上海科技大学
ShanghaiTech University

華東師範大學
EAST CHINA NORMAL UNIVERSITY

上海交通大學
SHANGHAI JIAO TONG UNIVERSITY

2025 International Workshop on Ultrafast & X-ray Science

WUFXS 2025

Program

Center for Transformative Science
ShanghaiTech University

State Key Laboratory of Precision Spectroscopy
East China Normal University

Key Laboratory for Laser Plasmas
Shanghai Jiao Tong University

Program

Friday, 09 May	
14:00-17:00	Site tour
18:00-21:00	Welcome reception
21:00 ~	Bar and food for late arrivals & discussion #8-307
Saturday, 10 May	
Session 1 (8:45-10:30) - Opening/FEL Sources	Chair: **Kiyoshi Ueda**
08:45-08:50	**Zhentang Zhao**, Shanghai Advanced Research Institute, Chinese Academy of Sciences *Welcome from SARI and SHINE.*
09:00-09:30	**Zhentang Zhao**, Shanghai Advanced Research Institute, Chinese Academy of Sciences *Shanghai HIgh repetitioN rate XFEL and Extreme light facility (SHINE).*
09:30-10:00	**Guorong Wu**, Dalian Institute of Chemical Physics, Chinese Academy of Sciences *Advances in Ultrafast Laser Technologies and Opportunities for Ultrafast Time-Resolved Studies.*
10:00-10:30	**Enrico Allaria**, Deutsches Elektronen-Synchrotron, Elettra Sincrotrone Trieste *Advancements and Emerging Programs in Ultrafast X-Ray Science at FERMI.*
Coffee break (10:30-11:00)	
Session 2 (11:00-12:30) – FEL Science I	Chair: **Xincheng Wang**
11:00-11:30	**Robin Santra**, Deutsches Elektronen-Synchrotron (DESY) *Imaging collective quantum fluctuations.*
11:30-12:00	**Daniela Rupp**, ETH Zurich *Coherent Diffraction Imaging of ultrafast melting dynamics in silver nanoparticles.*
12:00-12:15	1. **Ruichao Dong**, Shanghai Advanced Research Institute (SARI), Chinese Academy of Sciences *Competition between one- and two-photon processes in Ar irradiated by the Shanghai Soft X-ray Free Electron Laser.*
12:15-12:30	2. **Xiaojing Liu**, ShanghaiTech University *Status of a composite VMI at the AMO beam line of SXFEL.*
Lunch break (12:30-14:00)	
Session 3 (14:00-16:15) – ATTO Science I	Chair: **Yuhai Jiang**
14:00-14:30	**Zhiyi Wei**, Institute of Physics, Chinese Academy of Sciences *Attosecond Lasers-Toward Advanced Light Infrastructure for Froniter Researches*
14:30-15:00	**Caterina Vozzi**, Consiglio Nazionale delle Ricerche (CNR) *Ultrafast XUV Spectroscopy for Investigation of Materials.*
15:00-16:15	1. **Jing Zhao**, National University of Defense Technology *Raman time-delay in attosecond transient absorption of strong-field created ions.*
15:15-15:30	2. **Liang Li**, Huazhong University of Science and Technology *The diffraction principle for high harmonic spectroscopy in solids.*
15:30-15:45	3. **Yanlan Wang**, Innovation Academy for Precision Measurement Science and Technology (APM), Chinese Academy of Sciences *Probing ultrafast electron dynamics with polarization-gated attoclock.*
15:45-16:00	4. **Yijia Mao**, Shanghai Jiao Tong University *Coherent control of ion-electron entanglement in multiphoton ionization.*
16:00-16:15	5. **Hongcheng Ni**, East China Normal University *Over-Barrier Ionization Dynamics Studied by Backpropagation*
Conference Dinner (17:00-21:30)	

Sunday, 11 May	
Session 4 (08:45-10:45) – ATTO Science II	Chair: **Ahai Chen**
08:45-09:15	**Francesca Calegari**, Deutsches Elektronen-Synchrotron (DESY) *Controlling chemistry at extreme time scales.*
09:15-09:45	**Jing Chen**, University of Science and Technology of China *Correlated tunneling in high-order above threshold dissociative ionization of* H_2.
09:45-10:15	**Chengyin Wu**, Peking University *Investigating electron dynamics in condensed matter through time-resolved photoemission electron microscopy and high-order harmonic generation.*
10.15 -10:30 10.30 -10:45	1. **Song Li**, Shanghai Institute of Optics and Fine Mechanics (SIOM) *Bright Betatron X-ray Source Developed at SIOM Using Laser Wakefield Acceleration* 2. **Juan Guo**: on the journal “**Ultrafast Science**”.
Coffee Break (10:45-11:15)	
Session 5 (11:15-12:30) – Qutanum Light	Chair: **Feng He**
11:15-11:45	**Misha Ivanov**, Max-Born-Institut (MBI) *Attosecond approaches to the generation of quantum states of light from IR to XUV range.*
11:45-12:15	**Paris Tzallas**, Foundation for Research and Technology-Hellas, Institute of Electronic Structure & Laser *Generation of optical Schrödinger “cat” states using intense laser-matter interactions and applications in non-linear optics.*
12:15-12:30	1. **Sili Yi**, Max Born Institute *Non-classical bright states of light generated via light-matter entanglement in strong laser fields.*
Lunch Break (12:15-14:00)	
Session 6 (14:00-16:30) – FEL Science II	Chair: **Xiaojing Liu**
14:00-14:30	**Akiyoshi Hishikawa**, Nagoya University *Ultrafast electron-ion coincidence spectroscopy at SACLA BL1.*
14:30-15:00	**Daniel Rolles**, Kansas State University *Visualizing the three-dimensional arrangement of hydrogen atoms in organic molecules by Coulomb explosion imaging.*
15:00-15:30	**Nina Rohringer**, Deutsches Elektronen-Synchrotron (DESY) *Nonlinear x-ray scattering.*
15:30-15:45 15:45-16:00 16:00-16:15 16:15-16:30	1. **Victor Kimberg**, KTH Royal Institute of Technology *Observation of an associative state ultrafast dynamics in aqueous hydroxide.* 2. **Zheng Li**, Peking University *Quantumness of* γ*-ray and hard x-ray photon emission from 3D free-electron lattices.* 3. **Yanrong Liu**, Shaanxi Normal University *Quantum control of photodissociation dynamics via an embedding UV pulse.* 4. **Marvin Schmoll**, University of Freiburg *Temporal structure of non-consecutive harmonic combs at the seeded FEL FERMI.*
Session 7 Poster Session (16:30 - 22:00)	
16:30-18:00	Poster Session 1 & Coffee Break
18:00-20:00	Dinner
20:00-22:00	Poster Session 2 & Beer

Monday, 12 May	
Session 8 (8:45-10:15) -- FEL Science III	Chair: **Jianfei Tao**
08:45-09:15	**Christian Ott**, Max-Planck-Institut für Kernphysik *Excited-state-resolved coherent dynamics in intense SASE FEL pulses.*
09:15-09:45	**Marcus Dahlström**, Lund University *Time-Dependent Strong-Coupling Phenomena at XUV Wavelengths*
09:45-10:00	1. **Sergey P. Polyutov**, Siberian Federal University Complementarity in which-path resonant Auger scattering.
10:00-10:15	2. **Weijie Hua**, Nanjing University of Science and Technology *Simulating ultrafast nonlinear X-ray spectra with MCNOX.*
Coffee Break (10:15-10:45)	
Session 9 (10:45-12:30) - ATTO Science III	Chair: **Hongcheng Ni**
10.45-11:15	**Alexander Kuleff**, University of Heidelberg *Ultrafast non-adiabatic charge dynamics in (core) excited molecules.*
11:15-11:30	1. **Weifeng Yang**, Hainan University *Tracing spatiotemporal information in ultrafast dynamics.*
11:30-11:45	2. **Sizuo Luo**, Jilin University *Attosecond Spectroscopy Reveals Spontaneous Symmetry Breaking in* CO_2 *Photoionization.*
11:45-12:00	3. **Feng Li**, Nanjing University of Posts and Telecommunications *Interband-polarization dominated high harmonic generation from flat-band* $NbOCl_2$ *materials in strong laser fields.*
12:00-12:15	4. **Linxuan Zhang**, Institute of Applied Physics and Computational Mathematics *Few-photon single ionization of laser-cooled Rubidium atoms.*
12:15-12:30	5. **Xiaodan Mao**, East China Normal University *Photon Momentum Transfer and Partitioning: From One to Many.*
Lunch Break (12:30-14:00)	
Session 10 (14:00-16:00) -- FEMTO Science I	Chair: **Jian Wu**
14:00-14:30	**Henrik Stapelfeldt**, Aarhus University *Real time observation of the diffusion-limited formation of an ion-molecule complex.*
14:30-15:00	**Olivier Faucher**, Université Bourgogne Europe, CNRS *Impulsive molecular alignment under resonant excitation.*
15:00-15:15	1. **Chuncheng Wang**, Jilin University *Coherent vibronic pseudorotation near conical intersection*
Coffee Break (15:15-15:30)	
Session 11 (15:30-17:00) -- FEMTO Science II	Chair: **Kiyoshi Ueda**
15:30-15:45	1. **Xueguang Ren**, Xi'an Jiaotong University *Recent studies of ultrafast intermolecular coulombic decay and proton transfer in bio-relevant molecular complexes.*
15:45-16:00	2. **Pengju Zhang**, Songshan Lake Materials Laboratory *ICD in liquid water competes with proton transfer and non-adiabatic relaxation.*
16:00-16:15	3. **Xiaoqing Hu**, Institute of Applied Physics and Computational Mathematics *Impact of dimer environments on conical intersection dynamics via interatomic coulombic decay (ICD) reactions.*
16:15-16:30	4. **Anna D. Skitnevskaya**, A. E. Favorsky Irkutsk Institute of Chemistry, Siberian Branch of the Russian Academy of Sciences *Possibility of predominant ICD in aromatic dimers.*
16:30-16:45	5. **Kefei Wu**, Shanghaitech University *Laser coupling induced fine structure in vibrational structures of* O_2^+ *dissociation revealed by high resolution measurement.*
Concluding Remark (16:45-16:55): **Kiyoshi Ueda,** Tohoku University, ShanghaiTech University, East China Normal University	
Invitation to WUFXS2026 (16:55-17:05): **Jian Wu**, East China Normal University	

Joint Seminars: https://reg.shanghaitech.edu.cn/cts/wufx2025/announcement.php?aid=26	
Wednesday 07 May: Shanghai Jiaotong University Joint Seminar on Ultrafast Science Program: https://reg.shanghaitech.edu.cn/cts/wufx2025/show_detail.php?aid=269&type=pdf&sp=F01 Website: http://www.lps.ecnu.edu.cn/jsufs2025/	
Thursday 08 May: ShanghaiTech University Joint Seminar on Intense Field Science	
09:00-10:00	**Marcus Dahlström**, Lund University *Theory for Ultrafast Dynamical Photoemission: cases of dressed continuum, dressed atom and dressed ion*
10:00-11:00	**Alexander Kuleff**, Heidelberg University *Interatomic Coulombic Decay: The power of non-local relaxation mechanisms*
11:00-12:00	**Nina Rohringer,** Deutsches Elektronen-Synchrotron (DESY) *X-ray Superfluorescence.*
Friday 09 May: ShanghaiTech University Joint Seminar on AMO@XFEL	
09:00-10:30	**Daniel Rolles**, Kansas State University *Gas-phase photochemistry studied by time-resolved photoelectron spectroscopy with free-electron lasers.*
10:30-12:00	**Enrico Allaria**, *Elettra Sincrotrone Trieste* *User-tailored experiments at free-electron laser facilities: the experience at FERMI*
Friday 09 May, East China Normal University Joint Seminar on Ultrafast Science Program: https://reg.shanghaitech.edu.cn/cts/wufx2025/show_detail.php?aid=270&type=pdf&sp=F01 Website: http://www.lps.ecnu.edu.cn/jsufs2025/	

2025 International Workshop on

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Abstracts

Center for Transformative Science, ShanghaiTech University
State Key Laboratory of Precision Spectroscopy, East China Normal University
Key Laboratory for Laser Plasmas, Shanghai Jiao Tong University

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Advances in Ultrafast Laser Technologies and Opportunities for Ultrafast Time-Resolved Studies

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Ultrafast lasers with high average power, high pulse energy, and short pulse duration are widely used in scientific research, including ultrafast pump-probe spectroscopy, high-harmonic generation (HHG), deep-ultraviolet (DUV) and extreme-ultraviolet (XUV) light sources, and attosecond science[1]. Among these, ytterbium-doped lasers show the most promising development potential. Ytterbium-doped gain media (e.g., fibers, slabs, and thin disks) enable direct diode pumping and high optical-to-optical efficiency, significantly expanding the repetition rate and average power of ultrafast laser systems while maintaining both high average power and high single-pulse energy[2]. However, due to the limited gain bandwidth of ytterbium ions, the shortest achievable pulse duration is typically 200–300 fs, which restricts both peak power and temporal resolution in time-resolved experiments, thereby limiting their application scope.

To achieve shorter pulses, one of the main approaches is optical parametric chirped-pulse amplification (OPCPA) pumped by Yb lasers, achieving pulse durations to the 10-fs regime and broadening their applications[3–5]. Currently, commercial ytterbium lasers reliably deliver hundreds of watts, but OPCPA systems exhibit an overall efficiency of only ~10% from pump to final output, resulting in average powers of tens of watts. This low efficiency limits OPCPA's practical deployment.

Recent advancements in post-compression techniques leveraging nonlinear effects (e.g., self-phase modulation, supercontinuum generation, and soliton dynamics) have made significant progress. Notably, multipass cell (MPC) compression—using thin plates or noble gases—achieves exceptional system efficiencies (~90%). To date, MPC technology has demonstrated: sub-50-fs pulses at kW-level average power, pulse energies exceeding 200 mJ, and pulse durations below 10 fs[6–8].

Another development in ultrafast laser technology is the DUV and VUV laser pulse generation through resonant dispersive wave, with merits of high efficiency (~1%), broad wavelength tunability in a compact table-top device[9,10].

In this report, we review these technological breakthroughs and present ongoing developments at the Dalian Institute of Chemical Physics (CAS) and Shenzhen Institute of Advanced Light Source Facilities.

Keywords: ultrafast optics, high repetition rate, nonlinear optics

Ref.

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[2] P. Russbueldt, T. Mans, J. Weitenberg, H. D. Hoffmann, R. Poprawe. *Optics Letters*, **2010**, *35(24):* 4169-4171.
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[9] J. C. Travers. *Optics Communications*, **2024**, *555:*130191.
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Advancements and Emerging Programs in Ultrafast X-Ray Science at FERMI

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FERMI, operated by Elettra – Sincrotrone Trieste, is the first free-electron laser (FEL) facility that produces fully coherent pulses in the extreme ultraviolet (EUV) to soft X-ray range, utilizing two FEL lines with external seeding. This high degree of coherence and accurate control over the radiation properties have supported groundbreaking research in coherent control and attosecond science within the EUV-soft X-ray spectral range.

Currently, access to the attosecond timescale is achieved at FERMI through the control of the well defined radiation phase, instead of relying solely on pulse amplitude duration, which remains within tens of femtoseconds. Various techniques, including the use of phase-locked multicolor pulses and control of FEL pulse chirp, have allowed investigation of the response of matter at the few as time scale. This presentation provides a comprehensive review of recent findings and ongoing developments at FERMI, aimed at further enhancing these capabilities.

Coherent Diffraction Imaging of ultrafast melting dynamics in silver nanoparticles

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Via single-shot coherent diffractive imaging (CDI), the structure and dynamics of isolated nanosamples can be directly visualized: Intense and short pulses of X-ray free-electron lasers (XFELs) or intense highharmonic generation (HHG) based sources scatter off a free-flying nanostructure, forming an interference pattern that is captured with a large-area detector. With computer-based iterative phase-retrieval or forward-fitting methods a snapshot of the object's structure can be retrieved from the pattern [1]. This has opened a door for us to study intense laser-matter interaction with unprecedented detail. Silver nanoparticles were brought into the gas phase and heated with laser pulses tuned to their surface plasmon resonance. This approach allows for a uniform heating of the nanoparticles at comparably low laser intensities, avoiding strong-field effects like tunneling and electron impact ionization. We observe, depending on the heating laser's intensity, a wide range of processes from surface to full melting, internal boiling, cavitation, expansion and inflation, droplet vibrations, up to explosive boiling. Molecular dynamics simulations match the observed states and show that the systems travel on rather similar trajectories through the phase diagram, differing only in whether and where the stability limit of the metastable superheated liquid is crossed [2]. These results exemplify the maturity of time-resolved single-particle coherent diffraction imaging for investigating ultrafast dynamics in matter, being now on a level where we can really extract previously inaccessible physical quantities of matter under extreme conditions. [1] Colombo et al., SPRING, an effective and reliable framework for image reconstruction in single-particle Coherent Diffraction Imaging, under review at npj Computational Materials (2025), arxiv 2409.07413 [2] Dold, Reichenbach, Colombo et al., Melting, bubble-like expansion and explosion of superheated plasmonic nanoparticles, Physical Review Letters 134, 136101 (2025)

Competition between one- and two-photon processes in Ar irradiated by the Shanghai Soft X-ray Free Electron Laser

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Shanghai Soft X-ray Free Electron Laser (XFEL) facility [1], the first XFEL in China, has entered user operation, delivering soft X-ray pulses in self-amplified spontaneous emission mode with an intensity of the peak fluence of ~1.0×10−2μJ/μm2 at a focus point. With these conditions, at photon energy of 260 eV, we expect that sequential processes in which Ar2+ created by the Auger decay following the 2p photoionization may be resonantly excited to the 2p−13p−23d inner- shell excited state and thus resonant Auger decay produces Ar3+, which can also be competitively produced by the direct double Auger decay (DDAD) following the one photon 2p ionization of Ar. Ar4+ ions dominantly arise from Ar2+ with a resonant excitation and subsequent DDAD, involving two-photon absorption. The measured charge distributions as a function of the XFEL fluence are well reproduced by ab initio calculations [2] that include DDAD, normally neglected in the previous XFEL studies, and resonant Auger decay. The present results demonstrate that the low peak fluence of the XFEL pulses can be determined if the competition between the one- and two-photon processes is precisely described along with DDAD and resonant Auger decay.

Keywords: XFEL, Resonant excitation, Direct double Auger decay, Peak fluence

Ref:

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Attosecond Lasers-Toward Advanced Light Infrastructurefor Froniter Researches

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The emergence and evoulation of attosecond light pulses not only broken the shortest time limits, but also provide an unpreceedent tool for explore electron dynamics, it exhibits wide applicationsin materials science, information technology etc. Since we realized the first attosecondpulseswith 160 as duration in China in 2013 [1], continuous progresses have been made by improvingthe experiment systems and driver lasers, the pulse duration was shortened from86as [2] to72as. Rencet, by using sub-10fs driven laser of central at 900nm generated by optical parametricchirped-pulse amplifier, we further obtained attosecond pulse as ahort as 59as. Missiontotheemerging researchs and applications, a national facility-Synergetic Extreme ConditionUserFacility (SECUF), has been constructed with comprehensive extreme-condition experimental platform integrating ultralow temperatures, strong magnetic fields, ultrahigh pressures, andultrafast optical fields, which was opend for international users. Furthermore, the AdvancedAttosecond Laser Infrastructure (AALI) was launched recent, which aims to multifuncational attosecond and XUV beamlines with application research terminal stations. Driver lasers includeTi:Sapphire, fiber and thin-disk laser as well as OPCPA were developed with high averagepower, sub-10 fs pulse and tunable wavelengths. In this talk we will report the relavent progresstoward a robust and cutting-edge platform for froniter researches. Ref: [1] J. Zhan, P. Ye, H. Teng et al., Chin Phys Lett, 30: 093201(2013). [2] S. Zhong, H. Teng, X. Zhu, et al., Chin. Opt. Lett. 21, 113201 (2023).

Keywords: Attosecond pulses; Ultrafast optics; High-harmonic generation; Femtosecond;

Ref:

[1] J. Zhan, P. Ye, H. Teng et al., Chin Phys Lett, 30: 093201(2013).
[2] S. Zhong, H. Teng, X. Zhu, et al., Chin. Opt. Lett. 21, 113201 (2023).

Raman time-delay in attosecond transient absorption of strong-field created ions

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Strong-field ionization injects a transient vacancy in the atom which is entangled with the outgoing photoelectron. When the electron is ultimately detached, the ion is left in different excited states with partial coherence loss [1]. The preserved coherence of the system after interaction with intense short pulses significantly influences subsequent nonequilibrium dynamics and energy relaxation. Using attosecond transient absorption spectroscopy, we observe that the absorptions of the two spin-orbit split states of the krypton vacancy during their creation, are modulated at difference paces with a time delay of several hundred attoseconds. By developing a comprehensive theoretical model incorporating all relevant dynamics, we reveal that coherent Raman coupling of the ions with the remaining field leads to the time delay between the two resonant absorptions [2]. This discovery provides insight into laser-dressed bound-bound transitions and transient ionization dynamics, enriching the attosecond chronoscopy.

Keywords: Attosecond transient absorption spectroscopy; Raman time-delay

Ref:

[1] E. Goulielmakis, et al., “Real-time observation of valence electron motion.” Nature 466, 739 (2010)

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The diffraction principle for high harmonic spectroscopy in solids

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Recently, HHG has also been observed in solids, which makes it possible to extend the successful attosecond science to condensed-matter phase. To achieve this goal, an accurate understanding of the physical mechanisms underlying the solid HHG is essential. Compared to gas-phase atoms and molecules, solid media have a more complex electronic structure, and the interaction between the system and the external laser field is more complicated. Although the three-step model can qualitatively explain the plateau structure and cut-off law of the interband harmonics, there are still many phenomena that cannot be explained by the recollision trajectories. Indeed, some works have modified the saddle-point equations or recollision conditions by taking account of the width or separation of electron and hole wave packets. However, the harmonic emission is still described by the trajectory-based recollisions formulated by the saddle-point equations, and the coherence and interference of the electron wave packet during propagation is excluded.

Here, we focus on the semiclassical perspectives of solid HHG which aims to decode the phase information of coherent electron dynamics and lies at the heart of HHS. We present a theoretical framework with a four step model [1], and propose Huygens-Fresnel picture for describing HHG in solids [2]. The important role of the wave-like behavior of electron motion in HHG from solids has been demonstrated [3,4] and some novel strategies for applications of high harmonic spectroscopy from wave perspective will also be introduced.

Keywords: high harmonic generation, strong-field ultrafast optics, ultrafast electron dynamics;

Ref:

[1] L Li, et al., Phys. Rev. Lett. 122, 193901 (2019)

[2] L Li, et al., Phys. Rev. Lett. 127, 223201 (2021)

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[4] L Li, et al., Phys. Rev. Lett. 133, 116902 (2024)

Probing ultrafast electron dynamics with polarization-gated attoclock

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Tracing and controlling electron motion on its characteristic timescale is crucial for understanding the fundamental features of ultrafast photophysics and photochemical processes. Here, we discuss our progress in detecting the correlated electron emission times of atoms with a polarization-gated (PG) attoclock scheme. This PG pulse consists of two counter-rotating circularly polarized, time-delayed, and CEP stabilized few-cycle laser pulses [1]. The combined pulse has a time-dependent ellipticity in overlapping temporal region, in which the central portion exhibits approximately linear polarization while the remaining regions exhibit elliptical polarization. As a result, the PG pulse can effectively drive two electrons emission in a correlated way within the gate, while also possessing the property of angular streaking. First, by precisely controlling the interference of electron wave packets released at different times within one optical cycle with the PG pulse, we demonstrate the capability to extract ultrafast electron dynamics information on the subcycle timescale [2,3]. Then, we apply the PG attoclock to measure ionization time difference of two correlated electrons in the non-sequential double ionization of Ar atom. Our results indicate that the electron pairs are mainly emitted through two channels, with the most likely emission time differences of about 234±22 as and 1043±73 as, respectively [4]. Classical trajectory Monte Carlo model calculations well reproduce the experimental measurements and allow us to comprehend the effect of the final-state electron-electron repulsion in correlated-electron dynamics deeply. Our proposed PG attoclock scheme provides a powerful tool for studying electron correlation dynamics in strong-field atomic double ionization, and opens up possibilities for exploring ultrafast electron dynamics in complex systems.

Ref:

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Coherent control of ion-electron entanglement in multiphoton ionization

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Recent experimental and theoretical studies are reavling the importance of entanglement in ultrafast physics [1-3]. We present a novel strategy for dynamically manipulating quantum entanglement in photoionization processes by employing resonance-enhanced (2+1') multiphoton ionization of argon atoms with two time-delayed ultraviolet pulses. We achieve precise tuning of photoelectron-ion entanglement via quantum beat modulation.

Through systematic analysis of the inter-pulse delay-dependent anisotropy parameters of the photoelectron angular distributions, we demonstrate that the photoelectron-ion entanglement—quantified via reconstructed purity—can be controlled by tailoring the pulse delay. Our multi-configurational time-dependent Hartree-Fock (MCTDHF) and R-matrix time-dependent (RMT) calculations uncover a characteristic quantum interferometric phase shift between the $3s^2 3p^5 \varepsilon d$ ($^1F^o$) and $3s^2 3p^5 \varepsilon d$ ($^1P^o$) scattering channels, serving as a direct signature of entanglement. This protocal of entanglement manipulation can be further extended to atoms or even molecules.

Keywords: Entanglement manipulation, photoelectron angular distribution, anisotropy parameters, multielectron calculations;

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Over-Barrier Ionization Dynamics Studied by Backpropagation

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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. 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].

Keywords: over-barrier ionization, backpropagation, transverse tunneling momentum;

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Correlated tunneling in high-order above threshold dissociative ionization of H_2

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Comprehension of photon-triggered molecular processes is essential in the study of various important topics in physics, chemistry, and biology. Here we propose a correlated tunneling picture to understand the dissociative ionization process of molecules in intense laser fields based on a quantum model developed in the framework of many-body S-matrix theory including nuclear vibrational motion. In this quantum correlation picture, the single ionization of H_2 and the subsequent electron-ion recollision induced dissociation are considered as an entangled correlated process. It enables us to attribute the interference pattern in the joint-energy spectra to combined effects of single-slit diffraction and multi-slit interference of correlated electron-nuclear wave packets in the time domain. Our work opens a new avenue to understanding molecular dissociative ionization processes in external fields.

Keywords: molecular dissociative ionization, electron-nuclear correlation, intense laser field

Ref:

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Investigating electron dynamics in condensed matter through time-resolved photoemission electron microscopy and high-order harmonic generation

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The interaction between ultrafast laser pulses and condensed matter has been the subject of extensive investigation over the past decade [1]. This research encompasses ultrafast science, strong-field physics, condensed matter physics, and nano-optics, offering significant potential for groundbreaking scientific discoveries. Both time-resolved photoemission electron microscopy and solid-state high-order harmonic spectroscopy are powerful tools for probing ultrafast electron dynamics in condensed matter induced by femtosecond laser pulses. In this report, we present our recent advancements in probing and manipulating electron dynamics by controlling the interaction between micro-nano structures and synthetic laser fields with attosecond precision. We have successfully built an ultrafast extreme ultraviolet photoemission electron microscope [2]. Utilizing this advanced microscope with ultra-high temporal, spatial, and energy resolution, we have demonstrated the characterization and manipulation of electron pulses emitted from nanostructures on an ultrafast timescale [3,4]. We also built a compact solid-state high-order harmonic spectrometer with high sensitivity, which provides an all-optical ultrafast probe to monitor electron dynamics in condensed matter. By using this time-resolved high-order harmonic spectroscopy with the help of theoretical simulations, we have explored the determination and manipulation of electron dynamics within semiconductor materials [5,6], three-dimensional Dirac semimetals [7], and strongly correlated systems [8]. Our work demonstrates that time-resolved photoemission electron microscopy and solid-state high-order harmonic spectroscopy are effective for probing ultrafast electron dynamics in condensed matter induced by strong femtosecond laser pulses.

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Bright Betatron X-ray Source Developed at SIOM Using Laser Wakefield Acceleration

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With the progressive miniaturization and stabilization of laser wakefield accelerators, table-top X-ray radiation sources based on wakefield accelerators have shown enormous potential. The betatron radiation source, driven by ultrashort laser pulses, features a source size in the micrometer range, pulse duration in the femtosecond range, divergence angles in the milliradian range, and covers a broad spectrum exceeding tens of keV. It is applicable for high-contrast imaging of minute structures and for probing interdisciplinary ultrafast processes. Here we present the achievement of bright betatron hard X-ray radiation sources at Shanghai Institute of Optics and Fine Mechanics (SIOM), Chinese Academy of Sciences, utilizing a 1PW/0.1Hz laser system at Shanghai Superintense Ultrafast Laser Facility (SULF). Such a laser system can produce electron beams with charges above hundred pC and energies exceeding 1 GeV. Meanwhile, electron beams undergo the betatron oscillations in the wakefield, generating high-brightness X-ray radiation sources with critical energies ranging from 15 to 25 keV.

This advancement enables time-resolved imaging and spectroscopy at atomic and molecular scales, suitable for dense materials and biological specimens. The high brightness and energy levels enhance the efficiency and resolution, making these sources competitive with larger synchrotron facilities. The compact nature of table-top betatron X-ray sources offers significant cost and complexity reductions, fostering broader access and innovation across fields such as nuclear photonics, ultrafast chemistry, materials science, and biomedical research, etc. As this technology matures, its transformative impact on scientific and industrial applications is expected to grow.

Generation of optical Schrödinger "cat" states using intense laser-matter interactions and applications in non-linear optics

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Intense laser-matter interaction leads to high harmonic generation (HHG), where the low frequency photons of a driving laser field are converted into photons of higher frequencies. This process has enabled breakthroughs in AMO physics and attosecond science [1]. Until recently, it was described by classical or semi-classical approaches [2], ignoring the quantum nature of light.

Recent investigations conducted using fully quantized approaches have shown how the HHG process can be used for the generation of optical Schrödinger "cat" and entangled light states from the infrared to the extreme-ultraviolet spectral range [3-10].

Here, after a brief introduction, I will focus my talk on the most recent studies concerning the generation of optical "cat" states and their role in nonlinear optics [11]. I will also emphasize the potential of using fully quantum approaches in laser-driven semiconductor crystals to develop non-classical and entangled light states in the VUV spectral region with attosecond pulse duration [12]. Finally, I will briefly discuss some additional representative key approaches developed in the recent past by other groups in this research field [13-19].

Keywords: High harmonic generation, quantum optics, attosecond science;

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Non-classical bright states of light generated via light-matter entanglement in strong laser fields

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Recent observation of Schrödinger cat states in strong-field regime has built a bridge between quantum optics and intense laser physics and opened up an exciting possibility of using intense light-matter interactions to generate bright quantum states of light.

First, we demonstrate a novel route to generating harmonics in non-classical quantum states carrying massive number 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.

We then consider the solid-state systems driven by two-color (Pump-Control) fields. We demonstrate that by tuning the pump laser to control non-adiabatic transition between valence band and conduction band, one can control quantum properties of the generated light state, pathing the way for generation of quantum light state in the solid-state device by purely classical control. In addition, we explore interference between quantum light states generated at different moments controlled by the number and timing of pump lasers.

Furthermore, we discuss the cases for molecule system driven by one-color and two-color fields, highlighting the role of nuclear motion during the intense light-matter interactions. We analyze how the coupling of electronic and nuclear dynamics influences the quantum light state.

Keywords: intense laser physic, quantum optics;

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Ultrafast electron-ion coincidence spectroscopy at SACLA BL1

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The response of matter to extreme ultraviolet (XUV) and X-ray laser pulses has attracted considerable attention for many years because of its importance in various free-electron laser (FEL) applications, such as single-molecule diffraction and ultrafast spectroscopy. The absorption of such high-energy photons proceeds along different pathways consisting of different sequences of photoabsorption and core-hole decay. Understanding nonlinear processes in the XUV and X-ray laser fields has been challenging due to their complicated nature. Photoelectron spectroscopy is powerful in resolving the contributions from different ionization pathways, since the intermediate states populated in the multiple ionization can be determined from the kinetic energies of the ejected electrons. However, in some cases, careful analysis is required to distinguish certain responses of interest from other signals. To circumvent this difficulty, we used counterpart ion coincidence spectroscopy, which allows direct characterization of electrons emitted in a particular ionization pathway. We studied the multiphoton ionization of Xe [1] and Kr [2] in strong XUV-FEL fields at SACLA BL1 using a magnetic bottle-type spectrometer equipped with ion-extracting electrodes [3]. Combined with a pump-probe scheme, electron-ion coincidence spectroscopy has also been used to elucidate the details of the photoreaction dynamics of molecules [4], which will advance our understanding of ultrafast imaging by ultrashort intense XUV and X-ray pulses. These previous and ongoing studies will be presented in the talk.

Keywords: FEL, electron-ion coincidence spectroscopy, magnetic-bottle spectrometer;

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Visualizing the three-dimensional arrangement of hydrogen atoms in organic molecules by Coulomb explosion imaging

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Structure-sensitive methods based on femtosecond light or electron pulses are now making it possible to measure how molecular structures change during light-induced processes. Despite significant progress, high-fidelity imaging of nuclear positions remains a challenge even for relatively small molecular systems and, notably, regarding the positions of hydrogen atoms. X-ray induced Coulomb explosion imaging (CEI) may overcome this obstacle as its sensitivity does not depend on the mass of the imaged atoms. I will show several recent results demonstrating that CEI offers a powerful route to imaging nuclear motions (e.g. isomerizations) in organic molecules on ultrafast timescales, including differentiating photoproduct isomers whose structures primarily differ in the position of the hydrogens. I will also show how machine learning tools can use correlations in the high-dimensional CEI data to identify and distinguish contributions from different photoproducts with high fidelity.

Nonlinear x-ray scattering

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X-ray crystallography is essential for determining protein structures, but challenges persist in achieving sub-Ångström resolution, such as growing high-quality crystals, radiation damage, and the phase problem. Serial femtosecond crystallography at XFELs addresses the first two issues, but obtaining model-free atomic structures remains difficult. Single-wavelength anomalous dispersion helps to enhance phase sensitivity, but has limitations due to the addition of heavy atoms to the protein. A solution might involve enhancing the scattering properties and phase sensitivity of lighter atoms by elastic resonant scattering of hidden, transient resonances. In a first proof-of-principle experiment we demonstrate enhanced resonant scattering at the Cu L-edge of a superlattice of a Cu-containing multilayer mirror. The XFEL creates warm-dense copper and opens transient resonances by efficient creation of 3d holes through electron collisions. The polarization response includes a non-resonant term defined by the atomic scattering factor of Cu and a resonant elastic scattering contribution from 2p-3d transitions. In addition to the enhancement and phase sensitivity, the spectrally resolved scattering signal encodes electronic structure information of the warm-dense copper, in agreement to a recent transient absorption experiment of XFEL created warm-dense copper (L. Mercadier et al., Nature Physics 2024, https://doi.org/10.1038/s41567-024-02587-w). Ideally, enhanced resonant x-ray scattering involves a two-color attosecond x-ray pump-probe scheme: The pump creates population inversion in atoms via ultrafast 1s ionization, while the probe pulse targets the transient 1s-2p (Kα) resonance to scatter elastically with a significant dipole-matrix element. In high-intensity probes, the resonant part may enter the nonlinear regime, with a significant contribution to the effective scattering factor for Cu of $f'' \sim -i \cdot 2500 \cdot \rho\_{\mathrm{inv}}$. I will present our theoretical estimates for fs pump-probe enhanced resonant x-ray scattering in copper, along with recent experimental results in $CuSO_4$ single crystals. Experimental challenges, in particular the stochastic fluctuations of the scattering strength due to the limited transverse coherence of XFELs, will be discussed, that also have implications for high-resolution x-ray scattering for valence-electronic structure determination.

Observation of an associative state ultrafast dynamics in aqueous hydroxide

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The dynamics of chemical reactions in solution is of paramount importance in fields ranging from biology to materials science. Because the hydrogen-bond network and proton dynamics govern the behavior of water solutions, they have been the subject of numerous studies over the years. Here, we report the observation for a previously unknown associative state in the hydroxide ion guiding an ultrafast nuclear dynamics when a proton from a neighboring water molecule approaches the hydroxide ion, utilizing resonant inelastic soft X-ray scattering (RIXS) and quantum dynamical simulations. Analysis via state-of-the-art theory reveals a state mixing in the electronically excited states between aqueous hydroxide ions and the solvent. Our results give new insights into chemical bonding and ultrafast excited-state dynamics in the aqueous environment. This investigation of associative states opens up new pathways for spectroscopic studies of chemical reaction dynamics and lays the foundation of directly accessing the dynamic proton exchange in solution.

Keywords: ultrafast nuclear dynaics, RIXS, hydrogen-bond network;

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Quantumness of γ-ray and hard x-ray photon emission from 3D free-electron lattices

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Crystalline undulator radiation (CUR) is emitted by charged particles channeling through a periodically bent crystal. We show that entangled high energy photons of the order of 100 MeV can be generated from CUR, and obtain the quantum entanglement properties of the double-photon emission of CUR with a non-perturbative quantum field theory. We demonstrate that the crystalline undulator (CU) can induce a 3D free-electron lattice with pre-microbunched electrons, and the resulting free-electron lattice can enhance the entangled high energy photon emission for certain angles by phase matching. We also examine the effects of demodulation and dechanneling during the electron beam channeling process, and show the dependence of the dechanneling and demodulation lengths on the undulator parameters. We have also proposed a undulator tapering scheme to reduce the single-emission background at the energies of the entangled photons. In summary, we introduce an important quantum light source, the crystalline undulator, to the X-ray and gamma ray quantum optics. This novel quantum light source has important application potential in the field of high energy quantum optics

Keywords: Quantum optics; Free-electron laser; Quantum electrodynamics;

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Quantum control of photodissociation dynamics via an embedding UV pulse

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Ultraviolet (UV) photodissociation provides valuable insights into fragmentation patterns and photochemical reactions. However, the limited overlap between vibrational bound states and continuum states hinders effcient quantum excitation. We address this challenge by embedding the ground bound potential into the dissociative continuum using a frequency-selected UV pulse. This pulse creates vibrational resonances by coupling the dissociative continuum with unpopulated vibrationally excited levels of the ground state, without initiating photoexcitation itself. Our findings demonstrate that the photodissociation spectra can be significantly controlled by tuning the embedding pulse frequency to tailor the asymmetric profiles of the vibrational resonances. This is illustrated in our simulations of kinetic energy release spectra for both diatomic and polyatomic molecules. These proof-of-principle examples offer new opportunities for controlling the yield of photofragmentation and the pathways of photochemical reactions in various molecular systems [1].

Keywords: Photodissociation; Quantum control; Embedding UV pulse

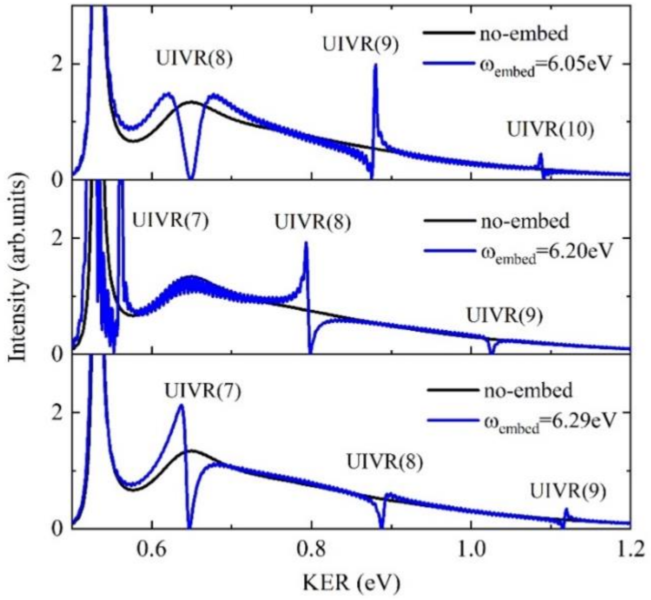

FIG. Kinetic energy release spectra for the embedding intensity $I_{embed}=1\times10^{12}$ W/cm^2.

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Temporal structure of non-consecutive harmonic combs at the seeded FEL FERMI

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Characterizing the temporal structure of pulse trains made from harmonic frequency combs requires knowledge about the relative phase of the individual harmonics. This measurement is commonly done creating photoelectrons by ionization with the harmonic comb and then letting them interact with a synchronized infrared (IR) probe field. Quantum path interference creates oscillations in the energy dependent yield, which can be used to reconstruct the phases [1]. For seeded free-electron lasers this method could also be demonstrated recently [2] but is significantly more challenging due to shot-to-shot delay jitter between the extreme ultraviolet (XUV) comb and IR probe pulse, requiring a single-shot attosecond timing tool to first extract the XUV-IR phase-delay. We present a further extension of this RABBITT-like measurement for a harmonic comb consisting of non-contiguous harmonics. Using transitions characterized by the exchange of several IR photons, we can extract the phases and reconstruct the temporal structure with attosecond resolution.

Keywords: attosecond,two-color photoionization, RABBITT, harmonic comb

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Excited-state-resolved coherent dynamics in intense SASE FEL pulses

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The nonlinear interaction of intense XUV FEL pulses with atoms and molecules enables the excited-state-specific control of localized (bound) electron dynamics. Since the strongly driven bound–bound transitions are typically embedded in the ionization continuum, new effects arise such as the FEL-control of autoionizing Fano line shapes [1] and AC Stark shifts of FEL-dressed ion multiplets [2]. Hereby the specific nature of the short-lived excited state is decisive, e.g., for the control of the emission direction of the eventually ionized electron leaving the system [3]. In this talk I will review these recent activities and especially focus on new results for the coherent control of the electron emission direction in helium with two-color SASE FEL pulses. Benchmarking the state-resolved FEL-driven dynamics in relatively small and isolated atomic systems is a key ingredient for the state-specific all-XUV measurement and control of ultrafast molecular dynamics [4–7].

Keywords: coherent XUV dynamics, state resolved spectroscopy, SASE FEL pulses

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Time-Dependent Strong-Coupling Phenomena at XUV Wavelengths

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Recent advances in free-electron laser (FEL) technology have intensified interest in coherent 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, c.f. Refs. [1,2]. The high intensity attainable by FEL pulses open up for previously inaccessible phenomena, such as nonlinear quantum interference effects in photoionization, and dressed-atom stabilization [3]. The coherent nature of seeded FEL pulses also opens up for ultrafast strong coupling in atoms and ions, which implies control over absorption and fluorescence processes [4]. In this talk, I will discuss how time symmetry of FEL pulse envelopes can be harnessed to control entanglement in photoionization [5]. Further, I will show how such envelopes can be used to reveal entanglement transfer from an electron-ion pair in photoionization, to an electron-photon pair in fluorescence [6].

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Complementarity in which-path resonant Auger scattering

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Different types of Young's double-slit experiments contain a significant amount of both particle and wave information running from full-particle to full-wave knowledge depending on the experimental conditions. We study [1] the Young's double-slit interference in resonant Auger scattering from homonuclear diatomic molecules where opposite Doppler shifts for the dissociating atomic slits provide path information. Our quantitative formulation of Bohr's complementarity principle—path information (W) vs interference (I) $W+I\leq 1$ is applied to two types of resonant Auger scattering experiments, with fixed-in-space and randomly oriented molecules. Special attentionis paid to the orientational dephasing in conventional Auger experiments with randomly oriented molecules.

This study was supported by the Ministry of Science and Higher Education of the Russian Federation, grant FSRZ-2023-0006.

Keywords: Young's double-slit experiments, Auger scattering, Bohr's complementarity principle.

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Simulating ultrafast nonlinear X-ray spectra with MCNOX

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We developed a program MCNOX[1,2] for computing and analyzing the ultrafast (attosecond and femtosecond) nonlinear X-ray spectra of molecules at the multiconfigurational quantum chemistry level. This tool is aimed at cutting-edge current and future photochemistry/photophysics applications enabled by X-ray free-electron lasers and high harmonic generation light sources. MCNOX can compute steady-state X-ray absorption spectroscopy (XAS) and three types of ultrafast nonlinear X-ray spectra: transient XAS,[3] all-X-ray four-wave mixing,[4] and stimulated Raman spectra.[5] It is especially capable of picking out major electronic transitions, and further computing the natural transition orbitals for these transitions, which help finally yield the physical and chemical insights from complex signals. Following the research paradigm of "electronic structure → molecular dynamics → signal," we have developed two earliest strategies based on "nonadiabatic molecular dynamics"[5] and "snapshots along the potential energy surfaces;"[3] to effectively simulate ultrafast nonlinear X-ray spectra, which have since been adopted by other researchers. Additionally, we are advancing a third computational strategy based on the full quantum wavepacket dynamics, achieving initial success in calculating steady-state X-ray spectra of diatomic molecules.[6]

Keywords: transient X-ray absorption, all x-ray four-wave mixing, stimulated Raman, multiconfigurational quantum chemistry

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Ultrafast non-adiabatic charge dynamics in (core) excited molecules

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When molecules are exposed to ultrashort laser pulses, coherent populations of several – often many – electronic states are typically generated, resulting in complex ultrafast dynamics in which electronic and nuclear motions are strongly coupled. A rapid redistribution of charge within the system is caused by the interplay between purely electronic charge migration and subsequent non-adiabatic processes, and the long-term structural evolution of the system is thereby influenced. In this talk, fully quantum simulations of ultrafast charge dynamics initiated by the excitation of polyatomic molecules will be presented. Electronic coherences created by both valence and core excitations will be examined, including cases where decoherence, revival of coherence, and transfer of electronic coherence through conical intersections are observed. The possibility for these dynamics to be traced by attosecond transient-absorption spectroscopy and time-resolved photoelectron spectroscopy will also be discussed.

Tracing spatiotemporal information in ultrafast dynamics

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We demonstrated that the quantum time delays of different ionization pathways in strong-field dynamics can be clearly resolved and extracted with semiclassical statistical methods and a large number of classical trajectories. Our results unified the seemingly opposing quantum Eisenbud-Wigner-Smith time delay and classical Coulomb-induced time delay by highlighting the same physical picture[1,2].We apply DL to perform Feynman's path integral approaches to demonstrate the classical-quantum correspondence[3]. We found that some quantum mechanical concepts such as "state", "excitation" and "resonance" in molecules can be expressed by statistical classical trajectories [4,5].

Keywords: attosecond physics; time delay; deep learning

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Attosecond Spectroscopy Reveals Spontaneous Symmetry Breaking in CO_2 Photoionization

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Spontaneous symmetry breaking induced by Renner-Teller distortion [1] in molecular CO2 photoionization is captured by attosecond photoelectron spectroscopy. The nondiabatic electron-nuclear coupling split the degenerate $A^2\Pi_u$ state of CO_2 molecules into two non-degenerate states with different geometries. These two states hold geometry-dependent shape resonances. By utilizing the vibrationally resolved RABBIT (Reconstruction of Attosecond Beating by Interference of Two-photon Transitions) spectroscopy [2]-[4], we captured the spontaneous symmetry breaking induced by the Renner-Teller effect by probing these two shape resonances. The RABBIT scan shows that the relative Wigner delays up to ~70 as between different vibrational states, which originate from the geometry-dependent shape resonances, with the combination of theoretical transition matrix analysis. The sub-10 as agreement with calculation allows us to determine the instantaneous time scale for the Renner-Teller splitting to occur.

Keywords: Attosecond Chronoscope, Renner-Teller Effect, Spontaneous Symmetry Breaking

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Flat-band $NbOCl_2$ materials in strong laser fields

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High harmonic generation (HHG) in solids is sensitively dependent on the band dispersions of crystals, which involves intraband and interband processes in conventional semiconducting materials. Here, we numerically simulate the HHG from a two-dimensional (2D) NbOCl2 material of a flat valence band in strong mid-infrared laser fields, which shows special harmonic signals that are predominantly originated from interband polarization. The suppressed intraband HHG is mainly attributed to its intrinsic heavy carrier with large effective mass. By bending the flat band under strain engineering, the HHG spectra of strained 2D NbOCl2 demonstrate typical features with below-gap harmonics and high-energy harmonics dominated by intraband and interband processes respectively. In addition, 2D NbOCl2 exhibits low exfoliation energy, excellent dynamical and thermal stability, and desirable optical property, which endow it with more experimental feasibilities.

Keywords: High-harmonic generation, NbOCl2 material, Flat band;

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Few-photon single ionization of laser-cooled Rubidium atoms

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In recent years, with the development of MOTReMi platform, laser-cooled alkali metal atoms have provided new insights into strong-field ionization, thanks to their unique shell structure and advantages in quantum manipulation of the Zeeman sublevels. In this talk, we will present some of our recent advancements along this direction, both experimentally and theoretically. In experiment, we use the magneto-optical trap (MOT) to prepare rubidium atoms at 5p excited states with a specified magnetic quantum number by destroying the symmetry of the cooling laser, and then use a 400-nm (or 800-nm) elliptically polarized femtosecond laser pulse to ionize the atoms and measure the photoelectron momentum distribution (PMD) with a reaction microscope (ReMi). Theoretically, we have developed an ab *initio* approach to treat the cooling and ionizing lasers on the equal foot. With the help of the simulations, we can determine the relative population of the ground-state and excited-state Rb atoms which exist simultaneously after laser cooling, by identifying the dominant ionization channels in various regions of the PMD observed in the experiment. Besides, we can also extract the relative amplitude and phase difference between the εs and εd ionization channels from the offset angle and interference structure in the PMD, achieving a *complete* measurement of the photoionization. In the two-photon near-resonance ionization, we will show how to use an elliptically polarized laser pulse to generate an unexpected three-fold AT splitting and control it with the ellipticity and helicity of the laser pulse. Applications of the scheme to realize selective population of dressed states (different branches of the AT splitting) and to extract additional phase shift (or time delay) induced by the Rabi oscillations through angular streaking will be also discussed.

Keywords: Laser cooling; few-photon ionization; *complete* measurement; AT splitting

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Photon Momentum Transfer and Partitioning: From One to Many

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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. 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].

Keywords: photon momentum, nondipole effect, above-threshold ionization

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Real time observation of the diffusion-limited formation of an ion-molecule complex

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Femtosecond time-resolved studies of molecular bond-breaking has been the subject of a vast number of studies since the pioneering work by Zewail and coworkers in the late 1980s and it remains a very active research area. Due to its unimolecular nature, bond breaking is fairly straightforward to explore experimentally.

Time-resolved studies of bond making between two molecules or atoms has also been explored but it faces one major challenge, namely how to clock the bimolecular reaction, i.e. measure when the two reactants meet because this is determined by diffusion – a process that is typically not controllable on ultrafast time scales. Previous works that explored bimolecular reactions used weakly bonded precursor complex of the two reactants to eliminate diffusion or introduced one of the reactants in a dense environment of reactants to minimize diffusion or allow it to be simulated by models.

In this talk, I will show recent experimental findings that allowed us to measure, in real time, the formation of the non-covalently-bonded Li^+-benzene complex, a textbook cation-π-system, starting from a Li^+ ion and a benzene molecule initially separated by 35 Å. Our experimental scheme employs nanometer-sized droplets of liquid helium to ensure a well-defined starting geometry for the diffusion process and ensuing bond formation, and to serve as a solvent into which the 1.5 eV binding energy of the cation-molecule complex can be dissipated. We measure the diffusion time directly and determine a diffusion velocity of 15 m/s, in fair agreement with fully atomistic modelling based on ring-polymer molecular dynamics simulations. Our work should enable real-time imaging of the stereodynamics, i.e. how the molecular orientation changes as the ion approaches.

Impulsive molecular alignment under resonant excitation

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Laser-induced molecular alignment has received significant attention over the past three decades [1]. Despite extensive investigation of alignment methodologies, the impact of electronic resonances has remained largely unexplored, with only a theoretical study reported in a 1995 paper by T. Seideman [2]. In this contribution, we will present what is, to the best of our knowledge, the first experimental observation of resonantly enhanced molecular alignment [3].

Our findings demonstrate that the optical coupling between two electronic states of the iodine molecule strongly affects the distribution of energies across various rovibronic states, resulting in a persistent planar alignment effect [4,5], which remains robust even above room temperature. As a generic phenomenon, this effect may have broader implication for other molecular systems.

Keywords: Molecular alignment, resonant absorption, femtosecond pulse, dipole moment, polarizability

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Coherent vibronic pseudorotation near conical intersection

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Electronic and vibrational coherence governs many light-driven dynamics in atto- and femtosecond processes, such as charge migration and energy transfer in molecules. As a paradigmatic example of aromatic molecules, the photophysics of benzene plays a vital role in many areas of chemistry and biology. The ultrafast charge transfer process and energy relaxation in complex aromatic molecules after photoexcitation are closely related to the non-adiabatic coupling between electrons and nuclei. One of the important non-adiabatic phenomena of benzene is the symmetry-induced (Jahn-Teller) conical intersection, which can lead to a large-amplitude internal motion within the ring molecule, which connects several stable isoenergetic structures separated by a saddle point and leads to the conformational changes of molecules. This dynamical evolution, dubbed pseudorotation, is a widely invoked kind of intramolecular motion. However, direct imaging of the JT-effect-induced vibronic pseudorotation in real time is still lacking.

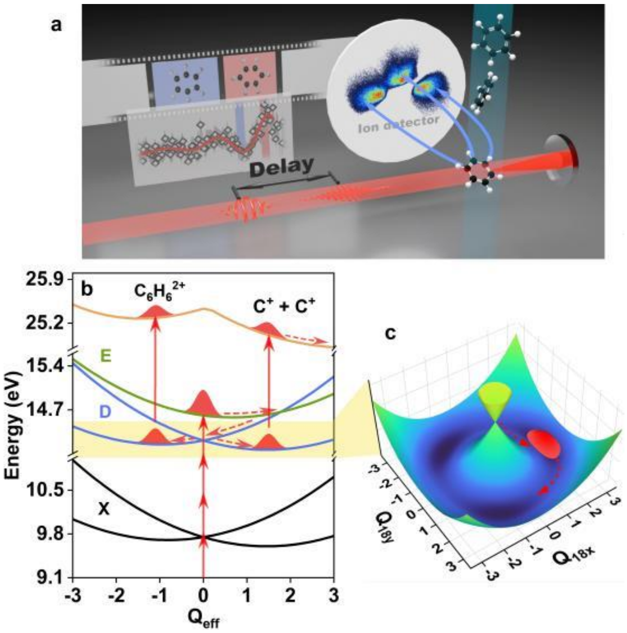

Figure 1: The experimental scheme a and potential energy surface of benzene b, the summary of the time-resolved structural evolutions for different electronic states. c shows the coherent vibronic motion of C6H6 wavepacket between three potential minima leads to the long-period oscillation of experimental ion yields.]

We study the coupled electronic-vibrational dynamics of a photoionized benzene molecule using the time-resolved Coulomb explosion method. An unusual long-period oscillation in the ion yields of dication and Coulomb explosion channels has been experimentally identified, and the theoretical simulation reveals this phenomenon as a ~600 fs oscillation of the nuclear wavepacket, which notably exceeds the period of any vibrational modes of the benzene cation. This exotic vibrational period originates from the quantum beating of interference between two vibronic states, arising from the Jahn-Teller coupling in the cationic excited states of benzene and the pseudorotation of the nuclear wavepacket. Furthermore, we achieved real-time visualization of the pseudorotation by directly capturing its transient structural evolution of the quantum beating between two vibronic states.

Recent Studies of Ultrafast Intermolecular Coulombic Decay and Proton Transfer in Bio-relevant Molecular Complexes

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Upon ionization in atomic and molecular complexes, charges can be dispersed across different units via interatomic or intermolecular relaxation processes. A prominent example of such mechanisms is the interatomic or intermolecular Coulombic decay (ICD) [1,2]. Beyond ICD, if the transferred energy exceeds the double-ionization threshold of the neighbor, double ICD (dICD) is initiated, resulting in double ionization of the neighbor with the emission of two low-energy electrons. Here, we verify dICD in bio-relevant molecular dimers consisting of benzene, pyridine, and water [3]. We found that in dICD the singlet states of the $C_6H_6^{++}$ dication are preferentially occupied upon energy transfer from the neighboring cation, which can lead to an enhancement of the ring-breaking fragmentation processes of the $C_6H_6^{++}$ dication.

When biomolecules are in an aqueous environment, a new relaxation mechanism may become operative, i.e. intermolecular proton transfer (PT) [4, 5]. We performed the experimental and theoretical studies of PT in hydrated pyrrole dimer upon double ionization of pyrrole molecule. This localized double ionization state leads to the PT from the initial doubly-ionized pyrrole to the neutral water forming the H_3O^+ + $C_4H_4N^+$ channel. Ab initio calculations and time-resolved measurements using fs laser were obtained to elucidate the mechanistic and dynamical details of ultrafast PT occurring in the hydrated biomolecular system. These results are essential to our understanding of radiation biological damage. More details will be presented at the conference.

Keywords: Intermolecular energy transfer, proton transfer, radiation biology

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ICD in liquid water competes with proton transfer and non-adiabatic relaxation

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Our understanding of radiation damage in aqueous systems remains surprisingly limited even after several decades of research. The recent observation of Intermolecular Coulombic Decay (ICD) in liquid water [1] raises the question of the efficiency of this process that is probably the most important direct source of low-energy electrons at the origin of radiation damage. To answer this question, we performed electron-electron coincidence measurements on liquid H_2O and D_2O sing a monochromatized high-harmonic-generation (HHG) light source. Electrons created by intermolecular Coulombic decay (ICD) were detected in coincidence with photoelectrons emitted from the inner-valence 2a1 shell. Remarkably, we find that the efficiency γ of ICD is below unity in both liquids and determine a relative efficiency of $\gamma(H_2O)/\gamma(D_2O) = 0.86 \pm 0.03$ [2]. Our ab-initio based calculations suggest that ICD stands in competition with proton transfer between neighboring water molecules and non-adiabatic relaxation, which can both close the ICD channel. A multi-scale stochastic model allows us to calculate ICD efficiencies, including the solvent effect. The combined experimental and theoretical results suggest that the higher ICD efficiency measured for heavy water is a consequence of the slower proton transfer and non-adiabatic transitions in D_2O, demonstrating the importance of nuclear motion in liquid-phase ICD. These results have implications for the understanding and modeling of radiation damage.

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Impact of Dimer Environments on Conical Intersection Dynamics via Interatomic Coulombic Decay (ICD) Reactions

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Conical intersections (CIs), degenerate points on molecular potential energy surfaces where electronic states cross, serve as pivotal gateways governing ultrafast photochemical processes such as photoisomerization, charge transfer, and radiationless decay. Through a synergistic theoretical-experimental approach, we unravel the Ar-mediated resonant charge transfer dynamics between He^{2+} and Ne atoms at nonadiabatic conical intersections: He^{2+} + Ne-Ar → He^{+} + Ne($2s^{-1}$)-Ar → ICD-driven fragmentation induced by 7.5 keV/u He^{2+} projectile collision [1]. Experimentally, the angularly resolved ion momentum imaging of Ne^{+} fragments generated through ultrafast interatomic Coulombic decay (ICD) of the excited Ne($2s^{-1}$)-Ar system serves as a critical probe for quantifying orientation-dependent reaction cross-sections. Theoretically, *ab initio* electronic structure calculations coupled with fully quantum mechanical time-dependent wavepacket propagation simulations provide atomistic insights into this stereodynamic process. Our findings reveal a striking fourfold enhancement in resonant charge transfer probability when He_2^{+} traverses perpendicularly through the NeAr cluster compared to parallel trajectories, demonstrating how anisotropic cluster geometries govern electron-nuclear coupling efficiencies at conical intersections.

Keywords: [Resonant charge transfer, Conical intersection, Interatomic coulombic decay]

Ref:

[1] Jiang. et al. Phys. Rev. A 100, 022705 (2019).

Possibility of predominant ICD in aromatic dimers

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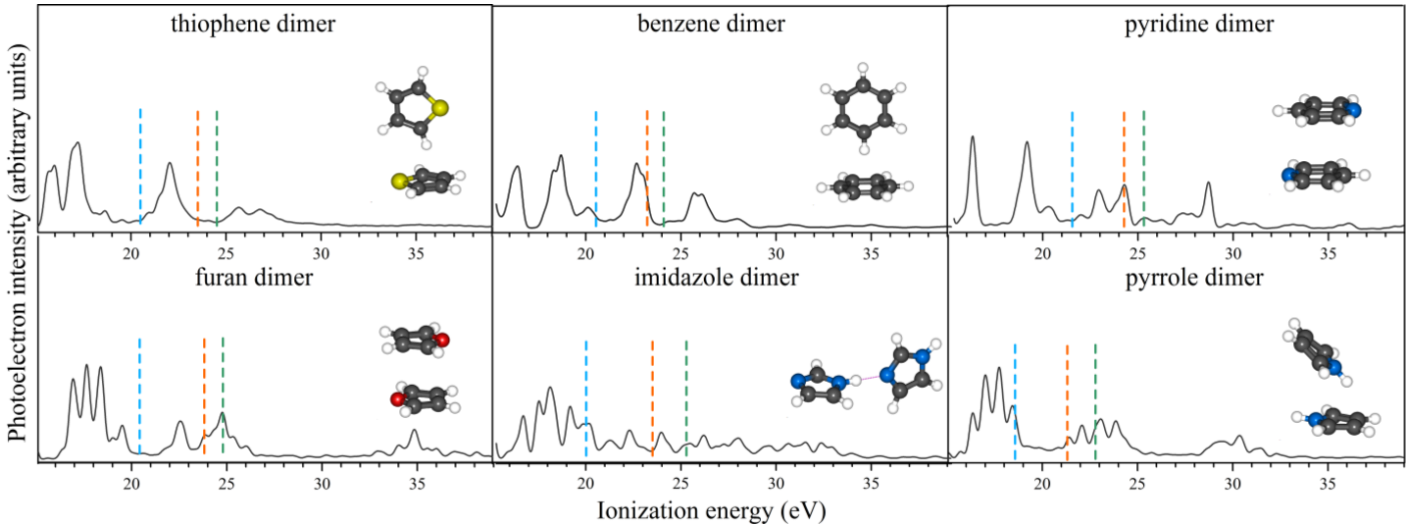
ICD [1] is a non-local auto-ionization process that occurs when highly excited cation-radicals, produced with ionization energies (IP) exceeding the double ionization threshold of the system, relax via ionization of the neighboring molecules. In most cases, ICD requires less energy than the corresponding local auto-ionization, namely Auger decay, while the latter is known to be faster when both channels are energetically possible. When the inner-valence IP band fits the gap between Auger and ICD thresholds, ICD can become the dominant decay channel for it, since the system does not have enough energy for local Auger decay. Such a case was recently found by us for the thiophene dimer, where the enhanced production of low-energy electrons was attributed to effective ICD from the states with inner-valence vacancy located predominantly on sulfur (Fig 1) [2]. In the talk, theoretical calculations illustrating these results will be presented in detail, and the cases of other aromatic dimers will be discussed (Fig 1).



[FIG. 1. Inner-valence IP spectra of heterocyclic dimers calculated at the ADC(3)/cc-pVDZ level. The energy thresholds of monomer double ionization, Auger and ICD decays, evaluated at the level of ADC(2)/cc-pVDZ, are shown as green, orange and blue dashed lines, respectively.]

Keywords: ICD (Intermolecular Coulombic decay), heterocycles, aromatic dimers, inner-valence ionization, Algebraic Diagrammatic Construction (ADC).

Ref:

[1] Jahnke T., Hergenhahn U., Winter B. et al. Chem. Rev. 2020, 120 (20), 11295–11369.

[2] Mootheril D. M., Skitnevskaya A. D., Ren X. et al. Commun. Chem. (under review)

Laser Coupling Induced Fine Structure in Vibrational Structures of O^{2+} Dissociation Revealed by High Resolution Measurement

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Vibrational structures in kinetic energy release spectrum of dissociation of molecules can reveal laser-induced dynamics. However, the resolution needed for measuring vibrational structure makes such kind of measurement still limited especially for molecules with complex electronic structures, such as oxygen molecules. Pioneering works by Zohrabi et al.[1] show laser-induced suppression of dissociation from certain vibrational level and further use this effect to identify dissociation pathway of molecules[2].

Here, we present our studies of laser-induced dissociation of O^{2+}. The O_2 target is ionized by laser and the KER spectra of O^{2+} are measured. We discovered additional fine structures within vibrational levels. Through experimental measurements and wave pack propagation calculation [3] we discovered the origin of such fine structures as laser coupling. This study advanced the understanding of laser-induced dissociation by joined experimental and theoretical studies.

Keywords: laser induced dissociation, kinetic energy release.

Ref:

[1] M. Zohrabi et al., Physical Review A **83**, 053405 (2011).

[2] T. Severt et al., Physical Review A **110**, 063112 (2024).

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L-edge fluorescence imaging of Kα spin-orbit doublet stimulated emission

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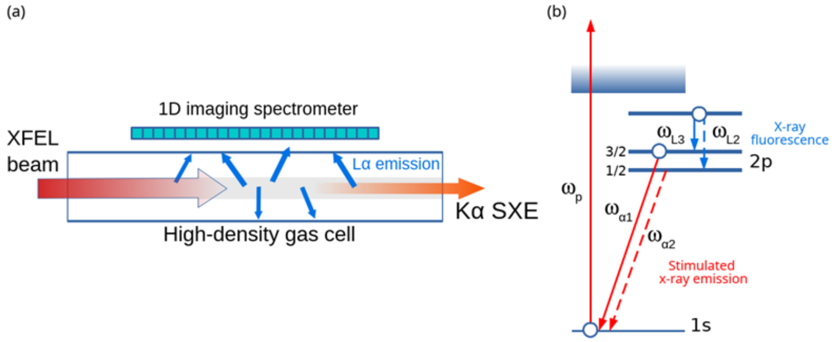
A new scheme for analyzing nonlinear effects in the x-ray range (Fig. 1a) is proposed and theoretically investigated by observing the propagation and amplification dynamics of Kα emission in a gas medium using one-dimensional (1D) imaging in the transverse direction.



In the scheme, the deep 1s orbital of an atom or molecule is ionized by a strong and short XFEL x-ray pulse, which induces Kα emission due to electron transition from the spin-orbit (SO) split 2p shell. When propagating along the gas cell under favorable conditions, stimulated x-ray emission of the Kα doublet can be induced. In this case, the relative population of the 2p1/2 and 2p3/2 final states will depend on the effects of x-ray lasing and the propagation length. In particular, since the spontaneous emission of the ωα1 line is twice as strong as that of the ωα2 line (Fig. 1b), under stimulated emission conditions, the first line will dominate while the latter will be suppressed. This phenomenon, in turn, has a significant effect on the L-edge fluorescence process that follows the Kα decay. Indeed, the suppression of the $\omega_{\alpha2}$ line leads to the dominance of the hole in the 2p3/2 state and thus to a significant amplification of the ωL3 line compared to the ω_{L2} line, as shown schematically in Fig. 1b. In our approach, we propose recording Lα fluorescence along the gas cell, which will allow tracking the effects of nonlinear propagation of Kα stimulated emission. The scheme offers a new alternative approach to tracking nonlinear effects in the x-ray range and can find application in various systems with SO-splitting, especially where direct observation of nonlinear phenomena is difficult.

This study was supported by the Ministry of Science and Higher Education of the Russian Federation, grant FSRZ-2023-0006.

Keywords: Kα emission, XFEL, nonlinear effects in the x-ray;

Generation of isolated white-light attosecond pulses in solids

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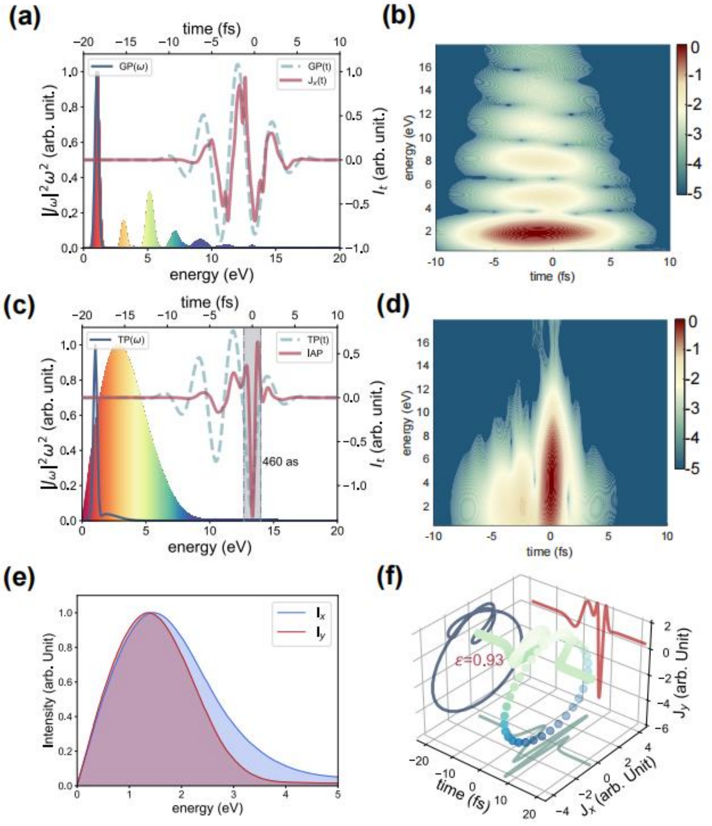
Attosecond-pump attosecond-probe spectroscopy (APAP) is the key to understanding electronic-dominated dynamics in light-matter interactions. However, the generation of IAPs capable of exciting neutral electronic excited states, which are more prevalent in nature, is still in its infancy, significantly limiting a comprehensive insights into attosecond-scale valence-electron wavepacket dynamics. Here, we theoretically demonstrate a novel scheme for the straightforward and compact generation of white-light IAPs covering the visible and ultraviolet regions, based on solid-state HHG.



Keywords: Isolated Attosecond Pulse; Solid-state High Harmonic Generation; Attosecond Science

Advancements and Emerging Programs in Ultrafast X-Ray Science at FERMI

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FERMI, operated by Elettra – Sincrotrone Trieste, is the first free-electron laser (FEL) facility that produces fully coherent pulses in the extreme ultraviolet (EUV) to soft X-ray range, utilizing two FEL lines with external seeding. This high degree of coherence and accurate control over the radiation properties have supported groundbreaking research in coherent control and attosecond science within the EUV-soft X-ray spectral range.

Currently, access to the attosecond timescale is achieved at FERMI through the control of the well defined radiation phase, instead of relying solely on pulse amplitude duration, which remains within tens of femtoseconds. Various techniques, including the use of phase-locked multicolor pulses and control of FEL pulse chirp, have allowed investigation of the response of matter at the few as time scale.

This presentation provides a comprehensive review of recent findings and ongoing developments at FERMI, aimed at further enhancing these capabilities.

Theoretical study of ultrafast dynamics of bimolecular interactions

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Bimolecular reactions involving intermolecular interations are much more general chemical reaction processes in nature in comparison to unimolecular reactions. Since the dimer systems have well-defined equilibrium geometries, the starting point of the light-driven bimolecular reaction can be precisely tracked in spatial and time domains. The formation of trihydrogen cations H_3^+ and its isotope D_3^+ from the hydrogen dimer represents a basic while pivotal instance among bimolecular reactions. Recent experimental advance [1] has allowed for precise temporal characterization of the formation dynamics of trihydrogen cations, pinpointing a reaction timescale on the order of hundreds of femtoseconds. Accordingly, our study systematically survey diverse theoretical approaches for numerically simulating bimolecular reaction dynamics, demonstrating the consistency and reliability of both classical and quantum methods in predicting the reaction timescale for D_3^+ formation [2]. Besides, by examining the H_2-D_2 dimer instead, We gain insights into the kinetic isotope effect in bimolecular reaction dynamics [3]. We have further conducted a systematic investigation to the bimolecular reaction of a heteromolecular dimer, H_2-CO [4]. The formation of the C-H bond is observed in the bimolecular reaction of two inorganic molecules, which is determined to be approximately 200 fs. Our theoretical study of ultrafast dynamics of bimolecular interactions can not only enhance the detailed understanding of bimolecular reaction processes, but also lay a foundation for subsequent research, which is expected to further understand and guide the complex processes in chemical reactions.

Keywords: bimolecular reaction, trihydrogen cations, kinetic isotope effect, C-H bond formation

Ref:

[1] L. Zhou et al., Nat. Chem. 15, 1229 (2023).
[2] H. Huang et al., Phys. Rev. A 109, 043107 (2024).
[3] M. Shi, H. Huang et al., Phys. Rev. X 14, 041001 (2024).
[4] Z. Jiang, H. Huang et al., Nat. Commun. 15, 2854 (2024).

eTraj.jl: Trajectory-Based Simulation for Strong-Field Ionization

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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. 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].

Keywords: strong-field ionization, electron trajectory, eTraj.jl

Ref:

[1] M. Zhu, H. Ni, and J. Wu, Comput. Phys. Commun. 311, 109549 (2025).

Subcycle Conservation Law in Strong-Field Ionization

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Exploiting the infinite-order continuous dynamical rotational symmetry of circularly or elliptically polarized classical light pulses, 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:

$$E = \omega L_z - I_p$$

where I_p is the ionization potential and ω is the angular frequency. In the asymptotic region, the conservation law becomes:

$$E = \omega L_z - I_p - U_p$$

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 on electron vortices to directly visualize the existence of the subcycle conservation law [1]. In what follows, we present two applications of this conservation law.

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:

$$\frac{1}{2}[p + A(t_s)]^2 + I_p + m\varepsilon\omega = 0,$$

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].

Keywords: subcycle conservation law, correlated spectrum of angular momentum and energy

Ref:

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[2] Y. Ma et al., Phys. Rev. A **111**, 033103 (2025).
[3] X. Mao et al., Phys. Rev. A **111**, 033113 (2025).

Comparison study of nanoplasmas ignition in helium nanodroplet induced by molecular and atomic dopants

Yue Hu[a,+], Zhengjun Ye[a,+], Jiaxuan Chen[a], Xinglei Long[a], Zhejun Jiang[a], Menghang Shi[a], Wenbin Zhang[a*], and Jian Wu[a*]

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The dopant-induced ignition of helium nanoplasmas under intense laser fields has emerged as a pivotal mechanism for generating extreme states of ionized matter [1]. Previous studies have demonstrated that dopants like metal atoms (K, Ca) [2,3] and rare gases (Xe) [4] significantly lower the ionization threshold of helium droplets and trigger the avalanche ionization in it by providing seed electrons and modifying local electric fields. However, these investigations remain confined to atomic dopants, while the role of molecular species, which may offer diverse electronic structures, enhanced electron donation, and tailored spatial configurations, remains unexplored. Understanding molecular dopants could unlock new pathways for controlling nanoplasma dynamics, optimizing energy absorption, and expanding applications in high-energy particle sources or ultrafast spectroscopy [4].

In this study, we conducted a comparison study on the ignition of helium nanoplasmas induced by the dopants of diatomic O_2 molecule and Xe atom, which have nearly the same first ionization energy. We found that when only a few molecules or atoms are doped, the overall ignition efficiency of O_2 dopant is lower than that of Xe, which can be ascribed to the weaker muliti-seed-electron providing ability of O_2 due to its higher second ionization energy. The mass effect also plays a role. We also observed that the O_2-doping-induced ignition shows a better performance with the increasing number of dopants for the less evaporation of He atoms. The dependence of relative abundances of He_n^+ ions from helium nanoplasmas on the number of dopants is also discussed. Furthermore, we check the effects from laser peak intensity and He cluster size on the difference between ignition efficiencies of Xe and O_2. We found that the former diminishes with the increasing number of dopants, and the latter is not significant on the relative abundances of He_n^+ ions.

This study highlights the critical role of dopant properties in optimizing helium nanoplasma ignition and underscores the untapped potential of molecular dopants. By exploring molecular species, which enable precise control over electron donation, spatial distribution, and ionization pathways, we can design more efficient ignition strategies. Such advancements promise to enhance the yield of energetic electrons and ions, pivotal for applications in compact particle accelerators, attosecond pulse generation, and laboratory astrophysics [5].

Key words： nanoplasma; avalanche ionization; helium nanodroplets; molecular dopants; femtosecond laser

+*These authors contributed equally*

Ref:

[1] Ostrikov, K., et al. *Reviews of Modern Physics*, *88*(1) (2016)., 011001.
[2] Heidenreich, A., et al. *New Journal of Physics* 18.7 (2016): 073046
[3] Heidenreich, A., et al. *Journal of Physics B: Atomic, Molecular and Optical Physics* 50.24 (2017): 244001.
[4] Medina, C., et al. *New J. Phys.* 23 (2021) 053011
[5] Schütte, B. *Progress in Ultrafast Intense Laser Science XIII*, (2017). 85-110.

Theoretical study of ultrafast dynamics of bimolecular interactions

Hao Huang[1], Hongcheng Ni[1,*], and Jian Wu[1,†]
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Bimolecular reactions involving intermolecular interations are much more general chemical reaction processes in nature in comparison to unimolecular reactions. Since the dimer systems have well-defined equilibrium geometries, the starting point of the light-driven bimolecular reaction can be precisely tracked in spatial and time domains. The formation of trihydrogen cations H_3^+ and its isotope D_3^+ from the hydrogen dimer represents a basic while pivotal instance among bimolecular reactions. Recent experimental advance [1] has allowed for precise temporal characterization of the formation dynamics of trihydrogen cations, pinpointing a reaction timescale on the order of hundreds of femtoseconds. Accordingly, our study systematically survey diverse theoretical approaches for numerically simulating bimolecular reaction dynamics, demonstrating the consistency and reliability of both classical and quantum methods in predicting the reaction timescale for D_3^+ formation [2]. Besides, by examining the H_2-D_2 dimer instead, We gain insights into the kinetic isotope effect in bimolecular reaction dynamics [3]. We have further conducted a systematic investigation to the bimolecular reaction of a heteromolecular dimer, H_2-CO [4]. The formation of the C-H bond is observed in the bimolecular reaction of two inorganic molecules, which is determined to be approximately 200 fs. Our theoretical study of ultrafast dynamics of bimolecular interactions can not only enhance the detailed understanding of bimolecular reaction processes, but also lay a foundation for subsequent research, which is expected to further understand and guide the complex processes in chemical reactions.

Keywords: bimolecular reaction, trihydrogen cations, kinetic isotope effect, C-H bond formation

Ref:

[5] L. Zhou et al., Nat. Chem. 15, 1229 (2023).
[6] H. Huang et al., Phys. Rev. A 109, 043107 (2024).
[7] M. Shi, H. Huang et al., Phys. Rev. X 14, 041001 (2024).
[8] Z. Jiang, H. Huang et al., Nat. Commun. 15, 2854 (2024).

Attosecond X-ray Core-level Chronoscopy of Aromatic Molecules

Jia-Bao Ji[1,*,†] and Zhaoheng Guo[2,3,4,*] on behalf of the LW94 collaboration team
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[3]*Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, CA, USA.*
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We conducted attosecond angular streaking to measure the relative photoionization time delays between N-1s and C-1s core shells 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 photoionization time delay into the momentum modulation in the polarization plane, which is recorded by the co-axial velocity map-imaging (c-VMI) spectrometer [2,3]. The atom-like core-level spectra avoid spectral congestion and convey information about the scattering dynamics from point-like sources.

We find that the delays systematically increase with the number of nitrogen atoms in the molecule and are featured by multiple resonances, which is supported by the calculation, as compared in Fig. 1. 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.

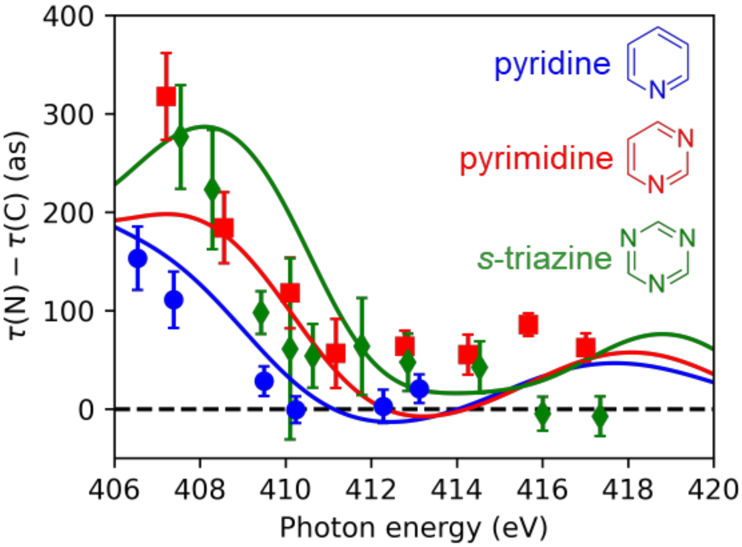

Figure 1. Measured relative photoionization time delays (markers with error bars indicating the 95% confidence intervals) compared to calculated N1s photoionization time delays (lines) including CLC delays for pyridine (blue circles), pyrimidine (red squares), and s-triazine (green diamonds).

Keywords: X-FEL, photoionization time delay, aromatic molecules, angular streaking

Ref:

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[2] Li S et al 2022 Science 375 285-290
[3] T Driver et al 2024 Nature 632 762-767

Advancements and Emerging Programs in Ultrafast X-Ray Science at FERMI

Enrico Allaria
on behalf of the FERMI team

FERMI, operated by Elettra – Sincrotrone Trieste, is the first free-electron laser (FEL) facility that produces fully coherent pulses in the extreme ultraviolet (EUV) to soft X-ray range, utilizing two FEL lines with external seeding. This high degree of coherence and accurate control over the radiation properties have supported groundbreaking research in coherent control and attosecond science within the EUV-soft X-ray spectral range.

Currently, access to the attosecond timescale is achieved at FERMI through the control of the well defined radiation phase, instead of relying solely on pulse amplitude duration, which remains within tens of femtoseconds. Various techniques, including the use of phase-locked multicolor pulses and control of FEL pulse chirp, have allowed investigation of the response of matter at the few as time scale.

This presentation provides a comprehensive review of recent findings and ongoing developments at FERMI, aimed at further enhancing these capabilities.

Vibrational-resolved attosecond photoionization delay of H2 molecules

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[Attosecond pulse serves as a powerful tool, enabling real-time observation and control of electron dynamics with high time resolution. Narrow-band attosecond pulse trains generated via high-harmonic generation (HHG) not only offer exceptional temporal resolution but also enhance the energy resolution to the molecular vibrational levels.]

In the RABBIT spectrum of the H2 molecule, five sidebands (SBs) corresponding to the X state were observed, specifically SB12 to SB20. Our high-energy resolution measurements clearly resolve all vibrational levels up to $v = 15$, as shown in Fig. 1. The ionization time delay for vibrational levels from $v = 1$ to $v = 6$ in the X state was extracted by isolating the contribution from the main band (MB), revealing a significant ionization delay for the lower sidebands (SB12 and SB14). In contrast, for higher-order sidebands (SB16 to SB20), the measurement indicates almost no ionization delay between vibrational levels. Additionally, the coupling between molecular vibrations and electron ionization, as well as the correlated motion of electrons and nuclei has been investigated. The contribution of vibration-dependent continuum-continuum transitions requires further study.

Keywords: [Attosecond photoionization, Ionization time delay, Molecular vibration]

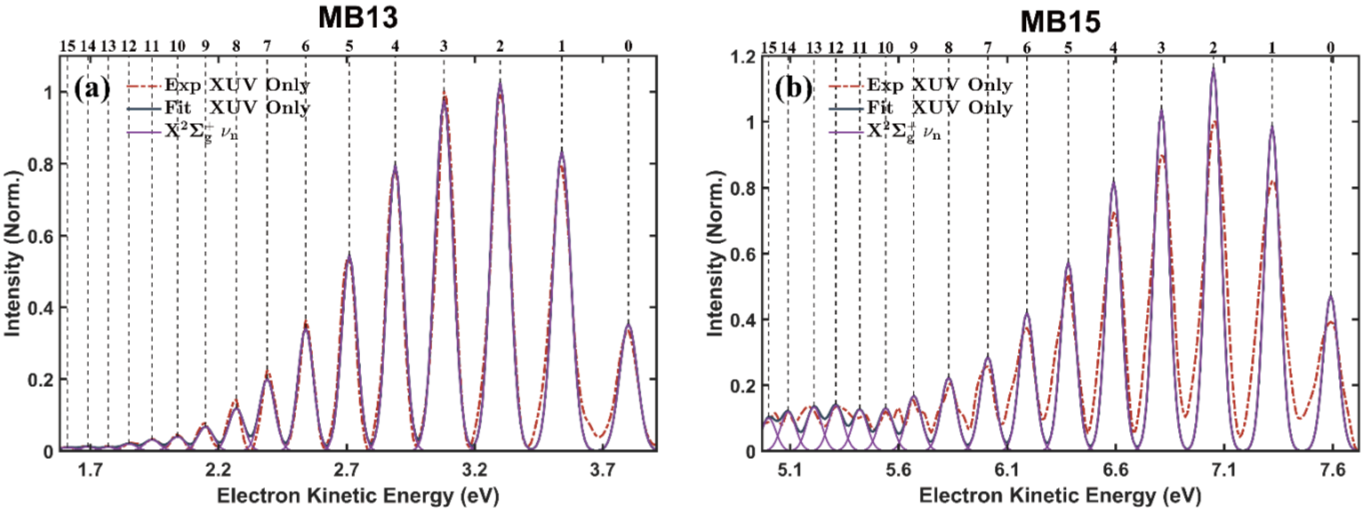

Figure 1. Photoelectron spectra obtained from XUV only from (a) H13 and (b) H15.

Attosecond XUV pump-control spectroscopy for state-selective tracking of dynamics in neutral molecules

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In a newly introduced pump-control scheme for attosecond XUV spectroscopy we focus on the time-domain interpretation of high-resolution absorption spectra [1]. Here, the molecular ground state reveals the system's dynamics through the associated dipole emission, thus acting as a self-probe for the intact neutral molecule, Fig.1(a). High-resolution experimental absorption spectra (4 meV at 15 eV) as in Fig.1(b) allow for the reconstruction of this time-dependent dipole response through a Fourier-based technique, previously developed for isolated atomic states [2]. The accessible ultrafast wave-packet dynamics, Fig.1(c), shows a modification upon interaction with a control NIR laser field of variable intensity.

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

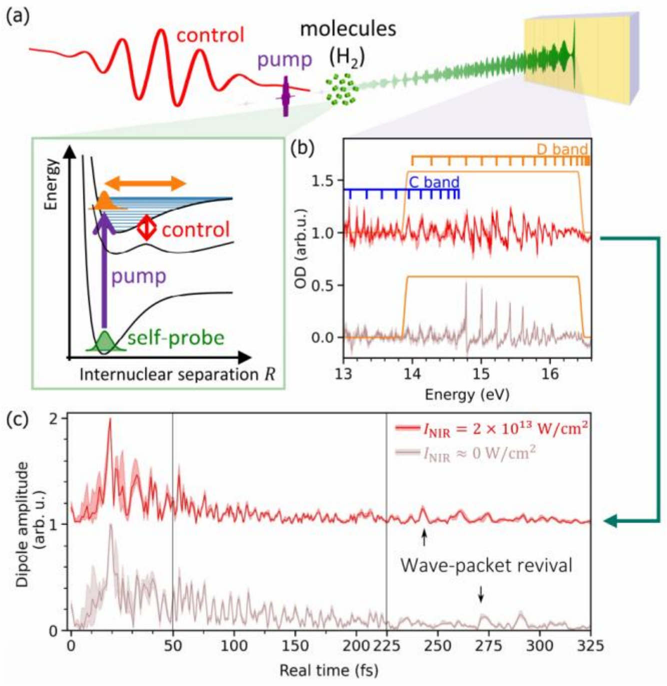

Fig. 1. (a) Experimental scheme for attosecond XUV pump-control spectroscopy in neutral molecules. (b) Measured optical density (OD) spectra of vibronic excitations in the neutral H2 molecule. (c) Reconstructed time-dependent dipole amplitude of the D 1Πu vibrational wave packet in an XUV-only configuration (lower curve) and for an NIR control field with $I_{NIR}\approx 2\times 10^{13}$ W/cm^2(upper curve), respectively.

Keywords: Wave-packet dynamics, Pump-control spectroscopy, Molecular self-probing

Ref:

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[2] V Stoß et al., Phys. Rev. Lett. 121, 173005 (2018)

Time-resolved imaging of methane fragmentation in strong laser fields

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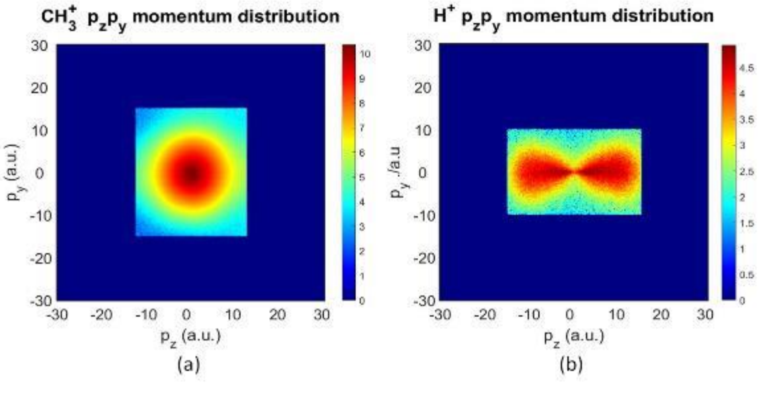
The ionization and dissociation dynamics of methane CH4 in strong laser fields are studied in a series of near-infrared (NIR) pump-probe experiments with a Reaction Microscope [1].

Upon few-cycle strong NIR pump pulses, methane is ionized to different cationic states (X or A states) and undergoes the Jahn-Teller distortions [2], leading to multiple dissociation pathways. A delayed probe pulse triggers Coulomb explosion (CE), dissociating the cations into identical final fragments through different intermediate geometries.

In two-body fragmentation, atomic hydrogen dissociates from methane as either neutral hydrogen H^0 or a proton H^+, characterized by distinct angular distributions as shown in Fig.1(a) and (b). Applying these angular-momentum characteristics to the pump-probe experiment, where both channels are mixed, we observe that the angular distribution of H^+ is initially aligned with the laser polarization but gradually becomes isotropic. This suggests that the branching ratio between the pathways from the X and A states changes as a function of time delay.



(a) (b)

Fig. 1 (a), (b) Angular momentum distributions from single-pulse measurements. The laser polarization is along the z-axis, with the molecular jet along the y-axis.

(a). H^0 from X state: $CH_4^+ \rightarrow CH_3^+ + H^0$ is released isotropically.

(b). H^+ from A state: $CH_4^+ \rightarrow H^+ + CH_3^0$ is dissociated along the laser polarization.

In three-body fragmentation of ($CH2^+$, H^+, H^+), four distinct fragmentation pathways are identified, providing insights into symmetry-breaking processes during molecular structural dynamics.

Keywords: Photofragmentation Jahn-Teller Effect;

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Complete experiment on atomic photoionization

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The amplitude and phase of a photoionization channel provide deep insights into the nature of ionization dynamics. In this work, we use a state-of-the-art in-house experimental setup, so called MOTReMi to perform one-photon single ionization into ϵs and ϵd with well controlled initial polarized state (*5p*) in Rb atoms, the simplest and cleanest benchmark experiment for complete measurements. We propose a novel method to retrieve the phase difference of the final-state wave function and the transition amplitude of two ionization channels from the photoelectron momentum distribution (PMD), where a 400 nm linearly / elliptically polarized femtosecond laser is used to ionize polarized Rb atoms in *5p* state with well controlled magnetic quantum numbers (m) by breaking down the symmetry of the laser cooling. From the tilt angle of the PMD and the interference structure of two outgoing ϵs and ϵd ionization channels, we are able to extract experimentally their relative amplitude and phase shift, in good agreement with ab initio calculations. The present scheme is rather robust in the sense that it is not sensitive to the ellipticity and intensity of the ionizing laser. This exhibits a benchmark one-photon single ionization study of the hydrogen-like atom, providing a complete measurement with full characterizations of intermediate and final state wave functions.

Keywords: photoionization time delay, magnetic quantum number control

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Laser Coupling Induced Fine Structure in Vibrational Structures of O_2^+ Dissociation Revealed by High Resolution Measurement

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Vibrational structures in kinetic energy release spectrum of dissociation of molecules can reveal laser induced dynamics. However, the resolution needed for measuring vibrational structure makes such kind of measurement still limited especially for molecules with complex electronic structures, such as oxygen molecules. Pioneering works by by Zoherabi et al.[1] shows laser induced suppression of dissociation from certain vibrational level and further use this effect to identify dissociation pathway of molecules[2].

Here, we present our studies of laser induced dissociation of O_2^+. The O_2 target is ionized by laser and the KER spectra of O_2^+ are measured. We discovered additional fine structures within vibrational levels. Through experimental measurements and wave pack propagation calculation [3] we discovered the origin of such fine structures as laser coupling. This study advanced the understanding of laser induced dissociation by joined experimental and theoretical studies.

Keywords: laser induced dissociation, kinetic energy release.

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Investigating Ultrafast Nuclear Dynamics in SF_6 via Nonlinear Spectral Dispersion in Resonant Auger Scattering

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In this study, we integrate experimental observations and theoretical models to elucidate the complex phenomena observed in the resonant S K-edge Auger spectra of SF_6. By constructing a two-dimensional spectral map based on photon and Auger electron kinetic energy, we reveal nonlinear spectral dispersion resulting from the interplay between electronic and nuclear degrees of freedom. This approach enables the extraction of detailed information about molecular potentials and ultrafast dynamics in the excited state, highlighting the significance of accounting for molecular geometry changes when interpreting spectral data. Our methodology offers a novel avenue for investigating molecular potentials and ultrafast dynamics in complex polyatomic molecules through resonant Auger spectroscopy. Theoretical modelling was supported by the Ministry of Science and Higher Education of Russian Federation (project FSRZ 2023-0006).

Keywords: ultrafast nuclear dynamics, nonlinear spectral dispersion, resonant Auger scattering, SF_6 molecule, molecular geometry changes

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Coherent control via ion-photoelectron entanglement transformation induced by Rabi oscillations

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Coherent control, enabled by the ability to manipulate the dynamics of a quantum system through independently tunable parameters, plays a crucial role in the study of ultrafast processes [1,2]. In this work, we employ an $\omega + 2\omega$ pulse pair to ionize neon, inducing Rabi oscillations. These oscillations drive the evolution of the ion-photoelectron entangled state [3], thereby enabling coherent control over the system.

We perform ab initio calculations to investigate the ionization dynamics of a neon atom subjected to an $\omega + 2\omega$ pulse pair. Following ionization, the system forms an entangled ion-photoelectron state, which undergoes transformation due to Rabi oscillations. By tuning the relative phase between the two laser fields, we achieve coherent control over the system's evolution. Our results reveal a strong dependence of photoelectron angular distributions (PADs) on the relative phase, exhibiting significant variations in asymmetry. Moreover, our approach is applicable to other multielectron systems, offering a pathway for controlled manipulation of quantum entanglement in ultrafast processes.

Keywords: Coherent control, entanglement manipulation, asymmetric photoelectron momentum distribution, multielectron dynamics.

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Femtosecond atomic-scale diffraction imaging of molecular quantum dynamics

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The pursuit of capturing molecular movies has been a longstanding goal in ultrafast science. Time-resolved ultrafast X-ray diffraction (UXD) and ultrafast electron diffraction (UED) have demonstrated significant advantages in probing molecular structural evolution and electron-electron correlations. Here, we report the simultaneous imaging of molecular structural and electronic dynamics at the quantum level using MeV UED, achieving unprecedented spatiotemporal resolution.

This high spatiotemporal resolution is enabled by combining gas-phase MeV UED with a super-resolution inversion algorithm, which is crucial for investigating nonadiabatic dynamics characterized by ultrafast timescales and subtle structural changes. Applied to the ring-opening reaction of 1,3-cyclohexadiene, our approach reveals two conical intersections (CIs) occurring within 100 femtoseconds, with a nuclear wave packet traversal time of approximately 30 femtoseconds and a C–C bond length difference of less than 0.4 Å. These findings provide a direct real-time visualization of CI dynamics, offering critical insights into bond breaking and ring-opening processes.

Additionally, we investigate the photoinduced dynamics of cyclobutanone under deep-UV laser excitation using MeV UED. Both elastic and inelastic scattering signals reveal a sub-picosecond photodissociation timescale and an excited-state lifetime of approximately 230 femtoseconds. Our results quantitatively determine the branching ratio of photoproducts and capture the complete photochemical pathway, spanning from the S_2 minimum, through the S_1/S_0 CI, to final dissociation. This study provides new insights into Norrish Type I reactions and serves as a benchmark for nonadiabatic dynamics simulations.

Keywords: diffraction, super-resolution, nonadiabatic, ring-opening, dissociation

Photoelectron Spin Texture in Strong-field Ionization Induced by a Linearly Polarized Laser Pulse

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Synopsis: We demonstrate a nontrivial spin texture of photoelectrons in the strong-field ionization of inert gas atoms, exhibiting a vortex structure relative to the laser polarization axis. The momentum-resolved polarization originates from the emergence of spin-correlated quantum orbits in the continuum. For direct electrons in few-cycle pulses, the nonvanishing initial transverse velocity of the electron is responsible for the polarization. In long pulses, however, the spin texture is essentially shaped by recollisions. Furthermore, the interference between direct and rescattering ionization leads to spin-polarized electron holography, offering an alternative method for extracting atomic fine structural information.

Spin-orbit coupling in bound states can create a strong correlation between the electron's spin and orbital angular momentum due to the degenerate atomic energy levels. This correlation, combined with significant angular momentum-dependent ionization probability, results in photoelectron polarization. When combined with circular dichroism in strong-field ionization, significant polarization along the laser polarization axis becomes possible.

In contrast to a circularly polarized pulse, the total polarization of the ionized electron vanishes when the laser pulse is linearly polarized. Nevertheless, we demonstrate the emergence of significant momentum-resolved spin polarization (spin texture). We trace the origin of the spin texture to the correlation between spin and the initial transverse velocity of the photoelectron at the tunnel exit, which gives rise to spin-dependent quantum orbits. For direct electrons, using saddle point approximation in spin-resolved strong-field ionization, we demonstrate that the spin polarization forms a vortex relative to the laser polarization axis:

$$\langle \boldsymbol{\zeta}(\boldsymbol{p}) \rangle \sim \frac{\boldsymbol{p} \times \boldsymbol{E}(t_r)}{|\boldsymbol{E}(t_r)|},$$

where $\boldsymbol{p}$ is the photoelectron momentum and $\boldsymbol{E}(tr)$ is the electric field at the instant of ionization. For rescattering electrons, the nontrivial momentum-resolved spin texture arises from forward rescattering of the spin-dependent quantum orbits and the associated quantum phase shift.

Moreover, the interference between direct and rescattered trajectories can lead to photoelectron holography of the spin polarization. As shown in Fig. 1, the typical spider structure is not only visible in the momentum distribution but also appears with enhanced resolution in the spin polarization.

The photoelectron spin texture is generally nontrivial in strong-field ionization, which can provide complementary information to the photoelectron momentum distribution. The nontrivial spin texture not only shows promise for enhancing the sensitivity of ultrafast electron spectroscopic techniques—such as photoelectron holography, attoclock, and streak camera measurements—but also may reveal novel methods for generating spin-polarized photoelectrons.

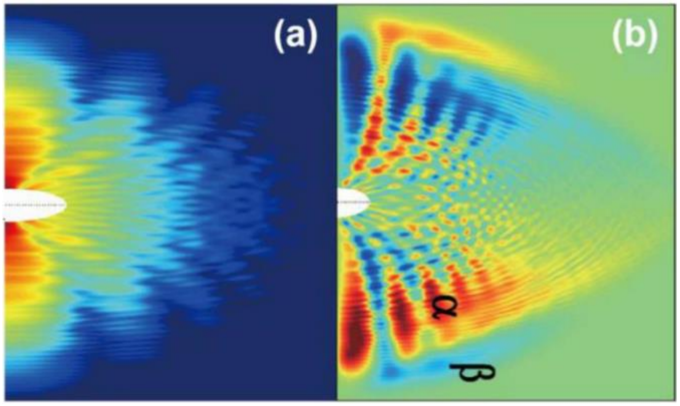

Figure 1. Photoelectron holography fringes observed in (a) photoelectron momentum distribution and (b) spin texture from a Xe atom.

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Correlation-Driven Anomalous Photoelectron Angular Distribution in Multiphoton Single Ionization of Argon

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In single ionization induced by photon absorption, a neutral atom is broken into a two-body complex consisting of an electron and an ion. Conserved physical quantities carried by the eliminated photons, such as energy and momentum, are shared by the newborn particle pair. Although linearly polarized lasers only transfer net-zero angular momentum projection alongside the polarization direction, the sharing of angular momentum may still render the complex in an entangled state. We take (4+1) REMPI of argon driven by an intense ultraviolet laser as a prototype, where the aforementioned interchange is possible through the Coulomb interaction between the photoelectron and the other electrons in the valence shell, and the multi-pathway interference may affect the probability to emit photoelectrons in $d_{0,\pm 1}$ partial waves, The ab initio theoretical study by numerically solving the multielectron time-dependent Schrödinger equation with multi-configurational time-dependent Hartree-Fock (MCTDHF) method [1,2] and R-matrix with time-dependence (RMT) method [3], along with the experiments employing the photoelectron detection technique of cold-target recoil ion momentum spectroscopy (COLTRIMS), robustly confirm the existence of such process at the four-photon resonance with 4p near a wavelength of 367nm [4].

Keywords: multi-photon ionization, multi-configurational time-dependent Hartree-Fock, electron entanglement

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Unveiling the ultrafast photodynamics of urea solutions probed by transient absorption spectroscopy

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Urea is a fundamental component in biological metabolic processes and industrial chemistry; investigations of the photoinduced reactions of urea are critical in uncovering the origin of life [1]. The isolated molecules could undergo complex ultrafast processes under the excitation of short-wavelength ultraviolet light (UV), e.g., electronic state transitions and vibrational relaxation [2]. In solution environments, the excited-state dynamics could perform much more complicated pathways due to the influence of hydrogen-bond between urea with solvent water molecules [3, 4]. However, in-depth study of the mechanism urea solution, especially excited by the UV lasers is still lacking.

Thus, we performed a femtosecond transient absorption spectroscopy measurement of urea solution, utilizing 266 nm pump excitation and broadband white-light continuum probe (300-750 nm). The excited-state dynamics of urea molecules in solution are systematically investigated employing concentration-dependent solvent comparison experiments to unravel solvent-solute interactions. From the 2D spectroscopy, at least two main mechanisms are identified utilizing the singular value decomposition analysis and global fitting method, exhibiting fast and slow processes lasting from fs to ps. A distinct absorption peak has been observed around hundreds of femtoseconds, indicating different decay lifetimes at different concentrations.

Keywords: Femtosecond Transient Absorption Spectroscopy, Urea Solution, Pump-Probe

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A liquid-phase high-order harmonic generation apparatus for investigating ultrafast dynamics in liquids

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High-order harmonic generation (HHG) is crucial in ultrafast science due to its ability to generate attosecond pulses and study the structure and dynamics of atoms, molecules, and solids on the electronic time scale[1]. While extensive studies in gases and solids HHG have unveiled their underlying microscopic mechanisms, liquid HHG remains in its infancy. Recent experiments have observed extreme ultraviolet (EUV) high-harmonic radiation from liquid samples[2,3], but the complex disordered nature of liquids complicates the development of a universal theoretical model for the observed harmonic features, necessitating further research.

In this work, we built a liquid HHG apparatus and conducted HHG experiments using liquid water, ethanol, and isopropanol as samples, driven by 800 nm laser pulses. The high-order harmonic spectra (HHS) of both liquid ethanol and isopropanol have a cut-off up to the eleventh order, while that of water extends to the thirteenth order. Additionally, the harmonic intensity of water shows a characteristic envelope with a sharp cut-off region following a plateau. These quantitative differences of the HHS spectra from different samples provide a reliable tool to unfold the ultrafast electronic dynamics, probing the subtile molecular structure in the bulk.

Keywords: Liquid, ultrafast, HHG;

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Design of an extreme ultraviolet monochromator for attosecond time-resolved photoelectron spectroscopy

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Attosecond time-resolved photoelectron spectroscopy is a key tool for studying ultrafast dynamics in molecules and materials. Its core is to capture transient electronic structure changes using extreme ultraviolet (XUV) light sources with high temporal resolution (from femtoseconds to attoseconds) and energy resolution. This paper presents the design of a XUV monochromator, based on high-order harmonic generation (HHG) and conical diffraction technology [1], which provides a high energy resolution and short pulse XUV light source.

The monochromator consists of three main parts: (1) XUV generation module: A lens focuses an 800 nm, 30 fs near-infrared laser pulse into the high-pressure region of a semi-infinite gas chamber [2] to generate XUV harmonics covering 10-50 eV; (2) Spectral separation module: A grazing-incidence plane grating spatially separates harmonics of different wavelengths, which are then collimated and focused by toroidal mirrors. A slit is used to select harmonics of specific energy; (3) Detection module: A microchannel plate (MCP) and fluorescent screen detector can replace the slit, enabling real-time monitoring of the spatial distribution of the harmonics.

In the future, optimizing grating efficiency and laser parameters could further enhance flux and extend the energy range, providing a more powerful tool for studying ultrafast dynamics in complex molecular systems and condensed matter.

Keywords: Extreme ultraviolet monochromator, Time-resolved photoelectron spectroscopy, High-Harmonic Generation;

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