International Conference on Many Particle Spectroscopy of Atoms, Molecules, Clusters and Surfaces

April 23-26 2024, Shanghai, China

MPS2024

About MPS

MPS (The International Conference on Many Particle Spectroscopy of Atoms, Molecules, Clusters and Surfaces) is a biennial meeting of scientists to discuss the physics and chemistry related to the interaction of light and charged particles (ions and electrons) with various forms of matter, ranging from atoms to nanoparticles and surfaces. In particular the latest experiments, techniques and theoretical results involving multiple particles, their interaction (correlation, scattering, dynamics, multiphoton absorption etc.) and detection (such as coincident experiments) are presented.

The history of MPS and its predecessors goes back to the year 1986. The latest MPS Conference was held in Turku, Finland in 2022. The most recent meetings have been held in Budapest (Hungary, 2018), Moscow (Russia, 2016), Metz (France, 2014), Berlin (Germany, 2012), Sendai (Japan, 2010), Paris (France, 2008), Rome (Italy, 2006).

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International Conference on Many Particle Spectroscopy of Atoms, Molecules, Clusters and Surfaces

Program Overview

April 23-26th 2024 Fudan University, Shanghai

April 24 (Wed)		
9:00-9:15	**MPS Opening, Chair: Baoren Wei**	
Chair: Dajun Ding		
9:15-9:45	**Toshiyuki Azuma, RIKEN, Japan**	*High-resolution spectroscopy of electronic K x rays from muonic atoms using transition-edge sensor microcalorimeters*
9:45-10:15	**Xueguang Ren, Xi'an Jiaotong University, China**	*Intermolecular energy and charge transfer in molecular complexes upon electron impact*
10:15-10:30	**Pengju Zhang, ETH Zürich, Switzerland**	*Unveiling the ultrafast relaxation dynamics of thymidine in aqueous solution via extreme ultraviolet time-resolved photoelectron spectroscopy*
10:30 ~ 11:00 Photo & Coffee Break		
Chair: Marc Vrakking		
11:00-11:30	**Mathieu Gisselbrecht, Lund University, Sweden**	*An insight into attosecond photoionization*
11:30-12:00	**Sebastian Eckart, Frankfurt University, Germany**	*Ultrafast preparation and detection of entangled atoms using a COLTRIMS reaction microscope*
12:00-12:30	**Rocio Borrego-Varillas, CNR-Istituto di Fotonica e Nanotecnologie, Italy**	*Ultrafast electron transfer in donor-acceptor molecules tracked by attosecond spectroscopy*

12:30-13:45 Lunch		
Chair: Károly Tőkési		
13:45-14:15	**Sergio Díaz-Tendero, Universidad Autónoma de Madrid, Spain**	*Peptide bonds formation in clusters of amino acids induced by ionizing radiation*
14:15-14:45	**Chuncheng Wang, Jilin University, China**	*Directly imaging excited state-resolved transient structures of water*
14:45-15:00	**Yun Li, American Physical Society**	*Meet Phys Rev editor – An introduction of Physical Review journals*
15:00-15:15	**Lishu Wu, Nature publishing office**	*Launching Nature Reviews Electrical Engineering*
15:15 ~ 15:45 Coffee Break		
Chair: Masahiko Takahashi		
15:45-16:15	**Arnaud Leclerc, Université de Lorraine, France**	*Calculation of molecular ionization cross sections using analytical Gaussian integrals*
16:15-16:45	**Shenyue Xu, CAS-Institute of Modern Physics, China**	*Heavy-ion-induced intermolecular Coulombic decay in hydrated biomolecules*
16:45-17:15	**Satoru Kanaya, Tohoku University, Japan**	*Application of electron-atom Compton scattering to measurements of individual atomic motions in polyatomic molecules*
17:15-17:45	**Alexander Zaytsev, Pacific National University, Russia**	*The parabolic quasi-Sturmian approach to single ionization of Helium by high energy protons*
17:45 ~ 19:00 Snack in poster region		

April 25 (Thur)		
Chair: Edwin Kukk		
9:00-9:30	**Oksana Travnikova, Sorbonne University, France**	*Exploring ultrafast dynamics with advanced electron-ion coincidence methods under soft and hard x-ray excitations*
9:30-10:00	**Marc Simon, Sorbonne University, France**	*Post Collision Interaction, Photoelectron recapture and Angular Momentum transfer in the hard X-ray domain. From the isolated Atoms and Molecules to condensed phase*
10:00-10:30	**Mizuho Fushitani, Nagoya University, Japan**	*Transient core-to-core resonances of Kr in intense EUV-FEL fields studied by electron-ion coincidence spectroscopy*
10:30-11:00 Coffee Break		
Chair: Xiangjun Chen		
11:00-11:30	**Kade Spicer, Curtin University, Australia**	*Energy and angular distributions of electrons produced in intermediate-energy proton-helium collisions*
11:30-11:45	**Nicolas Sisourat, Sorbonne University, France**	*Intermolecular Coulombic Electron Capture in Aqueous Systems*
11:45-12:00	**Anna Skitnevskaya, Irkutsk State University, Russia**	*Intermolecular Coulombic decay of inner-valence vacancies in biologically relevant systems. Theory meets experiment*
12:00-12:15	**Bocheng Ding, ShanghaiTech University, China**	*Mapping the potential energy curve of Auger final state from resonant decay in O_2 molecules*
12:15-12:30	**Pufang Ma, Fudan University, China**	*Studying of charge exchange cross section of highly charged ion interact with atom and molecule*
12:30-13:45 Lunch		

Chair: Xinwen Ma		
13:45-14:15	**Elena Gryzlova, Lomonosov Moscow State University, Russia**	*An efficient method of accounting polarization in sequential processes*
14:15-14:45	**Jiabao Ji, ETH Zürich, Switzerland**	*Relation between photoionisation cross sections and attosecond time delays*
14:45-15:15	**Victor Kimberg, Royal Institute of Technology, Sweden**	*Nonlinear spectral dispersion in resonant x-ray scattering for studying potentials and dynamics*
15:15 ~ 15:45 Coffee Break		
15:45-17:45	**Poster Section**	
18:15 ~ 21:00 Conference Dinner		

April 26 (Fri)		
Chair: Jianmin Yuan		
9:00-9:30	**Lynda Hutcheson, Queen's University, Ireland**	*Resonance Analysis of Strong Field Ionisation*
9:30-10:00	**Yu He, Max-Planck-Institut für Kernphysik, Germany**	*Controlling Photo absorption Interferometrically with Intense Laser Pulses from Microscopic to Macroscopic Gases*
10:00-10:30	**Tomoyuki Endo, National Institutes for Quantum Science and Technology, Japan**	*Capturing roaming molecular fragments in Formaldehyde*
10:30-11:00 Coffee Break		

Chair: Alexander Dorn		
11:00-11:30	**Matthieu Génévriez, Université Catholique de Louvain, Belgium**	*Double Rydberg states of alkaline-earth atoms: two-electron dynamics far from the nucleus*
11:30-11:45	**Weifeng Yang, Hainan University, China**	*Application of Semi-Classical Methods in Ultrafast Electron Dynamics Information Probing*
11:45-12:00	**Eemeli Aulis Eronen, University of Turku, Finland**	*Statistical analysis of X-ray spectra of aqueous triglycine*
12:00-12:15	**Jinfeng Chen, University of Science and Technology of China, China**	*Non-resonant elastic and inelastic x-ray scattering study of nitrous oxide*
12:15-12:30	**Dongdong Zhang, Jilin University, China**	*New source for tuning the effective Rabi frequency discovered in multiphoton ionization*
12:30-13:45 Lunch		
Chair: David Busto		
13:45-14:15	**Hongcheng Ni, East China Normal University, China**	*Time-resolved electron dynamics from the XUV to the IR regime*
14:15-14:45	**Divya Bharti, Max-Planck-Institut für Kernphysik, Germany**	*Multisideband Photoelectron Interferometry in Atoms and Molecules*
14:45-15:00	**Keyu Chen, Kansas State University, America**	*UV-induced ring conversion dynamics investigated by time-resolved Coulomb explosion imaging*
15:00-15:15	**Huanyu Ma, ShanghaiTech University, China**	*Complete characterizations of intermediate and final state wave functions with photoionization of polarized Rb*
15:15-15:45	**MPS Closing, Chair: Kiyoshi Ueda**	
15:45～21:00 City Center and Boat Dinner		

Abstracts of invited talks

High-resolution spectroscopy of electronic *K* x rays from muonic atoms using transition-edge sensor microcalorimeters

***T. Azuma*[1]**

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Muonic atoms, where a negative muon is captured onto the atomic orbitals, provide a unique window into strong electric field atomic physics because muons are 207 times closer to the nucleus than electrons and thus probe high Coulomb fields. Precision study of few-electron, high-Z ions is a privileged field for probing high-field, bound-state quantum electrodynamics (BSQED). We proposed an alternative method with exotic atoms and showed that transitions may be found between circular Rydberg states where nuclear contributions are vanishing while BSQED effects remain large [1]. Using superconducting transition-edge sensor (TES) microcalorimeters (5.2–5.5 eV FWHM resolution), we recently performed a proof-of-principle experiment at J-PARC(Japan) for high-precision muonic x-ray spectroscopy of the 5g-4f and 5f-4d transitions (BSQED contribution of 2.4 and 5.2eV, respectively) of muonic neon (μNe) atoms in the low-pressure gas phase without bound electrons. We determined the $5g_{9/2}$ - $4f_{7/2}$ transition energy to be 6297.08 ± 0.04(stat) ± 0.13(sys) eV, which agrees well with the most advanced BSQED theoretical prediction of 6297.26 eV [2].

Not only muonic x rays, we also succeeded in observing electronic *K* x-rays emitted from muonic atoms. We measured electronic K x rays from muonic iron (μFe) in a metal. The excellent energy resolution of the TES detectors allowed us to observe the asymmetric broad profile of the electronic characteristic K_α and K_β x-rays together with the hypersatellite K^h_α x rays around 6 keV. This signature reflects the time-dependent screening of the nuclear charge by the negative muon and the *L*-shell electrons, accompanied by electron side feeding. These data, assisted by a simulation, clearly reveal the electronic *K*- and *L*-shell hole production and their temporal evolution on the 10–20 fs scale during the muon cascade process [3,4].

More recently, we measured electronic *K* x rays emitted from muonic argon (μAr) atoms in the gas phase. We obtained a clear signature of the presence of muonic atoms accompanied by a few electrons. Not only He-like and Li-like muonic atoms but also H-like muonic atoms were identified from hyper-satellite K x rays.

These findings allowed us to revisit the research field of highly-charged ion spectroscopy and its interaction with matter. They also shed light on the interaction between a muon and electrons in the deexcitation process, which has not been well understood due to the lack of experimental tools.

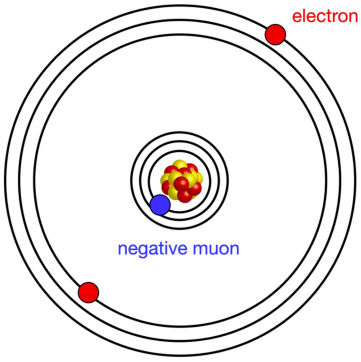

Figure 1. schematic structure of highly-charged muonic atoms.

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Recent studies of intermolecular Coulombic decay and proton transfer processes in bimolecular dimers by electron-impact ionization

***X. Ren*, J. Zhou, S. Jia, X. Xue, X. Hao**

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The radiolysis of biomolecules is of great importance to a range of chemical and biological processes. The biomolecules can be excited or even ionized when they are exposed to high energy ionizing radiation and the excited molecules are often unstable tending to dissociate. The presence of weakly bound neighbors may substantially reduce the probability for fragmentation of the initially ionized molecules, which can manifest as a protective effect in biomatter. On the other hand, the local environment in form of hydrogen-bonded molecules can also act as a catalyst and considerably affect the stability of molecular covalent bonds. However, the mechanisms and dynamics of ionization-induced relaxation processes in biomolecular systems remain largely unexplored due to the complex non-covalent interactions in these systems.

Recently, we studied the intermolecular energy and proton transfer in several bio-molecular dimers initiated by electron-impact. The experiments were performed using a COLTRIMS reaction microscope [1-3], where the outgoing electrons and ions are detected in coincidence and their momentum vectors are determined in three-dimensional kinematics. First, we investigate the non-local relaxation processes that follow inner-valence ionization in a range of molecular dimers involving various combinations of benzene, pyridine, and pyrimidine, which initiate an ultrafast inter-molecular Coulombic decay (ICD) process [4]. Multi-particle coincidence momentum imaging, combined with ab initio calculations, enables us to explore the principal orientations of these fundamental dimers and, thus, to elucidate the influence of N heteroatoms on the relative preference of the aromatic π-stacking, H-bonding, and CH−π interactions and their dependence on the number of nitrogen atoms in the rings (as shown in Fig. 1) [5].

When biomolecules are in an aqueous environment, a new relaxation mechanism may become operative, i.e. intermolecular proton transfer (PT) [6, 7]. We performed experimental and theoretical studies of PT in hydrated pyrrole dimer upon electron-impact double ionization of pyrrole molecule. In the experiment, ICD will lead to the dissociation channel H_2O^+ + $C_4H_5N^+$. Here, ICD can be initiated with the removal of a nitrogen or oxygen 2s inner-valence electron. Afterward, an electron from the outer-valence shell of $C_4H_5N^+$ or H_2O^+ fills the inner-valence vacancy, and the energy released ionizes neighboring molecule. While the localized double ionization of the complexes may cause 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 are also obtained to elucidate the mechanistic and dynamical details of ultrafast PT occurring in the hydrated biomolecular system. More details will be presented at the conference. We are grateful to NSFC for financial support.

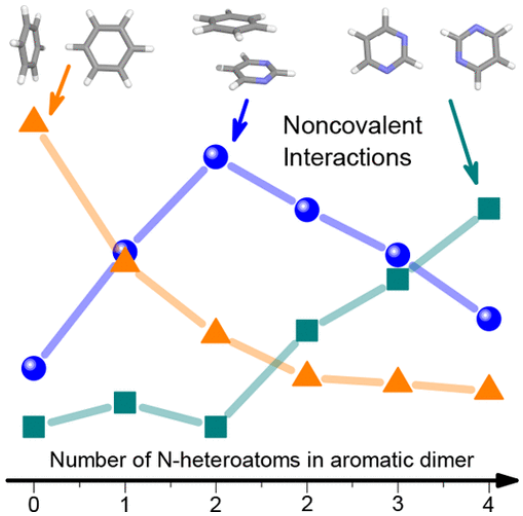

Figure 1. Measured structural preferences of the aromatic dimers.

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An insight into attosecond photoionization

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The discovery and development of attosecond light sources has opened the possibility to access experimentally the time scales on which electronic dynamics occur during photoionization. The photoemission time delay [1,2], which is today well understood as a dispersion dynamic of electron wave-packets in a potential well, has attracted much attention using the RABBIT technique [3]. In recent years it was possible to study individual ionization scattering channels [4,5] and to discuss, among other things, effects induced by the short-range potential. Here, we review our understanding of the two-photon photoionization process central to the RABBIT technique, and present key studies carried out in Lund.

Our measurements combine an XUV attosecond pulse train with a weak IR pulse. The resulting photoelectron spectrum is composed of a comb of odd harmonics originating from the ionization by the XUV pulse train with sidebands between the harmonic peaks which arise from two-photon transitions (XUV±IR). The kinetic energy of the emitted electrons is analyzed with a magnetic bottle spectrometer or a Velocity Map Imaging spectrometer (VMI). As the delay between the IR and XUV pulses is varied the amplitude of these sidebands oscillates with a phase equal to the phase difference between the two paths leading to the sideband, and thereby providing temporal information about the electronic wave-packet group delay.

Supported by theoretical calculations, we will show that the group delay of electronic wave-packet contains information from both light-matter interaction and photoionization. We will discuss how the use of time-frequency approach is essential to disentangle various ionization processes [6,7] and how to uncover the detail mechanisms upon ionization using angle-resolved measurement [4].

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Ultrafast preparation and detection of entangled atoms using a COLTRIMS reaction microscope

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We report on experiments with highly intense femtosecond laser pulses with tailored polarization to study entanglement of spatially separated atoms on femtosecond time scales.

Figure 1. Illustration of a circularly polarized laser pulse (orange) that selectively liberates electrons with a magnetic quantum number of $m = -1$ (blue) compared to electrons with $m = +1$ (red). Here, in a semi-classical picture, the $m = -1$ electrons are counter-rotating with respect to the electric field of the circularly polarized laser pulse.

Previously, it has been shown that circularly polarized light favors electrons with a certain magnetic quantum number in strong field ionization [1]. This preference is illustrated in Fig. 1 and was used to prepare and detect ring currents in single argon ions [2,3].

Building on these insights, we use a pump-probe scheme to prepare and detect ring currents in dissociating oxygen molecules. The laser pulses have intensities on the order of 10^{14} W/cm^2.

The pump pulse excites a ring current in molecular oxygen and simultaneously triggers the dissociation of the molecule into two spatially separated entangled oxygen atoms (see Fig. 2). The probe pulse allows us to investigate this pair of atoms on femtosecond time scales. We find that the valence electrons of the two atoms are entangled in their magnetic qunatum number [5].

The momenta of the liberated electrons and the ions are measured in coincidence using cold-target recoil-ion momentum spectroscopy (COLTRIMS) reaction microscopes [4].

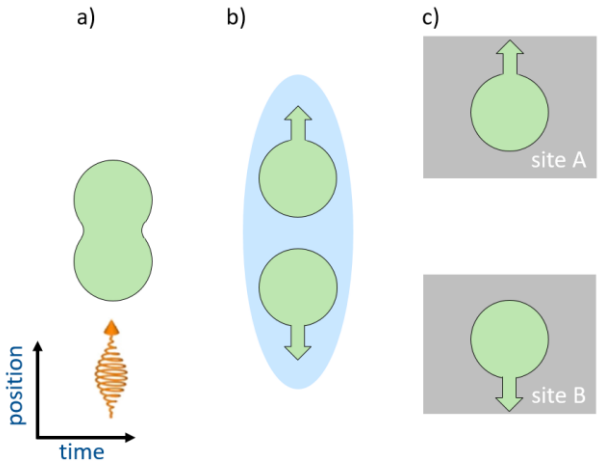

Figure 2. Illustration of the preparation of spatially separated atoms on femtosecond time scales. (a) The pump pulse with a defined helicity excites an oxygen molecule in its ground state by three-photon absorption. (b) The dissociating molecule possesses an electronic ring current with a sign that depends on the helicity of the pump pulse. (c) After a pump-probe time delay of 1.5 ps the two ground state oxygen atoms are separated by 15 nm and are investigated with the probe pulse.

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Ultrafast electron transfer in donor-acceptor molecules tracked by attosecond spectroscopy

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Photoinduced electron transfer (ET) and charge transfer (CT) are fundamental phenomena in physics, chemistry, and material science. A deeper understanding of these processes requires real-time tracking of electron and nuclear motion on the few-femtosecond time scales [1]. In this work, we investigate the electron transfer dynamics following prompt photoionization in nitroanilines, the simplest donor-acceptor systems, by XUV-near infrared (NIR) pump-probe attosecond spectroscopy, in combination with photo-electron photo-ion coincidence energy-resolved (PEPICO) measurements and advanced many-body quantum chemistry calculations. We explore three molecules with different functionalization of the push-pull groups: 4-nitroaniline; 4-NA; 3-nitroaniline; 3-NA; N,N-Dimethyl-4-nitroaniline, nd-NA.

Following XUV photo-ionization the molecule dissociates into several fragments. In Fig.1, we report the mass spectrum for the fragment NO^+: two side peaks are observed, with high kinetic energy release broadening (KE+ and KE-), which are a fingerprint of molecular dication dissociation by Coulomb explosion (CE) [2], as corroborated by synchrotron measurements. These shoulders display an ultrafast transient signal as function of the pump-probe delay, which is reported in Fig. 1c). A fit of the data reveals a rise time of ~10 fs and a relaxation decay with a time constant of 22 fs (4-NA), 23 fs (3-NA) and 28 fs (nd-NA).

We assess the origin of these timings through numerical simulations, using semi-classical trajectories at the CASSCF level of theory, performing few-switches trajectory surface hopping calculations as implemented in the SHARC method [3]. Simulations show that the molecule undergoes an ultrafast planarization of the amino group NH_2, which favors a CT from the lone pair of NH_2 toward the C-N bond in 10 fs.

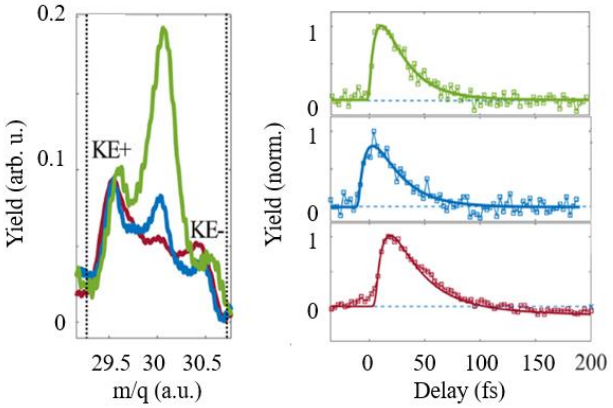

Figure 1. Mass spectra of the molecules after ionization by the XUV radiation in the region m/q=30 (left) and relative yield of the high kinetic energy NO^+ fragment as a function of the pump-probe delay for 4-NA (green), 3-NA (blue) and nd-NA (red).

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Peptide bonds formation in clusters of amino acids induced by ionizing radiation

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When ionizing radiation interacts with molecules or clusters in the gas phase, it induces their ionization and excitation. This phenomenon is observable regardless the type of ionizing radiation used, including collisions with multiply charged ions, energetic synchrotron radiation, or intense laser fields. The direct consequence of this interaction is the formation of molecular species with high degrees of excitation and ionization, leading to fragmentation into two or more positively charged moieties which repeal each other, the so-called Coulomb explosion.

However, alongside this process, a variety of other phenomena occur in competition; these include intramolecular charge transfer [1,2], hydrogen migration [3-7], roaming of a neutral fragment [8] or even intermolecular reactivity in a cluster forming larger species [9-12]. These reactions are triggered by ionization and excitation and can only be observed if they occur within the femtosecond timescale characteristic of the Coulomb explosion.

In this communication I will present a detailed study of the dynamics of these unexpected processes. Specifically, I will show how, through the integration of state-of-the-art experiments, where ions are detected in coincidence, with simulations based on quantum chemistry, we can infer a comprehensive understanding of such reactions and track their dynamics, thereby unambiguously characterizing competing channels. Among all the processes observed special attention will be paid to the formation of peptide bonds in clusters of amino acids after interaction with ionizing radiation.

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Directly imaging excited state-resolved transient structures of water

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Capturing the transient position of nuclei with sub-Angstrom and femtosecond spatiotemporal resolution for a specific electronic state can visualise the ultrafast structural dynamics of polyatomic molecules, which is a key step towards a better understanding and controlling of the chemical reaction. The CEI was developed decades ago to study the molecular structure by ionising the target to a highly charged state upon various radiation sources. The molecular structure prior to Coulomb explosion (CE) can be reconstructed from the momentum information of all the fragments in the molecular frame of reference. Benefited from the ion momentum imaging technique, CEI has equal sensitivity for the probing of light and heavy atoms.

However, retrieval the accurate electronic-state resolved structure of molecules with CEI is still elusive. A direct comparison study of the CEI induced by the ionisation of intense laser field and electron impact can provide solid experimental evidence to reveal the electronic state-resolved ultrafast nuclear relaxation dynamics of polyatomic molecules.

By developing the laser-induced electron recollision-assisted Coulomb explosion imaging approach and molecular dynamics simulations, snapshots of the vibrational wave-packets of the excited (A) and ground states (X) of D_2O are captured simultaneously with sub-10 picometer and few-femtosecond spatiotemporal precision.

We visualised that bond length and angle are significantly increased by around 50° and 10 pm, respectively, within 8 fs after initial ionisation for the A state of cation, and the R_{OD} further extends 9 pm within 2 fs along the ground state of the dication.

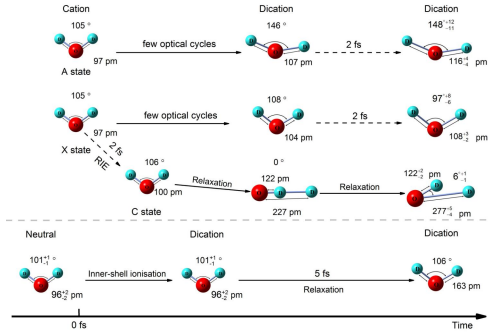

Figure 1. The summary of the time-resolved structural evolutions for different electronic states.

Moreover, the ultrafast nuclear relaxation along the autoionisation state of dication has been studied, where the R_{OD} can stretch more than 50 pm within 5 fs after double inner-shell ionisation induced by electron impact. These results provide comprehensive structural information for studying the fascinating molecular dynamics of water, and show rich electronic states-resolved nuclei dynamics triggered by the inner-shell and valence level ionisation. Our studies pave the way towards to make a movie of excited state-resolved ultrafast molecular dynamics and light-induced chemical reaction.

Calculation of molecular ionization cross sections using analytical Gaussian integrals

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The description of electronic continuum states plays a major role in the theoretical study of molecular ionization processes. It is well-known that Gaussian basis sets, thanks to their interesting mathematical properties, lead to very efficient calculations of bound molecular states. However, being quite local and monotonic, these basis sets have been - at best - considered in the literature as a second-class for describing long-range oscillating wavefunctions, such as those appearing in the context of fragmentation processes.

Over the past few years, we have developed optimized Gaussian-type orbitals with complex exponents (cGTOs), allowing for numerically reliable representations of the radial wavefunction of the outgoing electron in molecular ionization [1,2]. The originality of the method stands in the way we calculate the cross-sections. Indeed, using cGTO expansions for the continuum states allows us to derive closed-form expressions for all those integrals appearing in transition matrix elements. The initial molecular state can be either described by Slater type orbitals or Gaussian type orbitals. We have successfully applied this approach to the calculation of differential cross-sections for molecular ionization by photon or electron impact, in the case of highly symmetrical molecules (ammonia, water and methane) where monocentric approximations are sufficient [1,3]. As an example, Fig. 1 shows triple differential cross-section (TDCS) calculations for the electron- impact ionization (e,2e) of methane.

We are presently making progress toward a generalization of the approach to a multicentric description, stimulated by the fact that all the interesting mathematical properties of the Gaussian integrals, well-known in the case of real GTO, can be adapted to cGTOs [5].

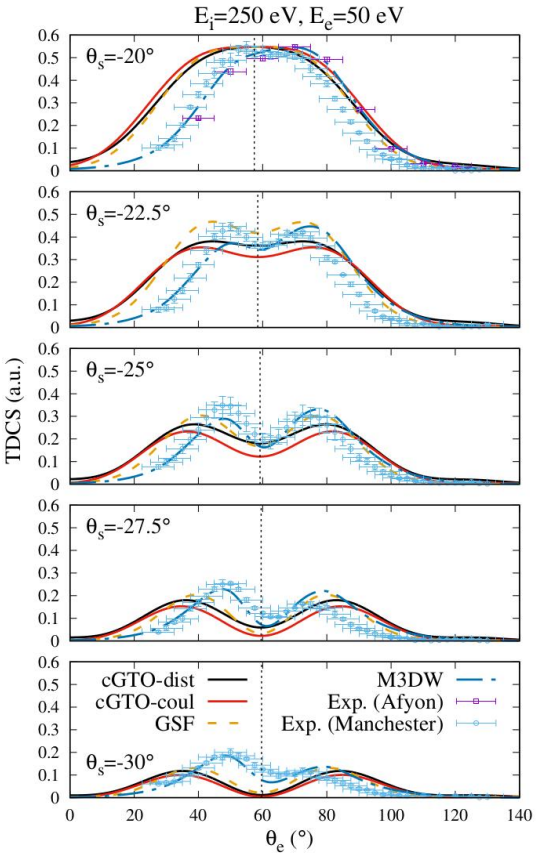

Figure 1. TDCS for ionization by electronic impact of the outer (1t2) valence orbital of CH_4, as a function of the ejection angle θ_e, for different values of the scattering angle θ_s with kinematical parameters of ref. [4]. The ejected electron is described either by a Coulomb wave (cGTO-coul) or a distorted wave (cGTO-dist). Results are compared with other theoretical results and experimental points [4].

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Heavy-ion-induced intermolecular Coulombic decay in hydrated biomolecule systems

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Heavy-ion-irradiation can cause severe damage to cell and DNA. Acting on cancer cells, heavy ion therapy servers as an efficient radiotherapy technique due to the ability of carrying a large amount of energy and accurate delivering these energies to the tumor area by controlling the depth of the so-called Bragg Peak, and the high biological effectiveness of killing cancer cells. Despite its importance in both fundamental research and clinical applications, the mechanistic details of the heavy ion irradiation are unclear.

A new reaction microscope is built up in the Heavy Ion Research Facility in Lanzhou (HIRFL), as shown in Figure 1. The supersonic target of this setup can provide not only atomic and molecular jets, but also the target of mixed clusters, providing the opportunity for investigation of fragmentation dynamics of biological relevant molecules and clusters. We have chosen pyrimidine solvated by a few water molecules (Pyr-nH_2O, n = 1-3) as the model system, and investigated the relaxation mechanisms after irradiated by heavy ion beam.

In our study, a harmful relaxation cascade that can occur in the DNA vicinity, and contribute to high biological effectiveness of heavy-ion-irradiation was identified. Ionization of a water molecule in its inner-valance shell initiates intermolecular Coulombic decay (ICD) [1], leading to dramatically enhanced production of harmful low energy electrons. As a consequence of ICD, intermolecular proton transfer [2] takes place between water molecules, producing a protonated water cluster and a reactive hydroxyl radical OH. More importantly, we found that the intensity of such relaxation process increases dramatically as the number of solvated water increasing, demonstrating that the biological damage of ICD is much more serve for the living tissues in aqueous environment compared to that have been widely investigated in biomolecules.

Details of this study will be presented in the conference.

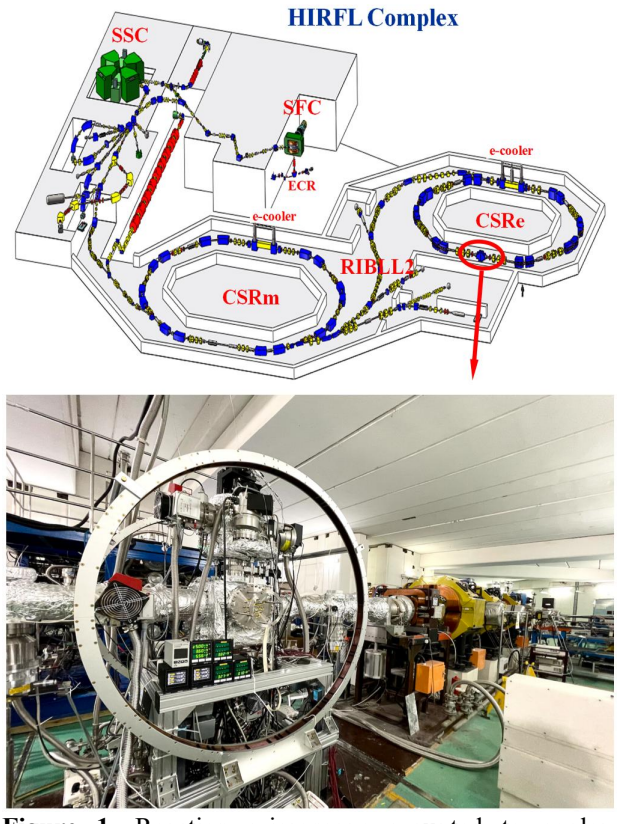

Figure 1. Reaction microscope mounted to cooler-storage ring of Heavy Ion Research Facility in Lanzhou.

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Application of electron-atom Compton scattering to measurements of individual atomic motions in polyatomic molecules

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Intramolecular atomic motion has long been attracting a great deal of interest, as it often governs the reactivity and functionality of the molecule in question. To observe such atomic motion directly, atomic momentum spectroscopy (AMS) or an electron-atom Compton scattering experiment was proposed in 2001 [1]. This is because the double differential cross section (DDCS) for the AMS scattering contains, within the plane-wave impulse approximation (PWIA), the momentum distribution ρ of each constituent atom i of a target molecule:

$$\frac{d\sigma^2}{dEd\Omega} \propto \sum_{i:atom} (Z_i)^2 \frac{M_i}{q} J_i(\boldsymbol{P}_i), \quad (1)$$

$$J_i(\boldsymbol{P}_i) = \int \rho(\boldsymbol{P}_i)\delta\left(E - \frac{q^2}{2M_i} - \frac{\boldsymbol{P}_i \cdot \boldsymbol{q}}{M_i}\right) d\boldsymbol{P}_i . \quad (2)$$

Here $\boldsymbol{q}$, Z_i and M_i are the momentum transfer and the nuclear charge and mass of the scattering atom i, respectively. $\boldsymbol{P}_i$ is the initial momentum of the scattering atom. Indeed, the unique ability of AMS has recently been embodied by a series of attempts. They involve developments of a highly sensitive apparatus [2], a quantum chemistry-based AMS theory for diatomic molecules [3] and a data-analysis protocol to disentangle the experimental response of the intramolecular atomic motion from the instrumental response function [3-5], as well as elucidation of the range of the validity of PWIA [4]. However, application of AMS is still limited to diatomic molecules. The reason behind this situation is the lack of a quantum chemistry-based AMS theory for polyatomic molecules.

The above-mentioned circumstances may be due to the difficulties in decomposing the atomic momenta of molecular vibrational and rotational motions into the momentum of each constituent atom. We have found an idea to overcome the difficulties and eventually obtained a quantum chemistry-based AMS theory for polyatomic molecules. Furthermore, the accuracy of the theory has been tested and confirmed by comparison with AMS experiments on the CH_4 molecule, conducted at a scattering angle of 135° and at an incident electron energy of 2.0 keV [6].

As an example of the results, an experimental electron energy loss (EEL) spectrum of CH_4 is presented in Figure 1 (a). It can be seen that two bands appear at around 0.3 and 3.7 eV, which are assigned to those of the C and H atoms, respectively. The momentum distribution due to the intramolecular motion of the H atoms was subsequently obtained and the result is presented in Figure 1 (b), together with associated distribution calculated by using the AMS theory developed here. The good agreement between experiment and theory evidently demonstrates the accuracy of the theory as well as the experimental ability of AMS to map intramolecular atomic motion for polyatomic molecules. In the talk, we will discuss details of the present work and future possibilities of AMS involving our currently running projects.

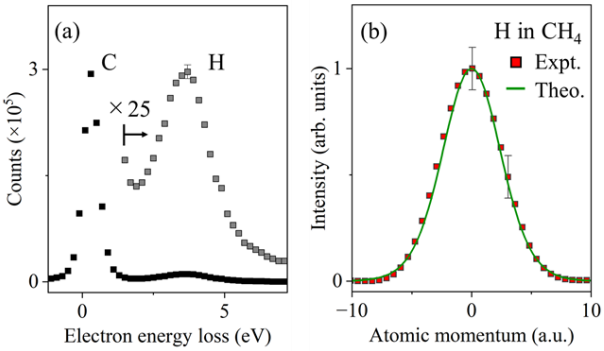

Figure 1. (a) An electron energy loss spectrum of CH_4. (b) A momentum distribution due to intramolecular atomic motion of the H atom in CH_4.

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Single ionization of helium by high-energy protons using the parabolic quasi-Sturmian approach

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We investigate theoretically ion-atom ionizing collisions. For this purpose, we have developed a parabolic quasi-Sturmian approach [1] and applied it to the single ionization of helium by intermediate and high energy protons. The aim is to further understand this fundamental process by looking at fully differential cross sections (FDCS) for which measurements are available (on a relative scale for 1 MeV protons, and on an absolute scale for 75 keV protons).

In the framework of our approach [1], the proton-electron interaction is treated as a perturbation, and the transition amplitude is extracted directly from the asymptotic behavior of the solution of an inhomogeneous Schrödinger equation for the Coulomb three-body system $(e^-, \mathrm{He}^+, p^+)$. The driven equation is solved numerically by expanding in convolutions of the parabolic quasi-Sturmians [1] for the two-body (p^+, He^+) and (e^-, He^+) systems. We call these basis functions *convoluted* quasi-Sturmians (CQS).

Our calculated FDCSs converge pretty fast as the number of terms in the expansions is increased, and are in overall reasonable agreement with other theoretical results. At the same time, there are significant discrepancies between theoretical predictions for FDCSs and experimental data. For example, for 1 MeV protons, even well-established theories fail to give a proper account of the experimentally observed binary peak position. The situation gets worse for lower proton energies (see, e.g., [2]). Our CQS results for the 75-keV protons FDCSs (without the use of the pe-interaction cutoff proposed in [3]) are presented in Fig. 1. For comparison, we show the results of calculations [3] within the 3C model, which correctly takes into account the Coulomb wave function asymptotic behavior.

Thus, for 1 MeV incident protons, the CQS approach yields angular distributions in agreement with other theoretical calculations and in satisfactory agreement with relative scale experimental data. At lower incident energies, our FDCS results are reasonable as far as the absolute scale is concerned, but important shape discrepancies are present indicating that our description is incomplete. We are considering improving our model by including the electron-capture channel.

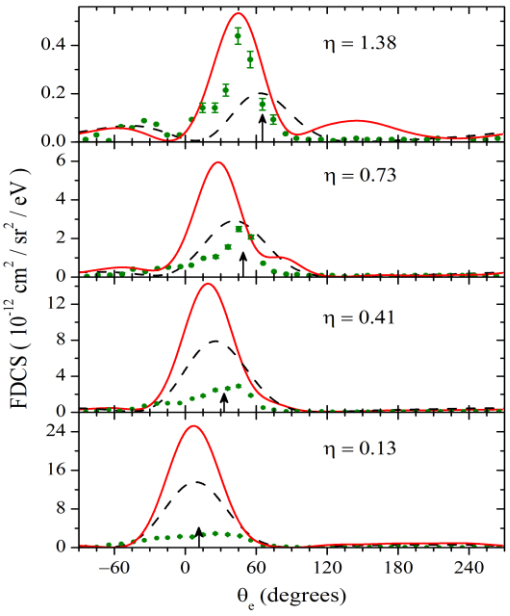

Figure 1. Our calculated FDCSs (solid line) are compared to experimental data [2] for single ionization of helium by 75 keV protons in the collision plane, for different transverse momenta η. The ejected electron energy is $E_e = 5.4$ eV. Dashed lines represent 3C calculations [3].

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Exploring ultrafast dynamics with advanced electron-ion coincidence methods under soft and hard x-ray excitations

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Photoexcitation of molecules by soft X-rays creates core holes with lifetimes of a few femtoseconds, which may trigger considerable dynamics for light nuclei. Ultrafast dissociation (UFD) is an extreme example, when dissociation of intermediate states occurs before electronic decay. [1] High-resolution electron spectroscopy can be used to reveal signatures of UFD in Auger decay spectra.

Alongside high-resolution photoelectron, Auger and photon emission spectroscopies, research on the photoionization and photoexcitation dynamics of isolated species often employs coincidence techniques, which involve simultaneous detection of multiple charged particles originating from the same ionization event. Coincidence techniques contribute to a more complete understanding of the dynamics of photon-molecule interactions and the resulting decay pathways. Electron-ion coincidence methods have been used extensively over the last two decades, primarily in the soft X-ray range. The most recent state-of-the-art electron-ion coincidence setup, GPES, permanently installed at the soft X-ray FinEstBeAMS beamline of the MAXIV synchrotron radiation facility in Sweden, employs hemispherical electron analyzer coupled with 3D ion momentum imaging spectrometer. [2] It allows measurements with vibrational resolutions. Moreover, correlation of kinetic energies of Auger electrons with kinetic energy release of the ions allows describing partitioning of the excess energy between internal degrees of freedom in dissociating molecular fragments. [3-4]

Momentum vector correlation allows imaging the nuclear dynamics associated with molecular dissociation. One example employing cold target recoil-ion momentum spectroscopy (COLTRIMS) measurements, performed at BESSY II in Germany, demonstrates a dissociation pathway during UFD process, where light linkages fling in a catapult-like manner leaving heavy fragments behind.

Hard X-ray photons (>1 keV) may reach deeper-lying core electrons. The lifetime of deep-core-hole states is of the order of 1 fs or below and their decay occurs via a series of subsequent relaxation steps. In general, intermediate states with one or multiple holes in the shallower-core electronic shells are generated in the course of such cascades, which undergo UFD processes. [5-6]

Very recently, we have developed a new coincidence setup MUSTACHE, which will allow us implementing high-resolution advanced coincidence techniques into the studies of nuclear dynamics processes induced by X-rays. MUSTACHE setup, which was developed for fast Auger-electron - ion coincidence measurements at the GLAXIES beamline of the synchrotron SOLEIL in France, is unique allowing studies in the hard x-ray regime for electrons with kinetic energies above 1 keV. Note, that neither magnetic bottle nor COLTRIMS coincidence setups can detect electrons of high energies (above ca. 500 eV) with reasonable efficiency and energy resolution. MUSTACHE combines 3D ion momentum imaging from COLTRIMS setups with high-resolution high-energy photoelectron spectroscopy akin to HAXPES techniques.

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Post Collision Interaction, Photoelectron recapture and Angular Momentum transfer in the hard X-ray domain. From the isolated Atoms and Molecules to condensed phase

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Inner-shell photoionization just above the inner shell ionization threshold leads to the emission of a slow photoelectron and a fast Auger electron. Post Collision Interaction is a well-established process leading to energy exchange between the photoelectron and the Auger electron. Photoelectron kinetic energy is diminished by the same amount of kinetic energy gained by the Auger electron. It can be theoretically described by taking into account that the photoelectron is first leaving a singly charged and later a doubly charged ion when it is overtaken by the Auger electron. The photoelectron is then feeling a higher Coulombic field from a doubly charged ion meaning that the photoelectron kinetic energy is decreased. When the Auger electron overtakes the photoelectron, because of the screening by the photoelectron, it feels the Coulombic field from a singly charged instead of a doubly charged ion, meaning it is accelerated.

Because of the short core-hole lifetime of deep inner-shell and of the high kinetic energy of the Auger electrons, it turns out that the PCI effect in the HAXPES regime is very strong [1] leading eventually to the recapture of the photoelectron [2]. We have recently performed PCI studies in condensed phase [3] demonstrating that the scattering of the photoelectron and the polarization screening are playing an important role in PCI in condensed phase. Angular momentum transfer between the photoelectron and the Auger electron has been observed for the first time on argon [4] leading to perturbed photoelectron angular distribution.

Recent studies on comparison of PCI effect on 2 step relaxation after 1s ionization of krypton (Kα decay followed by LMN) and L shell ionization have been recently published [5-6]. I will present new results on PCI on krypton measured at APS (Argonne, US) and at PETRA III (Hamburg, Germany). 2D map of PCI distortion have been measured at both thresholds. These results allow to underline very different recapture processes of the photoelectron in both cases. I will also present results obtained au European XFEL on PCI on Double Core Hole states and recent results measured at SOLEIL.

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Transient core-to-core resonances of Kr in intense EUV-FEL fields studied by electron-ion coincidence spectroscopy

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Understanding nonlinear optical responses of atoms and molecules is crucial for deepening our knowledge of the fundamental physical and chemical processes of matter in intense laser fields. Recent advances in free electron laser (FEL) technology in the extreme ultraviolet (EUV) and X-ray regions have made it possible to investigate nonlinear phenomena induced by light with high-frequencies. Numerous studies have revealed that multiple multiphoton ionization is a typical nonlinear response in these wavelength region [1]. In the multiphoton processes, it has been found that nonlinear optical response involving inner-shell electrons can open new resonant channels which are inaccessible in neutral states. Such "hidden" resonances associated with inner-shell electrons have been unveiled for atomic transitions including 1s-2p core-to-valence transition of Ne^+ [2] and Rydberg transition from $3l$ (l = s, p and d) states of multiple charged Xe ions [3]. However, resonant transitions between inner-shell states are yet to be clearly identified. Herein, we present the first observation of core-to-core resonance of the Kr^+ $3d^{-1}$-$3p^{-1}$ transitions following the initial 3d photoionization (Fig.1(a)) by identifying Auger electron signals via the 3p core hole decays using electron-ion coincidence technique [4].

All the experiments were carried out at the soft X-ray beamline (BL1) of RIKEN SACLA, Japan. Ultrashort EUV-FEL pulses (~120 eV, 30 fs) were focused by KB mirrors to an effusive Kr gas introduced in a magnetic bottle type electron-ion spectrometer. Electrons generated in the interaction region were guided to a time-of-flight (TOF) tube by inhomogeneous magnetic fields from a cone-shaped permanent magnet and homogeneous magnetic fields by a solenoid coil, and detected with a microchannel plate (MCP) detector placed in the other side of the TOF tube. Counterpart ions were extracted into the TOF tube by an application of pulsed voltages to ion-collection electrodes 1 μs after the laser irradiation, and detected with the same MCP detector. In these measurements, a retardation voltage of −57 V was applied to improve the energy resolution of 3p Auger electron peaks.

A TOF mass spectrum of Kr in intense EUV-FEL (120.5 eV, 1.2×10^{14} W/cm^2) shows highly charged Kr^{z+} ions (z = 1-4) where the Kr^{4+} ion formation requires at least two-photon absorption. An electron spectrum observed in coincidence with Kr^{3+} ions (Fig. 1(b)) exhibits two prominent peaks at 78 and 92 eV in addition to a valence photoelectron peak (Kr^+ $4p^{-1}$ at 107 eV. The relative intensities and energy positions of the two peaks are well reproduced by stick spectra of the $3p_{3/2}$ Auger peaks associated with the $3d^{-1}4s^{-1}$ and $3d^{-1}4p^{-1}$ states. Since the 3p photoionization cannot occur by one-photon process with $h\nu$ = 120.5 eV, the 3p core-hole formation is interpreted due to a transition from the $3p^63d^9$ to the $3p^53d^{10}$ states by the second photoabsorption following the initial 3d photoionization. The 3p Auger electron signals disappeared when the photon energy was set to 124.5 eV, showing the resonant character of the $3d_{5/2}{}^{-1}$-$3p_{3/2}{}^{-1}$ transition (energy difference: 120.6 eV). Since the core-to-core resonant transition is not specific to Kr, the present study clearly shows that this type of hidden resonance is important for understanding nonlinear optical responses of matter in the high-frequency regions.

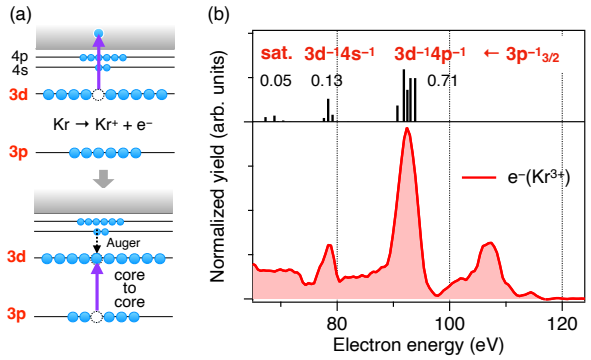

Figure 1. (a) Schematics of core-to-core transition of Kr. (b) Electron spectrum in coincidence with Kr^{3+} ions and stick spectra of $3p_{3/2}$ Auger electrons.

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Differential ionisation in intermediate-energy proton-helium collisions

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Modelling differential ionisation for ion-atom collisions remains a significant challenge. This is especially true for intermediate-energy collisions, where two-centre effects need to be accounted for. Experimental data for singly differential cross sections (SDCS) and doubly differential cross sections (DDCS) have been available for decades, however, theoretical methods have been applied at only select energy and angular regions and agreement is inconsistent. The challenge for theory lies in the accurate representation of the continuum.

We investigate single ionisation for the simplest four-body system, proton collisions with helium atoms, using the wave-packet convergent close-coupling (WP-CCC) method. In this approach, the total scattering wave function is expanded in terms of both target- and projectile-centred basis states. Each basis consists of a number of bound eigenstates and a continuum discretised using wave-packet pseudostates. The two-centre expansion allows us to differentiate between direct ionisation of the target and electron capture into the continuum of the projectile. To describe the helium target, a correlated two-electron wave function is used. Previously, we have calculated the integrated ionisation cross section [1]. The results displayed excellent agreement with experimental data.

Calculations of the singly differential cross sections (SDCS) differential in the electron ejection energy, electron ejection angle, and scattered projectile angle [2], and the doubly differential cross sections (DDCS) differential in the ejected electron energy and angle [3], and the ejected electron energy and scattered projectile angle [4] were performed at intermediate energies (50–300 keV). Excellent agreement between the WP-CCC results and experimental measurements was found for both SDCS and DDCS. The WP-CCC method is the first theory to accurately describe the electron emission spectra for the proton-helium system at both a singly and doubly differential level across the intermediate-energy range investigated. Moreover, this study represents the first calculation of differential ionisation in a non-perturbative manner.

We have now turned our attention to the study of the fully differential cross section (FDCS) in particular kinematic regimes such as electron ejection in the narrow forward direction and in coplanar geometry. This presents a significant challenge as the calculation becomes very complicated and computationally time-consuming. However, preliminary results are encouraging. A kinematically complete solution of the problem should help deepen the existing understanding of the dynamics governing ionisation in proton collisions with helium.

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An efficient method of accounting polarization in sequential processes

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An intense vacuum ultraviolet radiation (VUV) generated by the modern Free-Electron Lasers (FELs) possesses an unique combination of parameters: intensity, coherency, duration and polarization. The last is often and unfairly overlooked in the consideration of complex evolution of a sample, when there are a number of competitive processes such as photoionization, Auger-decay and fluorescence. The major obstacle is that direct accounting of the polarization implies enormously increasing number of equations to be solved. For example, in the ionization of a noble gas from a valence shell within LS-coupling scheme there are two channels $np^5\,{}^2Pks\,{}^1P$ and $np^5\,{}^2Pkd\,{}^1P$ to be treated in "unpolarized" case, but there are twelve allowed combinations of ionic M_f and electron' m magnetic quantum number in "polarized" case. To overcome the obstacle one may apply some symmetry properties, for example, to postulate that for linearly polarized radiation there is no difference between positive and negative values of the magnetic quantum numbers.

Here we propose a new method [1] based on solution of the analogue of a conventional system of rate equations formulated in terms of statistical tensors [2]. The approach accounts polarization of both radiation and intermediate ionic states, and suits to consideration of evolution of an atom irradiated by linearly or circularly polarized field involving excitation, ionization and relaxation (Auger and fluorescence) processes.

The illustrative calculations were performed for sequential multiple ionization of krypton, for photon energy below 3d excitation (90 eV) and for the energy corresponding resonance excitation of $3d^9 4s^2 4p^6 5p[3/2]_1$ resonance (91.2 eV). The calculation manifests essential redistribution of ionic yields and re-arrangement of photoelectron spectrum depending on polarization (see fig.1).

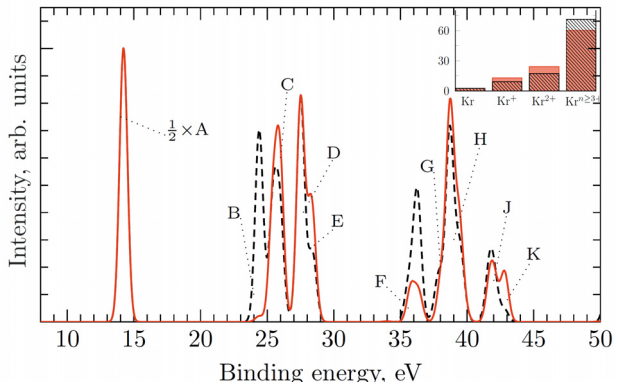

Figure 1. Photoelectron spectrum in three-photon triple ionization of Kr by linearly polarized (solid line) and unpolarized (dashed line) radiation. In the insertion there are ionic yields for polarized (colored) and unpolarized (shaded) cases.

In figure 1 the photoelectron spectrum simulated for the frequency ω=78 eV, fluence $F=10^3$ ph/Å^2 for linearly polarized and unpolarized radiation is presented. The photon energy is chosen near the Cooper minimum where the alignment of Kr^+ tends to the edge value -1.4. One can see that polarization may suppress or even completely smear out some particular line in photoelectron spectrum (lines B and F in fig. 1).

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Relation between photoionization cross sections and attosecond time delays

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We derive a Kramers-Kronig-like relation between photoionization cross sections and Wigner-like time delays and demonstrate its validity for (anti)resonances. This new concept defines a topological analysis and thereby sheds new light on a long-standing controversy regarding the sign of the photoionization delay near the Ar 3s Cooper minimum (CM) [1].

We show that for a Fano resonance:

$$\sigma_{\mathrm{F}}(\epsilon) = \sigma_{\mathrm{a}} \frac{(\epsilon+q)^2}{\epsilon^2+1} + \sigma_{\mathrm{b}},$$

where $\epsilon = (E - E_{\mathrm{r}})/(\Gamma/2)$ is the relative energy, the time delay can be expressed by:

$$\tau_{\mp}(E) = \frac{2}{\Gamma}\left\{\mp \frac{1/\gamma}{[(\epsilon+Q)/\gamma]^2+1} + \frac{1}{\epsilon^2+1}\right\},$$

where $r = \sigma_{\mathrm{b}}/\sigma_{\mathrm{a}}$, $Q = q/(r+1)$, and $\gamma = \sqrt{r(r+q^2+1)}/(r+1)$. The sign of the first term in the parathesis depends on the winding number (WN) of the trajectory of the transition amplitude in the complex plane about the origin: τ_- for WN = 0 (not encircling the origin) and τ_+ for WN = 1 (encircling the origin).

The Ar 3p CM is a typical case of WN = 0, and the time delay from the Fano model agrees well with the RPAE calculation and the photorecombination experiment [3], as shown in **Fig. 1**. Conversely, for the Ar 3s CM, RPAE gives the trajectory with WN = 1, whose delay also coincides with the Fano model of WN = 1. However, the RABBIT experimental result [4] shows better agreement with the Fano model of WN = 0, which may originate from the small couplings that shift the trajectory subtly and the WN flips from 1 to 0. This demonstrates that the topological properties of transition amplitudes are sensitive and play essential roles in attosecond time delays.

In the general case, when the trajectory does not encircle the origin on the complex plane, the following relation holds:

$$\tau(E) = \frac{1}{2}\mathcal{H}\left\{\frac{1}{\sigma(E)}\frac{\partial\sigma(E)}{\partial E}\right\},$$

where $\mathcal{H}$ is the Hilbert transform. Our work bridges traditional photoionization spectroscopy with attosecond chronoscopy.

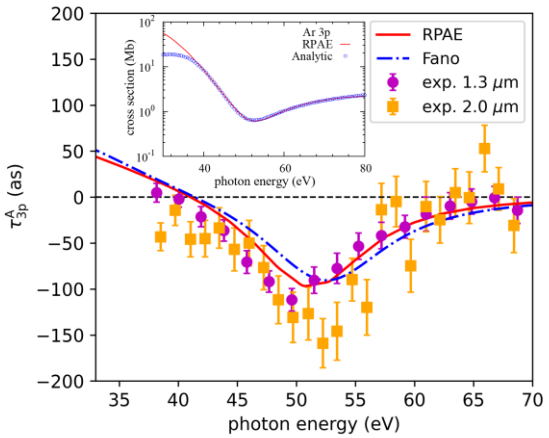

Figure 1. Fano-modelled time delay for Ar 3p CM (WN = 0), compared to RPAE calculation [2] and photorecombination experiment [3] at various wavelengths. The inset plot compares the computed and modelled cross sections.

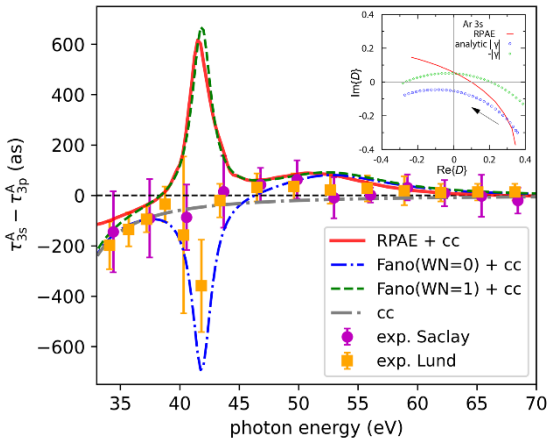

Figure 2. Relative time delay between Ar 3s and Ar 3p near Cooper minima, with Fano model of WN = 0 and 1, compared to RPAE calculation [2] and RABBIT experiment [4]. The inset plot compares the trajectories on the complex plane.

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Nonlinear spectral dispersion in resonant x-ray scattering for studying potentials and dynamics

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Resonant x-ray scattering made a sufficient contribution to understanding of femtosecond nuclear dynamics. It stands out for a distinctive capability to probe the ultrafast dynamics through the scattering duration control, enabled by energy detuning from x-ray absorption resonances and observed via variation in spectral dispersion laws. Here I show manifestations of these phenomena in our recent works focusing on nuclear dynamics.

An intricate dynamics within the short-lived core-excited states at SK-edge of the SF_6 is unveil in KLL Resonant Auger Scattering (RAS), that showcases diverse dispersion laws among spectral lines, capturing ultrafast molecular dynamics in $S1s^{-1}6t^{1}_{1u}$ core-excited state with the conspicuous absence of dynamics in $S1s^{-1}7t^{1}_{1u}$ state. The 2D spectral map emerges as a versatile tool for studying change in molecular geometry upon the decay. Employing a theoretical model we demonstrate how the characteristics of the spectral dispersion laws (Raman, non-Raman, anti-Raman) mirror the relative gradients of intermediate and final states in RAS (Fig. 1). This approach is considered as a potent method for mapping molecular potentials.

The spectral dispersion law characteristics were applied to uncover dynamical features using vibrationally-resolved RAS spectra of ammonia recorded in coincidence with the NH_2^+ fragment's kinetic energy release (KER), produced during dissociation in the core-excited or final state. The median distribution of KER, derived from the coincidence data, shows three distinct branches as a function of Auger electron kinetic energy. With the help of theoretical simulations, the deviation from the Raman dispersion law was attributed to the redistribution of the available energy to the dissociation energy and excitation of the vibrational degrees of freedom in the fragment. It is shown that for each vibrational line the dispersive behavior is very sensitive to the uncertainty in KER causing the competition between the Raman and Auger dispersions.

In theoretical simulations of a UV-pump x-ray-probe setup (UVX-PP) [2], we show that frequency detuning of the pump UV pulse acts as a camera shutter by regulating the process duration. This two-photon absorption with long overlapping pulses, allowing for high spectral resolution, thereby provides information about ultrafast dynamics of the nuclear wave packet. Case study of the CO shows that the interference of intermediate vibrational states reveals nuclear dynamics details using the propagation duration controlled by the UV detuning, where intersection of dispersive and non-dispersive spectral bands result in destructive interference.

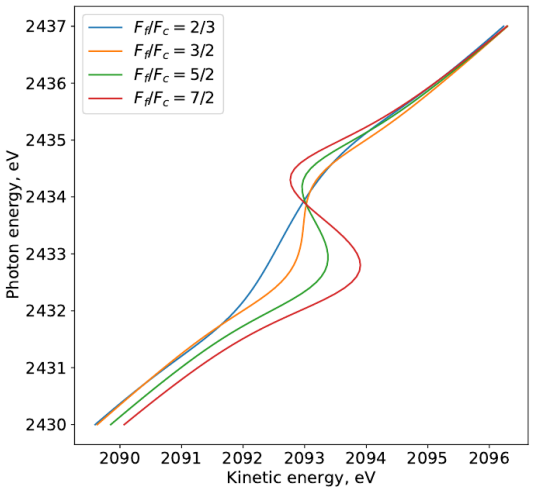

Figure 1. Theoretical modeling for the dispersion law of the RAS (lines follow peak's center of gravity) for several values of relative gradients of a final F_f and core-excited F_c potential energy curves (see legends).

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Resonance Analysis of Strong Field Ionization in Xenon

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Attosecond Transient Absorption Spectroscopy (ATAS) is well suited for the time-resolved investigation of strong-field ionization (SFI) since the all-optical approach probes the system whilst it is being ionized, allowing state-specific view into the intrinsic attosecond dynamics of strong-field initiated electron motion. [1] Using extreme ultraviolet (XUV) spectrometers with high resolution, absorption lines can not only be investigated with respect to their line strength (amplitude) but also their specific shape (phase). The continuous transition from a symmetric Lorentzian to an asymmetric Fano is a result of a change in phase of the underlying dipole [2].

Here, we investigate the attosecond scale strong field ionization of xenon atoms by few-cycle (4.5 fs FWHM) near-infrared (NIR) laser pulses obtained from both experiment and R-matrix with time-dependence (RMT) calculations [3,4]. We look at the build-up of specific 4d core orbital to 5p valence orbital ionic resonances and extract attosecond time-resolved amplitude and phase oscillations of the state-specific XUV dipole response as a function of the NIR intensity.

First, by looking specifically at the oscillations in the dipole phase, we identify an indirect ionization pathway via an XUV core-excited virtual state which competes with the direct ionization pathway. Specifically, we observe delay-dependent NIR-half-cycle oscillations of the line-shape asymmetry whose amplitude decreases with increasing NIR pump intensity. This effect is due to the depletion of the neutral ground state, and hence a weakening of the interfering virtual pathway, which is confirmed by calculating the remaining neutral Xe population.

Second, we consider an alternative approach wherein we consider the amplitude and phase of the dipole response simultaneously by plotting them together in the complex plane. This allows us to analyze the subtle differences between experiment and theory. We may also attribute the complex-plane behavior to interference between different residual ion states populated by the laser field.

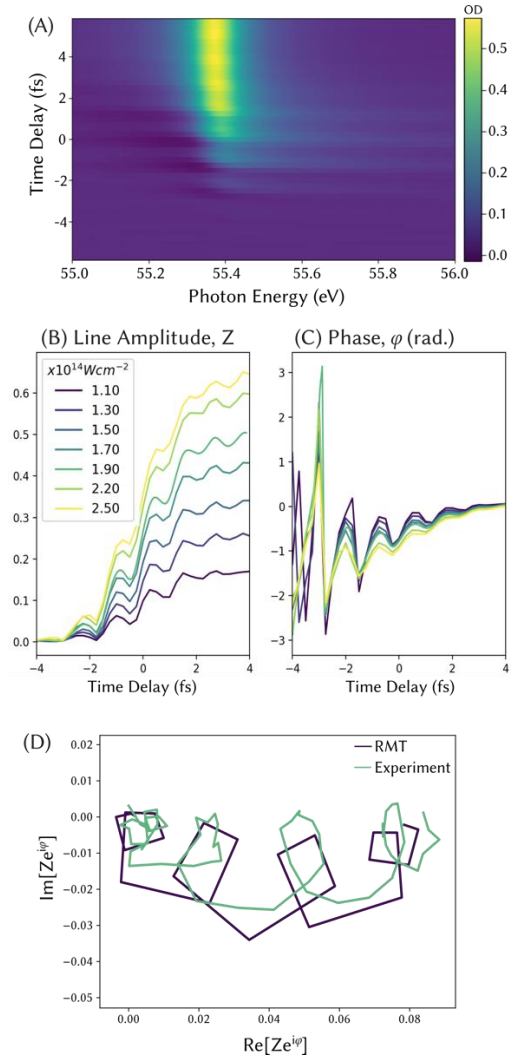

Figure 1. (A) Attosecond build up of the Xe^+ $5p_{3/2}^{-1} \rightarrow 4d_{5/2}^{-1}$ resonant transition during NIR strong-field ionization calculated using the *ab initio* RMT approach, and quantifying (B) the amplitude and (C) the phase of the fitted absorption line shape for different NIR intensities. (D) The amplitude and phase of the fitted absorption line plotted together on the complex plane for both experiment and RMT calculations for the lowest NIR intensity.

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Coherent control of photoabsorption with intense laser pulses: from optically thin to thick media

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Attosecond transient absorption spectroscopy (ATAS) offers state- and element-specific access to resonant transitions in strong laser fields, and has been established as a powerful tool for tracking electrons in atoms and molecules. What ATAS measures is the transmitted laser spectrum, which results from the interference between the incident field and the newly generated one radiated by the induced dipole oscillation. This dipole-emitted field persists on a timescale determined by the dephasing time, and may reinforce or cancel the initial spectrum, giving rise to the modification of spectral profiles.

The interferometric nature of ATAS makes it amenable to coherent control through tailoring the dipole-emitted electric field with external fields. Microscopically, it can be achieved from the single-particle level by implementing laser-induced amplitude and phase modification of the dipole response. When a rather dense medium is employed, it has been revealed that propagation effects can lead to complex spectral reshaping that is substantially different from the dilute-medium expectation

Here we describe two kinds of scenarios for the coherent control of photoabsorption by combining pulse propagation and external laser manipulation. By temporally confining the dipole-emitted field through emptying the population of the excited state after its excitation, we achieve a local enhancement of absorbance in transient absorption spectroscopy [1,2]. By imposing a phase shift to the dipole emission, we demonstrate the transition of absorption profiles from natural Lorentzian to Fano-like, which then become broader with further emergence of new spectral structures, finally turning back to near-Lorentzian lines in optically dense helium, both theoretically and experimentally [3]. The underlying mechanism is revealed and applies broadly to other systems. These results represent a substantial step for the collective interactions of matter with ultrashort laser pulses from understanding to control.

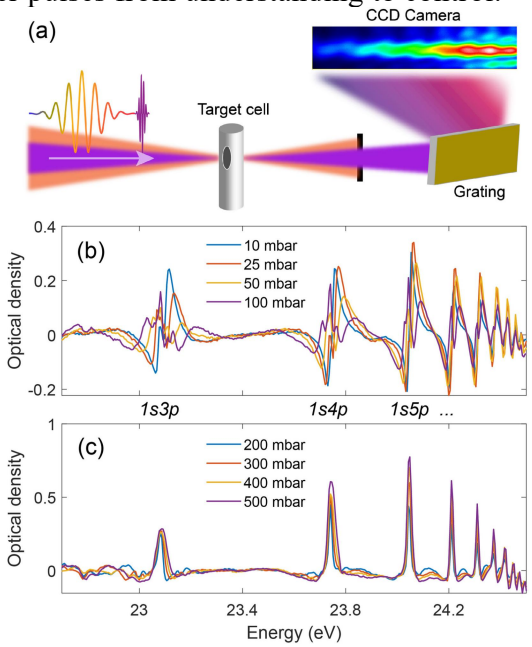

Figure 1. (a) Sketch of the experimental ATAS scheme. (b), (c) Measured absorption profiles for helium as a function of backing-gas pressure.

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Capturing roaming molecular fragments in Formaldehyde

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In conventional dissociation reactions, molecules follow the minimum energy paths from reactants to products. Alternative dissociation pathway called "roaming" was discovered in the photodissociation reaction of formaldehyde (H_2CO) [1]. In the roaming pathway, the roaming fragment (H atom) walks around the remaining HCO and then dissociates as H_2. Since the first observation of roaming in 2004, footprints of roaming have been reported in a wide variety of molecular systems [2-5]. However, roaming fragments themselves have not been observed directly because each molecule follows its own pathway. Recently, we have successfully captured roaming fragments in the dissociation of D_2CO by using a combination of Coulomb explosion imaging and quasi-classical trajectory simulation [6].

Coulomb explosion imaging experiments were performed at Advanced Laser Light Source (INRS-EMT, Canada) on the multi-kHz beam line. Ultraviolet pump pulses (304 nm) were obtained by frequency quadrupling of the output of an optical parametric amplifier. Time-delayed ultrashort intense probe pulses (8 fs) were obtained by pulse compression with a pressure-gradient hollow-core fiber and chirped mirrors. The pump and probe pulses were focused on an effusive molecular beam of D_2CO. The ions generated by the probe pulse were collected with a uniform-electric-field ion momentum imaging spectrometer.

Figure 1 shows the two-dimensional plots of the total kinetic energy release (KER) with the angle between two deuteron momentum vectors for three-body breaking (D^+, D^+, CO^+) channel at different pump-probe time-delays Δt. The onset of molecular dynamics occurred already before 100 fs. This indicates that the S_1/S_0 relaxation occurs five-orders of magnitude faster than that previously assumed [7]. The existence of the fast relaxation process was verified by photoelectron experiments at Nagoya University and *ab initio* calculation. From the simulation including all critical steps (S_1/S_0 relaxation, dynamics on the S_0 state, and Coulomb explosion), we could identify the areas corresponding to roaming, molecular and radical dissociation channels in the two-dimensional plots as shown in the lower right subplot in Fig. 1. The broad angular distribution in the KER range below 5 eV reflects that one deuteron moves freely around the parent molecule in the radical dissociation channel. The limited angular distribution in the 5-12 eV corresponds to weak interaction between D and DCO in the roaming channel. The narrow angular distribution around 150 degrees reflects the strong repulsive force between two deuterons in the molecular dissociation channel. The present results provide a new method for detecting weak statistic dynamics hidden in overwhelming background and could be extended to a vast variety of molecular systems.

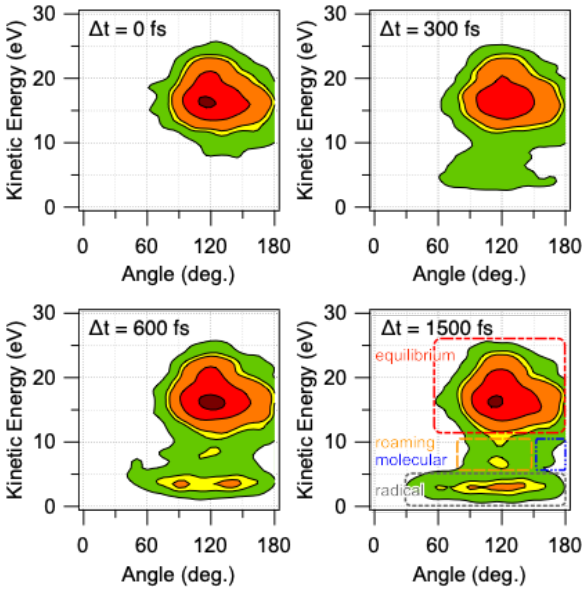

Figure 1. Two-dimensional plots of kinetic energy versus deuteron momentum angle at the pump-probe time-delay Δt. The distribution at Δt = -300 fs was subtracted as background. Indicated in the lower right subplot are the areas corresponding to each reaction pathway identified by using ab initio calculations.

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Double Rydberg states of alkaline-earth atoms: two-electron dynamics far from the nucleus

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The motion of two electrons excited far away from the nucleus is the result of the subtle balance between their mutual Coulomb repulsion and the Coulomb attraction of the residual ionic core. In highly excited "double Rydberg" states (DRS), strong electronic correlations give rise to complex two-electron dynamics that range from chaotic motion to quasi-stable orbits depending on the relative degree of excitation of the two electrons and on the details of electronic correlation [1-3]. DRS thus represent an ideal test bed to explore the quantum mechanical three-body Coulomb problem. However, a detailed theoretical description of the energies and dynamics of DRS, combined with systematic experimental studies in the time- or frequency domains, is lacking and hampers our understanding of a seemingly simple problem: how two electrons move far away from the nucleus.

Theoretically, the recent development of the method of configuration interaction with exterior complex scaling (CI-ECS) has paved the way to accurately treating high-lying doubly excited states [2]. The method relies on a two-active-electron approach, on the use of exterior complex scaling to treat the hundreds of open channels in which DRS can autoionize, and on optimized numerical basis functions to make the CI expansion as compact as possible. In this way, the energies, lifetimes and wavefunctions of the very dense manifold of DRS resonances just below the double ionization threshold ($E\sim$ -0.5 meV) can be calculated, providing a detailed information on the associated electron correlations and dynamics [3].

In this talk, I will present the recent advances achieved with combined theoretical CI-ECS and experimental approaches in the study of the DRS of the Sr atom. I will explain (i) how such states are prepared through sequential, resonant, multiphoton excitation of the two valence electrons of Sr; (ii) how they are measured through multiple double-ionization mechanisms; and (iii) how the signatures of electronic correlations are visible in the dense and complex DRS spectra. A particularly interesting class of DRS are planetary states, in which the two-electron motion resembles the motion of two planets orbiting a star. The strong interaction between the electrons induces a dynamical localization of the electronic wavefunction (Fig. 1) and long autoionization lifetimes [1, 3]. Planetary states will be identified in the experimental spectra and their spectacular two-electron dynamics will be visualized with the calculated wavefunctions.

The results presented in this talk will provide insight into the physics of DRS and the underlying three-body problem [3], ultimately paving the way to controlling and manipulating atoms in such states at the quantum-state level and using them for, e.g., nondemolition measurements or simulation [4].

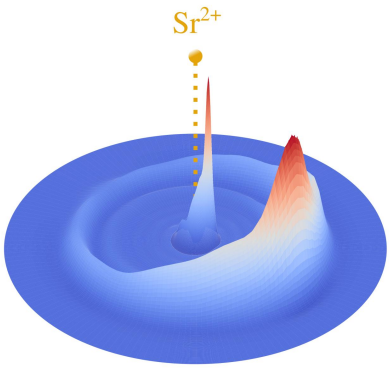

Figure 1. Two-electron composite probability density for a planetary state of the Sr atom below the N=8 ionization threshold. The composite density was obtained by summing, for each electron, the two-electron density calculated while fixing the other electron at its most probable radial distance r.

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Abstracts of contributed talks

Unveiling the ultrafast relaxation dynamics of thymidine in aqueous solution via extreme ultraviolet time-resolved photoelectron spectroscopy

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It is believed that nucleobases are purposeful selected as the fundamental building blocks of DNA, the essential macromolecule of life. After the solar radiation, DNA bases prefer to rapidly dissipating the absorbed energy before harmful reactions take place. This so-called photoprotection mechanism manifests as the ultrafast electronic relaxation of the optical bright $^1\pi\pi^*$ state to its ground state (S_0). Interestingly, a noticeable contribution from a long-lived optical dark $^1n\pi^*$ was observed during the photodeactivation of some pyrimidine nucleobases by transient absorption spectroscopy [1], a finding that seems to contradict the photoprotection principle but remains unverified by utilizing ultraviolet time-resolved photoelectron spectroscopy [2]. Moreover, investigating the relaxation dynamics of nucleobases in aqueous solution, is of the utmost importance to understand the role of photoprotection mechanism in the natural DNA environment.

In the present work, we study the ultrafast relaxation dynamics of thymidine in aqueous solutions using extreme ultraviolet time-resolved photoelectron spectroscopy (XUV-TRPES). The thymidine solution (0.05M) was firstly excited by an UV (266nm) pulse, and then ionized by an XUV pulse with its photonenergy equals to 23.2 eV, ensuring the ionization from all electronic states participated into the relaxation processes. The decay profile of the photoelectron intensity integrated over all possible excited-states is shown in Figure 1.

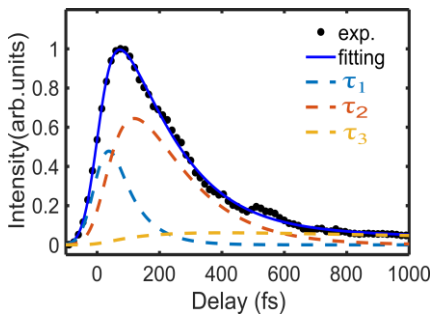

Figure 1. The decay profile of the photoelectron intensity integrated over all possible excited-states.

It is clearly observed that, the experimental result is very well fitted by three decay components with different lifetimes, where the longest-lived component contributes the least. To interpret our results, we performed QM/MM TRPES simulations using the Cobramm platform[3]. The nucleobase and sugar with the surrounding waters were treated at the QM and MM level, respectively. Both manifolds of QM states were obtained at the accurate CASSCF/CASPT2 level of theory within the MM point charged field [4]. The population dynamics is reported in Figure 2. It is clearly shown that, the $^1\pi\pi^*$ state population decays rapidly within the first 100fs and then exhibits a plateau until 200fs. Interestingly, the $^1n\pi^*$ state population increases to its maxium around 100fs and shows a monotonous decaying character, which indicates that the $^1n\pi^*$ state is only transiently populated. The decay of the population of other states is considerably slower compared to that of $^1\pi\pi^*$ and $^1n\pi^*$, which may correspond to the long-lived signals in Figure 1.

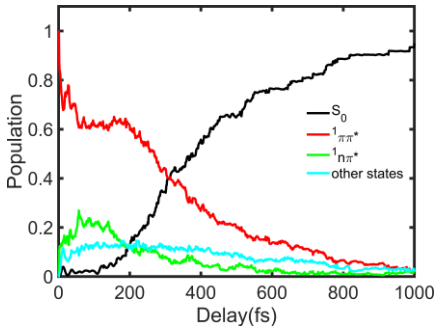

Figure 2. Electronic states population dynamics.

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Intermolecular Coulombic Electron Capture in Aqueous Systems

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Intermolecular Coulombic Electron Capture (ICEC) is an environment assisted process in which a free electron can efficiently attach to a quantum system by transferring the excess energy of the electron capture to a neighbor ionizing it (see Fig. 1).

ICEC is expected to be a general process that controls damage of living cells under ionizing irradiation. Furthermore, water being the solvent of life, it is therefore highly relevant to investigate ICEC in aqueous systems. The latter can thus reveal the potential impact of ICEC on radiation damage.

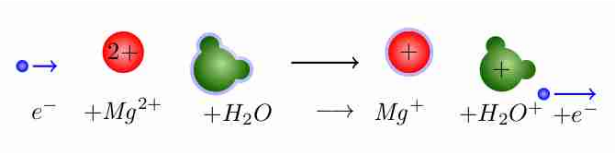

Figure 1. Scheme of the ICEC process: The magnesium dication recombines with a free electron to form a singly charged ion. The excess energy is transferred to the water molecule which is ionized. Figure taken from [1].

In the last 5 years, our group has developed the methodology and numerical tools to investigate ICEC in various systems, ranging from rare gas dimers to micro-hydrated cations [1-4]. More recently, we have focused our attention to ICEC in aqueous systems. In this contribution, we will give an overview of our works and summarize our current knowledge of ICEC.

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Intermolecular Coulombic decay of inner-valence vacancies in biologically relevant systems. Theory meets experiment.

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The ionization of the molecule triggers numerous relaxation processes, which become even more diverse as the vacancy becomes deeper in energy. The variety is further increased by the involvement of neighboring molecules or the solvation environment. If the internal energy of the molecule is sufficient, the secondary electron can be ejected. In the case of organic molecules, this is already possible with inner-valence ionization. The fastest and most probable of these processes is Auger decay. In the presence of a neighbor, intermolecular Coulombic decay (ICD) can occur by transferring the relaxation energy from the primary ionized molecule to the neighbor and ionizing it. Relaxation can also lead to electron transfer mediated decay (ETMD), in which two vacancies are created on neighboring molecules while the originally ionized one is entirely reduced. Although the latter two processes are nonlocal relaxation processes, they are known to play an important role in redistributing the harmful effects of ionizing radiation between biomolecules as well as to the environment [1, 2], and the secondary electrons produced are known to have harmful effects on biological tissues. Thus, understanding the underlying mechanisms is not only of fundamental importance, but can also be used to control such processes, e.g. for medical purposes.

The system can flow from the single to the double ionized state if the latter is energetically preferable. The energetically open channels can be determined by comparing the single and double ionization spectra. In our work, the propagator methods (based on the theory of Green's function) in the algebraic-diagrammatic construction (ADC) approximation are used to compute the IP and DIP spectra. Such approaches are well suited for the theoretical treatment of excitation, ionization, and double ionization in molecules and molecular clusters, providing the opportunity to model the spectra in wide energy diapasons, which, among other advantages, provides the opportunity to estimate the secondary electron spectra comparable to the experimental data.

We have shown that the scenario of decay of the inner-valence vacancy depends on it's location and the type of non-covalent interactions with neighboring molecules [3, 4]. While for heterocycles all: Auger, ICD and ETMD are operative and compete with each other, for water molecules only nonlocal variants are available. As a consequence, ionization of water in the vicinity of the organic molecule can lead to single and double ionization of the latter by ICD and ETMD, respectively, or can be redistributed over the solvation shell by the same schemes. An analysis of such possible competing processes will be given based on our recent theoretical results considering competing Auger, ICD and ETMD processes in dimers and hydrated clusters of heterocyclic molecules. The comparison with the experimental data will be discussed.

This work has been supported by Russian Science Foundation (grant № 23-23-00485) https://rscf.ru/en/project/23-23-00485/.

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Mapping the potential energy curve of Auger final state from resonant decay in O_2 molecules

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With the x-ray photon energy below the ionization threshold, a molecule can be promoted to a dissociative state. When the lifetime of the Auger decay is comparable to that of dissociation processes, there are two types of decay features, one is atomic feature, anther is molecular feature [1].

With the data acquired using the EPICEA setup at PLEIADES beamline at synchrotron SOLEIL, in Fig. 1, we plotted the correlation between the Auger electron kinetic energy and ion kinetic energy at the photon energy of 539.75 eV.

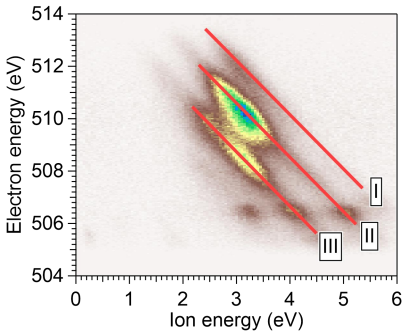

Figure 1. The correlation between the Auger electron kinetic energy and ion kinetic energy.

There are three features lead to three dissociation limits I, II and III, respectively. With the help of energy-conservation law, we can identify the dissociation processes. The dissociation limits energies and the Assignment of fragments are listed in Table 1.

Table 1. The dissociation limit energy and Assignment of fragments.

	diss.limit. (eV)	Assignment
I	20.69	$(O)^1D+(O^+)^4S$
II	22.04	$(O)^3P+(O^+)^2D$
III	23.74	$(O)^3P+(O^+)^2P$
	24.01	$(O)^1D+(O^+)^2D$

According to Table 1, we concluded that dissociation limits II and III could contain the atomic features, while I contain on molecular features. To further disclose where Auger decay happens, we plotted the correlation between the electron energy and the electron emission angle in molecule for the dissociation limit I in Fig. 2.

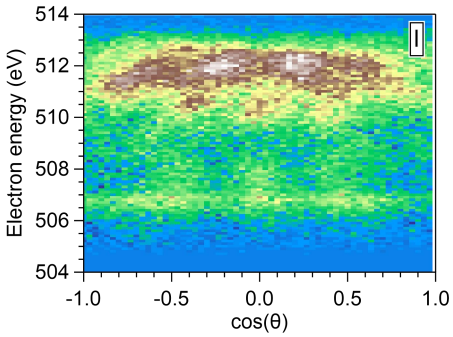

Figure 2. The correlation between the electron energy and angle of dissociation limit I.

In Fig. 2, we observed several double-slit interference patterns. It can be seen that the deduced internuclear distance depends on the electron kinetic energy. According to the potential energy curve of core excited state [3], we can map the potential energy curve of Auger final state, as shown in Fig. 3.

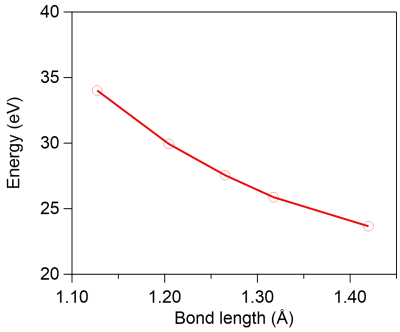

Figure 3. The calculated potential energy curve of Auger final state.

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Studying of charge exchange cross section of highly charged ion interact with atom and molecule

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Electron capture (EC) of highly charged ions (HCIs) with neutral atoms and molecular species is a fundamental process that occurs in various plasma environments. It affects the ionization balance and leads to spectral line emissions in astrophysical regions or fusion devices. For example, Ne is one of the main impurities appearing at various relative energies in different ionization stages in the plasmas. Furthermore, the EC between the Ne^{8+} ion and O_2, N_2, CH_4, H_2 and He is the dominant collision process in the solar wind. For these applications, there is a strong demand for accurate data on the cross sections of EC.

The experiment is performed on the 150 kV highly charged ion collision platform at Fudan University to measure the total and state-selective EC cross sections [1,2]. The HCIs beam produced by an ECR ion source collides with neutral gases in a gas cell to measure the total EC cross sections. The systematic uncertainties of our experimental and measurement procedures are analyzed in detail, and eventually the experimental error of absolute cross sections of single electron capture (SEC) is reduced to lower than 9%. As for the state-selective EC cross sections, the measurements were performed using a cold target recoil ion momentum spectroscopy (COLTRIMS) apparatus [3].

The absolute total cross sections of EC in collisions of He^{2+} with He have been studied in energy ranges from 3.5 to 50 keV/u [4]. The present result of He^{2+}- He collision shows good consistency with previous measurements, which verifies the reliability of the experimental system and paves the way for precise measurements of EC cross sections for a variety of ions and neutral gases. On this basis, the total EC cross sections for Ne^{8+}- O_2, N_2 and CH_4 were performed, as shown in Figure 1, filling in the gaps of the EC cross section data in the corresponding energy region.

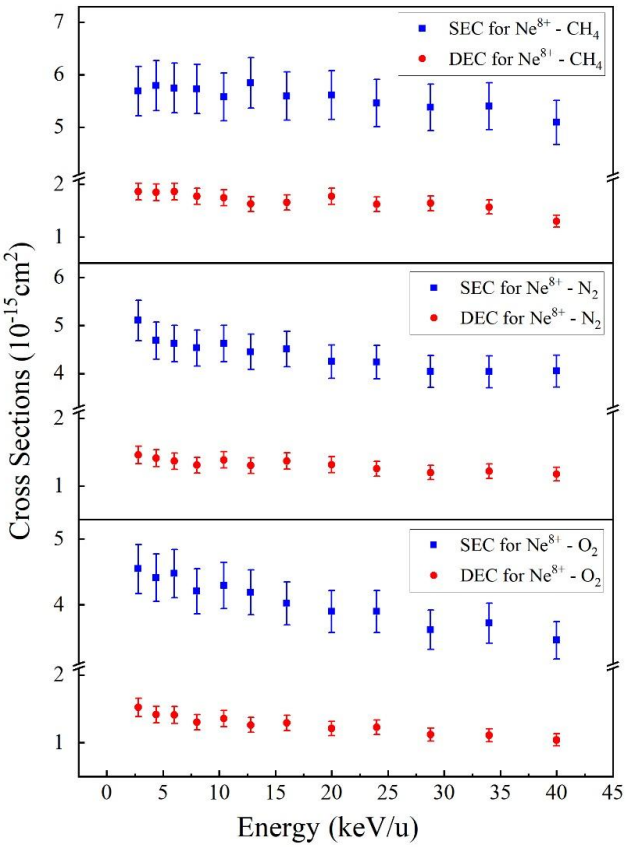

Figure 1. Measured absolute SEC (filled square ■) and DEC (filled circle ●) cross sections for Ne^{8+} colliding with CH_4, N_2, and O_2. The errors are given at the 1σ level.

Furthermore, the total and *n*-resolved state-selective cross sections of SEC between Ne^{8+} and H_2 and He were measured in the impact energy range between 8 and 40 keV/u, which is of interest for fusion research.

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Application of Semi-Classical Methods in Ultrafast Electron Dynamics Information Probing

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Ultrafast pump-probe techniques enable the measurement of time delays in various systems, such as atoms, molecules, and solids, during the process of light emission with unprecedented temporal resolution. Strong-field photoelectron spectroscopy encodes spatiotemporal information about electron and ion dynamics. To extract this dynamic information, we employed semi-classical methods and experimental data, combined with phase-controlled two-color femtosecond laser pulses, to observe a Feynman resonance time delay of 140 attoseconds in multiphoton ionization of atoms[1]. Additionally, through semi-classical imaging, we discovered that photoelectrons can be temporarily recaptured into quasi-bound states by atomic potentials after appearing in a continuum state[2]. They remained in this state for several hundred attoseconds before being emitted again. We introduced, for the first time, a deep learning-based strong-field Feynman path integral method, incorporating deep neural networks to solve complex problems in strong-field attosecond physics[3]. We proposed a self-referenced molecular attoclock scheme, revealing the transient resonant ionization radius of Ar-Kr+ dimer molecules and measuring the time delay of electron binding in the molecular resonant state as 3.5±0.04 fs[4].

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Statistical analysis of X-ray spectra of aqueous triglycine

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The folding of proteins is a complex and complicated question [1], affected by both intramolecular and intermolecular interactions. The process is ultimately dependent on the primary order of the amino acids and the surrounding solvent. While diffraction allows for atomistic structure determination of proteins, it requires a crystalline sample. Instead, owing to its localized mechanism, X-ray spectroscopy maintains its sensitivity to atomistic structure also in the liquid phase.

A liquid system allows for the movement of relatively strongly interacting molecules, which leads to a broad distribution of possible configurations. These individual configurations have been computationally observed to have significantly different X-ray spectra, only their ensemble mean predicting the experimentally observed spectrum. Although X-ray spectroscopy does not permit full structural reconstruction, its principles allow for discovery of structural information, especially when combined with statistical analysis [2-4] of computational results.

In this contribution, we present our ongoing statistical analysis of computational X-ray spectra of aqueous triglycine. Classical molecular dynamics (MD) was used to obtain the configuration distribution of the system (see Fig. 1a, visualised with the Ramachandran plot in Fig. 1b). Then, X-ray absorption spectra were calculated with density functional theory (Fig. 1c).

The main goal of our study [4] is to determine if X-ray spectroscopy could be used to gain new structural information of aqueous tripeptides. As the systems are complex, simple tools like linear correlations between the internal coordinates of the system and its X-ray absorption spectrum do not deliver satisfying results. We approach this problem by applying neural networks and a specialized dimensionality reduction method, as already done with a different system [3]. The results are compared to X-ray Raman scattering spectra of liquid triglycine at the nitrogen K edge.

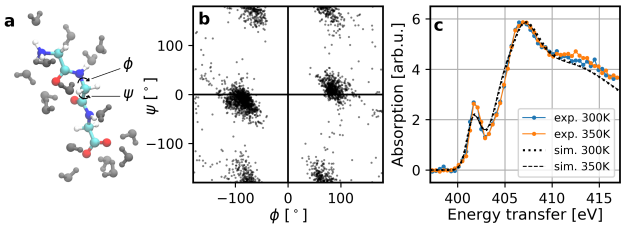

Figure 1. Results for the study of triglycine(aq). **a**: An example configuration and its Ramachandran angles φ and ψ. **b**: Ramachandran plot for an MD trajectory. **c**: Experimental X-ray Raman scattering spectra and the mean of simulated X-ray absorption spectra at two temperatures.

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Non-resonant elastic and inelastic x-ray scattering study of nitrous oxide

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X-ray scattering technique is one of the most powerful tools to study the structures or excitation dynamics of condensed and gaseous materials. For the structure determination, the elastic (coherent) x-ray scattering by atoms and molecules in the target is the physical basis of the standard experimental methods like x-ray diffraction. Hence, knowledge of this elementary scattering process is of great importance in the structure analysis [1].

In contrast, the inelastic x-ray scattering is used to reveal the excitation dynamics of the target. Since 2010, non-resonant inelastic x-ray scattering (NRIXS) technique has been employed to study the valence- and inner-shell excited states in atoms and molecules [2,3]. Particularly, inner-shell excitation of atoms and molecules can bring us many novel physical insights due to its uniqueness for different elements and the abundant decay channels of the core-excited states, including radiative decay and nonradiative Auger decay. Hence, the study of the inner-shell excitation of atoms and molecules has attracted widespread interest, for which many experimental techniques such as electron energy loss spectroscopy, x-ray absorption spectroscopy, resonant inelastic x-ray scattering and resonant Auger spectroscopy have been adopted.

Nitrous oxide (N_2O) has a linear configuration of N_t-N_c-O at the ground state, the core-excitation properties of the two N atoms are expected to be distinct due to their different chemical environment. Furthermore, the structural information, such as the bond lengths and the binding effect, can be extracted from the elastic X-ray scattering experiment.

In this work, based on the X-ray scattering technique, the elastic X-ray scattering spectra of N_2O and the NRIXS spectra of the K-shell excitations of the two N atoms have been measured at the scattering angles of 20˚-100˚, an incident photon energy of about 10 keV and an energy resolution of about 1.3 eV. The energy difference between transitions of N_t: 1s→3π and N_c: 1s→3π have been derived, while the elastic squared form factor (ESFF) and the generalized oscillator strengths (GOSs) of these transitions have been determined at squared momentum transfers in 0.84-17.2 a.u.

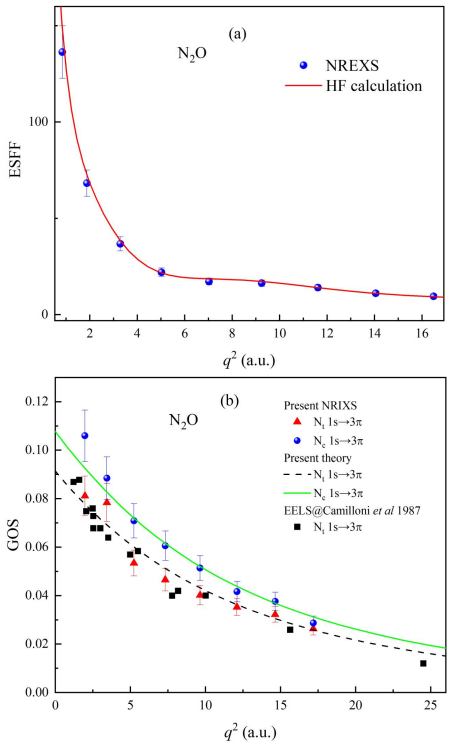

Figure 1. (a) The ESFF of N_2O. (b) The GOSs of N_t: 1s→3π and N_c: 1s→3π excitations.

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New source for tuning the effective Rabi frequency discovered in multiphoton ionization

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A detailed account for the Autler-Townes (AT) effect [1] measured in the photoelectron momentum distribution of multiphoton ionization of potassium atoms is presented. Beyond the normal AT splitting, at higher intensities we observe significant deviation from the Rabi frequency. To explain the experimental observations, we treat the 4s-4p as a simple two-level system and the ionization process as a pure loss. The effective Hamiltonian can be written as:

$$H = H_0 + H' \quad (1)$$

where $H_0 = \begin{pmatrix} 0 & \frac{1}{2}\Omega_{Rabi} \\ \frac{1}{2}\Omega_{Rabi} & 0 \end{pmatrix}$, and $H' = \begin{pmatrix} 0 & 0 \\ 0 & i\Gamma \end{pmatrix}$, Ω_{Rabi} and Γ represent the coupling strength of the two-level system with the laser field and the ionization rate, respectively. The energy splitting of such system can be related to the renormalized effective Rabi frequency as:

$$\Delta E = \Omega_{Rabi}^{eff} = \sqrt{\Omega_{Rabi}^2 + (i\Gamma)^2} \quad (2)$$

The comparison between our model and the experimental results is plotted in Fig. 1. The agreement is excellent.

To summarize, an effective Rabi frequency formulae taking into account the ionization process is derived, which indicates that decay processes causing loss of populations will affect the Rabi frequency which in turn change the coupling strength.

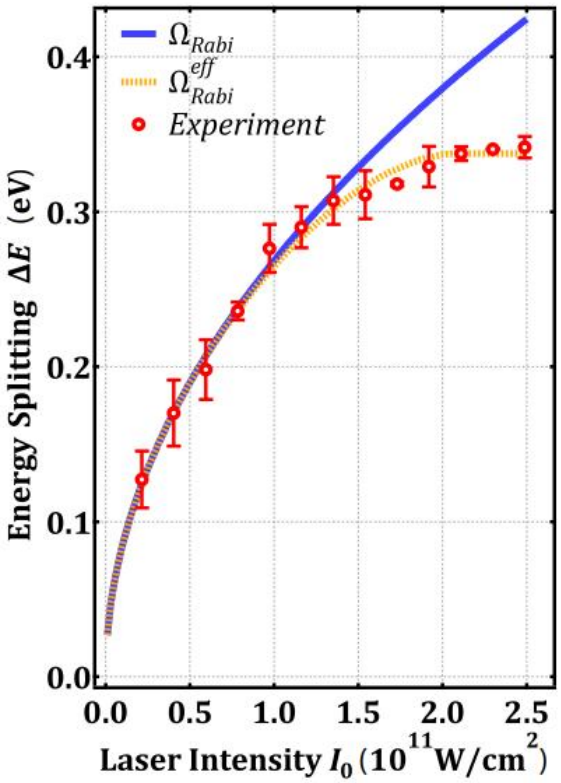

Figure 1. The energy separation of the Aulter-Townes splitting as a function of laser intensities. The red dots represent the experimentally measured kinetic energy difference of the photoelectron. The error bar is the statistical error from ten individual measurements. The blue line is the Rabi frequency. The orange broken line is the effective Rabi frequency in Eq. 2.

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UV-induced ring conversion dynamics investigated by time-resolved Coulomb explosion imaging

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Ultrafast ring-conversion reactions driven by ultraviolet (UV) light play an important role in many chemical and biological processes. To obtain a time-domain picture of these reactions, they have been recently studied using time-resolved photoelectron spectroscopy (TRPES) [1] and ultrafast electron diffraction (UED) [2].

Here we apply a complementary approach, time-resolved Coulomb explosion imaging (CEI) [3,4] for studying UV-induced ring conversion dynamics in two five-membered heterocyclic molecules, dihydrofuran (C_4H_6O) and thiophenone (C_4H_4OS). For thiophenone, we excite the molecule to the S2 electronic state using a 266 nm pump, which results in C-S bond extension, ring opening and ultrafast nonradiative decay back to the electronic ground state [1], where different families of photoproducts are formed [2]. We probe the evolving molecular structure using multiple ionization and CE induced by intense, femtosecond X-ray or near-infrared (NIR) laser pulses and detecting four or more ionic fragments in coincidence (see Fig. 1a-f) [4]. For dihydrofuran, which can be excited to the $\pi\pi^*$ state using 200 nm UV pump, the molecule deforms on its way to the conical intersection leading to O-C bond extension, ring opening or ring puckering [5]. Here, we probe the dynamics employing CEI by intense NIR probe pulses (see Fig. 1g-i). For both molecules, the CEI patterns reveal the dynamics unfolding on sub-200 fs time scale. Comparing the experimental data with the results of CE simulations, we can directly visualize the UV-induced changes in the ring structure of the molecules.

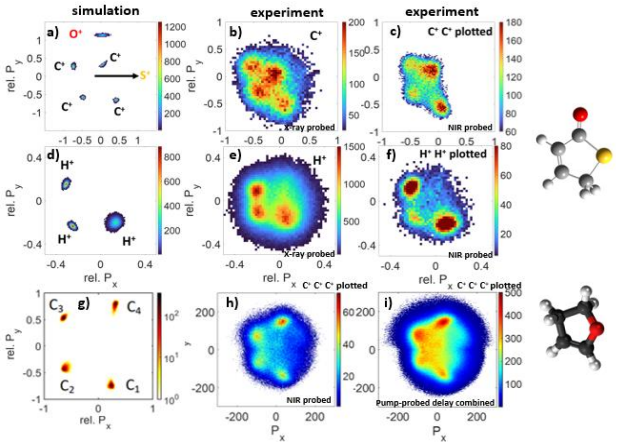

Figure 1. (a-f) Molecular-frame momentum distributions (MFMD) for CEI of thiophenone. In all panels, the S^+ momentum vector defines the $+x$ direction, whereas the (x,y) plane is determined by the S^+ and O^+ momenta. Panels (a-c) show the momenta of C^+ ions and (d-f) – of H^+ ions in that reference frame. (a,d) show the results of the CE simulations, (b,e) present the results obtained with the 2.7 keV X-rays, and (c,f) – with 800 nm NIR pulses.
(g-i) MFMD for C^+ ions in CEI of dihydrofuran. The O^+ momentum defines the $+x$ direction, and the (x,y) plane is determined by the momenta of O^+ and one of the C^+ ions. (g) CE simulations for the unpumped molecule; (h) experimental data for NIR pulse only; (i) UV pump – NIR probe data integrated over delay range of [-300, 1020] fs.

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complete characterizations of intermediate and final state wave functions with photoionization of polarized Rb

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Alkali atoms play a crucial role in the study of atomic physics and quantum optics and have attracted increasing attention in the past few decades with the rapid advancements of cold atom technology. In this work, a Rb-MOTRIMS (magneto-optical trap recoil ion momentum spectroscopy) setup [1] is adopted for experimental photoionization of cold Rb atoms, which provides a very high degree of target polarization. For polarized atoms, a "complete" quantum-mechanical description of the photoionization process can be presented.

In the MOTRIMS, in addition to Rb atoms in the $5S_{1/2}$ state, a certain fraction of excited Rb atoms ($5P_{3/2}$) are effectively populated by the cooling lasers. It is thus possible to compare directly the ionization mechanism of the two states exposed to the same laser. The measured recoil-ion momentum distribution (RIMD) in the polarization plane (x-z plane) of the ionizing laser is presented in Fig.1 (a). RIMD of Rb^+ exhibits rich ring-like structures and these energies correspond to single-photon ionization of the $5P_{3/2}$ state (pr = 0.2a.u.), two photon (pr = 0.39a.u.) and three-photon ionizations of the $5S_{1/2}$ state (pr = 0.6a.u.), respectively. The corresponding TDSE simulation result shown in Fig.1 (b) can qualitatively reproduce the experimental results. On the other hand, the intensity-dependent scaling law of the ion yields ratio η_{5S}/η_{5P} can serve as an indication for the onset of strong-perturbative few photon ionization, as presented in Fig.1 (c). Although the RIMD and ionization yields obtained by TDSE agree well with the experimental results, there are still noticeable discrepancies in the change of the near zero momentum dip of one-dimensional momentum distribution with the laser intensity, as shown in Fig.1 (d), which could stem from the electron-electron correlation [2].

Further, to get a deeper understanding of the underline physics, a complete experimental reconstruction of photoionization dynamics is required, i.e., both the amplitude and the phase of different ionization channels. Recently, the Coulomb wave function phase difference between the final state channels are extracted by changing the chirality of the ionized laser and controlling the magnetic quantum number of the $5P_{3/2}$ state. More details will be presented in the meeting.

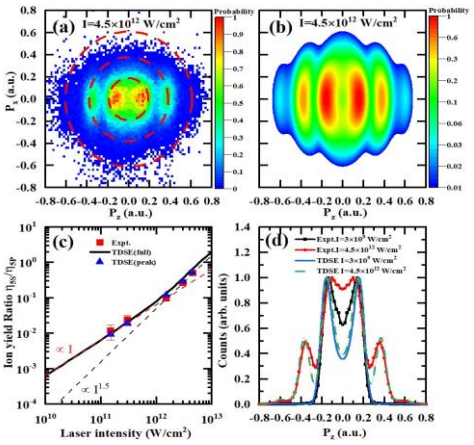

Figure 1. Experimental and theoretical results of momentum distribution and ion-yield ratio.

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Abstracts of posters

Excitation dynamics of the $5p^5nln'l'$ states in Ba+e^- collisions

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Ionization of the $5p^6$ subshell in barium leads to the forming a large class of the $5p^5nln'l'$ states, most of which are located between the second ionization limit and the lower excited levels of Ba^{2+} ion. For these states, which play an important role in double ionization [1, 2], there are still no cross-sections and their classification is largely contradictory (see [3, 4] and references therein).

Because the formation of Ba^{2+} ions in the ground state is the only de-excitation channel for $5p^5nln'l'$ states, their cross-sections are proportional to the intensity of the corresponding lines in the Auger spectra. Thus, measurements of spectra with appropriate energy resolution over a wide range of impact energies make it possible to obtain the energy dependence of the cross-sections. The latter contains direct information about the nature and efficiency of electronic transitions. Recently, this method was used to study ionization of the $4p^6$-subshell in strontium [5].

Using the apparatus and measuring procedure [5], we obtained preliminary data on the ionization cross-sections for the $5p^5nln'l'$ states, which are represented in the Auger spectra by the most intense and well-separated lines (see numbered lines in the spectrum in Figure 1). The measurements were carried out for incident electron energies of 27.5, 32.5, 52.5, and 102.5 eV. The relative uncertainty in determining the line intensity generally did not exceed 20%.

As can be seen from Figure 2, the different

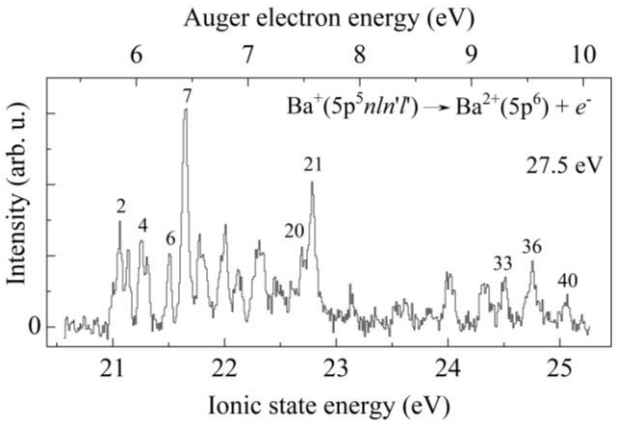

Figure 1. Barium $5p^6$ Auger spectrum measured at 27.5 eV incident electron energy.

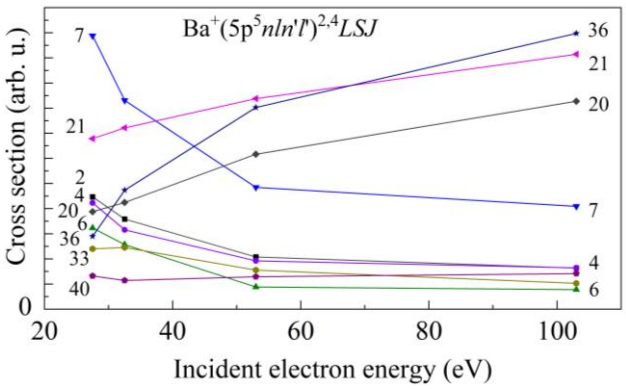

Figure 2. The ionization cross sections for the $5p^5nln'l'$ states in Ba^+.

shapes of the cross-section are observed. Cross-sections with maxima at low impact energies should reflect the formation of states with a dominant quartet character [5]. That is consistent only with the identification of lines 2 and 4 with the quartet states $5d^2(^3F)^4D_{3/2}$ and $5d(^3P)6s^4P_{3/2}$ [3]. The shape of the cross-sections 6, 7, and 33 contradicts their identification in [3] as $5d(^3P)6s^2P_{1/2}$, $5d(^3P)6s^2P_{3/2}$ and $5d(^1D)6s^2D_{5/2}$ doublet states. The cross-section 40 possesses a quartet-like behavior at low impact energies, but its maximum is around 100 eV. That may indicate the presence of a weak quartet component in the $5d^2(^3P)^2S_{1/2}$ + $5d^2(^3P)^2D_{5/2}$ state [3].

Direct ionization of the $5p^6$ subshell with forming the $6s^2\ ^2P_{1/2,3/2}$ states is described by the cross-sections of lines 20, 21, and 36. A remarkable enhancement of the cross-sections 20 and 21 at low energies may be due to the presence of the quartet components (0.557 $5d^2(^3F)^4F_{3/2}$ for the state 21[3]).

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Measurements of absolute cross sections for ionization and fragmentation of chlorine and/or fluorine-based ozone-damaging molecules

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The experimental determination of absolute electron impact ionization cross sections for molecules is a difficult task and has been restricted to a limited range of species.

Knowledge of absolute ionization cross section is important for a variety of applications, including calibration and normalization of experimental and theoretical data. The direct measurement of absolute cross sections is challenging. A sophisticated calibration procedure is required in order to convert the measured relative data into absolute values. On the theory side the accurate quantum mechanical treatment of the ionization cross section is generally possible for atoms and atomic ions. Since calculations for molecular species are much more challenging several approximate semi-empirical and semi-classical models have been developed.

The aim of this study within the scope of the 21GRD02 BIOSPHERE* project, is to understand the influence of energetic electrons produced, e.g., by cosmic radiation (CR) on the composition and chemistry of the Earth's atmosphere. Here in particular processes influencing the stratospheric ozone layer are of interest. Therefore, we consider electron impact ionization of the ozone depleting chlorofluorocarbons (CFCs) and hydrochlorofluorocarbons (HCFCs) but also hydrofluorocarbons (HFCs) which are intended to replace these substances but, themselves, are potent greenhouse gases. Following the control on the production and use of CFCs under the provisions of the Montreal Protocol, the production and use of hydrofluorocarbons (HFCs) increased significantly [1].

Electron impact ionization cross sections have been measured as a function of the incident electron energy from threshold up to 1 keV. For absolute calibration known quantities of the species under study and a reference gas (argon, krypton, xenon), for which absolute cross sections are known, were mixed. As an example, Figure 1 shows absolute cross sections for the production of various ionic fragments from HCFC-22 ($CHClF_2$).

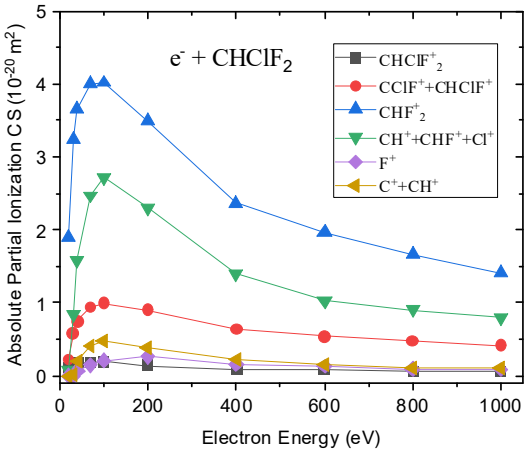

Figure 1. Absolute cross sections for production of various ions from $CHClF_2$.

Clearly, the yield of the molecular fragments including the ozone active fragments Cl and F is highest at low impact energies below 100 eV. This coincides with the most likely energies of secondary electrons from cosmic rays. The strongest fragmentation channel is $CHClF_2 \rightarrow CHF_2^+ + Cl$ from which only the residual ion is detected.

Our results will give new insight into the mechanism of electron collision dynamics in ozone-damaging molecules.

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Structural and dynamical studies of CH–π bonded CH_4–C_6H_6 dimer by ultrafast intermolecular Coulombic decay

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The CH-π bond is widely present in nature and plays a crucial role in the crystal packing of organic compounds and the construction of high-performance materials. The CH_4-C_6H_6 dimer, as a typical CH-π bonded system, has attracted considerable attention due to the ubiquity of methyl and phenyl groups in biorelated molecules [1]. When aromatic π systems are ionized, the positive charges are often found to participate in CH–π bonding and strengthen the binding ability due to the strong induction effect in the cation. In contrast, if the ionization proceeds in the deeper inner-valence orbital, the system can undergo significant molecular and structural transformation via ultrafast electronic relaxation, e.g. inter-molecular Coulombic decay (ICD) [2]. We investigated the ionization and dissociation dynamics of the CH_4-C_6H_6 dimer by electron impact, confirming that the ICD process also plays a significant role in CH-π complexes.

In the experiment, the product ions (CH_4^+ and $C_6H_6^+$) resulting from the double ionization of the CH_4-C_6H_6 dimer were detected in coincidence using a reaction spectrometer. The study suggests that the CH_4^++$C_6H_6^+$ channel may originate from two ionization mechanisms, sequential ionization (SI) and intermolecular coulombic decay (ICD). As shown in Figure 1(a), for the SI, the incident electron successively knocks out one outer-valence electron from CH_4 and C_6H_6, leading to the repulsive CH_4^++$C_6H_6^+$ ion pair through coulomb explosion. In ICD, after an electron is removed from the inner-valence $2a_1$ orbital in CH_4 or the $2a_{1g}$ orbital in C_6H_6, one outer-valence electron fills the inner-valence hole and energies released ionize the neighboring molecule leading to the two-body fragmentation of CH_4^++$C_6H_6^+$ (Figure 1(b)). We find that the ICD process induced by the inner-shell ionization of CH_4 and C_6H_6 (σ_{ICD}(200 eV) = 0.436 Å²) dominates over the SI (σ_{SI}(200 eV) = 0.046 Å²) by calculating the absolute cross sections for the two ionization processes. Furthermore, through AIMD simulations of the CH_4^+-$C_6H_6^+$ dication dissociation dynamics, we presented the real-time dynamical evolution of CH_4-C_6H_6 dimer dications, including translational, rotational, and vibrational dynamics. By the Coulomb explosion imaging, we determine the global-energy minimum structure of the CH_4-C_6H_6 complex in the gas jet, where the CH_4 molecule lies on the benzene ring and one CH bond is pointing to one of the carbon atoms. This work is expected to have a potential implication for crystal structure imaging with various radiation sources [3].

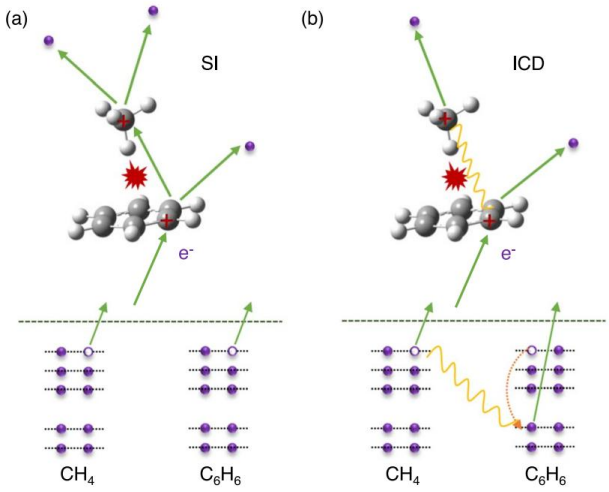

Figure 1. Schematic of electron impact-induced (a) sequential ionization and (b) ICD. The green solid and orange dashed arrows denote electron emission and intramolecular de-excitation, respectively. The yellow wavy lines denote intermolecular energy transfer.

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Electron impact induced ultrafast H_2 formation from tetrahydrofuran

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Studies on the ionization and fragmentation dynamics of isolated molecules induced by electron impact not only promote the understanding of radiation damage but also provide the clues of the formation mechanism of new molecules in the interstellar medium (ISM), planetary atmospheres, and plasma etching processes [1,2]. The hydrogen dynamics for H_2 molecule formation is the focus of interest, which could provide valuable insights into molecular hydrogen formation in atmospheres and ISM, and stimulate the hydrogen energy storage and utilization.

The tetrahydrofuran molecule (THF, C_4H_8O) is a five-membered aliphatic molecule with one oxygen heteroatom and four (CH_2) units, which is an important prototype of the biomolecules. In the present work, we report the formation mechanisms of hydrogen from THF and the subsequent detailed fragmentation dynamics.

The experiment was performed using a multiparticle imaging spectrometer (reaction microscope) which has been introduced comprehensively in previous studies [3]. The final-state charged particles, including fragment ions and one scattered electron, are detected in coincidence. The reaction channel for hydrogen formation was determined by time-of-flight (TOF) correlation map, as shown in Fig.1. First, the THF molecule is doubly ionized by electron impact:

$e^- + C_4H_8O \rightarrow 3e^- + C_4H_8O^{2+}$

Afterwards, the molecule H_2 is ejected, and the remaining $C_4H_6O^{2+}$ dication dissociates into two positive cations by Coulomb repulsion:

$C_4H_8O^{2+} \rightarrow H_2 + C_4H_6O^{2+}$

$C_4H_6O^{2+} \rightarrow CHO^+ + C_3H_5^+$

The two-body fragmentation channels $C_2H_4^+ + C_2H_4O^+$ and $CH_2O^+ + C_3H_6^+$ are also observed as two sharp coincidence lines. The reaction channel for hydrogen formation shows broad correlation features due to the unobserved neutral fragments that carries additional momenta. The two undetected hydrogen atoms are determined to form molecular hydrogen H_2 by theoretical calculations.

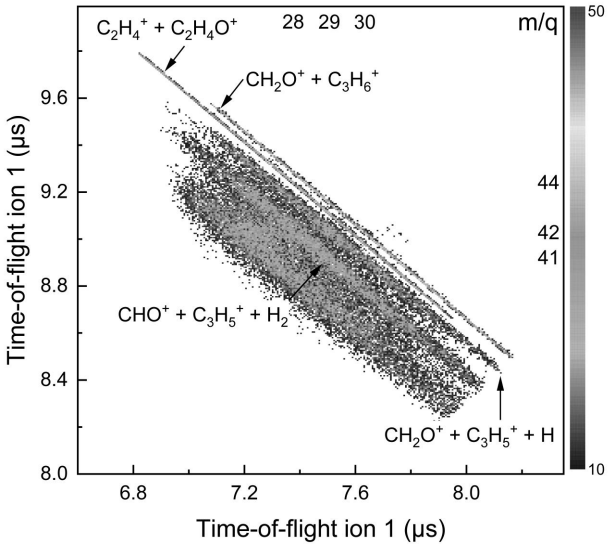

Figure 1. Correlation of the time-of-flight of the two measured ions.

Ab initio molecular dynamics (AIMD) simulations combined with high-level potential energy surfaces calculations (PESs) determined fragmentation mechanism of the hydrogen molecule production channel. The H_2 formation and ejection occurs in ultrafast time scales (50 fs), followed immediately by the ring opening via C–O bond cleavage, and finally Coulomb-explosion to the final ionic fragments.

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Electron-impact ionization of atoms and molecules in asymmetric kinematics

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The fundamental aim of basic research is to identify and comprehend the strength and significance of interactions among elementary particles. Given that the universe is predominantly made up of atoms, molecules, and charged particles, the exploration of interactions and collisions involving charged particles within atoms and molecules has been a central focus since the inception of quantum mechanics. The electron impact ionization of atoms and molecules is a fundamental process in collision physics. The triple differential cross-section (TDCS), which represents the most stringent test for theoretical models, is particularly investigated. Consequently, there have been increasing efforts to understand and model the fundamental interaction of the process called (e,2e) where very impressive results have been found for atoms and molecules.

In this work, we present two new developed models designated as 2CWZ and 3CWZ to study the dynamics of electron impact ionization of atoms and molecules [1, 2]. In these models, we utilize a Coulomb wave with variable charges Z(r) instead of an effective charge to describe the two outgoing electrons in the case of 2CWZ model, and all three electrons in the case of 3CWZ model. Additionally, the post-collision interaction (PCI) is incorporated and precisely treated at all orders of perturbation theory. We note that our models are a kind of approximate distorted wave approaches with PCI. The spherical static potential which enables to resolve the Schrödinger equation in the true distorted wave description, is used instead to calculate the variable charge Z(r). The results are systematically compared with recent experimental data and other theoretical predictions across various kinematic scenarios.

Figure 1 presents the calculated TDCS in coplanar asymmetric geometry. This situation is characterized by a large recoil momentum absorbed by the atom, indicating strong participation of the recoil ion in the reaction and necessitates a rigorous modeling of the process.

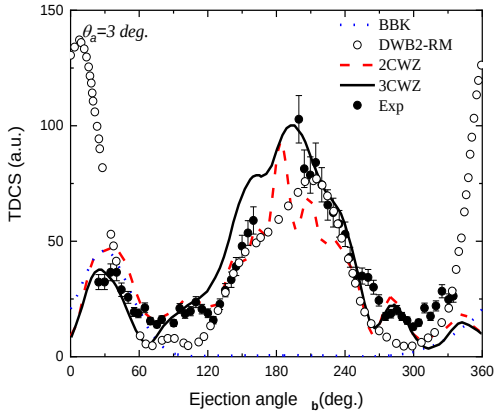

Figure 1. TDCS, in 10^{-4} au, for the electron impact ionization of argon 3p. The projectile is scattered with an energy E_a = 500 eV in coincidence with the ejected electron having an energy E_b = 205 eV. The data have been normalized to the 2CW and 3CW calculations. The DWB2-RM results have been multiplied by 0.34.

It is evident that, the DWB2-RM model effectively describes various aspects of the TDCS. However, in all instances, it displays a binary peak that is notably larger than that of the recoil region, which contradicts the observed data. Additionally, the 2CWZ and 3CWZ models demonstrate a relatively accurate reproduction of the TDCS across most sections of the angular distribution. It is worth noting that BBK has previously been shown to inadequately capture the recoil region. In interpreting these findings, it's important to note that DWB2-RM is a robust model with the inherent capability to provide a generally accurate description of this reaction. However, it falls short in accounting for (PCI), which is crucial in this context. Unfortunately, this omission explains the observed shortcomings in Figure 1, contrary to the 3CWZ model.

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Exploring vibronic couplings by (e, 2e) spectroscopy

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Couplings between nuclear and electronic motions, known as vibronic coupling, play pivotal roles in molecular systems [1-3]. This significance is particularly pronounced when two or more electronic states involved are degenerate or near-degenerate. The intrinsic nature of vibronic coupling lies in the mixing of electronic states induced by nuclear motion, which gives rise to various phenomena including intensity borrow of electronic transitions [4], Jahn-Teller (JT), Renner-Teller and pseudo-Jahn-Teller (PJT) effects [5], ultrafast nonadiabatic transitions [6], etc. Ethane, a molecule of D_{3d} symmetry in its neutral electronic ground state, is a prototypical molecule showing features of vibronic coupling [7, 8]. The typical (E⊗e) JT and ((A+E)⊗e) PJT effects exist among the doubly degenerate X ^2E_g and the A $^2A_{1g}$ electronic state of the ethane radical cation through e_g vibrational modes (ν_7, ν_8, ν_9). Theoretically, the Köppel, Domcke, and Cederbaum (KDC) model is employed to elucidate the irregular line shapes observed around 11-13 eV in the photoelectron spectrum of ethane [7, 8]. In these studies, the calculated vibronic coupling constants exhibit generally good agreement, except for the PJT constants, where notable discrepancies emerge. The PJT coupling constants, which describe the degree of mixing of X ^2E_g and A $^2A_{1g}$ electronic states, can be inferred from the binary (e, 2e) experiment.

In the present work, the valence shell binding energy spectrum and electron momentum profiles (EMPs) of ethane are measured by our high-sensitivity binary (e, 2e) apparatus, together with theoretical calculations involving vibronic couplings. Considering JT and PJT couplings in the X ^2E_g and A $^2A_{1g}$ states, the triple differential cross sections (TDCS) for X ^2E_g and A $^2A_{1g}$ can be expressed as $\sigma \propto d_1 \rho_{1e_g} + d_2 \rho_{3a_{1g}}$, where d_1 and d_2 denote vibrational average mixing coefficients. We plot the mixing coefficients $d_{3a_{1g},3a_{1g}}(\boldsymbol{Q})$ calculated by B3LYP as a function of dimensionless normal coordinate of the x component of e_g mode ν_7 (Fig. 1). The PJT coupling constant λ' is estimated to be 0.18 eV. Furthermore, by taking into account vibronic couplings both in the neutral and ionized electronic states, the theoretical EMPs reproduce the experiment very well, indicating the reliability of our vibronic couplings model (see Fig. 2).

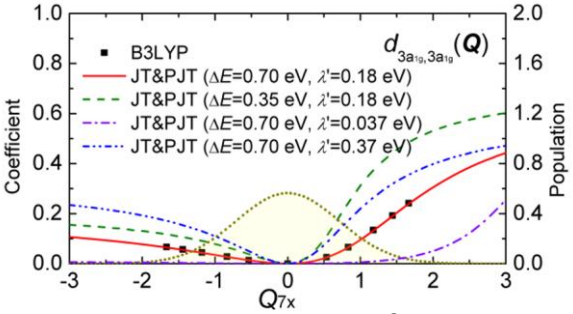

Figure 1. The mixing coefficient for A $^2A_{1g}$ along Q_{7x}. Square points are based on B3LYP calculations. The lines correspond to the diagonalization of the vibronic Hamiltonian, with the exception of the dotted line, which is the vibrational population of neutral ethane in the electronic ground state.

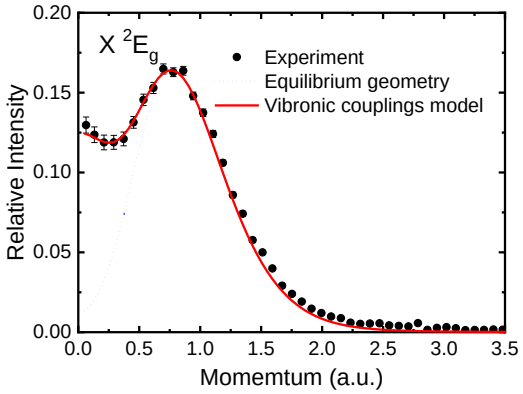

Figure 2. Experimental and theoretical momentum profiles of ethane for X ^2E_g ionization band.

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Precision Measurements of the $^2P_{1/2}$-$^2P_{3/2}$ fine-structure splitting in B-like S^{11+} and Cl^{12+} at SH-HtscEBIT

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Precision measurements of the fine structure transition energy of boron-like ions are considered important not only to investigate the fundamental physics [1], including QED effects, electron correlation effects, relativistic effects and atomic nuclear effects, but also to provide key atomic data for astrophysics [2]. Additionally, the M1 transition of the boron-like ions are considered for highly charged ion optical clocks [3].

We present the experimental results as well as the theoretical calculation of the electric dipole-forbidden transition $^2P_{3/2}$ - $^2P_{1/2}$ in boron-like S^{11+} and Cl^{12+} ions. The experiments were per-formed by using a high-resolution Czerny-Turner spectrometer at the SH-HtscEBIT [4]. Emission lines from S^{11+} and Cl^{12+} ions are shown in Figure 1 (a) and (b). The observed peaks belong to the $^2P_{3/2}$ - $^2P_{1/2}$ transition of B-like S^{11+} and Cl^{12+} ions are indicated. All random uncertainties including line position errors and standard deviation of wavelength calibration function have been calculated as a "root sum of squares," whereas systematic calibration errors and uncertainties from calibration lines have been added linearly. The final transition wavelengths of S^{11+} and Cl^{12+} are determined as 760.9635(29) nm and 574.1539(26) nm (in air), respectively. Additionally, the M1 transition energies in S^{11+} and Cl^{12+} were evaluated within the ab initio QED framework to compare with the experimental data. The present experimental results agree with the theoretical calculations and provide a possibility to test QED effects and correlation effects with high accuracy in few-electron highly charged ions. The details can be found in Ref. [5].

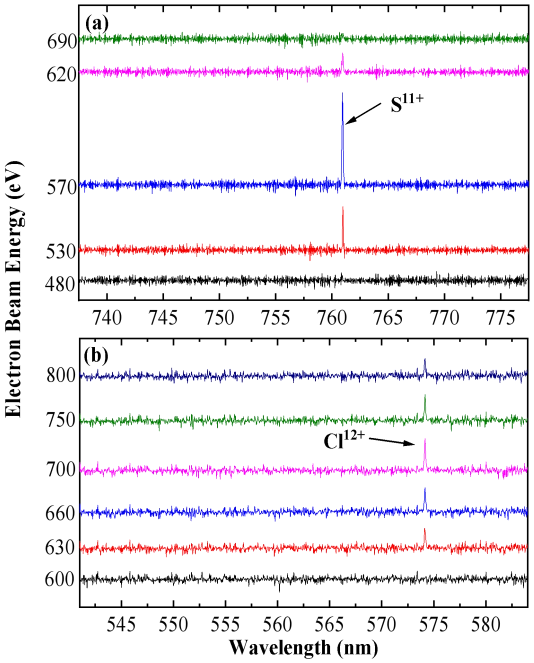

Figure 1. Emission spectra of B-like ions. (a) for S^{11+} in the range of 737–777 nm and (b) for Cl^{12+} in the range of 541–584 nm. The nominal electron beam energies are labeled in the vertical axis respectively for each ion with different colors.

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Predicting pharmacological activity by using frontier electron density in momentum space

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In drug screening, quantitative structure-activity relationship (QSAR) has been widely used. It can predict pharmacological activity by relating a set of phyco-chemical properties of test molecules, such as energy, hydrophilicity, to the biological activity through data training, though it provides no understanding of why. One may desire a simple but quantitative prediction method with physical chemistry-based principle. We have proposed a new attempt in this study, the momentum representation of frontier orbital-based similarity index (MR-FOSI) [1,2], in which assumes that spatial pattern of an outermost orbital molecular orbital governs chemical reactivity. MR-FOSI can be directly related to relative activity of target molecule X with respect to standard reference molecule R and is defined as

$$I_{\mathrm{MR-FOSI}}[\mathrm{R},\mathrm{X}] = \frac{2\int \rho^{\mathrm{R}}(\boldsymbol{p})\,\rho^{\mathrm{X}}(\boldsymbol{p})d\boldsymbol{p}}{\int[\rho^{\mathrm{R}}(\boldsymbol{p})]^2 d\boldsymbol{p} + \int[\rho^{\mathrm{X}}(\boldsymbol{p})]^2 d\boldsymbol{p}}$$

Here, $\rho(\boldsymbol{p})$ denotes the electron density of a frontier orbital wavefunction in momentum space obtained by Fourier transform of the corresponding position-space one. The key behind MR-FOSI is utilization of momentum-space frontier orbital, sensitive to the region far from nuclei governing long-range interaction.

We have so far applied MR-FOSI into several carbamates, in which have a wide range of toxicity strength measured by $\log(1/\mathrm{LD}_{50})$ $(\mathrm{mol/kg})^{-1}$ [3]. LD_{50} is the amount of a material give rise to 50% death of a group of rats in vivo. The position-space wavefunctions are computed in DFT/B3LYP at 6-311G** level. As depicted in Figure 1., MR-FOSI associated with lowest unoccupied molecular orbital (LUMO) reproduce well the relative toxicity between molecules R and X, having same prediction level with QSAR study [4]. In the suggested documents [5], carbamates play as am electron acceptor and hence unoccupied molecular orbitals can thus be a frontier orbital, playing a role in toxicity mechanism. On the other hand, compared with utilization of 29 phyco-chemical properties in the QSAR, MR-FOSI only uses momentum-space frontier orbital as a phyco-chemical property.

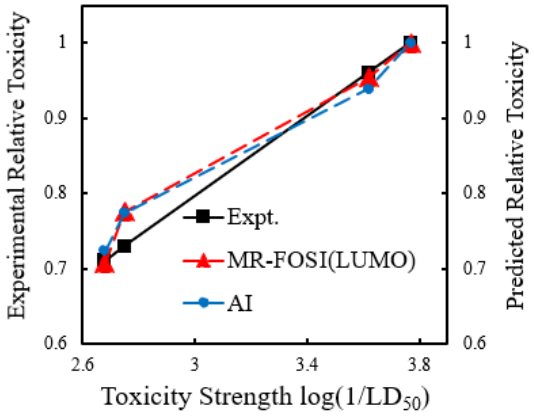

Figure 1. Toxicity prediction in carbamates.

To clarify applicable range of the MR-FOSI, we have also applied it into phospholipid compounds with incomparable anti-HIV activity [6]. In contrast to carbamate study, the MR-FOSI calculation with summation of some outer shell molecular orbitals obtains an excellent anti-HIV activity prediction. In the talk, further analysis of the results will be discussed.

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Intermolecular Coulombic decay in organic dimers

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Inter-atomic/intermolecular Coulombic decay (ICD) serves as a crucial electronic relaxation mechanism following inner-valence ionization of atoms or molecules that possess weakly bound neighbors. Several experimental studies of ICD have been conducted in various atomic and molecular clusters using electron-ion coincidence spectroscopic techniques. It is established that ICD plays a role in mediating radiation damage in biologically relevant molecules. Experimental observations have identified that ICD can instigate alpha-cleavage in hydrated tetrahydrofuran (THF) subsequent to inner valence ionization of the O $2s^{-1}$ orbital in water and THF [1].

In this investigation, we explore intermolecular Coulombic decay (ICD) subsequent to inner valence ionization caused by electron collisions with an energy of 109 eV in thiophene dimers and pyridine-water clusters, which contains the heteroatoms sulfur and nitrogen, respectively. Experimental findings show that the energy liberated during relaxation to the inner valence vacancy is transferred to the adjacent molecule, mainly through ICD, resulting in the ionization of the outer valence orbital of the neighboring molecule. The resulting dicationic state, with charges on both neighboring molecules, instigates the Coulomb explosion of the dimer. Ionic fragments are detected in coincidence with one of the outgoing electrons, employing the 'Reaction microscope,' [2] a multi-particle momentum spectrometer. In the off-line analysis the particles' momentum vectors are reconstructed.

The electron energy loss spectrum observed in the Coulomb explosion of the thiophene dimer exhibits an onset at 20 eV (Fig. 1a). This indicates the generation of an inner valence vacancy, specifically C $2s^{-1}$, initiating the energy transfer process. This spectrum is compared with the energy loss spectrum associated with the monomer inner valence ionization channel, leading to fragmentation and the production of C_3H^+ and $C_2H_2^+$ ions. The striking similarity between these spectra strongly suggests the occurrence of energy transfer through intermolecular Coulombic decay (ICD) to the neighboring weakly bound molecule. The likelihood of an alternative electron transfer process following an Auger decay can be dismissed, as the observed onset is below the double ionization potential where Auger decay typically takes place. Similarly, in the energy loss spectrum for the Coulomb explosion of the pyridine-water dimer (Fig. 1b), which shows an onset at 26.5 eV, a comparable pattern is observed to that of the O 2s inner valence ionization of water, resulting in the production of O^+ ions. A comparison with the theoretical single ionization spectrum for the pyridine-water complex supports the conclusion that an inner-valence vacancy at D_2O (O $2s^{-1}$) in close proximity to this energy range can relaxes through ICD [3]. The lack of analogous monomer inner valence ionization channels in pyridine suggests that the inner valence vacancies in pyridine may decay through the Auger channel. Consequently, the observed intermolecular Coulombic decay (ICD) channel is attributed to the energy transfer from D_2O to pyridine.

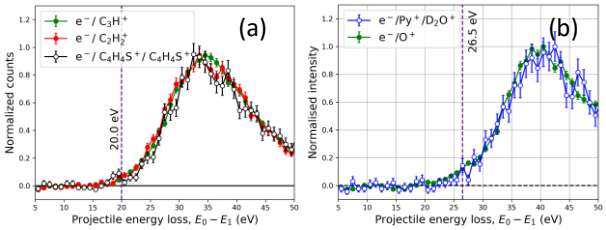

Figure 1. Projectile energy loss spectra: (a) Coulomb explosion of thiophene dimer and C 2s inner valence ionization of thiophene monomer, (b) Coulomb explosion of pyridine-D_2O dimer and O 2s inner valence ionization of D_2O.

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Investigation of the valence-shell excitations of CS_2 by high-energy electron scattering

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Synopsis Absolute generalized oscillator strengths (GOS) of the valence-shell excitations of carbon disulfide (CS_2) have been determined by a joint investigation of a fast electron impact experiment and an equation of motion coupled cluster with singles and doubles (EOM-CCSD) calculations. Moreover, the integral cross sections (ICS) of the valence-shell excited states from their excitation thresholds to 5000 eV have been obtained by means of the BE-scaling method. The oscillator strengths and integral cross-sections complement the fundamental data of carbon disulfide and supply important applications in astrophysics and atmospheric physics.

Carbon disulfide (CS_2) is one of the important species in astrophysical observation, which was identified in the emission spectra of Venus [1] and several comets [2, 3]. Moreover, about 5 tg yr^{-1} of OCS in the earth's atmosphere could be generated from CS_2 [4, 5], and CS_2 is the primary precursor of the atmosphere pollutant SO_2 [6]. Therefore, the dynamic parameters of CS_2, including the optical oscillator strengths (OOS), GOS and ICS, are crucial for modeling the astronomically observed spectra and understanding its evolution in the earth's atmosphere.

In this work, the GOSs of the valence-shell excitations of CS_2 have been determined at an incident electron energy of 1500eV and an energy resolution of 80meV. Besides, The GOSs for the $^1\Delta_u$, $^1\Sigma_u^+$ and $4\sigma_g{}^1\Pi_g$ states were calculated using the equation of motion coupled cluster with single and double excitations (EOM-CCSD), which give a cross-check and help to determine the excitation character. Moreover, the OOSs have been obtained by extrapolating the GOSs to the limit of the zero squared momentum transfer $K^2 \rightarrow 0$, and the ICSs of the valence-shell excitations from the threshold to 5000 eV have been obtained systematically with the aid of the BE scaling method. Present electron energy loss spectrum at 3.5° and generalized oscillator strength density (GOSD) of CS_2 are shown in the Fig.1.

The present reported dynamic parameters of CS_2 can serve as the basics input parameters of the theoretical models and help to explain the astronomical observations.

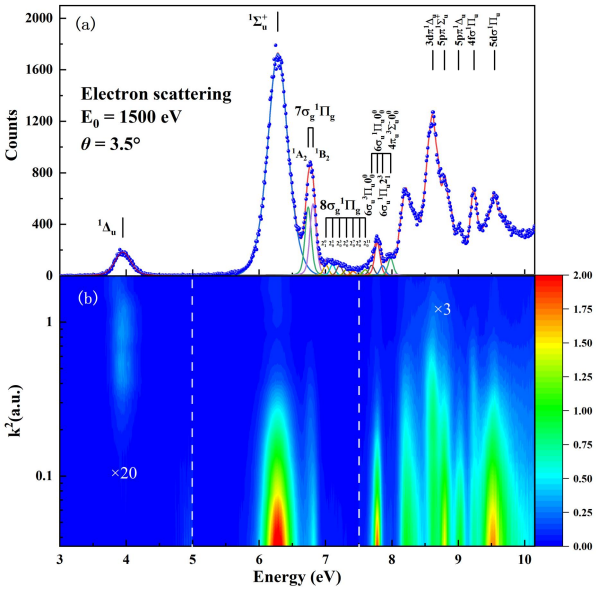

Figure 1. Electron energy loss spectrum at 3.5° and GOSD of CS_2.

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Fragmentation dynamics of formic acid induced by 1 keV electron impact

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The dissociative ionization of organic molecules induced by photons or charged particles involves multiple physical and chemical processes such as bond rearrangement, isomerization, and proton transfer, etc. Formic acid is a typical organic molecule which can form dimers through double hydrogen bonds at room temperature, like base pairs in DNA double strand. Recent researches by intense laser [1-3] have demonstrated that the ionized formic acid undergoes abundant fragmentation processes including bond rearrangement and intramolecular proton transfer. Two competing mechanisms, intermolecular charge transfer and intermolecular Coulombic decay, were found to lead to the dissociation of dimer dication induced by electron impact [4, 5].

In this work, we have studied the fragmentation dynamics of formic acid after double or triple ionizations induced by 1 keV electron impact utilizing an ion momentum imaging spectrometer [6]. In Fig.1, abundant dissociation channels have been identified including neutral hydrogen and oxygen emitted channels. Six two-body dissociation channels for formic acid and its dimer have been investigated, as well as the three- or multi-body fragmentation channels.

The deprotonation $HCOOH^{2+} \rightarrow H^+ + HCO_2^+$ hydroxyl formation channels $HCOOH^{2+} \rightarrow HO^+ + HCO^+$ are the preference two-body dissociation processes with the total branching ratio more than 95%. Apparently, the hydrogen and water formation channels, i.e., $HCOOH^{2+} \rightarrow H_2^+ + CO_2^+$ and $HCOOH^{2+} \rightarrow H_2O^+ + CO^+$, are originated from the proton migration during the Coulomb explosion process. For the deoxygenation channel $HCOOH^{2+} \rightarrow O^+ + H_2CO^+$, the kinetic energy release distribution has two peaks indicating that there may be two dissociation processes including a direct C=O bond cleavage process and a possible proton migration process. For the dimer, we have observed the symmetry dissociation channel $(HCOOH)_2^{2+} \rightarrow HCOOH^+ + HCOOH^+$ while the intermolecular proton transfer channel $(HCOOH)_2^{2+} \rightarrow HCOOH \cdot H^+ + HCOO^+$ only has a few events due to its quite small probability. Information on more dissociation channels will be presented in the poster.-

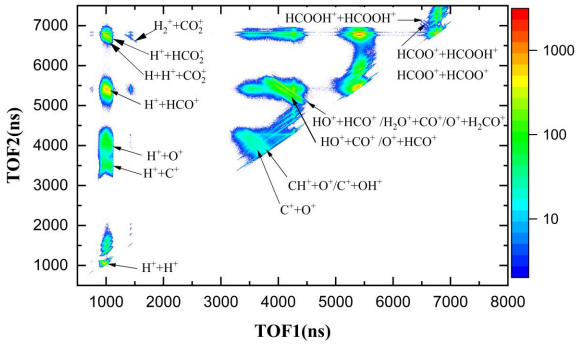

Fig.1 Time of flight (TOF) correlation map of the product ions form the dissociative ionization of formic acid.

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Classical description of the ionization of carbon by electron impacts

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In this work, we calculate the total ionization cross sections of carbon atoms by 10 to 1000 eV electron impact using a three-body classical trajectory Monte Carlo (CTMC) method. The present calculations are restricted to the L-shell due to the large difference in the ionization potentials of the K-shell electrons, whose ioniza tion cross sections are negligible in the energy range explored. Results obtained by means of the simple addition rule of the C(2s) and C(2p) orbitals are shown in Figure 1 and compared to the experimental data of Brook et al [1], and to the theoretical predictions of the generalized oscillator strength formulation of the Born approximation [2], the time-dependent close coupling (TDCC), the R-matrix-with-pseudo-states (RMPS), the time-independent distorted wave method (TIDW) and the B-spline R-matrix with-pseudostates (BSR) [3, 4]. Good agreement is obtained with the data at impact energies greater than about 100 eV. In contrast, electron emission as the threshold region is approached seems to be overestimated. A closer inspection of the classical dynamics at low impact energies reveals the formation of a transient double bound electron system [5]. It is well known that classical two-electron systems are unstable and their resulting dynamics are not expected to be accurately reproduced. In a first approach to the problem, we analyze the results obtained by switching-off the e-e interaction during the transient double bound state. Results are included in Figure 1 and clearly evidence an improvement with respect to the standard CTMC description at the time they highlight the need of further studies.

Acknowledgements
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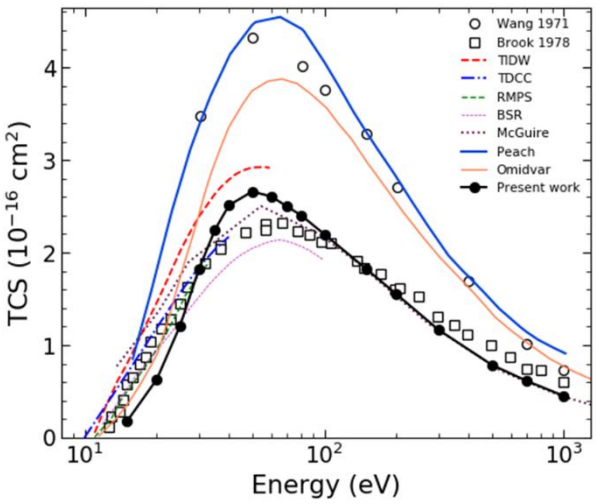

Figure 1. Total ionization cross section as a function of the impact energy for electron-carbon collisions. Expt. data from Ref. [1]. Theories as stated in the text

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Determination of electron inelastic mean free path and stopping power of hafnium dioxide

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With the development of the semiconductor industry, HfO_2 has received much more attention as a representative high-*k* dielectric material which has low leakage current and good thermal stability. For example, due to excellent optical and thermal properties, HfO_2 has emerged as an ideal material for the improvement of absorption efficiency and thermal stability of solar energy. HfO_2 also plays a key role in optical coating materials field for its high transmittance in the ultraviolet to near-infrared band, and high laser damage threshold. Therefore, it is important to know the IMFP and stopping power of HfO_2 especially for the electron microscope analysis of this material. The inelastic mean free path (IMFP) and the stopping power are basic and very important physical quantities describing the transport properties of electrons in materials. For instance, it can help determine the thickness of HfO_2 film which is an important factor affecting the performance of HfO_2 as a semiconductor device or optical element in practical applications.

In this work we present highly accurate electron IMFP and stopping power data of the hafnium dioxide by applying the relativistic dielectric response theory [1]. Our calculations were based on the energy loss function (ELF) of hafnium dioxide derived from the reflection electron energy loss spectroscopy (REELS) spectra.

The probability of the energy loss is determined by the dielectric response function $\varepsilon(q,\omega)$ as a function of the frequency ω and the wavenumber q of the electromagnetic disturbance. The full Penn algorithm (FPA) [2] was used to expand the optical energy loss function, $\mathrm{Im}\{-1/\varepsilon(\omega)\}$, into the (q,ω) -plane. Furthermore, we also applied the super-extended Mermin algorithm [3] to calculate the IMFP and stopping power for comparison with the results of the FPA model.

We have shown that the FPA and SMA results are almost identical at high electron kinetic energies above ~70 eV. However, they differ significantly below 70 eV. We have shown that the bandgap value has a significant influence on IMFP and stopping power data. Therefore, for compound semiconductors, the bandgap must be considered in order to obtain accurate IMFP and stopping power data for applications.

Acknowledgements

This research was supported by the Education Ministry through "111 Project 2.0" (BP0719016), and by European Cost Actions CA18212 (MDgas). This work was also supported by the National Institute for Materials Science under the Support system for curiosity-driven research, JSPS KAKENHI Grant Number JP21K14656 and Grant for Basic Science Research Projects from The Sumitomo Foundation. The work was also support by the by the Bilateral Relationships between China and Hungary in Science and Technology (S&T) under the project number 2021-1.2.4-TÉT-2021-00055.

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Energy loss function of samarium determined from the reflection electron energy loss spectroscopy spectra

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Lanthanides have nowadays become of vital importance in advanced materials and technology. Applications in laser science, solar cells, fluorescent lamps and a new organic light-emitting diodes components, as well as luminescent probes are strongly related with their optical and/or electronic properties.

Samarium and its compounds are among the most frequently used lanthanides in the investigations during the last years. But the precise excitation property, especially the plasmon structure of samarium is still not known. It is not surprising because all lanthanides are highly reactive elements and interact strongly with oxygen and hydrogen. So, experimentally it is really a challenging to obtain accurate results. The excitation properties are intrinsically embodied in the energy loss function (ELF), $\mathrm{Im}\left\{-1/\varepsilon(\omega)\right\}$, which is clearly related with the frequency dependent complex dielectric function $\varepsilon(\omega)$. Da et al. [1] developed a reverse Monte Carlo (RMC) method for the derivation of the ELF and thereby the dielectric function and optical constants of solids in a much wider photon energy range than that of the usual optical measurements. The RMC method combines a Monte Carlo modelling of electron transportation for REELS spectrum simulation with a Markov chain Monte Carlo calculation of parameterized ELF.

Our RMC technique was used to obtain the electron energy loss features buried in the REELS spectra of samarium [2]. The accuracy of the ELF was checked by applying the Thomas-Ritchie-Kuhn (*f*-sum rule) and the perfect-screening sum rules (*ps*-sum rule). We found that the *ps*- and *f*-sum rules with our ELF, imaginary part of the complex dielectric function ε_2, and extinction coefficient k fulfils very accurately and reach the nominal values with 0.2% and 2.5% accuracy, respectively. We were able to separate the contribution from the bulk and the surface excitations. We show the detailed excitation characteristic in the optical data in the energy range between 3 and 60 eV. We found a surface plasmon mode at 10 .4 eV and the corresponding bulk mode 14.3 eV.

Acknowledgements

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Impact parameter and kinematic information for differential ionization of argon by positron and electron impacts

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It is well known that the reversal of directions of the Coulomb field for positron and electron impact ionization produces trajectory changes for the projectile. Pre-collision field effects result in a larger impact parameter for positron impact with respect to that for electron impact. In addition, post-collision effects between the post-collision particles introduce additional trajectory effects. In order to test various theoretical models for ionization, comparisons on the differential level are used. For electron impact, a large amount of experimental information, ranging from singly to fully differential, is available whereas for positron impact, relatively few differential studies have been performed. Also, unlike the case for heavy ion impact where impact parameter information can be obtained, post-collision effects effectively prohibit such studies for lepton impact.

However, the Classical Trajectory Monte Carlo method allows us to investigate such kinematic effects and also to obtain information as a function of impact parameter. As an initial study, such information for ionization of argon 3p electrons by 1 keV positrons and electrons is presented. Similar to that used by Sparrow and Olson [1], the argon atom was modeled as a single 3p electron and a central core potential and interactions between all particle pairs are taken into account. Unlike previous studies, the present study also provides information about the impact parameter and the scattering and ejection directions, not just the angles but also whether the directions are "positive" or "negative", i.e., toward, or away from the central core.

We have presented studies of the ionization cross sections in collisions between electron and positron impact with Ar(3p) target. The calculations were performed classically using the three body CTMC approximation. We found that our present CTMC model, where the target atoms were described within the single active electron approximation, describes reasonably well the ionization cross sections and agrees with existing experimental data. We have shown that the energy distributions, both for electron and positron impact, have the same shape and structure. At the same time, the angular distributions behave completely different which we suggest is associated with a projectile-target core interaction. The ionization probabilities as a function of impact parameter were also presented. We found different impact parameter probability distributions. For the case of positron impact the distribution is symmetric, for the case of electron impact the distribution is asymmetric.

Acknowledgments

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Three-body fragmentation dynamics of $C_6H_6^{3+}$ induced by electron-impact ionization

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Six-membered aromatic rings as the basic unit for building a variety of compounds play an important role in biochemistry, interstellar medium and astrophysics [1]. When these molecules are ionized, more complex ring-opening reactions can be initiated, such as successive ring-breaking and proton emission. As an archetypal aromatic molecule, the ionization and subsequent dissociation of benzene (C_6H_6) can produce various isomers, which have been discovered to exist in the interstellar medium, and such radical ions may be an important source for many smaller carbonaceous fragments in space [2]. Therefore, understanding of the dissociation dynamics of benzene cations is of significant interest for elucidating how these radical species arise.

In the present work, we reported a joint experimental and theoretical study of the three-body fragmentation dynamics of $C_6H_6^{3+}$ induced by 200 eV electron-impact ionization. The experiments were performed using a reaction microscope which was particularly designed for electron-collision experiments [3,4]. The fragment ions are detected in coincidence, the complete three-body fragmentation channels are identified in the two-dimensional time-of-flight (TOF) coincidence map (see Figure 1). They are described as

(1) $C_6H_6^{3+} \longrightarrow H^+ + C_3H_2^+ + C_3H_3^+$

(2) $C_6H_6^{3+} \longrightarrow H^+ + C_2H_3^+ + C_4H_2^+$

(3) $C_6H_6^{3+} \longrightarrow H^+ + C_2H_2^+ + C_4H_3^+$

The formation of three channels all involves the ring-opening reactions and the breaking of C-H bonds. Combining the momentum correlation maps between fragment ions, Dalitz plots, Newton diagrams, and ab initio molecular dynamics simulation, we analysis the fragmentation dynamics and provide a detailed picture of dissociation mechanisms of three channels.

The dissociation processes in all three channels include two sequential dissociation mechanisms, corresponding to proton emission in the first and second steps, respectively. The kinetic energies of the protons from two sequential pathways are different. The molecular dynamics simulations show that these processes all involve ring-opening reactions in the first step, then the trication undergoes two-step Coulomb explosion and forms three cations. The mechanisms revealed in our study provide detailed information on the ring-opening fragmentation dynamics of the triply ionized benzene molecule induced by electron impact and thus expand our understanding of radiation chemistry and biology.

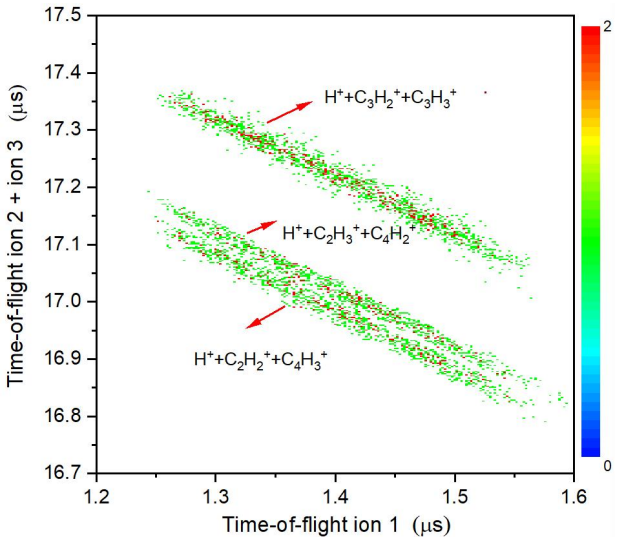

Figure 1. The triple-ion coincidence time-of-flight coincidence map induced by 200 eV electron-impact ionization. The TOF of the first detected recoil ion was plotted along the horizontal axis and the sum of TOF of the second and third ions along the vertical axis.

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Molecular ionization dissociation of the $ArCH_4$ dimer by electron impact

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Interatomic Coulombic decay (ICD) is an important energy transfer process, where the excess energy from the de-excitation of the excited atom (or ion) A within a cluster may be transferred to the neighboring atom (or molecule) B and induces its ionization. ICD generally exists in weakly bound systems, such as the van der Waals cluster, the hydrogen bond cluster, and the solution [1], it acts as a powerful tool to enhance the ionization cross-section, to detect the chemical environment of an ion, and to investigate the radiation damage on the molecular level.

It has been widely accepted that ICD-induced molecular fragmentation occurs through a two-step process, involving ICD as the first step and dissociative-electron-attachment (DEA) as the second step. However, the theoretical calculations by Cederbaum [2] and Chiang et al [3] recently proposed a one-step mechanism, where ICD directly causes the dissociation of a molecule with high efficiency. If this mechanism is validated, the fragmentation efficiency induced by ICD will increase significantly.

Selecting the $ArCH_4$ dimer as a prototype system, we perform the fragmentation experiment by employing a transversal reaction microscope at Institute of Modern Physics, Chinese Academy of Sciences. By detecting two ions and one electron in coincidence, we can reconstruct the kinetic energy release (KER) and electron energy spectrum (EES) after $ArCH_4$ dissociation, and then investigate the corresponding fragmentation dynamics [4].

As shown in Figure 1, besides the well-known ionization pathways induced by ICD producing the Ar^+/CH_4^+ ion pair(channel A), a coincident island (channel B) representing the Ar^+/CH_3^+ ion pair is also observed. Through the comparison of the KER and EES of channel B with that of channel A, we demonstrate that channel B is from the ionization dissociation reaction induced by ICD. More details, after the Ar atom is simultaneously ionized and excited, its excess energy is transferred to CH_4, populating the CH_4^+ into excited vibrational states. The molecular ion dissociates subsequently into the CH_3^+/H ion pair. The yield of channel B is 1.7 times larger than that of channel A, indicating it is of high efficiency[4].

On the one hand, via this new pathway, it will be possible to construct an antenna-receiver complex to enhance the cross-section of the molecular ionization dissociation by at least one order.

On the other hand, this new decay pathway offers a novel approach to directly break covalent bonds in DNA molecules, bypassing the processes involving DEA. It has potential applications in developing new radiotherapy.

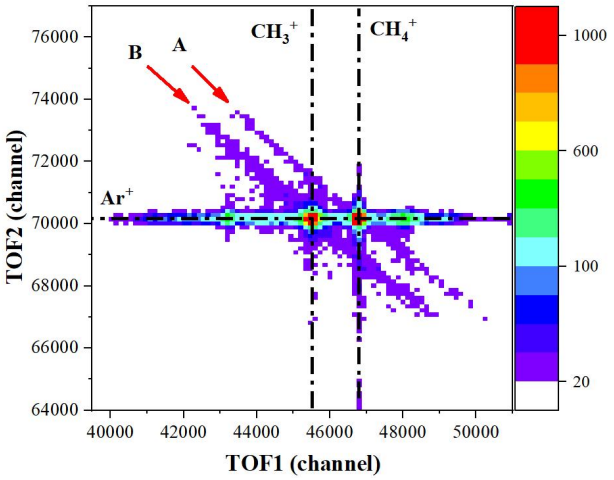

Figure 1. Two-dimensional TOF correlation map. The arrow marked with A is the ionization pathway inducing the Ar^+/CH_4^+ ion pair, while the arrow marked with B is the ionization dissociation pathway inducing the Ar^+/CH_3^+ ion pair.

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Fragmentation dynamics of $BrCN^{q+}$ (q = 2-6) induced by 1-keV electron impact

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Fragmentation dynamics of multiply charged polyatomic molecules have drawn continuous interest in the past decades [1-6]. It was found that the chemical bonds of the multiply charged polyatomic molecules break either through non-sequential or sequential processes. Identification and elucidation of the fragmentation processes are crucial for the understanding of multi-body quantum correlation dynamics. One of major issues is to identify the dissociation process and to understand its mechanism.

In this work, the fragmentation dynamics of $BrCN^{q+}$ (q = 2-6) induced by 1-keV electron impacts were studied by employing an ion momentum imaging spectrometer. [7-8] Six two-body and eleven three-body fragmentation channels were identified. The pathways of the two-body fragmentations are analyzed with the assistance of the potential energy curves (PECs) calculated by the complete active space self-consistent field (CASSCF) method, while the fragmentation mechanisms of three-body fragmentation channels are clarified using the Dalitz plot, Newton diagram, and native frame method. The sequential fragmentation processes are observed i.e., $BrCN^{3+} \rightarrow N^+ + BrC^{2+} \rightarrow Br^+ + C^+ + N^+$, and $BrCN^{m+} \rightarrow Br^{l+} + CN^{2+} \rightarrow Br^{l+} + C^+ + N^+$ ($m = l + 2$ and l = 1-3). The kinetic energy release (KER) for the intermediate dications, BrC^{2+} and CN^{2+}, from the sequential mechanism, are determined and the electronic states of the intermediate molecular ion CN^{2+} are discussed. For the channels leading to higher charge states of the light elements, only the concerted fragmentation mechanism is observed. For the concerted fragmentation of all the three-body fragmentation channels, the experimental KERs are lower than the predictions by the Coulomb explosion model. The reason may be ascribed to the ultrafast dissociation and electron transfer at the very beginning of the dissociation which results in a prolonged internuclear distance compared to the equilibrium geometry.

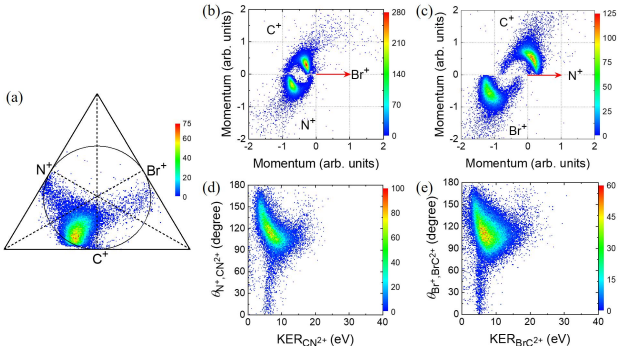

Figure 1. Experimental results for channel $Br^+ + C^+ + N^+$. (a) Dalitz plot, (b) and (c) Newton diagrams with Br^+ and N^+ as references, respectively. (d) and (e) Native frame plots assuming sequential breakup of $BrCN^{3+}$ with CN^{2+} and BrC^{2+} as the molecular intermediates, respectively.

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The structural measurement of heteroaromatic thiophene dimer by ultrafast electron collision ionization

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An accurate account of the non-covalent interactions between molecular species involving heteroaromatic moieties, for example, thiophene, is paramount to a range of fundamental science and cutting-edge applications. The characteristic poly-conjugated nature of thiophene polymer allows the intermolecular delocalization of p-electrons resulting in the reduction of the band-gap, which determines its electronic and nonlinear optical properties. Knowledge of the structural aspects of thiophene aggregations dictates how molecules arrange around one another and sheds light on the factors influencing electronic overlap that are critical to electronic materials' functionality.

In this work, we focus on the structural studies of thiophene dimer by Coulomb explosion. Coincidence momentum imaging provides a kinematically complete description of the Coulomb explosion process, whereby the three-dimensional momentum vectors of all product ions resulting from a single parent ion are determined [1]. The dynamical information is closely related to the positions of charges and nuclei prior to the dissociation in the molecular frame, which enables us to deduce the initial configuration information of the molecular systems. It is worth noting that the nuclei must be ``frozen" before the Coulomb explosion to prevent structural deformation, which requires an ultrafast ionization process.

One effective method of achieving ultrafast ionization is electron collision, where the outer-valence electrons of molecular clusters can be quickly stripped off by two different mechanisms. One involves the sequential ionization (SI) of an outer valence electron from each molecule. The other pathway involves inner-valence ionization, followed by ultrafast radiationless energy transfer between the excited species and its neighbor. The energy transferred leads to the emission of one electron of the neutral neighbor, which is known as intermolecular Coulombic decay (ICD) [2]. Both ionization processes proceed on the femto-second timescale, during which the slower nucleus motion of the molecular complexes can be neglected.

Here, we clarified the minimum energy configuration of the thiophene dimer through a Coulomb explosion induced by electron impact (200 eV) [3]. The measured kinetic energy release (KER) of the Coulomb explosion channel is compared with the ab initio calculations of three different conformers of thiophene dimers (see Figure 1). The consistency between calculated results of different initial configuration and measured KER reveals the most stable structure of the supersonic cold gas-phase thiophene dimer, and the result is further supported by the measured PEC of the thiophene dimer.

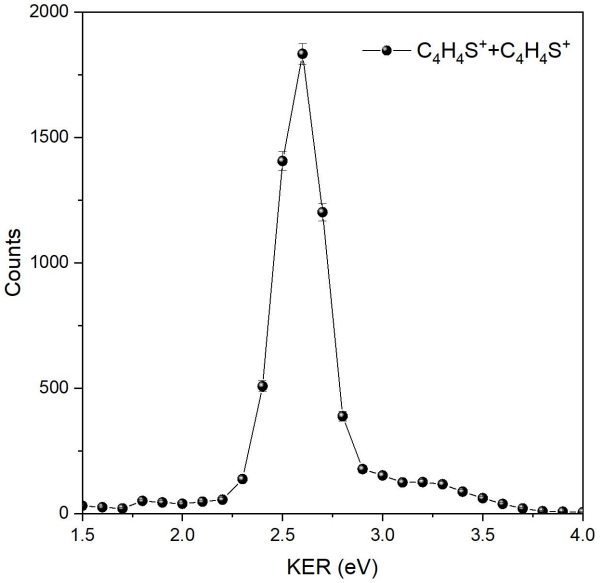

Figure 1. Measured and calculated KER of $C_4H_4S^+$+$C_4H_4S^+$ fragmentation channel.

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(e, e + ion) study on electron-induced dissociative ionization of CO

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Electron-induced dissociative ionization of molecules is a fundamental process in molecular physics. As a typical heteronuclear diatomic molecule, studies on such process of CO have been widely conducted [1-4], and most of them are limited to measure and analysis kinetic energy release of fragmentation ions, without distinguishing the dication states. The complex electronic states of CO^+ and CO^{2+} near the double ionization threshold gives rise to three mechanisms of produce of $C^+ + O^+$: autoionization of O^* followed by dissociation of CO^{+*}, predissociation mainly from $X^3\Pi$, $a^1\Sigma^+$ and $b^1\Pi$ states of CO^{2+}, and direct dissociation mainly from $^3\Sigma$, $c^1\Delta$ and $2^3\Pi$ states of CO^{2+} [5]. To investigate these three mechanisms, a state selected dissociative ionization experiment has to be performed.

In this work, electron-induced dissociative ionization of CO has been investigated by employing an (e, e + ion) spectroscopy. The apparatus consists of an electron gun, an energy-dispersive electron spectrometer, and an ion momentum imaging spectrometer. An electron beam produced by the electron gun is chopped at a frequency of 100 kHz by applying rectangular voltage pulses to a deflector electrode. Electrons scattered by an effusive molecular beam at angle of 5° with respect to the incident beam direction are decelerated by electrostatic lens subsequently dispersed by a hemispherical analyzer and detected by a one-dimensional position-sensitive detector. Triggered by detection of scattered electron, a pulsed electric field is applied to the scattering region to push fragment ions towards the momentum imaging spectrometer. The fragment ions are analyzed by a Wiley-Mclaren type time-of-flight mass spectrometer followed by a two-dimensional time- and position-sensitive detector. The momenta of ions are then determined from its time of flight and arrival positions at the detector. By selecting different electron energy loss regions, as shown in Fig. 1(a), three mechanisms of produce of $C^+ + O^+$ had been observed appearing successively near the double ionization threshold, and they can be distinguished by the kinetic energy release distributions. Noticeable difference from the angular distributions of C^+ ions relative to the momentum transfer direction for different energy loss regions has also been observed.

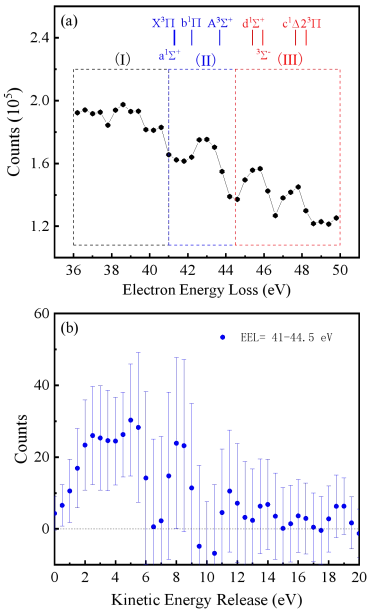

Figure 1. (a) Electron energy loss spectrum measured at 5° scattered angle by 648eV electrons impacts, vertical bars indicate relevant vertical ionization potentials reported in Ref. [5]. (b) Kinetic energy release distributions of $C^+ + O^+$ from region(II) in (a).

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Pump-probe investigation of photo-induced ring-opening of 1,3-cyclohexadiene by electron momentum spectroscopy

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The photo-induced ring-opening of 1,3-cyclohexadiene (CHD) to 1,3,5-hexatriene (HT) is a textbook electrocyclic reaction in organic chemistry. It acts as a model for photobiological synthesis of vitamin D in the skin [1]. This ring-opening reaction has been an issue of many experimental studies over the years [2-4]. As the CHD is pumped from the ground state (S_0) to the excited state (S_1), as shown in Fig.1(a), it may pass the conical intersection through two pathways: one is back to the CHD ground state, while the other undergoes the ring-opening and leads to the open-chain isomer with a quantum yield of about 0.3[5]. The nuclear motions were directly observed by the ultrafast X-ray scattering [2] as well as the ultrafast electron diffraction [3], while the recent time-resolved photoelectron spectroscopy study have explored the evolution of excitation-deexcitation process from energy standpoint [4].

In the present work, an experiment on CHD was carried out using time-resolved electron momentum spectrometer (TREMS) [6]. The ground state (S_0) of CHD was pumped to the excited state (S_1) by 267 nm laser pulses with 5 kHz repetition rate. An 800 eV pulsed electron beam (10kHz repetition rate) is used to ionize the target. The repetition rate of the pump laser is half of the pulsed electron beam, therefore the electron density as a function of binding energy and momentum of the target electron are measured both with laser on and off. By subtracting the laser-on data from that of laser-off, the difference of the electron density between CHD and HT can be obtained. Fig.1 (b) shows the difference in binding energy spectra(BES), and Fig.1(d) shows the difference of the summed electron momentum distributions (EMDs) over the binding energy range from 5 -17 eV covering all the outer valence orbitals. We also performed the theoretical calculations by SAC-CI method, the summed EMDs of all the outer valence orbitals for CHD and HT are shown in Fig.1(c). The theoretical results of the difference in BES and EMDs are plotted in Fig.1 (b)(d) for comparison. The agreement between experiment and theory clearly reveals the direct observation of the electron density change in photo-induced ring-opening of CHD to HT. The details will be presented in the poster.

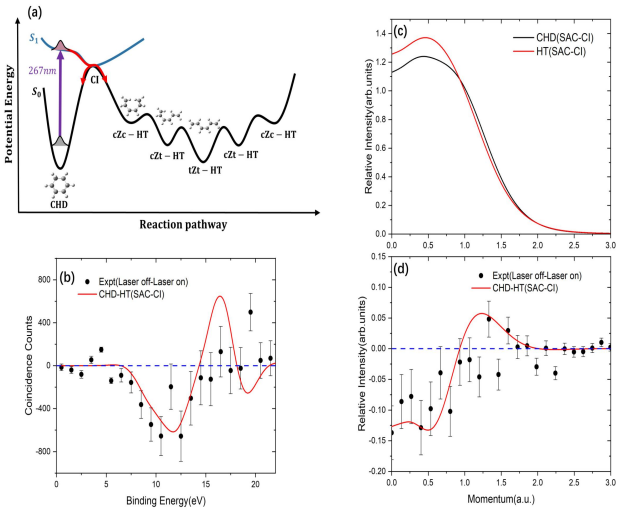

Figure 1. (a) Schematic of the photo-induced ring-opening reaction of CHD. (b) The difference in binding energy spectra between CHD and HT. (c) The summed electron momentum distributions for all the outer valence orbitals of CHD and HT calculated by SAC-CI. (d) The difference of the summed electron momentum distributions of CHD and HT over the binding energy range from 5 -17 eV.

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First Experiments with the CSR-ReMi, the Reaction Microscope in the cryogenic storage ring CSR

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The CSR-ReMi is a dedicated in-ring reaction microscope for the cryogenic storage ring CSR at the MPIK in Heidelberg [1]. It was designed to perform experiments on the cold atomic, molecular or cluster ions stored in the CSR.

A reaction microscope (ReMi) [2, 3] is a combined electron and ion spectrometer. Superimposed electric and magnetic fields guide both electrons and ions to time- and position-sensitive detectors. For the CSR-ReMi, multi-channel plate detectors with delay line readout are used, because they are multi-hit capable and provide high detection efficiency, acceptance and resolution. This enables the energy- and angular-resolved detection of charged particles that are created in single collision events. Through the coincident detection of all collision fragments kinematically complete data-sets on the reaction dynamics can be collected. In addition to the electron and recoil ion detector the CSR-ReMi is equipped with a projectile detector, which is located downstream after the CSR bend section and can detect neutralized particles from the ion beam.

The integration of the CSR-ReMi into the CSR was finalized in July 2023. Thereafter, the CSR was cooled down to cryogenic temperature (<15 K) for the autumn beamtime 2023. The CSR-ReMi was successfully commissioned and first experiments were performed under cryogenic conditions in November 2023. In the commissioning experiments emphasis was given to electron-transfer and electron-loss reactions in collisions of the stored ion beam with neutral atoms or molecules provided by a supersonic gas jet. As an example, in Figure 1 the ion detector image is shown for the collision of a 30 keV Ar^+ ion beam on Ar atoms. We identified the three visible lines with electron capture into ground and excited states of argon. By coincident detection of the neutralized projectiles out of the ion beam we additionally get time-of-flight information on the recoil ions.

This contribution will present this and other selected results from the commissioning beamtime.

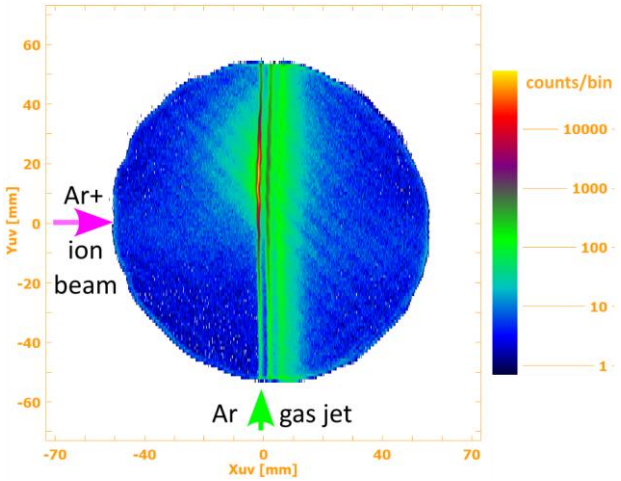

Figure 1. Ion detector image for collisions of 30 keV Ar^+ ions on Ar. Directions of the Ar^+ ion beam (magenta) and the Ar target gas jet (green) are indicated by the arrows. The distribution is offset relative to the marked ion beam position because of the initial velocity of the gas jet.

The commissioning of the laser incoupling line of the CSR-ReMi is planned for March 2024 with the intention to perform experiments on photo-detachment with atomic and molecular anions.

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Wave-packet convergent close-coupling approach to ion-atom collisions: progress update

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Recent advances in experimental techniques have resulted in detailed and highly accurate kinematically complete measurements of electron emission in ion-atom and ion-molecule collisions. However, theory lags far behind and cannot describe the experiments on fully differential ionisation. Even the description of experimental data on energy and angular distributions of electrons ejected in intermediate-energy ion collisions with simple atomic and molecular targets has remained an insurmountable problem for over five decades. We have developed a two-centre continuum-discretised coupled-channel method that provides the first accurate solution to the problem [1-4]. The method has been applied to calculate the doubly differential cross sections for ionisation in proton collisions with He and H_2 in a wide range of projectile energies. In particular, we presented (i) energy and angular distribution of emitted electrons [1,4] and (ii) energy distribution of electrons as a function of scattered-projectile angle [2,3]. Excellent agreement between the obtained results and the experimental data is found indicating the importance of capture into the continuum in the dynamics of ionisation.

The approach known as the wave-packet convergent close-coupling (WP-CCC) method is based on expansion of the total scattering wave function using a two-centre pseudostate basis. This allows one to take into account all underlying processes, namely, direct scattering and ionisation, and electron capture into bound and continuum states of the projectile. The normalised wave packets constructed from the Coulomb waves are used to discretise the continuous spectrum of the target and projectile atoms. This has a particular advantage in calculating differential ionisation cross sections since the discretisation of the continuum can be as dense as necessary and the distribution of the resulting wave packets can be arbitrary to cover the most important regions.

The method is also capable of providing benchmark data on the total and state-selective cross sections for various processes taking place when fully stripped ions collide with atomic and molecular targets [5,6]. Furthermore, we have developed a new method to treat collisions involving multielectron targets. The method allows one to reduce the multi-electron target to an effectively single-electron one and then use the standard WP-CCC approach. The method has been used to calculate integrated charge-exchange and ionisation cross sections for proton collisions with alkalis. Obtained results are in very good agreement with experiment available for charge exchange. The method has since been extended to collisions of dressed ions with atomic hydrogen.

We report on current progress in applications of the WP-CCC approach to collisions of various fully and partially stripped ions with H, He and H_2.

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State-selective electron capture in Ar^{16+}–H($1s$) collisions for charge-exchange recombination spectroscopy

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A number of ions are being considered as the primary seeding impurities for future fusion reactors [1]. Impurity seeding will be required for machines such as ITER or DEMO for radiative cooling, plasma control and diagnostics. For instance, the seeding of impurity ions will help maintain the temperature in the diverter region through radiative cooling. Charge-exchange recombination spectroscopy (CXRS) is a diagnostic technique capable of measuring the impurity ion density, ion temperature and plasma rotation within the fusion plasma including ITER [2, 3]. Measurements of the helium ash density, which can be used to determine the effectiveness of the fusion reaction, remains a major challenge. The CXRS technique requires accurate state-resolved electron-capture cross sections for collisions between the seeded impurity ion and the neutral hydrogen beam injected during diagnostics.

In the case of argon seeding, Ar^{16+} is found to be the most important ion for the present fusion devices operating in the temperature range of 1–5 keV as it is present within most of the plasma, even away from the centre of the tokamak. For this projectile, capture into states with a final-state principal quantum number, $n = 14$ and 15 is found to be the most important to consider. This is because the transitions from $n = 16 - 14$ of Ar^{16+} or Ar^{15+} are in the visible spectrum, which is well suited for CXRS diagnostics. Experimental measurements for this collision system are not readily available in the literature. The classical trajectory Monte Carlo (CTMC) method has been employed by Errea *et al.* [4] and Schultz *et al.* [5] to calculate state-selective cross sections for capture into the $n = 14$ and 15 states. However, their results differ by over an order of magnitude at the low energies. In addition, their cross sections peak at different energies. McDermott *et al.* [2] have used the CTMC method to calculate the density profiles of the seeded argon impurities. They reported that the data by Errea *et al.* [4] underestimate the density profile whilst those by Schultz *et al.* [5] are larger by over an order of magnitude. In contrast to this, the X-ray measurements of Schlummer *et al.* [6] reported better agreement with the Schultz *et al.* data [5] on the intensities for high $1snp - 1s^2$ transitions in He-like argon.

We look into this problem using the wave-packet convergent close-coupling approach. In the past, we have modelled electron capture in Ne^{10+}–H($1s$) collisions [7]. We found that in collisions of highly charged ions with hydrogen, capture into states with n significantly larger than the charge of the projectile still makes a non-negligible contribution to the total electron-capture cross section. Therefore, the calculations become computationally very demanding due to the size of the basis required to get convergent results. The calculations for Ar^{16+} are even more challenging because the cross sections for electron capture into the $n = 14$ and 15 states are several orders of magnitude smaller than the total electron-capture cross section. Thus, it is extremely difficult to calculate these cross sections accurately. We report our results which shed light on the situation.

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Three-body fragmentation dynamics of cyclopropane trication following 5.8 MeV/u Ni^{19+} impact

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Fragmentation dynamics of molecules have received much attention in recent decades, not only because of their fundamental importance but also because of their applications in many fields. Among the molecules investigated in the past, the hydrocarbon molecules, which widely exist in the interstellar medium and play an important role in industry, have attracted special interest. While the chain-structure hydrocarbon molecules have been the focus of most of the previous work [1-3], the three-body fragmentation of the ring-structure hydrocarbon molecules has been reported only for a few cases [4, 5].

In this work, the three-body fragmentation dynamics of cyclopropane induced by 5.8 MeV/u Ni^{19+} ion collisions were studied using a cold target recoil ion momentum spectroscopy (COLTRIMS). Two three-body dissociation channels including deprotonation channel $C_3H_6^{3+} \rightarrow H^+ + CH_2^+ + C_2H_3^+$ and isomerization channel $C_3H_6^{3+} \rightarrow H^+ + CH_3^+ + C_2H_2^+$ were identified. The Dalitz plots, Newton diagrams and the kinetic energy release (KER) distribution are used to study the dissociation mechanisms.

For the channel $C_3H_6^{3+} \rightarrow H^+ + CH_2^+ + C_2H_3^+$, two oblique stripe structures marked by red and gray dashed rectangle are observed in the experimental Dalitz plot shown in Fig. 1(a), implying that different fragmentation mechanisms may be involved in this channel. In Fig. 1(b), we show the Newton diagram normalized to the momentum of H^+ (black arrow). A clear circular structure marked by red lines exhibits in the Newton diagram, which is the evidence of a sequential fragmentation process via $C_3H_6^{3+} \rightarrow H^+ + C_3H_5^{2+} \rightarrow H^+ + CH_2^+ + C_2H_3^+$. There is another weak circular structure marked by gray dashed semicircles in the Newton diagram, indicating that a different sequential fragmentation process occurs in this channel.

The Newton diagrams and the Dalitz plots for another channel $C_3H_6^{3+} \rightarrow H^+ + CH_3^+ + C_2H_2^+$ also displays similar patterns (not shown here), implying that the dominant dissociation dynamics for isomerization channel might be similar for the deprotonation channel, i.e., only two sequential fragmentation pathways are observed for both the two channels. Our preliminary analysis suggests that the dominance of the sequential fragmentation processes may be attributed to the projectile velocity which leads to different electron lost mechanism of molecule target.

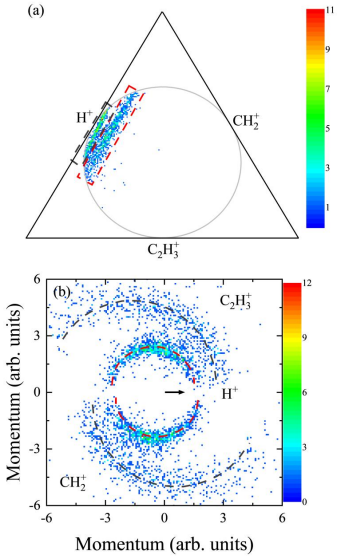

Figure 1. Experimental results for channel $C_3H_6^{3+} \rightarrow H^+ + CH_2^+ + C_2H_3^+$ (a) Dalitz plot and (b) Newton diagram.

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The TrapREMI: A reaction microscope inside an electrostatic ion trap for collision measurements

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We present an overview of the TrapREMI, a novel experimental setup designed to study atomic and molecular ion reactions crossed with various projectile beams. We report the initial experiments conducted with the TrapREMI focused on atomic collision dynamics.

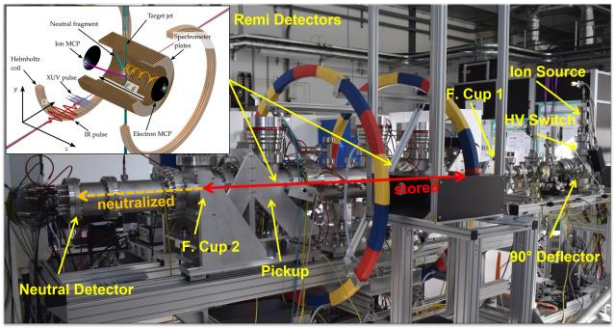

Figure 1. Image of the TrapREMI setup. A EIBT beam-transport line follows the ion source. The REMI comprises several spectrometer electrodes, three detector modules, and a pair of Helmholtz coils, see inset. In the TrapREMI, the ion beam is brought to interact with IR laser pulses or a neutral jet. The overall setup is about 5 m long and 2 m in height.

The apparatus, detailed in Figure 1 and introduced in [1], integrates an electrostatic ion beam trap (EIBT) [2,3] with a reaction microscope (REMI) [4,5]. This combined setup allows for the storage of a bunch of hot atomic or molecular ions crossed with an atomic gas jet or a laser pulse. This unique combination facilitates coincident measurements of the resulting reaction products (ions, electrons, and neutrals) and the subsequent reconstruction of their momentum vectors. First, an ion bunch is injected into the EIBT via a radio-frequency ion source. The ion bunch is stored in an oscillatory motion under UHV for up to several milliseconds (Fig. 2a). The ions oscillate through the REMI and interact with the gas jet at the center of the detectors (see inset Fig. 1). The charged products are then guided by an extraction field toward the time-position-sensitive detectors of the REMI while the neutral products are detected at the end of the EIBT.

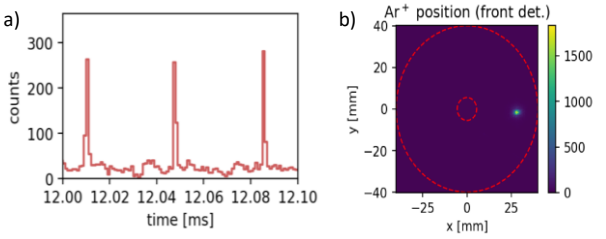

Figure 1. a) Stored ion bunch signal. b) Momenta distribution of the recoil Ar ion after the Ar^+-Ar collision. The ion bunch travels across the small red circle. The gas jet direction is from left to right.

Initial experiments investigated the collision dynamics of stored fast argon ions (2-3 keV) as projectiles incident on various atomic and molecular targets. First observations indicated that the singly-charged ions capture electrons from the neutral gas beam. These measurements performed with the stored ion beam showed less background signal compared to a continuous beam without storage (Fig. 2b). Ion/neutral-coincidence measurements of reaction products from Ar^+-Ar, Ar^+-N_2, and Ar^+-Ne were performed to validate the effectiveness of the new setup.

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Study of single electron capture in O^{6+} + He collisions

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Electron capture (EC), as an indispensable collision mechanism when impact velocities are less than approximately one a.u., has received close attention from both fundamental physics and applied physics for nearly half a century. Propelled by development of ion source technology, heavier and higher-charged ions are being utilized in collision experiments. In this work, we have experimentally and theoretically studied the cross sections of single electron capture (SEC) in collisions between O^{6+} and Helium, which is a significant system in various plasma environments such as interstellar medium or tokamak [1, 2]. The results of high-resolved cross-section study can provide fundamental database for fine spectral analysis and promote the progress of theory.

The measurement is carried out based on the Highly Charged Ions Collision Platform at Fudan University constructed with a 14.5 GHz electron cyclotron resonance (ECR) ion source. Absolute total and state-selective cross sections are achieved for the removal electron captured into $O^{5+}(1s^2nl)$ from $He(1s^2)$ in the energy range of 2.625 – 37.5 keV/u by a cold target recoil ion momentum spectroscopy (COLTRIMS). A two-active-electron semi classical asymptotic-state close-coupling method (SCASCC) is also applied in the respective calculations. The detailed scheme of this study can be found in our published article [3].

Figure 1 shows the partial results of this work. The present measured and calculated state-selective SEC cross sections as a function of impact energy are plotted together with other published data. As for $n = 3$ and $n = 5$ channels, the present calculation achieves better agreement with experimental ones. However, an obvious divergence between two pairs of measured and calculated results appears below 10 keV/u. In the energy region we have measured, $n = 3$ is the dominant capture channel, while as the impact energy increases, the removal electron will be more likely to stay in higher excited states of projectile, which gives a hint that large basis sets involving more high energy states are needed in calculations in order to obtain more accurate results, especially for weak channels.

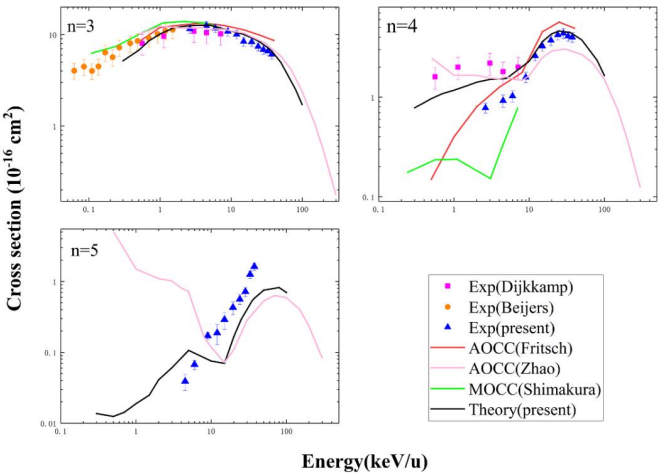

Figure 1. The SEC cross sections as a function of the impact energy. Experiments: present (blue triangles); Dijkkamp et al [4] (pink squares); Beijers et al [5] (orange circles). Theory: present (black line); Fritsch and Lin [6] (red line); Shimakura et al [7] (green line); Zhao et al [8] (pink line).

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Calculation of isotope shift electronic factors for neutral Tl and Ag atoms within the relativistic and high-order electronic correlation effects framework

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The study of isotope shifts in the energies of electronic transitions in atoms can provide a reliable source of information about one of the most important properties of the nucleus — the root-mean-square (rms) charge radius[1]. Currently, there is a large quantity of results on isotope shifts in various atomic systems obtained in the experiment. In particular, similar experiments for various electronic states were carried out for the thallium atom[2, 3]. However, to determine the difference in the rms charge radii from the isotope shift experimental data, one has to know so-called electronic factors, which can only be obtained via theoretical methods. For many atomic systems, these factors have rather big uncertainties or have not been calculated at all.

Isotope shifts have two contributions: field shift (associated with a change in the rms charge radius of the nucleus) and mass shift (associated with the recoil effect). The mass shift contribution can be further split into normal (NMS) and specific (SMS) terms.

In the present work, we refine the values of the field shift constant, and also calculate the normal and specific mass shift constants for the $6p^2P_{3/2} \to 7s^2S_{1/2}$, $6p^2P_{1/2} \to 6d^2D_{3/2}$ and $6p^2P_{1/2} \to 7s^2S_{1/2}$ transitions in the neutral thallium at a high level of accounting for the effects of electron correlation (up to the contribution of quadruple cluster amplitudes of the relativistic coupled cluster method). We also calculate the electronic constants for the $5s^2S_{1/2} \to 5p^2P_{1/2}$ and $5s^2S_{1/2} \to 5p^2P_{3/2}$ in the neutral silver within the same level of theory. Furthermore, we compare theoretical and experimental values of King-plot parameters for $6p^2P_{3/2} \to 7s^2S_{1/2}$ and $6p^2P_{1/2} \to 6d^2D_{3/2}$ transitions in Tl. Finally, we recalculate rms charge radii for wide range of Tl isotopes using our results for electronic factors. The estimated theoretical uncertainties for rms charge radii are reduced and amounted to 2.6%. Results on Tl electronic factors were previously reported in[4].

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Calculation of differential ionisation in proton collisions with H_2

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Accurate calculation of differential cross sections for ionisation in proton collisions with molecular hydrogen presents a significant challenge. Currently available calculations rely on perturbative methods and ignore strong coupling effects between the various reaction channels, leading to poor agreement with the experimental data.. For example, measurements [1] of the the singly differential cross section (SDCS) and doubly differential cross section (DDCS) for ionisation differential in the energy and angle of the electron have been available for over 50 years [1], but theoretical results are only available for selected electron energies and angles where agreement with the data is inconsistent. More recently, the DDCS as a function of the projectile-scattering angle and electron energy has also been measured [2]. Until now, only perturbative methods have been applied to calculate the differential cross sections for ionisation in $p + H_2$ collisions, demonstrating inconsistent agreement with the experimental data.

We have developed a wave-packet convergent close-coupling (WP-CCC) method to calculate differential cross sections for ionisation in $p + H_2$ collisions. The approach expands the total scattering wave function in terms of both target and projectile-centred basis states. Substituting this expansion into the Schrödinger equation for the scattering system leads to a set of coupled differential equations to solve for the unknown expansion coefficients. The latter are then used to calculate the differential cross sections. The two-centre expansion allows us to determine direct ionisation and electron capture into the continuum of the projectile. Both components contribute significantly to the ionisation process at intermediate energies.

The WP-CCC method has been applied to calculate all types of the molecular orientation-averaged SDCS [3] and DDCS [4] for single ionisation in proton collisions with molecular hydrogen. Very good agreement with experimental data was found for the SDCS as a function of the electron energy and the SDCS as a function of the electron angle. The experimental data on the DDCS as a function of the energy and angle of the emitted electron is also very well described by the present approach, resolving a long-standing theoretical challenge. The WP-CCC approach to differential ionisation is the first theoretical method capable of accurately describing the DDCS across the entire kinematic regime of the emitted electron in $p+H_2$ collisions, which currently available perturbative methods are unable to accurately reproduce [5]. We have also calculated the DDCS as a function of the projectile-scattering angle and electron energy, providing the first non-perturbative results for this cross section [6].

Having established the robustness of the present approach we are now focussing on the fully differential cross section (FDCS) for ionisation. This is the most detailed description of the ionisation process and presents a major theoretical challenge. Currently available perturbative calculations differ significantly from experimental measurements. Our hope is that the sophisticated WP-CCC method can provide insight into the underlying physics and resolve these discrepancies.

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Fragmentation mechanism of hydrocarbon dications or trications

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The fragmentation mechanism of hydrocarbon molecules is a critical elementary topic in physical, chemical, and biochemical reactions. Especially, the typical H migration or roaming process occurring in tens to hundreds of femtoseconds could open some novel dissociation channels. To probe the detailed fragmentation mechanism of hydrocarbons, the cold target recoil ion momentum spectrum (COLTRIMS) is established [1], and the *ab-initio* quantum chemical calculations (e.g., transition state theory (TST) and *ab*-initio molecular dynamics (AIMD)) are also used to provide theoretical support.

In our investigation, the symmetric and isomerized asymmetric fragmentations of C_2H_6 and C_2H_4 dications have been investigated by collisions between 3-keV/u Ar^{8+} and 18keV/u Ne^{8+}, respectively [2,3]. As shown in Figure 1(a, b), the isomerized and symmetric fragmentation $CH_4^+ + CH_2^+$ and $CH_3^+ + CH_3^+$ were observed. With the help of the measured kinetic energy release distribution (KERs) and calculated minimum energy path (MEP), the isomerized fragmentation channel is contributed by an electronic triplet state, and an H-induced isomerization occurs before fragmentation. The triplet state also contributes to the direct symmetric fragmentation $CH_3^+ + CH_3^+$ with lower energy. For the symmetric fragmentation with higher energy, to our surprise, H-induced isomerization plays a special role in the reaction pathway, which is contributed by the electronic singlet state.

The formation of H_3^+ from C_2H_6 is investigated by the impact of 300 eV electron beam [4]. With the simulation of the potential energy curve, MEP, and AIMD, the formation of H_3^+ is reported. In addition to the short-time TS mechanism, H_3^+ is also formed by the H and H_2 roaming mechanisms. It is also illustrated, compared to the TS mechanism, the roaming process takes a much longer time, exhibits larger amplitude motion, and requires the displacement of more H atoms.

Besides the investigation of the H-migration induced fragmentation mechanism, the three-body Coulomb explosion mechanism of $C_2H_4^{3+}$ is studied, three different fragmentation channels with various concerted and sequential dissociation mechanisms are reported [5]. Delayed dissociation channels of metastable dications $C_2H_4^{2+}$ and $C_3H_4^{2+}$ (allene) are also observed using COLTRIMS [6]. Besides the two-body channels, i.e., $H^+ + C_2H_3^+$ and $H_+ + C_3H_3^+$, the delayed deprotonation is also observed in many-body channels, i.e., $H + H^+ + C_nH_2^+$ (n = 2, 3) and $2H/H_2 + H^+ + C_3H^+$. The lifetimes of the metastable intermediate dications CnH_3^{2+} and $C_3H_2^{2+}$ are estimated.

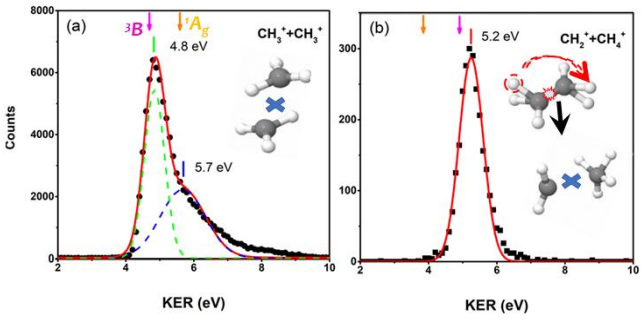

Figure 1. Fragmentation sketches and KERs of (a) $C_2H_6^{2+} \rightarrow CH_3^+ + CH_3^+$, (b) $C_2H_6^{2+} \rightarrow CH_2^+ + CH_4^+$.

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Interaction of protons with noble gas atoms: Total and differential ionisation cross sections

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Electron processes have a crucial impact on various fields of study, such as radiation physics, atomic and molecular structures, and fusion plasma research. In the context of tokamaks, neutral atom beams are used as diagnostic tools [1].

The aim of our present work is to provide total and differential cross sections for single ionization in collisions between H+ with Ne(2p) Ar(3p) Kr(4p), Xe(4d) and Xe (5p).

To model our collision systems we used the 3-body classical trajectory Monte Carlo technique. The interactions among the particles are taken into account with the Garvey model potential [2]. The target was split into a single active electron and the target core consisting of the nucleus and remaining non-active electrons. The projectile H^+ was the third particle. This model potential takes into account the effective charge of the target, incorporating the screening effect of non-active electrons. The classical equation of motions were solved numerically using the adaptive Runge-Kutta method, the step size depends on the initial parameters of all particles [3]. We present results both for the charge transfer and ionisation total cross section as a function of the impact energy. Moreover, we also present single (SDCS, see Figure 1.) and the double (DDCS) differential cross sections as functions of the ejected electron energies and angles at an impact energy of 35 keV.

We compared our cross sections with the existing experimental data, and with the previous theoretical data like first Born approximation (FBA), Oppenheimer-Brinkman-Kramers (OBK) approximation, and the results of the two-state two-center atomic expansion model [4].

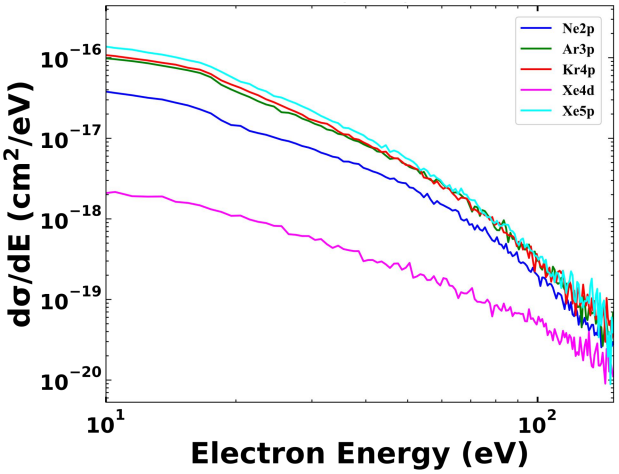

Figure 1. Single differential cross section (SDCS) as a function of the ejected electron energies at impact energy of 35 keV.

Acknowledgements

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Interaction of singly charged sodium ion with nitrogen atom: Total and differential ionisation cross sections

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The electron processes play a significant role in radiation physics, the study of atomic and molecular structures, fusion plasma, and other research domains. In tokamak, atoms are used as a neutral diagnostic beams, such as helium and nitrogen [1].

In our work, we present total and differential crosss sections for single ionization in collision between Na^+ ions with N(2p) atom.

We modelled the collision system as a three body system using Garvey model potential [2]. The target is separated into a single active electron and nitrogen core (i.e., nitrogen nucleus and its remaining non-active electrons), the projectile Na^+ with its electrons are considered as a single particle. This model potential invokes the effective charge for a given particle, hence, it considers the screening effect of the non-active electrons. The equations of motion of the collisions system are solved numerically using classical trajectories Monte Carlo CTMC method [3].

We present the total ionisation cross section as a function of the impact energy in the energy range between 10 keV to 100 MeV. Moreover, we present the single (SDCS) and double (DDCS) differential cross sections for impact energy range of 30-60 keV as a function of the ejected electron energies and angles.

We found that the dominant contribution of the Double Differential Cross Sections (DDCS) are achieved by electrons with energies below 10 eV and ejection angles under 20 degrees. Moreover, electrons ejected with energies larger than 20 eV have shown very small angular dependence [4].

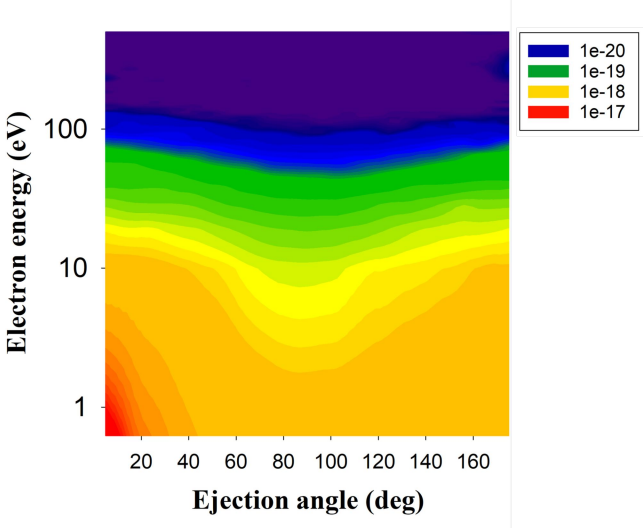

Figure 1. Double differential cross section (DDCS) of the single ionization in collision between Na^+ ions with N(2p) atom as a function of the ejected electron energies and ejection angles at impact energy of 30 keV.

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Single ionization dynamics in relativistic Fe^{26+} + He collisions

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Atomic single ionization by ion impact is one of the most fundamental few-body processes, having been extensively studied for decades [1]. The extraction of fully differential cross sections (FDCS) from kinematically complete measurements has proven indispensable as the most sensitive and stringent test for relevant theoretical models. However, the projectile energies of most studies have predominantly remained below 10 MeV/u, save for only two studies conducted at relativistic energy region [2,3]. Among these, significant inconsistencies have been observed, particularly in the perpendicular plane of scattering coordinates, which challenge the most sophisticated theoretical models, such as the Continuum Distorted Wave-Eikonal Initial State (CDW-EIS) theory. Until now, considerable efforts have been made to solve this problem [4-7], yet the so-called 'C^{6+} puzzle' remains controversial, fueling debates and investigations in the field.

Our study is carried out at the advanced facilities at the CSRe of Heavy-Ion Research Facility at Lanzhou (HIRFL), where we have established an in-ring reaction microscope. The single ionization dynamics of Fe^{26+}+He collisions at the projectile energies of 25 and 120 MeV/u is studied. The experiment employs a stored Fe^{26+} ion beam intersecting with a cold helium gas target emitted from a supersonic jet. A pulsed projectile beam, operating at a 1 MHz repetition rate with sub-3 ns pulse durations, facilitates precise timing resolution of the ionization events.

Our data analysis has successfully generated the three-dimensional momentum components of the recoil ion and electron. Figure 1(a) displays the recoil momentum distribution in the plane orthogonal to the beam direction at a projectile energy of 120 MeV/u, revealing the isotropic ejection of recoil ions in the transversal plane. Figure 1(b) illustrates the electron energy spectrum resulted from single ionization at the same projectile energy.

Now the analysis of the experimental data is ongoing and we will present fully differential results at this conference.

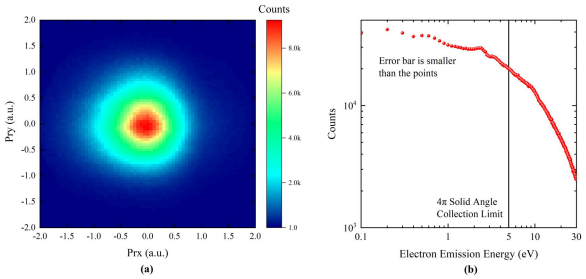

Figure 1. Experimental results of 120 MeV/u Fe^{26+}+He (a) Prx-Pry recoil ion momentum spectrum, (b) Electron emission energy spectrum.

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Parabolic wave packets for time propagation of atomic hydrogen in an electric field of short laser pulses

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We propose parabolic wave packets (PWP) as a basis to solve the time-dependent Schrödinger equation (TDSE) for atomic hydrogen in a laser radiation field. Within the framework of the time-dependent variational principle (TDVP) [1], the TDSE is transformed into a set of first-order differential equations for the parameters of the PWP.

Thanks to the asymptotic properties of the basis functions, which are Sturmian basis functions of parabolic coordinates [2], the ionization amplitude is expressed in closed form in terms of the PWP parameters. The robustness of the method is first validated with a test model with analytical solution, and then applied to the ionization process of atomic hydrogen in half- and few-cycle laser pulses.

The use of parabolic coordinates, suited to the axial symmetry of the linear polarization, makes the expansion numerically efficient. The main purpose of our work is to analyze the capabilities of the TDVP formulated for PWP. The method is tested by comparing with results obtained by other methods. For example, Fig. 1 shows our results for the laser impulse case [3, 4] with $\omega = 0.3$ a.u. and $I = 10^{15}$ W/cm^2.

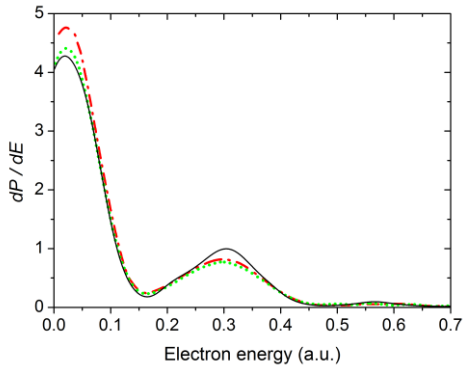

Figure 1. Photoelectron spectrum of atomic hydrogen after its irradiation with the four-cycle pulse. Solid curve: this work for the time step $\Delta t = 0.002$; dotted curve: results from [3]; dash-dotted curve: results from [4].

In turn, Fig. 2 demonstrates the behavior of the calculated ionization spectrum with decreasing the time step.

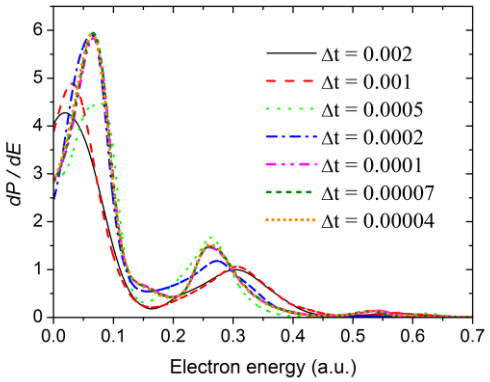

Figure 2. Convergence behavior of the photoelectron spectrum as the time step Δt decreases.

As can be seen from the figure, the final result differs markedly from the initial behavior presented in Fig. 1. The time-dependent basis functions appear to be quite flexible, so that $N = 54$ basis functions are already enough to perform such a study.

It should be added that within the framework of the method, there are not explicit restrictions on the size of the spatial region in which the solution is sought.

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Argon dimer dissociation induced by slow and low-charge-state ion collisions

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Weakly bound systems provide possibilities to study nonlocal electronic decay processes, which present unique physical and chemical properties [1]. Here we report the experimental observation of a nonlocal decay process, i.e., nonadiabatic-coupling-mediated charge transfer (NCMCT), in argon dimer. This process can be effectively produced and manipulated by the double electron capture in low-charge-state oxygen ion collisions. Compared to other common decay mechanisms of Ar_2 dication, the NCMCT process involves distinguishing collision dynamics and results in notably different kinetic energy releases (KERs) for the $Ar^+ + Ar^+$ channel (see Fig. 1). It is due, revealed by theoretical calculations, to the special potential energy curve of the responsible Ar^{2+} – Ar state that possesses many crossing points and thus nonadiabatically couples with the Ar^+ – Ar^{+*} states in a wide internuclear distance range. Such an NCMCT process is expected to be a general process occurring in weakly bound systems when highly excited. The present work provides a prime collision system to explore the charge transfer in ion – molecule reactions and also brings insight into cluster dissociation mechanism.

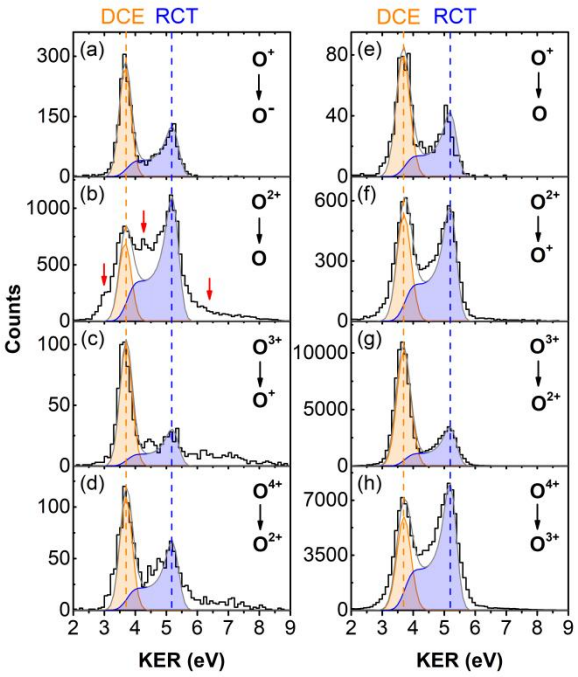

Figure 1. KER distributions for the Ar+ + Ar+ channel under 15q keV O^{q+} (q = 1-4) collisions. The left and right panels correspond to the double capture and transfer ionization channels, respectively. The vertical dashed lines indicate the peak positions of the direct Coulomb explosion (DCE) and radiative charge transfer (RCT) processes. The red arrows mark the new KER structures.

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Correlation effects in the theory of g-factor and hyperfine splitting of highly charged ions

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Studies of the g-factor of highly charged ions have seriously developed recently [1]. Joint theoretical and experimental results for H-like ions have provided the most accurate value of the electron mass to date [2]. There are reasons to believe that further refinement of theory and experiment will lead to an independent determination of the fine structure constant α [3,4], and to tests of quantum electrodynamics beyond the Furry picture in the strong coupling region [5]. It will also make it possible to determine the magnetic moments and radii of nuclei [6]. Recent studies have also demonstrated the determination possibility of the lifetime of the thorium-229 isomeric state by measuring the g-factor of H-like and Li-like ions [7]. This approach has important implications for the development of the so-called nuclear clocks [8].

Another effective method for the study of QED effects is the hyperfine splitting (HFS) in highly charged ions. Many experiments have been carried out with H-like ions, see e.g. [9], and the corresponding theory has been developed. However, the QED contribution to the HFS is masked by the Bohr-Weisskopf effect. To overcome this problem, it was proposed to use a specific difference of the HFS values in H-like and Li-like ions with the same nucleus [10]. By far the most accurate theoretical value of the specific difference for Li-like and H-like bismuth 209Bi was obtained in [11]. Calculations for Li-like ions have been extended to a wide range of nuclear charge Z [12], and calculations of the HFS in B-like ions have also been carried out [13].

At present, the interelectronic-interaction contributions of the first and second order to the g-factor and HFS of Li-like and B-like ions are calculated within the framework of the bound-state QED. Higher-order contributions are accounted for within the Breit approximation by nonperturbative methods. The computations in this work were performed using a novel approach built on the recursive formulation of perturbation theory with a finite basis set of many-electron wave functions constructed in the form of Slater determinants [14]. Recently, our group has applied this method to calculations of electron correlation effects in the g-factor of Li-like ions for a wide range of Z [15]. Also, in [12] this method was applied to calculate the HFS of Li-like ions. In this work, we present the g-factor theoretical values of the excited states $(1s)^2\,2p_{1/2}$ and $(1s)^2\,2p_{3/2}$ of Li-like ions for Z = 10 – 92 with an accuracy of 10^{-6}. Results on the hyperfine splitting of the ground state of Li-like ions and the ground and first excited states of B-like ions are also presented.

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Measuring the quantum state of photoelectrons

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The photoelectric effect is a fundamental quantum phenomenon that is used in various spectroscopy and microscopy techniques. The advent of attosecond science has opened the possibility to study the dynamics of photoemission on its natural time scale by measuring the spectral amplitude and phase of the emitted photoelectron wave packets. This description of the photoelectrons is appropriate when they can be described by a wavefunction, i.e. the photoelectron quantum state is pure. However, in many cases the photoelectron quantum state is mixed and it must be described by a density matrix.

In this work, we implement photoelectron quantum state tomography, KRAKEN, to measure the density matrix of photoelectrons emitted from helium and argon atoms [1]. Our technique uses an ultrashort extreme ultraviolet pulse to ionize the targets and a delayed tunable bichromatic infrared pulse with frequency components ω_1 and ω_2 spaced by $\delta\omega=\omega_1-\omega_2$ to measure interferometrically the coherences between different continuum states populated by the extreme ultraviolet pulse [Fig. 1a)]. By recording photoelectron interferograms for different values of $\delta\omega$, different coherence in the photoelectron density matrix can be obtained. Our measurements show that, while the quantum state of photoelectrons emitted from helium atoms is pure [Fig. 1b)], in argon the photoelectrons are in a mixed quantum state [Fig. 1c)] due to ion-photoelectron entanglement induced by spin-orbit interaction [2]. Using the measured photoelectron density matrix in argon, we quantify the degree of electron-ion entanglement. This work paves the way for the development of photoelectron quantum state spectroscopy of matter and studying entanglement and decoherence in photoionization.

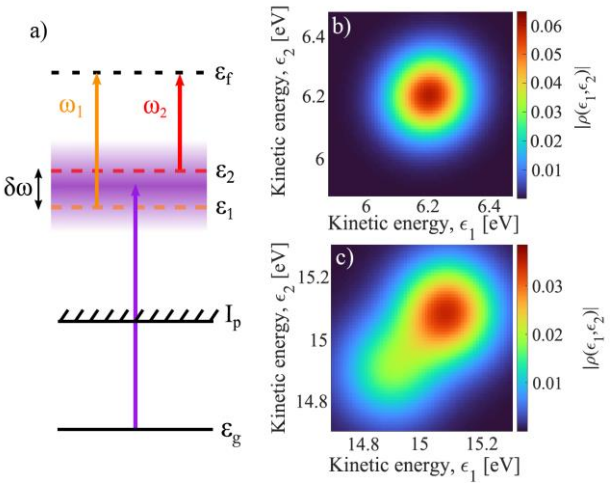

Figure 1. a) Principle of the KRAKEN protocol. b,c) Experimentally reconstructed density matrices in helium (b) and argon (c).

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quantum state tomography of molecules by ultrafast diffraction

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The ability to capture the structural dynamics of molecules at the atomic level has revolutionized our understanding of chemical reactions and biological processes. However, most experiments only provide partial information on the nuclear wavepackets, leaving the entire quantum state inaccessible. In this work, inspired by crystallographic phase retrieval, we introduce a new framework for characterizing the entire quantum state of molecules using ultrafast coherent diffraction from rotational wavepackets. By reconstructing the density matrix, which encodes the amplitude and phase of the wavepacket, researchers can create a quantum molecular movie from experimental data. We demonstrate the feasibility of this approach with N2 and discuss the theoretical and experimental aspects of the framework, including its potential applications in studying molecular dynamics. This work will inspire further research in quantum state tomography and its applications in chemistry, physics, and biology.

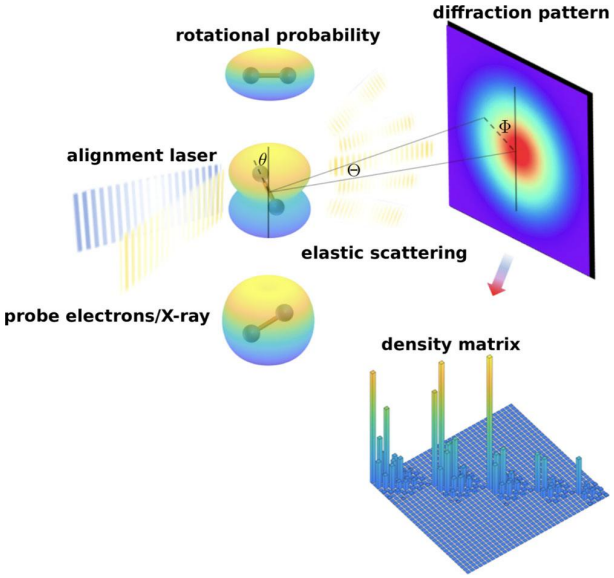

Figure 1. Schematic drawing of quantum tomography by ultrafast diffraction.

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Simulation of the Coulomb dynamic of the fragments at irradiation of water molecule by intense electromagnetic pulse

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The progress in generation of intense short electromagnetic pulse allow observation of exotic states both atomic and molecular species, such as double core-hole states. In molecules absorption of a few VUV or X-Ray photons initiate a complex dynamic leading sometimes to Coulomb decay of the molecule.
This work was stimulated by the recent investigation [1], where the fragments of water decay: protons and oxygen ion were in coincidence. In the report we will present a theoretical model to describe the dynamic of charged water fragment and corresponding electron emission spectrum.

Water molecule irradiated by X-Ray pulse (1 KeV) is ionized primarily from inner shell. The configuration of $H_2O(z=+1)$ differs on neutral molecule in a minor way, but first absorption followed by Auger-decay or second absorption. The ion $H_2O(z=+2)$ tends to relax to the equilibrium configuration (fig.1 (a)) and vibrates according to bending (686 cm^{-1}), symmetrical (1144 cm^{-1}) and asymmetrical modes (1672 cm^{-1}). Calculations were carried out in the GAMESS program using the ROHF method, taking into account electron-electron correlations using the MP2 method. If the next ionization occurs before the dissociation then the molecules acquires an essential charge (z=+3 or +4) and the fragments repulse due to the Coulomb interaction. The geometry reached at $H_2O(z=+2)$ evolution to the moment of decay provides us with the initial conditions for solving of the equations of the classical mechanics. The fragments remain in the field for a while may be ionized further.

In figure 1(b) there is simulation of the proton momentum detected in coincidence with oxygen ion O^2+. The calculations are performed for the 40-fs pulse with $\sin^2$ envelope and intensity 10^{16} W/cm^2. The bright spots correspond to the momentum expected from the three-body repulsion. The green rings are due to geometrical relaxation of the molecule and vibration modes accounted in the simulation. In panel (c) there is the spectrum of electron emission for the same conditions.

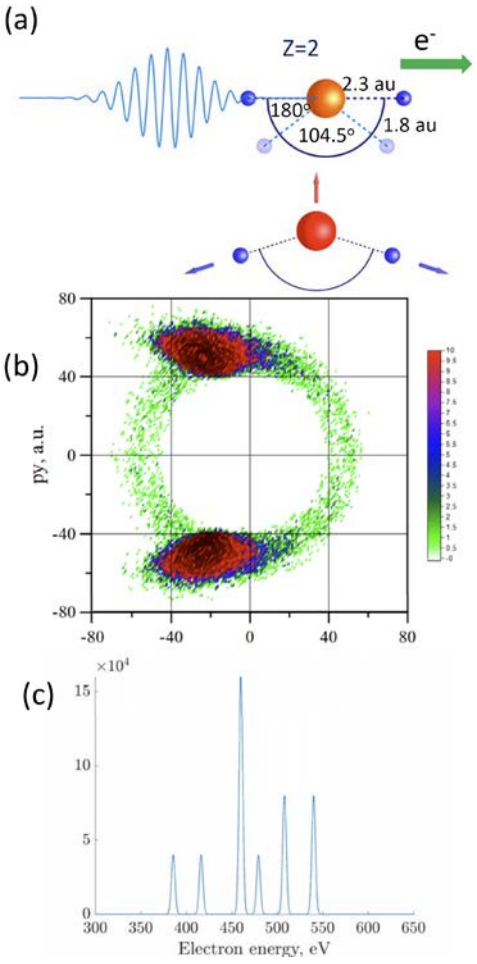

Figure 1. (a)The sketch of ionic dynamic; (b) Newton diagrams for two protons and O^{2+} momentum; (c) the electron emission spectrum.

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Investigation of non-perturbative multiphoton effect in attosecond photoionization of helium atoms

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The development of attosecond metrology is central to the understanding of ultrafast electron dynamics in matter from gas to the condensed phase with attosecond temporal resolution. Traditionally, RABBITT employs a perturbative, near-IR pulse, i.e. with intensity on the order of 10^{10} to 10^{11} W/cm^2 in atoms and a little bit higher in molecules and clusters [2,3,4]. However, limiting the NIR intensity to remain within the perturbative regime increases the difficulty of the experimental data acquisition. A direct way to improve the efficiency of experimental measurements is to increase the intensity of the NIR field for a higher photoelectron yield. But, when the perturbative limit is no longer valid, extra transition pathways become available, and the resulting RABBITT spectra will comprise additional partial waves with higher angular momenta.

The general approach to the Reconstruction of Attosecond Beating By Interference of Two-photon Transition (RABBITT) technique is based on perturbation theory. Here, we extend the traditional RABBITT measurements into a nonperturbative regime with an NIR intensity of TW level. Experimentally, we observe the oscillation amplitude of the sideband under the frequency of 4ωNIR as well as the standard 2ωNIR. The laser-assisted photoionization through the intermediate 1s4p- and 1s5p-Rydberg states leads to distinct angle-resolved photoelectron phase shift distributions. Our partial-wave decomposition approach demonstrates that these differences originate from the contribution of g0-wave, further signifying a higher-order interaction involving one XUV photon and three NIR photons, which consist well with the time-dependent R-matrix simulations.

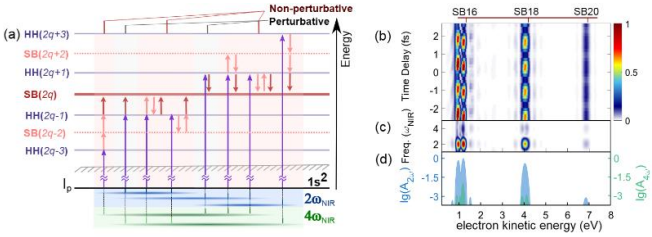

Figure 1. (a) Schematic diagram of transition map of the photoionization in helium atoms. (b) RMT simulated RABBIT-FPT spectrum in helium atoms with a NIR intensity of 1.0×10^{12}W/cm^2. (c) FFT amplitude spectrum of (b). (d) The logarithmic scaled gated FFT amplitudes of $2\omega_{NIR}$ and $4\omega_{NIR}$ oscillations.

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Visualize the vibronic coupling in Auger final states in N_2 molecule

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Normally, the motion of the ground state molecule can be described well using the Born–Oppenheimer approximation, in which the vibronic coupling is not included. On the other hand, the molecule is usually in excited state when involving the chemical reactions. Due to the density of the states increases, vibronic coupling is a critical mechanism in chemical reactions. However, its quantitative evaluation is challenging due to mathematical complexity and programming difficulty, and its experimental proof is often elusive due to overlap among neighboring states.

To address these challenges, we conducted an experiment focusing on exciting different vibrational levels (v = 0, 1, 2) of the N 1s → π_g* core-excited state in N_2 molecules. By separating the resonant Auger decay channels leading to the lowest dissociation limit in the two-dimensional energy correlation maps, we aimed to provide experimental proof of vibronic coupling. Only involving the vibronic coupling between 1 $^2\Pi_g$ and 2 $^2\Pi_g$ resonant Auger final states, the experimental kinetic energy release spectra of these channels at different vibrational quantum numbers can be interpreted well, as shown by the green line and the red line in Figure 1. This ultimately providing the first experimental proof of vibronic coupling between the two resonant Auger final states.

This experimental proof is significant as it contributes to a better understanding of vibronic coupling, which plays a critical role in chemical reactions. The experimental approach and results presented offer valuable insights into advancing the understanding of vibronic coupling and its implications for chemical reactions.

In conclusion, this work provides an in-depth exploration of vibronic coupling in Auger final states in the N_2 molecule, offering valuable contributions to the field of chemical reactions and molecular dynamics.

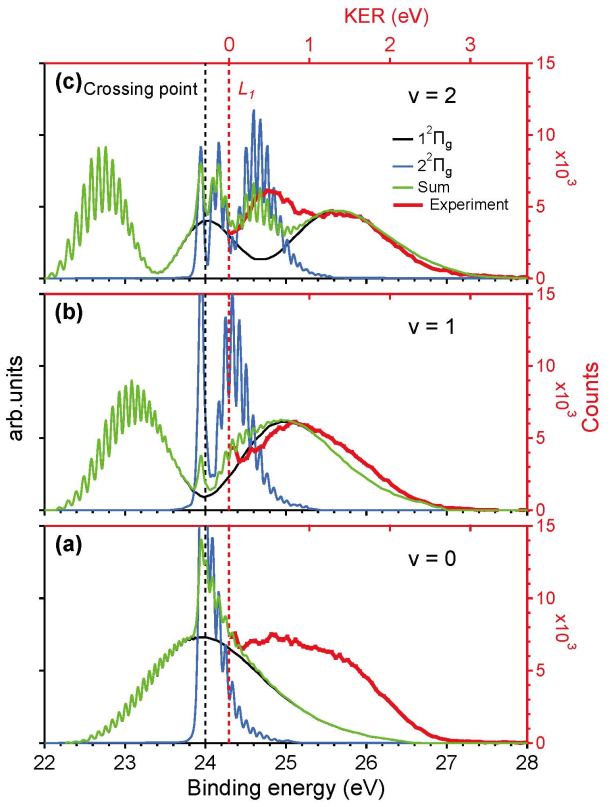

Figure 1. Comparing the experimental KER and theoretical resonant Auger electron spectra. (a)–(c) The theoretical resonant Auger electron spectra to the 1 $^2\Pi_g$ and 2 $^2\Pi_g$ states from Ref. [1] are shown by a black and a blue line, respectively. The green line indicates the total contribution of the 1 $^2\Pi_g$ and 2 $^2\Pi_g$ states to the KER spectrum. The vertical dashed red and black lines indicate the lowest dissociation limit L_1 and the crossing point between the 1 $^2\Pi_g$ and 2 $^2\Pi_g$ states, respectively.

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"Mustache" electron-multi-ion coincidence setup -- first results at the tender X-ray regime

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Various multiparticle coincidence techniques have proven invaluable for studying the structure and dynamics in small quantum systems. Various designs have their particular pros, cons and compromises; in the "Mustache" electron-multi-ion coincidence endstation design presented here, the emphasis is on the high electron energy resolution across a very broad kinetic energy range. This is achieved by combining a Scienta hemispherical energy analyzer with a modifiel Wiley-McLaren type ion time-of-flight spectrometer that provides, in addition to the ion mass data, a fair 3D resolution also for the ion momenta.

The "Mustache" setup (Fig. 1) is particularly targeted for the use of synchrotron radiation at the "tender" X-ray regime. The electron-energy-resolved coincidence data across the broad energy range provide numerous advantages, allowing to select various ionization sites from valence to deep core levels, or concentrate on Auger, multi-spet Auger or satellite structures in the spectra. It also allows for vibrationally resolved photoelectron-photoion coincidences involving deep core levels.
A main challenge at the tender X-ray regime is the diminished transmission of high-energy electrons. During the first commissioning measurements at the GALAXIES beamline [1] of the Soleil synchrotron, the peformance of the setup was tested under a wide range of conditions. The results are presented here for Argon, the CS_2 and N_2 molecules.

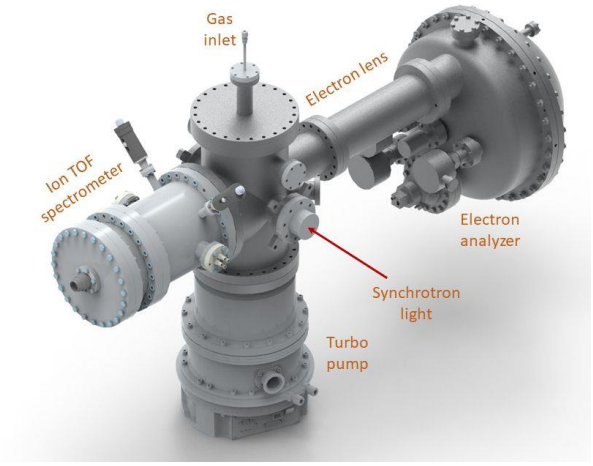

Figure 1. "Mustache" electron-ion coincidence endstation.

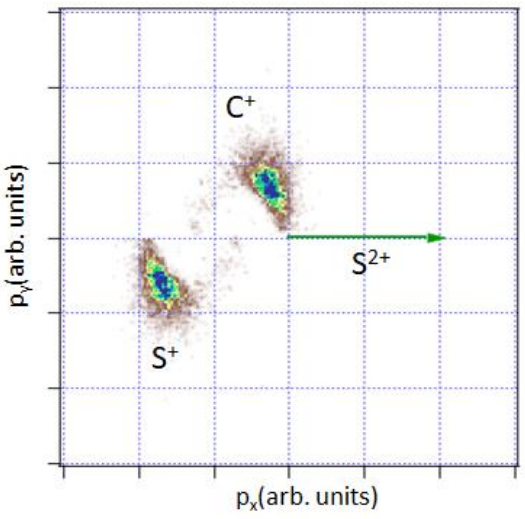

Figure 2. Newton plot of the dissociation of CS_2 after sulfur 1s ionization.

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Spectroscopy of atomic ensembles in a waveguide

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The study of optical collective effects in atomic ensembles represents one of the most promising fields in quantum physics [1]. It has found a number of applications in quantum metrology, frequency standardization, and quantum information science.

In the recent decades, the study of optical properties of atomic ensembles coupled with a cavity or waveguide attracts considerable attention [2]. It is caused by the fact that a cavity offers an exciting tool to control over the light-matter interaction. Light-matter interface in the presence of nanophotonic structures, such as nanofibers, photonic crystal cavities, and waveguides propose future applications for quantum metrology, scalable quantum networks, and quantum memory cells.

In our group, called “optical spectroscopy of quantum systems”, we work on many particle spectroscopy of atomic ensembles located in a waveguide.

In particular, we found out the effect of incomplete spontaneous decay — when the atomic excited state population asymptotically approaches to a nonzero value at large times. It happens under the conditions when the atomic transition frequency is larger than the cutoff frequency of a waveguide and far from the vicinities of the cutoffs [3]. The discovered effect is explained by the emergence of the dark state, which is nondecaying due to polarization selection rules. It was revealed for a single-mode waveguide with rectangular cross section both in a single-atom case and a diatomic case when the long-range dipole-dipole interaction plays a significant role.

Furthermore, we have calculated the transmission of disordered atomic ensemble in a waveguide. It has been done on the basis of self-consistent quantum microscopic treatment taking into account 3D arrangement of atoms and the vectorial nature of the electromagnetic field [4]. We have found that the nature of light transport essentially depends on the transverse sizes of a waveguide. A single-mode waveguide with small transverse sizes exhibits Anderson localization of light, which manifests itself in exponential decrease of the transmittance with increasing in the thickness of atomic sample. An increase of the transverse sizes breaks exponential law, so that in a few-mode waveguide we observe complicated dependence of the transmission on the thickness. The reason of this is complex interplay between Anderson localization of light and diffuse radiation transfer. With further increasing of the transverse sizes of a waveguide, the transmission scaling obeys hyperbolic law, which indicates on the regime of diffuse transfer in a multi-mode waveguide.

In recent years, we work on the investigation of the Lamb shift which depends on the position of an atom in a waveguide [5]. Spatial dependence of the Lamb shift is explained by the inhomogeneity of the spatial structure of the modes of the electromagnetic field. We have analyzed how it modifies the character of the dipole-dipole interaction. Our results demonstrate that the difference in the Lamb shift for two atoms located at different points can be comparable to the natural linewidth of the atomic transition or even exceed it. Therefore, the collective dynamics of an atomic system can be significantly affected by the Lamb shift.

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Geometric phase effect in attosecond stimulated x-ray Raman spectroscopy

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Conical intersections (CIs) are diabolical points in the potential energy surfaces generally caused by point-wise degeneracy of different electronic states, and give rise to the geometric phases (GP) of molecular wave functions. The direct observation of CI requires high spectral resolution and ultrafast time resolution, which makes it extremely difficult to measure the GP effects on the molecular dynamics.

We theoretically propose and demonstrate that the transient redistribution of ultrafast electronic coherence in attosecond Raman signal (TRUECARS) spectroscopy [1] could be capable of detecting the GP effect in excited state molecules by applying two probe pules including an attosecond and a femtosecond X-ray pulses, as shown in Figure 1. The two coherent X-ray pulses induce an off-resonant stimulated X-ray Raman process between the electronic states involving the CI. By varying the time delay of the probe light with respect to the pump light, the coherence of the wave packet can be visualized by the TRUECARS technique.

The existence and removal of GP can strongly influence the topological structure of the potential energy surface and further the symmetry of the wave packet in the dynamic process. Since the coherence of the wave packet depends on its symmetry, the Raman transitions in the TRUECARS process are finally affected. In special cases, we show that the appearance of GP can forbid the Raman transitions and cause the corresponding TRUECARS signal to vanish. This vanishing signal provides a direct way to reveal the GP effect.

The mechanism is based on a set of symmetry selection rules in the presence of non-trivial GP. The model of this work [2] can be realized for probing geometric phase effect in the excited state dynamics of complex molecules with appropriate symmetries, using attosecond light sources such as free-electron X-ray lasers. By monitoring initial conditions of the wave packet, the activity of Raman transition can be suppressed and the TRUECARS signal vanishes due to the GP effect. This offers an approach to observe the GP effect of CI and provides a new direction for studying the GP caused by CI with ultrashort coherent X-ray sources.

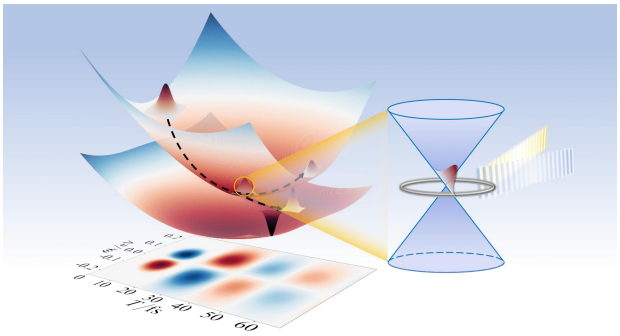

Figure 1. Schematic of molecular dynamics with GP effect. First a nuclear wave packet is stimulated from the ground state to the upper state of a CI, and is symmetric about the coupling mode. When the wave packet passes a symmetry-allowed CI, nonadiabatic coupling and GP effect result in antisymmetric wave packet in the lower state and symmetric wave packet in the upper state. In this case, the expectation of polarization operator as well as TRUECARS signal vanishes and the stimulated Raman process is forbidden.

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Influence of atomic motion on the dynamics of pulsed resonant fluorescence of cold atomic ensembles

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Improvements in techniques for cooling atomic gases in atomic traps make their use very promising for applications in fundamental science and in various area of technology such as quantum information, metrology, the development of frequency standards, and so on.

Practically all of applications envisioned for cold atomic ensembles are based on their interaction with electromagnetic radiation. This interaction has several features associated with collective polyatomic effects. First, cooled atomic ensembles have very large optical thickness even at a low atomic density and, hence, multiple light scattering in them must be considered. The second reason is a spatial disorder, which allows the formation of atomic clusters (quasi-molecules) consisting of several closely spaced atoms. The dipole–dipole interaction and recurrent multiple scattering of light inside a cluster lead to the formation of collective sub- and super-radiant quantum states, which are very sensitive even to small and slow displacements of atoms [1]. And finally, due to low velocities of atoms, interference effects can be observed during multiple scattering despite the absence of any ordering in the atomic cloud [2].

Consistent consideration of all these features is possible in the frame of the coupled dipole (CD) method [3,4]. This method describes the intricate collective dynamics of all atoms of the system. For this reason, it is usually simplified. In most cases the motionless scatterers approximation is used. The displacements of atoms are simulated, firstly, by averaging over their random spatial distribution, and, secondly, by introducing a random frequency shift of their transitions, modeling Doppler effects [5].

In this work, we consider a generalization of the CD method, solving a dynamic problem, explicitly taking into account the displacement of atoms over time. As a specific problem we analyze the influence of atomic movement on the dynamics of fluorescence of dilute ensembles excited by weak resonant pulsed radiation.

We show that three main phases can be distinguished in the evolution of fluorescence. These are the stage of super-radiance, the stage of diffuse radiation trapping, and the stage when sub-radiation is determined by the emission of quasi-molecules. The movement of atoms affects fluorescence at all these stages, even at temperatures 10-100 μK typical for magneto-optical traps (MOT).

Heating leads to a nonmonotonic time dependence of super-radiation. At certain time intervals, instead of slowing down, we observe an acceleration of decay. At the trapping stage, the main factor affecting the fluorescence rate is the diffusion of the radiation frequency caused by multiple scattering of light. The spectrum of the secondary radiation reveals significant broadening upon heating of the ensemble. The most interesting results are found for sub-radiation of diatomic clusters. For the temperature typical for MOTs, the sub-radiation effect is enhanced for moving atoms. This result is explained by the influence of two factors. First, by a modification of the decay rate of each eigenstate of the dimer with a change in the distance between atoms, and second, by possible nonadiabatic transitions between different sub- and super-radiant states.

Note that model calculation based on simulation of Doppler effects by random shifts of atomic energetic levels does not reproduce these results.

The work was supported by the Foundation for the Advancement of Theoretical Physics and Mathematics “BASIS”. Analysis of the broadening of the spectrum of secondary radiation was supported by the RSF (Grant No. 21-72-10004).

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Multiple-core-hole resonance spectroscopy with ultraintense x-ray pulses

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Understanding the interaction of ultraintense and ultrafast x-ray pulses with heavy atoms is crucial for gaining insights into the structure and dynamics of matter. One key aspect of nonlinear light-matter interaction is its dependence on the photon energy, but there has been no systematic study at x-ray free-electron lasers (XFELs) so far. We present a joint theoretical and experimental study of highly charged xenon ions after interaction with XFEL pulses [1] scanning the photon energy over a wide range, which enables us to map out the transient resonances occurring during the complex charge-up pathways. Massively hollow atoms [2] featuring up to six simultaneous core holes determine the spectra at specific photon energies and charge states. The extraction of resonance spectra is facilitated by the fact that the ion yields become independent of the peak fluence beyond a saturation point. Our study lays the groundwork for novel spectroscopy of transient atomic species in exotic, multiple-core-hole states that have not been explored.

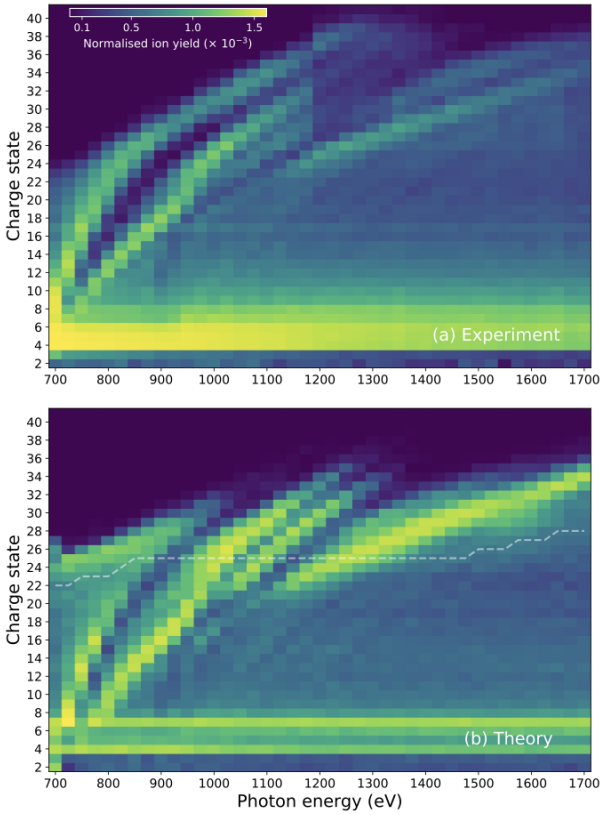

***Figure 1**. Experimental and calculated xenon charge-state distributions as a function of photon energy.*

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excited states dynamics of endohedral metallofullerenes

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The electronic excited state dynamic process of metallofullerene widely exists in its photophysical or photochemical process. From a basic research perspective, metallofullerene is a typical three-dimensional quantum many-body system. Studying its excited state dynamics will promote the understanding of the excitation/decay dynamics of quantum many-body systems in radiation environments such as strong lasers and high-energy electron beams. From a perspective of practical applications, when metallofullerenes were applied to functional devices including single-molecule quantum devices, organic photovoltaic devices, and catalytic devices, their electronic excited state dynamics process plays an important role in its photoelectric conversion efficiency, catalytic efficiency, etc.

In this talk, we will report the ultrafast electronic excited state dynamics of the endohedral metallofullerenes Be@C_{60} and Lu_2@C_{76} by using the time-dependent Kohn-Sham (TDKS) method combined with surface-hopping methods. Through detailed comparison and analysis, the influence of energy levels and fullerene cage size on the decay dynamics was revealed.

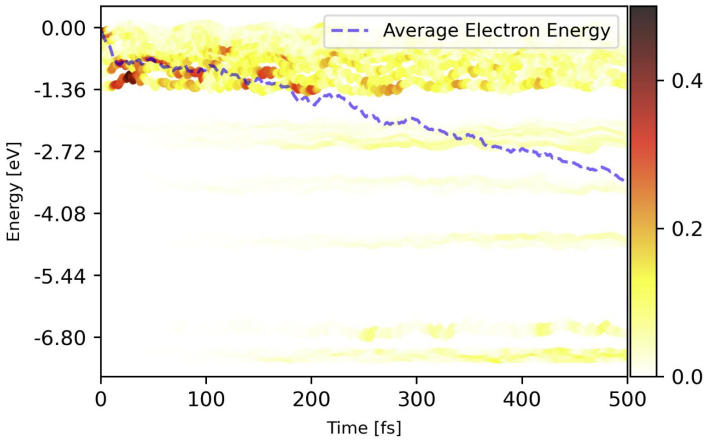

Figure 1. excited states decay dynamics of Be@C_{60}.

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Time-resolved Imaging of Methane Fragmentation in Strong Laser Fields

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With a Reaction Microscope (ReMi) [1] the ionization and dissociation dynamics of CH_4 in strong laser fields was studied in a series of pump-probe experiments. By employing a spatial light modulator (SLM) [2], we successfully compressed the laser pulses to below 10 fs and achieved a precise control over the time-delay between the two pulses. Upon strong-field ionization, the molecule undergoes fragmentation and Coulomb explosion (CE) [3] and the corresponding ionic fragments are collected with the ReMi.

For example, in the case of CE, the initial inter-nuclear distances can be determined via the measurements of final kinetic energies, and together with the angular distribution between the different fragments the molecular structure can be traced. With pump-probe measurements, the evolution of the molecular geometry is visualized as the function of time. Figure 1 displays the kinetic energy release (KER) for the fragmentation pathway $CH_4 \rightarrow CH_3^+ + H^+ + 2e^-$ as a function of the delay between the two laser pulses. Due to dissociation of the fragments after single ionization of CH_4, the KER decreases noticeably with rising pulse delay, and thus, also the radius of the KER-ring in Fig 1. Furthermore, the angular distribution of the detected ions is clearly non-isotropic but peaked along the laser polarization axis.

Beyond these observations, our measurements involve further fragmentation pathways, encompassing not only bond breaking but also formation of new bonds. Notably, the coincident detection of H_2^+ ions with CH_2^+ indicate the formation of a bond between two protons within the pulse duration, i.e., within 10 fs. To validate this further, we also examined the channel leading to $CH_2^+ + H^+ + H^+$ as illustrated in the Newton-plot in Figure 2. Whenever the final kinetic energies of two protons are identical, which is indicative for CE of H_2 or H_2^+ created in the probe pulse, the corresponding distance between $H^+ + H^+$ closely resembles the typical features observed in strong-field ionization of H_2. The three distinct peaks in the proton energy (Fig. 2b) match very well with the dissociation, charge resonance-enhanced ionization (CREI), and CE obtained in experiments with H_2 [4].

More details and further results will be presented in the poster.

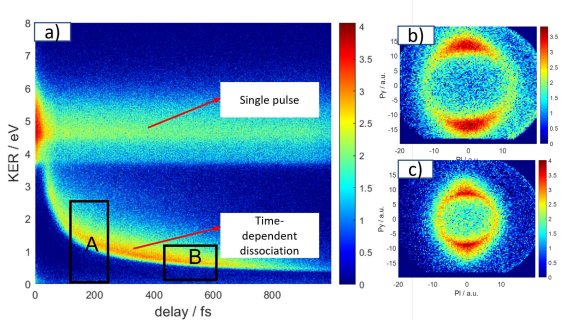

Figure 1. a) KER release of $CH_3^+ + H^+$ dissociation with time delay. b), c) Momentum distribution of H+ for different time delays.

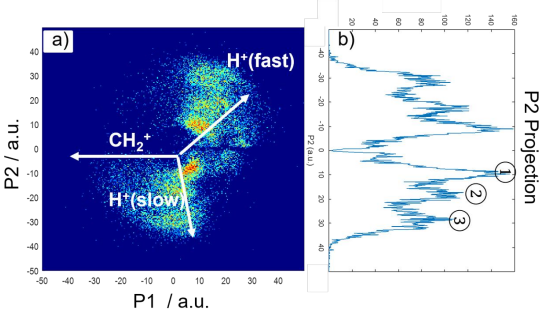

Figure 2. a) Newton Plot for Coulomb explosion into $CH_2^+ + H^+ + H^+$. b) Momentum projection of H^+, presenting three peaks at 9.0, 17.5, and 30 a.u., corresponding to distances between $H^+ + H^+$ of 22.7, 6.0, 2.5 a.u., respectively.

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Observation of an Associative State in Aqueous Hydroxide

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Understanding the behavior of chemical reactions in solutions is crucial across various disciplines, including biology and materials science.
The way protons behave in water, in particular, plays a significant role in many unique properties and processes, such as proton movement in biological systems and the reactions between acids and bases.[1,2]
In this study, we describe the discovery of a previously unidentified ***associative state*** of the hydroxide ion **OH^-**, which is responsible that a proton from a neighboring water molecule approaches the hydroxide ion.
This is in stark contrast to a dissociative behavior reported for pure water.[3]
This finding was made possible through the use of high resolution resonant inelastic soft X-ray scattering (RIXS) and highly sophisticated quantum dynamical simulations.
Advanced theoretical analysis has uncovered a state mixing of electronically excited states between the hydroxide ions and their surrounding water molecules.
Our findings provide fresh perspectives on chemical bonds and the behavior of molecules in excited states within water-based environments.
By exploring these associative states, we pave the way for new approaches in spectroscopic investigations of how chemical reactions unfold, establishing a groundwork for directly studying how protons are exchanged in solutions.

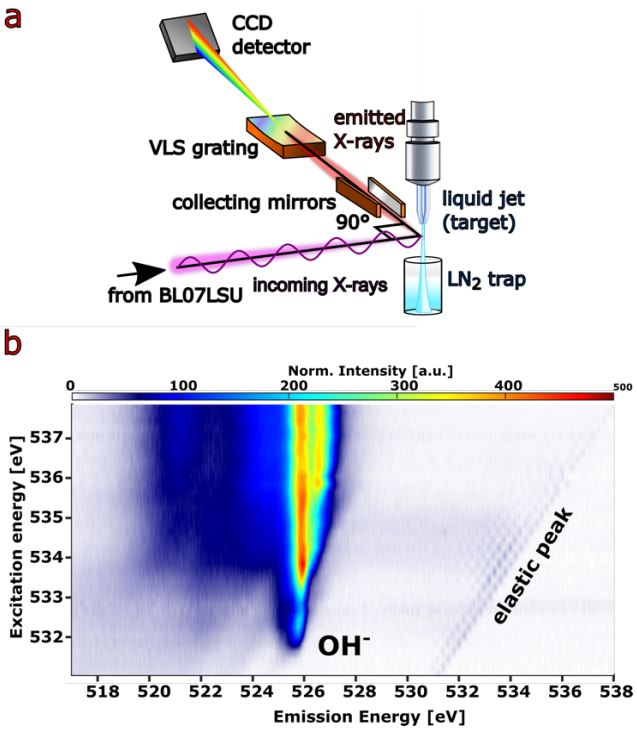

Figure 1. a) Schematic illustration outlines the key components and layout of the experiment. Monochromatic X-rays the beamline at the Spring-8 synchrotron facility excites on the oxygen K-edge of the liquid microjet.
b) RIXS spectra of aqueous OH- solution at the oxygen K-edge reveal emission signals at excitation energies below 533 eV primarily originate from hydroxide ions, demonstrating the site-specific sensitivity of RIXS.

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Two-photon ionization of localized atoms by Bessel light

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The technique of laser radiation generation is being developed in various directions, and its state-of-the-art characteristics such as temporal and longitudinal coherency, frequency, intensity, and pulse duration are spectacular. It helps researchers to delve into light-matter interaction at deeper scales and to study subtle effects that previously were inaccessible. One of the tools to advance our knowledge about light-matter interaction is twisted radiation, i.e., pulses with a definite projection of orbital angular momentum – particularly, Bessel light [1, 2]. This type of electromagnetic waves could be interpreted as a superposition of plane waves with their wave vectors lying on the surface of a cone.

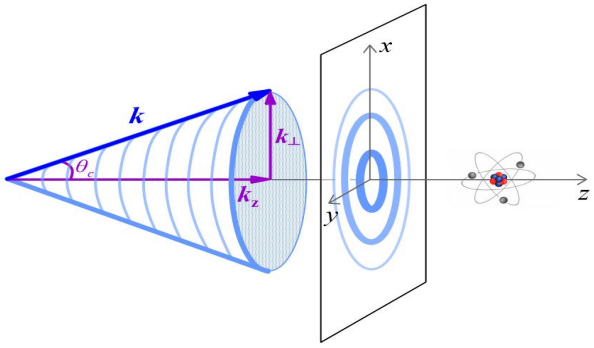

Figure 1. The overview of the Bessel beam propagating along z-axis with wave vector $\boldsymbol{k}$ and cone angle θ_c. Intensity profile of the beam consisting of concentric rings is schematically illustrated in xy-plane. Target atom is localized on the z-axis.

Our work is devoted to a specific process of atomic two-photon ionization by the Bessel beams for a case when atom is localized on the beam propagation axis (see Fig. 1).

In order to calculate the probability of the process and the angular distribution of emitted photoelectrons, we derive the appropriate expressions in dipole approximation using quantum theory of angular momentum and connection between plane-wave and twisted wave ionization matrix elements, similar to the method employed in our previous study [3].

Theoretical results for Bessel light two-photon ionization of helium and neon atoms will be presented at the conference.

The research was funded by the Russian Science Foundation (Project No. 23-62-10026) and by the Ministry of Science and Higher Education of the Russian Federation (Project No. FEME-2024-0005).

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Double-to-single ionization ratio of Ar by 500 keV/u He^{2+} impact

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Recently, we investigated the double-to-single ionization ratio of Ar induced by 500 keV/u He^{2+} impact using a newly built cold target recoil ion momentum spectroscopy (COLTRIMS) mounted on the 3 MV tandetron accelerator. With the help of the supersonic gas jet, the single-collision conditions are ensured. By employing the position spectra of recoil ions, the random coincidence events are largely eliminated.

The ratios of $\sigma(Ar^{2+})/\sigma(Ar^{+})$ as a function of E/M (keV/u) obtained in this work as well as the available theoretical and experimental data are shown in figure 1. In the 10-175 keV/u energy range, the time-dependent density functional theory (TDDFT) values [1] generally agree with the experimental data [2]. However, the basis generator method (BGM) [3] values and the continuum distorted wave eikonal-initial-state (CDW-EIS) [4] values without capture contribution overestimate the $\sigma(Ar^{2+})/\sigma(Ar^{+})$ and *pure* $\sigma(Ar^{2+})/\sigma(Ar^{+})$ experimental data [2, 5], respectively, especially in the 200-2000 keV/u energy range. The present result agrees well with the theoretical values at 500 keV/u impact energy. Tails produced by multiple collisions have been observed on the main peak of the TOF spectra in the existed experiment [5]. The discrepancy between different experiments may be ascribed to the multiple collisions, which could lower the double-to-single ionization ratios.

Further identification of the detection efficiencies of the present experiment is needed. Some effects are not considered in the existed theory, such as autoionization, electron-capture, and electron correlation. More precision theoretical and experimental studies plane to perform.

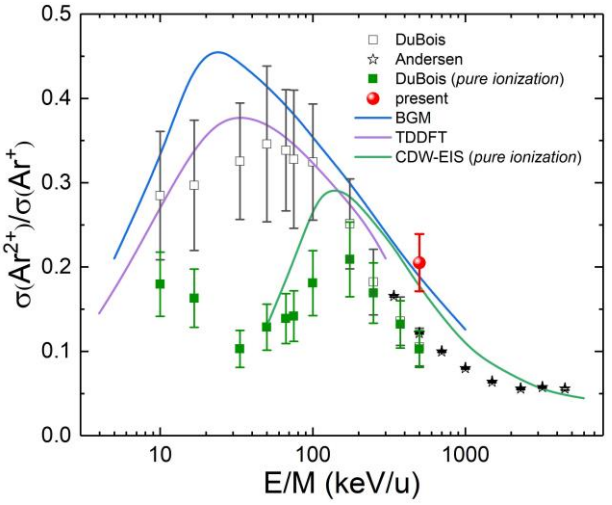

Figure 1. The ratios of double-to-single ionization cross section of Ar by He^{2+} as a function of impact energy. Symbols, Experiment: black open squares, DuBois [2]; black open stars, Andersen et al. [5]; green full squares, *pure* ionization by DuBois [2]; red full circle, this work. Curves, Theory: blue line, BGM theory (target response). [3]; purple line, TDDFT theory. [1]; green line, CDW-EIS theory, pure ionization. [4].

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linear polarization of L-RR and dielectronic hydrogenlike satellite lines

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Recently, the polarization measurement of L-shell radiative recombination (L-RR) x rays was performed on the Tokyo-EBIT [1], and the reported theoretical result performed by the flexible atomic code (FAC) was higher than the experimental value. We present joint theoretical investigations on the linear polarization of x rays emitted from the L-RR into Kr, and Bi ions by two distinct theoretical approaches, i.e., the present relativistic calculations and the revised FAC (NFAC) using the density matrix formalism, in which the relativistic and multipole effects are fully considered.

The energy and atomic number Z dependences of L-RR x-ray polarization are further investigated. Our results are closer to those for L shells than previous calculations. However, deviations still exist between the theoretical calculations and experimental results. It is found that the effect of the configuration interaction (CI) and the finite nuclear size on the polarization of L-RR x rays is not substantial. Detailed calculation results can be found in Figure. 1. More theories and experiments are required to explain these deviations.

Furthermore, a systematic investigation of dielectronic satellite lines in hydrogenlike ions with Z values ranging from 9 to 92 is carried out, with a focus on the effects of the Breit interaction and the generalized Breit interaction on their linear polarization.

We present a detailed analysis of the polarization behaviors and resonance strengths of the satellite lines, facilitating the spectral analysis under the EBIT measurement conditions. Emphasizing the second-step radiative decay process in dielectronic recombination of hydrogenlike ions, the alignment parameters of the singly excited states are determined by using both the deorientation factor formalism and density-matrix theory. Finally, to propose the feasible polarization measurement of the dielectronic satellite lines of hydrogenlike ions at EBITs, the effective polarization is calculated. These calculations indicate a promising opportunity for understanding the effect of the Breit interaction as well as differentiating the formalism of the deorientation factor and the density-matrix theory for the calculations of dielectronic satellite lines.

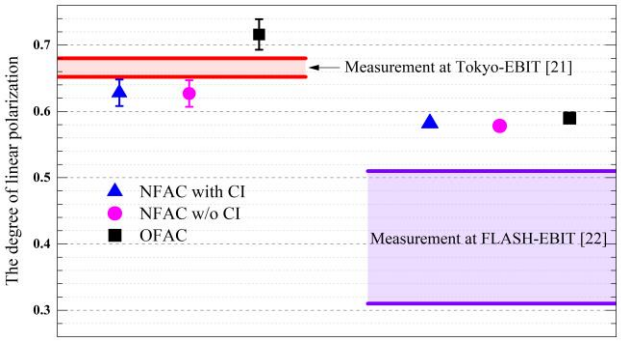

Figure 1. Measurements (Bi ions at 51.1 keV [1] and Kr ions at 8.76 keV [2]) and theoretical calculations of L-RR x-ray linear polarizations. The blue triangles and pink circles represent the calculations of NFAC with and without CI, respectively. The black squares represent the calculations of the OFAC which are reported in previous papers.

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Suppressing electron disorder-induced heating of ultracold neutral plasma via optical lattices

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Ultracold neutral plasma (UNP) provides a promising platform for investigating strongly coupled and high energy density plasma (HEDP) [1-3]. The extremely low temperature of UNPs enables them to be in or near the strongly coupled regime. However, the Coulomb coupling strength of electrons is limited mainly by disorder-induced heating (DIH). Therefore, suppressing DIH and improving electronic Coulomb coupling strength are of great significance for research in UNPs and HEDPs.

In this work, we propose a scheme to suppress the DIH of electrons, which is based on pre-ordering UNPs by loading atoms into three-dimensional optical lattices. We simulate the evolution dynamics of UNP with different initial lattice types and plasma densities using classical molecular dynamics method with open boundary condition. The results show that for experimentally achievable condition, electronic DIH is suppressed by a factor of 1.3, and the Coulomb coupling strength can reach to 0.8 which is approaching the strong coupling regime, as shown in Fig. 1. We also show that the degree of electron temperature reduced is independent on plasma density and lattice types. Suppressing electronic DIH via optical lattice may pave a way for the research of electronic strongly coupled plasma. Moreover, decreased electron temperature will improve the coherent length and brightness of electron beam based on UNPs.

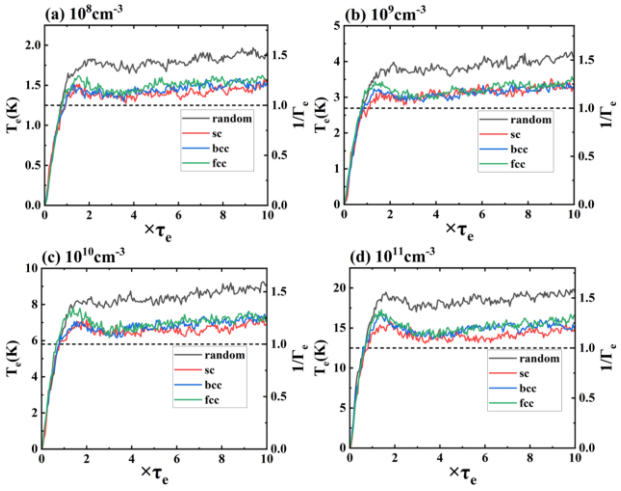

Figure 1. Temperature and Coulomb coupling strength evolution of electrons at different cubic lattice geometries and densities: (a) 10^8 cm^{-3}, (b) 10^9 cm^{-3}, (c) 10^{10} cm^{-3}, (d) 10^{11} cm^{-3}. The legend represents initial different ion density distributions. Black dotted line implies Coulomb coupling strength of 1.

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state-selective single and double electron capture in slow He^{2+}-Ne collisions

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The charge transfer from atoms to ions is the dominant processes in slow highly charged ions (HCI) with neutral atoms. For the many electrons atomic neon target, the study of the state-selective electron only a few works were reported based on the cold target recoil ions momentum spectroscopy (COLTRIMS). Zhang et al. reported the single electron capture (SEC) to quintuple-electron captures using recoil-ion momentum spectroscopy in N^{7+}-Ne collisions at the energy of 7.5 keV/u. Xue et al. reported the study of the SEC and double electron capture (DEC) in 3.2 keV/u Ar^{16+}-Ne collisions. Xu et al. reported the SEC in slow N^{6+}-Ne collisions. For the SEC mentioned above, the projectile capture *2p* of neon target is the dominant reaction channel. In 2021, Wang et al. reported the SEC and DEC processes of He^{2+} ions colliding with Ne atoms by utilizing the full quantum-mechanical molecular-orbital close-coupling method in the energy region of 2 eV/u to 20 keV/u. Their results shown that the dominant reaction channel of SEC is $Ne^{+}(2s2p^{6}\ ^{2}S)$ + $He^{+}(1s)$ in the considered energy region, i.e., a *2s* target electron transfer to the ground states (*1s*) of He^{2+} projectile. For DEC, due to the Demkov-type couplings between DEC channel $Ne^{2+}(2s^{2}2p^{4}\ ^{1}S)$ + $He(1s^{2})$ and the dominating SEC channel $Ne^{+}(2s2p^{6}\ ^{2}S)$ + $He^{+}(1s)$, the DEC cross sections increase with increasing impact energies. Hence, in this work, we use a COLTRIMS for a kinematically complete study of SEC and DEC in He^{2+}-Ne collisions in the energy region of 1 keV/u to 30 keV/u. The orbital angular momentum number *l*-resolved state-selective cross section and angular differential cross section were obtained for the SEC and DEC. Also, our experiments confirm the theoretical calculations, but there are some discrepancies. The detailed results will be present this conference.

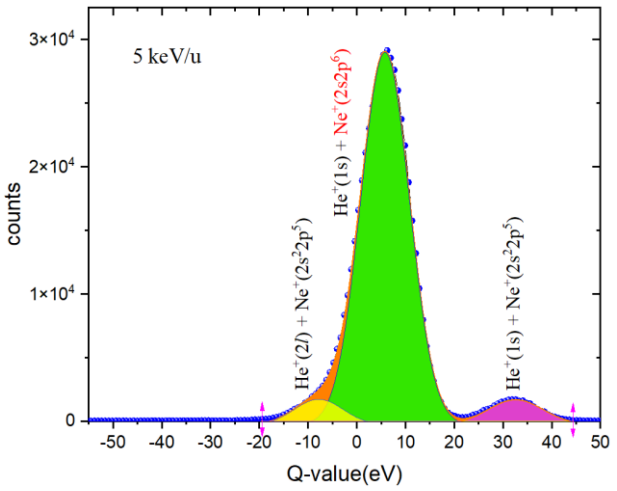

Figure 1. The Q-value spectrum of SEC in He^{2+}-Ne collisions at impact energy of 5 keV/u. The different reaction channels marked in Figure.

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Probing ionization dynamics of Argon by attosecond photoelectron spectroscopy

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

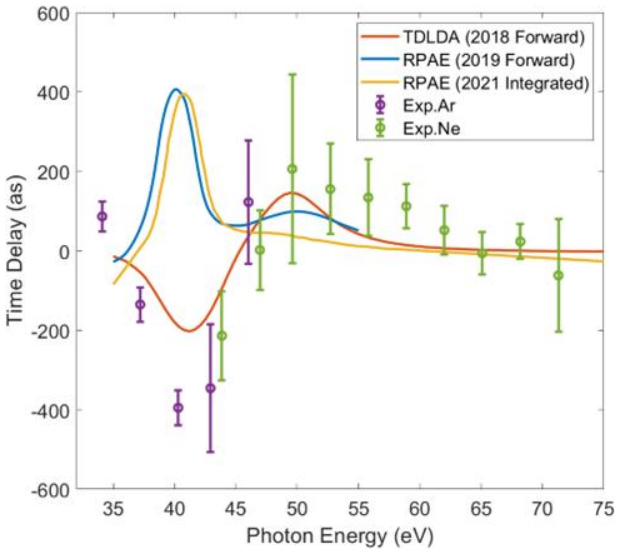

Figure 1. Measured and calculated ionization time delay between 3s and 3p valance shells of Argon.

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Unraveling photodissociation dynamics by sub-femtosecond ultraviolet pulses: insights into fragmental kinetics and carrier-envelope phase characterization

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The duration of laser pulses and the carrier-envelope phase (CEP) play a crucial role in shaping kinetic energy release (KER) spectra. In this study, we performed theoretical calculations on pulse duration-dependent KER spectra, ranging from hundreds to sub-femtoseconds, focusing on the MgH+ scenario. Two crucial findings emerge from this study [1].

Firstly, diminishing pulse duration leads to a transformation of sharp KER peaks into a diverse array of broad structures (see Figure 1). This metamorphosis is intricately influenced by pulse frequency, the initial vibrational state, and the complex interplay between the pulse's energy spectrum and the energy-dependent transition matrix element.

Secondly, within the sub-femtosecond regime, both counter-rotating and rotating terms [2] play a substantial role, amplifying the CEP effect's significance. To characterize the CEP effect accurately, we propose a novel pump-probe method. This method analyzes the KER spectrum generated through the simultaneous excitation of long pulses with known parameters and sub-cycle pulses with unknown CEP values. Leveraging the unique structure of the modulated coherent spectrum, we derive a nonlinear relationship associated with the CEP. This nonlinear dependence serves as a valuable complement to the standard approach for characterizing the CEP of isolated attosecond pulses (see Figure 2).

The insights derived from this work carry significant implications for advancing our comprehension of ultrafast phenomena and their applications across various scientific domains.

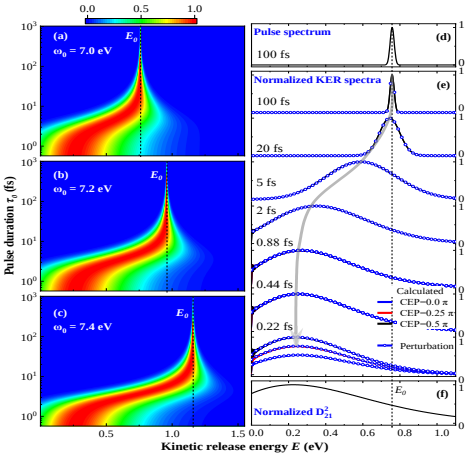

Figure 1. The One-dimensional (right) and two-dimensional (left) photofragment KER spectra.

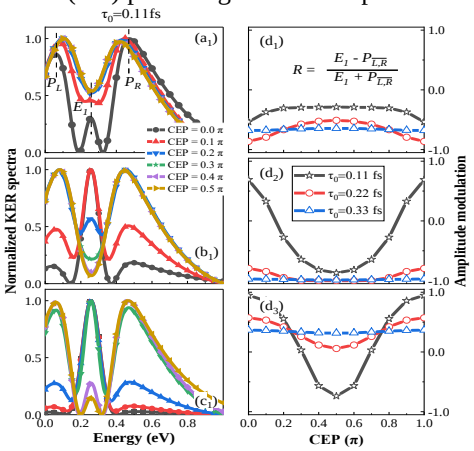

Figure 2. The normalized coherent KER spectra.

References

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Time	Wednesday, April 24	Thursday, April 25	Friday, April 26
9:00	**MPS Opening**	Oksana Travnikova	Lynda Hutcheson
9:15	Toshiyuki Azuma		
9:30		Marc Simon	Yu He
9:45	Xueguang Ren		
10:00		Mizuho Fushitani	Tomoyuki Endo
10:15	Pengju Zhang		
10:30	Photo & Coffee Break	Coffee Break	Coffee Break
11:00	Mathieu Gisselbrecht	Kade Spicer	Matthieu Génévriez
11:30	Sebastian Eckart	Nicolas Sisourat	Weifeng Yang
11:45		Anna Skitnevskaya	Eemeli Aulis Eronen
12:00	Rocio Borrego-Varillas	Bocheng Ding	Jinfeng Chen
12:15		Pufang Ma	Dongdong Zhang
12:30	Lunch	Lunch	Lunch
13:45	Sergio Díaz-Tendero	Elena Gryzlova	Hongcheng Ni
14:15	Chuncheng Wang	Jiabao Ji	Divya Bharti
14:45	Yun Li	Victor Kimberg	Keyu Chen
15:00	Lishu Wu		Huanyu Ma
15:15	Coffee Break	Coffee Break	**MPS Closing**
15:45	Arnaud Leclerc	**Poster Section**	City Center and Boat Dinner
16:15	Shenyue Xu		
16:45	Satoru Kanaya		
17:15	Alexander Zaytsev		
17:45	Snack in poster region	Banquet (18:15)	