# The 2024 International Workshop on

# Ultra-Fast Science

## ( WUFS 2024 )

21-23 April 2024

| Shanghai |

East China Normal University

# East China Normal University

East China Normal University (ECNU) is located in Shanghai---a renowned international metropolis with architectural landmarks that capture the eye with new aesthetic designs. The architectural designs are emblematic of the skyline over the city, revealing a nurturing culture of people and an abundance of resources for boosting the city.

Founded in Shanghai in October 1951, ECNU is one of the prestigious universities in China. In 2017, ECNU was selected as one of the Category A universities in China's Double First-Class University Program, a higher education initiative launched by the Ministry of Education aiming to develop elite Chinese universities into world class institutions by the end of 2050.

Adhering to the strategy of "Interdisciplinary, Internationalization and Informatization", the "Three Programs" – "Developing education excellence, Promoting interdisciplinary academic excellence, and Serving national strategies", and the mission - "Education, Civilization, Development", ECNU has made tremendous achievements in talent training, scientific research, community service, and international exchanges in the recent decades.

ECNU's motto is: "Seek Truth, Foster Originality, and Live up to the Name of a Teacher", with a mission to foster "Creativity, Character, Community". Today, ECNU is striving to transform itself into a world-class university, with a number of first-class disciplines and well-coordinated discipline programs development. The university also intends to lead in the development of China's teacher education, part of its faculty of education.

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# HOST

East China Normal University

## Workshop Chair

**Chair:** Prof. Jian Wu (East China Normal University)
**Co-Chair:** Prof. Kiyoshi Ueda (Tohoku University)

## Local Organizing Committee

Dr. Hongcheng Ni
Dr. Wenbin Zhang
Dr. Hui Li
Dr. Peifen Lu

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## Special Sponsorship: APS Poster Award

APS Advancing Physics

# GENERAL INFORMATION

**Conference Venue:** The Main Hall of the Optics Building in the ECNU Campus
**Address:** Optics Building, 500 Dongchuan Road, Minhang District, Shanghai（上海市闵行区东川路 500 号光学大楼）

## Speaker Preparation

Keynote talks are allotted 40 minutes, invited talks 25 minutes, and oral talks 20 minutes, including Q&A. We kindly request all oral speakers to arrive at the session room 15 minutes before their scheduled talk time for the uploading and verification of their presentation slides. Please note that the presentation language for all talks is English.

## Poster Preparation

Authors are required to be present at their poster during the session for interactive discussions. Please ensure that your poster ID is prominently displayed on the poster board. Students and early-career researchers have the chance to win the APS Poster Award, which will be announced during the workshop banquet. Poster presenters are kindly reminded to take down their posters at the conclusion of the session. The conference staff will not be responsible for collecting posters left behind.
**Poster session:** Monday, Apr. 22, 16:55-18:20
**Poster board size:** 0.95 m (length) * 2.47 m (height), recommended poster size: 0.8m*1.2m
**Set-up time:** Monday, Apr. 22, 08:00-16:55

## Registration

The registration will be open from 14:00 to 21:30 on April 21st at Marriot Courtyard.
If you are late for the registration, it is still possible right at the workshop venue in the Optics Building of the ECNU campus during the entire workshop.
Regarding the on-site payment, we accept credit card or cash payments for workshop registration fees.

## Welcome Reception

The welcome reception of the "2024 International Workshop on Ultra-Fast Science" (WUFS2024) is open for all registered attendees. A ticket is provided together with the badge.
**Location:** Rice Space 紫叶广场大米先生 , right next to Marriot Courtyard
**Time:** Sunday, Apr. 21, 17:00-20:30

## Commute

A shuttle bus will transport you between the conference venue at the Optics Building of ECNU campus and the Marriott Courtyard Hotel. The bus will depart from the hotel at 08:00 on the mornings of April 22nd and April 23rd.

## Workshop Banquet

The workshop banquet of the "2024 International Workshop on Ultra-Fast Science" (WUFS2024) is open for all registered attendees. You will have the opportunity to enjoy a delightful artistic program during the banquet.
A ticket is provided together with the badge.
**Location:** Ballroom, 3F Marriott Courtyard ( 紫竹万怡酒店三楼宴会厅 )

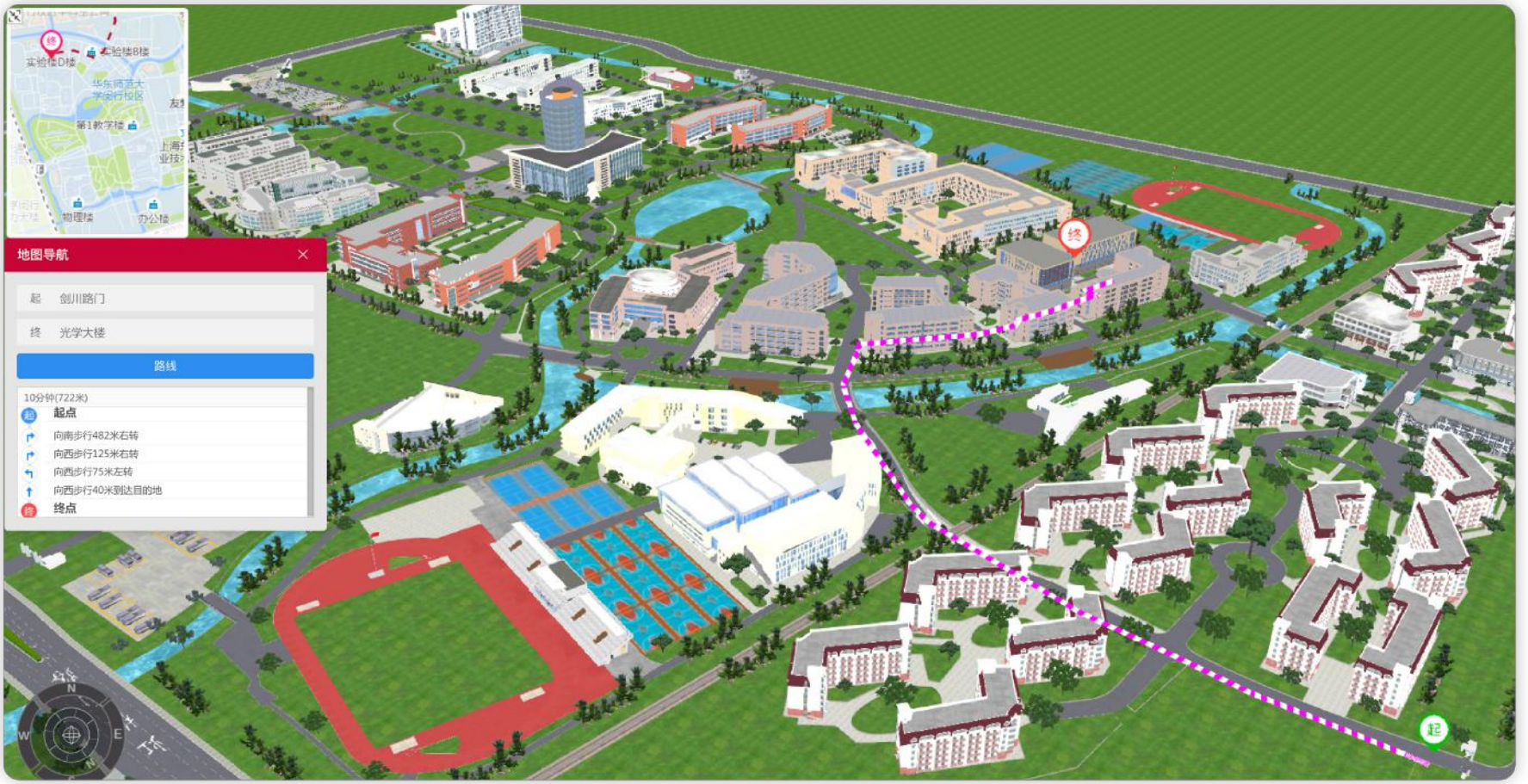
## APS Poster Award

The winners will be selected by reviewers on site during the poster session and announced during the workshop banquet.

## Excursion

On April 23rd, you are welcome to join us in the excursion to the Yuyuan Garden.
If you wish to go for the excursion, you may fill in a questionnaire during registration on April 21st.
Your passport number or ID number is needed when you fill in the questionnaire, as it is necessary for insurance during the excursion.
For those who opt not to participate in the excursion, you are dismissed from the workshop beginning the afternoon of April 23rd.
After the excursion, a shuttle bus will take the participants who will attend the following MPS2024 conference to the corresponding conference site.
For those who do not attend MPS2024, another shuttle bus will take you from the Yuyuan Garden back to the Marriot Courtyard hotel.
If you want to leave right from the Yuyuan Garden, you are also free to go.

# CONFERENCE MAP




Navigation map for those living in the Artels, located at the North Gateway of the ECNU Campus.

# PROGRAM AT A GLANCE

| Program | Sunday Apr.21 | Monday Apr.22 | Tuesday Apr.23 | Location |
|---|---|---|---|---|
| Registration | 14:00-21:30 | | | Hotel lobby |
| | | 08:00-18:00 | 08:00-12:00 | 3F, Optics Building |
| Technical Session | | 8:30-16:55 | 8:30-12:00 | 3F, Optics Building |
| Poster | | 16:55-18:20 | | 3F, Optics Building |
| Banquet | | 18:30-20:30 | | Marriott Courtyard |
| Lab Tour | | 10:20<br>14:55 | | B1, Optics Building |
| Welcome Reception | 17:00-20:30 | | | Rice Space |
| Excursion | | | 13:00-18:00 | Yuyuan Garden |

# PROGRAMS

| | |
|---|---|
| **22 April 2024, Monday** | |
| **Location: Main Hall, 3F, Optics Building, ECNU Campus** | |
| **Session 1** | |
| **Chair: Jian Wu, East China Normal University** | |
| 8:30-8:40 | **Opening Address** |
| 8:40-8:50 | Workshop Photo |
| 8:50-9:30 | **Ultrafast phases (Keynote)**<br>Reinhard Dörner, University of Frankfurt |
| **Session 2** | |
| **Chair: Michael Meyer, European XFEL** | |
| 9:30-9:55 | **Ultrafast powerful laser and driven radiation source (Invited)**<br>Yuxin Leng, Shanghai Institute of Optics and Fine Mechanics |
| 9:55-10:20 | **Probing ultrafast electron dynamics with attoclock (Invited)**<br>Xiaojun Liu, Innovation Academy for Precision Measurement Science and Technology |
| 10:20-10:35 | Coffee Break |
| **Session 3** | |
| **Chair: Till Jahnke, Max Planck Institute for Nuclear Physics** | |
| 10:35-11:00 | **Threshold fragmentation with multiphoton XUV pulses (Invited)**<br>Jan M. Rost, Max Planck Institute for the Physics of Complex Systems |
| 11:00-11:25 | **Attosecond coincidence spectroscopy in isotopic mixtures (Invited)**<br>Ioannis Makos, University of Freiburg |

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|---|---|
| 11:25-11:45 | **Strong-field ionization of laser-cooled Rubidium atoms (Oral)**<br>Difa Ye, Institute of Applied Physics and Computational Mathematics |
| 11:45-12:05 | **Probing ionization dynamics of Helium and Argon by attosecond photoelectron spectroscopy (Oral)**<br>Sizuo Luo, Jilin University |
| 12:05-13:00 | Lunch |
| **Session 4** | |
| **Chair: Xue-Bin Bian, Innovation Academy for Precision Measurement Science and Technology** | |
| 13:00-13:40 | **Attosecond chemistry: imaging coupled electronic and nuclear motions in molecules in real time (Keynote)**<br>Fernando Martín, Autonomous University of Madrid |
| 13:40-14:05 | **Strong-field ionization with quantum light (Invited)**<br>Yunquan Liu, Peking University |
| 14:05-14:30 | **Strong field excitation/ionization probed in attoseconds (Invited)**<br>Zengxiu Zhao, National University of Defense Technology |
| 14:30-14:55 | **Coherence and non-adiabaticity tracked with time-resolved x-rays (Invited)**<br>Oriol Vendrell, Heidelberg University |
| 14:55-15:10 | Coffee Break |
| **Session 5** | |
| **Chair: Kiyoshi Ueda, Tohoku University** | |
| 15:10-15:35 | **Ultrafast few-photon physics in few-body quantum systems: From tests of fundamental theory, via state-specific spectroscopy, to attosecond phase control of Bell-like states (Invited)**<br>Thomas Pfeifer, Max Planck Institute for Nuclear Physics |
| 15:35-16:00 | **Quantum coherence and entanglement in attosecond atomic and molecular photoionization (Invited)**<br>Vitali Averbukh, Imperial College London |
| 16:00-16:25 | **Measuring the quantum state of photoelectrons (Invited)**<br>David Busto, Lund University |

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|---|---|
| 16:25-16:40 | **How to publish with Nature journals (Editor)**<br>Lishu Wu, Nature Publishing Group |
| 16:40-16:55 | **Meet Phys Rev editor — Inside Physical Review X (Editor)**<br>Yun Li, American Physical Society |
| 16:55-18:20 | Poster |
| 18:30-20:30 | Banquet @ Marriott Courtyard |

| 23 April 2024, Tuesday | |
|---|---|
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| **Session 6** | |
| **Chair: Edwin Kukk, University of Turku** | |
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| 9:10-9:35 | **Carrier transport in 2D perovskite quantum wells (Invited)**<br>Shengye Jin, Dalian Institute of Chemical Physics |
| 9:35-9:55 | **Few-cycle pulse generation with post pulse compression by multiple thin water films (Oral)**<br>Pengfei Lan, Huazhong University of Science and Technology |
| 9:55-10:10 | Coffee Break |
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| **Chair: Hongcheng Ni, East China Normal University** | |
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| 10:35-11:00 | **Ultrafast two-electron orbital swap in Li initiated by attosecond pulses (Invited)**<br>Feng He, Shanghai Jiao Tong University |
| 11:00-11:20 | **Imaging the ultrafast coupled electron-nuclear dynamics of light-induced molecular fragmentation (Oral)**<br>Tian Wang, Nation Research Council |
| 11:20-11:40 | **Light-induced ultrafast dynamics of cold molecules in helium nanodroplets (Oral)**<br>Wenbin Zhang, East China Normal University |

| Session 8 | |
|---|---|
| **Chair: Jian Wu, East China Normal University** | |
| 11:40-12:00 | **Concluding Remarks** |
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| 13:00-18:00 | Excursion @ Yuyuan Garden |
| 18:00-20:30 | Welcome Reception @ MPS2024 |

# ABSTRACTS

## Ultrafast phases

Kang Lin[1,2], Sebastian Eckart[1], Hao Liang[3], Alexander Hartung[1], Sina Jacob[1], Jan Kruse[1], Dörte Blume[4], Lothar Ph. H. Schmidt[1], Markus S. Schöffler[1], Till Jahnke[1], Maksim Kunitski[1], Reinhard Dörner[1]

[1]*Insitute for Nuclear Physics, Goethe University Frankfurt, Germany*

[2]*School of Physics, Zhejiang Key Laboratory of Micro-Nano Quantum Chips and Quantum Control, Zhejiang University, Hangzhou 310058, China*

[3]*Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany*

[4]*Center for Quantum Research and Technology, University of Oklahoma, 440 W. Brooks St., Norman, OK 73019, USA*

**Synopsis**

Quantum particles moving in free space are characterized by wavepackets which have a characteristic time evaluation of their phase. We will show two examples how movies of such wavepackets including their phase can be made on femtosecond time scale.

- Time evolution of free moving Helium atoms emitted from rotating Helium Clusters [1]
- Time evolution free moving electron wavepackets emitted by tunnelionization [2]

**References**

[1] M. Kunitski, Q. Guan, H. Maschkiwitz, J. Hahnenbruch, S. Eckart, S. Zeller, A. Kalinin, M. Schöffler, L. Ph. H. Schmidt, T. Jahnke, D. Blume, R. Dörner Ultrafast manipulation of the weakly bound helium dimer, Nat. Phys., 17 (2021) 174-178

[2] Kang Lin, Sebastian Eckart, Hao Liang, Alexander Hartung, Sina Jacob, Qinying Ji, Lothar Ph. H. Schmidt, Markus S. Schöffler, Till Jahnke, Maksim Kunitski, Reinhard Dörner Ultrafast Kapitza-Dirac effect, Science, 383 (2024) 1467-1470

# Ultrafast powerful laser and driven radiation source

**Yuxin Leng, Xu Yi, and Ruxin Li**

*Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China*

A series of ultra-intense and ultra-fast lasers (0.2PW, 1PW and 10PW lasers) have been developed in SIOM (Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences), which have been in opening operation for the internal and international end users. Some laser driven particle accelerator and radiation have been carried out based on these lasers recently. For example, the laser driven electron accelerator has been achieved with near GeV energy and <1% energy spread. Further, a free-electron lasing using a laser wakefield electron accelerator has been demonstrated.

# Probing ultrafast electron dynamics with attoclock

**YanLan Wang, RenPing Sun, ShaoGang Yu, XuanYang Lai, Wei Quan, XiaoJun Liu***

*State Key Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Innovation Academy for Precision Measurement Science and Technology, Chinese Academy of Sciences, Wuhan 430071, China*

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Tracing the motion of the electrons on its characteristic timescale provides an effective way to identify and understand the essential features of many ultrafast photophysics and photochemical processes. Among attosecond metrology techniques, the attoclock [1,2] holds particular significance due to its self-referencing and compatibility with femtosecond lasers. In this talk, we will present our recent advancements in probing ultrafast electron dynamics of atoms and molecules with improved attoclock schemes [3-6].

**References**

[1] P. Eckle, M. Smolarski, P. Schlup, J. Biegert, A. Staudte, M. Schöffler, H. G. Muller, R. Dorner, and U. Keller, Nat. Phys. **4**, 565 (2008).

[2] P. Eckle, A. N. Pfeiffer, C. Cirelli, A. Staudte, R. Dörner, H. G. Muller, M. Buttiker, and U. Keller, Science **322**, 1525 (2008).

[3] Y. L. Wang, S. G. Yu, X. Y. Lai, H. P. Kang, S. P. Xu, R. P. Sun, W. Quan, and X. J. Liu, Phys. Rev. A **98**, 043422 (2018).

[4] W. Quan, V. V. Serov, M. Z. Wei, M. Zhao, Y. Zhou, Y. L. Wang, X. Y. Lai, A. S. Kheifets, and X. J. Liu, Phys. Rev. Lett. **123**, 223204 (2019).

[5] R. P. Sun, Y. L. Wang, Y. Zhou, S. G. Yu, S. P. Xu, X. Y. Lai, W. Quan, and X. J. Liu, Phys. Rev. A **105**, L021103 (2022).

[6] Y. L. Wang, S. G. Yu, R. P. Sun, H. P. Kang, S. P. Xu, X. Y. Lai, W. Quan, and X. J. Liu, to be published.

# Threshold fragmentation with multiphoton XUV pulses

**Jan M Rost**

*Max Planck Institute for the Physics of Complex Systems, Dresden, Germany*

Kinetic energy spectra $P(E)$ of charged particles resulting from interaction with intense XUV pulses may exhibit a characteristic shape governed by a Wigner power law [1] onset near threshold, $P(E) \propto E^{\alpha}$, followed by a Gaussian tail $P(E) \propto e^{-(E-E_o)^2/\Delta E^2}$.

Interaction of complex targets with intense laser pulses typically results in a disintegration of the target. Many charged particles, electrons and ions, leaving the interaction region, often with Maxwellian energy (velocity) distribution with an exponential tail, $P(E) \propto e^{-E/E_0}$, characteristic for some kind of thermalization through mutual interaction.

However, as we will demonstrate, also the exact opposite may happen: namely a Gaussian tail and a threshold onset with a Wigner power law. The latter implies that the charged particle leaves a neutral compound behind, while the Gaussian tail can have different dynamical reasons, as we illustrate with two examples: The first addresses escaping electrons and introduces a *zero-energy photo-electric effect* in the interaction of intense XUV pulses with negative ions. The second one has been already seen experimentally in an experiment at FLASH, where dimer ions have been detected with a *Wigner-Gauss shaped* kinetic energy spectrum following photo-absorption in fullerenes with photons of roughly 20 eV.

**References**

[1] E. P. Wigner, Phys. Rev. **73**, 1002 (1948).
[2] A. C. LaForge etal, submitted (2024).

# Attosecond coincidence spectroscopy in isotopic mixtures

**Ioannis Makos[1], Dominik Ertel[1], David Busto[1,2], Marvin Schmoll[1], Jakub Benda[3], Hamed Ahmadi[1], Matteo Moioli[1], Fabio Frassetto[4], Luca Poletto[4], Claus Dieter Schröter[5], Thomas Pfeifer[5], Robert Moshammer[5], Zdeněk Mašín[3], Serguei Patchkovskii[6], Giuseppe Sansone[1]**

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*[6]Max-Born-Institute for Nonlinear Optics and Short Pulse Spectroscopy, Berlin, 12489, Germany.*

Attosecond photoelectron interferometric techniques have been employed over the past decade to investigate dynamics in molecular systems following photoionization. These techniques offer valuable insights into electron correlation effects and coupled electronic-nuclear dynamics [1]. Additionally, the combination of two-color interferometric techniques with photoelectron-photoion coincidence spectroscopy allows for angle-resolved studies in the recoil frame, revealing information about the anisotropy of the molecular potential [2].

In this study, we investigate photoionization dynamics in an isotopic gas mixture of methane ($CH_4$) and deuteromethane ($CD_4$), utilizing attosecond coincidence spectroscopy. The absorption of an extreme ultraviolet (XUV) photon originating from an attosecond pulse train populates a manifold of cationic states. An additional absorption or emission of an infrared (IR) photon gives rise to a two-color photoionization spectrogram. A collinear configuration, presenting an attosecond stability, is utilized to control the relative delay between the XUV and the IR pulses, which co-propagate in a reaction microscope where they interact with the gas target [3].

The investigation of the mixture of $CH_4$ - $CD_4$ using attosecond photoelectron interferometry gives access to the nuclear response of the two isotopologues, since the electronic properties of the two molecular systems are not significantly affected by the two-color field. By looking into the angle-integrated amplitude and contrast of the oscillations of the photoelectron peaks, we obtain information regarding the nuclear dynamics upon photoionization, as well as the effect of nuclear motion on the correlated electronic-nuclear dynamics [4]. Additionally, we investigate the angle-resolved phase of the photoionization process by fitting the oscillating component and the phase of the two-color signal using complex asymmetry parameters [5].

**References**

[1] a) G. Sansone *et al.*, Nature **465**, 763 (2010); b) R. Pazourek, S. Nagele, and J. Burgdörfer, Rev. Mod. Phys. **87**, 765 (2015); c) M. Nisoli *et al.*, Chemical reviews **117**, 10760 (2017).
[2] a) J. Vos *et al.*, Science **360**, 1326 (2018); b) H. Ahmadi *et al.*, Nat Commun **13** (2022); c) X. Gong *et al.*, Phys. Rev. X **12** (2022).
[3] D. Ertel *et al.*, The Review of scientific instruments **94** (2023).
[4] a) X. Gong *et al.*, Nat Commun **14**, 4402 (2023); b) D. Ertel *et al.*, Sci. Adv. **9**, eadh7747 (2023).
[5] a) D. Ertel *et al.*, J. Phys. Chem. A **128**, 1685 (2024); b) S. Saha *et al.*, Phys. Rev. A **104** (2021).

# Strong-field ionization of laser-cooled Rubidium atoms

**Linxuan Zhang[1,2 †], Huanyu Ma[3,4,5 †], Xincheng Wang[3, *], Zhihan Zou[3,4,5], Rujin Lv[4,5], Ahai Chen[3], Kiyoshi Ueda[3,6], Difa Ye[1, *], Yuhai Jiang [3,4, *]**

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Alkali metal atoms such as Rubidium (Rb) are generally applied to achieve Bose-Einstein condensation and fabricate atomic clock due to their specific energy levels suitable for laser cooling and magneto-optical trapping. If subsequently exposed to another ultrashort laser pulse, they can generate cold ions and electrons nearly instantaneously, which offers a fascinating playground to the study of ultracold neutral plasmas and hybrid atom-ion systems, a candidate for quantum computation. In the above studies, the knowledge about the ionization of cold atoms first cooled by a relatively longer yet weaker laser and subsequently stripped by another shorter yet stronger laser field is crucial and fundamental.

Recently, with the help of a magneto-optical trap recoil ion momentum spectroscopy (MOTRIMS) platform, we are able to extend the study of various strong-field dynamics such as the above threshold ionization and electron recollision, to alkali atoms. In our work, photoionization of rubidium atoms cooled in a magneto-optical trap, characterized by the coexistence of the ground $5S_{1/2}$ and excited $5P_{3/2}$ states, is investigated experimentally with a 400-nm (or 800-nm) femtosecond laser pulses at intensities of the order of $10^{12}$W/cm$^2$. The recoil-ion momentum distribution (RIMD) of $Rb^+$ exhibits rich ringlike structures. Numerical simulations by solving the time-dependent Schrödinger equation (TDSE), developed to take into account the quantum coherence introduced by the six cooling laser beams, can successfully explain the experimental measurements, not only disentangling the contribution by the ground and the excited states but also helping to determine the population of the excited state during the cooling cycle. When applying an elliptically polarized strong laser field, we show that the angular streaking technique can be applied to retrieve the Coulomb phase shift (or equivalently, the Wigner time delay) of the photoelectric effect, and to study the Autler-Townes splitting in the time domain.

We also study the multiple ionization of Rb atoms driven by circularly polarized intense laser fields and find that although the outmost electron is loosely bound and can be easily ionized during the rising edge of the laser pulse, it is not just a spectator witnessing the subsequent multiple ionization of the ion. This early released electron (the shepherd electron) is driven back and forth at the vicinity of the inner shell electrons and can lead to some novel phenomena: (i) a double-knee structure on the ion yield curve; (ii) a halo structure on the differential ion momentum spectrum; (iii) the emergence of a transient hollow atom. The findings might have applications in coherent extreme-ultraviolet light amplification.

**References**

[1] Shiwei Liu, Difa Ye, Yuhai Jiang, and Jie Liu, Communications Physics **6**, 282 (2023).
[2] Huanyu Ma, Xincheng Wang, Linxuan Zhang et al., Physical Review A **107**, 033114 (2023).
[3] Linxuan Zhang et al., in preparation.

# Probing ionization dynamics of Helium and Argon by attosecond photoelectron spectroscopy

**Sizuo Luo,* Mingxuan Li, Huiyong Wang, Jieda Zhang, Dajun Ding**

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**Synopsis**

The photoionization of helium and argon, along with the phenomenon of laser-assisted interference of electrons and the ionization time delay of electrons from two shells, has been investigated using attosecond photoelectron spectroscopy.

Attosecond photoelectron spectroscopy (APS) has emerged as a powerful tool for investigating ultrafast electron dynamics in atoms and molecules. In this study, we employ APS to probe the ionization dynamics of two noble gases, helium (He) and argon (Ar), shedding light on their distinct electron behavior. By employing attosecond time-resolved techniques, we investigate the intricate interplay of laser field-induced ionization processes of He and the ionization time delay between 3s and 3p valance shells across the Cooper minimum of Ar. Our experimental setup allows for the characterization of attosecond-scale electron dynamics, providing insights into the fundamental mechanisms governing ionization in noble gases. The observed photoelectron spectra from He exhibit energy-domain fringes corresponding to interference from temporal slits, modifiable by additional IR laser fields. The relative attosecond delay between electrons from the 3s and 3p shells validates predictions of the RPAE[1,2] (Random Phase Approximation with Exchange) and reveals discrepancies with the results from TDLDA[3] (Time-Dependent Local Density Approximation) as given in Fig 1, indicating potential electron correlation effects between inner shells that may influence the photoionization matrix element of 3s. The findings presented herein not only deepen our understanding of fundamental atomic processes but also offer valuable insights into the development of advanced laser-driven technologies and attosecond science applications.

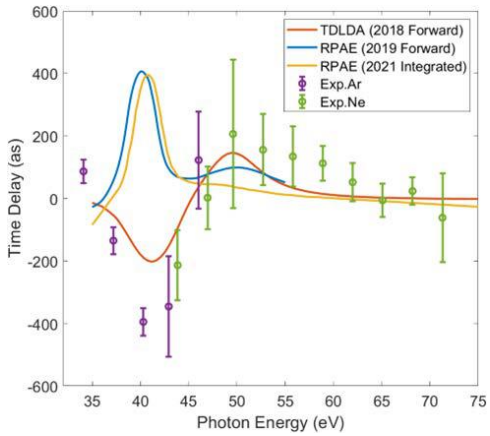


Fig.1. Measured and calculated ionization time delay between 3s and 3p valance shells of Argon.

**References**

[1] Jimmy Vinbladh et al., Phys. Rev. A **100**, 043424 (2019).

[1] C. Alexandridi et al., Phys. Rev. Research **3**, L012012 (2021).

[2] Liang-Wen Pi et al., Appl. Sci. **8**, 322 (2018).

# Attosecond chemistry: imaging coupled electronic and nuclear motions in molecules in real time

**Fernando Martín**[1,2]

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From water to DNA, all chemical reactions involve breaking and forming bonds, in which atomic nuclei are forced to live close to each other or to separate forever. But this is the consequence of the way electrons move. Thus, chemical reactivity results from the combined action of the "fast" electronic motion and the "slow" motion of atomic nuclei. Following the motion of the latter was possible by the end of the twentieth century with the help of femtosecond laser pulses. With the advent of attosecond light pulses at the dawn of the twenty first century, access to the time scale of electronic motion, i.e., the ultimate time scale responsible for chemical transformations, was finally at our reach. This was accomplished in 2010 [1] for the simplest molecule in nature, hydrogen, and, in 2014 [2], for phenylalanine amino acid. Since then, the field has grown exponentially, leading to a discipline called attochemistry [3]. In this talk, I will review some of the most recent experimental and theoretical achievements in attosecond chemistry. All these guided by theoretical modelling [4], which has been an essential ingredient since the very beginning of this discipline. Attochemistry is still at its infancy, but its long-term goal, achieving control of chemical processes by acting on electronic motion at its natural time scale does not seem to be a remote possibility anymore.

**References**

[1] G. Sansone, F. Kelkensberg, J. F. Pérez-Torres, F. Morales, M. F. Kling, W. Siu, O. Ghafur, P. Johnsson, M. Swoboda, E. Benedetti, F. Ferrari, F. Lépine, J. L. Sanz-Vicario, S. Zherebtsov, I. Znakovskaya, A. L'Huillier, M. Yu. Ivanov, M. Nisoli, F. Martín, and M. J. J. Vrakking, Nature **465** 763 (2010).
[2] F. Calegari, D. Ayuso, A. Trabattoni, L. Belshaw, S. De Camillis, S. Anumula, F. Frassetto, L. Poletto, A. Palacios, P. Decleva, J. B. Greenwood, F. Martín, and M. Nisoli, Science **346**, 336 (2014).
[3] M. Nisoli, P. Decleva, F. Callegari, A. Palacios, and F. Martín, Chem. Rev. **117**, 10760 (2017).
[4] A. Palacios and F. Martín, WIREs Comput. Mol. Sci. **10**, e1430 (2020).

# Strong-field ionization with quantum light

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Strong-field physics is a prosperous research field that accesses the ultrafast electronic processes of atoms and molecules occurring with the natural electronic time resolution, while the quantum effects of driving light have been rarely revealed. Very recently, the establishment of a full-quantum description has aroused, but mainly concentrated on high harmonic generation. As the intrinsic basic process of high harmonic generation, exploring the quantum-optical nature in strong-field ionization is indispensable for understanding the intense-light-matter interaction.

In the talk, we show the study on photoelectron momentum distribution with squeezed state of light by developing a quantum-optical-corrected strong-field approximation model 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 [1]. Then we extend the strong-field ionization theory to the regime of spin-squeezed light and show that the squeezing of spin substantially alters the ultrafast dynamics of tunneling electrons. The formation of additional interference channels leads to a single-lobe photoelectron momentum spectrum in phase-spin-squeezed state laser, which can be utilized to establish the quantum-statistical attoclock. Furthermore, we discuss the quantum optics nature of strong-field ionization with two kinds of bunched lights, i.e., bright squeezed vacuum and thermal light, and show that the bunched lights not only enhance the ionization rates of photoelectrons, but also alters its statistics of momentum.

In addition, we will also present the work on the quantum effects induced by the free-electron–photons interaction and proposed a scheme to generate optical cat states based on the quantum interference of multi-path interactions [2]. By performing a projection measurement on the electron, the state of light collapses into a non-Gaussian state with oscillating Wigner negativity, and the optical cat states are successfully generated at the oscillation peaks.

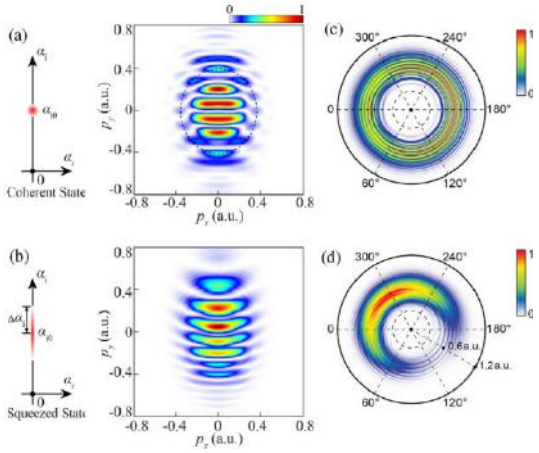


Fig. 1. Photoelectron momentum distribution driven by linearly polarized (a) coherent state, (b) squeezed state of light, and circularly polarized (c) coherent state, (d) squeezed state of light.

**References**

[1] Y. Fang, F. Sun, Q. He, Y. Liu, Phys. Rev. Lett. **130**, 253201 (2023).

[2] F. Sun, Y. Fang, Q. He, Y. Liu. Sci. Bull. **68**, 1366-1371 (2023).

# Strong field excitation/ionization probed in attoseconds

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Strong field excitation/ionization has been long considered the other extreme of light-matter interaction, in contrast to resonant excitation by weak continuous light used in quantum optics. In this talk, I will focus on the bridging of the two extremes and demonstrate that their interplay plays a critical role in strong field quantum optics due to the multiple dimensional freedom of ultrafast dynamics such as electronic, vibration, rotation and spin-orbit coupling. I will briefly introduce our recent achievement on the generation of isolated attosecond pulse with 51 as duration and then emphasize on the attosecond probing of the ionic population dynamics, the evolution of quantum coherence and the resulted coherent emission with all of which involves of the quantum optics behaviors of atoms and molecules in strong laser fields.

**References**

[1] L. Wang, G. Bai, X. Wang, J. Zhao, C. Gao, J. Wang, F. Xiao, W. Tao, P. Song, Q. Qiu, J. Liu, and Z. Zhao, Nature Communications **15**, 27205 (2024).

[2] Q. Zhang, H. Xie, G. Li, X. Wang, H. Lei, J. Zhao, Z. Chen, J. Yao, Y. Cheng, and Z. Zhao, Communications Physics **3**, 50 (2020).

[3] H. Lei, J. Yao, J. Zhao, H. Xie, F. Zhang, H. Zhang, N. Zhang, G. Li, Q. Zhang, X. Wang, Y. Yang, L. Yuan, Y. Cheng, and Z. Zhao, Nature communications **13**, 4080 (2022).

# Coherence and non-adiabaticity tracked with time-resolved x-rays

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**Synopsis**

We consider theoretically the concept of electronic coherence in non-adiabatic molecular dynamics and how this can be triggered and probed using ultrafast x-rays.

Under adiabatic conditions, the electrons follow the nuclei instantaneously and, in turn, the nuclei move on a single potential determined by the electronic cloud. In this regime, the electronic states are well separated compared to typical vibrational energies. However, most photo-physics and photo-chemistry take place under conditions where this approximation breaks down, with dramatic consequences for the short-time dynamics after photo-absorption, the stability of photo-excited molecules, and the underlying mechanisms of charge and energy transfer triggered by light.

In this talk, we shortly review the theoretical description of quantum molecular dynamics, the relation between non-adiabatic phenomena and electronic coherence following photo-absorption, and opportunities to track these dynamics using ultrafast x-rays. [1,2,3].

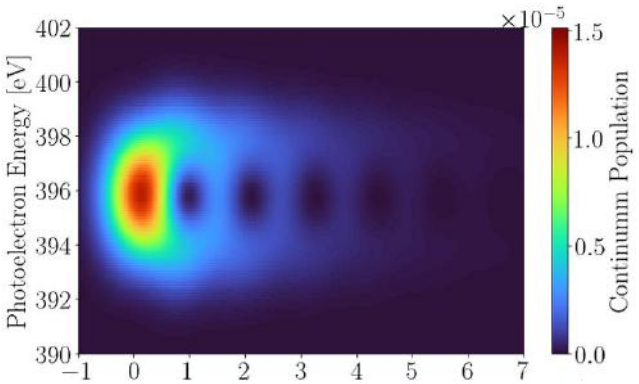


Fig. 1. Photoelectron yield of coherently core-excited $N_2O$ at the nitrogen K-edge

**References**

[1] Freibert, A.; Mendive-Tapia, D.; Huse, N.; Vendrell, O. *Femtosecond X-Ray Absorption Spectroscopy of Pyrazine at the Nitrogen K-Edge: On the Validity of the Lorentzian Limit.*
J. Phys. B: At. Mol. Opt. Phys. 2021, **54**, 244003 https://doi.org/10.1088/1361-6455/ac3846.

[2] Freibert, A.; Mendive-Tapia, D.; Huse, N.; Vendrell, O. *Time-Dependent Resonant Inelastic X-ray Scattering of Pyrazine at the Nitrogen K-Edge: A Quantum Dynamics Approach*.
J. Chem. Theory Comput. 2024, **20**, 2167-2180 https://doi.org/10.1021/acs.jctc.3c01259

[3] Rodriguez-Cuenca, E; Picón, A; Oberly, S; Kuleff, A.I.; Vendrell, O. *Core-hole Coherent Spectroscopy in Molecules*, (under review)

# Ultrafast few-photon physics in few-body quantum systems: From tests of fundamental theory, via state-specific spectroscopy, to attosecond phase control of Bell-like states

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**Synopsis**

Extreme-ultraviolet light from high-harmonic generation (HHG), a free-electron laser (FEL) and their combination are employed to observe fundamental nonlinear and time-dependent quantum effects in atoms and small molecules for testing our theory understanding,

The interaction of light with atoms is a testing ground for our understanding of fundamental quantum physics. At low light intensities, precision spectroscopy of atoms and ions allows for verification of quantum electrodynamics and tests of the standard model. Moving to higher intensities, nonlinear interactions of atoms and small molecules with two or multiple photons already challenge our understanding of time-dependent quantum dynamics, as the coupling of several excited or even continuum states to light substantially increases complexity and thus the demand on theory, but at the same time creates new opportunities to advance our understanding of (sub-)atomic quantum control. To experimentally shed light on these processes, we employ a range of experimental methodologies including reaction microscopes and time-resolved spectroscopy of small quantum systems driven by visible, HHG, and FEL light,and combinations thereof.

Angle-resolved two-photon ionization of helium near a single-photon forbidden doubly excite state was performed by recoil-ion imaging with a reaction microscope [1]. This was enabled by a ghost-imaging related spectroscopy technique that we implemented at the free-electron-laser (FEL) FLASH at DESY, Hamburg. Recording the statistically produced extreme-ultraviolet (XUV) spectrum for every shot of the FEL, it was possible to enhance the spectral resolution and to select resonant events.

By combining FEL with HHG light in a transient-absorption experiment, we tracked ionic and neutral-atom states of dissociating oxygen molecules as a function of time [2]. This allowed us to directly measure the life time of a predissociative state, which decays by the tunneling of the oxygen atoms out of the local minimum of a molecular potential energy curve.

We also performed two-particle coincidence detection of electrons and ions in a reaction-microscope experiment on hydrogen molecules interacting with a combination of HHG and visible laser pulses [3]. We observed a characteristic asymmetry of the relative emission direction of electrons and protons varying on

an attosecond time scale. The origin of this phenomenon was found in the ultrafast generation and phase control of parity-entangled states of a free and a bound electron.

**References**

[1] M. Straub, T. Ding, M. Rebholz, G. D. Borisova, A. Magunia, H. C. Lindenblatt, S. Meister, F. Trost, Y. Wang, S. Palutke, M. Braune, S. Düsterer, R. Treusch, C. H. Greene, R. Moshammer, T. Pfeifer, and C. Ott
*Differential Measurement of Electron Ejection after Two-Photon Two-Electron Excitation of Helium*
Physical Review Letters **129**, 183204 (2022)

[2] A. Magunia, M. Rebholz, E. Appi, C. C. Papadopoulou, H. Lindenblatt, F. Trost, S. Meister, T. Ding, M. Straub, G. D. Borisova, J. Lee, R. Jin, A. von der Dellen, C. Kaiser, M. Braune, S. Düsterer, S. Ališauskas, T. Lang, C. Heyl, B. Manschwetus, S. Grunewald, U. Frühling, A. Tajalli, A. B. Wahid, L. Silletti, F. Calegari, P. Mosel, U. Morgner, M. Kovacev, U. Thumm, I. Hartl, R. Treusch, R. Moshammer, C. Ott, and T. Pfeifer
*Time-resolving state-specific molecular dissociation with XUV broadband absorption spectroscopy*
Science Advances **9**, eadk1482 (2023)

[3] F. Shobeiry, P. Fross, H. Srinivas, T. Pfeifer, R. Moshammer, A, Harth,
Sub-femtosecond optical control of entangled states
arxiv.org/abs/2110.06668

# Quantum coherence and entanglement in attosecond atomic and molecular photoionization

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**Synopsis** Atomic and molecular photoionization by attosecond pulses may lead to creation of coherent superpositions of ionic states and as a result to coherent dynamics of the electron hole taking the form of hole migration in molecular ions or electron cloud breathing in atomic ions. The quantum entanglement between the photoelectron and the ion which precludes the ionic state coherence can be probed and verified by the Bell tests.

Molecular photoionization by attosecond pulses may lead to creation of coherent superpositions of ionic states by coupling of a series of such states to the same continuum wavefunction of the photoelectron. The temporal extent of the resulting coherent hole dynamics that can be seen as a migration of the electron hole across the molecular volume [1] is limited by decoherence stemming from the zero-point energy of the nuclear vibrations, the nuclear motion itself, as well as by the electronic decay of the ionic wave packet components above the double ionization threshold. In this presentation, I will discuss the theoretical interpretation of the recent hole migration experiments performed at the LCLS [2] and FLASH [3] X-ray free electron laser facilities and targeting the electronic observables, such as time-dependent Auger electron signal [4]. I will also describe our progress in developing the *ab initio* many-electron theoretical tools, such as B-spline ADC and Fano-ADC [5,6], that allow us to gain insight into the mechanisms of the onset and decay of the coherent hole dynamics. Combining application of such *ab initio* tools with analytical modelling has led us to propose a number of new spectroscopic approaches for direct observation of coherent many-electron dynamics in ionized systems, such as interferometric measurement of Auger decay dynamics and of quantum revivals below the double ionization threshold [7,8], as well as pump-probe spectroscopy of Auger decay [9]. While these schemes still await their experimental realization, attosecond measurement of the resonant Auger decay of a coherent superposition of core-excited states has been very recently achieved by the angular streaking technique [10].

A central role in the generation of the ionic coherence belongs to the quantum entanglement be-tween the photoelectron and the atomic or molecular ion. We have developed and simulated numerically a Bell test for probing the quantum entanglement in photoionization [11]. We have designed and simulated the quantum protocol for entanglement quantification for the case of noble gas atoms photoionized by ultrashort, circularly polarized infrared laser pulses in the strong-field regime, demonstrating robust violation of the Bell inequality. The Bell test developed in our work detects entanglement between the

internal states of the $Ar^+$ and the spin states of the photoelectron by exploiting the spin polarization of the photoelectron beam.

**References**

[1] Calegari F *et al.* 2016 *J. Phys. B: At. Mol. Opt. Phys.* **49**, 142001

[2] Barillot T *et al.* 2021 *Phys. Rev. X* **11**, 031048

[3] Schwickert D *et al.* 2022 *Sci. Adv.* **8**, eabn6848

[4] Cooper B *et al.* 2014 *Faraday Discussions* **171**, 2014

[5] Averbukh V and Ruberti M 2018 *Theoretical and Computational Chemistry Series* **13**, 68

[6] Kolorenč P and Averbukh V 2020 *J. Chem. Phys.* **152**, 214107

[7] Khokhlova M *et al.* 2019 *Phys. Rev. Lett.* **122**, 233001

[8] Tarrant J, Khokhlova M and Averbukh V 2022 *J. Chem. Phys.* **157**, 054304

[9] Kissin Y, Ruberti M, Kolorenč P and Averbukh V 2021 *PCCP* **23**, 12376

[10] Li S *et al.* 2022 *Science* **375**, 285

[11] Ruberti M, Averbukh V and Mintert F, arXiv:2312.05036

# Measuring the quantum state of photoelectrons

**Hugo Laurell[1], Sizuo Luo[1], Robin Weissenbilder[1], Mattias Ammitzböll[1], Shahnawaz Ahmed[2], Hugo Söderberg[1], C. Leon M. Petersson[3], Vénus Poulain[1], Chen Guo[1], Christoph Dittel,[4,5,6], Daniel Finkelstein-Shapiro[7], Richard J. Squibb[8], Raimund Feifel[8], Mathieu Gisselbrecht[1], Cord L. Arnold[1], Andreas Buchleitner[4,5], Eva Lindroth[3], Anton Frisk Kockum[2], Anne L'Huillier[1], David Busto[1]**

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**Synopsis**

We use quantum state tomography to measure the density matrix describing the quantum state of photoelectrons emitted from helium and argon atoms. A reduced photoelectron purity is measured in argon due to ion-photoelectron entanglement.

The photoelectric effect is a fundamental quantum phenomenon that is used in various spectroscopy and microscopy techniques. 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[1]. This description of the photoelectrons is appropriate when they can be described by a wavefunciton, i.e. the photoelectron quantum state is pure. However if the photoelectron quantum state is mixed, the photoelectron must be described by a density matrix.

In this work, we use photoelectron quantum state tomography to measure the density matrix of photoelectrons emitted from helium and argon atoms. The technique relies on ionization with a short extreme ultraviolet pulse and probing with a tunable bichromatic probe pulse [2]. Our measurements show that, while the quantum state of photoelectrons emitted from helium atoms is pure, in argon the photoelectron are in a mixed quantum state due to ion-photoelectron entanglement induced by spin-orbit interaction [3]. Our results pave the way for the development of photoelectron quantum state spectroscopy.

**References**

[1] R. Pazourek *et al.*, Rev. Mod. Phys. **87**, 765 (2015)

[2] Laurell *et al.*, Phys. Rev. Research **4**, 033220 (2022)

[3] Laurell *et al.*, arXiv : 2309. 13945 (2023)

# Attosecond chemistry of isolated and solvated molecules1

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**Synopsis**

Experimental advances in the field of attosecond chemistry, including their recent expansion into the liquid phase and molecular clusters will be presented.

Attosecond charge migration is a purely electronic process that causes a periodic rearrangement of the charge distribution in molecules. It is driven by a coherent superposition of electronic states and therefore takes place on attosecond time scales (see, e.g. [1]). Charge migration usually couples to structural dynamics and can then result in permanent charge transfer. I will discuss the experimental observation of decoherence and revival of attosecond charge migration driven by nuclear motion, as well as the transfer of electronic coherence through a conical intersection in the neutral silane molecule [2]. These results demonstrate a broadly applicable approach to inducing and probing charge migration in neutral molecules, opening the door to controlling molecular dynamics on the electronic time scale, which defines the field of "attochemistry". One of the remaining challenges for attochemistry experiments is their extension to complex systems, such as larger molecules, molecular aggregates or nanoparticles in solution. X-ray spectroscopy offers an attractive approach to this goal, owing to its element specificity and site sensitivity. In this context, I will discuss the development of table-top soft-X-ray spectroscopy, which has led to the current world record of the shortest pulse of light (43 as) [3] and its application to observing the rearrangement of unoccupied molecular states during chemical reactions [4]. Turning from the gas phase to the liquid phase, I will discuss recent results on the observation of femtosecond proton transfer in ionized urea dimers in aqueous solution [5]. We find that ionization of concentrated aqueous urea solutions initially creates an electron hole that is delocalized over urea dimers. This hole subsequently localizes and defines which molecule transfers the proton. This is, to our knowledge, the first direct observation of charge-directed reactivity. The partial delocalization of electron holes created by ionization is not unique to urea dimers, but likely to be a general phenomenon in hydrogen-bonded systems. Applying attosecond interferometry to a liquid jet, we have measured delays of 50-70 as between photoemission from liquid water and isolated water molecules. These delays were found to be dominated by the first two solvation shells of the ionized molecule [6]. Applying attosecond coincidence spectroscopy to water clusters, the delays between size-resolved water clusters and water monomers has been measured. These measurements have revealed a linear relationship between the measured time delays and the spatial extension of the electron hole, which increases from the monomer to the tetramer, but does not further increase towards larger clusters [7]. These measurements suggest that photoionization delays are sensitive to the spatial delocalization of electronic wave functions, which opens the perspective of tracking attosecond electron-hole dynamics and how they control charge-directed reactivity in solutions. Most recently, using nitrogen K-edge transient absorption, we have observed the creation of electronic coherence at conical intersections in pyrazine and its complete decoherence when pyrazine is dissolved in aqueous solution [8].

**References**

[1] P. M. Kraus et al., *Science* **350,** 790 (2015) [2] D. Matselyukh et al., *Nature Physics* **18**, 1206 (2022) [3] T. Gaumnitz et al., *Opt. Exp.* **25**, 27506 (2017) [4] Y. Pertot et al., *Science* **355**, 264 (2017) [5] Z. Yin et al. *Nature* **619**, 749 (2023) [6] I. Jordan et al., *Science* **369**, 974 (2020) [7] X. Gong et al., *Nature* **609**, 507 (2022) [8] Y.-P. Chang et al., arXiv:2402.10508 (2024).

# Carrier transport in 2D perovskite quantum wells

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Two dimensional (2D) layered hybrid perovskites are naturally formed multiple quantum well (QW) materials, which hold great promise for many optoelectronic devices. In this presentation, I will introduce our recent works on carrier transport dynamics in 2D layered perovskites. By using time-resolved spectroscopic techniques, we found that the carriers within a QW plane performed a trap-enabled transport over a few micrometers, which is a distance comparable with that in 3D perovskites; on the other hand, the electron charges in a QW plane can overcome the energy barrier and transfer to the adjacent QWs trough the Auger-recombination of excitons. These results enlighten the fundamental photophysical properties of 2D perovskites and provide guidelines for designing this class of materials.

# Few-cycle pulse generation with post pulse compression by multiple thin water films

**Pengfei Lan**
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High-energy, few-cycle laser pulses are essential for numerous applications in the fields of ultrafast optics and attosecond physics. In this talk, different from the traditional hollow-core fibers and multiple thin solid plates, I will present a method to compress the pulse duration of the multi-cycle Ti:sapphire laser pulse to 4 fs by using multi-thin liquid films as the nonlinear media. I will show that continuum covers a range from 380 to 1050 nm, corresponding to a Fourier transform limit pulse width of 2.5 fs, when 35 fs Ti:sapphire laser pulse is applied on the multi-thin liquid films. The output pulses are compressed to 3.9 fs by employing chirped mirrors. Furthermore, a continuous high-order harmonic spectrum up to the 33rd order is realized by focusing the compressed laser pulses to Kr gas jet. Finally, I will present an all-optical method to measure the pulse duration of the isolated attosecond pulse.

# Control of Attosecond Entanglement and Coherence

**Marc Vrakking**

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Attosecond science is a branch of ultrafast laser physics that aims to investigate and possibly control electronic motion on its natural timescale by means of pump-probe experiments. Attosecond pulses are formed by the process of high-harmonic generation. Their generation and characterization were recently recognized by the 2023 Physics Nobel Prize, which was given to Anne L'Huillier, Pierre Agostini and Ferenc Krausz.

Attosecond pulses have wavelengths in the extreme ultra-violet (XUV) to soft X-ray spectral range. Accordingly, attosecond pulses are ionizing radiation for any medium (solid, liquid or gaseous) that is placed in its path. Photoionization splits a quantum system under investigation into an ion and a photoelectron. The ion and photoelectron will commonly display quantum-mechanical entanglement, which influences the coherence that attosecond pump-probe experiments rely on. In my talk I will discuss experimental and numerical work demonstrating the role of ion-photoelectron entanglement in attosecond pump-probe experiments, by taking as an example the vibrational and electronic wave packet dynamics that is induced in $H_2^+$ cations upon ionization of $H_2$ by an attosecond laser pulse [1-4]. I will show how tailoring the properties of the attosecond pulses (i.e. forming a pair of these pulses, or chirping these pulses) can be used to control the degree of ion-photoelectron entanglement that occurs, as indicated by the degree of vibrational, respectively electronic coherence that can be observed in the ion. In the calculations, the conclusions are furthermore supported by evaluation of the purity and a Schmidt decomposition of the ion + photoelectron wave function that results from the ionization process.

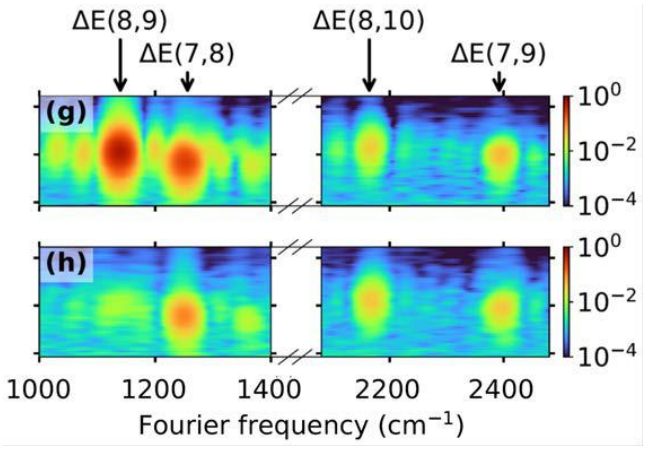


Fig. 1. Fourier Transform Power spectra revealing vibrational coherences that occur in a pump-probe experiment on H2 molecules. The upper and lower data set was recorded using two different delays between the attosecond pulse trains that were used to ionize the molecule. The absence of a number of vibrational coherences in the lower figure is due to ion-photoelectron entanglement.

**References**

[1] M.J.J. Vrakking, *Control of Attosecond Entanglement and Coherence.* Physical Review Letters, 2021. 126(11): p. 113203.

[2] L.-M. Koll, et al., *Experimental Control of Quantum-Mechanical Entanglement in an Attosecond Pump-Probe Experiment.* Physical Review Letters, 2022. 128(4): p. 043201.

[3] M.J.J. Vrakking, *Ion-photoelectron entanglement in photoionization with chirped laser pulses.* Journal of Physics B, 2022. 55(13).

[4] L.-M. Koll, et al., *The role of ion-photoelectron entanglement in electron localization following attosecond ionization of $H_2$ (working title).* (in preparation), 2024.

# Ultrafast two-electron orbital swap in Li initiated by attosecond pulses

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A universal mechanism of ultrafast 2-electron orbital swap is discovered through 2-photon sequential double ionization of Li. After a 1s electron in Li is ionized by absorbing an extreme ultraviolet photon, the other 2 bound electrons located on 2 different shells have either parallel or antiparallel spin orientations. In the latter case, these 2 electrons are in the superposition of the singlet and triplet states with different energies, forming a quantum beat and giving rise to the 2-electron orbital swap with a period of several hundred attoseconds. The orbital swap mechanism can be used to manipulate the spin polarization of photoelectron pairs by conceiving the attosecond-pump attosecond-probe strategy and thus serves as a knob to control spin-resolved multielectron ultrafast dynamics.

References

[1] H. Jiang, Z. H. Zhang, Y. Li, C. Ruiz, and F. He, Ultrafast Two-Electron Orbital Swap in Li Initiated by Attosecond Pulses, Ultrafast Sci. **3**, 0028 (2023).
[2] Y. N. Yang, S. Q. Chen, Z. H. Zhang, H. Jiang, M. Chen, Y. Li, and F. He, Harmonic Suppression Induced by Three-Electron Dynamics of Li in Strong Laser Fields, Phys. Rev. Lett. **131**, 183201 (2023).

# Imaging the ultrafast coupled electron-nuclear dynamics of light-induced molecular fragmentation

**Tian Wang[1], Nida Haram[1], Zack Dube[1], Yonghao Mi[1], Fatemeh Mousavi Karimi[1], Andrei Yu. Naumov[1], Giulio Vampa[1], Caterina Vozzi[2], Albert Stolow[3], Michael Schuurman[4], David M. Villeneuve[1], Paul B. Corkum[1], André Staudte[1]**

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**Synopsis**

By using Cold Target Recoil Ion Momentum Spectroscopy and a pump-probe experiment, we follow the breaking of the molecular bond in dissociating $Br_2$ by simultaneously observing photoion and photoelectron. The different dynamical stages are identified by the photoelectron momentum spectra.

In a chemical-bond breaking process, the valence electrons within the bond undergo rearrangement, leading to a corresponding rearrangement and separation of the atoms in a molecule[1]. Various methods can be employed to independently measure the dynamics of valence electrons [2,3] and atoms [4] in such instances. However, observing the dynamic coupling between valence electrons and atoms during a chemical reaction necessitates the simultaneous monitoring of both[5].

As a representative example of a photochemical reaction, the neutral dissociation of $Br_2$ has been employed to investigate diverse imaging methods for tracking the dynamics of valence electrons and atoms during bond breaking[1,6-8]. This dissociation process occurs in the excited C state of $Br_2$ which is prepared by single-photon absorption of 400nm laser pulse: $Br_2 + h\nu_{400nm} \rightarrow Br_2^* \rightarrow Br + Br$.

Here, we report on simultaneous measurements of photoion and photoelectron momentum spectra from strong-field ionization of the neutral dissociating $Br_2$ by using COLd Target Recoil-ion Momentum Spectroscopy (COLTRIMS). Fig. 1(a) shows the evolution of the ion-core charge distribution during the transition from molecule to atom. Fig. 1(b-d) displays the photoelectron momentum distributions from different dynamical stages. The differential analysis on these photoelectron spectra further supports the findings of photoion observations. Moreover, we observe the convergence of the photoelectron spectra during the end of dissociation, which concludes a dissociation duration of about 125 fs. The Semi-classical simulations reveal that the distinction of molecule and atom by photoelectron momentum spectra originates in the different electron rescattering on the charge distributions in molecular and atomic ion cores. Our findings open a new avenue for imaging molecular dynamics through concurrent examination of photoion and photoelectron spectra.

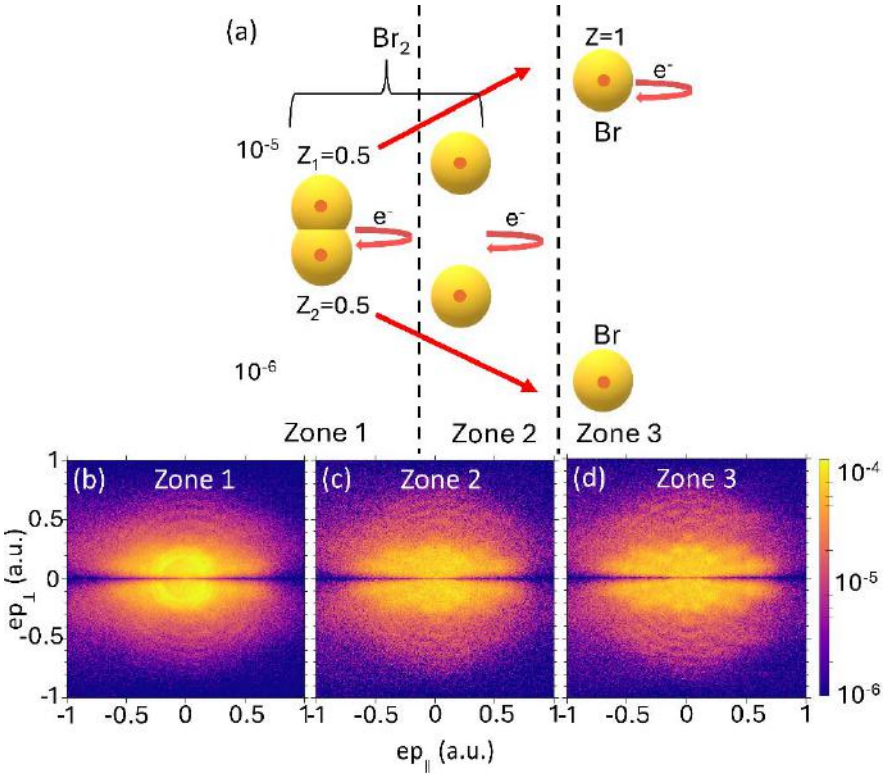


Fig. 1. (a) Illustration of the breaking up of $Br_2$ with the evolution of the ion-core charge distribution. The red curved arrows indicate the electron recollision after ionization. (b-d) photoelectron momentum distributions from the different dynamical stages of $Br_2$ dissociation.

**References**

[1] W. Li *et al,* PNAS **107**, 20219 (2010).

[2] E. Goulielmakis *et al,* Nature **466**, 739-743 (2010).

[3] S. Haessler *et al,* Nature Physics **6**, 200-206 (2010).

[4] A. Barty *et al,* Nature Photonics **2**, 415-419 (2008).

[5] J. Yang *et al,* Science **368**, 885-889 (2020).

[6] Ph. Wernet *et al,* Physical Review Letters **103**, 013001 (2009).

[7] H. J. Wörner *et al,* Nature **466**, 604-607 (2010).

[8] A Rouzée *et al,* Journal of Physics B **46**, 164029 (2013).

# Light-induced ultrafast dynamics of cold molecules in helium nanodroplets

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**Synopsis:** The superfluid helium nanodroplets can cool down the molecules to 0.37 K and serve as an ideal nanoreactors for light-induced interaction between the embedded cold molecules and helium environment. Driven by strong laser fields, we explore how strong the interaction of a charged molecular ion is with its helium surroundings by real-time tracking the ultrafast vibrational decoherence dynamics of in-droplet $D_2^+$ molecule. We also investigate the matter wave nature of a single cold in-droplet molecule, exhibiting as localization and delocalization behaviors depending on the molecule's mass.

Superfluid helium ($^4$He) nanodroplets with an extremely cold environment at 0.37 K and a broad transparent spectral range are ideal nanoreactors for light-induced physical and chemical reactions of the embedded molecules. Most information of the in-droplet molecules is obtained from the spectroscopy measurements, although the dynamics are the key for one to understand and further control the chemical reactions. The newly developed reaction microscopy of helium-nanodroplet ($He_N$) target recoil ion momentum spectroscopy ($He_N$TRIMS) allows us to explore the ultrafast dynamics of in-droplet molecules driven by femtosecond laser pulses [1,2], where the electrons and ions ejected from an in-droplet molecule are measured in coincidence.

By performing pump-probe experiments in the $He_N$TRIMS, we capture in real time the collision-induced ultrafast dissipation of vibrational nuclear wave packet dynamics of $D_2^+$ ion embedded in a $He_N$ [3]. Our results show that, differing from the behaviors of the in-droplet neutral molecules, the charged ions in the $He_N$ interior strongly couple to the He solvent via ion-He collisional interactions, leading to the extremely fast collisional dissipation dynamics within ~140fs. Our findings underscore the crucial role of ultrafast collisional dissipation in shaping the molecular decoherence dynamics during solution chemical reactions, particularly when the solute molecules are predominantly in ionic states.

Furthermore, we explore the spatial scale of the matter wave of a cold molecule with respect to the $He_N$ by diagnosing the angular nodal structures in the photoelectron momentum distributions (PMDs) [4]. The preserved and nodal structures in the PMDs allows us to identify and characterize the delocalization of the lightest $H_2$ and localization of heavier $D_2$ and $O_2$ molecules, whose de Broglie wavelength is comparable to or smaller than droplet size.

**References**

[1] J. Qiang et al, "Femtosecond Rotational Dynamics of $D_2$ Molecules in Superfluid Helium Nanodroplets", Phys. Rev. Lett. **128**, 243201 (2022).

[2] L. Zhou et al, "Enhancing Strong-Field Dissociation of $H_2^+$ in Helium Nanodroplets", Phys. Rev. Lett. **130**, 033201 (2023).

[3] J. Qiang et al, "Femtosecond Collisional Dissipation of Vibrating $D_2^+$ in Helium Nanodroplets", Phys. Rev. Lett. **132**, 103201 (2024).

[4] Z. Ye et al, "Localization and Delocalization of a Single Molecule in a Helium Nanodroplet", in preparation.

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