2 for a) zigzag and b) armchair orientation of the laser polarization. The dotted lines show linear fit. The dependence of the absolute value of the slope in the linear fit on the laser intensity for c) zigzag and d) armchair.

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High Harmonic Generation from Localized Electronic States in Solids

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Synopsis High harmonic generation (HHG) in solids reveals ultrafast electronic dynamics, but localized electronic states (LESs) are less explored. Here, we investigate HHG from LESs in solids, uncovering unique spectral features absent in trivial bulk materials. We identify the circular dichroism of the dipole transition in HHG from nanosized topological insulators, the enhancement of HHG yield in heterojunctions, and excitonic effects in pump-probe spectra. These findings highlight the potential of LESs for tailoring HHG spectra and enhancing HHG efficiency, advancing ultrafast optoelectronics.

High harmonic generation (HHG) in solids has emerged as a powerful tool for probing ultrafast electronic dynamics and strong-field interactions in condensed matter systems [1]. While extensive research has focused on HHG from delocalized Bloch states, the role of localized electronic states (LESs), induced by topological edges [2], interface effects [3], or Coulomb interactions [4], remains less explored despite their significant influence on material properties. In this work, we investigate the mechanisms and spectral features of HHG from LESs in solids, employing the time-dependent Schrödinger equation and the semiconductor Bloch equation to model the strong-field optical interactions. Our results demonstrate that LESs introduce distinct spectral features in the harmonic emission, which are absent in trivial bulk materials.

We identify the distinct circular dichroism of the dipole transition in HHG from nanosized topological insulators with localized topological edge states [5]. This circular dichroism is mapped onto the large ellipticity of HHG and can manifest the topological order of the bulk states. Additionally, we demonstrate that the HHG yield in the second plateau is higher in heterojunctions compared to homogeneous materials, and it increases with the built-in potential barrier of the heterojunction. Furthermore, the formation process of excitons in solids can also be encoded in the HHG spectra of pump-probe experiments.

Our findings provide new insights into the interactions between intense laser fields and condensed matter systems, emphasizing the potential of LESs for tailoring HHG spectra and enhancing harmonic efficiency. By bridging attosecond science and condensed matter physics, our work paves the way for exploiting LESs in ultrafast optoelectronic devices.

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Theoretical description of attosecond momentum-resolved resonant x-ray scattering of photo-excited molecules

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Synopsis We explore the capabilities of attosecond momentum-resolved resonant inelastic x-ray scattering (RIXS) for imaging charge migration. We theoretically describe an experiment in which an optical pump pulse induces electron dynamics in sexithiophene through photo-excitation, and an attosecond x-ray probe pulse resonant to the Sulfur K-edge is employed to probe the dynamics. We analyze the probability of scattering of a photon with a certain momentum and connect it to the properties of electron dynamics being probed.

Attosecond RIXS is capable of imaging charge density and charge migration in the considered system [1,2]. An optical pump pulse excites the system into a superposition of excited states, consequently inducing charge density migration within the molecule. A resonant X-ray probe pulse, tuned to the sulfur K-edge, with momentum k_{in}, is scattered from a molecule. We propose to measure the momentum of the scattered photon (k_F) via this process.

We use the second-order time-dependent perturbation theory to describe the interaction of an ultrashort x-ray pulse with a nonstationaty electronic system. We apply the Configuration Interaction method to obtain the excited and core-excited states. The probability of observing the scattered photon, $P(t,Q)$, depends on the time delay between the pump and probe pulses and on the difference between the momenta of the absorbed and emitted photons ($Q=k_{in}-k_F$).

We calculate this probability in the Q-space for different time delays and at various emitted photon frequencies. We demonstrate that the signal contains information about the time-dependent electron density and analyze how to retrieve it.

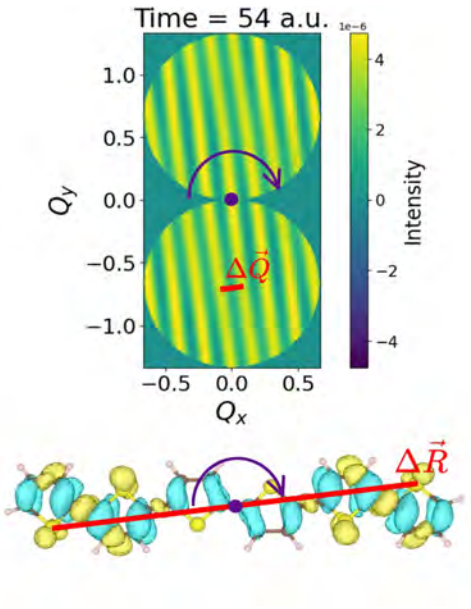


Figure 1. Comparison of $P(t,Q)$ with a charge density.

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Quantitative characterization and manipulation of quantum entanglement in multiphoton ionization

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Synopsis We investigate the $(2+1')$ resonance-enhanced multiphoton ionization of argon atoms driven by two ultraviolet laser pulses with variable time delays. By tuning the inter-pulse delay, we demonstrate controlled manipulation of photoelectron purity, which is directly encoded in the photoelectron angular distributions (PADs). Through a systematic analysis of the PADs' anisotropy parameters, we achieve quantitative reconstruction of the photoelectron purity and simultaneously extract the phase difference between coherently populated configurational states.

Quantum entanglement between photoelectrons and residual ions in photoionization processes has emerged as a key phenomenon in attosecond science, with profound implications for quantum information and ultrafast dynamics characterization [1,2]. Here we demonstrate active control of ion-photoelectron entanglement through quantum beat manipulation in $(2+1')$ resonant-enhanced multiphoton ionization (REMPI) of argon atoms.

The protocol initiates with coherent excitation of the $4p$ orbital, where the angular momentum coupling between the ion and the excited electron splits the state into $3s^2 3p^5 4p$ (^{1}S^e) and $3s^2 3p^5 4p$ (^{1}D^e). Subsequent ionization from this superposition state generates photoelectrons with mixed $m = 0, \pm 1$ orbital components, whose ratio can be dynamically controlled through the inter-pulse delay τ. Our combined R-matrix with time-dependence (RMT) and multi-configurational time-dependent Hartree-Fock (MCTDHF) calculations reveal striking quantum beat modulations in photoelectron angular distributions (PADs). A systematic analysis of the τ-dependent anisotropy parameters in PADs can decode the photoelectron purity, enabling the entanglement monitoring, as visualized in Fig. 1. Besides, we identify a characteristic phase shift between the $3s^2 3p^5 \varepsilon d$ (^{1}F^o) and $3s^2 3p^5 \varepsilon d$ (^{1}P^o) scattering channels, serving as a quantum interferometric signature of entanglement.

While demonstrated for argon, this protocol establishes a general framework applicable to complex atoms and molecules, opening new avenues for quantum state engineering in strong-field ionization processes.

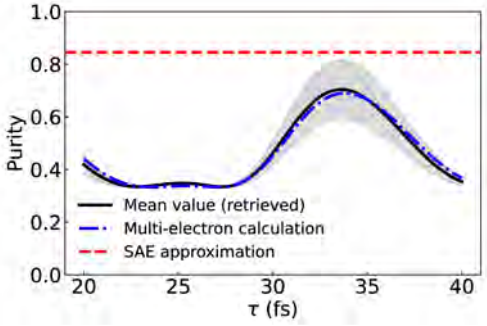


Figure 1. Photoelectron purity retrieved from the PADs (gray area), from multi-electron calculations (blue), and from calculations under single-active electron approximation (red).

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Ultrafast Optical Control of Multi-Valley States in 2D SnS

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Synopsis We theoretically investigate optical valley control in 2D SnS monolayers. In contrast to conventional materials that typically rely on a single valley pair, SnS offers the possibility of exploiting multiple valley states, thereby significantly enhancing the degrees of freedom for valley manipulation. Moreover, SnS monolayers exhibit both linear and circular dichroism enabling flexible valley-selective excitation by simply adjusting the light polarization. Notably, we demonstrate the precise sub-cycle optical control of electron dynamics at multiple valley points on femtosecond timescales.

Bulk monochalcogenides (MC) have layered structures, featuring D_{2h}^{16} space group with puckered layers. Experimental findings have demonstrated valley-selective linear dichroism, revealing unique valley-specific optical transition rules [1]. Interestingly, 2D layers of MC, consisting of mono- to a few layers, have been synthesized [2]. The transition from bulk to monolayer results in the non-centrosymmetric C^7_{2v} space group. This raises the intriguing question: can monolayers of MCs exhibit spin(s)-valley(τ) coupling (s·τ) physics, where the helicity of light controls both spin and valley characteristics, ultimately giving rise to circular dichroism [3]? To address this question, we study optical valley control in 2D SnS monolayers using the time-dependent density functional theory. We also propose the ultrafast optical control of multi-valley selection on femtosecond timescales, paving the way for advancements in valleytronics.

For a laser pulse with a wavelength of 1127 nm and a foot-to-foot pulse length of 8 optical cycles, the maximum valley polarization is achieved along the X-Γ-X' ($\theta = 0°$, *x*-polarization) and Y-Γ-Y' ($\theta = 90°$, *y*-polarization) directions, indicating strong valley selectivity in high-symmetry directions. *x*/*y*-polarized light primarily excites the corresponding V_X/V_Y valley [Fig. 1(a)].

The combination of inversion symmetry breaking and spin-orbit coupling in 2D SnS yields Berry curvatures of opposite signs at the X and X' points. As a result, circularly polarized light with the same wavelength and pulse duration as stated in the last paragraph can drive s·τ to simultaneously manipulate spin and valley at X and X' points [Fig. 1(b)], a feature rarely observed in conventional materials.

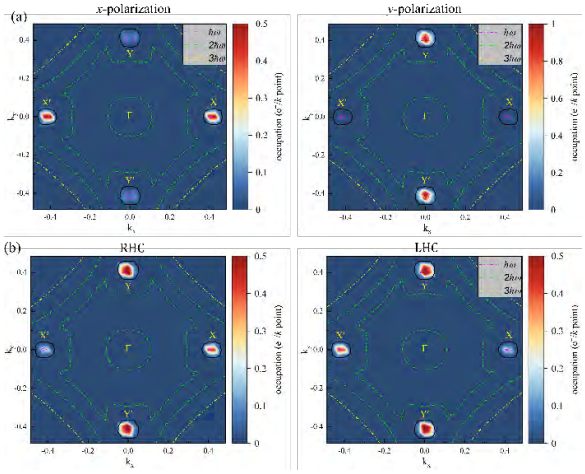


Figure 1. Excited electron populations in the first Brillion zone of the conduction band at the end of a pulse with field strength of 10^{10} W· cm^{-2}. (a) linearly polarized pulse (b) circularly polarized pulse.

We also propose a method for rapidly switching between multiple valley states. This involves utilizing ultrashort (3 optical cycles) carrier-envelope phase controlled linearly polarized pulses with a wavelength of 3100 nm. Bragg scattering results in a clear valley asymmetry between X-X' and at Y-Y' valley points (not shown). Thus, a robust control of multi-valley in 2D SnS at sub-cycle even in the absence of opposite Berry curvature and time reversal symmetry.

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Sub-cycle switchable light-induced Floquet topological flat bands in Dirac system and its dynamics

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Synopsis In recent years, Floquet engineering has provided a novel paradigm for utilizing periodic driving to manipulate the topological states of quantum materials. In this paper, through theoretical modeling and numerical calculations, we propose and demonstrate the feasibility of realizing topological flat bands in optically driven two-dimensional lattices. Here, we have developed a sub-cycle-resolved Floquet-Bloch electronic dynamics theory. Based on this theory, we find that elliptically polarized light as well as trefoil light with specific frequencies and polarizations can dynamically modulate the electronic band structure, inducing topological flat bands with non-zero Chern numbers in momentum space.

Under elliptically polarized or trefoil-shaped optical fields, the interplay of PT-symmetry breaking and rotational-symmetry disruption induces anisotropic renormalization of light-driven Floquet bands, leading to the emergence of Chern-insulating flat bands with quantized Chern numbers.

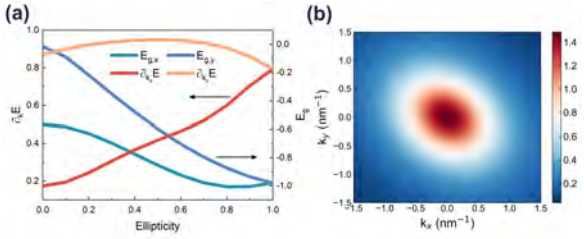


Figure 1. Elliptically polarized light induced (a) flat bands and (b) anisotropic Berry curvature.

This renormalization mechanism, attributed to optically engineered complex next-nearest-neighbor hopping terms enables unprecedented control over nonequilibrium topological phases.[1]

Crucially, in trefoil-field-driven systems, the sub-cycle tunability of the field's relative amplitude and phase permits ultrafast switching between Floquet-trivial and Chern-insulating phases on femtosecond timescales.

Our findings not only establish a universal framework for manipulating Floquet topology via multiscale symmetry engineering but also bridge the gap between ultrafast photonics and robust quantum transport. The proposed paradigm opens avenues for designing reconfigurable topological optoelectronic devices, fault-tolerant quantum architectures, and energy-efficient switching technologies, marking a pivotal step toward dynamical control of quantum matter.

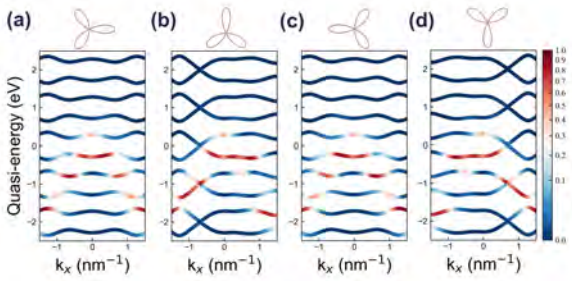


Figure 2. Trefoil lightwave-induced sub-cycle switchable light-induced Floquet topological flat bands

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Pseudospin-driven polarimetric singularities in high harmonic generation of black phosphorus

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Synopsis Pseudospin exhibits opposite orientations in the electron and hole states of black phosphorus (BP). Using the quantum master equation (QME), we reveal a unique polarimetric singularity in BP's high harmonic generation (HHG), unlike in graphene or transition-metal dichalcogenides (TMDCs). This singularity arises from pseudospin selectivity, governed by the symmetry between pulse polarization and pseudospin orientation. These pseudospin-induced harmonic singularities suggest the integration of pseudospintronics into the light-matter dynamics of 2D materials.

BP consists of two sublattices, resulting in an electronic degree of freedom known as pseudospin [1]. In particular, unlike graphene, BP is a bipolar pseudospin semiconductor, where the electron and hole states exhibit opposite pseudospin polarization, as well established by angle-resolved photoemission spectroscopy (ARPES) experiments [2]. Furthermore, pseudospin-selective Floquet band engineering has been actively explored using tr-ARPES, and its feasibility has been well demonstrated [3,4].

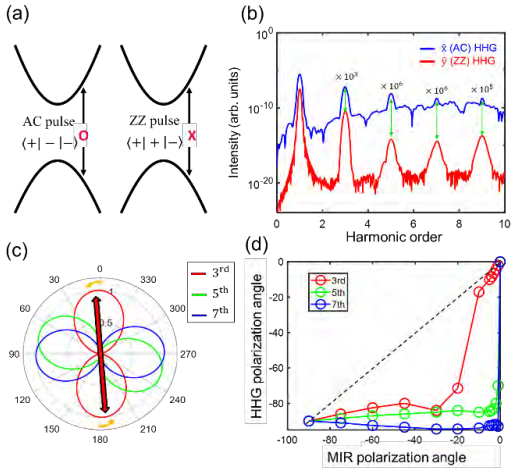


Figure 1. (a) According to the symmetry of matrix element $\langle \boldsymbol{c}|\boldsymbol{A}\cdot\boldsymbol{P}|\boldsymbol{v}\rangle$, AC(ZZ) pulse induce even(odd) function. This symmetry induces a strong intensity difference of HHG. (b) Calculated HHG in BP. (c-d) Due to the optical selection rule, when the MIR pulse polarization is rotated by 1° (orange arrow), the 5th and 7th HHG pulses exhibit a significant deviation along the x-axis.

This pseudospin selection rule is determined by the symmetry of the matrix element $\langle f|A\cdot P|i\rangle$, which consists of the final state, pulse polarization, and initial state. In conventional ARPES studies, the final state corresponds to the photoelectron state, while the initial state is a valence or conduction band state. However, in HHG, the electron dynamics occur within the material, meaning that the states are strictly confined to the conduction $|+\rangle$, and valence $|-\rangle$, bands. Consequently, the intensity of interband transitions is entirely determined by the polarization of the pulse.

To verify the application of this pseudospin-based optical selection rule in HHG, we performed QME calculations based on a monolayer BP tight-binding Hamiltonian with a bandgap of approximately 1.6 eV. By varying the mid-infrared (MIR) pulse polarization direction (0.26 eV) along the x and y axis, we observed intensity differences of 10^3 to 10^6 magnitude in the 3rd, 5th, and 7th harmonics of HHG. Due to the HHG intensity difference caused by the pseudospin selection rule along the x and y directions, even a 1-degree tilt of the incident MIR pulse from the y-axis results in the emitted HHG being directed along the x-axis. Furthermore, when the bandgap and the integer multiples of the pulse energy become comparable, interband transitions become dominant in the 5th and 7th harmonics, leading to the emergence of polarimetric singularities.

Extending the concept of pseudospin selection from conventional tr-ARPES to HHG within the framework of light-matter interaction is expected to provide deeper insights into ultrafast dynamics, including Floquet states.

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Revealing the Berry phase under the tunneling barrier

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Synopsis We introduce the manifestation of a complex geometric phase accumulated by an electron wavepacket during the tunneling mechansim in solid-state systems. We experimentally resolve this phase by inducing an internal interferometer, composed of two electrons trajectories, while resolving their relative complex phase via HHG spectroscopy.

Whenever a quantum system undergoes a cyclic evolution governed by a change of parameters, it acquires a phase factor, known as the geometric phase. Its most common formulation, the Berry phase [1], is responsible for many phenomena ranging from condensed-matter physics [2], fluid mechanics and optics. When this phase is accumulated during evolution in a *complex* parameter space, the geometric phase itself becomes complex, with its imaginary part leading to amplification or attenuation of the wave under study. This imaginary part of the geometric phase exhibits interesting properties such as gauge-independence even for open loops in parameter space [3].

When the electronic wavefunction undergoes tunneling between the valence and the conduction bands in a solid state system, it propagates in complex parameter space. In the presence of a non-zero Berry curvature such dynamics leads to the accumulation of a complex geometric phase. This phase can be attributed to the geometric phase accumulated under the potential barrier, with the electron-hole wavepacket being attenuated or amplified accordingly.

We experimentally probe this phase [4] by performing high harmonic generation (HHG) spectroscopy in an inversion-broken high bandgap crystal (α-quartz) which experiences a non-zero Berry curvature [5]. Along Γ-K, the complex geometrical phase breaks the symmetry between two tunneling events evolving along the first and second half cycles of the laser field. Adding a second harmonic perturbation, polarized parallel to the fundamental field, allows us to probe the complex properties of this phase. The second harmonic field breaks the symmetry between concecutive half cycles – slightly enhancing the barrier along one half cycle, while suppressing it along the second half cycle. Such perturbation works along or against the symmetry breaking imposed by the imaginary Berry phase. It allows to probe this phase and map it into the HHG spectrum.

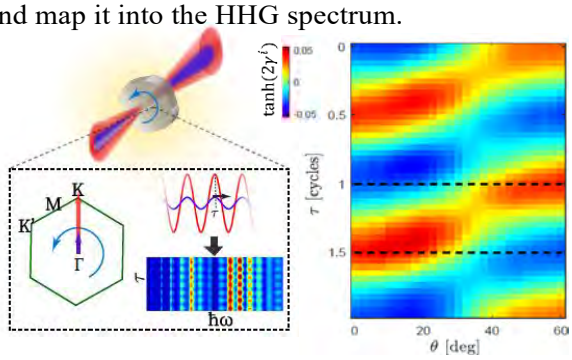


Figure 1: Experimental setup along with extracted imaginary geometric phase modulation signal per crystal angle and SH delay [4].

Resolving the imaginary geometric phase in solid state systems opens up new perspectives in the study of geometrical phases in light driven systems – where the fundamental properties of the solid change during less than one optical cycle.

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Polarization-dependent High-Harmonic Generation in Organic Molecular Crystals

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Synopsis In this work, we demonstrate efficient high-harmonic generation (HHG) up to the 17th order from an organic molecular crystal (OMC) with perfectly aligned molecules. The harmonic yield is the strongest for laser polarizations that are aligned along nearest-neighbor directions. Simulations show that HHG in OMCs can be applied to probe the strength of the weak intermolecular coupling.

High-harmonic generation (HHG) can be applied to probe the electronic and crystalline structure, as well as ultrafast dynamics of various inorganic solids. [1, 2]

In this work, we explore HHG in a new class of targets: thin organic molecular crystals (OMCs), focusing on pentacene crystals. Compared to covalent and ionic-bonded crystalline solids, the molecules that bond to form OMCs are much weaker coupled, which is reflected by flat electronic bands. At the same time the perfect alignment of the molecules in OMCs could open up the opportunity of efficient HH spectroscopy of rigid large organic molecules, without the need for complicated alignment techniques.

Surprisingly, we find that the pentacene crystal withstands high laser intensities and allows for efficient HHG up to order 17. Experimental measurements of harmonic yields, as well as time-dependent density functional theory simulations, reveal a strong dependence on the polarization direction of the driving field. Higher harmonic orders are particularly sensitive to intermolecular couplings, including nearest and next-nearest neighbors, as indicated in Fig. 1 for the 13th order. Single molecule calculations fail to reproduce the observed effect.

Tight-binding model calculations show that weaker intermolecular couplings require higher harmonic orders to probe the crystal structure. Our findings highlight the spectroscopic potential of HHG to probe intermolecular couplings in OMCs, marking an important advance toward all-optical techniques for studying electronic properties and dynamics in complex organic materials.

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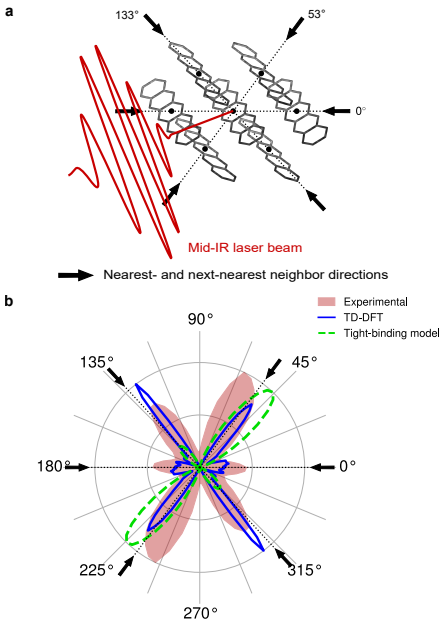


Figure 1. **a**, Alignment of the pentacene crystal with the laser propagating into the plane. **b**, Normalized harmonic yield for the 13th harmonics depending on the polarization of the driving field obtained experimentally (red), by time-dependent density functional theory (blue), and with a simple tight-binding model (dashed green).

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Impact of Solvation on the Ultrafast Ring-Opening Dynamics of Furfural

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Synopsis A comprehensive study on solvation-dependent ring-opening dynamics, comparing the ultrafast UV-induced photoreactions in the gas phase and pure liquid environments of furfural is presented. By combining time-resolved photoelectron spectroscopy (TRPES) with quantum mechanics/molecular mechanics (QM/MM) surface-hopping simulations, an unprecedented agreement between experiment and theory has been reached.

Understanding the role of solvation in excited-state molecular dynamics is essential for controlling photochemical reactivity. A key benchmark system in this context is the UV-induced ring opening of aromatic heterocycles such as furfural. Previous studies using transient absorption [1] and photoelectron spectroscopy [2] have provided insights into the photochemistry of furfural in the gas phase. Advancing such studies to the liquid phase, we present a comprehensive investigation of the effects of solvation on these ring-opening dynamics. By combining a tabletop monochromatized high-harmonic source with heatable in-vacuum sample delivery and liquid microjet technology, we directly compare the ring-opening dynamics in the gas and neat liquid phases under otherwise identical experimental conditions. To complement our experimental approach, in-depth ab-initio QM/MM surface hopping calculations provide a unified theoretical framework for both phases of matter, achieving excellent agreement with experimental observations. Notably, we reveal that the underlying mechanism of ring opening changes significantly from the gas to the liquid phase, both in timescale and chemical pathway as seen in panels a) and b) of figure 1. This work represents a major step forward in the study of ultrafast photochemical dynamics, offering new insights into solvation-mediated electronic relaxation dynamics on ultrafast timescales.

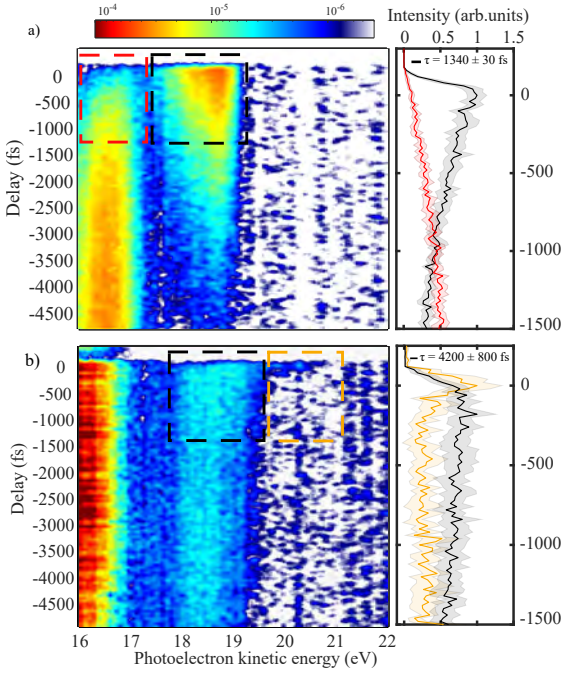


Figure 1. TRPES of furfural in gas phase (a) and pure liquid phase (b). The colored boxes denote the integrated lineouts in the adjacent panels respectively. The region between 16 and 17 eV in (b) panel is dominated by the broadened ground state signal fluctuations. The indicated lifetimes are fitted using an exponentially modified Gaussian.

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All-attosecond transient reflection spectroscopy

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Synopsis The large configuration space and interaction strength of solid state systems presents challenges for time-resolved spectroscopy and require attosecond temporal resolution. Here we introduce all-attosecond transient reflection spectroscopy, enabling core excitation in both the pump and the probe steps. We present first results on XUV-excited ionic crystal insulators, shining light on the quasi-particle dynamics of core-excitons.

Solid state materials are true many-body systems, often reaching electron densities beyond 10^{20} cm^{-3}. In the temporal domain, this large interaction space can present itself as a variety of ultrafast dynamics following interaction with light that lie beyond the PHz barrier.

Previously, transient absorption and reflection spectroscopy using attosecond XUV pulses have been shown to be an insightful tool in understanding these dynamics, such as photoinjection [1] and thermalization of carriers [2], charge localization [3] and exciton decoherence [4]. These experiments combine the high spectral resolution and core-level sensitivity of XUV transient spectroscopy with excellent temporal resolution. The latter so far has been limited by requiring a strong low-frequency pulse (UV to THz range) to accompany the relatively weak attosecond XUV pulse to provide the transient signal.

A longstanding ambition in attosecond science has been to perform all-attosecond pump-probe spectroscopy, eliminating the need for strong laser fields while enabling specific excitation of valence and / or core levels in both the pump and the probe steps. Recently, we have conducted the first all-attosecond transient absorption spectroscopy experiments in atoms using a table-top HHG source [5, 6].

Here we present the first demonstration of all-attosecond transient reflection spectroscopy (AATRS), with results obtained for ionic crystals. An AATRS map recorded in BaF_2 is shown in Fig. 1. Preliminary analysis suggests a shift in energy of core-exciton quasi-particle resonances following one-photon interband transition across the insulator's bandgap (~10eV).

Our results demonstrate that all-attosecond pump-probe spectroscopy can be transferred to solids without significant sample degradation. This opens the door to study electron dynamics in a wide range of materials from a core-level perspective and across large bandwidths on the attosecond domain. For instance, it may be possible to observe core-hole decay and charge transfer processes in solids directly in the time domain.

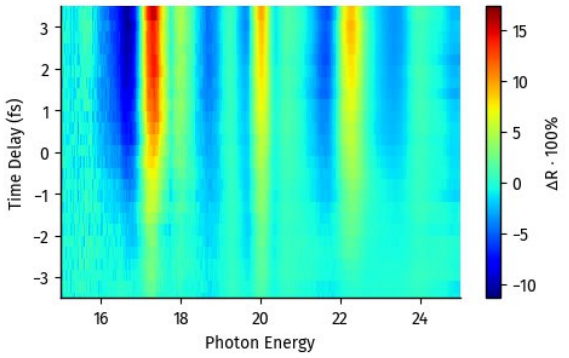


Figure 1. All-attosecond transient reflection spectrum of BaF2 in the Ba^{++} 5p core-excited region, showing strong shifts in the core-exciton resonance reflectivity due to carrier excitation by the XUV pump.

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Efficient quantum state tomography of photoelectrons via polychromatic infrared pulses

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Synopsis Photoelectron quantum state tomography allows measuring the density matrix describing photoelectrons emitted following the ionization of atoms or molecules by ultrashort extreme ultraviolet light pulses. In this work, we present a new quantum state tomography protocol based on two-photon ionization of the target by an extreme ultraviolet pulse and a polychromatic infrared pulse allowing the measurement of the photoelectron quantum state with only two measurements.

The advent of attosecond science has opened the possibility to study the dynamics of photoemission on its natural time scale by measuring the spectral amplitude and phase of the emitted photoelectron wave packets. However, this approach turns out to be insufficient if the photoelectron is not described by a pure state. For mixed states, the characterization of the photoelectron quantum state requires measuring the photoelectron density matrix using quantum state tomography (QST).

We present a QST protocol based on two-photon ionization by an ultrashort extreme ultraviolet (XUV) light pulse and a polychromatic infrared (IR) pulse. This technique is a variation of the KRAKEN scheme [1,2] that uses IR pulses with only two spectral components. The experimental principle is illustrated in fig.1: an ultrashort extreme ultraviolet pulse is used to ionize the target generating a photoelectron that are probed by a tunable delayed IR. In the original KRAKEN, the probe is bichromatic with frequencies ω_1 and ω_2 spaced by $\delta\omega = \omega_2 - \omega_1$. By collecting the photoelectron spectrogram for different delays τ between the XUV and IR, we can isolate the signal oscillating at the frequency $\delta\omega$ which is proportional to the subdiagonal $\langle\epsilon|\rho|\epsilon + \delta\omega\rangle$ of the density matrix [2]. These measurements can be repeated for different values of delta omega in order to reconstruct different subdiagonals of the density matrix..

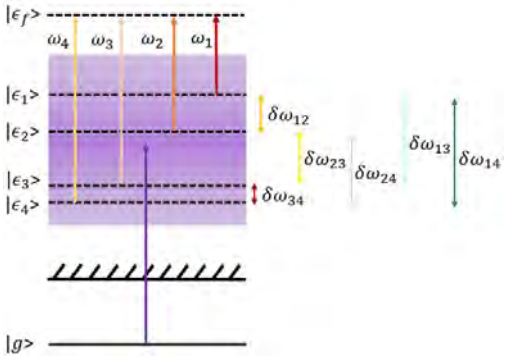


Figure 1. Principle of the Poly-KRAKEN protocol. With a measurement, it reconstructs 6 subdiagonals of the density matrix and the corresponding complex conjugate.

This procedure is time consuming and imposes strong stability constraints on the experimental setup. The poly-KRAKEN technique aims to solve this issue: we exploit a polychromatic IR and therefore collect multiple subdiagonals corresponding to different $\delta\omega_{ij}$ with the same measurement, fig.1. The set of IR frequencies are selected according to the Golomb ruler ensuring that each beating frequency is unique. We will present the reconstructed density matrix for helium and argon and how they can be used to determine whether the ion and the photoelectron are entangled or not.

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Quantum tomography of nonperturbative harmonic light from solids

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Synopsis We present the first measurement of the full quantum state of non-perturbative third harmonic generation from graphite using a novel homodyne detection scheme. Our results show non-classical characteristics including anti-squeezing and signatures of squeezing.

The intersection of quantum and extreme nonlinear optics leads to fascinating phenomena in the emerging field of extreme nonlinear quantum optics. While high-harmonic generation (HHG) has gained significant interest as a non-perturbative light-matter interaction, and quantum optics explores subtle interactions between quantas of light and matter, combining these fields challenges conventional assumptions about the quantum nature of light in nonlinear processes [1-3].

A main gap in the field remains measuring the quantum state of light generated by non-perturbative nonlinear processes, which requires a sub-cycle quantum light tomography approach. Our research bridges this gap by presenting the first complete measurement of the quantum state of light from a non-perturbative nonlinear process. We measure (Fig. 1(a)) the quantum state of 3rd harmonic driven by a mid-infrared laser out of highly ordered pyrolytic graphite (HOPG). The emitted harmonic is in the visible regime, where the quantum efficiency of detectors is sufficiently high for homodyne tomography. The vanishing bandgap of HOPG generates a non-perturbative 3rd harmonic, which we characterize in a novel homodyne scheme. This scheme mixes the signal with a strong, phase-locked coherent state local oscillator (LO), derived from a zinc oxide (ZnO) crystal. The ZnO generates a perturbative third harmonic LO that shares the same properties as the signal except its quantum light state. This allows reconstruction of the signal's Wigner function (Fig. 1(b)), revealing non-classical characteristics includind anti-squeezing and traces of squeezing (Fig. 1(c)).

In conclusion, our work provides a novel method to study non-perturbative HHG in solid-state materials. The measurement and comparison of the quantum properties of light generated by perturbative and non-perturbative processes will shed new light on the fundamental light-matter interactions in solid-state HHG.

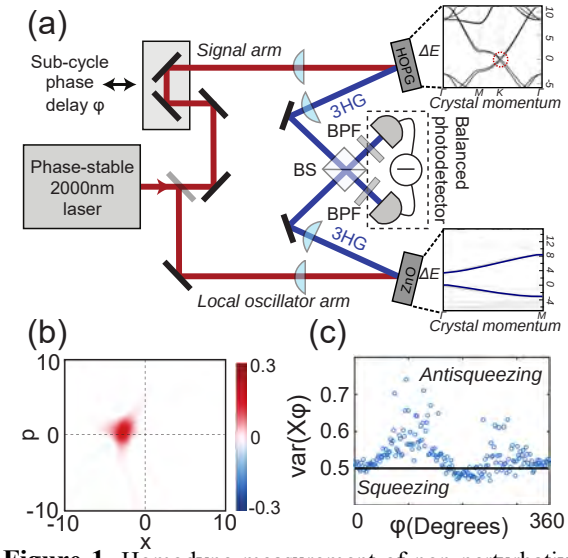


Figure 1. Homodyne measurement of non-perturbative harmonic. (a) Experimental setup: Two 3rd harmonic pulses, non-perturbative (HOPG) and perturbative (ZnO), generated by a 2 μm femtosecond laser and interfere in a homodyne scheme measuring the signal as a function of sub-cycle phase delay φ (BS: 50:50 beam splitter, BPF: bandpass filter). Insets: band structures. (b) Measured Wigner function. (c) Quadratures variance as a function of φ, showing anti-squeezing and slight squeezing.

We envision the generation of non-classical femtosecond and attosecond light pulses in the ultraviolet and extreme ultraviolet using HHG, opening up new frontiers for quantum optics.

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Photoelectron emission from silver clusters on substrates

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Synopsis

This study examines how intense laser light interacts with silver nanoclusters, driving electron emission via plasmonic effects. Our near-field calculations around faceted particle reveal "hot spots" at particle corners, with field enhancement depending on nanoparticle shape, orientation, and substrate interactions. TDDFT simulations analyze plasmon resonance effects on emission dynamics. These insights increase our understanding of nanoscale light-matter interactions, with potential applications in ultrafast electron sources and plasmonic nanodevices.

Plasmon-assisted photoemission occurs when strong localized electromagnetic fields from surface plasmons enhance electron emission under laser illumination. This process is vital for ultrafast electron sources, optoelectronics, and nanoplasmonic sensors [1] and plays a key role in biological sensing [2], cancer therapy [3], and photocatalysis [4], making its study highly relevant.

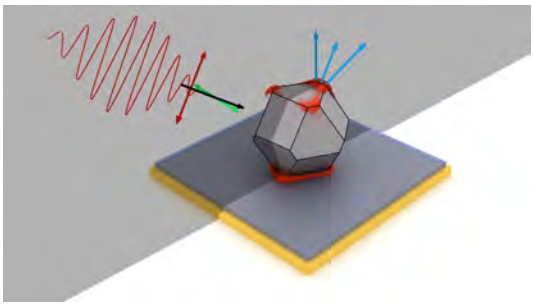


Figure 1. Model of a particle on substrate. The enhancements of the field at the top and bottom of the particle are evident.

We investigate photoelectron emission from silver clusters (5–15 nm) deposited on silica substrates with a thin oxidation layer. The particles are illuminated by an 800 nm laser at an intensity of 10 GW/cm^2. Our goal is to understand how nonlinear plasmon excitation influences photoemission, the role of localized field enhancements ("hot spots") in the emission dynamics and the cluster visibility in PEEM, and the effect of the substrate on the near-field distribution. We further explore the possibility of externally controlling plasmon-assisted photoemission.

To characterize the local field distribution, we use the Green dyadic method, which accurately describes the enhancement of the electric field around nanoparticles. This approach helps to identify regions of maximum enhancement, to determine field decay rates, and analyze the influence of particle shape and orientation on near-field properties.

To investigate the role of plasmon resonance in electron emission, we perform time-dependent density functional theory (TDDFT) simulations in one dimension along the primary emission direction, coinciding with regions of highest field enhancement, such as sharp corners.

These results deepen our understanding of localized field effects in plasmon-assisted photoemission, with future work extending the analysis beyond TDDFT.

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Element-Specific Phonon Dynamics in Molybdenum Dioxide Revealed by Transient Absorption Spectroscopy

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Synopsis Our combined experimental and computational study of molybdenum dioxide provides element-specific insights into atomic-scale dynamics driven mainly by two optical A_g phonon modes. The 6 THz mode is mainly associated with molybdenum displacements and the 11 THz mode with oxygen displacements, which allows element-specific analysis. Variations in measured phonon amplitudes across the extreme ultraviolet (XUV) spectrum of the transient absorption spectrum reveal changes in the density of states (DOS), as corroborated by density functional theory (DFT).

In the analysis of attosecond transient absorption spectroscopy measurements, time-dependent DFT [1] or the computationally intensive Bethe-Salpeter equation [2] are often used to model absorption changes. Here, we demonstrate that a simpler, time-independent DFT approach is sufficient to capture phonon-induced fingerprints in transient absorption.

In our experiment, optical A_g phonon modes are excited in thin film MoO_2 by an intense near-infrared femtosecond pulse and probed using a spectrally broad XUV pulse train spanning 30 to 60 eV (Fig. 1a). This energy range is dominated by transitions from the shallow core-level state Mo 4p to the conduction band states Mo 4d (Fig. 1b). The observed overall increase in change in optical density (ΔOD) is mainly driven by the local field effect, as previously demonstrated for molybdenum diselenide [3]. The dynamics of hot excited carriers around time zero are well captured by a convolution of the PDOS of the Mo 4p core-level state and that of the Mo 4d states (Fig. 1c). The absorption spectrum is modulated by oscillations attributed to the 6 THz and 11 THz phonon modes. The phonon amplitudes vary throughout the probed energy range, seen by taking the average over the energy indicated by the horizontal error bars and fitting it to the sum of two cosine at two frequencies multiplied by an exponential decay. The 11 THz mode induces absorption changes that peak around 38 eV, while the changes induced by the 6 THz mode are nearly constant throughout the observed energy range (Fig. 1d). These energy-dependent phononic fingerprints arise from phonon-induced modifications of Mo 4d states: the 6 THz mode directly alters them through Mo vibrations, while the 11 THz mode indirectly alters them through O displacements.

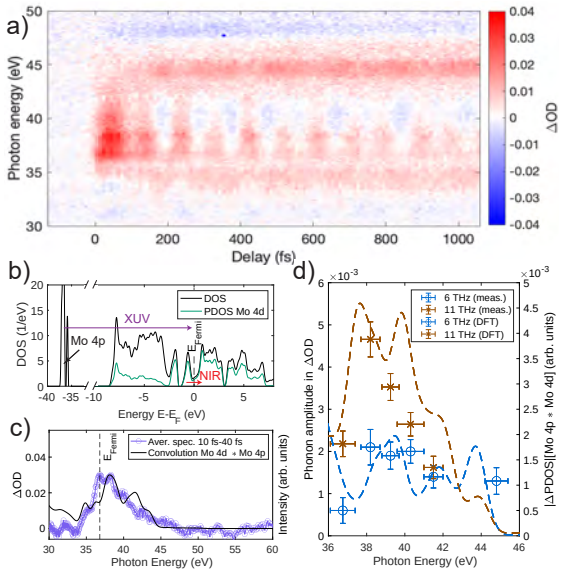


Figure 1. a) Transient absorption spectrogram. b) DOS and PDOS of Mo 4d states. c) Averaged spectrum vs. Mo 4p $*$ Mo 4d. d) Measured 6 THz and 11 THz phonon amplitudes vs. calculated phonon-induced absolute ΔPDOS (Mo 4p $*$ Mo 4d).

To conclude, we provide element-specific insights into phonon-driven modifications near E_F using transient absorption spectroscopy and DFT.

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Excitation and deexcitation dynamics in high harmonic generation of correlated electron systems

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Synopsis We investigate the high harmonic generation of the one-dimensional Hubbard model in the dissipative open quantum environment within a frame of the Lindblad master equation. In the limit of a small interatomic distance, we find the Dicke superradiance, the coherent stimulated emission triggered by the spontaneous emission, as well as the broadband emission mimicking the blackbody radiation in each harmonic multiple. Further, we find a strong destructive interference between the broadband emission and superradiance, which makes a scale down of the effective scattering time.

High harmonic generation (HHG) has been in the spotlight for the application to the ultrafast dynamics and the nonequilibrium phenomena as well as the coherent light conversion, and for the spectroscopic availability to explore the fundamental physics of materials. Recently, as an important feature of the electron-electron scattering, the broadband emission in addition to odd-order harmonics is observed in 2H-$NbSe_2$ [1]. The broadband emission is generated from hot carriers, which is found to follow the blackbody radiation at an effective temperature. Besides, an interplay between the electron scattering and the optical driving enhances the nonperturbative nature of high harmonics.

We utilize the Lindblad master equation [2] combined with one-dimensional Hubbard model and investigate the electron dynamics of HHG in dissipative open quantum systems. Especially at a weak or moderate correlation, as the Lindblad coupling constant γ regarded as the electron-environment scattering strength turns on, each harmonic becomes to be suppressed systematically due to a strong destructive interference between the broadband emission and the Dicke superradiance. The broadband emission is structureless emission due to hot carriers prepared by the electron-electron scattering. In particular, the Dicke superradiance [3] could be separated from the whole HHG spectra and found to drastically increase as the interatomic distance decreases. Furthermore, such a destructive interference is also found to make a scale down of the electron scattering time, eventually leading to the few-femtosecond time scale of T_2 which is consistent with the recent results using the quantum master equation [4].

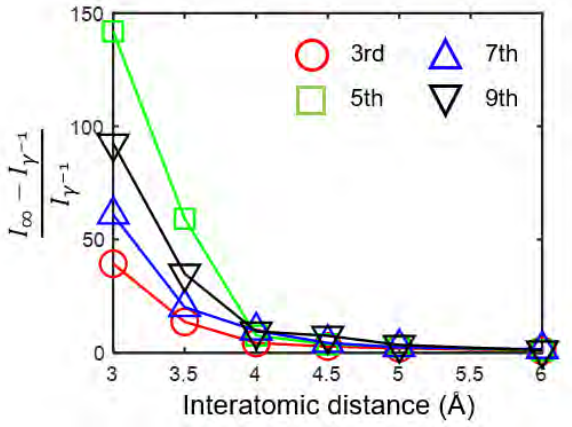


Figure 1. Normalized differences between harmonic strengths at $\gamma^{-1} = \infty$ and 100 fs for the third, fifth, seventh, and ninth order harmonics with respect to *d*.

Revealing the emergent properties of the Dicke superradiance offers insights into coherent and collective quantum effects induced through the many-body electron dynamics. The present work should be a milestone for the dissipative many-body electron dynamics of correlated electron systems, advancing the next generation quantum technologies.

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First-principles Simulation for Subcycle Reciprocal Image of Electron Density

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Synopsis We perform atomistic simulations of sub-cycle electron dynamics under an intense laser field. We find that the time-dependent evolution of the reciprocal-space image of the electron density reveals electronic hopping around chemical bonds. This finding demonstrates the potential of combining intense light with diffraction measurements to achieve attosecond temporal resolution and sub-nanometer spatial resolution for probing atomic-scale electronic motion.

The development of intense coherent light source technologies enables us to observe sub-cycle dynamics of electrons in materials on a time scale reaching attosecond resolution [1]. While all-optical measurements offer fine temporal resolution for electron motion, most of them lack sub-nanometer spatial resolution. Diffraction measurements with an intense light pump [2] are well-suited to complement all-optical methods for observing electronic motion at the atomic scale.

We perform first-principles simulations of sub-cycle reciprocal-space images of electron density based on time-dependent density functional theory for monolayer hBN and graphene under an intense electric field to obtain the real-space electron density. The reciprocal-space electron densities are then obtained by applying spatial Fourier transforms. The applied electric field is linearly polarized along y-axis, with a duration of 14 fs (FWHM), photon energy of 0.4 eV, and peak intensity of 1 TW/cm².

Figure 1(a) shows the ground state (GS) electron densities, represented as white isosurface renderings, while Fig. 1(d) displays their differences from the GS at the time of the field maximum, rendered as a color volume. The applied external field distorts the electron distribution, as indicated by the red (increased density) and blue (decreased density) regions. The reciprocal-space distribution of the monolayer hBN in Fig. 1(b) exhibits larger values compared to graphene in Fig. 1(e), reflecting the spatially localized electron density. The smoothest components of the reciprocal-space distribution for the monolayer BN exhibit an almost field-linear oscillation, as shown in Fig. 1(c). The antiphase oscillation between the (1,1) and (1,-1) components suggests that electrons hop between two B-N chemical bonds in response to the applied electric field. In contrast, such field-linear oscillations do not appear in Fig. 1(f) for graphene. The imbalance among the three components after the external pulse indicates a breaking of three-fold symmetry due to the strong excitation.

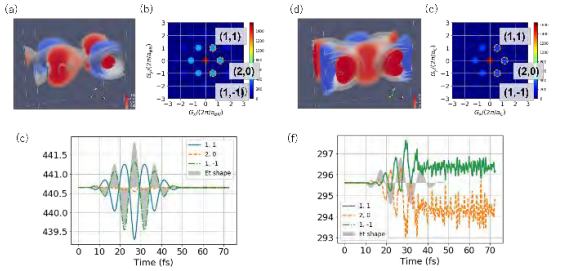


Figure 1. Real- and reciprocal-space electron densities (top) and time-evolution of the reciprocal components (bottom) for monolayer hBN (left) and graphene (right).

The simulation of atomistic-scale, sub-cycle electronic motion in reciprocal space is indispensable for uncovering the physics underlying the time-resolved diffraction images obtained in experiments.

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Novel quantum states of exciton-Floquet composites: electron-hole entanglement and information

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Synopsis. In this study, ultrafast coherent mixing of the exciton and Floquet states is investigated through a theoretical description of time-resolved and angle-resolved photoemission spectroscopy (tr-ARPES) in 2D semiconductors. Two branches of novel quantum states are found in the form of bosonic exciton-Floquet composites, which result from the exchange coupling due to the Coulomb interaction. Furthermore, tr-ARPES is directly employed for the density matrix element of the biparticle subsystem of photoelectron and hole, and electron-hole entanglement and information are further explored.

Recent advancements in pump-probe spectroscopy have facilitated the observation of the subfemtosecond motion of carriers and have promoted the investigation of time-resolved ultrafast light-matter interactions[1]. In particular, the dynamics of Floquet and exciton states have been fascinating phenomena in semiconducting optoelectronics. However, they have been separately dealt with in most previous studies [2,3], although a single resonant pumping can generate both states simultaneously. In this work, we investigate the ultrafast exchange between exciton and Floquet dynamics driven by weak optical pumping. In addition, from the viewpoint of the quantum processing of photoelectron and hole, the electron-hole entanglement and information in the excited electron-hole system are examined.

We construct a monolayer Haldane Hamiltonian, a prototypical two-band semiconductor of the 2D hexagonal lattice, extended to incorporate the Coulomb interaction between electron and hole, $V_{\mathrm{el-h}}$, and photoelectron Hamiltonian, H_{PES} as described in [4]. The wavefunction of the quantum system is obtained from the time-dependent Schrödinger equation, $i\frac{\partial}{\partial t}|\psi(t)\rangle = H|\psi(t)\rangle$, with a single exciton approximation. The theoretical tr-ARPES data is then obtained from the photoelectron components in $|\psi(t)\rangle$.

We extract the Floquet-like (FLL) and exciton-like (EXL) contributions from the tr-ARPES. As shown in Figure 1, EXL features arise, particularly at absolute detuning energies less than ~ 0.05 eV, and more interestingly, FLL features become discontinuous at a negative detuning energy (i.e., ~ -0.03 eV). Such a discontinuous change is an actual indication of strong mixing of exciton and Floquet states, and we figure out that the mixing is induced by $V_{\mathrm{el-h}}$. These FLL and EXL features suggest two branches of novel quantum quasiparticles, i.e., exciton-Floquet composites, which are created by the ultrafast mixing of exciton and Floquet states in the first-order excitation process. Furthermore, the time-derivative of the entanglement entropy evaluated from the tr-ARPES data rapidly increases around detuning$= -0.03$ eV (B in the right side panel of Figure 1), meaning faster decoherence of states, which occurs due to many-body interaction. It indicates that the Coulomb-induced mixing of exciton and Floquet state plays a key role in not only the ultrafast exciton and Floquet dynamics but also their information processing.

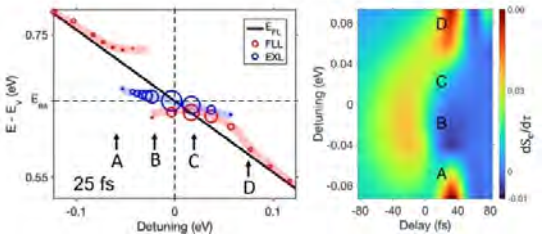


Figure 1. (Left) EXL and FLL of tr-ARPES at the pump-probe delay, 25 fs. (Right) the time derivative of the entanglement evaluated by the tr-ARPES.

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Diatomic Molecular Vibrations in Intense Laser Fields: An Analytical Perspective

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Synopsis We develop a mathematical framework to describe a Morse potential under strong harmonic excitation, incorporating permanent and field-induced dipole moments and their gradients into the interaction term of the molecular Hamiltonian. Using the Kramers-Henneberger frame, we derive analytical expressions for bond-length modifications and shifted energy eigenvalues. As an application, we analyze H_2 and LiH under near- or mid-IR laser fields (~10^{13} W/cm^2), gaining insights into their structural and energetic responses under these conditions.

Diatomic molecules subjected to intense laser pulses exhibit a range of nonlinear interactions, strongly dependent on the relationship between laser parameters and molecular properties. While theoretical models for molecular interactions with laser fields are well established, analytical approaches in the strong-field regime remain scarce and often rely on approximations, whereas numerical methods become increasingly costly for infrared (IR) wavelengths [1,2].

In this contribution, we introduce a general mathematical framework to describe a Morse potential under intense IR excitation. Our approach incorporates both permanent and field-induced dipole moments, along with their gradients, into the Hamiltonian, validated through density-functional theory calculations. Consequently, our model is applicable to both homonuclear and heteronuclear diatomic molecules, as well as certain alkali metal dimers. As a demonstration, we apply our method to H_2 and LiH, showing that an IR laser field in the intensity range significantly shifts vibrational levels (see Figure 1) and alters the equilibrium internuclear distance.

Our results reveal that the dipole gradient and polarizability gradient play a crucial role in these effects. We derive exact analytical expressions for vibrational energy levels and bond-length modifications within the Kramers-Henneberger frame, valid across a broad wavelength range [3].

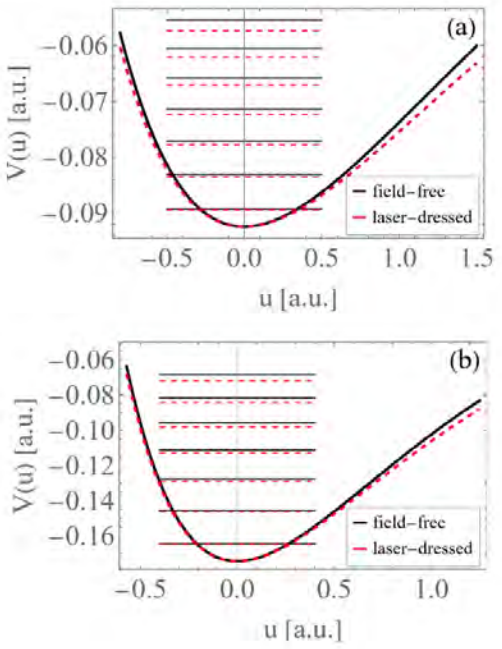


Figure 1. Field-free (black solid curve) and laser-dressed (red dashed curve) Morse potentials of LiH (a) and H_2 (b). The horizontal lines indicate the first seven energy levels for each potential, following the same line styles. The applied laser parameters are $I_0 = 2.8 \cdot 10^{13}$ W/cm^2 and $\lambda = 1500$ nm.

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Probing Band Structure Properties through HHG in MgO: The Role of Ellipticity and Bloch Confinement

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Synopsis Bloch oscillations and Bloch confinement are some of the most intriguing and counter-intuitive phenomena of electronic band theory. Solid-state HHG can accurately probe such phenomena due to its ultrafast timescale and high spatial sensitivity. We experimentally resolve the onset of Bloch confinement via HHG in MgO. Controlling the field's ellipticity manipulates the 2D electron trajectories, allowing us to probe their localization and the interplay between the laser field and the periodic potential.

Strong-field-driven electric currents in condensed-matter systems are pushing the boundaries of attosecond science. While strong-field dynamics in gases have been extensively studied over the past two decades, their role in solids remains an emerging frontier. In gas phase, electrons follow the electric field, which dictates the fundamental properties of the interaction. In solids, however, the periodic crystalline structure fundamentally alters their behavior [1]. Under sufficiently strong laser fields, electrons in solids undergo Bloch oscillations—a localized motion where they oscillate back and forth across the crystal lattice due to the electron's momentum reversing at the Brillouin zone (BZ) edges [2].

Inter-band High harmonic generation (HHG) in solids serves as an extremely accurate probe of such localization owing to the Angstrom resolution of the electron-hole recombination process. Manipulating the temporal-structural properties of this mechanism allows us to study the role of the band structure and identify the localization of the electron-hole wavefunction.

Our experimental scheme is based on HHG spectroscopy in MgO with an elliptically polarized field, as previously done in [3]. In gas phase systems, increasing the fundamental field's ellipticity leads to a drastic decrease in the harmonic signal, due to the lateral displacement of the electron from its parent ion [4]. Here, we utilize this tool to probe the localization of Bloch electrons. Our experimental results reveal that when the crystal is rotated away from its main symmetry axis, increasing the ellipticity leads to a dramatic increase of the signal. We attribute this increase to Bloch confinement—along this axis the drift between the electron and hole is compensated by the lateral drift induced by the band dispersion, leading to the confinement of the electron-hole wavefunction. As we investigate the angular dependence of this confinement for various harmonic orders (Fig. 1), we directly probe the band dispersion and can resolve anomalous features of the band structure.

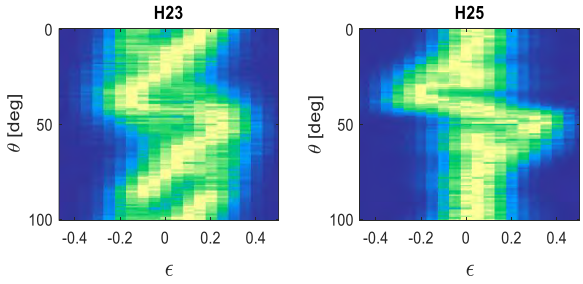


Figure 1. Angular mapping of the ellipticity dependence of harmonics 23 and 25 (λ=2.1μm) in MgO. The main symmetry axis is aligned with **0**°.

Our findings provide new insights into the role of band structure anisotropy in wave function localization and HHG in solids. By leveraging ellipticity-dependent HHG as a sensitive probe, we offer a novel approach for mapping electronic band dispersion advancing the understanding of ultrafast light-matter interactions.

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Beyond Electric-Dipole Treatment of Light-Matter Interactions in Materials: Nondipole Harmonic Generation in Bulk Si

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Synopsis We derive an accurate beyond electric-dipole light-matter formalism that does not impede on Bloch's theorem and thus allows for describing emerging strong-field light-driven scientific applications in condensed matter systems. Through first-principles microscopic simulations of harmonic generation in bulk Si, our approach reveals nondipolar spectroscopic signatures arising from induced transverse currents. Nondipole signatures are perturbative even-ordered harmonics and the onset of nonperturbative higher-order harmonics.

Our understanding of foundational light-matter interactions, spectroscopic principles, and photonic processes relies almost exclusively on the dipole approximation, which neglects the spatial dependency of electromagnetic fields. It breaks down both in the short-wavelength limit, as the wavelength subceeds the atomic length scale, and in the long-wavelength limit, when an intense field induces a non-negligible beyond-dipole $v \times B$ transverse Lorentz force interaction.

With emerging lightwave electronic and quantum material science applications relying on orbital angular momentum vortex beams, plasmonic nanostructures, or angle-resolved photoelectron spectroscopy, the need for beyond-electric dipole descriptions is imminent. However, such progress is hindered in condensed matter systems as full treatment of the spatial dependencies of the fields impedes on the periodic translational symmetry of Bloch's theorem.

We provide a first-principles formalism that accurately accounts for leading-order nondipole light-matter interactions in the strong-field regime, without violating the periodic symmetry considerations of Bloch's theorem [1]. The formalism is applied to study nondipolar spectroscopic features arising from harmonic generation in crystalline Si. A nondipole transverse current is revealed to generate perturbative scaling even-ordered nondipole harmonics, alongside higher-order nonperturbative nondipole harmonics when approaching the laser damage threshold.

Figure 1 depicts harmonic spectra from Si driven with three different laser wavelengths. Nondipole even-ordered harmonics arise in the novel nondipole strong-field approximation formalism, that are of above-bandgap interband nature in (a), and below-bandgap intraband nature in (b-c) scaling with $S(2\omega)/S(\omega) \propto I\lambda^2$. At increasing field intensity, a perturbative fourth and non-perturbative sixth harmonic emerge.

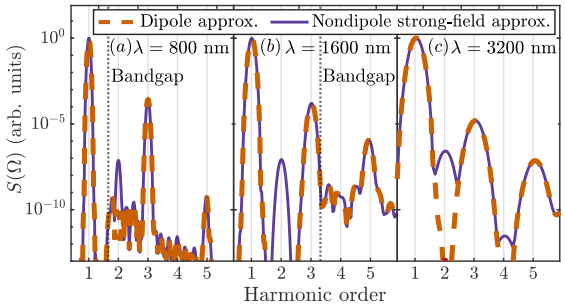


Figure 1. (a-c) HHG spectra from Si driven by a 100 fs $\sin^2$ pulse of 10^{10} W/cm^2 for 800, 1600, and 3200 nm, respectively. Simulations are done within both the electric dipole and the nondipole strong-field approximation.

The formalism, originally derived for atoms [2], allows for extending any electric dipole approximated theory to include nondipole interactions by modifying the electromagnetic vector potential $A(t)$ with an associated nondipole correction $A^{\mathrm{ND}}(t)$ that can be obtained at negligible computational cost. An implementation is already available in the open-source Octopus code [3].

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Attosecond-resolved Non-dipole Photoionization Dynamics

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Synopsis We measured the time-dependent non-dipole effects of photoionization using a self-referenced attosecond photoelectron interferometry with 3D electron momentum resolution. We show that the magnetic component of the IR field leads to electron subcycle motion in the light propagation direction in the 15 pm range, whereas the electric quadrupole transition leads to an oscillating forward-backward asymmetry of the photoelectron in sideband with 15±10 as delay compared to the dominating electric dipole transition.

Non-dipole effects in photoionization are relatively small from the infrared to the soft X-ray domains and are usually neglected in studies on light-matter interactions. All non-dipole observations reported so far were limited to single-pulse, single-color measurements. Here [1] we advance attosecond time-resolved spectroscopy into the non-dipole interaction regime. An XUV attosecond pulse train is superposed with a moderately strong (6×10^{13} W cm^{-2}) IR field. Expanding the Hamiltonian coupled to the IR field up to $1/c$, we have the M_1 (magnetic dipole) and the E_2 (electric quadrupole) terms that are contributing besides the dominating electric dipole term, leading to the forward-backward asymmetry of the electron momentum distribution and the sideband electron yield, respectively. Both effects depend on the XUV-IR delay. Experimentally, using a self-referenced attosecond photoelectron interferometry on helium atoms with 3D electron momentum resolution, we resolve the electron subcycle motion along the light propagation direction in the 15 pm range driven by the magnetic component of a near-infrared laser field. Furthermore, we measure a time delay of 15±10 as between the electric-dipole and electric-quadrupole transitions by resolving the asymmetry of the photoelectron forward-backward yields with attosecond resolution. These fundamental findings are supported by ab initio calculations based on the non-dipole time-dependent Schrödinger equation.

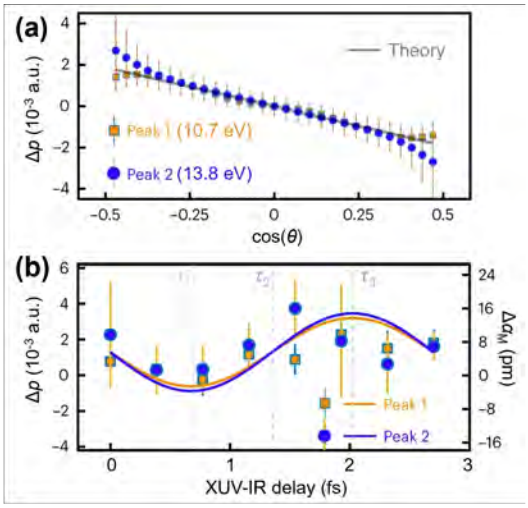


Figure 1. (a) The forward–backward momentum shift induced by the magnetic field of the IR pulses, referenced to the XUV-only case. The shifts are extracted from the time-delay integrated spectra. (b) The forward–backward ($\theta = 70°/110°$) momentum shift as a function of the time delay τ between the XUV and IR pulses. The right y-axis indicates the displacement $\Delta\alpha_M(\tau)$. The error bars are determined by the 95% confidence level of the fitting.

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Rabi oscillations, photo-emission and entanglement: A time-resolved picture

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Synopsis We investigate concerted Rabi and photo-emission dynamics through an original pulse sequence with a focus on the photo-electron/ion entanglement mechanism. We illustrate this with simulations on a model atom of helium with two active electrons. We assess its validity by taking the limit of our pulse sequence when it converges towards a single pulse. Our work establishes, both analytically and through simulations, the link between Rabi dynamics, photo-emission and entanglement in the photo-electron spectrum.

The development of free electron lasers gives access to ultra-short, intense extreme ultraviolet light pulses which allow the experimental study of non-linear effects in this energy range. For instance, the photo-ionization of an atom with a photon energy simultaneously driving Rabi oscillations in the ion manifests as a doublet in the photo-electron spectrum due to the dressing of the ionic states [1].

An interesting feature of such a situation is the generation of entanglement between the ionic core and the photo-electron [2]. There, the light pulse plays two roles, i.e., ionizing the atom and dressing the ion. This makes the investigation of the entanglement build-up mechanism difficult to address, in particular in terms of time and length scales.

In this work, we address this issue by considering an original pulse configuration in simulations on a model helium atom. We use two different photon energies to unambiguously discriminate the Rabi dynamics from the photo-emission process. Also, we consider, for the ionizing field, a sequence of two short pulses instead of a single longer one. By controlling the delay τ_{XUV} between the ionizing pulses, this allows uncovering the mechanism behind the generation of entanglement in the time domain.

Moreover, since this scheme creates electron wave-packets with the same energy but at different times, τ_{XUV}-dependent interferences are observable in the photo-electron spectrum. We established, both numerically and analytically, a link between the contrast C of these interferences and the purity P of the reduced density matrix of the ion as

$$C^2 = 2P - 1, \tag{1}$$

see Fig. 1. Thus, our original scheme allows a direct measurement of the purity in the spectra.

Finally, we show that the contrast directly corresponds to the overlap between the states populated by each of the two short pulses. Hence, it allows performing a quantum tomography of the Rabi states.

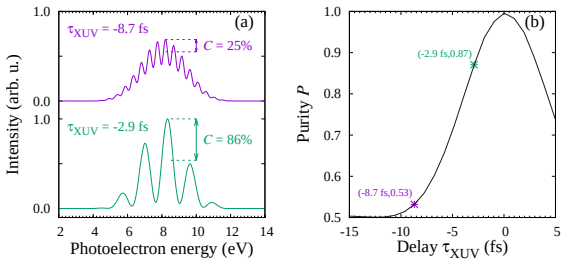


Figure 1. (a) Photo-electron spectra for two different τ_{XUV} delays. (b) Final purity as a function of τ_{XUV}.

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Strong-field control over valley polarization and topology

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Synopsis We demonstrate valley polarization control of inversion symmetric bulk materials by a trefoil polarized optical pump. The induced anisotropy is detected via polarimetry of a background free second harmonic signal and probes the optically controlled topological phase transition.

By utilizing electronic degrees of freedom, combined with ultrafast switching speeds, light-wave electronic devices provide a platform for encoding states for information processing while exploiting the classical and quantum properties of photons [1]. Leveraging the bandgap extrema of two-dimensional materials, resonant excitation with circular polarized light creates an anisotropic carrier population between neighboring "valleys" which can be switched upon subsequent excitation. However, to truly enable control at sub-cycle speeds a novel approach to valleytronics has been proposed in Ref. [2]. By combining counter rotating circular polarized optical fields at fundamental and second harmonic frequencies a three-fold symmetric so called "trefoil" shaped polarization state is created. Originating in the temporal and spatial inversion symmetry breaking by the field & crystal a complex phase is induced during second neighbor hopping, akin to the Haldane model. The consequence is a reduction of the bandgap either at the K or K' valley depending on the trefoil angle, realizing strong-field valley polarization control in h-BN monolayers [3].

Using a toy model based on gapped graphene we confirm the universality of this approach and extend the method to multilayer (bulk) 2H-MoS_2 where resonant valley control fails, due to the inversion symmetry of the sample. In a non-collinear geometry, the background-free second harmonic (SH) generation probes the valley polarization from the asymmetric populations between K and K', induced by the trefoil [4]. Polarimetry in Fig.1 (b) confirms the probe SH polarization state parallel with the anomalous current along Γ-K-K'. Rotation of the trefoil in Fig.1 (d) selects the valleys, as seen by the maximized probe signal, which even distinguishes K or K' alignment.

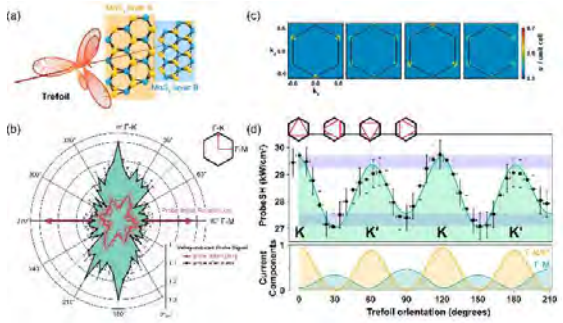


Figure 1. (a) Illustration of bi-circular trefoil field incident on 2H-MoS2 with layer B rotated by 180°. (b) Polarimetry of probe second harmonic generation before & after pump-probe overlap. (c) Calculated carrier distribution in k-space summed over all conduction bands for trefoil orientations indicated by insets. (d) Measured probe SH as function of trefoil rotation with simulation as green line. Bottom: decomposition into polarization components along Γ-M and Γ-K, K'.

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Reduction of Chromatic Aberration in High Order Harmonic XUV Beams using a Temporal Flat Top

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Synopsis We synthesised a temporal 'flat top' laser pulse and used it to control HHG. The temporal flat top profile was obtained using a collinear setup comprising birefringence optical element stacking. We use this pulse to generate high harmonics in Argon gas, and demonstrate that the tailoring of the laser driver enables a fine spectral tuning of the high harmonics over several eV. Using this temporal flat top, we measured a reduction of the chromatic aberration of the high harmonics employing the SWORD technique.

Attosecond pump-probe experiments require high flux attosecond sources with a high beam quality. This is paramount to many applications such as high-resolution imaging [1] and attosecond metrology [2]. However, the HHG process produces a chromaticity between the high harmonic wavefronts due to microscopic and macroscopic phases acquired by the XUV field. This leads to an inhomogeneity in the wavefronts and chromatic foci when the high harmonic beam is focused on target. This is due to these phase terms being proportional to the laser intensity profile. This induces a harmonic wavefront dependent upon $\alpha_q^{(j)} \nabla I(r, z, t)$. The high harmonic wavefronts are therefore harmonic order q dependent and high harmonic quantum path (j) dependent [3]. They can be controlled by tailoring the spatio-temporal intensity gradient of the laser pulse to a constant intensity profile such as with flat tops.

To achieve a temporal flat top pulse and minimise the $\frac{\partial I}{\partial t}$ phase term, we used a collinear set up made of a pair of birefringent wedges and a birefringent plate. We successfully characterised the temporal flat top profile using Fourier Transform Spectral Interferometry (FTSI) [5]. We achieved many different temporal syntheses including the flat top. We compared the high harmonic spectra for all temporal profiles and demonstrated control over the high harmonic photon energy and wavefront, characterised using the SWORD method [4].

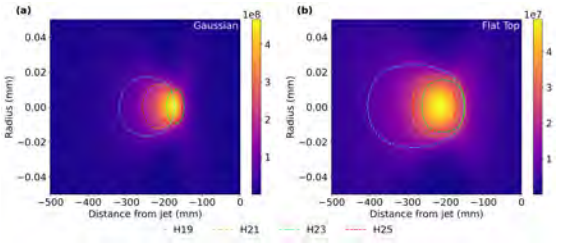


Figure 1. Comparison of the H19-25 source point overlap, reconstructed by SWORD, using (a) the Gaussian intensity profile and (b) the temporal flat top.

As shown in Fig.1, we observe a clear improvement of the overlap of the H19-25 source point reconstruction using the temporal flat top as compared to the Gaussian envelope. Our measurement proves that a temporal flat top can reduce the chromatic aberration in the produced XUV radiation, and reduce the spatio-temporal couplings in attosecond pulses. These results show a potential future optimisation of the wavefront of the high harmonic source.

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Progress towards revealing attosecond dynamics in strongly correlated systems

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Synopsis Correlated insulators, such as Mott and charge transfer insulators, exhibit insulating behaviour due to strong electron-electron interactions. We investigate the sub-cycle electron-electron dynamics in NiO, a charge transfer insulator, using HHG spectroscopy. We present preliminary results on sample fabrication, HHG measurements from NiO nano-films, and concepts for manipulating attosecond electron dynamics using intense, tunable mid-IR pulses.

Correlated insulators are a class of materials that should conduct electricity under conventional band theories. However, due to electron–electron interactions, they are insulators. In such systems, the sub-cycle dynamics are dictated by electron-electron correlations [1]. Solid state HHG spectroscopy in the mid-infrared (MIR) regime can manipulate the attosecond time scale dynamics of electrons, and therefore the fundamental electronic properties of the system [2]. We aim to apply HHG as a unique probe to resolve and control the sub-cycle evolution of electron-electron interactions in correlated materials.

Nickel oxide (NiO) is a typical charge transfer insulator that has been the subject of recent studies [3]. The sub-cycle modifications in the electron-electron interactions can be resolved via two-colour HHG spectroscopy. By adding a weak second harmonic field we intend to "shape" the optical cycle and thereby manipulate the electron dynamics on attosecond time scales.

A key challenge is producing high-optical quality crystalline nanofilms for our studies. We produced crystalline (<111> orientation) NiO films on Sapphire via electron beam evaporation followed by annealing at 600 K. Figure 1 shows the measured HHG spectrum from a 100 nm NiO layer, using 3.9 um pulses from a tunable, 50 kHz OPCPA system. Performing two color HHG spectroscopy, so far applied to resolve sub-cycle dynamics in a large variety of systems [4], will enable exploration of the rich electronic dynamics in correlated materials.

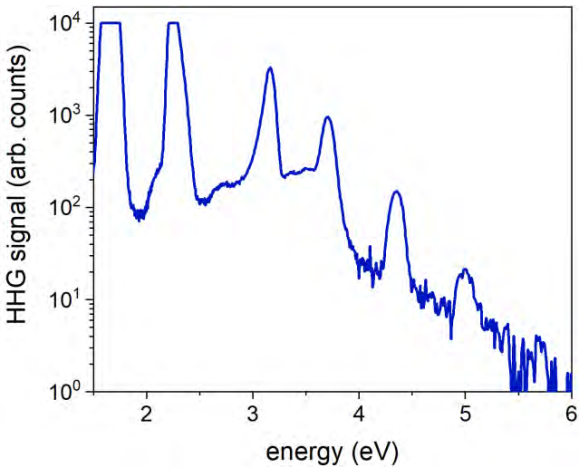


Figure 1. HHG spectrum from a crystalline 100nm NiO layer on Sapphire substrate using 3.9 um, 60 fs, 4 uJ pulses at 50 kHz, focused to a peak intensity of ~ 10^{12} W/cm^2.We observe HHG peaks up to 6 eV, well above the 4.3 eV bandgap of the material.

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Electron Trajectory Analysis through Carrier-Envelope Phase Effects in High Harmonic Generation from ZnO

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Synopsis We observe spectral interference between the short and the long trajectories produced in high-harmonic generation (HHG) from ZnO. This spectral interference is directly affected by the carrier-envelope phase (CEP) of the driving 3 µm, 66 fs pulse (∼ 7 cycles). This work shows that CEP, even with multicycle pulses, can be a tool to distinguish the different electron trajectories.

The study of HHG in solids provides insight into the fundamental physics of light-matter interactions and holds promise for various applications, e.g. the development of compact, table-top, controllable XUV and soft X-ray sources. Varying the CEP of the driving field allows for temporal sub-cycle electron resolution and control and CEP effects have already been studied with a long pulse, i.e. not a typical few-cycle pulse [1].

We collect high harmonics generated from a ZnO sample (300 µm thick, c-cut) with a 3 µm laser beam with pulse duration of 66 fs. The intensity is controlled with a wire-grid polariser and the CEP is varied with a f-to-2f interferometer. The spectrum of the 11th harmonic is visible on Figure 1. As the intensity increases, the harmonic spectrum splits and broadens. We observed the same behaviour in reflection. Moreover, we do not see any corresponding spectral shift of the fundamental nor of the harmonics, rendering unlikely propagation to be the cause of this splitting and broadening. Similar behaviour has been observed by Kim *et al.* [3] in sapphire and attributed to spectral interference of short and long electron trajectories.

We plot on Figure 1 the normalised difference between the CEP-dependent harmonic spectrum, and the CEP-averaged spectrum as outlined in [2]. The peak and the dips of the harmonic spectrum correspond to different CEP variations despite using a long pulse. We demonstrate that the CEP dependence encodes information on the long and short electron trajectories in high-order harmonics.

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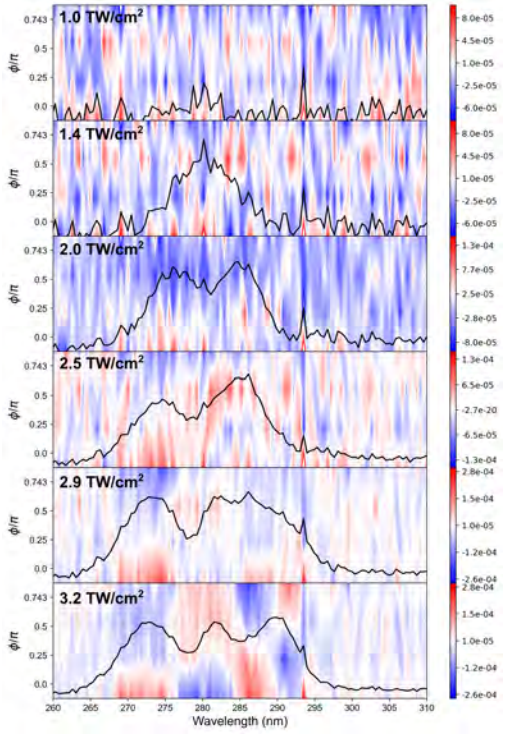


Figure 1. Harmonic 11 generated from ZnO for different intensities. The colormap corresponds to the normalised difference of the harmonic spectrum. The black curve is the spectrum taken for CEP value 0.

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Phase transitions induced by quantum vacuum

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Synopsis Quantum light emission by strongly driven matter opens a yet unexplored route toward generating non-classical photonic states. However, a major obstacle in its theoretical treatment is that the light states involved necessitate a potentially infinite Hilbert space. This constrains the analysis to weak feedback between generated light and matter. We employ novel tools to show how strong light-matter feedback leads to concurrent formation of a correlated state of matter and a quantum state of light, the light heralding the quantum phase transition in the matter induced by vacuum fluctuations.

Quantum photonic states generated on an attosecond scale are attracting a heightened attention. In addition to lending themselves well to storing and manipulating quantum information, they turn out [1, 2] to herald quantum correlations in the emitter, imprinting them on the generated photonic states. Yet, these states remain an almost uncharted territory due to the complexity entailed by both observing them in experiment and accurately predicting them in theory. The latter has to do with the sheer dimension of the Hilbert space involved. Indeed, a Hilbert space necessary to faithfully describe a driven system which emits photons into a quantum electromagnetic vacuum is, strictly speaking, infinite.

One way to get around this limitation is to constrain our description to the perturbative limit, as in [2, 3]. However, it is insufficient for describing situations involving multiple re-emissions and re-absorption of photons. We adopt a novel non-perturbative approach which allows us to contain the growth of the Hilbert space for the quantum light modes, putting ourselves in a position to simulate the behaviour of driven systems.

As discussed in [3], the non-classical properties of the final state are most pronounced when the emission occurs into a mode resonant with one of the emitter's dipole transitions. We consider a system consisting of several (1-10) indistinguishable qubits with no explicit coupling between them, driven classically by a perturbatively weak light field, and coupled strongly to a common resonant narrow-band waveguide. This combination ensures that after a photon is emitted by one qubit, it can be reabsorbed by another while still retaining entanglement with the original one, giving rise to quantum correlations between the two.

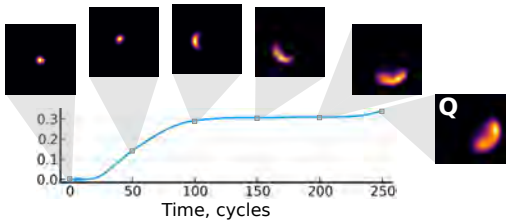


Figure 1. Average values of the correlator $\hat{C} = \mathbf{S}_i\mathbf{S}_j - \mathbf{S}_i^2$ during the laser pulse, normalised to its expectation value for the highly non-classical Dicke state. The insets show the Husimi functions of the central mode at the respective time moments.

The correlated qubits undergo superradiance-like collective transitions, prompting the spontaneous formation of a correlated light-matter state. Fig. 1 shows how the correlator for the many-body state grows over time and how this, in turn, gives rise to highly non-classical photonic emissions, manifesting as a nontrivial Husimi function corresponding to a cat-like state.

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Time resolved Coupled Electron-Nuclear Dynamics of Methane Cation upon Photoionization Using an XUV Attopulse

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Synopsis Ultrashort pulses enable coherent excitation of multiple electronic states, creating a superposition of interfering electronic wave functions. By tuning pulse parameters, electronic density localization between nuclei can be controlled, opening new possibilities for steering bond rearrangement and chemical reactivity once the nuclear dynamics kicks in. In this study, we investigate the quantum and semiclassical dynamics of Jahn-Teller-driven relaxation and dissociation of methane ionized by an XUV attosecond pulse, revealing pathways for ultrafast structural rearrangement and fragmentation.

Ionizing a neutral molecule with an XUV attopulse forms a highly excited cation in a coherent superposition of electronic states that undergoes ultrafast relaxation via different nuclear coordinates. Investigating the dynamics of such non-equilibrium coupled electron-nuclear dynamics is crucial for advancing our understanding of ultrafast dynamics which is a central challenge of attochemistry. Here, we report a quantum dynamical study on the three lowest states of CH_4^+/CD_4^+ cations suddenly ionized using an XUV attopulse.

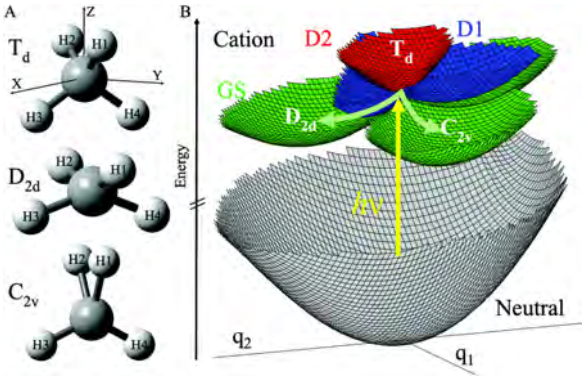


Figure 1. Geometries of the cation and PES of three lowest electronic states of cation with the GS of the neutral molecule (Ref. [1]) .

Two nuclear coordinates are included in the quantum dynamics for describing the initial state resulting from photoionization process and the subsequent structural rearrangement. [1] The ground state of the cation, D0, is unstable in T_d geometry that corresponds to the equilibrium geometry of the neutral molecule. The sudden ionization of CH_4 or CD_4 leads to a superposition of the three electronic states of the cation that are degenerate in the T_d geometry. An ultrafast, non-adiabatic dynamics triggered by the Jahn-Teller effect therefore sets in right after the ionization. [1-5] We report on two relaxation channels on D0. First, the cation structurally rearranges to lower symmetry geometries, first to D_{2d} and then to C_{2v}, the cation ground state equilibrium geometry. Second, a relaxation channel leading to the dissociation $CH_4^+ \rightarrow CH_3^+ + H$ ($CD_4^+ \rightarrow CD_3^+ + D$) is opened on D0 for sufficiently high energies of the XUV ionization pulse. Our multistate quantum dynamics investigates how isotopic substitution influences both these relaxation processes. We complemented our reduced dimension quantum dynamical (QD) study with semi-classical non adiabatic Surface Hopping (SH) dynamics which describes the nuclear motion in full dimensionality on the three lowest states of the cation. In the SH calculations, the cation undergoes rapid relaxation to D0 resulting in an ultrafast dissociation in agreement with our QD simulations.

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Loss of contrast in attosecond temporal double slit interferometry from atomic photoionization

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Synopsis The loss of contrast in the photoelectron momentum distribution (PMD) in attosecond temporal double-slit interferometry from atomic photoionization is theoretically investigated. When a weak, linearly polarized few-cycle infrared (IR) laser pulse is introduced, the contrast diminishes predominantly along the polarization of the IR pulse. These results highlights the potential loss of contrast in attosecond streaking experiments when shaped electron vortices are employed.

Attosecond interferometry, through the Ramsey interference of matter wave packets, provides valuable insights into the quantum dynamics of atomic and molecular systems, making it an essential tool in ultrafast science. Recently, vortex-shaped electron wave packets [1-4], arising from the interference of time-delayed electron wave packets with different magnetic quantum numbers produced by few-cycle ionizing pulses, have garnered significant attention due to the highly sensitive properties of free electron vortices in light-matter interactions [5]. These shaped electron vortices encode phase information in their wave packets, making them promising candidates for attosecond streaking [6] experiments, where they can serve as highly sensitive probes for uncovering timing information. However, the interaction of the infrared field used in attosecond streaking with the ionizing pulses that generate electron vortices requires thorough investigation.

In this work, we employ the R-Matrix with Time Dependence (RMT) method [7] to theoretically demonstrate the loss of contrast in attosecond temporal double-slit interferometry from atomic photoionization. Specifically, we examine how contrast diminishes in the momentum distribution of ejected electrons produced via single- and multiphoton photoionization by two time-delayed, oppositely circularly polarized attosecond pulses. By introducing a weak, linearly polarized few-cycle IR laser pulse—similar to those used in attosecond streaking experiments—we show that the loss of contrast occurs predominantly along the polarization axis of the infrared laser. Figure 1 shows the PMD results of photoelectrons ionized from ^{1}S ground state of Helium atom.

This study provides a proof of concept for the potential loss of contrast in attosecond streaking experiments when shaped electron vortices are employed. Our findings underscore the importance of understanding the interplay between ionizing pulses and infrared fields in preserving contrast in ultrafast experiments.

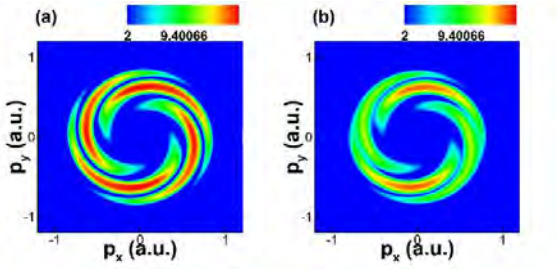


Figure 1. PMDs for electrons ionized by two counter-rotating circularly polarized pulses (Right-Left Circularly Polarized) with 15 eV photon energy delayed in time for 976 as (a) without the presence of IR field and (b) with the presence of a single cycle IR field with photon energy 1.55 eV polarized in the x-direction. Four-arm spiral pattern is obtained in accordance to what has been observed in [3].

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Ultrafast charge migration in solvated molecules

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Synopsis We investigate the effect of a liquid environment on attosecond charge migration (CM) in fluoroaniline. By comparing these results with gas-phase fluoroaniline, we analyze how solvation influences CM behavior, providing insights into solvent-induced modifications in ultrafast CM.

Charge migration (CM) in molecular systems has been extensively studied in the gas phase, yet its behavior in the condensed phase remains largely unexplored. Understanding the role of solvation in attosecond CM is crucial—both fundamentally, in identifying differences between CM in gas-phase and liquid phase, and practically, in determining whether CM can be effectively measured in solution and how it varies with different solvents.

We created a liquid environment by placing the molecule fluoroaniline in a methanol solution through Molecular Dynamics simulation [1]. Using time-dependent density functional theory implemented in a developmental version of NWChem [2], we simulate core-hole ionized CM in fluoroaniline, both in its gas phase and solvated in methanol (CHOH), to examine solvation effects. In both cases, initiating a hole in the fluorine (F 1s) sub-shell results in periodic valence hole oscillations between the phenyl ring and amine group, with a time period of 800-850 attoseconds, highlighting key similarities. However, in solution, we observe a finite dephasing time, in contrast to the gas phase where no dephasing is observed, as illustrated in Fig. 1.

To further investigate solvation effects, we replaced methanol with furan, a less polar solvent that lacks hydrogen bonding interactions. This change led to a longer dephasing time in CM, as illustrated by the blue curve in Fig. 1. Overall, we see that while the CM period is almost comparable across all cases (800-850 as), dephasing times differ, faster in methanol than in furan.

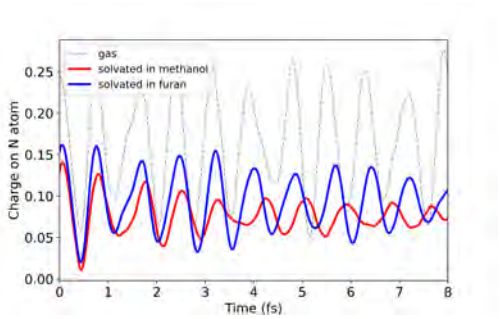


Figure 1. Charge migration in fluoroaniline, shown as the time-dependent charge on the N atom following 1s core-hole ionization: Gas-phase (gray-dotted), solvated in methanol (red), and solvated in furan (blue) .

These findings confirm that solute-solvent interactions play an important role in modifying CM behavior, as evidenced by the solvent-dependent dephasing. This demonstrates that CM can be tailored by choosing solvents with different chemical properties, paving the way for further studies on controlling ultrafast CM in complex environments.

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Update on the Modular Multiscale Approach (MMA) to simulate high harmonic generation in gases

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Synopsis We present an update on the development of the MMA simulation tool. This code simulates high harmonics generation (HHG) in gases for linearly polarised driver in cylindrically symmetric geometry. A pipeline of modules is implemented, comprising the infrared driver's propagation, a TDSE microscopic solver, and the diffraction integral for the XUV radiation. The code supports arbitrary shapes of the input pulses and media with density modulation (thus HHG in gas cells and jets). The code comes along with interactive tutorials and is deployable on various hardware and compiler architectures.

We are developing an open-source numerical model [1], which describes the multiscale problem of HHG in gas phase in its full complexity: macroscopic scale for field description and microscopic scale for laser/matter interaction. The development is motivated and accompanied by laboratory applications.

The nature of the problem allows us to pipeline the subtasks: (a) the propagation of the driving laser (unidirectional pulse-propagation), (b) the Time-dependent Schrödinger equation (TDSE) for the microscopic response, and (c) the diffraction-integral approach to aggregate the macroscopic XUV field from the microscopic emitters. Because the numerical computations are expensive, all modules use MPI or multithreading parallelisation. The main computational routines are written in low-level c/Fortran, complemented by Pythonic high-level for pre- and post-processing. The computational interface is the HDF5 data structure, which reduces data-handling for visualisations and post-processing. For example, the driver's phase and intensity profile might be masked by phenomenological SFA dipoles to quickly estimate the XUV signal. Complementary to the main pipeline, all the modules are available as stand-alone applications. Namely, the pulse propagation can be used to study, e.g., laser filamentation, the TDSE is available as a dynamic library for Python, and the computation of the XUV diffraction integral comes with its own interface. The recent developments allow multiplatform deployability (intel/GNU/AppleClang) on various supercomputers. Pythonic interfaces make the computational routines easy to use and present the data in an understandable form. The code is accompanied by jupyter tutorials to make it more user-friendly.

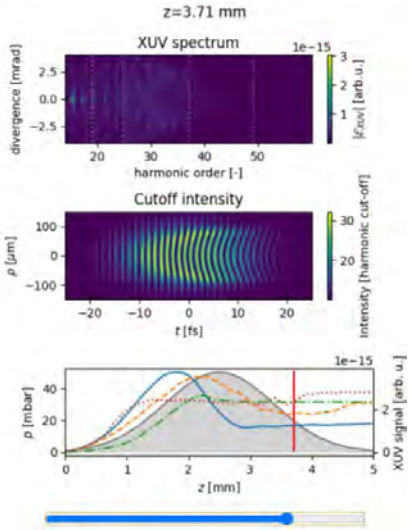


Figure 1. An example of the output for HHG in a gas jet by an 800-nm, 15-fs (1/e), laser pulse with 3.6×10^{16} W/cm^2 peak intensity. (upper panel): spatially resolved XUV spectrum containing HHG interference structures [2]; (middle panel) the profile of the driving pulse; (bottom panel): pressure profile of the jet, scaled cumulative XUV signals for H19(-), H25(--) H37(-·), H49(··).

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Proposal for High-Energy Cutoff Extension of Optical Harmonics of Solid Materials Using the Example of a One-Dimensional ZnO Crystal

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Synopsis We propose a novel approach based on the subcycle injection of carriers to extend the high-energy cutoff in solid-state high harmonics and verify through coupled solution of Maxwell propagation equation and singlecell Semiconductor Bloch equations.

Beyond the tremendous exploration of high harmonic generation (HHG) in gases during the last several decades, HHG in solid phases has been realized recently in experiment, which triggers rapidly-emerging activities as complementary approaches to attosecond pulse metrology. However, to realize the generation of attosecond pulses in solids and etc., the spectra of solid HHG are far from ideal and the characteristics should be optimized, such as the cut-offs and polarizations, in further researches.

A novel approach based on the sub-cycle injection of carriers by EUV pulse shows that the high-energy cut-off of solid high harmonic generation is extended to the secondary plateau employing infrared+ultraviolet (IR+UV) pulses within the framework of singlecell semiconductor Bloch equations (SC-SBE). The underlying mechanism can be schemed in our extended three-step model. Furthermore, considering that copropagating harmonics in solids with finite thickness (~100 μm) can serve as the injecting UV pulse as in the singlecell scheme, propagation induced self-injection process can be achieved in real laser-solids interaction and theoretically verified using coupled simulation of Maxwell equation and semiconductor Bloch equations (MP-SBE). Fig. 1(a) illustrates the singlecell (SC)， reflected and transmitted harmonic spectra, respective-ly. As can be seen, the reflected harmonics are in close analogy to that of the SC-SBE simulation, implying that propagation effect barely influences the characteristics of reflected harmonics. However, transmitted spectrum shows: (1) nonlinear gain in the first plateau; (2) extended high-energy cutoff in the second plateau. The nonlinear gain can be explained be the competition between absorption and phase-matching during propagation, while the ex-tension of cut-off origins from self-injection of carrier driven by harmonics generated in the front region, as shown in Fig. 1(b).

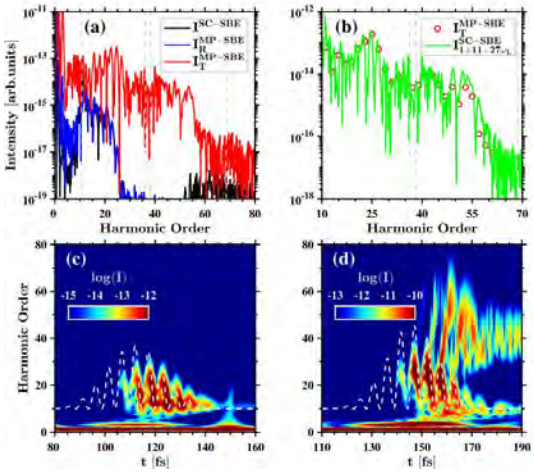


Figure 1. (a) Harmonic spectra of single-cell SBE (black), reflected (blue) and transmitted (red) field. (b) Comparison between transmitted wave and IR+UV scheme from SC-SBE simulation. Time-frequency analysis of harmonic emission from SC-SBE and MP-SBE.

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Primary Events of the MLCT Dynamics of Photoexcited Iron(II)-Tris(2,2'-Bipyridine) in Solution Probed by Transient Soft X-ray Absorption Spectroscopy

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Synopsis The metal-to-ligand charge transfer dynamics in photoexcited $Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$ in aqueous solution is investigated by time-resolved soft X-ray absorption spectroscopy at the nitrogen K-edge using a table-top extreme high-order harmonic generation setup combined with a liquid flat jet. In our combined experimental and theoretical investigation, we unambiguously identify spectral signatures of the short-lived MLCT state before the long-lived ^{5}T quintet state is reached. Our study paves the way towards the investigation of ultrafast charge flow dynamics in solution.

$Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$ is a prototypical transition metal complex that is known to undergo ultrafast metal-to-ligand charge transfer and spin-crossover dynamics upon visible irradiation. Over the past 20 years, the ultrafast electronic and spin dynamics in photoexcited $Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$ has been intensively studied using various methods, from transient visible and ultraviolet absorption spectroscopy [1,2] to ultrafast Fe L- and K-shell x-ray emission and absorption spectroscopy [3-5]. Multiple electronic excited states, such as singlet and triplet metal-to-ligand charge transfer states (^{1,3}MLCT), triplet ligand field excited states (^{3}T), and quintet ligand field excited states (^{5}T), have been suggested to play a role in the charge and spin crossover mechanism. However, despite the numerous studies, the decay mechanism responsible for the transition form the initially populated ^{1}MLCT state to the final high-spin (HS) ^{5}T remains actively debated.

Here, we employed a newly developed femtosecond, table-top soft X-ray source that provides broadband pulses in the photon energy range of 250 eV to 450 eV to investigate the ultrafast MLCT dynamics in photoexcited $Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$ in solution. Upon 400 nm excitation, a new absorption band is observed in the transient X-ray absorption spectrum at a photon energy of 400.3 eV (see Fig.1). This absorption band decays with a ~130 fs time constant and is accompanied by the appearance of a novel absorption band at 399.4 eV with a rising time constant of ~160 fs, which can be assigned to the formation of molecules in the high-spin quintet ^{5}T state. By comparing our measurements to calculated N 1s spectra for different electronic excited states, we unambiguously identify the fast-decaying feature at 400.3 eV as N 1s transitions occurring in photoexcited molecules in the short-lived ^{1,3}MLCT manifold. Our work opens the door towards the investigation of femtosecond charge-transfer processes in solution by transient absorption spectroscopy in the X-ray water window spectral range.

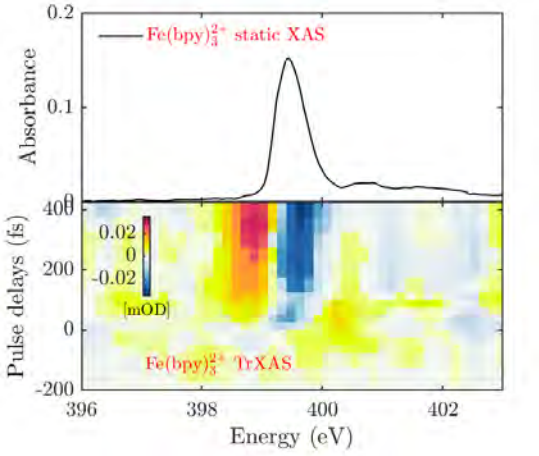


Figure 1. (Top panel) Steady-state absorption spectrum recorded in $Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$ in aqueous solution near the N K-edge. (Bottom panel) Transient X-ray absorption spectrum upon 400 nm excitation of $Fe^{II}(2,2'\text{-bipyridine})_3^{2+}$.

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Coherent Phonon-Driven Modifications of Orbital-Specific Local Fields

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Synopsis The presented study utilizes attosecond transient absorption spectroscopy (ATAS) to explore the dynamic response of local field effects (LFEs) on the photoabsorption of extreme ultraviolet (XUV) light in $Ti_3C_2T_x$ MXene. Our study reveals orbital-specific modifications of LFEs through coherent phonons probed by the shallow-core Ti *3p* state which cannot be explained by independent particle approximation (IPA).

Element-specific LFEs [1] remain hidden with most studies focusing on energy loss spectroscopy [2]. Here, we reveal *d*-orbital-specific LFEs for the first time in the functionalized MXene crystal $Ti_3C_2T_x$ (T_x: O, OH, F). Moreover, we demonstrate that the independent particle picture fundamentally fails to capture our observations due to its omission of collective effects, particularly LFEs and electron correlations.

We present a combined experimental and theoretical study providing insights into LFEs primarily modified by near-infrared (NIR) excited coherent A_{1g} phonons. The Ti *3p* shallow-core level serves as an optimal probe for LFEs, enabling us to resolve the interplay between distinct *d*-orbitals, particularly d_{xz} and the more localized d_{z^2}, during A_{1g} phonon-driven coherent oscillations. The energy region of our transient absorption measurement in Fig. 1a below the Fermi level E_F corresponds to a mixture of d_{xz} and d_{z^2} orbitals, while higher-energy levels exhibit almost pure d_{z^2} and fully pure d_{xz} states, as demonstrated by the calculated difference between the dominant *d*-orbitals, see Fig. 1b.

We solve the Bethe-Salpeter Equation (BSE) at an elevated electron temperature of 1000 K, incorporating lattice elongations obtained via Ehrenfest dynamics in time-dependent density functional theory (TDDFT) following NIR excitation. This approach successfully reproduces the measured changes in optical density (ΔOD). However, agreement with experiment is only achieved when LFEs are included in the BSE, whereas the IPA completely fails to capture our observations.

This highlights the high sensitivity of our measurement to LFEs induced by coherent atomic displacements via the A_{1g} phonon mode (see Fig. 1c).

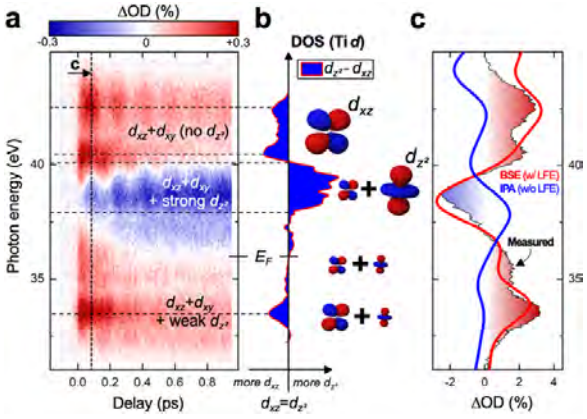


Figure 1. a) Measured ΔOD in $Ti_3C_2T_x$ after background subtraction. b) Orbital-resolved Ti DOS for T_x: O, showing $d_{z^2} - d_{xz}$ difference. c) BSE/IPA for T_x: O compared with measured ΔOD at 90 fs delay (label c in subfigure a).

Our study not only demonstrates the capability of attosecond transient spectroscopy to probe the LFEs in an element-specific manner but also showcases their sensitivity to phononic fingerprints even in individual *d*-orbitals with excellent theoretical agreement for the first time.

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Photoionization time delays in H_2O

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Synopsis Current work deals with the RABBITT simulation by taking continuum-continuum coupling into account. Photoionization time delays are extracted from the vibrationally resolved spectra in H_2O. We show that including nuclear motion leads to strong variation of the time delays.

Attosecond science has had a meteoric rise in the past two decades thanks to advances in the ultra-fast laser technology. This has allowed for controlling and observing electron dynamics in atoms and molecules in real time and to explore the phenomena happening on their natural time scales. In particular, the attosecond photoionization time delay has motivated a large number of studies in both atomic and molecular systems.

In the present work, we use the XCHEM methodology along with nuclear motion to simulate RABBITT spectra (recontruction of attosecond beatings by interferences in two-photon transitions) and calculate the corresponding photoionization delays in H_2O. We look at the lowest three lying cationic states of H_2O^+: $\tilde{X}^2B_1$, $\tilde{A}^2A_1$ and $\tilde{B}^2B_2$. The XCHEM approach[1, 2] includes electron correlation in both the target and neutral states, as well as correlation of the outgoing electron with those of the target. This is necessary to include in RABBITT as the absorption of the second photon happens when the electron is ionized, so the continuum-continuum transitions must be evaluated. XCHEM has already been shown to simulate the one-photon photoionization in the energy regions from the first cationic state to higher energies satisfactorily with the available literature.[3]

The channel resolved RABBITT spectra along with the photoionization time delays are presented up to an electron kinetic energy of 31 eV where comparison with other theoretical and experimental results is possible.[4] Despite a smooth nature of Wigner delays for the $\tilde{X}^2B_1$ and $\tilde{A}^2A_1$ cation states of H_2O the two-photon time delays show strong deviations and this seems to be exaggerated by the inclusion of the nuclear motion. The importance of both, including the continuum-continuum coupling and nuclear motion for such calculations of photoionization time delays is thus underlined.

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First-principles calculations of high-harmonic generation in amorphous Si and SiO

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Synopsis We study HHG in amorphous Si (a-Si) and SiO (a-SiO) using time-dependent density functional theory. aSi and a-SiO have a closed band gap, thus even with photon energies below the crystalline Si (c-Si) band gap, they undergo single-photon excitation under femtosecond pulses. Compared with c-Si, HHG spectra in a-Si and a-SiO show reduced intensity due to structural disorder, with a-Si having the highest and a-SiO the lowest cutoff energy.

Study on high-harmonic generation (HHG) is expanding from the gas phase to solids [1], further to the liquid phase [2], amorphous materials [3], and laser-induced heating effects [4]. The diversity of the underlying HHG mechanisms is under active debate.

This study investigates HHG in a-Si and a-SiO as well as c-Si with defects using time-dependent density functional theory with SALMON [5] on the Fugaku supercomputer. We consider femtosecond pulses of 14.4 fs duration and wavelengths of 1030, 1800, and 3000 nm. The simulation cells contain 512 and 552 atoms for a-Si and a-SiO, respectively. While even-order harmonics appear in c-Si with point defects (figure not shown) [6], the effect is more pronounced in amorphous materials. Figure 1 compares the HHG spectra of c-Si (without defects), a-Si and a-SiO for a 3000 nm pulse at a peak intensity of 10^{12} W/cm^2. Structural disorder and composition strongly influence on HHG, with a-Si and a-SiO having lower intensities than c-Si, but a-Si having the highest cutoff energy. Lattice distortion disrupts the periodic potential and shortens electron coherence time, reducing spectral intensity in amorphous materials. The cutoff energy of the first plateau for c-Si, a-Si, and a-SiO aligns with the prediction from the simple three-step model for c-Si, whereas the second plateau exhibits a higher cutoff energy for a-Si and a-SiO. These trends support previous modeling studies that suggested that the shape of the HHG spectra depends on the degree of structural disorder [7]. The plateau broadening in a-Si and a-SiO may result from electron density gradients or local field enhancement. These findings contribute to a systematic understanding of how liquid, amorphous, and crystalline phases influence HHG.

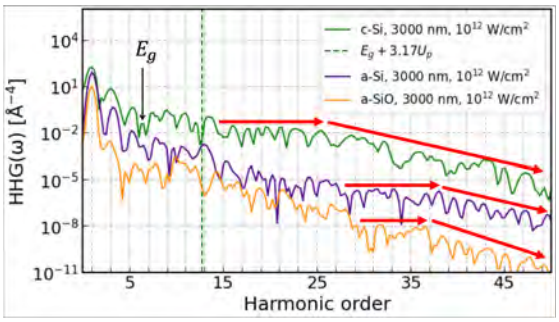


Figure 1. HHG spectra of c-Si (green), a-Si (purple), and a-SiO (orange).

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Ultrafast correlation dynamics during high-harmonic generation in multi-electron atoms

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Synopsis We study the role of electronic correlations during HHG in multi-electron atoms. By comparing fully correlated time-dependent simulations of noble gases (He, Ne) and of alkaline earth atoms (Be, Mg) we identify prominent features of the influence of correlations on both the tunneling as well as the recombination step. We identify species-specific signatures of correlations during tunneling imprinted on the high harmonic spectrum.

Originally viewed as a process involving one single active electron, the influence of multi-electron effects on the HHG spectrum has lately been extensively studied (see e.g. [1, 2]). In this work, we quantify the time-dependence of electron-electron correlations on ultrafast time scales using correlation measures from quantum information theory. One prominent measure is the von Neumann entropy $S_1(t)$ of the distribution of the occupation numbers of the natural orbitals, i.e. the eigenstates of the one-particle reduced-density matrix.

By explicitly solving the time-dependent Schrödinger equation with the multi-configurational time-dependent Hartee-Fock (MCTDHF) [3] method, we obtain fully converged results for He, Ne, Be and Mg. Using driving fields adjusted to the ionization potential of each atom, such that the driving occurs in the strong-field regime and double-ionization is negligible, we compare the HHG yields for the different atomic species. We find that, during tunneling, correlations systematically increase for noble gas and decrease for alkaline earth atoms (Figure 1). In analogy to the semiclassical three-step model, we identify how correlations affect the ionization as well as recombination process for each individual natural orbital. We provide an intuitive picture for the dynamics of $S_1(t)$ based on the shape and the dynamics of the natural orbitals. Using the classical Larmor formula, we show that the HHG yield is obtained as a sum of the contributions from the driven natural orbitals weighted by their time varying occupation numbers.

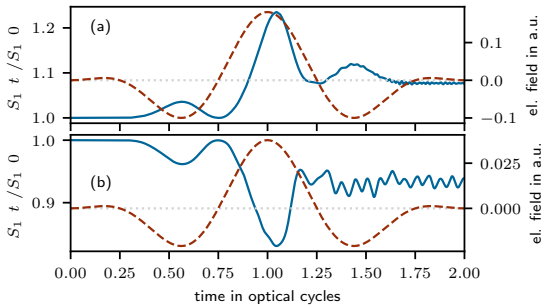


Figure 1. Time-dependent evolution of the entropy S_1 (blue) for helium (a) and beryllium (b) relative to the value in the ground state. The two-cycle laser field is plotted in dashed red as a reference.

We find that during recombination the natural orbitals of the multi-electron atom oscillate phase shifted relative to each other. In this collective motion correlations play an important role. We demonstrate how the effects of correlations both during tunneling as well as recombination imprint themselves on the HHG spectra.

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Electron-nuclear energy sharing through low-energy inelastic recollisions in dissociative strong-field ionization of deuterium

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Synopsis We present coincidence measurements of the dissociative ionization of D_2 in an intense 515 nm laser field with a half-frequency perturbation. The recorded phase-dependent photoelectron spectra strongly depend on the D^+ kinetic energy release (KER) and reveal the role of laser-driven electron recollisions. Our results indicate that inelastic recollisions facilitate bond breakage even at low D^+ energies via vibrational excitation.

Recollisions are known to contribute to high KER (i.e. several eV) ion emission from D_2 [1]. Low-KER ion emission (i.e. up to few eV), however, is typically understood to result from single or multiphoton absorption from the field. Here, we show experimentally that recollisions can contribute to the dissociation of D_2^+, even at KER ($\mathrm{KER} = E_{D^+} + E_D$) down to 0 eV. For such low KER, our data implies that the photoelectron excites a dissociating vibrational state in the molecular ion upon recollision. Our experiment uses a two-color technique based on a second harmonic field (515 nm, 50 fs, 10^{14} W/cm^2) and a perturbative fundamental (1030 nm, $\approx 10^{10}$ W/cm^2). The perturbation induces phase-dependent sidebands in the ATI spectrum, generated by the 515 nm light. Both, main ATI peaks and sidebands depended on the relative two-color phase, allowing us to use phase-of-the-phase spectroscopy [2]: The experimental data are Fourier-transformed with respect to the relative phase, and the oscillation amplitude, and the oscillation phase (the "phase-of-the-phase", PP) are plotted as a function of the electron momenta in Figure 1: The PP map for D2+ (Fig. 1(a) right) shows various color patterns corresponding to direct electron emission, forward, and backward scattering. In stark contrast, the PP map for D^+ (Fig. 1(b) right) shows only a single color pattern, which coincides with the one observed for backscattered electrons in Fig. 1(a) (right) - though, at substantially lower momenta. In addition, a narrower angular distribution is observed for D^+, again indicating reduced scattering momentum. Hence, the photoelectron transfers energy to the ion via inelastic recollision. The inelastic nature of the rescattering is confirmed by the energy correlation between ions and electrons, detected in coincidence, extending down to 0 eV ion energy. The similar yield between 0 and 1 eV KER indicates that vibrational excitation, driven by inelastic recollisions, contributes to these low energies.

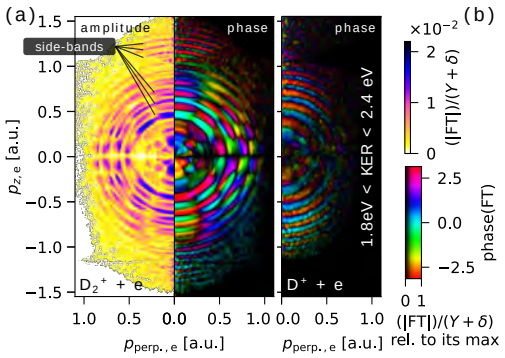


Figure 1. Phase-dependent ATI momentum distributions of electrons detected in coincidence with (a) D_2^+ and (b) D^+ ions (1.8 eV < KER < 2.4 eV). In (a), the relative-phase contrast (oscillation amplitude) is shown on the left, and the phase-of-the-phase on the right. In (b), only the phase-of-the-phase is displayed [3].

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Experimental Realisation of Synthetic Chiral Light

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Synopsis We have created *synthetic* chiral light using a non-collinear 800 nm scheme with an orthogonally polarized 400 nm component in one beam. In the overlap region, the field is *locally* chiral: the tip of the electric-field vector traces a chiral trajectory in time. By recording the non-linear emission from BBO and the sub-cycle control over the two-colour phase delay, we demonstrate the generation and control of our chiral field.

Traditional chiroptical methods rely on the chirality of circularly polarised light (CPL) to determine molecular handedness. However, due to the large disparity between the pitch of the light's helix and the size of the molecules, the interaction of CPL with chiral molecules is weakly enantiosensitive.[1] Synthetic chiral light is an efficient alternative to CPL.[2] This new type of chiral light is locally chiral, i.e. the electric-field vector traces a 3D chiral Lissajous figure in time, that drives ultrafast chiral currents on the molecular scale. The nonlinear response of chiral molecules to such tailored fields is orders-of-magnitude more enantiosensitive than with CPL.[1, 2]

We have created locally chiral light in our laboratory. Our setup uses two 50 fs non-collinear 800 nm beams linearly polarised in the plane of the optical table. One of the two beams has an additional orthogonally polarised 400 nm component. The beams are then focused and overlapped spatially and temporally (Fig. 1, top left).

To characterise our chiral field, we recorded the nonlinear response of a BBO crystal. Fig. 1, top right, shows the far-field emission from the BBO imaged on a piece of white card. Each nonlinear process emits in a specific direction due to the non-collinear geometry and conservation of linear momentum. The second harmonic at position 0 and the two third harmonic generation (THG) signals at -1 and 1 confirm overlap of the two 800 nm beams. A non-collinear sum-frequency generation signal (SFG) also appears at position 1 and confirms the overlap of the 400 nm and 800 nm components in opposite beams. This validates the simultaneous overlap of all components and demonstrates the generation of a locally chiral field.[2, 3]

The chirality of the field is controlled by the relative phase of the 800 and 400 nm field components. Fig. 1, bottom, shows the intensity of the ~266 nm signal at position 1 as a function of the 400 nm delay. A clear, reproducible interference effect between the SFG and THG is measured. This demonstrates the stability and fine control over the chirality of our field. [2, 3]

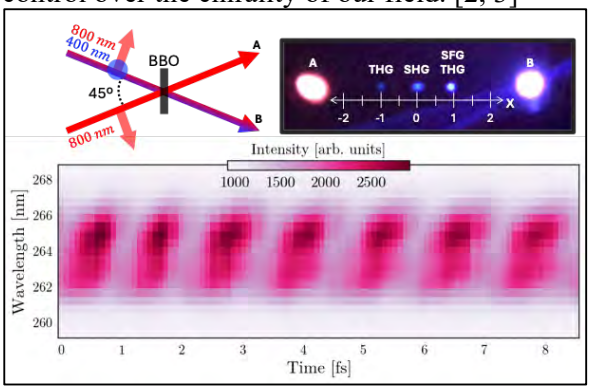


Figure 1. Top left, experimental scheme. Top right, emission from the BBO imaged on white card. Bottom, intensity of the ~266 nm emission at position 1 as a function of the 400 nm delay.

We aim to further demonstrate the unique interactions of our locally chiral field with chiral biomolecules to create new methods for highly efficient detection and control of chirality.

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Ab initio calculation of real-space light-driven current in graphene under strong laser fields

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Synopsis Using an ab initio method, i.e., time-dependent density functional theory, we study a directional electric current induced in graphene by a strong few-cycle laser pulse and its real-space distribution. Our results reproduce the current direction reversal originating from the cancellation of two residual currents in opposite directions, previously predicted by Morimoto *et al.* [1]. Further, our simulations reveal a complex electric field dependence of the three-dimensional real-space current distribution.

The interaction between strong laser fields and graphene will lead to the ultrafast nonlinear dynamics as a result of its unique Dirac cone structure in reciprocal space. Higuchi *et al.* have experimentally found that the peak field intensity of the few-cycle laser pulse can modulate the carrier-envelope phase-dependent residual current due to the asymmetric conduction-band population [2]. Using the nearest-neighbor tight-binding model (TB), Morimoto *et al.* reveal that the formation of two currents in opposite directions with different pathways in graphene canceling each other causes a decrease of the total residual current from a real-space perspective [1].

Since time-dependent density functional theory (TDDFT) does not require an explicit overlap integral in TB, it avoids errors in current continuity. In addition, it complements the out-of-plane view of graphene that is missing in the real-space two-dimensional model. We use the SALMON TDDFT software package [3] to calculate the current response after the pulses at a wavelength of 800 nm with different peak field amplitudes and to visualize the three-dimensional current density distribution. In Fig. 1, at the peak field amplitude of 3.5 V/nm, the total residual current is a positive value close to zero, representing a field where the direction reversal occurs, while two currents in opposite directions along the frame and the center of the hexagonal structure, respectively, are verified to cancel each other at the position of carbon nuclei in the XY plane (graphene plane), which supports the prediction by Morimoto *et al.* [1]. When the current density is projected onto the XZ plane, the tiny positive current is found slightly above and below the basal (XY) plane of σ bonds rather than within it. This work supports the explanation of the direction reversal in real space by TDDFT and further reveals its three-dimensional spatial distribution.

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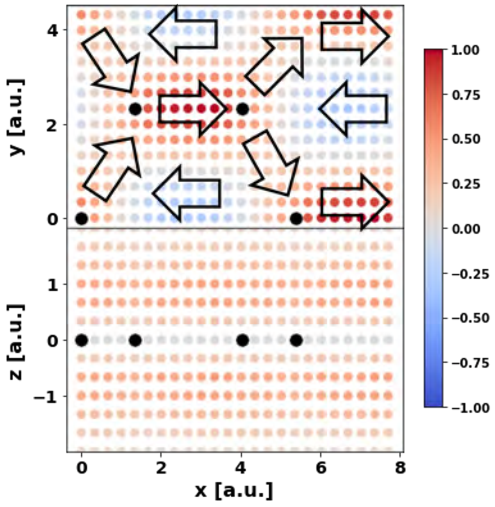


Figure 1. Projection of residual current distribution component along the x direction which is the direction of laser polarization (parallel to C-C bonds) to the XY and XZ plane in an orthogonal unit cell of graphene. Arrows show the direction of the current and the black circles represent the position of the carbon nuclei. A normalized coefficient is used for the ratio of the distribution.

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Photoionization of Chiral Molecules by Sub-Cycle Shaped Laser Fields

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Synopsis We present results from a versatile two-color photoionization setup with full control over polarization, relative intensities, and phase locking of a fundamental field ω and its second harmonic 2ω. This setup enables systematic studies of chiral photoionization dynamics under tailored field configurations, revealing their impact on chiral asymmetries in photoelectron angular distributions.

Chiral photoionization is a fundamental process in light-matter interactions, crucial for understanding chiral molecules, whose enantiomers can only be distinguished through their response in chiral environments. Probing its dynamics with high temporal (attosecond) and spatial (ångström) resolution provides a deeper understanding of chiral effects, but requires advanced experimental techniques. This, for instance, can be achieved with laser-induced electron diffraction (LIED) [1].

In this work, we present results from a versatile experimental setup that enables precise control over the polarization states, relative intensities, and phase locking of two collinear ω and 2ω fields. By exploring various field configurations—combinations of linear, elliptical, and circular polarizations, as illustrated in Fig. 1—we analyze their impact on chiral photoionization on different chiral molecules.

Starting with an elliptical strong-field infrared pulse, as used in [1], we introduce a perturbative 2ω field aligned with the ellipse's major axis, as in (a), allowing us to modulate the electric field strength and extract chiral phase shifts from holographic interference patterns [2]. Then, rotating the 2ω field's polarization by 90°, as in (b), we control the direction of electron collisions LIED. Furthermore, switching to bicircular counter-rotating fields, as in (c), can enhance the recollision probability, amplifying the LIED signal [3]. Meanwhile, co-rotating fields, as in (d), enable attosecond delay measurements via the HASE method [4], which we aim to compare to the RABBITT-like method [5]. Orthogonally linearly polarized ω and 2ω fields, as in (e), create a chiral field that reverses handedness every half laser cycle [6]. In this last configuration, we examine recollision effects and perturb electron trajectories by tuning the 2ω ellipticity. Finally, we explore unconventional setups, such as the "fish-like" configuration, as in (f), to optimize chiroptical responses.

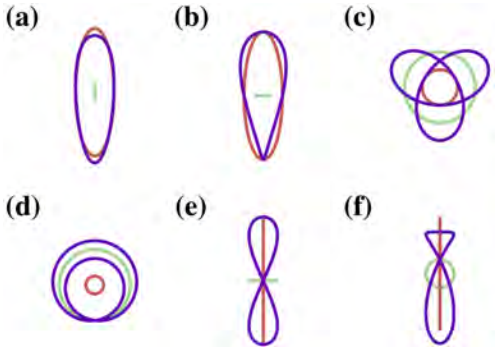


Figure 1. Schematic of ω (light red) and 2ω (light green) field configurations and their superposition (purple).

This approach provides a powerful framework for probing and manipulating chiral electron trajectories in real time. It not only enables precise measurements but also opens new ways for controlling chiral light-matter interactions on sub-optical-cycle timescales.

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Ga3d and Se3d spin orbit delays in GaSe solids

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Synopsis Photoemission delays from Ga 3d and Se 3d fine structure components are extracted by RABBITT, despite the small fine structure splitting. The large delays revealed by these measurements show the need for adequate theory beyond ballistic electron transport.

The reconstruction of attosecond beating by interference of two-photon transitions (RABBITT) is one of the standard methods for probing photemission dynamics in atoms, molecules and solids [1]. Both, atomic scale effects in solid state photoemission and the transport of the electron wave packet in the material affect the total photoemission delay [2]. A unified description of this process is still open and therefore spin-orbit delays, which have been reported for Xe and Kr ions [3], as well as the van-der-Waals crystals Bi_2Se_3 and Bi_2Te_3 [4], are ideal to investigate the interplay between atomic and material induced delays in the photoemission process.

Here we report spin-orbit delays for two core levels of GaSe and discuss the consequences for a unified theoretical model of photoemission delays. The fine structure splitting in the Ga 3d and Se 3d shells of $\Delta E_{LS,\mathrm{Ga3d}} = 0.5\mathrm{eV}$ and $\Delta E_{LS,\mathrm{Se3d}} = 0.84\mathrm{eV}$ together with broadening due to the XUV linewidth leads to strongly overlapping RABBITT-spectrogramms of both spin-orbit split components. Despite this the extraction of spin-orbit delays is possible and yields $\tau(\mathrm{Ga3d_{5/2}}) - \tau(\mathrm{Ga3d_{3/2}}) = -164 \pm 49\mathrm{as}$ and $\tau(\mathrm{Se3d_{5/2}}) - \tau(\mathrm{Se3d_{3/2}}) = -97 \pm 43\mathrm{as}$.

Such large delays for energetically close ground states cannot be explained by ballistic transport of photoelectrons in the material, where small differences in kinetic energy and inelastic mean free path lead to a small relative photoemission delay. This is corroborated by the observation of the delays between Ga3d and Se3d spin-orbit split states. The relative delay between Ga and Se 3d differs between $-80 \pm 21\mathrm{as}$ for the $j = 5/2$-states and $-4 \pm 55\mathrm{as}$ for the $j = 3/2$-states respectively although the involved kinetic energies of the photoelectrons are almost identical. The obvious difficulty to explain such large relative delays by transport phenomena indicates that local atomic scale delay mechanisms also are relevant for spin-orbit delays in solid state photoemission.

This tentative assignment is further supported by the observation that the Se3d spin-orbit delay reported here for GaSe agrees well with those found in Bi_2Se_3 and WSe_2. The same material independence has also been observed for the Bi5d spin-orbit delay Bi_2Se_3 and Bi_2Te_3 [4]. Seemingly the material properties and in particular the rather different continuum band structure of the various materials has less impact on the spin-orbit delay then the local electronic structure of the emitter atom. This opens an interesting perspective with respect for further investigating of the spin-orbit delays in solids, seeing to what extent atomic contributions and solid-effects affect photoemission delays.

The observation of spin-orbit delays and their material independence lead to a breakdown of the picture of both, ballistic electron transport and the three-step-model of photoemission. These results demand for a more complex model like the one-step photoemission [5], where effects like the final-state bandstructure as well as local atomic effects can be included, to explain the observations of time-resolved attosecond spectroscopy on solids.

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High-Order Harmonic Generation in Pentacene

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Synopsis High-order harmonic generation is experimentally demonstrated from pentacene, a prototype organic crystal. This novel target class shows efficient harmonic generation far beyond the lowest electronic transition energy. Polarization dependent harmonic yield measurements indicate a significant contribution of inter-molecular couplings to the generation process.

High-order harmonic generation (HHG) is an extremely nonlinear optical response of materials exposed to intense light fields. It can be used to probe electronic structures and dynamics in matter on ultrashort timescales. HHG has established as a powerfull all-optical spectroscopic tool in condensed matter systems, allowing investigation of electronic and crystal structures [1, 2].

In this work, we extend HHG spectroscopy to a target class that was so far untouched by HHG experiments: organic molecular crystals (OMCs), consisting of perfectly aligned, large organic molecules. As a prototype organic crystal we chose pentacene. Compared to covalent and ionic bonds in inorganic crystalline solids, organic molecules in OMCs are weakly coupled via van-der-Waals interaction or hydrogen bonds. This results in rather flat electronic bands and excited states localized on a few neighboring molecules. In this work, we experimentally address the question, if pentacene crystals survive the high field strengths necessary for HHG and if the generation process is dominated by isolated molecules or by crystal effects.

We find that pentacene crystals show highly efficient HHG when driven in the mid-infrared. Order 17, the highest order observed with a photon energy of approx. 5.3 eV, is well above the lowest electronic transition energy of 1.85 eV. To investigate the origin of the harmonics, the harmonic yield was measured as a function of the laser polarization. The strongly polarization dependent yield is shown in Fig. 1. For all harmonics, two maxima are visible at directions (indicated by black arrows at 50° and 130°) that connect nearest neighbors. Interestingly, the higher harmonic orders 11 and 13 are particularly sensitive to the couplings even to next-nearest neighbors, as they show a third principle direction at 0°. In contrast, no enhancement is visible in directions of the single molecular transitions. This indicates the superior importance of intermolecular couplings compared to single molecule responses, although the intermolecular coupling is weak.

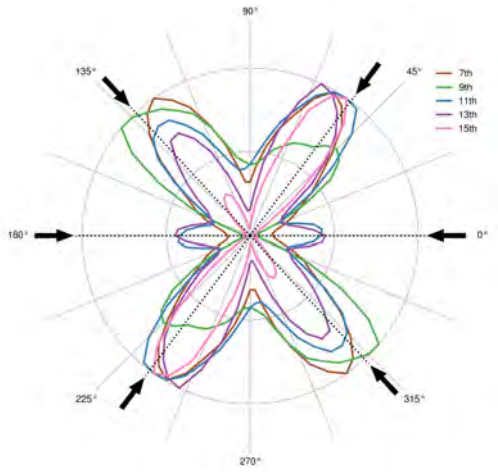


Figure 1. Yields of harmonics 7 to 15 as function of the driving field polarization. Enhanced harmonic emission is observed in directions of nearest and next-nearest neighbors, marked by black arrows.

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A universal approach to saddle-point methods in attosecond science

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Synopsis
Strong-field processes like HHG and above-threshold ionisation are naturally described in terms of highly-oscillatory integrals which we often tackle using saddle-point methods. These methods simultaneously simplify the calculations and let us understand the physical processes in terms of quantum orbits. However, applying saddle-point methods for fields without clear symmetries has remained challenging, up until now. Here we introduce Picard-Lefschetz theory as a universal and robust link between the time integrals and the semi-classical trajectories, for arbitrary driving laser fields.

Light-matter interactions in the strong-field regime, such as high-harmonic generation (HHG), typically give rise to highly-oscillatory integrals, which are often solved using saddle-point methods. Not only do these methods promise a faster computation, but they also inform a more intuitive understanding of the process in terms of quantum orbits, as the saddle points correspond to interfering quantum trajectories (i.e., Feynman's path integral formalism).

Despite these advantages, a sound understanding of how to rigorously apply saddle-point methods to highly-oscillatory integrals in high dimensionality with algorithms that work uniformly for arbitrary configurations and laser fields, remains conspicuously absent. This hinders our ability to keep up with state-of-the-art experimental setups which increasingly rely on tightly-controlled laser waveforms.

We introduce the key ideas of Picard-Lefschetz theory – the foundation of all saddle-point methods – and their implementation. Using HHG and above-threshold ionisation driven by two-colour laser fields as examples, we show how those ideas provide a universal and robust approach for a fast computation of the integrals, as well as a widely-applicable algorithm to derive the relevant semi-classical quantum orbits that underlie the physical processes. We demonstrate this for setups that involve spectral caustics, as well as intensity scans for a two-colour field beyond the perturbative regime ("colour switchover"), which were previously rendered impossible to solve with saddle-point methods due to coalescences and branch cuts.

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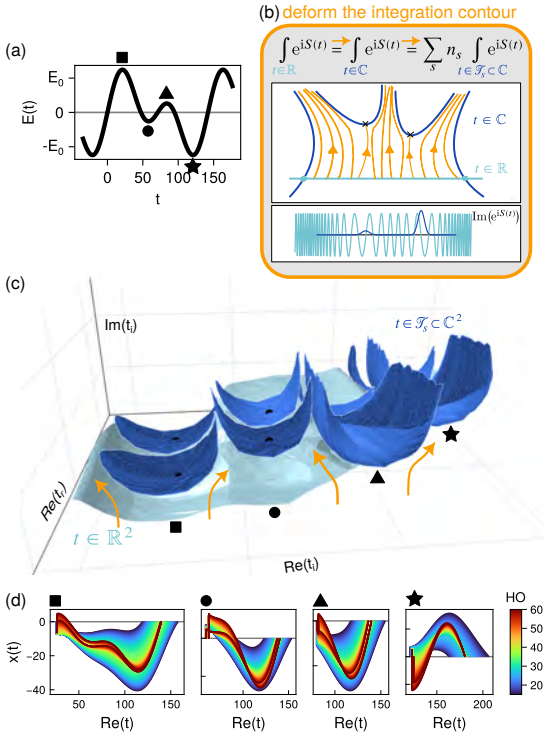


Figure 1. For arbitrary driving fields (panel (a)) we can solve time integrals by deforming the integration contour from the real into the complex domain, so as to minimise oscillations of the integrand (b). In the case of HHG we integrate over ionisation and recombination times, hence the deformed contour is embedded in $\mathbb{C}^2$. A projection in $(\mathrm{Re}(t_i), \mathrm{Im}(t_i), \mathrm{Re}(t_r))$ for harmonic order 25 generated from the above field is shown in (c), and the corresponding electron trajectories for a range of harmonic orders in (d).

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Relativistic calculations of electron–parent ion entanglement using the KRAKEN protocol

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Synopsis In systems with multiple open ionisation channels, such as with valence shell fine-structure splitting, ejected electrons are not pure wave-functions, but instead need to be described using density-matrix formalism. With recent advances in attosecond science, methods such as KRAKEN have been developed to fully reconstruct the one-photon density matrix. This is done by interferometric measurements after the addition of a probe pulse. The present communication uses Relativistic Random Phase Approximation with Exchange to directly calculate the one-photon density operator and its reconstruction.

In most atomic systems, single ionisation-channel continua are the exception, rather than the norm. When multiple ionisation channels are open, the purity γ of the state density operator can provide information on the electron–Parent Ion entanglement. This information may not be extracted using only an XUV pump, but can be obtained with the addition of a probe pulse. This is the principle behind pump-probe methods, which allow for the extraction of time-dependent information by varying the pump-probe delay.

KRAKEN is a recently proposed [1] and implemented [2] interferometric pump-probe method aimed at reconstructing the one-photon density operator $\hat{\rho}_{\mathrm{XUV}}$. In KRAKEN, an XUV pump is combined with a two-colour IR probe, allowing for interference measurements of the population at any two intermediate energies ε_1 and ε_2. The retrieved information is assumed to be representative for the one-photon wave packet, allowing the density matrix elements $\langle\varepsilon_1|\hat{\rho}_{\mathrm{XUV}}|\varepsilon_2\rangle$ to be reconstructed.

From the point of view of theory, both the one- and two-photon matrix elements can be directly obtained, without the need for any of the assumptions relied on in the KRAKEN protocol. Thus, both $\hat{\rho}_{\mathrm{XUV}}$ and its KRAKEN reconstruction may be calculated—and compared—directly. The present work makes use of Relativistic Random Phase-Approximation with Exchange (RRPAE) [3] to simulate KRAKEN in rare-gases, where the fine-structure splitting of the ground state as well as channel-dependent resonance features results in a lowering of the purity. Both the $\hat{\rho}_{\mathrm{XUV}}$ and its KRAKEN reconstruction are calculated and compared to evaluate the assumptions present in the method.

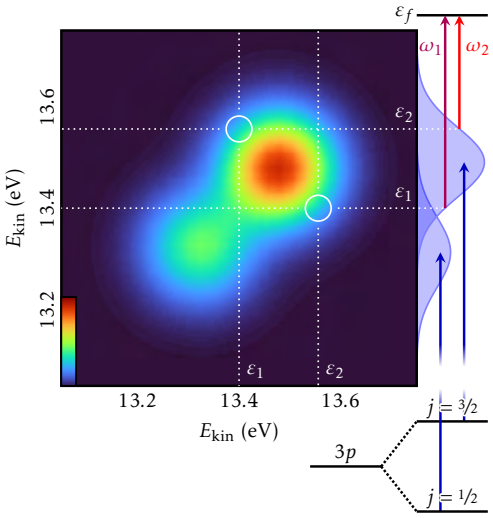


Figure 1. The magnitude of the calculated argon one-photon density operator $\hat{\rho}_{\mathrm{XUV}}$. Two maxima are present, due to fine-structure splitting of the $3p$ orbital. The matrix elements $\langle\varepsilon_1|\hat{\rho}_{\mathrm{XUV}}|\varepsilon_2\rangle$ can be reconstructed from the beatings induced at a final energy ε_f by a two-colour IR-probe. The two-photon ionisation paths are shown to the right.

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Probing the final state continuum on atomic length scales by attosecond time-resolved photoelectron spectroscopy

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Synopsis Photoelectron dynamics in Bi_2Te_3 and Bi_2Se_3 are investigated by RABBITT and substantial photoemission delays between fine structure components of the Bi 5d core levels were determined. These delays cannot be interpreted in terms of atomic photoionization delays or nearly-free photoelectron transport delays. Instead, one-step photoemission theory is needed to explain the experimental observations revealing that the emitter atom is probing variations of the photoemission final state continuum on an atomic length scale.

By attosecond time-resolved photoelectron spectroscopy photoemission dynamics in solids can be observed on its natural time scale [1]. This access to the timing of photoemission challenges the theoretical understanding of photoemission dynamics fundamentally [2]. Recently, RABBITT experiments on gas-phase Xe and Kr atoms revealed significant photoionization delays between different fine structure states of the same p-shell, which are not yet fully understood [3]. In this study we expand these studies to significantly more complex systems, i.e., solid-state photoemission from Bi_2Te_3 and Bi_2Se_3 van-der-Waals crystals. For the first time photoemission delays between different core-level fine structure components in a solid are resolved with attosecond precision.

For this study several hundreds of RABBITT spectrograms were measured on both materials. In Bi_2Te_3 the RABBITT oscillations reveal spin-orbit delays in the Bi 5d initial state band of $\tau(\mathrm{Bi5d_{5/2}}) - \tau(\mathrm{Bi5d_{3/2}}) = 30 \pm 13\,\mathrm{as}$, while the corresponding spin-orbit delay in Bi_2Se_3 is $\tau(\mathrm{Bi5d_{5/2}}) - \tau(\mathrm{Bi5d_{3/2}}) = 31 \pm 10\,\mathrm{as}$.

Since both photoemission channels originate from the same emitter atom and are only weakly separated in final state energy, spin-orbit delays in solids provide unique access to the observation of photoemission delays which cannot be interpreted by nearly-free transport of photoexcited electrons. The remarkable agreement of the Bi 5d spin-orbit delays in two different materials suggests, that these delay differences are dominated by transition matrix effects, i.e., intra-atomic contributions. However, calculated photoionization delays in isolated Bi atoms do not predict spin-orbit delays in this order of magnitude. In contrast, taking photoelectron scattering in the solid into account via *ab initio* one-step photoemission theory [4] predicts substantial spin-orbit delays in this energy range.

This approach includes the effect of evanescent waves in photoemission. The character of these photoemission final states in band gaps and Cooper minima can have a strong influence on the photoemission delay. This questions the wave packet picture of electron transport based on propagating partial waves. However, the surprising match of spin-orbit delays in two materials suggests, that the final state continuum alone cannot account for our observation. Instead, it is the emitter atom localized close to the surface that probes the final state continuum with atomic spatial resolution.

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Ultrafast charge carrier dynamics in semiconductor nanostructures resolved in time, space and energy

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Synopsis An energy resolved PhotoEmission Electron Microscope (PEEM) is combined with a High-order Harmonic Generation (HHG) eXtreme Ultra-Violet (XUV) light source driven by few-cycle Near-Infrared (NIR) laser pulses delivered at 200 kHz repetition rate. Using a pump-probe interferometer, we study ultrafast charge carrier dynamics in nanostructures with femtosecond (NIR-NIR) and attosecond (NIR-XUV) resolution.

PEEM is a powerful tool for studying electronic states in nanoscale solid state components, in both real-space and *k*-space (momentum-space) [1]. Combined with a pulsed light source, time-resolved PEEM (TR-PEEM) offers fundamental insights into charge carrier dynamics, relevant for both next-generation high speed electronics [2] and increasing the efficiency of solar cells [3]. We present our recent progress in combining TR-PEEM with a 200 kHz attosecond light source.

A pump-probe setup is coupled to the PEEM to study the temporal dynamics, utilizing two ultrashort (<6 fs) near-infrared pulses with variable delay. The pulses are used to produce a comb of phase-locked odd order harmonics to probe charge carrier dynamics with attosecond resolution. The beamline includes an XUV spectrometer and a 3D momentum electron spectrometer, enabling characterization of the incident XUV radiation to a high degree. Figure 1a shows the set-up schematically. The XUV is generated in an Ar gas jet at (1) and recombined with the NIR, before being focused into another gas jet (2) for XUV characterization. The light is then focused onto the sample surface in the PEEM at (3), producing photoelectrons (PEs) which are captured by electrostatic lenses and detected at (4).

The PEEM is equipped with a Time-of-Flight (ToF) tube and a delay-line detector, and we are able to simultaneously image the spatial distribution and resolve the energy spectrum of the PEs. Figure 1b shows an InP NW imaged in the PEEM using the NIR laser. Figure 1c shows a scanning electron micrograph of similar InP NWs.

We study III-V semiconductor nanowires (NWs), whose small dimensions lead to localized enhancements of the electromagnetic field, thus increasing the probability of non-linear effects such as multiphoton electron emission. Our previous TR-PEEM studies [4] on NWs have demonstrated the feasibility but suffered from a lack of a proper energy spectrum, highlighting the importance of the ToF unit in studying such systems.

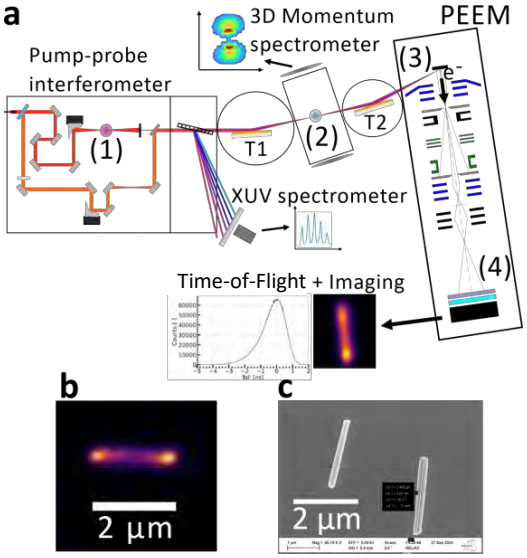


Figure 1. (a) Set-up schematic. T1,T2: Toroidal mirrors. (b) InP NW imaged in the PEEM. (c) InP NW imaged in a scanning electron microscope.

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Scaling of the fractional angular momenta of a light polarization Möbius strip in extreme nonlinear optics

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Synopsis The rotational symmetries of a light beam are linked to its angular momentum. Specifically, spin and orbital angular momentum (SAM & OAM) correspond to the rotational invariance of the polarization vector and of the spatial distribution of the light field, respectively. Both take values that are integer multiples of $\hbar$. However, in certain cases, a generalized angular momentum (GAM), linear combination of SAM and OAM, naturally emerges with fractional values of $\hbar$. Through theory and experiments, we have demonstrated that half-valued GAM is conserved during high-harmonic generation.

Standard decomposition bases, such as Hermite-Gaussian or Laguerre-Gaussian modes with left- and right-handed polarization, are usually well-suited for describing any light beam. These modes serve as eigenmodes of both the Spin and Orbital Angular Momentum (SAM and OAM) operators, yielding quantized values of helicity as $\pm\hbar$, and of the OAM as $\ell\hbar$, with $\ell \in \mathbb{Z}$. Recently, Ballantine et al. [1] observed that, under specific conditions, light beams may not be eigenvectors of either the SAM (S_z) nor OAM (L_z) operators individually but rather of linear combinations of them, reading $J_{\gamma,z} = L_z + \gamma S_z$, with γ a rational value determined by the beam's symmetries. This concept of "Generalized Angular Momentum" (GAM) opened new possibilities for fundamental tests, beam shaping, quantum optics... However, it has received little attention. Notably, it was predicted to exhibit well-defined behavior in extreme nonlinear optics [2], but this has yet to be experimentally tested.

Investigating GAM in nonlinear optics presents the challenge of identifying a nonlinear medium that preserves the polarization state while also producing wavelengths suitable for GAM measurement. In previous work, we demonstrated that High Harmonic Generation (HHG) provides an ideal platform for studying OAM conservation laws [3, 4]. Here, we report the development of experimental methods to measure the GAM of a light beam in the extreme ultraviolet (XUV) spectral range [5]. Our observations confirm that GAM scales linearly with the harmonic order in HHG [5]. Remarkably, even when harmonics are superpositions of different SAM and OAM states, they still carry a well-defined fractional GAM charge, making GAM the best quantum number for certain nonlinear interactions.

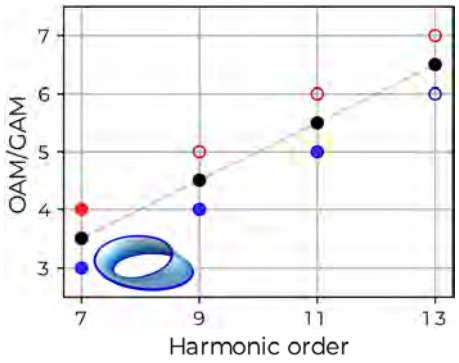


Figure 1. Experimental observation of the scaling of angular momentum in HHG. Each odd harmonic frequency is made of two underlying beams with opposite SAM (red and blue dots) carrying different OAM. Conversely, the GAM is single valued (black dots), with half integer values, and scales linearly with the harmonic order.

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Calculations of Impulsive Stimulated X-ray Raman Scattering in Liquid Water

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Synopsis We model the impulsive stimulated x-ray Raman scattering in water molecules in the liquid phase upon interaction with an intense attosecond x-ray pulse, considering two different configurations: monomers and dimers. We compare the x-ray Raman spectral signatures and population dynamics for these configurations.

Impulsive stimulated x-ray Raman scattering (ISXRS) can be used to excite neutral molecules to valence states, providing a tool to study attosecond charge dynamics [1,2]. Here we simulate the ISXRS in a liquid water sheet upon interaction with an intense attosecond x-ray pulse. During ISXRS, the water molecules interact twice with the x-rays, leading to a first excitation from the initial state into a core-excited state and then to a valence-excited state.

We model the ISXRS in monomer and dimer configurations to explore the effects of the hydrogen bonding present in the dimer. A classical molecular dynamics simulation was performed to extract a set of dimer geometries, on which the electronic states wavefunctions were obtained using a quantum mechanics/molecular mechanics (QM/MM) model. In these calculations, the water molecules were described by a complete active space self-consistent (CASSCF) method. The microscopic response of the QM structures to x rays was computed by solving the time-dependent Schrödinger equation (TDSE). To simulate the propagation effects on the x-ray pulse as it traverses the liquid sheet, we follow the unidirectional pulse propagation equation derived from Maxwell's equations [3].

Figure 1. shows the ISXRS signal for the monomer and the dimer at different propagation distances through the water sheet. The ISXRS signal is quite different for the monomer and the dimer, while in the monomer it exhibits 2 sharp distinct emission peaks, in the dimer it exhibits a wider single emission feature. This is due the geometrical averaging in the dimer calculations, where each geometry exhibits distinct emission peak energies.

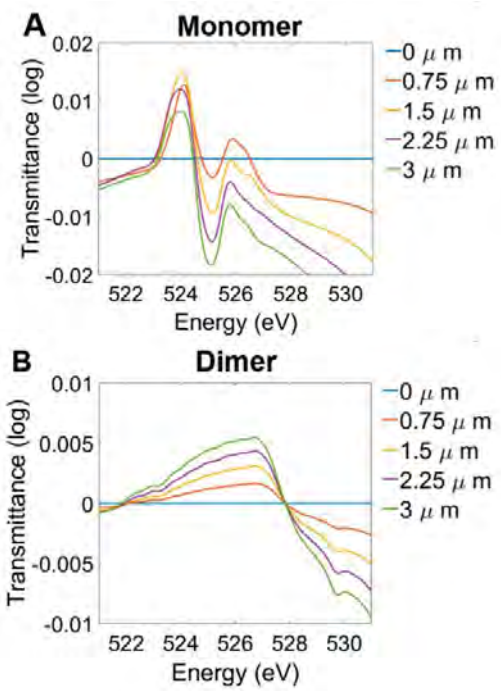


Figure 1. Computed transmittance (ratio of transmitted intensity I_{out}/I_{in}) of an x-ray pulse as it travels through the liquid water sheet considering the monomer (A) and dimer (B) configurations, as a function of the propagation distance. We use an input x-ray pulse of 80 PW/cm^2 peak intensity, 529 eV central energy and 7 eV FWHM.

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On the use of a self-homodyning experiment to study the quantum nature of High Harmonics generated in solids

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Synopsis Recently, independent theoretical and experimental investigations have revealed non-classical effects in the high harmonic radiation [1, 2, 3]. In this context, performing a homodyne detection experiment to measure the quadratures and reconstruct the Wigner function of each individual harmonic will allow us to gain a deeper understanding of the quantum nature of HHG. In this paper we present the self-homodyne setup we use, the challenges of such an experiment and our most recent results, including the calibration of an industrial homodyne detector for ultrashort pulses.

Among the many quantum photonic sources currently available, high harmonic generation (HHG) remains a relatively unexplored area. In addition to the fundamental interest in understanding the physics behind HHG, a better understanding of the quantum nature of this process has potentially broad implications for the rapidly developing field of quantum technologies. Indeed, recent studies have demonstrated its potential for quantum technologies such as quantum imaging.

Our team has recently provided the first evidence for the existence of superbunching and non-classical correlations between high harmonics with a Hanbury-Brown-Twiss-like experimental setup, which led to a violation of the classical Cauchy-Schwarz inequality [2]. However, this experiment provided only partial information about the quantum state of the harmonics. To obtain a complete reconstruction and knowledge of the wave function of a high harmonic, one solution is to measure its quantum Wigner function by means of a homodyne detection experiment.

The principle of such an experiment is to make interferences between an unknown state and a powerful local oscillator on a beamsplitter, and then to subtract the intensities measured by two photodiodes at the output of the beamsplitter. The result of this subtraction is a voltage proportional to the quadrature of the unknown states. With enough measurements, it is possible to fully reconstruct its quantum Wigner function.

The two main difficulties behind homodyne detection are a low-noise electronics (which is sensitive to the length of the pulses) and the generation of a local oscillator with spectral and temporal modes close enough to the modes of the studied states. With the self-homodyning technique [4] we use, the local oscillator is generated under conditions similar to the generation of the studied state, which helps the mode matching between the two beams. We have also calibrated an industrial *Newport* homodyne detector for very short pulses and compared the difference in behaviour between harmonic and nanosecond pulses around 700 nm. This experiment is not yet guaranteed to work, but we are providing useful insights and will present our latest progress on the challenge of measuring the quantum Wigner function of high harmonics generated in solids.

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Spatial mode transformations of high-order harmonic generation in transition metal dichalcogenides

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Synopsis In this work, we make use of the lattice symmetries of transition metal dichalcogenide (TMD) monolayers to locally influence the polarization and phase of the harmonics emitted by high-order harmonic generation (HHG). High-order harmonics were generated from mono- and polycrystaline molybdenum disulfide (MoS2) monolayers with an infrared femtosecond pulse. We control the orbital angular momentum (OAM) and spin angular momentum (SAM) of the generation beam by using a liquid crystal Q-plate. We then measure the OAM and the full polarization map of the emitted harmonics.

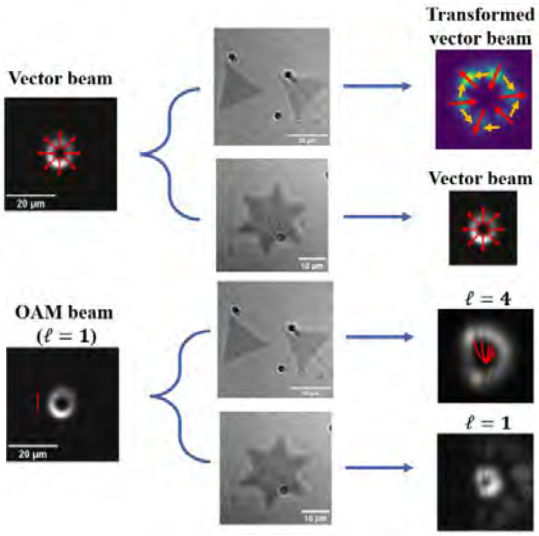


Figure 1. Illustration of the mode conversion of the 4th harmonic by mono- and polycristaline MoS2 monolayers. The top row depicts the transformation of a vector beam while the bottom row shows the conversion of OAM.

Since the first observation of high-order harmonic generation (HHG) from bulk crystals by S.Ghimire *et al.* in 2011, HHG in solids has been a growing field in ultrafast optics and condensed matter physics. Linear and nonlinear optical properties of TMDs were often studied in the past in the attempt to observe excitonic effects or the effect of valley selectivity. In the context of HHG, Liu et al. [1] have shown that even and odd harmonics could be generated from molybdenum disulfide (MoS_2) monolayers. The intensity and the polarization of the harmonics depend on the angle between the the polarization of the laser used for the generation and the symmetry axes of the crystal. This evolution is periodic and repeats every 60 degrees.

In this work, we made use of the anisotropic response of MoS2 to realize spatial mode conversions. On one hand we observe that, monocrystaline MoS2 monolayers behave as a polarization converter on even harmonics (4th harmonic), while they conserve the generation beam's OAM. On the other hand, we show that polycrystaline MoS2 monolayers can behave as a phase mask, which effectively modifies the OAM of the generated beam. This work illustrates the impact of local crystal symmetries during the strong field interaction and the potential of HHG from solids to generate structured light.

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Attosecond transient reflectivity and absorption from an Ytterbium laser driven High Harmonic Generation Source

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Synopsis We present Attosecond Transient Reflectivity Spectroscopy (ATRS) setup driven by Yb- doped laser, offering enhanced stability, higher repetition rate, and improved signal to noise ratio (SNR). Preliminary MgO measurements confirm its potential for attosecond-resolved reflectivity studies in condensed matter systems.

Over the past decade, attosecond transient absorption (ATA) and reflection spectroscopy (ATRS) have become essential techniques for studying ultrafast charge carrier dynamics, excitonic effects, and electron correlations of condensed matter systems[1-3]. Traditional ATRS setups rely on attosecond XUV pulses generated via high-harmonic generation (HHG) in gases, typically driven by Ti:Sapphire laser sources. Recently, new generations of XUV sources based on Yb laser sources have emerged, offering higher repetition rates, better stability, spatial quality and robustness. However, these systems have yet to be adapted in ATAS/ATRS to fully leverage their potential.

To address these challenges, this work presents a Yb-doped femtosecond laser-based ATRS setup. The laser system operates at 1030 nm, delivering 4 mJ, 430 fs pulses, CEP stable at 5 kHz which are subsequently compressed to 5 fs via a multi-pass cell (MPC) and hollow-core fiber setup, achieving an output energy of 1.5–2 mJ . Pulse characterization is performed using D-scan measurements.

We have conducted preliminary attosecond-resolved reflectivity measurements on MgO using a 1030 nm pump pulse, with spectral calibration performed via absorption in helium. Core-excitons corresponding to the $L_{2,3}$-edge in MgO were observed, along with a 2ω periodicity in the time-dependent signal, as shown in Figure 1. This is consistent with previous experiments using an 800 nm pump [2] and demonstrates the attosecond capability of the setup.

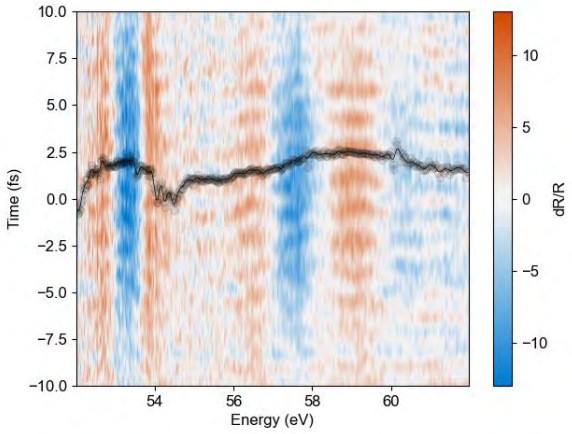


Figure 1: Preliminary data showing the transient reflectivity trace of magnesium oxide. The black curve shows the phase of the 2ω oscillating component.

The new setup provides more stability, high average power and higher repetition rate which can significantly improve SNR, demonstrating its advantages over Ti:Sapphire driven ATRS setups.

We will present detailed characterization of the XUV source stability, and assess improvements in transient reflectivity sensitivity enabled by this new laser system.

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Two-color high harmonic spectroscopy of ultrafast Dirac electron dynamics

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Synopsis We report the first observation of high harmonic generation (HHG) from highly oriented pyrolytic graphite (HOPG). Two-color HHG-spectroscopy reveals strong intraband-interband current coupling in the gapless Dirac cone, providing a novel all-optical probe of sub-laser-cycle carrier dynamics under strong fields.

Dirac materials with their linear energy-momentum dispersion near the Dirac points are ideal platforms for studying strong-field nonlinear optics and attosecond dynamics in two-dimensional systems, as well as material topology and electronic correlations. The gapless band structure allows electrons to be efficiently accelerated by strong laser fields without bandgap restrictions, enabling high-harmonic generation (HHG) driven by THz and mid-infrared light [1]. This unique electronic landscape with massless Dirace fermions opens new avenues for exploring unconventional HHG that differs significantly from bulk solid-state systems [2]. However, gapless 2D materials like graphene typically have a low damage threshold, making experiments highly challenging.

In our work, we show that highly oriented pyrolytic graphite (HOPG), a synthetic form of graphite, is a promising alternative experimental platform. Its ordered stacking of graphene layers preserves many of the remarkable electronic properties of graphene, including high electron mobility, and a vanishing bandgap at the Fermi energy, while offering structural robustness. In our experimental and theoretical study, we demonstrate ω - 2ω two-color spectroscopy as a powerful technique to study HHG in HOPG, leveraging its unique electronic structure to probe novel aspects of HHG beyond those observed in gapped solids.

Our setup combines a 2-μm laser pulse (60 fs duration) with an orthogonally polarized second harmonic pulse (10% relative intensity). Nonperturbative harmonics were generated from an HOPG surface in a reflection geometry. We measure the harmonic intensity as a function of the relative time delay Δt between the two pulses. Strikingly, the maximum signal of the 4th harmonic occurs ~20 fs before the maximal temporal overlap of the two pulses (Fig. 1(a)). Our numerical analysis indicates that this shift originates from the strong coupling between intraband and interband currents in the gapless Dirac cone. Unlike gapped semiconductors, where interband polarization primarily governs above-bandgap harmonics, the absence of a bandgap in HOPG allows significant electronic excitation. Consequently, intraband population dynamics strongly influence HHG. A comparative measurement of the 6th harmonic in ZnO exhibits a symmetric delay dependence, with the maximum signal occurring at perfect pulse overlap. The shift in the peak harmonic signal for HOPG thus serves as a key indicator of the strong coupling between interband and intraband currents.

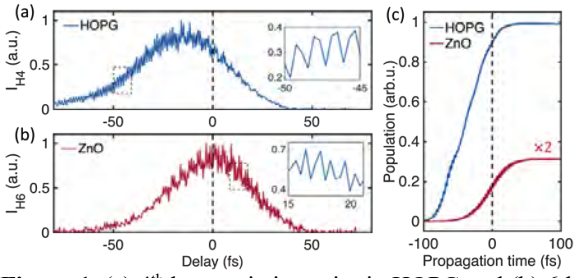


Figure 1. (a) 4th harmonic intensity in HOPG and (b) 6th harmonic in ZnO as a function of two-color delay. (c) Simulated population dynamics in HOPG and ZnO.

We also find that HOPG exhibits a highly asymmetric two-color modulation contrast -as the delay decreases, the modulation depth gradually decreases since the laser encounters a reduced number of pre-excited carriers. Simulated population dynamics for HOPG comfirm the asymmetric behavior (Fig.1(c)). The correlation between harmonic modulation depth and excitation provides a powerful all-optical method for studying population dynamics in Dirac systems under strong-field conditions.

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Electron–ion covariance spectrometry for molecular dynamics

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Synopsis We present a covariance spectrometer for electron velocity map imaging together with high-resolution mass spectra. Results are shown for different molecular samples investigated using few-fs UV, attosecond XUV or few-cycle NIR pulses.

One of the goals of attochemistry is to understand the role of ultrafast electronic processes triggered by the absorption of light for photochemical reactions. Excitation to a non-stationary state by an attosecond or few-femtosecond pulse enables time-resolved probing of molecular dynamics with methods such as absorption-, photoelectron- and mass spectrometry. The former two may give information about the state when the probe was absorbed, while the mass spectrum reveals the final state of charge localization and broken bonds.

Here we present the capabilities of a two-sided spectrometer (Fig. 1a) that combines 2D velocity map imaging (VMI) of electrons with a reflectron-type mass spectrometer[1], intended for covariance analysis. For this purpose, we record not just the average spectra but single-shot data and analyse the correlations between photoelectrons and fragment ions. Conceptually this allows one to filter the electron spectrum based on the mass spectrum, as briefly demonstrated in Figure 1b–c with the fragmentation of acetone by UV-excitation and NIR-photoionization.

The VMI settings are typically chosen to accept electrons up to 8 eV or 33 eV depending on whether UV or XUV radiation is used. Simulations show reflectron mass resolutions from 150 u for 2 eV dissociation fragments to 1500 u for room-temperature samples. Alternatively, cations can be sent to the VMI and mass-specific ion velocity map images be extracted from TOF–image-covariance analysis (analogously to a 3D-detection system). For ions, we show kinetic energies in the meV energy range (accepting up to 3 eV).

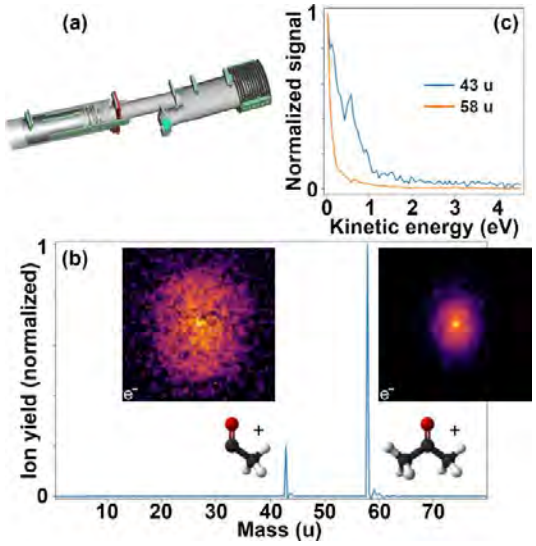


Figure 1. (a) Overview of the spectrometer design. (b) Mass spectrum of acetone. (b-insets & c) Electron velocity map images covariance-filtered by whether acetone or its 43 u fragment ion was detected.

The spectrometer is equipped with injection systems for gas- and vapour-phase samples and has already been used in several few-femtosecond time-resolved studies of UV-excited dynamics in neutral molecules [2, 3]. We also intend to combine this versatile apparatus with UV and XUV beamlines operating at up to 200 kHz and for instance study UV-induced electron dynamics in molecules solvated in water clusters.

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Control of Graphene by Two-Colour Laser Fields

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Synopsis We present a rigorous analytical description allowing to understand how to control valleytronics of graphene interacting with bichromatic fields with tailored polarisations. Our study provides a complete understanding of how varying the amplitude, phase difference and polarisation of ω and 2ω fields yields calculable varying current and valley polarisation, with potential applications to many 2-dimensional materials.

Control of valley polarisation and current within 2-dimensional materials on the attosecond scale have the undeniable potential to revolutionise our future of material engineering. Recent experiments have shown exciting results for the quantum-material graphene [1], in which two inequivalent valleys are formed in the band structure, accessible and controllable by light pulses in the form of bichromatic fields.

Although demonstrations have proven this to be an effective method of manipulating valleytronic configurations [2], the underlying vital mechanism of this process is yet to be understood, and only by diagnosing these systems with analytical methods can we access the heart of valleytronics which is to obtain control of valley selections on sub-femtosecond time scales.

In this work we model graphene as a Floquet topological insulator, through the Taylor series second expansion around the K points of the Haldane model which provides us with a way to break sublattice inversion symmetry and time-reversal symmetry in order to create a bulk band gap. This edge state is necessary for measuring an interband electronic current. With the main driving force behind this working coming from Floquet theory [3], we are able to analyse not just fields of varying amplitude, but varying phase difference and polarisation, allowing for true transparency of ultrafast electron dynamics in graphene. A beautiful advantage of Floquet theory is the ability to not just observe the valence and conduction bands, but the all the possible individual contributions that the bands have with the ω and 2ω fields.

This work provides a foundational framework for analytical control that can be extended to other 2-dimensional materials for which valleytronics triumphs. The resulting working will be made publically available as an open-source Mathematica notebook.

The relationship of attosecond physics with 2-dimensional quantum-materials is ready to take the next step, with the ultrafast control of irradiated graphene being a proposal to a long lasting marriage between the two fields.

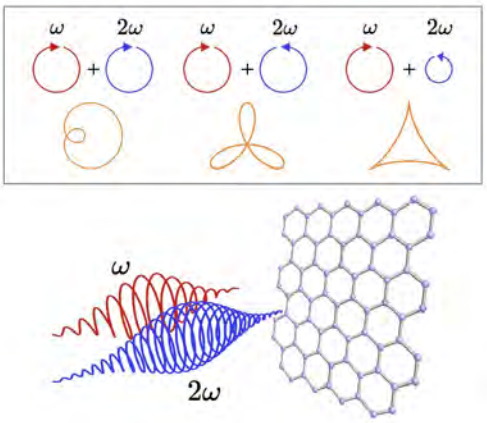


Figure 1. Above shows a few options of bichromatic fields with different symmetry that reflect the geometrically triangle symmetry between the K and K' points in the Brillouin zone of graphene. Below, a sketch to visualise this process.

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A nonlinear optical chirality for high-harmonic generation

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Synopsis The quantification of the chirality of a light field is a long-standing problem in optics, with existing solutions capturing both linear-optical interactions (through the optical chirality and helicity) as well as few-photon perturbative nonlinear processes, through nonlinear correlation functions. However, an optical chirality that captures nonperturbative dynamics such as HHG remains lacking. Here we present such a nonperturbative optical chirality, built through a suitable orientation averaging of the standard SFA integral, which fully reflects the universal features of chiral HHG dynamics.

Chiral light-matter interactions hold enormous potential, as they allow us to access the full scope of chemistry – without restriction to the small set of symmetric achiral molecules – using photo-driven processes. In order for light-matter interactions to be chirally sensitive, the light itself needs to be chiral. Thus, the quantification of the chirality of an optical field has been a long-standing goal.

For linear optics, optical chirality [1] and optical helicity [2] capture all chiral interactions, and have been the subject of intensive study, including their conservation properties, optical singularities, and interactions with matter.

For nonlinear optics, the discovery of giant enantiosensitivity in HHG [3] has spurred an explosion in applications of synthetic chiral light (SCL) – multi-colour combinations whose electric-field Lissajous figures are chiral over time – to drive chiral processes. It has also prompted the creation of nonlinear chiral field correlation functions, denoted $h^{(n)}$ [3], which encode the chirality of the light field in a way which captures the dynamics involved in few-photon nonlinear processes.

However, the existing nonlinear measures of field chirality are only directly applicable to few-photon nonlinear processes of perturbative nature, and, as such, they do not have any direct bearing on HHG or other nonperturbative processes. This raises the need, therefore, of a nonlinear optical chirality which does capture the nonperturbative dynamics involved in HHG and its sibling processes, and which can explain the mechanisms that underlie HHG driven by SCL.

Here we present such a nonlinear optical chirality, through a suitable orientation-averaging procedure applied to the universal SFA expression for HHG [4]. This orientation averaging respects the usual form of SFA, with a new prefactor, which fully captures the chiral (and achiral) geometric and structural responses of the molecule to the dynamical forcing provided by the driving field. This reveals the dynamical mechanisms that underlie enantiosensitive HHG, and allows clean and powerful ways to tailor chiral fields in order to maximize their chiral selectivity.

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Enantiosensitive exceptional points of open chiral systems

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Synopsis We introduce the concept of enantiosensitive exceptional points – non-Hermitian degeneracies whose position in parameter space is sensitive to the handedness of an open system. By investigating different physical realizations of the concept, we display its generality and pave the way for novel topological methods for the detection of molecular chirality based on non-Hermitian physics.

Non-Hermitian (NH) Hamiltonians describe open systems, where energy and/or matter is exchanged between the system and its surrounding environment. Exceptional points (EPs) are branch points in the parameter space at which the complex eigenvalues of the Hamiltonian become degenerate and the corresponding eigenstates coalesce, separating the parity-time ($\mathcal{PT}$) symmetric and $\mathcal{PT}$-broken phases of the system. The presence of EPs in NH physics leads to a plethora of remarkable phenomena observed in a number of different experimental platforms, from atomic and molecular media to photonic and optical platforms [1].

Here, we study the influence and role EPs play in chiral ($\mathcal{P}$-breaking) NH systems. Specifically, we show that the position of EPs of open chiral systems is naturally enantiosensitive, is highly tunable, and can be engineered to enhance the detection of chiral molecules. Displaying the generality of our concept, we investigate different physical realizations, paving the way for novel chiral sensitive methods relying on EPs and NH physics [2].

First, we show that a three-color chiral field can induce enantiosensitive topological population transfer between adiabatic states of a chiral molecule, both in the adiabatic as well as non-adiabatic regime. Then, we show how EPs can strongly enhance typically weak beyond-dipole chiral effects that rely on the interference between electric and magnetic dipole transitions, leading to large differences in residual population of dissociating chiral molecules subject to circularly polarized fields. Finally, we consider the interaction of a birefringent twisted optical fiber with a solution of chiral molecules, where the typical NH sub-linear scaling of the response to an external perturbation, a role played here by the enantiomeric excess of the solution, paves the way for novel ultrasensitive chiral sensing approaches.

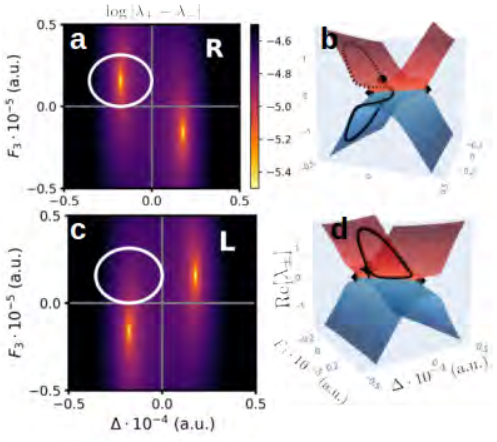


Figure 1. a,c) Absolute difference of the eigenvalues of an open chiral molecule coupled by a three-color laser field for right-handed (a) and left-handed (b) enantiomers. The white line indicates the path in parameter space enclosing the EP. b,d) Adiabatic evolution on the path for right- (b) and left-handed (c) molecules, showing the enantiosensitive topological population transfer.

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Topological valley control of graphene by tailored two-colour fields using a light and fast SBE solver

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Synopsis Our work focuses on ultrafast electron dynamics in graphene under a bichromatic laser field. We develop an open-source code solving the semiconductor Bloch equations (SBE) with a Haldane Hamiltonian and Lindbladian dephasing. We benchmark our results with previously published data, showing tight control of graphene valley polarisation, current, and harmonic generation by tuning the relative phase of structured fields with the symmetry of the underlying lattice. Next steps include expanding the SBE code to incorporate orbital angular momentum beams for spatial control over topological phases.

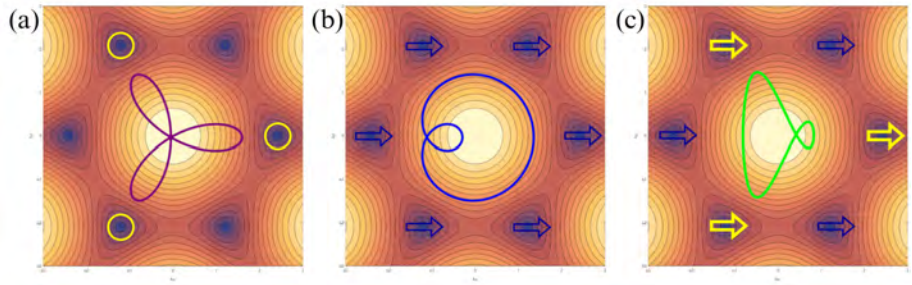


Figure 1: Schematics for the graphene bandgap behaviour around K points: (a) valley polarisation due to a counterrotating $\omega - 2\omega$ field, (b) current due to a corotating $\omega - 2\omega$ field, and (c) valley current due to circular ω and linear 2ω field.

Work in graphene, a single-atom-thick layer of carbon atoms arranged in a honeycomb lattice, has led to significant breakthroughs in the field of valleytronics. This is due to graphene's excellent electronic transport properties, unique topological characteristics, and the presence of an additional degree of freedom – the valley pseudospin, opening up the possibility to encode information via population imbalances at the Dirac points.

We develop a Python-based open-source software package and demonstrate its benchmarking with the findings in [1, 2]: these demonstrated that despite having zero bandgap and zero Berry curvature (properties previously considered essential for valley selection) it is possible to induce a current and valley polarisation in graphene-like materials by using ultrafast structured light [Fig.1]. Depending on the symmetry and orientation of the lattice under different driving field configurations, we can control valley-selective excitations and dynamics by scanning over the relative phase and polarisation using bichromatic ω and 2ω fields. We solve the semiconductor Bloch equations (SBE) for a Haldane Hamiltonian within the tight-binding approximation, and we model dephasing with a Lindbladian. We calculate the dynamics of the density matrix to extract the relevant observables, i.e. valley population and current, and compare our results with [2], which also uses the SBE approach but treats the dephasing phenomenologically, as well as to TDDFT and experimental results from [1].

Next steps include expanding the SBE code to account for a new degree of freedom by adding the spatial profile of vortex beams. The aim is to control different induced topological phases using spatial variations in the driving light.

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Competition between proton transfer and resonant intermolecular Coulombic decay in water dimers

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Synopsis Recent experiments on H_2O and D_2O dimers showed that the resonant ICD process is affected by the nuclear dynamics following excitation. The simultaneous opening of the proton (or deuterium) transfer channel makes the two processes enter in competition, causing the ICD lifetime to change significantly between $(H_2O)_2$ and $(D_2O)_2$, with the latter being shorter. The slower motion of the deuterium allows the heavy water dimer to pass more time in a higher decay region, ensuring a faster ICD. We carried out nuclear dynamics simulations to explore the competition between these two ultrafast processes.

Intermolecular Coulombic decay (ICD) is an ubiquitous, electron-correlation driven relaxation process in weakly bound systems, including liquid water. First proposed in 1997 by Cederbaum *et al.* [1], ICD has been proven to often dominate over other relaxation channels [2]. In ICD, an inner valence hole created upon ionization is refilled by an outer valence electron and the excess energy is released by ejection of a second electron from the neighbouring unit. When the process is triggered not by ionization but by a photoexcitation, it's defined as resonant ICD (RICD) [3]. Recent pump-probe experiments on RICD in water dimers [H. J. Wörner, private communication] showed that the process is substantially more efficient in deuterated water, hinting to the role of the proton transfer following excitation.

We analysed the potential energy curves of the initial and final RICD states of a water dimer along the proton transfer coordinate. The results show that the excitation of the proton donor water molecule can trigger a nuclear dynamics which slows down the RICD channel.

Using the multi-configurational time-dependent Hartree (MCTDH) [4] approach, with non-Hermitian Hamiltonians containing the decay rates of RICD obtained by the Fano-CI approach [5], we simulated the decay process initiated both by exciting the proton donor or the proton acceptor water molecule. The nuclear dynamics simulations shed light on the competition between RICD and the fast proton transfer, both occurring on a femtosecond timescale.

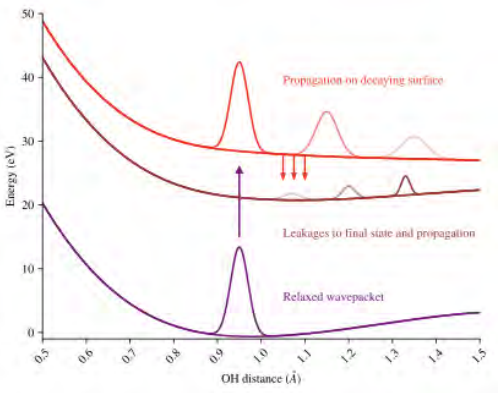


Figure 1. Three-states model for the nuclear dynamics simulation: the initial wavepacket, relaxed on the electronic ground state (purple curve), is transferred vertically to an averaged decaying state (red curve, built from the band of coupled inner-valence excited states of the water dimer, excited on the proton donating molecule) and propagated with a non-Hermitian Hamiltonian containing the RICD decay rates. This way population is transferred to the final ICD states (brown curve) to simulate the decay.

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Reconstruction and characterization of single particle entanglement dynamics in photoionization

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Synopsis We study signatures of entanglement between energy and emission angle in time-resolved atomic photoionization. We illustrate our analytical results on numerical simulations and available experimental measurements. We investigate how this entanglement manifests as decoherence in partially resolved measurements and study the implications for their time domain interpretation. Finaly, we develop an adapted entanglement witness, based on Bell inequalites, pushing further the link between attosecond and quantum optics.

Attosecond time-resolved spectroscopy allows the observation in real time of photoemission driven by the correlated interactions between the photoelectron and its parent ion, using experiments that are essentially based on interferometric schemes. In their common implementations, one assumes coherent processes, in particular at the level of the pump/probe light sources but also of the studied phenomena.

Here, using simulations and revisiting experimental results published in [1], we investigate how entanglement between the photoelectron energy E and emission angle θ builds up during photoemission, in the case of resonant and near-resonant 2-photon transitions in He, see Fig. 1(a).

We first show how this entanglement manifests as a loss of coherence when only E is resolved in the measurements [2]. This may prevent a direct reconstruction of the photoelectron dynamics out of spectral amplitudes infered from standard interferometric measurements (e.g. RABBIT [3]) *without θ resolution*. We thus apply an original method that we developed to circumvent this limitation in a very general way [4], and show that a proper analysis of the photoelectron's asymptotic *reduced density matrix* does give complete access to its dynamics.

In addition, we show that the system can be created and detected in a near maximally entangled state, violating Bell's inequalities, using the Clauser–Horne–Shimony–Holt (CHSH) test [5], see Fig. 1(b). To proceed a step further, we propose a robust entanglement witness opening the way to unambiguously assess the entanglement of total density matrices of statisticals mixtures in this context. This witness was tested in situations where we took into account classical decoherences inherent to, e.g., a limited experimental resolution.

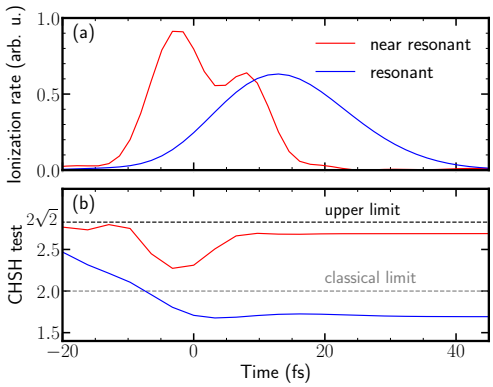


Figure 1. Reconstruction of ionization rate (a) and Bell test for the (E, θ) entanglement (b) on experimental time-resolved near-resonant (red) and resonant (blue) 2-photon ionization of He.

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Attosecond Resolved Intramolecular Charge Transfer in Neutral Donor-Acceptor Organic Molecules

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Synopsis We simulate the early steps of ultrafast charge dynamics induced by a broadband UV pulse in donor-acceptor organic molecules and show how to probe it with XUV attosecond pulses.

Since the first models of organic solar cells were proposed more than 40 years ago, the search for new materials with the ability to produce a charge separation after interaction with UV and visible light, necessary for photovoltaic applications, has continuously kept the attention of the scientific community. Organic photovoltaic devices usually achieve charge transfer by using molecular complexes containing electron donor and acceptor groups. To capture the time evolution of this process at the early stages, i.e., during the first few tens of femtoseconds, ideally attosecond time-resolution is required.

In this work, we have theoretically simulated a pump-probe scheme consisting of a broadband few-fs UV pump pulse (currently available in several laboratories [1, 2]) and an XUV attosecond probe pulse to investigate the early stages of charge transfer in the donor-acceptor molecules para-nitroaniline (PNA) and meta-nitroaniline (MNA). Our theoretical approach describes (i) the coherent excitation of the electronic states of the molecules and (ii) the ensuing coupled electron and nuclear dynamics resulting from the initial coherent superposition. It also provides (iii) time-resolved photoelectron spectra as those expected to be produced in the very near future in several attosecond laboratories. Excitation in (i) is described by solving the time-dependent Schrödinger in a basis of fully correlated electronic states of the neutral molecule. The couple electron and nuclear dynamics that follows in step (ii) is simulated by using an extension of the trajectory surface hoping method that has been developed for the occasion to account for the coherent superposition of electronic states produced in step (i) (which is not possible by using currently available methods based on classical trajectories) and the non adiabatic transitions that can take place during the process (e.g., through conical intersections). Finally, ionization by the probe pulse in (iii) is described by using an extension of the static-exchange RAS-DFT approach that provides a similarly accurate description of the remaining molecular cation and explicitly accounts for the ejected electron by solving the appropriate scattering equations within the static exchange and DFT approximations. We will show how the charge dynamics ignited by the broadband UV pulse is imprinted in the calculated time-resolved photoelectron spectrum and provide a few guidelines for future experimental effort in this direction.

As an example, Fig. 1 shows the evolution of the permanent dipole of MNA resulting from excitation with a 4 fs UV pulse with central frequency 5 eV. Pronounced charge oscillations can be observed during the first 40 fs.

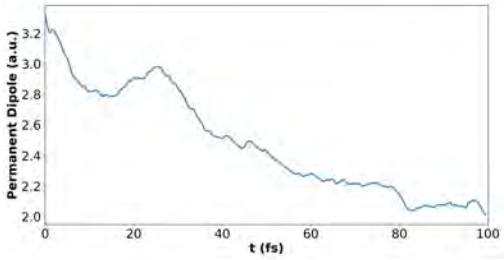


Figure 1. Evolution of the permanent dipole of MNA resulting from excitation with a 4 fs UV pulse with central frequency 5 eV

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Relativistic core-level molecular photoionization in heavy molecules

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Synopsis A relativistic methodology capable to describe core-level molecular photoionization in heavy molecules is presented. The exploration of a series of iodine molecules allows us to investigate the chemical and the structural information encoded in different angle- and spin-resolved photoionization observables.

Recent developments in Synchrotron and Free Electron Laser facilities have enabled the investigation of light-matter interaction processes in new energy and time domains. The unprecedented control of such powerful radiations has also opened the possibility of designing interferometers aiming at exploring ultrafast electron dynamics induced by photoionization [1]. One of the driving forces has been the possibility of probing the electron dynamics locally inside the molecular targets.

Currently, it is possible to measure the so-called core-level molecular photoionization time-delays (MPTDs), giving access to local information about the chemical and structural composition of the molecular target. However, up to now, and to the best of our knowledge, the investigation of core-level MPTDs is largely limited to molecules containing "light" atoms such as C, O and N [2], although, molecules containing heavy atoms are preferred in many experiments due to their large absorption yields compared to molecules containing only light atoms [3].

From a theoretical point of view, the correct description of the core-level MPTDs in molecules containing heavy atoms requires the description of the molecular photoionization process within a relativistic treatment. The first step towards the development of such relativistic methodology consists in solving the molecular Dirac equation. However, this is not a trivial task. In the present work, we propose an approach that adapts existing non-relativistic methods for molecular photoionization by incorporating a stable numerical solution of the two-center Dirac equation in the continuum that avoids the appearance of "spurious states" common in other relativistic implementations [4].

For the time being, the method presented here allows one to investigate photoionization of heavy diatomic molecules such as HI, LiI, NaI, IF, ICl, IBr, etc., using an effective relativistic DFT potential. This approximation allows us to explore angle- and spin-resolved photoionization observables such as cross sections, asymmetry β -parameters and the one-photon Wigner delays. Moreover, by investigating a series of iodine molecules, chemical and structural information can be quantified from the I 4d core-levels [5], *see* Table 1. Finally, the possibility of incorporating a second photon in our calculation is discussed in order to describe pump-probe scenarios aiming at providing photoionization time-delays [6].

Table 1. Spin-orbit and ligand-field splittings of the I 4d core-levels in IBr molecule (in atomic units) computed with the reference code DIRAC and with our methodology "RTDmol".

Term	DIRAC	RTDmol	diff.
Π1/2	1.911749	1.912206	2.4E-04
Δ3/2	1.895999	1.896025	1.4E-05
Σ1/2	1.849793	1.849965	9.2E-05
Π3/2	1.841600	1.841509	4.9E-05
Δ5/2	1.828402	1.827960	2.4E-04

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Observation of nonadiabatic coupling induced by Floquet-Bloch states in high harmonic generation from large-bandgap materials

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Synopsis Nonadiabatic coupling dynamics arise in semiconductors when strong-field light-matter interaction involves multiple electronic bands. Here, we propose a scheme to observe nonadiabatic electronic dynamics associated with the emergence of the Floquet-Bloch states (FBS) using high harmonic spectroscopy on a magnesium oxide (MgO) monocrystal. Our approach provides a first observation of the nonadiabatic coupling between the first conduction band (CB1) and the lower FBS of the second conduction band (CB2) in MgO. Via this nonadiabatic coupling, we access the morphology of the bandgap encoded directly into the high harmonic spectrum.

We investigate the influence of the nonadiabatic coupling between the solid's electronic bands and their FBSs [1, 2] in large-bandgap materials by applying HHG spectroscopy. In the experiment, we employ a strong ultrashort infrared laser pulse to produce HHG around the second plateau area below the damage threshold of the crystal. We demonstrate that high-order harmonic 12 (HH12) and HH13 can perfectly probe the nonadiabatic coupling process between the FBS and conduction band at the edge of the Brillouin zone, where the bandgap between the first few conduction bands is smaller than one laser photon energy. By rotating the crystal, we map the morphology of the bandgap along the MgO's Γ-X and Γ-K directions, as shown in Fig.1. This mapping is demonstrated by a distinct arc-like structure of HH12 in the HHG spectrum, which is consistent with our time-dependent Schrödinger equation simulation in MgO under the experimental condition [3]. Based on these findings, we propose a new quantum path scenario for HHG that includes nonadiabatic coupling mediated by the FBS at the edges of the Brillouin zone. Our results pave the way for resolving nonadiabatic coupling in a complex quantum system, and for reconstructing the band morphology, thereby opening new avenues in HHG spectroscopy in solid-state physics.

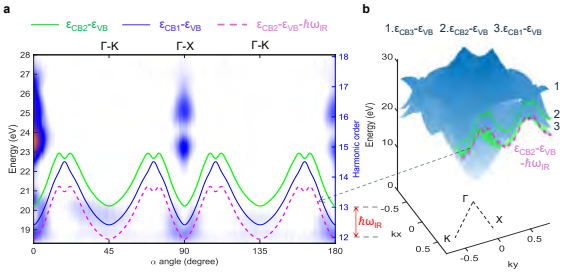


Figure 1. (a) Experimental HHG spectrum as a function of the crystal orientation. Calculated bandgap curves at the edge of the Brillouin zone: $\epsilon_{CB2} - \epsilon_{VB}$ (green) and $\epsilon_{CB1} - \epsilon_{VB}$ (blue). Corresponding energy gap for FBS: $\epsilon_{CB2} - \epsilon_{VB} - \hbar\omega_{IR}$ (purple). **(b)** Calculated 3D bandgap structure of MgO, highlighting the same bandgap curves at the edge of the Brillouin zone.

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Towards attosecond photoemission electron microscopy: Spatially-resolved strong-field electron acceleration from a nanostructure

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Synopsis Photoemission electron microscopy (PEEM) offers the possibility to study ultrafast charge carrier dynamics in single nanostructures. While attosecond pulses can enhance this technique with the highest temporal resolution, the combination turns out to be challenging. Here, we observe field-driven electron emission and acceleration from well-defined regions of a nanoantenna in PEEM by using a long-wavelength, few-cycle driving field at a high repetition rate. This is probably the most important prerequisite for attosecond time-resolved photoemission experiments in nano-localized near-fields.

The non-equilibrium dynamics of charge carriers at interfaces and in application-relevant nanoscale devices after optical excitation remains elusive, as methods with combined attosecond to femtosecond temporal resolution and few nanometer spatial resolution are still under development. Attosecond photoemission electron microscopy (PEEM) is a promising idea [1], but despite recent progress [2] is still not realised [3]. Here, we show how to circumvent the so-called space charge problem, which is central to maintaining a high spatial, angular and energy resolution in a time-resolved experiment.

Our setup consists of a home-built optical parametric amplifier system that delivers CEP-stable few-cycle pulses at a central wavelength of 2 µm at a repetition rate of 200 kHz with sufficient pulse energy to generate high-order harmonics. A small fraction of the light pulses is focused onto a monocrystalline gold flake containing a double nanohole structure with an apex radius of 9 nm. The pulses lead to photoemission of electrons which are recorded with our time-of-flight PEEM. We observe a distinct change in the energy and emission angle of the photoelectrons after leaving the near-field region of the sample (Fig. 1d). We attribute this to a light-field driven acceleration, which has not been observed before in photoemission electron microscopy. In a next step, we will combine our results in a time-resolved experiment using attosecond pulses to record transient near-fields of nanostructures after ultrafast optical excitation.

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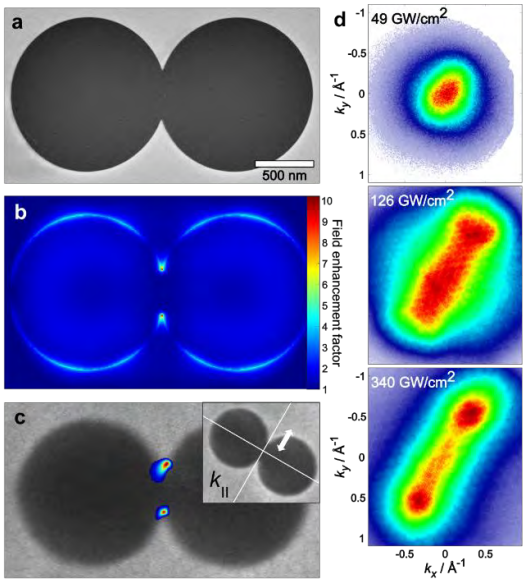


Figure 1. Field-driven photoelectron emission from an isolated nanostructure. (a) Scanning electron micrograph of the nanoantenna milled into a monocrystalline Au flake. (b) Numerical simulation of the local field enhancement upon optical excitation at a wavelength of 2 µm. (c) Combined PEEM image of laser-driven emission (color) and Hg lamp driven emission (gray). Inset: Rotation of the sample for the k-space investigation in d. (d) Laser-driven photoemission with different intensities from the nanostructure. The emission is recorded in momentum space, capturing the acceleration of photoelectrons with increasing light intensity.

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Ultrafast ethylene dication formation through resonant enhanced multiphoton ionization

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Synopsis We study the non-adiabatic evolution of the ethylene cation, generated by the absorption of an XUV-pump pulse, by monitoring the dication formation induced by a delayed NIR pulse. The experiment reveals a prominent peak in the dication yield around 15 fs delay. We combine surface-hopping methods with *ab initio* simulations of the ion's resonantly-enhanced multiphoton ionization (REMPI), using ASTRA, a new close-coupling molecular-ionization code. The peak results from the increase in the REMPI signal, as the C=C bond elongates, combined with population depletion due to non-adiabatic relaxation.

Ultrashort extreme-ultraviolet (XUV) pulses used in conjunction with near infrared (NIR) probe pulses allow us to monitor complex molecular dynamics in real time, e.g., to study non-adiabatic relaxation processes of molecular ions relevant in astrophysics [1].

We present a theoretical study of the ethylene dication formation using an attosecond-XUV-train pump/NIR probe excitation scheme, aimed at reproducing recent experiments. The broad-band pump ionizes the neutral molecule to several cationic states (D0-D4), triggering a non-adiabatic nuclear dynamics. The delayed NIR promotes the subsequent multi-photon ionization to produce the dication. We employ advanced trajectory surface hopping calculations to obtain the time-dependent D0-D4 populations and structural rearrangements [2], and subsequently compute the average multi-photon ionization yield from those cationic states by solving the time-dependent Schrödinger equation (TDSE) in presence of the NIR pulse. To solve the TDSE we use ASTRA, a new implementation of the close-coupling method designed to reproduce molecular photoionization observables from the interaction with ultrashort ionizing pulses [3]. The simulations for multiple NIR intensities confirm the experimentally observed peak in the dication yield in the 10-20 fs time delay region [4]. The main contributions to the dication signal comes from the D1 and D2 cations: while their REMPI ionization rate is enhanced by the C=C bond elongation, their population is rapidly depleted by non-adiabatic relaxation processes, resulting in the observed peak. We acknowledge s

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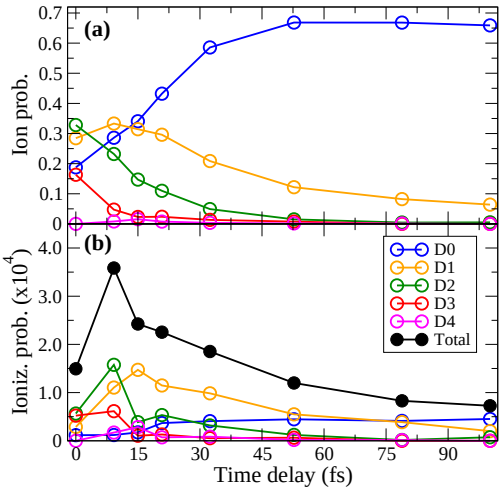


Figure 1. Population evolution of ethylene cations (a) and ionization yield at 7 TW/cm^2 (b).

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Adapting polarization gating for generation of isolated attosecond pulses after multiple mirror reflexions

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Synopsis Polarization gating relies on using pulses with time-dependent polarization. It is very sensitive to optical elements such as mirrors or beam splitters that can significantly modify the polarization. We show that polarization gating can be used despite multiple reflections on 45° mirrors. We describe how to measure and compensate for polarization-dependent effects. After pre-compensation, polarization gating of CEP (carrier-envelop phase) controlled few cycle pulses leads to a strong evolution of the harmonic spectrum with the CEP compatible with the emission of isolated attosecond pulses.

Polarization gating[1, 2] is a common approach for XUV emission confinement via high order harmonic generation (HHG) in gas that can lead to isolated attosecond pulse emission [3- 5]. It relies on modulating temporally the polarization of a driving pulse to ensure that its polarization is linear for a short period of time, close to half the optical period of the driver to achieve isolated attosecond pulse generation. As HHG in gases is usually maximum for linear polarization, modulating the polarization from elliptic to linear to elliptic temporally confines the XUV emission in a controllable time window.

Pulses with time-dependent polarization can be created with polarization sensitive interferometers or birefringent plates [2]. Propagation of these pulses to the medium where harmonics are generated can be problematic, especially when propagation involves many optical elements that are polarization-dependent like beam splitters or mirrors with non-normal incidence.

We used an experimental setup including 7 reflexions on metallic mirrors at 45° [6]. We measured the relative dephasing and delays induced by these mirrors between the S (vertical) and P (horizontal) polarizations. The observed delay, larger than 2 fs, is very significant when few-cycle pulses are used.

Without pre-compensation, applying a polarization gate before the 7 reflexions had very little impact on the XUV spectrum showing no signature of XUV confinement (Fig. 1. a) . With proper pre-compensation (Fig. 1. b) , we observed a strong impact of the gate on the spectrum that translates as a large sensitivity of the XUV spectrum on the CEP similar to [3]. For some CEP values, the harmonic spectrum in the cutoff region is continuous and shows broad harmonics after a $\pi/2$ CEP shift. These signatures are compatible with the emission of isolated attosecond pulses or pairs of pulses depending on the CEP [3], as expected with such short polarization gates when propagation effects can be neglected.

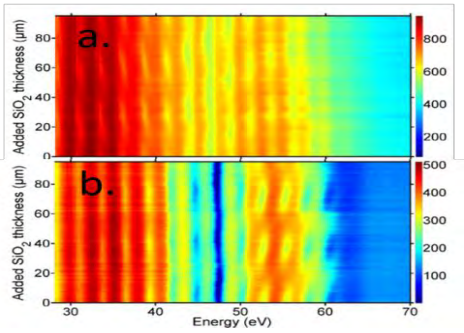


Figure 1. CEP dependence of the XUV spectra generated in Ar jet with a short polarization gated pulse: a). Without compensation of polarization dependent propagation. b). With propagation effects pre-compensation.

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High-order frequency mixing: harvesting bright XUV from propagation

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Synopsis High-harmonic generation (HHG) in gases became our trusty table-top source of coherent XUV light, however it possesses fundamental limitations for brightness. One of the main obstacles for harvesting brighter XUV from the longer propagation in media is the photoionisation, which causes the blue shift of the driver, which disturbs the coherent growth of XUV with propagation. We show, theoretically, that high-order frequency mixing, occurring when a strong driver is accompanied by a weak low-frequency one, produces much brighter XUV than HHG, gaining from the blue-shift compensation.

The generation of bright, coherent XUV light via frequency conversion of intense laser drivers is a problem of both fundamental and technological importance. Increasing the intensity of the generated high harmonics by raising the intensity of the driving field only works up to a point: at high intensities, rapid ionisation of the medium limits the conversion efficiency.

We identify the dominant limiting mechanism – the combined effect of phase matching and the blue shift of the driving field during its propagation through a rapidly ionising medium [1, 2]. We introduce the blue-shift length, which sets the upper bound for the quadratic intensity growth of the harmonics.

We study analytically and numerically (solving the propagation equation coupled with the TDSE) the behaviour of the macroscopic HHG signal with propagation distance. We show that its quadratic growth is limited by the shortest of three lengths: absorption, coherence, or blue-shift length. Thus, we define three regimes of HHG, corresponding to the dominant limiting mechanisms (see Fig.1(a)).

Moreover, we show that this seemingly fundamental restriction can be overcome by using an additional generating weak mid-IR field. For suitable combinations of frequencies of the generating fields, the corresponding high-order frequency-mixing (HFM) process does not suffer from the blue shift of the drivers and phase mismatch [1], and thus its efficiency grows quadratically with propagation (see Fig.1(b)).

Our results open a new route for highly efficient generation of XUV light, the first step of which has been taken already via an observation of high-order parametric generation [3]. Moreover, HFM offers new handles for XUV control such as the control of the carrier envelope phase of the emitted attosecond XUV pulses [4, 5].

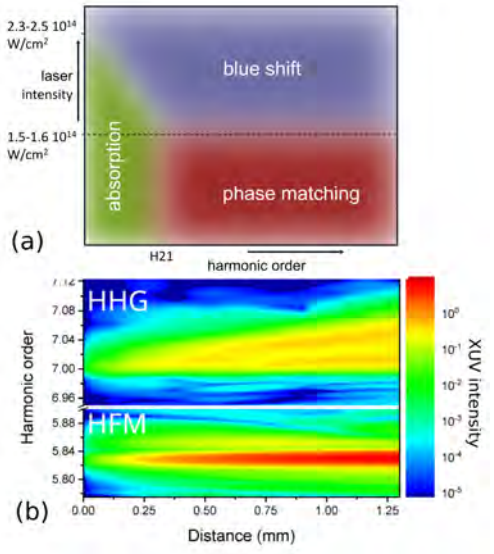


Figure 1. (a) Diagram showing dominant mechanisms for HHG in argon. (b) TDSE calculation for the propagation of HHG and HFM in argon for $2 \cdot 10^{14}$W/cm^2 of 330nm driver with weak 1980nm.

This work is done in tight collaboration with Vasily Strelkov.

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Observing coherent electronic wavepackets in molecules using attosecond transient absorption spectroscopy

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Synopsis Experimentally recorded attosecond transient absorption spectroscopy (ATAS) of molecules in the gas phase, featuring coherent electronic dynamics triggered by strong-field excitation will be presented. While iodomethane (CH_3I) is found to exhibit a characteristic beating that starts right after excitation, the observed interference pattern in aluminum bromide (Al_2Br_6) arises after a delay of 175 fs.

Electronic coherence has long been predicted to be a sensible property to achieve coherent control of chemical reactions [1]. One way to detect a coherent superposition between electronically excited states is ATAS [2]. In this contribution, our latest findings on two different molecular electronic coherences will be presented.

First, Figure 1a) shows how a strong-field-ionizing 5 fs pulse creates a coherent superposition of two cationic states separated by a spin-orbit splitting of 630 meV in CH_3I, probed with four transitions into two common 4d excited states of iodine [3]. In Figure 1b), the interference pattern of these transitions is shown, which does not appear to decay up to a delay of 80 fs.

Second, first transient absorption data of Al_2Br_6 in the gas phase will be presented. As shown in Figure 1c), similar oscillations in the magnitude of the transient absorption signal are apparent. Figure 1d) highlights that the delay-dependent interference pattern only becomes prominent after 175 fs, which roughly aligns with the appearance of new sharp lines at 64.3 and 65 eV, assigned to neutral bromine atoms. We preliminarily assign the oscillatory features to probing a coherent superposition of excited electronic states after bifurcation of the vibronic wavepacket. To our knowledge, this is the first time such a feature has been observed experimentally, possibly providing a view onto non-Born-Oppenheimer dynamics during a chemical reaction in quantum-mechanical detail.

While the periods of the oscillations in both molecules appear to be on the same order of magnitude, in one case the interferences are present immediately after the interaction with the electric field, whereas in the other the interference pattern only becomes observable after molecular dynamics have taken place. This shows that observing interferences of the coherent electronic wavepacket in common final states can be preceded by nuclear motion, leading to different delay-dependent signatures.

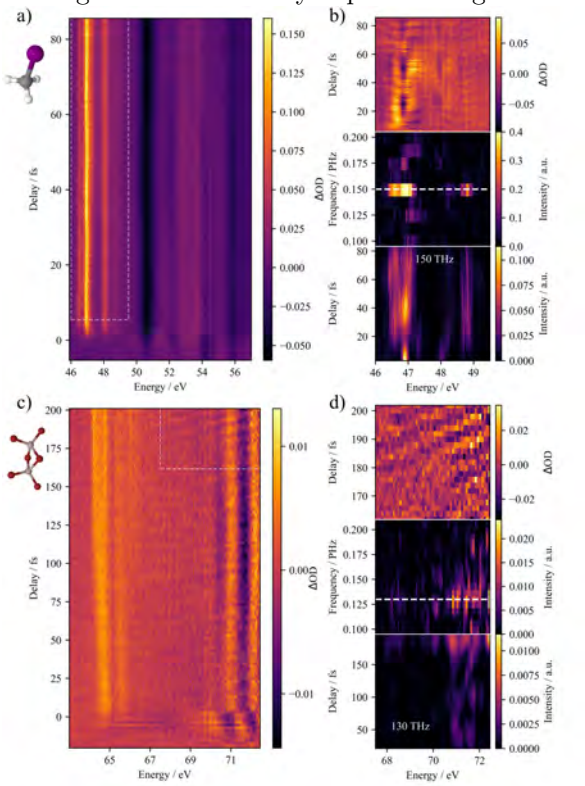


Figure 1. Attosecond transient absorption data of a) CH_3I and c) Al_2Br_6. Zoomed-in transient absorption map after subtraction of the delay-mean, its fast Fourier transform along the delay axis and a time-frequency analysis at a frequency of b) 150 THz and d) 130 THz, respectively.

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eTraj.jl: Trajectory-Based Simulation for Strong-Field Ionization

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Synopsis We present a general, efficient, flexible, and out-of-the-box solution for trajectory-based simulation program named after `eTraj.jl` using the Julia programming language.

The dynamics of light-matter interactions in the realm of strong-field ionization has been a focal point and has attracted widespread interest. We present the `eTraj.jl` program package, designed to implement established classical/semiclassical trajectory-based methods to determine the photoelectron momentum distribution resulting from strong-field ionization of both atoms and molecules. The program operates within a unified theoretical framework that separates the trajectory-based computation into two stages: initial-condition preparation and trajectory evolution. For initial-condition preparation, we provide several methods, including the Strong-Field Approximation with Saddle-Point Approximation (SFA-SPA), SFA-SPA with Non-adiabatic Expansion (SFA-SPANE), and the Ammosov-Delone-Krainov theory (ADK), with atomic and molecular variants, as well as the Weak-Field Asymptotic Theory (WFAT) for molecules. For trajectory evolution, available options are Classical Trajectory Monte-Carlo (CTMC), which employs purely classical electron trajectories, and the Quantum Trajectory Monte-Carlo (QTMC) and Semi-Classical Two-Step model (SCTS), which include the quantum phase during trajectory evolution. The program is a versatile, efficient, flexible, and out-of-the-box solution for trajectory-based simulations for strong-field ionization (see Figure 1). It is designed with user-friendliness in mind and is expected to serve as a valuable and powerful tool for the community of strong-field physics [1].

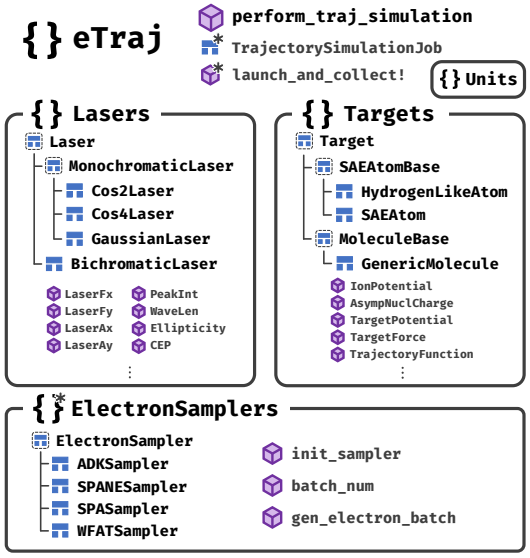


Figure 1. Scheme of the eTraj package

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Self-Consistent Extraction of Photoabsorption Time Delays in Attosecond Streak Camera

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Synopsis We present a self-consistent iterative method to extract the intrinsic photoabsorption time delay.

Photoionization can generally be decomposed into a photoabsorption process plus a half-scattering process (see Figure 1). The streaking time delay observed in the attosecond streak camera, correspondingly, can be decomposed into a photoabsorption time delay and a continuum time delay. We propose a self-consistent method to account for the potential-laser coupling effect and extract the intrinsic absorption time delay, without invoking an implicit assumption of a constant continuum correction made in previous streaking studies. The concept is based on an iterative technique starting from an initial guess of the absorption time delay and using classical electron trajectories to consistently remove the coupling-induced momentum shift to the streak signal. We show that the self-consistent iterative algorithm converges quickly and is robust against different initial guesses. The method is applied to laser-induced ionization of atoms to obtain the absorption time delay in resonant two-photon ionization and to explore possible time delays in above-threshold ionization. The results confirm an absorption time delay linearly dependent on the ionizing pulse duration in resonant two-photon ionization. The method is also shown to perform better than conventional methods and to identify a small negative absorption time delay (less than 2 attoseconds) in above-threshold ionization [1].

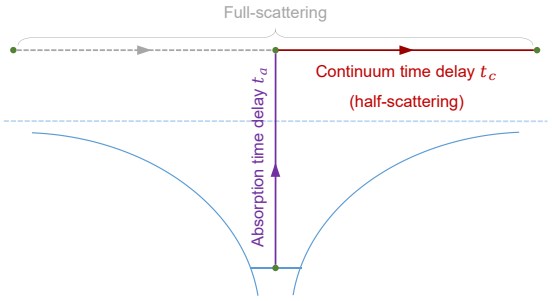


Figure 1. Scheme of the photoionization time delay. The full photoionization process in attosecond streak setting includes photoabsorption from the XUV pulse and a half-scattering process in the continuum, shown by the solid line. The observed streaking time delay in the attosecond streak camera $t_s = t_a + t_c$, where t_a and t_c denote time delays related to absorption of XUV photons and dynamics in the continuum, respectively. The full scattering process is shown as the horizontal line.

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Analysis of low-energy electron interferences in Helium

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Synopsis Studying the interference of photoelectrons that are ionized from a coherent superposition of states is a powerful method for characterizing the dynamics of bound electron wave packets. Iterative numerical methods can extract information from experimental spectrograms, mapping photoelectron energy against delay between a short attosecond pulse train and a probing infrared field. Here, we present our progress in developing such methods for the case of Helium.

The development of broadband extreme ultraviolet sources has made it possible to study complex superpositions of both bound and continuum electron states via pump-probe schemes [1, 2]. In our case, a short extreme-ultraviolet (XUV) attosecond pulse train is used to excite simultaneously several bound states of the Rydberg p-series in Helium close to the ionization threshold. A delay controlled, few-cycle, near-infrared, broad bandwidth ($1.2 - 2\,\mathrm{eV}$) probe pulse subsequently ionizes into the continuum. Due to the energy difference between states, contributions from different ionization pathways will have a different phase and can interfere in the continuum. This causes quantum beats as well as direct-indirect interference to appear in the signal while scanning the delay between the exciting attosecond pulse and the probe.

Spectrograms, scanning the pump-probe delay while detecting photoelectrons with a home-made 3D momentum spectrometer, have been recorded and are being analyzed. To this end, we have developed a numerical method aiming at extracting the (partial) density matrix for the state prepared by the pump and the ionization amplitude due to the probe without any prior knowledge of the state preparation and spectral profile of the probe. The numerical method uses a Monte Carlo approach to retrieve the ionization amplitude and has been benchmarked against generated data within a range of photoelectron resolutions, showing good performance and robustness under our experimental conditions.

Figure 1 shows an experimental dataset along with the fit retrieved by the method. The method reproduces the signal strength well, however, the experimentally observed phase of the quantum beat between the 4p and 5p singlet states, appearing between $0.6 - 0.8\,\mathrm{eV}$ could not be reproduced. Such a limitation is not found for generated data, and systematic studies of various effects including the statistical noise and the preparation of the bound states with the attosecond pulses will be discussed.

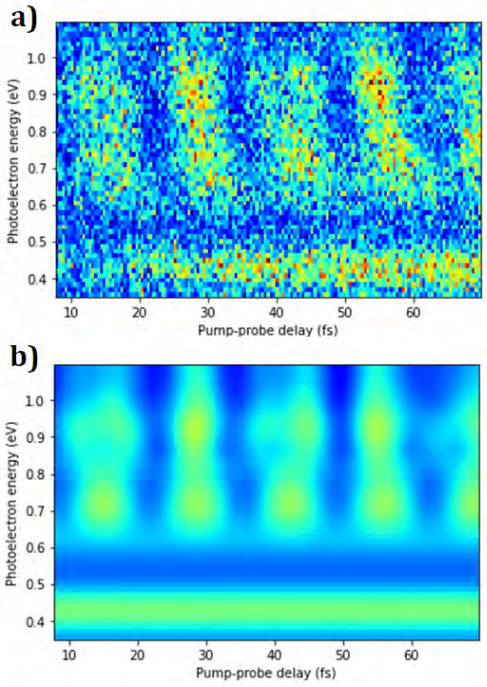


Figure 1. a) An experimental pump-probe delay scan. b) The retrieved fit.

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Probing Laser Dressing-Induced Dynamics in Acetonitrile with Soft X-ray Attosecond Transient Absorption Spectroscopy

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Synopsis Unlike atomic systems, molecular strong-field interactions show complex couplings beyond standard theories. Soft X-ray attosecond transient absorption spectroscopy is a powerful technique for the investigation of such dynamics. In this study, we probe acetonitrile using a soliton self-compressed single-cycle mid-infrared pump and reveal transient features at the C K-edge that are linked to light-induced states and AC Stark shifts with femtosecond beatings. Ongoing studies at the N K-edge, combined with field reconstructions, promise deeper insight into laser-dressed dynamics in acetonitrile.

Recent advances in attosecond transient absorption spectroscopy (ATAS) in the water window have enabled unprecedented exploration of ultrafast electron dynamics with site specificity [1]. Such measurements are now capable of revealing complex strong-field interactions in molecules, including light-induced states (LIS) and AC Stark shifts of core-edge transitions. Unlike atomic systems [2], where these effects are well understood [3], molecular interactions introduce non-trivial couplings that challenge conventional theories. Understanding these dynamics is crucial for advancing ultrafast light-matter control and attochemistry.

Here, we report a soft X-ray (SXR) ATAS investigation using a soliton self-compressed single-cycle mid-infrared (MIR) pump to probe a prototypical molecular system: acetonitrile (ACN). ACN is particularly well-suited for this study due to its highly polarizable valence states that originate from the strong dipole moment induced by the N lone pair and its linear structure. Furthermore, its high ionization potential (12.2 eV) makes it an ideal platform for investigating laser dressing effects below the ionization threshold, allowing us to isolate and study these dynamics without the interference of ionization-induced processes.

Our preliminary results reveal multiple laser dressing-induced transient signals associated with core-to-valence excitation localized at different sites of the ACN molecule. The resulting SXR absorption features in Fig. 1a and their C K-edge transients in Fig. 1b are assigned as follows: A: C 1s→π^*C≡N, B: C 1s→C−H, C: C 1s→σ^*C−C, D: C 1s→σ^*C≡N, E: N 1s→π^*C≡N [4]. Observed pump-induced changes at the C K-edge of the absorption spectrum have also shown time-dependence in Fig. 1b.

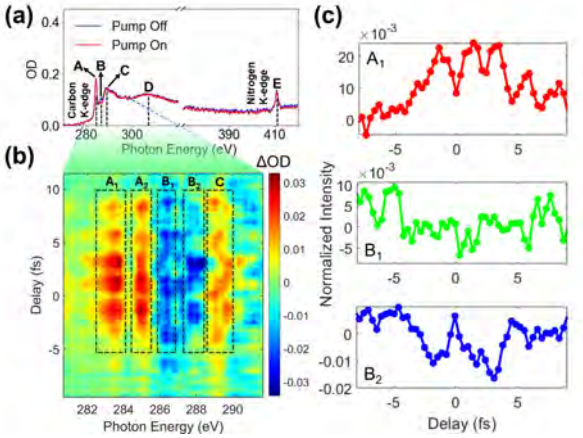


Figure 1. (a) ATAS spectra of ACN at pump off and pump on instances. (b) 2D ATAS map and (c) lineouts of the C K-edge of ACN.

The underlying mechanisms driving the 2-fs oscillations (A_1, Fig. 1c) and the anti-symmetric counter-beating splitting of the negative transient absorption features (B_1 and B_2, Fig. 1c) are under active investigation, alongside the time-dependent dynamics at the N K-edge. These results are expected to give crucial information on the nature of valence electronic states in a strongly polarizable molecular system as ACN.

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Generation of high-order harmonics with hybrid spin and transverse orbital angular momentum from a crystalline solid

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Synopsis We theoretically study the high harmonic generation of solids driven by the spatiotemporal optical vortex. We develop a semiconductor quantum-orbit model to demonstrate the conservation of angular momenta in the HHG process. Harnessing the bicircular spatiotemporal vortex light field, we generate high-order harmonics with the hybrid spin and transverse orbital angular momentum.

Light beams carry two distinct manifestations of angular momenta which can be distinguished under the paraxial approximation: the spin angular momentum (SAM) and the orbital angular momentum (OAM). The SAM of light exhibits its inherent "twisting" or polarization. In contrast, the OAM of light is conveyed through an optical vortex. Recently, the spatiotemporal optical vortex (STOV), which carries transverse OAM, has attracted significant attention and expanded the functionality of optical vortices.

In this work, we explore the conservation and modulation of SAM and transverse OAM in high harmonic generation (HHG) of crystalline solids [1]. First, we quantitatively demonstrate the conservation law of transverse OAM in HHG process with semiconductor Bloch equations and the semiconductor quantum-orbit model derived from the recollision model, elucidating the electron motion under crystal potential fields at the attosecond scale. Then we utilize the specific symmetries of crystals to modulate multiple absorption channels, generating high-order harmonics with hybrid SAM and transverse OAM components. Furthermore, by adjusting the intensity ratio of two light beams, we achieve a precise control over the proportion of different transverse OAM components for a certain harmonic. Our work provides an intuitive and clear physical picture for the transfer and conservation of SAM and OAM in HHG of crystalline solids. It also offers new insights for further exploring the interactions between anisotropic materials and optical Hopfions, optical skyrmions, and other novel structured light fields.

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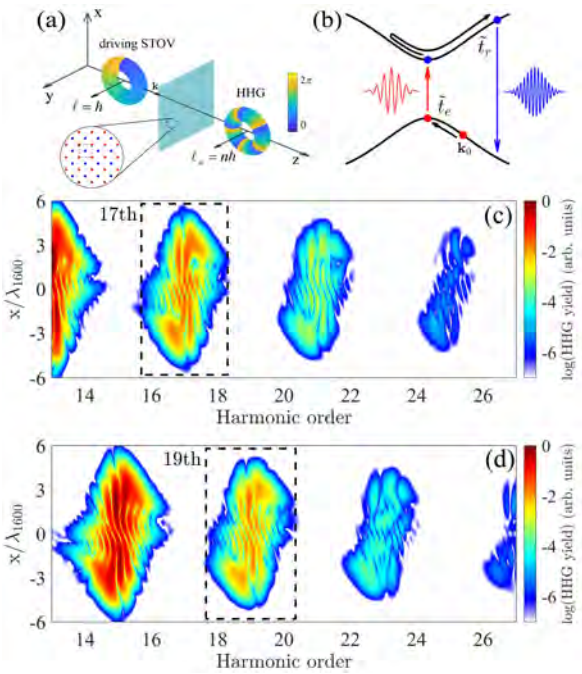


Figure 1. (a) Schematic for HHG by a fundamental STOV interacting with the checkerboard lattice. The transverse OAM obeys the conservation law in this process. (b) Schematic of the semiconductor quantum-orbit model [2]. The spatially resolved HHG spectrum driven by two-color counter-spin and counter-vorticity STOVs is simulated, with (c) and (d) corresponding to the left and right circular polarization components, respectively.

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Ultrafast Light-induced Spin and Orbital Magnetization in Two-dimensional Semiconductors

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Synopsis We demonstrate ultrafast magnetism in 2D non-magnetic semiconductors through valley-polarized excitations from circularly polarized laser pulses. By tuning the energy and polarization of the laser, we show selective control over orbital and spin contributions.

The quest for rapid control of magnetism has propelled advancements in the field of femtomagnetism. In this study, we theoretically demonstrate that a net magnetic moment can be induced in a two-dimensional (2D) non-magnetic semiconductor on a femtosecond time scale, even in the absence of spin-orbit coupling. Two-dimensional hexagonal semiconductors, such as transition metal dichalcogenides, possess a valley degree of freedom, where valleys are local minima in the conduction bands having opposite Berry curvatures at opposite corners of the Brillouin zone. Optical excitations with circularly polarized laser pulses in these materials result in valley-polarized excitations, a cornerstone of the field of valleytronics. We illustrate how these valley-polarized excitations lead to the induction of an orbital magnetic moment on sub-femtosecond time scales, under both resonant and multi-photon resonant conditions. Furthermore, in the presence of spin-orbit coupling, the net magnetic moment is determined by competing contributions from valley-polarized spin and orbital magnetic moments. Moreover, we show that the spin and orbital magnetic moments can be distinctly controlled by tuning the polarization properties of the laser pulse. This work opens new avenues for ultrafast information processing and storage technologies based on the orbital degree of freedom of magnetism.

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Manipulating Molecular Dynamics with Polarization-Shaped Laser Pulses

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Synopsis Light-molecule interactions play a crucial role in photochemistry, at the heart of which is the breaking and formation of chemical bonds. Our polarization-shaped scheme aims to understand and manipulate the fundamental mechanisms behind the light-induced molecular dynamics and bimolecular reactions.

For molecules exposed in strong laser fields, valence electrons absorb photons and escape from the ionic potential, while the remaining cation interacts with the strong field and dissociates as nuclear fragments. In this respect, if we construct an ultrafast strong laser pulse characterized by time-dependent polarizations, the time information of ejected electrons and nuclear fragments can be recorded experimentally, and full-dimensional molecular dynamics are explored and manipulated in principle.

In this presentation, we demonstrate the powerful applications of the polarization-shaped laser pulse in understanding and manipulating light-induced molecular dynamics and bimolecular reactions. The target molecular systems ranges from simplest hydrogen monomer and dimer to complex dimers. Our work opens the opportunity towards the real-time tracking and coherent control of the laser-induced molecular dynamics and stereo-dynamical bimolecular reaction using waveform-manipulated light fields.

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Non-Cascade Random Walk in Solid High Harmonic Generation

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Synopsis The well-known random walk, including classical itself and the quantum counterpart, has laid the foundation for applications in many realms of science and technology. Here, we report the experimental realization of non-cascade random walk in the orbital angular momentum space of light using solid high-harmonic spectroscopy. Our results propose a novel view of algorithm simulation based on solid-state materials and introduce a potential platform for non-cascade computations.

Random walk was first proposed by K. Pearson in 1905, when A. Einstein provided a quantitative explanation and outlined its statistical properties based on molecular kinetic theory. Quantum walk, as the quantum counterpart of classical random walk, is characterized by quantum superposition and interference effects. Hitherto, photonic implementations of random walks have widely expanded from real space to discrete orbital angular momentum space. However, the current realization of both classical and quantum random walks requests a cascade characteristic, which implies that the disturbance of preceding random walk will influence the next step propagation [1, 2].

In this work, we observe the high harmonic generation process in solids driven by a structured femtosecond laser pulse. The spin and orbital angular momenta carried by the fundamental wave act as a coin and a high-dimensional Hilbert space where a walker move. The highly nonlinear process driven by the structured laser pulse provide an appropriate platform to demonstrate the non-cascade random walk in the orbital angular momentum space. The random walk in high harmonic generation demonstrates a district distribution from conventional classical and quantum walks. Our work displays a novel utilization through the interaction between laser and the solid material, providing a bridge between the fields of high-harmonic spectroscopy and the quantum information.

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Robust Discrimination Between Solid-State High Harmonic Generation Mechanisms Based on Real-Space Dynamics

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Synopsis We introduce a gauge-invariant, real-space method using maximally localized Wannier functions to distinguish between inter- and intraband high harmonic generation (HHG) in solids. Our approach reveals that intraband HHG scales with the lattice-site separation (and hence appears at higher harmonic orders), whereas interband HHG is dominated by transitions between nearby sites. Our method provides a gauge-invariant and computationally efficient means for distinguishing between distinct harmonic generation mechanisms in condensed matter.

High harmonic generation (HHG) in solids has emerged as a powerful tool for probing ultrafast electron dynamics and developing compact extreme ultraviolet light sources. Unlike gas-phase HHG, where a single recombination step dominates HHG emission, the solid-state counterpart can involve multiple mechanisms, including injection current, intraband acceleration, and interband electron-hole recombination. While the light emitted by these mechanisms exhibits distinct spectral characteristics, the definitions of these mechanisms is gauge-dependent and numerically distinguishing these different mechanisms can be computationally expensive [2].

We present a novel, gauge-invariant approach based on a real-space description using maximally localized Wannier functions to discriminate HHG mechanisms through the dependence of each mechanism on the separation between Wannier states by expressing the total momentum operator as a sum over contributions connecting Wannier states with different lattice site separations. Our analysis shows two distinct regimes, as shown in Fig. 1. For (a) intraband HHG, energy conservation requires that the frequency of emission Ω satisfies $\Omega \leq F_0|R|$, where F_0 is the driving electric field amplitude and R is the separation between Wannier states. For (b) interband HHG, the cross-section for recombination rapidly decreases with the separation between states, clustering emission near zero separation.

We validate our gauge-invariant method through numerical simulations in two-dimensional systems. This real-space approach provides clear physical insight into the distinct mechanisms of HHG in solids, circumvents many of the numerical challenges encountered in traditional reciprocal-space methods, and will be critical for advancing the interpretation of HHG experiments in ultrafast condensed matter physics.

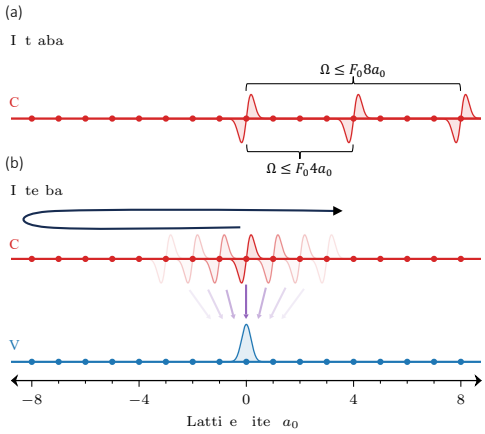


Figure 1. (a) Intraband HHG emission results from radiative transitions between Wannier orbitals separated by a distance R in the presence of an electric field with amplitude F_0, for which the frequency of emission $\Omega \leq F_0 R$. (b) Interband HHG emission results from radiative interband transitions, for which the cross section rapidly decreases with $|R|$.

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Electronic dynamics created at conical intersections and its dephasing in aqueous solution

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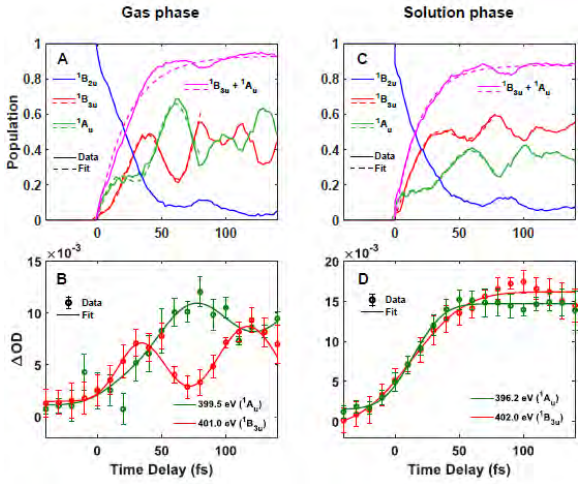
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Synopsis In this work, we performed ultrafast time-resolved X-ray absorption spectroscopy on UV-excited gas and solution phase pyrazine, demonstrating that there is large-amplitude rearrangement of the electronic structure caused by the conical intersection and dephasing of electronic dynamics due to solvation.

Understanding the electronic dynamics of molecular systems is important for many fields in photophysics and photochemistry. This is especially challenging in the vicinity of a conical intersection, where the Born-Oppenheimer approximation breaksdown. In this work [1], we present two experimental breakthroughs: (1) the observation of electronic and vibrational dynamics corresponding to a circular rearrangement of the electronic structure created by conical intersection dynamics and (2) the sub-40 fs dephasing induced by aqueous solvation.

Using a state-of-the-art table-top HHG source covering the entire water window [2], we performed time-resolved X-ray absorption spectroscopy (XAS) of single-UV-photon-excited gas and solution phase pyrazine samples at both the carbon and nitrogen K-edges. In the gas phase, we observed antiphased quantum beats indicating population transfer between the $^1B_{3u}(np^*)$ and $^1A_u(np^*)$ excited states, which is not observed in the solution phase due to decoherence and dissipation mechanisms present from solvation.

Figure 1. (A,C) In gas and solution phase, respectively, populations of the diabatic $^1B_{2u}(pp^*)$, $^1B_{3u}(np^*)$ and $^1A_u(np^*)$ states based on FSSH trajectories. (B) Time-dependent ΔOD of gaseous pyrazine for the bands centered at 399.5 eV and 401.0 eV, which are associated with the $^1A_u(np^*)$ and $^1B_{3u}(np^*)$ characters, respectively. (D) Time dependent ΔOD of 5M aqueous pyrazine for the bands centered at 396.2 eV and 402 eV, which are associated with the $^1A_u(np^*)$ and $^1B_{3u}(np^*)$ characters, respectively.



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In Search of Lost Tunneling Time

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Synopsis The Larmor clock and the attoclock offer distinct approaches to measuring tunneling time, yielding conflicting results. We reconcile these discrepancies by mapping the attoclock onto a static tunneling model and applying the weak-value interpretation of the Larmor clock.

Tunneling time, the duration for a quantum particle to traverse a potential barrier, remains debated in quantum mechanics, as time itself is not an observable. Various operational definitions highlight different aspects of tunneling.

Attoclock experiments, using ultra-short, nearly circularly polarized laser pulses, have renewed interest in tunneling time in strong-field ionization [1, 2, 3, 4]. However, they were shown to measure not the tunneling time but the Coulomb interaction's effect on the electron [3, 4]. In contrast, a recent experiment in A. Steinberg's group unambiguously measured the Larmor tunneling time using a Bose-Einstein condensate (BEC) of ^{87}Rb atoms tunneling through an optical barrier [5]. Applying the Larmor time concept to strong-field ionization resulted in a nonzero tunneling time [2, 7], contradicting attoclock experiments [4].

We resolve this contradiction by analyzing the attoclock in a rotating reference frame and mapping it onto a static one-dimensional tunneling model. Using the weak-value interpretation of the Larmor clock, we calculate the Larmor tunneling time during the ionization process and find a nonzero value over a range of field strengths, consistent with Steinberg's measurements [5, 6] and prior theoretical predictions [2, 7]. Our results show that while the Larmor time is well-defined inside the barrier, it remains constant once the electron exits, independent of where it is measured.

Our model further allows us to define a position-resolved attoclock tunneling time, which, while nonzero at the tunnel exit, vanishes at the detector (Fig. 1). This means that the attoclock does not resolve the local Larmor time but instead a non-local phase time. The lack of a position-resolved measurement in attoclock experiments prevents direct access to the tunneling time at the barrier exit.

Thus, the tunneling time is not necessarily imprinted on the electron wavefunction in strong-field ionization. Instead, it can only be rigorously determined by detecting the spatial phase of the electron at the barrier exit, which is achievable with an experimental setup akin to the BEC tunneling experiment [5].†

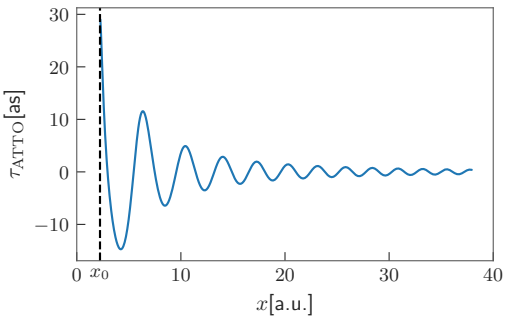


Figure 1. Attoclock time as a function of position. As the electron's distance to the ionic core increases, the attoclock time approaches zero.

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Enatiosensitive Geometric Phases in Multi-Photoexcitation Processes

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Synopsis We report enatiosensitive geometric effects in multiphoton excitation and ionization processes and identify new efficient enantio-sensitive observables of purely geometric origin. We show that (i) enantio-sensitive molecular orientation by photoexcitation is proportional to the curvature and can reach 60%, (ii) the enantio-sensitive photoexcitation yield and free induction decay in chiral molecules driven by locally chiral fields encode the enantio-sensitive geometric phase. Our results uncover geometric and topological properties of ultrafast non-equilibrium chiral electron dynamics.

We introduce the concept of temporal geometry, which encompasses the geometric and topological properties of temporal shapes, i.e. trajectories traced by the tip of a time-dependent vector in vector space [1]. The connection $\vec{A}(\vec{E}) = \langle\psi|\,\nabla_{\vec{E}}\,|\psi\rangle$, which detects sensitivity of non-linear response encoded in the laser field-driven electron wave-function $\psi(\vec{E})$ to the trajectory traced by the laser field polarization $\vec{E}$, emerges as a key geometric property of an ultrafast non-linear response. We show that the curvature $\vec{\Omega}_{\rm exc} = \nabla_{\vec{E}} \times \vec{A}(\vec{E})$ controls the enantio-sensitive molecular orientation upon photoexcitation of randomly oriented molecules by circularly polarised field, which can be probed by photoionization with linearly polarised field (Fig. 1).

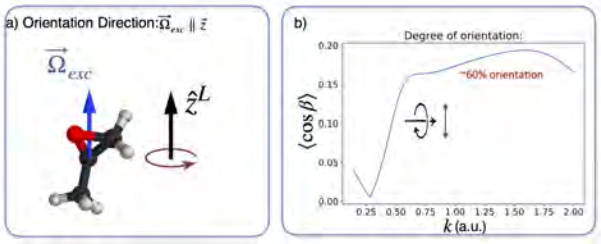


Figure 1. (a) $\vec{\Omega}_{exc}$ defines molecular orientation: in randomly oriented ensemble the molecules in which $\vec{\Omega}_{exc}$ is parallel to the light's spin direction $\hat{z}^L$ are preferentially excited. (b) The degree of orientation (expectation value $\langle\cos\beta\rangle$, β is the angle between $\vec{\Omega}_{exc}$ and $\hat{z}^L$) as a function of the photoelectron momentum k.

Next, we show that all geometric quantities: the connection, the curvature and the geometric phase become enantio-sensitive if chiral molecule is driven by locally chiral fields [2] – the fields with polarization vectors drawing chiral Lissajous figure in time during one cycle. We find that chiral electron currents excited by such fields lead to accumulation of enantio-sensitive geometric phases which can be detected in enantio-sensitive photoexcitation yield.

Finally, we consider a rotating chiral molecule, where the two photoexcited states are coupled via photoionization continuum by locally chiral light. We show that enantio-sensitive geometric phase (i) can be controlled via molecular rotations, (ii) is imprinted onto induced polarization and (iii) can be detected via free induction decay. By identifying non-vanishing enantio-sensitive geometric phase associated with temporal geometry of ultrafast chiral electron currents and linking it to specific observables our work lays the foundation for exploring attosecond topological phenomena driven by electronic currents in gas phase chiral molecules. The access to geometric phases via several observables established here may allow us to unlock a quantized chiral response, which could bring an attractive new property of topological robustness to chiral measurements. We gratefully acknowledge ERC-2021-AdG project ULISSES, grant agreement No 101054696.

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Ultra-fast nonlinear optical response of chiral molecules with a focus on conformer sensitivity

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Synopsis We model the nonlinear response of chiral molecules to linearly polarized, tightly focused laser pulses. Focusing creates a longitudinal field component, causing the field to acquire a transverse spin, which enables a strong chiroptical response via purely electric-dipole interactions. This results in the emission of odd (even) harmonics through achiral (chiral) channels, whose interference is modulated by the carrier-envelope phase of the driving field. We study serine conformers and examine how their molecular phase influences the polarization of emitted harmonics.

Chirality is a fundamental geometric property, present from molecular to macroscopic scales. Traditional chiroptical methods rely on weak magnetic interactions, limiting their efficiency. We aim to develop chiral recognition methods based solely on electric-dipole interactions, offering enhanced enantiosensitivity [1].

We investigate the carrier-envelope phase (CEP) dependence of enantio-sensitive observables within the electric dipole approximation by numerically analyzing the nonlinear response of randomly oriented chiral molecules in the gas phase. Using time-dependent density functional theory (TDDFT), we model their interaction with few-cycle, tightly focused, CEP-controlled linearly polarized laser pulses. Tight focusing induces a longitudinal field component, creating a forward-elliptically polarized field [2]. This drives a chiral response perpendicular to the polarization plane, leading to the emission of even-order chiral harmonics in addition to the odd-order achiral harmonics. Their CEP-dependent interference results in enantiosensitive non-linear optical rotation [2]. Here we explore the sensitivity of the CEP-dependent signal to chiral molecular conformations and the uniqueness of the CEP molecular markers.

We focus on the chiral dynamics of essential amino acids, using serine as a prototypical case. We analyze its three dominant conformers in the gas phase, with relative populations of 43.7%, 18.8%, and 14.8% [3]. By placing a polarizer before the detector one can convert enantio-sensitive polarization properties into an enantio-sensitive intensity distribution, which can be considered as chiral "QR codes" mapping the chiral dichroism of emitted harmonics as a function of CEP of the incident light. We show that chiral dichroism (CD) vs CEP has different pattern for different conformers (note that for the same harmonic order the CD maximizes at different CEP values in serineI and serine II, Fig 1) reflecting different molecular phase accumulated due to ultrafast electron dynamics in two conformers and making chiral QR codes suitable for molecular fingerprinting.

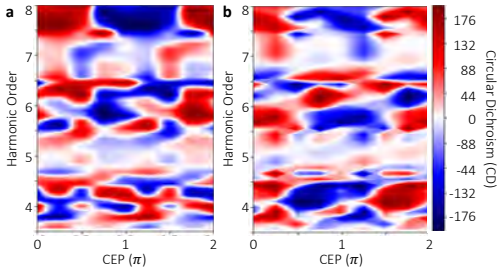


Figure 1. Chiral "QR codes": Chiral Dichroism (CD) of the emitted harmonics as a function of the CEP of incident light for a) serineI and b) serineII conformers.

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Spin and orientation locking in photoionization of chiral molecules

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Synopsis We extend the concept of geometric magnetism emerging in photoionization of chiral molecules (*Commun. phys.* **6**, 2023, 257) by including the photoelectron spin, and show that the detection of photoelectron spin defines the enantio-sensitive orientation of the cation. This effect can be regarded as a manifestation of chirality induced spin selectivity (CISS).

Chirality induced spin selectivity (CISS) was first discovered in 1999 and has now become an umbrella term for a wide range of phenomena wherein molecular chirality imparts significant spin selectivity to electron processes [1]. Several theoretical models regarding CISS have been proposed, however, a complete quantitative theory still remains elusive [2]. Here, we focus on an unexplored mechanism underlying the interplay of molecular chirality and electron spin in photoionization by circularly polarised light mediated by geometric magnetism [3].

Geometric magnetism in photoionization of chiral molecules addresses the dynamical origin of enantio-sensitive observables in one or multi-photon ionization from the emergence of the *curvature* $\vec{\Omega}$, associated with laser-driven electron dynamics, and its functional dependence on the molecular orientation with respect to the driving laser field with non-zero spin angular momentum [3]. The *curvature* is a dynamical property quantifying dichroism in photoionization from a superposition of electronic states $|i\rangle, |j\rangle$, [3], i.e,

$$\vec{\Omega}_{ij} = \langle i|\hat{\vec{\mathsf{F}}}|j\rangle \tag{1}$$

$$\hat{\mathsf{F}}_a = i\epsilon_{abc}\hat{\mathsf{d}}_b\hat{\mathsf{P}}_{\mathcal{E}}\hat{\mathsf{d}}_c \tag{2}$$

$$\hat{\mathsf{P}}_{\mathcal{E}} = \int d\Theta_k \, |\psi^{(-)}_{\vec{k}}\rangle\langle\psi^{(-)}_{\vec{k}}| \tag{3}$$

where, $\hat{\mathsf{d}}$ is the dipole operator, $\hat{\mathsf{P}}_{\mathcal{E}}$ is the energy projector resulting from averaging over all directions of the photoelectron momentum $\int d\Theta_k$ for a fixed photoelectron energy $\mathcal{E} = k^2/2$ and 'Raman-like' transitions occuring via the continuum states $|\psi^{(-)}_{\vec{k}}\rangle$. The operator $\hat{\vec{\mathsf{F}}}$ is a time-odd pseudovector, therefore, its diagonal matrix elements vanish which means that the *curvature* vanishes in time-even states for any stationary time-even state [3], e.g., the ground state of molecules.

One way to enable the *curvature* is to excite a current in molecular bound states prior to photoionization by creating coherences between electronic states $|i\rangle, |j\rangle$. In this case, the *curvature* $\vec{\Omega}_{ij}$ will control the enantio-sensitive orientation of the cations (MOCD - molecular orientation circular dichroism), which is an example of enantio-sensitive charge directed chemical reactivity. An alternative way to enable the *curvature* is to involve time-odd states by resolving photoionization on the photoelectron spin. In this case, we should expect enantio-sensitive molecular orientation to occur even after one-photon ionization from the ground state of a randomly oriented molecule, provided that we distinguish the photoelectron spin.

Here, we extend the concept of geometric magnetism to include the photoelectron spin in one-photon ionization, and report a curious effect which we refer to as enantio-sensitive spin-orientation locking. This effect can be regarded as a manifestation of CISS wherein the detection of photoelectron spin defines the enantio-sensitive orientation of the cation.

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An entanglement perspective on molecular Auger decay

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Synopsis After inducing core photoionization in a model diatomic molecule via an ultrafast light-pulse, we investigate the dynamics of entanglement during the system's time evolution. The molecule is primarily modeled and parameterised as an oversimplified version of the Beryllium dimer (but some exploration of the parameter space is also performed). This results in a low molecular dissociation energy, making possible to examine qualitative trends of the interplay of core-photoionization, Auger decay, and molecular dissociation through the entanglement lens.

The Auger process is well known as a spectroscopic fingerprint for elemental identification, but it also provides a very interesting stage to study electron entanglement. To gain theoretical insight into this latter aspect, we address entanglement development in a model molecule, after core photoionization induced by a ultrashort light-pulse, and its subsequent disappearance due to decoherence effects arising from the molecule nuclear dynamics and electronic de-excitation via Auger decay and electronic shake processes.

As system of interest we consider a simplified dimer molecule coupled to a discretised continuum, with the parameters in the Hamiltonian tailored to resemble some features of a real Beryllium dimer. In particular, the dissociation energy in our model is very close to that of Be_2 molecule, which allows us to examine possible connections between entanglement, photoionization, Auger-decay, and induced molecular dissociation.

In more detail, our model consists in a Hubbard dimer with several spin-orbitals/site (where the inter-site electron hopping depends on the internuclear distance, see Figure 1) in the presence of a continuum to accommodate the photo- and Auger electrons. Both the nuclear dynamics and the continuum states are described in one-dimension. Following an initial excitation via a short laser pulse (see Figure 1), the time evolution of the systems is performed numerically via the short iterated Lanczos algorithm.

We characterize the systems dynamics in terms of several observables and quantities; among these, the photo- and Auger electron energy distributions (see Figure 1), the atomic and molecular state occupations, the temporal behavior of the nuclear wavepacket, the one-particle reduced electronic density matrix (of interest for KRAKEN-type experiments [1, 2]) and, most relevant, the concurrence in the photo- and Auger continuum. Finally, although dealing primarily with a situation resembling the case of Be_2, we also explore a range of parameters values, to contrast different qualitative behaviours.

While certainly oversimplified in several ways, our model permits to perform a one-step, equal footing and full quantum treatment of all the different degrees of freedom involved, namely the multi-electron correlated dynamics, the quantum nuclear motion, and the photo/Auger electron continuum states, allowing for a qualitative and yet approximation-free exploration of entanglement dynamics.

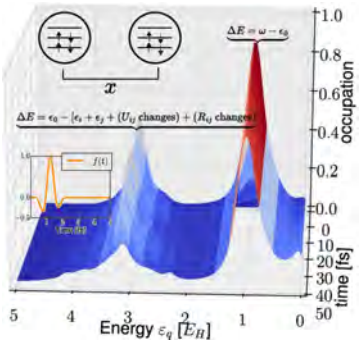


Figure 1. An example of the energy dispersion of the photo- and Auger electrons as function of time. A schematic of the model dimer molecule and the temporal profile of the light pulse are also shown.

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Photoelectron Spectra Calculated From Absorption

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Synopsis We demonstrate a way to calculate photoelectron spectra on a numerical domain much smaller than the actual extension of the wave function.

Simulating and analyzing unbound quantum systems tend to be demanding. One reason is the fact that obtaining the proper scattering states may be far from trivial. Another reason is that wave functions are not confined. As they extend very far, their description requires a very large numerical domain.

We present photoelectron spectra calculated using the method introduced in [2] and [3], and expanded in [1]. We have coined this approach photoelectron spectra calculated from absorption via the density operator (PESCADO).

We augment the Hamiltonian in the time-dependent Schrödinger equation with a complex absorbing potential (CAP),

$$H \to H_{\text{eff}} = H - i\gamma(r).$$

The norm of the wave function will decrease whenever it has a spatial overlap with the CAP. Since we know which parts have been taken out, we are able to analyse them as they are removed, and thus accumulate information about the absorbed particle.

We aggregate the outgoing waves by, formally, rewriting the state in terms of a density operator, and coherently accumulate its diagonal elements of interest. Within such a formulation, any global phase factor becomes superfluous. To the extent that the time-dependence of the scattering states resides in such a factor, the diagonal elements in question become, in effect, time-independent.

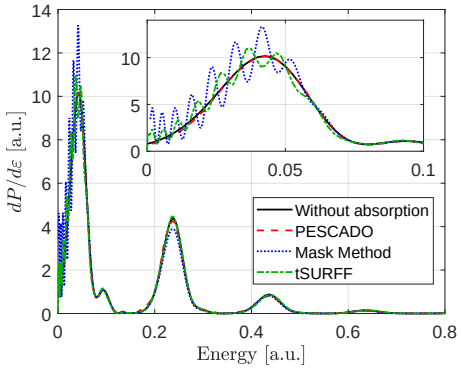
From this, ionization probabilities doubly differential in energy and ejection angle may be approximated by projecting onto the time-independent scattering states. To determine the energy distribution alone, $\frac{\partial P}{\partial \varepsilon}$, we can integrate out the angular dependence, which yields

$$\frac{\partial P}{\partial \varepsilon} \approx \frac{2}{\hbar}\text{Re}\sum_{\ell,m}\int \mathrm{d}r \int \mathrm{d}r' \psi^*_{\varepsilon,\ell}(r)\zeta_{\ell,m}(r,r')\psi_{\varepsilon,\ell}(r')$$

with $\zeta_{\ell,m}(r,r') = \int_0^T \gamma(r) f_{\ell,m}(r;t) f^*_{\ell,m}(r';t)\,\mathrm{d}t$, where $f_{\ell,m}(r;t)$ is the radial part of the wave function.

In the figure the resulting $\frac{\partial P}{\partial \varepsilon}$ can be seen. We have compared it with results from the Mask[4] and tSURFF[5, 6] methods for the same scenario. We have also included results with an untruncated numerical domain.

PESCADO works well, and has near complete overlap with the untruncated numerical domain. For higher order above threshold ionization peaks, both Mask and tSURFF provide well converged results. However, for the first peak, it is clearly seen how the photoelectron spectra oscillate quite vigorously. This artefact is attributable to their neglect of the Coulomb potential in the scattering states.



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Non-adiabatic dynamics investigation of $Cr(CO)_4$(2,2'-bipyridine)

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Synopsis Photochemical simulations of $Cr(CO)_4$(2,2'-bipyridine) revealed involvement of triplet states in the photodissociation process, contrary to previous understanding of the system. Photoelectron and X-Ray absorption spectra were produced to find a sensitive probe of this ultrafast process.

Transition metal complexes with a d^6 electron configuration and electron-accepting ligands exhibit rich excited state chemistry. The interplay of Metal-to-Ligand Charge Transfer (MLCT) and Metal-Centred states of both singlet and triplet characters leads to branching mechanisms where both phosphorescence and photodissociation can occur. This is exemplified by the widely studied, strongly phosphoresecent $[Ru(2,2'\text{-bipyridine})_3]^{2+}$ cation, which photodissociates under acidic experimental conditions, despite being generally considered to be stable [1].

For the species of $M(CO)_4$(diimine) (M = Cr, Mo, W), the photodissociation from singlet MLCT states is more facile, occuring on an ultrafast 100 fs timescale, and understood to occur in competition with intersystem crossing to triplet MLCT states [2]. Understanding the nature of these mechanisms is crucial for the design of luminescent probes and photocatalysts, as in each case the competing reaction is unwanted: For photocatalysis the long-lived phosphorescent trapping states are a kinetic bottleneck, while for luminescent probes photodissociation process is a substrate-depleting side reaction.

In our work, we investigate the photoprocesses of the simplest of these compounds: $Cr(CO)_4$(2,2'-bipyridine). We have performed non-adiabatic dynamic simulations using the Trajectory Surface Hopping method. Our results have shown that photodissociation can occur from both singlet and triplet MLCT states [3]. This result contradicts the previously established mechanism, wherein photodissociation occurs exclusively on the singlet MLCT state.

The experimental study of these systems is very challenging due to the presence of dense electronic state manifolds, and an ultrafast timescale. We have modelled two spectroscopic measurements of transient X-Ray absorption and photoelectron spectra with the goal of finding an experimental technique that is sensitive to the photodissociation process. Results of our studies provide additional insights for the ultrafast photomechanisms of d^6 transition metal complexes.

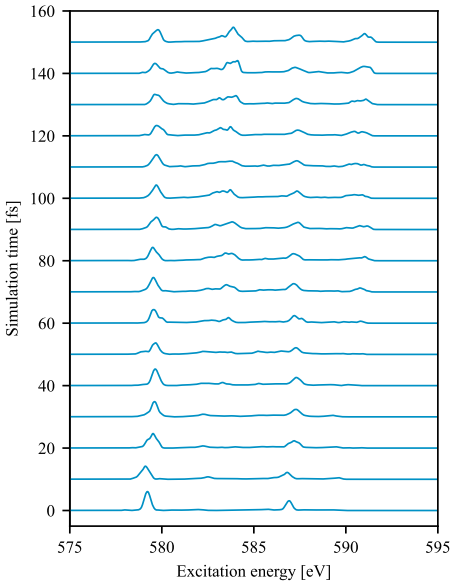


Figure 1. Transient X-Ray absorption spectrum of $Cr(CO)_4$(2,2'-bipyridine)

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