The 5th International Symposium on Intense Short Wavelenght Processes in Atoms and Molecules (ISWAMP)
Paris, France, July 20-22, 2019

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ABSTRACTS BOOKLET

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Welcome!

This is an exciting year for the intense short wavelength community:

The European XFEL has begun operations and exciting new results are being obtained.

The 2018 Nobel prize in Physics was awarded to Gérard Mourou and Donna Strickland "for their method of generating high-intensity, ultra-short optical pulses" that are used to generate short wavelength radiation.

It is therefore a good year to attend the biennial ISWAMP meeting in Paris!

Organized as a satellite of the International Conference on Photonic, Electronic and Atomic Collisions (ICPEAC), the 5th International Symposium on Intense Short Wavelength Processes in Atoms and Molecules (ISWAMP) is held on the Cité Universitaire campus in the 'Maison Deutsch de la Meurthe' from July 20 to July 22, 2019.

Don't miss out on the opportunity to discuss the exciting new developments in the ISWAMP field through the plenary and posters sessions.

Bienvenue !

C'est une année passionnante pour la communauté intense des ondes courtes:

Le XFEL européen a commencé ses activités et de nouveaux résultats intéressants ont été obtenus. Le le prix Nobel de physique 2018 a été attribué à Gérard Mourou et Donna Strickland "pour leur méthode de génération d'intensité élevée, impulsions optiques ultra-courtes" qui sont utilisées pour générer un rayonnement de courte longueur d'onde.

C'est donc la bonne année pour assister à la réunion bisannuelle de l'ISWAMP à Paris!

Organisé en tant que satellite de la Conférence internationale sur les collisions photoniques, électroniques et atomiques (ICPEAC), le 5ème Symposium international sur les processus intenses en ondes courtes dans les atomes et molécules (ISWAMP) se tiend sur le campus de la Cité Universitaire à la Maison Deutsch de la Meurthe les 20-22 juillet 2019.

Ne manquez pas l'occasion de discuter des nouveaux développements intéressants dans le domaine ISWAMP et d'assister aux sessions plénières et posters.

Organizing Committee

Chairperson:

John Bozek	Synchrotron SOLEIL, Gif-sur-Yvette, France

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John Costello	Dublin City University, Dublin, Ireland
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Dajun Ding	Jilin University, Changchun, China
Alexei Grum-Grzhimailo	Lomonosov Moscow State University, Moscow, Russia
Igor Litvinyuk	Griffith University, Brisbane, Australia
Michael Meyer	Universität Hamburg, Hamburg, Germany
Robert Moshammer	Max-Planck-Institut, Heidelberg, Germany
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Antony Starace	University of Nebraska, Lincoln, USA
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Oksana Travnikova	LCPMR, CNRS, Sorbonne Université, Paris, France
Stéphanie Yao	Synchrotron SOLEIL, Gif-sur-Yvette, France

The 5th International Symposium on Intense Short Wavelength Processes in Atoms and Molecules (ISWAMP)

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Programme

Saturday, July 20th

11:00 – 13:50	Welcome registration, coffee
13:50 – 14:00	Opening
	Chair: **Kiyoshi Ueda**
14:00 – 14:40	Plenary talk Attosecond measurements with ultrafast X-rays: The present and future prospects ***Jonathan Marangos*** *- Imperial College, London, UK*
14:40 – 15:10	Invited talk Self-referenced attosecond streaking ***Daniel Haynes*** *- Max-Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany*
15:10 – 15:30	Attosecond Pulse Shaping at FERMI FEL ***Praveen Kumar Maroju*** *– Albert Ludwigs Universität, Freiburg, Germany*
15:30 – 16:00	*Coffee break*
	Chair: **Igor Litvinyuk**
16:00 – 16:30	Invited talk Coherent X-ray diffraction nanoscale imaging with single attosecond-short FEL pulses ***Tais Gorkhover*** *- Pulse Institute, SLAC, Menlo Park, USA*
16:30 – 17:00	Invited talk Crystalline defects in single Xe clusters probed by single-shot X-ray diffraction ***Akinobu Niozu*** *- Kyoto University, Kyoto, Japan*
17:00 – 17:30	Invited talk Measuring the atomic recoil to study stimulated X-ray emission in Ne ***Ulli Eichmann*** *- Max-Born Institute, Berlin, Germany*
17:30 – 18:00	Invited talk Tracing fragmentation dynamics upon absorption of very intense X-ray pulses ***Rebecca Boll -*** *European XFEL, Hamburg, Germany*

Sunday, July 21st

Chair: **Robert Moshammer**

09:00 – 09:40	Plenary talk Attosecond soft-X-ray spectroscopy in the gas and liquid phases ***Hans Jakob Wörner*** *- ETH Zürich, Zürich, Switzerland*
09:40 – 10:10	Invited talk Exotic dense-matter states pumped by ultra-Intense X-ray radiation of relativistic laser plasma ***Sergey Pikuz*** *- Joint Institute for High Temperatures, Moscow, Russian Federation*
10:10 – 10:40	*Coffee break*

Chair: **Alexei Grum-Grzhimailo**

10:40 – 11:10	Invited talk Time-dependent wavefunction-based methods for intense laser-driven multielectron dynamics ***Takeshi Sato*** *- University of Tokyo, Tokyo, Japan*
11:10 – 11:40	Invited talk Analytic description of high-order harmonic generation in the adiabatic limit ***Mikhail V. Frolov*** *- Voronezh State University, Voronezh, Russian Federation*
11:40 – 12:00	Attoclock on atomic and molecular hydrogen ***Anatoli Kheifets*** *- Australian National University, Canberra, Australia*
12:00 – 12:20	A semiclassical study of the holographic interference in atomic photoionization ***Diego German Arbó*** *- Institute for Astronomy and Space Physics IAFE, Buenos Aires, Argentina*
12:20 – 14:00	*Lunch*

Chair: **John Costello**

14:00 – 14:40	Plenary talk Attosecond Science: Past, Present and Future ***Marc Vrakking*** *- Max-Born Institute, Berlin, Germany*
14:40 – 15:10	Invited talk Strong-field physics in the molecular frame: imaging with short-wavelength electrons ***Jochen Küpper*** *- DESY, CFEL and Universität Hamburg, Hamburg, Germany*
15:10 – 15:30	Autoionizing Rydberg states of H_2 molecules in strong laser field ***Yonghao Mi*** *- Max-Planck Institute for Nuclear Physics, Heidelberg, Germany*
15:30 – 16:00	*Coffee break*

Chair: **Dajun Ding**

16:00 – 16:30	Invited talk Circularly polarized X-rays via high order harmonics ***André D. Bandrauk*** *– Université de Sherbrooke, Sherbrooke, Canada*
16:30 – 17:00	Invited talk Probing Rydberg-Rydberg interactions in N_2 by ultrafast EUV-NIR photoelectron spectroscopy ***Akiyoshi Hishikawa*** *- Nagoya University, Nagoya, Japan*
17:00 – 17:30	Invited Talk Nonlinear emphasizing of electron spin polarization caused by two-pathway interference in VUV photoionization ***Elena V. Gryzlova*** *- Lomonosov Moscow State University, Moscow, Russian Federation*
17:30 – 19:00	Posters Session
19:30 – 21:30	Conference dinner at La Coupole, 102 Boulevard du Montparnasse, 75014 Paris

Monday, July 22th

Chair: **Carlo Callegari**

09:00 – 09:40	Plenary talk New science opportunities at the Small Quantum Systems (SQS) Instrument at the European XFEL ***Michael Meyer*** *- European XFEL, Schenefeld, Germany*
09:40 – 10:10	Invited talk The Maloja endstation for atomic, molecular and non-linear science at SwissFEL ***Kirsten Schnorr*** *- Paul Scherrer Institut, Villigen, Switzerland*
10:10 – 10:40	*Coffee break*

Chair: **Oksana Travnikova**

10:40 – 11:10	Invited talk Status and perspective of X-ray Free Electron Lasers in China ***Zhentan Zhao*** *- Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai, China*
11:10 – 11:40	Invited talk The TMO instrument: Opportunities and plans for time-resolved atomic, molecular and optical science at LCLS-II ***Peter Walter*** *- LCLS, SLAC, Menlo Park, USA*
11:40 – 12:10	Invited talk *Echo enabled harmonic generation at the FERMI Free-Electron Laser* ***Carlo Spezzani*** *– Elettra Sincrotrone Trieste, Trieste, Italy*

12:10 – 14:00	*Lunch*
	Chair: **Marc Simon**
14:00 – 14:30	Invited talk Time-resolved photoionization and fragmentation dynamics of fullerenes studied with FELs ***Nora Berrah*** *- University of Connecticut, Storrs, USA*
14:30 – 14:50	Ultrafast buildup of magneto-electric excitations by polarization-structured laser pulses ***Jamal Berakdar*** *- Martin-Luther University, Halle-Wittenberg, Halle, Germany*
14:50 – 15:20	*Coffee break*
	Chair: **John Bozek**
15:20 – 15:50	Invited talk Complete reconstruction of bound and unbound electronic wavefunctions in two-photon double ionization ***Paolo Antonio Carpeggiani*** *- Technische Universität Wien, Wien, Austria*
15:50 – 16:10	Nonsequential two-photon ionization of inner-shell electrons in neutral atoms ***Andrey V. Volotka*** *- Helmholtz-Institut Jena, Jena, Germany*
16:10 – 16:20	*Conclusion*

ABSTRACTS

Saturday, July 20th, 2019

The 5th International Symposium on Intense Short Wavelenght Processes in Atoms and Molecules (ISWAMP)
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Saturday, July 20th

Chairs: **Kiyoshi Ueda and Igor Litvinyuk**

PT-01	Attosecond measurements with ultrafast X-rays: The present and future prospects ***J. Marangos***
IT-01	Self-referenced attosecond streaking ***D. Haynes***
OC-01	Attosecond pulse shaping at FERMI FEL ***P.K. Maroju***
IT-02	Coherent X-ray diffraction nanoscale imaging with single attosecond-short FEL pulses ***T. Gorkhover***
IT-03	Crystalline defects in single Xe clusters probed by single-shot X-ray diffraction ***A. Niozu***
IT-04	Measuring the atomic recoil to study stimulated X-ray emission in Ne ***U. Eichmann***
IT-05	Tracing fragmentation dynamics upon absorption of very intense X-ray pulses ***R. Boll***

Attosecond Measurements with Ultrafast X-rays – The Present and the Future Prospects

J. Marangos

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ABSTRACT

We will motivate and describe recent progress on measuring non-adiabatic molecular dynamics associated with sudden ionization or excitation of molecules and polymers and unfolding on timescales from sub-femtosecond to few-femtosecond. First we will discuss the physical circumstances and nature of these processes, especially in terms of quantum coherence and decoherence, and then set-out what is required from incisive measurements to strengthen our understanding of these fundamental photo-physical events.

Three methods have been used in our recent research based on ultrafast strong-field optical lasers and ultrafast X-ray lasers. In the first of these we have studied dynamics of strong field molecular ionization in some prototypical polyatomic organic molecules using an extension of the chirp-encoded high harmonic spectroscopy technique we introduced some years ago[2]. Using an 1800 nm OPA system to overcome the limits set by low ionization saturation intensity to HHG spectroscopy in organic molecules[3] we have developed a two-dimensional technique in which scans of HHG spectrum versus laser intensity can be used to extract electronic and nuclear dynamics in strong-field ionized benzene and substituted benzenes. By hollow fibre pulse compression of our OPA and optimization of HHG generation conditions in Ne and He gas we have demonstrated the generation of soft X-ray attosecond pulses with flux sufficient for XANES spectroscopy up to the O K edge at 540 eV[4]. This source was used to study the X-ray spectrum of an organic-optoelectronic polymer (P3HT)[5] and we are now carrying out time resolved measurements to study the dynamics of exciton formation and decay in these materials.

The problem of measuring transient hole dynamics, arising from electron correlation driven processes such as charge migration, following sudden X-ray ionization of a molecule has long been an open question in attosecond science. We have developed a direct measurement concept of such transient hole dynamics[6] that we have recently demonstrated in isopropanol using an X-ray pump-X-ray probe method at LCLS employing the fresh slice two colour/two pulse mode. We report the first results on the evolution of an inner valence hole state in this molecule. Future prospects for sub-femtosecond resolved linear and non-linear X-ray measurements using X-ray FELs will be motivated by very recent results from our collaborations at LCLS.

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[4] A.S.Johnson *et al, Science Advances*, **4**, 3761 (2018)
[5] A.S.Johnson *et al, Structural Dynamics*, **3**, 062603 (2016)
[6] B.Cooper *et al, Faraday Discussion*, **171**, 93 (2014)

Self-referenced Attosecond Streaking

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ABSTRACT

With the advent of X-ray free-electron lasers (XFELs), it is now possible to investigate and manipulate matter in novel new ways[1]. Exotic states of matter, such as double-core-excited molecules, can only be realised and studied at XFELs. Pioneering nonlinear spectroscopic techniques which exploit these new possibilities hold tremendous potential for ever-deeper interrogation and understanding of fundamental processes[2,3].

However, these states are typically constrained by the lifetime of the core-hole, which for many systems relaxes predominantly via Auger decay. Although the Auger decay lifetime has been inferred from spectral linewidths[4], and the decay process has been time-resolved using table-top X-ray sources[5], it has never been possible to measure it *in situ* – that is, in the highly-excited XFEL-pumped environment used to create and manipulate exotic states. Whilst XFEL pulses are in principle short enough to interrogate sub-femtosecond dynamics[6], timing jitter precludes the application of established techniques such as streaking spectroscopy, which might be used to measure ultrafast electron dynamics including Auger decay.

Here, we present a novel technique which will enable the full potential of XFELs to be realised: Self-referenced attosecond streaking. This approach permits the extension of table-top attosecond spectroscopy to large scale FEL facilities, despite ubiquitous timing and phase jitter. The technique exploits the macroscopic trends present in an entire dataset: By constructing a map of the interaction of two temporally distinct peaks with the streaking field, one can identify the amplitude and phase of that field on a single-shot basis, even when those parameters were uncontrolled during the experiment.

In the first demonstration of this method, we report measurements of the Auger decay lifetime in neon, observing a lifetime of $3.5 \pm \frac{0.3}{0.5}$ fs for the KLL decay channel. To our knowledge, this measurement and its unprecedented sub-femtosecond precision represent the fastest event ever to be temporally resolved at an XFEL. This achievement paves the way for further study of the electron dynamics which drive highly excited states of matter.

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Attosecond Pulse Shaping at FERMI FEL

P.K. Maroju[1] , C. Grazioli[2], M. Di Fraia[3] , M. Moioli[1], D. Ertel[1], H. Ahmadi[1], O. Plekan[3], P. Finetti[3], E. Allaria[3] , L. Giannessi[3,4], G. De Ninno[3,5], C. Spezzani[3], G. Penco[3], A. Demidovich[3], M. Danailov[3] R. Borghes[3], G. Kourousias[3], C.E. Sanches Dos Reis[3], F. Billé[3], A.A. Lutman[6], R.J. Squibb[7], R. Feifel[7], P. Carpeggiani[8], M. Reduzzi[9], T. Mazza[10], M. Meyer[10], S. Bengtsson[11], N. Ibrakovic[11], E.R. Simpson[11], J. Mauritsson[11], T. Csizmadia[12], M. Dumergue[12], S. Kühn[12], N.G. Harshitha[12], D. You[13], K. Ueda[13], M. Labeye[14], J. Egebjerg Bækhøj[14] , K.J. Schafer[14], Elena V. Gryzlova[15], A.N. Grum-Grzhimailo[15], K.C. Prince[3], C. Callegari[3], and G. Sansone[1]

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ABSTRACT

Free-Electron Lasers (FELs), which can generate radiation in extreme ultraviolet and X-ray range along with unprecedented intensities, opened up opportunities for investigation of the valence and core electron dynamics. The temporal coherence and the high intensities have made possible the single-shot diffraction [1] imaging of non-crystalline samples. However, until recently the longitudinal coherence was not demonstrated which is essential for coherent control experiments [2]. In this work, we present the first demonstration of generation and characterisation of Attosecond Pulse Trains (APTs) at the seeded FEL FERMI exploiting the ability to produce multiple phase locked harmonics. We also demonstrate the ability to manipulate the amplitude and phase of the generated APTs independently.

REFERENCES

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Coherent X-ray Diffraction Nanoscale Imaging with Single Attosecond-short FEL Pulses

T. Gorkhover

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Crystalline Defects in Single Xe Clusters Probed by Single-shot X-ray Diffraction

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ABSTRACT

Characterizing crystalline defects in nanoparticles is of crucial importance for their applications in science and technology, since the defects potentially alter their desired properties. Single-shot X-ray diffraction with X-ray free electron lasers (XFELs) is a promising method to investigate the structure of nanoparticles, in which diffraction patterns of single nanoparticles are obtained with exposures to single XFEL pulses.

In this study, we carried out a wide-angle X-ray scattering experiment on Xe clusters at an XFEL facility SACLA[1] in Japan. Single Xe clusters (average radius r ~ 65 nm) were irradiated by 11.2-keV XFEL pulses, and the diffracted X-rays were recorded on a shot-by-shot basis with a multiport CCD sensor detector. The diffraction patterns contain Bragg spots corresponding to fcc {111}, {200}, {220} and hcp {101} reflections. In order to extract detailed structural information from the diffraction patterns, we developed a new analysis method that focuses on angular correlations[2] between plural Bragg spots in single-shot diffraction patterns. The analysis has revealed clear angular correlations between the Bragg spots, which provide rich structural information on the clusters. We carried out a simulation to quantitatively evaluate the angular correlations and obtained evidence of stacking faults in the Xe clusters.

This work was done in collaboration with Y. Kumagai, T. Nishiyama, H. Fukuzawa, K. Motomura, M. Bucher, Y. Ito, T. Takanashi, K. Asa, Y. Sato, D. You, Y. Li, T. Ono, E. Kukk, C. Miron, L. Neagu, C. Callegari, M. Di Fraia, G. Rossi, D. E. Galli, T. Pincelli, A. Colombo, T. Kameshima, Y. Joti, T. Hatsui, S. Owada, T. Katayama, T. Togashi, K. Tono, M. Yabashi, K. Matsuda, C. Bostedt, K. Nagaya and K. Ueda. This study was supported by the X-ray Free Electron Laser Utilization Research Project and the X-ray Free Electron Laser Priority Strategy Program of the MEXT, by JSPS, the Proposal Program of SACLA Experimental Instruments of RIKEN.

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2. D. Mendez et al., *IUCrJ* **3**, 420-429 (2016).

Measuring the Atomic Recoil to Study Stimulated X-ray Emission in Ne

U. Eichmann

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ABSTRACT

In this talk I will discuss our recent experiment at the European XFEL in Hamburg, where we investigated stimulated X-ray emission in Ne. Rather than detecting charged particles or scattered light we use a different experimental approach by focusing on those atoms that survive the interaction with the XFEL in a bound excited (metastable) state. Our experimental setup allows for the measurement of the deflection of the atoms, which is induced by the photon momentum transfer. As a result we are able to clearly distinguish spontaneous radiative processes from recoilless stimulated X-ray emission (a two-photon process) at a single atom level.

In the experiment we excited Ne atoms in the vicinity of the 1s ->3p inner shell transition, which then either decay radiatively on the 2p->1s transition in a spontaneous process to a bound excited state or stimulated emission is enforced by the XFEL pulse (near resonant Raman process) leaving the atom in a bound excited state. Our experimental findings, particularly the ratio of stimulated versus spontaneous emission as a function of the XFEL photon energy can be explained quantitatively within a three-level density-matrix formalism. Further analysis shows that the ratio depends significantly on the time and spectral structure of the XFEL pulses.

The novel experimental concept opens up perspectives on further experiments on nonlinear processes in the X-ray regime, which will be discussed.

Tracing Fragmentation Dynamics upon Absorption of Very Intense X-ray Pulses

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ABSTRACT

X-ray free-electron lasers (XFELs) have opened up unprecedented possibilities in different scientific areas. Key features of FEL radiation are the very short pulse duration and the high number of photons per pulse, leading to extreme intensities. When isolated molecules absorb an intense, femtosecond X-ray pulse, a complex interplay of multiple inner-shell photo absorptions, Auger decays, and nuclear motion takes place. All processes happen on timescales similar to the pulse duration.

The Small Quantum Systems (SQS) instrument at the soft X-ray branch of the European XFEL is dedicated to the investigation of nonlinear phenomena and ultrafast dynamics of gas-phase atoms, molecules, and nanoparticles. A wide range of experimental techniques for spectroscopy using electrons, ions and photons are available. User operation started in November 2018.

In one of the first user experiments, we have investigated the charge rearrangement and fragmentation dynamics of polyatomic molecules upon absorption of very intense soft X-ray pulses. Of particular interest were molecules containing a single heavy atom that acts as a localized absorber due to the strongly increased absorption cross section. Under typical experimental conditions at SQS, extremely high charge states have been observed in isolated heavy atoms, for example, up to 42 charges in a xenon atom. In a heavy-atom containing molecule, the charge is initially located on the heavy atom, but can spread to the other atoms in the molecule very quickly, thus charging up the complete molecule, which subsequently undergoes Coulomb explosion. Its fragmentation can be experimentally observed by means of 3d ion momentum imaging in a reaction microscope (REMI). Due to the high repetition rate of the European XFEL, several ions originating from the same molecule can be detected in coincidence, and kinetic energies and angles between ions can be determined. In this way, we were able to observe a complete picture of the rich dynamics occurring in a molecule with eleven atoms. We see indications that protons are emitted very early in the X-ray pulse, and that the pulse duration may have been significantly shorter than in our earlier experiments at the LCLS using soft and hard X-rays [1-5].

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ABSTRACTS

Sunday, July 21st, 2019

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Paris, France, July 20-22, 2019

Sunday, July 21st

Chairs: **Robert Moshammer, Alexei Grum-Grzhimailo, John Costello and Dajun Ding**

PT-02	Attosecond soft-X-ray spectroscopy in the gas and liquid phases ***H. J. Wörner***
IT-06	Exotic dense-matter states pumped by ultra-Intense X-ray radiation of relativistic laser plasma ***S. Pikuz***
IT-07	Time-dependent wavefunction-based methods for intense laser-driven multielectron dynamics ***T. Sato***
IT-08	Analytic description of high-order harmonic generation in the adiabatic limit ***M. V. Frolov***
OC-02	Attoclock on atomic and molecular hydrogen ***A. Kheifets***
OC-03	A semiclassical study of the holographic interference in atomic photoionization ***D. G. Arbó***
PT-03	Attosecond science: Past, present and future ***M. Vrakking***
IT-09	Strong-field physics in the molecular frame: Imaging with short-wavelength electrons ***J. Küpper***
OC-04	Autoionizing Rydberg states of H2 molecules in strong laser field ***Y. Mi***
IT-10	Circularly polarized X-rays via high order harmonics ***A. D. Bandrauk***
IT-11	Probing Rydberg-Rydberg interactions in N2 by ultrafast EUV-NIR photoelectron spectroscopy ***A. Hishikawa***
IT-12	Nonlinear emphasizing of electron spin polarization caused by two-pathway interference in VUV photoionization ***E. V. Gryzlova***

Attosecond Soft-X-ray Spectroscopy in the Gas and Liquid Phases

H. J. Wörner

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ABSTRACT

Attosecond time-resolved spectroscopy has the potential to address fundamental open questions in chemical sciences. Although the first two decades of research have led to very important advances towards this goal, the techniques of attosecond spectroscopy still need to overcome two gaps. The first gap is the complexity gap, i.e. the challenge of applying attosecond spectroscopy to complex molecules. A promising approach to overcome the complexity gap consists in exploiting the element-, site- and spin-sensitivity of X-ray spectroscopy. We have recently demonstrated the potential of table-top X-ray absorption spectroscopy with a water-window high-harmonic source, observing the temporal evolution of unoccupied molecular orbitals, as well as molecular shape resonances during chemical reactions [1]. Compressing the mid-infrared driving pulses to less than 2 optical cycles, we have demonstrated the extension of this table-top source to fully cover the oxygen K-edge with fluxes sufficient for time-resolved measurements [2]. Using the same technique, we have also demonstrated the generation of isolated attosecond pulses, which have established a new record of the shortest light pulses ever measured (43 attoseconds) [3].

The second gap is the extension from the gas to the liquid phase, which is the relevant to the vast majority of chemical and biophysical processes. I will discuss the first observation of extreme-ultraviolet high-harmomic generation from liquids, achieved through the application of ultrathin (0.6-2 μm) flat microjets [4]. Exploiting the geometric separation of liquid- and gas-phase harmonics inherent to our experimental geometry, we have observed a nearly linear scaling of the high-harmonic cut-off with the driving electric field, a non-perturbative scaling of all high-harmonic orders and a consistently broader ellipticity dependence of the liquid-phase harmonics compared to the gas-phase harmonics. Finally, I will present the first extension of attosecond time-resolved spectroscopy from molecules [5] to liquids [6], reporting on photoemission delays of liquid compared to gaseous water. The measured time delays range from 50-70 attoseconds and are shown to mainly originate from the solvation of water molecules, with liquid-phase electron scattering playing a minor role. These combined developments set the stage for attosecond time-resolved studies of molecular systems of chemical complexity.

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Exotic Dense-matter States Pumped by Ultra-Intense X-ray Radiation of Relativistic Laser Plasma

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Time-dependent Wavefunction-based Methods for Intense Laser-driven Multielectron Dynamics

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ABSTRACT

High-field physics and ultrafast science are rapidly progressing toward a world-changing goal of direct measurement and control of the electron motion in matters. To theoretically investigate intense laser-driven multielectron dynamics, the multiconfiguration time-dependent Hartree-Fock method [1-3] has been developed, which, though powerful, suffers from exponential increase of the computational cost with respect to the number of electrons.

To overcome this difficulty, more cost-effective time-dependent multiconfiguration methods have been developed [4-7], which substantially extend the applicability of accurate multielectron simulations. Furthermore, we have recently succeeded in formulating time-dependent coupled-cluster method using time-dependent orbitals, called TD-OCC method [8], which, based on Kvaal's pioneering work [9], realizes gauge-invariant (equivalent for length-gauge and velocity-gauge treatment of laser-matter interaction) and size-extensive (uniform accuracy for increasing number of particles) description of multielectron dynamics.

In this talk, I will summarize theory, and present our full three-dimensional implementation of the methods for atoms and molecules [10-12]. The efficient implementation enabled *ab initio* description, beyond single-active-electron approximation or time-dependent density functional theory, of high-field phenomena directly relevant to experiments, e.g, nonsequential double ionization of noble gas atoms, high-harmonic generation (HHG) enhanced by laser-induced electron recollision [13, 14], and laser polarization-dependent multichannel and multielectron effects on HHG from molecules.

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Analytic description of high-order harmonic generation in the adiabatic limit

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Synopsis Analytic description of high-order harmonic generation is discussed in terms of *real* classical trajectories. Based on the self-consistent analytical model in the adiabatic limit we show how these trajectories can be introduced and parameterized in terms of *real* starting and returning times. The accuracy and applicability of suggested description are discussed for bicircular few-cycle laser pulses.

The theoretical description of high harmonic generation (HHG) (as well as other nonlinear processes in a strong nonperturbative laser field) encounters several obstacles, which remain a challenge even a half-century after the advent of strong field physics. The key challenge is the need to describe the nonperturbative laser field on the same footing as the field of the ionic core, as together they govern the electron dynamics. This can be achieved by numerical solution of the time-dependent Schrodinger equation (TDSE), however numerical simulations are feasible only in limited ranges of the laser parameters and thus it is not predictive so far.

The ability of analytical methods for description HHG are much wider, however almost all of them have drawbacks, which restrain their application in the original form to the real system. Indeed, the original strong field approximation (SFA) presents transition amplitude in the form of Born-series in atomic potential and thus electronic dynamics may be considered only in the plane-wave approximation. Moreover, effects of long-range Coulomb potential are not taken into account correctly in all three steps of harmonic formation [1]. Nevertheless, the SFA led to a very important insight into the theoretical description of strong field phenomena, namely, the applicability of the quantum orbit approach (QOA). In terms of the QOA, the HHG amplitude is represented as a sum of partial amplitudes, each of which is associated with a *complex* closed electron trajectory in the laser field. The QOA provides a natural means of including the Coulomb-induced corrections to the HHG amplitude within quasiclassical perturbation theory.

The study of strong field phenomena has also been greatly advanced by exactly solvable analytical models. Here we shall discuss, our recently develop an analytic description of the HHG amplitude for the case of a laser field having an arbitrary spatial and temporal wave form. This description is based on the analytical model and low-frequency approximation [2]. Suggested analytical model allows to consider more accurately the effects of an atomic potential in a strong laser field, while low-frequency approximation helps to estimate transition amplitude in the tunneling (or adiabatic) limit and introduce Coulomb-induced corrections. By applying this model and low-frequency approximation we present the laser-induced dipole moment as a coherent sum of partial dipole moments, whose properties (direction, phase, and magnitude) are determined by the classical (*real*) times of ionization and recombination. These times determine the closed classical trajectories along which the ionized electron starts, with minimal kinetic energy, in the laser field and returns back with the kinetic energy corresponding to harmonic emission with frequency Ω. The partial dipole moment can be written as a product of three factors: the ionization factor, the propagation factor, and the exact photorecombination amplitude. We test the accuracy of this theory by comparison with results of numerical solution of the 3D time-dependent Schrödinger equation for time-delayed, few-cycle ω and 2ω counterrotating mid-IR pulses and discuss features of HHG in the bicircular field [3].

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Attoclock on Atomic and Molecular Hydrogen

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ABSTRACT

The attoclock measurements of the tunneling time have attracted a very considerable attention [1]. These measurements and further experiments [2] could be interpreted in terms of a finite tunneling time whereas the theory [3,4] firmly pointed to instantaneous tunneling. In the latest development, the attoclock offset angle was wholly attributed to the Coulomb field effect [5] and the hydrogen atom measurement returned the upper bounds on the tunneling time not exceeding 2 attoseconds [6]. My presentation will highlight these and other recent developments. In particular, the new set of attoclock measurements and calculations on the molecular hydrogen [7] will be presented and discussed. The estimate of the tunneling time in H_2 obtained in this work does not exceed 10 attoseconds.

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A Semiclassical Study of the Holographic Interference in Atomic Photoionization

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ABSTRACT

We show novel results on the interference structure in atomic ionization by near-infrared lasers. A theoretical study of the interference pattern imprinted on the doubly differential momentum distribution of the photoelectron due to atomic ionization induced by a short laser pulse is developed from a semiclassical standpoint. We use the semiclassical two-step model of Shvetsov-Shilovski *et al.*[1] to elucidate the nature of the holographic structure. Three different types of trajectories are characterized during the ionization process by a single cycle pulse with three different types of interferences. We show that the holographic interference arises from the ionization yield only during the first half cycle of the pulse, whereas the coherent superposition of electron trajectories during the first half cycle and the second half cycle gives rise to two other kinds of intracycle interference. Although the picture of interference of a reference beam and a signal beam is adequate, we show that our results for the formation of the holographic pattern agree with the glory rescattering theory of Xia *et al.*[2]. We probe the two-step semiclassical model by comparing it to the numerical results of the time dependent Schrödinger equation.

We show a successful way of how to scrutinize quantum interferences taking place in the subfemtosecond domain. We use both quantum and semiclassical models to show that our results essentially agree with the results of the glory rescattering trajectories of Xia *et al*. We identify the glory trajectories as the limiting ones between rescattering (nearside) and non-rescattering (farside) trajectories [3]. However, the picture of interference of a reference beam and a signal beam still persists. Besides the well-known holographic interference, there are two other types of intracycle interferences corresponding to different types of electron trajectories involved in the process: trajectories that rescatter and that do not. The general appearances of these types of intracycle interferences differs from the found in the literature (essentially using the strong field approximation) due to our accurate way to include the effect of the Coulomb potential of the remaining core [4].

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Attosecond Science: Past, Present and Future

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ABSTRACT

The last decade has witnessed spectacular progress in the way that attosecond laser pulses generated by high-harmonic generation (HHG) are used to study time-resolved electron dynamics. Starting from an initial emphasis on the generation and characterization of attosecond laser pulses, the field of attosecond science has within the last few years witnessed a dramatic growth in the number of application areas where attosecond pump-probe spectroscopy is pursued, or where techniques that have been developed in the context of the emergence of attosecond science are used to great advantage. A particularly significant example of the latter is the widespread use of time-resolved core-level spectroscopy using large bandwidth extreme ultra-violet (XUV) and soft X-ray pulses, which is finding application in both gas phase and condensed phase experiments.

In my talk I will present an overview of these developments, using a number of experiments that were recently performed at the MBI as illustration. I will discuss, among other things, experiments where femtosecond and attosecond time-resolved core level spectroscopy is used to investigate photodissociation dynamics and field-driven electron dynamics in the CH3I molecule[1, 2], as well as experiments on magnetization dynamics in thin Co/Pt films[3] and experiments on the coupling of electronic and nuclear motion in ionic crystals[4]. Moreover, I will show how the time seems ripe for novel applications of time-resolved core level spectroscopy in the condensed phase that venture deeply into the water window[5].

At the same time, attosecond seems to be on the verge of significantly extending its arsenal of available techniques. Besides the emerging possibilities to generate attosecond pulses at free electron laser facilities, these opportunities include, on the laboratory-scale, the development of attosecond pump-probe spectroscopy at high repetition rate[6-8], permitting the use of coincidence techniques, the realization of attosecond pump-attosecond probe spectroscopy, using high-harmonic sources that are able to reach previously inaccessible intensities[9], as well as novel ways to tailor the spatio-temporal distribution of XUV light produced by HHG[10].

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Strong-field Physics in the Molecular Frame: Imaging with Short-wavelength Electrons

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ABSTRACT

The interaction of strong laser fields with matter intrinsically provides powerful tools, such as laser-induced electron diffraction, to image transient dynamics with an extremely high spatiotemporal resolution. In strong-field physics, the initial conditions of this interaction are generally considered a weak perturbation. We investigated strong-field ionization of stateselected and strongly laser-aligned molecules and showed, for the first time, a full real-time picture of the photoelectron dynamics in the presence of the laser field and the molecular interaction. We demonstrated that the molecular potential defines the initial conditions of the photoelectron at birth and has a dramatic impact on the overall strong-field recollision dynamics: it sets the clock for the emission of electrons with a given rescattering kinetic energy. This result represents a new benchmark for the seminal statements of molecularframe strong-field physics. Our findings have strong impact on the interpretation of selfdiffraction experiments, where the photoelectron momentum distribution is used to retrieve molecular structures. Furthermore, the resulting encoding of the time-energy relation in molecular-frame photoelectron distributions shows the way of accessing a deeper understanding of electron transport during strong-field interactions and probing the molecular potential in real-time.

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https://www.controlled-molecule-imaging.org/publication/scientific

Autoionizing Rydberg States of H_2 Molecules in Strong Laser Field

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ABSTRACT

When an H_2 molecule is exposed to a strong laser field, the following two single ionization channels may occur: 1) bound ionization, in which one electron is freed by the laser field and the molecule ends up as a bound molecular ion and a free electron, and 2) dissociative ionization, where the H_2 molecule breaks up into a proton, a hydrogen atom and a free electron. The Born-Oppenheimer approximation gives rise to the two-step mechanism [1], which describes the laser-induced dissociation of H_2 molecules as follows: the molecule is ionized by the laser field in the first step and then dissociates as the vibrational wave packet is excited from the bound $1s\sigma_g$ state to the repulsive $2p\sigma_u$ state, which means the dissociative ionization is independent from the bound ionization. On first approximation, the electrons from the two channels should thus be the same.

By using a reaction microscope [2] and ultrashort laser pulses, we investigated channel-selective electron emission from strong-field ionization of H_2 [3]. Measuring electrons and ions in coincidence, we distinguished the bound and dissociative ionization channels and extracted electron energy spectra separately for the two channels. We observed the electron yield from bound ionization (blue curve) exhibited an enhancement in the low-energy (0-0.6 eV) spectrum. In the high-energy region, both normalized spectra agree within our experimental resolution. The difference of bound and dissociative ionization in low-energy electron spectra proves that the two-step model is not complete.

We interpret this enhancement in the electron yield for bound ionization as being due to the population and subsequent decay of autoionizing states. For H_2 molecules, the autoionzing states represent a series of high-lying Rydberg states and are characterized by a simultaneous electronic and vibrational excitation. Instead of populating the H_2^+ bound state and being excited to the repulsive state for dissociation, the molecule can be excited to the above-mentioned autoionizing state located above the ionization limit. Therefore, an autoionization process occurs, in which the vibrational energy of the nuclei is transferred to the excited electron, which is freed and appears in the electron energy spectrum.

The vibrational autoionization is a process of electronic-nuclear coupling after strong-field excitation, which may be used in the future to better understand the breakdown of the Born-Oppenheimer approximation.

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Circularly Polarized XRays via High Order Harmonics

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ABSTRACT

Attosecond(1 asec=10**-18s) pulses at photon energies corresponding to the fundamental edge of matter,the soft XRay region about 200 eV permit the probing,imaging of electron dynamics from atoms,to molecules and solid state.A soft XRay pulse duration of 43 asec has now been achieved using intense femtosecond (1fs=10**-15s) linearly polarized driving pulses [1].The main source of linear polarization asec pulses is high order harmonic generation,.HHG, a highly nonlinear nonperturbative response of bound electrons to ultrashort(fs) intense(I>10**14 W/cm**2) mid-IR laser pulses.HHG is now understood as due to laser induced electron recollision of an ionized electron with its parent cation[2].Recollision is suppressed in circular polarization whereas as shown as early as 1995 combinations of bichromatic counter-rotating circularly polarized intense ionizing pulses with frequencies w1/w2=n1/n2 for n integer induce recollision and copious harmonics[3-4] of circular polarization[5].Circular bichromatic HHG has shown to be **universal** with a first maximum intensity up to photon energies Ip+2Up and a maximum energy cut-off at Ip+3.17Up where Ip=ionization potential, ponderomotive energy Up depends at intensity I as (in atomic units) I/4w**2,w=(w1+w2)/2 [6].High level simulations performed with molecular TDSE,s(Time Dependent Schroedinger Equations) show that circularly polarized HHG, from which one can generate asec circular pulses is most efficient when the net symmetry of intense high frequency bichromatic pulses is compatible with molecular symmetry[7].This allows for the generation of circular quantum electronic currents ,new sources of asec magnetic field pulses[6].The new ability to generate circularly polarized XRay asec pulses based on circular HHG in a table top set-up[8] is currently helping to develop a new X-Ray light science such as circular polarized Laser Induced Electron Diffraction ,LIED,for the imaging of attosecond electron dynamics in molecules[9].

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Probing Rydberg-Rydberg Interactions in N_2 by Ultrafast EUV-NIR Photoelectron Spectroscopy

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ABSTRACT

The ultrafast dynamics of molecular nitrogen (N_2) just below the ionization threshold [1,2] has been investigated by time-resolved photoelectron spectroscopy using a single harmonic centered at hv= 15.38 eV. The evolution of the Rydberg wavepacket launched by the ultrashort EUV pulse is probed by a time-delayed femtosecond NIR laser pulse. The observed photoelectron spectra show two series of vibrational peaks to the ground $X^2\Sigma_g^+$ state and the first excited $A^2\Pi_u$ state of N_2^+. Among these, two photoelectron peaks with the vibrational quantum numbers $v_X^+ = 4$ and $v_A^+ = 1$ exhibit clear anti-phase oscillation with a period of 300 fs (Fig.1), showing that two Rydberg states converging to the $X^2\Sigma_g^+$ and $A^2\Pi_u$ ionic states interact with each other, thus causing periodic switching in the population of the ion core states [3].

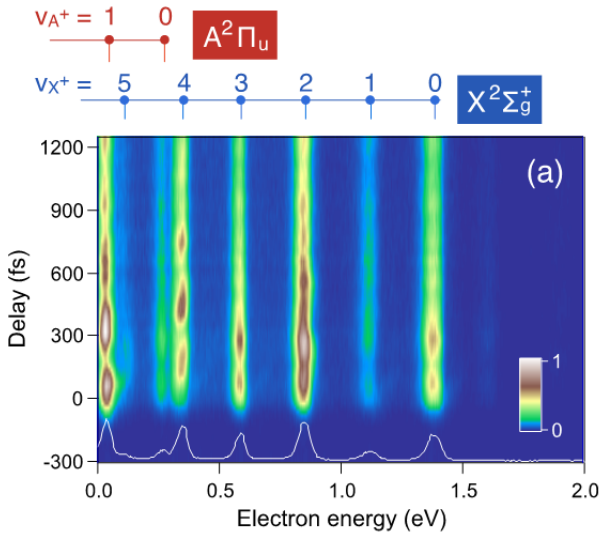

Figure 1. EUV-NIR photoelectron spectrogram of Rydberg N_2 plotted with the pump-probe time delay Δt and the photoelectron energy. Shown on the bottom is the photoelectron spectrum recorded at Δt = 1000 fs. The peak assignments are shown on the top.

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Nonlinear Emphasizing of Electron Spin Polarization Caused by Two-pathway Interference in VUV Photoionization

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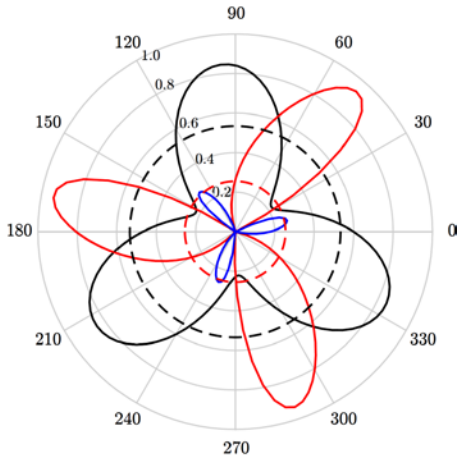
ABSTRACT

Photoelectron spin polarization is a fundamental phenomenon in atomic physics [1]. It is a powerful tool of photoelectron spectroscopy and is of great importance for many applications. Detection of the spin polarization may reveal tiny details of atomic structure and dynamics of interaction of radiation with matter. Main reasons of scarce studies of the photoelectron spin polarization in experiments with isolated atoms are related to the crucial decrease of efficiency of spin-sensitive detectors and the fact that essential spin polarization is often reached at photon energies in the minima of the cross sections. The advent of FELs generating brilliant coherent XUV pulses with variable polarization may open new avenues in studies, control, and applications of the photoelectron spin polarization.



PAD (solid black, arb. un.) and degree of spin polarization in bichromatic ionization with coherent (blue and red solid) and incoherent (dashed) harmonics. Section is given in the plane perpendicular to the field propagation direction.

We consider ionization of an atom by combined fields of the fundamental laser frequency (ω) and its second (2ω) harmonic. By manipulating relative polarization, phase and strengths of the harmonics one may efficiently control the photoelectron spin polarization. This possibility was recently discussed for alkali atoms in the optical range [2] and the first results are available for photoprocesses in condensed matter [3]. Control is achieved due to interference between two-photon (by ω) and one-photon (by 2ω) ionization pathways. An example of photoelectron angular distribution (PAD) and spin polarization in such bichromatic ionization of neon by circularly polarized harmonics (ω=16.67 eV) is presented in the figure. We consider cases when harmonics are either incoherent (PAD and spin polarization are axial symmetric) or they are coherent. In the latter case the PAD shows a three-lobe pattern, and in some directions the degree of spin polarization becomes nearly three times larger than for the incoherent beams, and approaching unity Noteworthy, maximum of the spin polarization occurs not in a minima of the cross section, thus it makes it easier to observe.

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ABSTRACTS

Monday, July 22nd, 2019

The 5th International Symposium on Intense Short Wavelenght Processes in Atoms and Molecules (ISWAMP)
Paris, France, July 20-22, 2019

Monday, July 22th

Chairs: **Carlo Callegari, Oksana Travnikova, Marc Simon and John Bozek**

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IT-13	The Maloja endstation for atomic, molecular and non-linear science at SwissFEL ***K. Schnorr***
IT-14	Status and perspective of X-ray Free Electron Lasers in China ***Z. Zhao***
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New Science Opportunities at the Small Quantum Systems (SQS) Instrument at the European XFEL

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ABSTRACT

The Small Quantum Systems (SQS) scientific instrument at the European XFEL dedicated to investigations on atoms, molecules, clusters and nanoparticles started user operation at the end of 2018. The instrument is located at the SASE3 soft X-ray undulator, which produces intense FEL radiation in the photon energy range between 250 eV and 3000 eV. In the first months of operation the accelerator was delivering up to 2000 pulses per second to the SQS experimental station with pulse energies between 1 and 5 mJ. These characteristics in combination with a pulse duration of about 25 fs provide an ideal basis for numerous investigations involving site-selective core excitations and focusing on non-linear phenomena and the dynamics of ultrafast processes.

In the talk, the results obtained in the commissioning as well as in the first campaign of user operation will be reviewed highlighting the excellent performances of the SQS instrument. Three experimental stations, AQS (Atomic-like Quantum Systems), NQS (Nano-size Quantum Systems) and REMI (REaction MIcroscope), which are optimized for different samples and different type of experiments, were successfully brought into operation and are available for the scientific community. The small spot size of about 1 micron achieved in the interaction region with a set of highly polished mirrors favors investigations of multi-photon processes. Multiple ionization, above threshold ionization (ATI) and double core hole (DCH) formation in atoms and molecules were investigated by high-resolution electron and electron-ion coincidence spectroscopy using the AQS and REMI endstation, respectively, and taking special advantage of the high number of pulses available at European XFEL. In addition, remarkable new studies on resonances excited by one- and two-photon processes and of the subsequent relaxation dynamics were made possible by the easy wavelength tunability of the SASE3 undulator. For larger systems, such as clusters and nanoparticles, mass-resolved ion spectra upon excitation with intense XUV radiation a well as the first diffraction patterns were recorded using a pnCCD 2D imaging detector and marking the promising opening for this type of investigations.

For the upcoming experiments, a synchronized optical laser will be available for time-resolved studies of dynamical processes. Pulse durations down to 15 fs and a synchronization of better than 10fs between the FEL and the optical laser pulses are envisaged. The extension of the present performances will be centered around the production of circularly polarized FEL radiation and the operation in two-color modes, i.e. the production of two different XUV wavelength with variable temporal delays, enabling various XUV-XUV pump probe experiments.

The Maloja Endstation for Atomic, Molecular and Non-linear Science at SwissFEL

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ABSTRACT

A new endstation for atomic, molecular and non-linear (AMO) science, Maloja, is currently built at the upcoming soft X-ray branch Athos [1] of the Swiss Free-Electron Laser (FEL) SwissFEL. The Athos undulator section has small chicanes in between every undulator segment and tuneable undulators, enabling novel and advanced FEL operation modes. Specifically, a mode to generate ultrashort pulses from a few femtoseconds down to hundreds of attoseconds, high-power operation with up to 8 mJ pulse energy at 250 eV, two-color X-ray pump-probe, or the combination of these schemes will be available. Exploiting these capabilities opens new exciting opportunities for the field of AMO science. For instance, charge transfer and migration in molecules shall be investigated on the few-femtosecond time scale with site-selectivity employing X-ray pump-probe schemes with pulses tuned to specific absorption edges. With a photon-energy range from 250 to 1800 eV, Athos covers essential absorption K-edges, such as C, N and O as well as L-edges of 3d transition metal elements, like Ti, Mn, Fe, and Cu to allow for a broad spectrum of targets. In addition to a synchronized optical pump-probe laser, which is intended to cover the wavelength regime from THz to UV radiation using secondary sources, a high-harmonic generation source is planned to be implemented into the endstation, in order to extend the available photon energies into the XUV regime. Thus, optical-light driven valence processes as well as XUV-initiated valence and inner-valence dynamics can be probed with soft X-rays or vice versa.

The Maloja endstation will allow the investigation of a variety of targets ranging from gas-phase atoms, molecules and clusters to aerosols and liquids, which requires a large degree of flexibility. A set of vacuum chambers compatible with the foreseen sample environments will be available and can be combined with one or multiple detection techniques: electron- and ion-spectroscopy, absorption and emission spectroscopy and X-ray scattering.

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Status and Perspective of X-ray Free Electron Lasers in China

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ABSTRACT

Two X-ray free electron laser (FEL) facilities are currently under construction in China. The soft X-ray free electron laser facility (SXFEL) is a 1.5GeV C-band linac based seeded FEL with the output photon energy up to 0.4 keV. This facility was funded with two independent projects, the test facility (SXFEL-TF), consisting of an 840MeV C-band linac and a HGHG/EEHG undulator line, is in the FEL commissioning, and the user facility (SXFEL-UF), by upgrading the linac energy to 1.5 GeV and adding an additional SASE undulator line and five experimental instruments, is under construction, aiming to operate for users in 2020. In the meantime, an 8GeV CW superconducting linear accelerator based hard X-ray FEL facility (SHINE), with maximum FEL pulse repetition rate up to 1 MHz and radiation photon energy covering 0.4 – 25 keV range, is in construction also at Zhangjiang High-tech Park of Shanghai now. This 3 km long facility is to be built in a tunnel located 30 m underground, and it includes 3 undulator lines and 10 Phase-I experimental end-stations. This talk presents an overview of the SXFEL and SHINE projects.

The TMO Instrument: Opportunities and Plans for Time-resolved Atomic, Molecular and Optical Science at LCLS-II

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Synopsis The new designed Time-resolved Atomic, Molecular and Optical Science end station, will be configured to take full advantage of both the high per pulse energy from the copper accelerator (120 Hz) as well as high average intensity and high repetition rate (1 MHz) from the superconducting accelerator. TMO will support many experimental techniques not currently available at LCLS and will have two X-ray beam focus spots. Thereby, TMO will support AMO science, strong-field and nonlinear science and a new dynamic reaction microscope.

The unique capabilities of LCLS, the worlds first hard X-ray FEL, have had significant impact on advancing our understanding across a broad range of scientific fields, from fundamental atomic and molecular physics, to condensed matter, to catalysis and structural biology. A major upgrade of the LCLS facility, the LCLS-II project, is now underway. LCLS-II is being developed as a high-repetition rate X-ray laser with two simultaneously operating, independently tunable FELs. It features a 4 GeV continuous wave superconducting linac that is capable of producing uniformly spaced (or programmable) ultrafast X-ray laser pulses at a repetition rate up to 1 MHz spanning the energy range from 0.25 to 5 keV. Furthermore, the XLEAP sub-femtosecond soft X-ray pulse generation program is scalable to LCLS-II repetition rate[1].

The Time-resolved atomic, Molecular and Optical Science (TMO) instrument, one of the three new LCLS-II instruments with a energy range from 0.25 to 2 keV, will support AMO science, strong-field and nonlinear science, and a new dynamic reaction microscope. TMO will support many experimental techniques not currently available at LCLS and will have two X-ray focus spots.

At the first focus spot of the TMO instrument we will offer the possibility to install standardized modular stations (roll in and out) which can be set up, aligned and commissioned outside the hutch and then get installed for operations. These highly standardized stations will be optimized for performing high energy, high resolution, time- but also angular-resolved photoelectron spectroscopic measurements by using high resolution iTOF and eTOF, double-sided-VMI [2] or coaxial-VMI [3].

At the second focus spot a new reaction microscope endstation will house a COLTRIMS type spectrometer to accommodate extreme vacuum, sub-micron focus spot size, and target purity requirements as dictated by coincidence experiments. The accumulation of data will be performed on an event-by-event basis using the 1 MHz full repetition rate of LCLS-II.

We would like to present some of the important science opportunities, new capabilities and instrumentation being planned for NEH 1.1 (TMO) at LCSL-II.

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Echo Enabled Harmonic Generation at the FERMI Free-Electron Laser

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ABSTRACT

FERMI is a seeded Free-Electron Laser (FEL) user facility at the Elettra laboratory in Trieste [1], operating in the VUV - soft X-ray spectral range. It produces ultra-short and ultra-bright pulses characterised by a high shot-to-shot wavelength stability, high degree of longitudinal and transverse coherence and low temporal jitter with respect to the secondary laser sources used in pump-probe experiments. The machine design is based on the High-Gain Harmonic Generation (HGHG) scheme, which makes it possible to frequency up-convert an external optical laser interacting with a relativistic electron bunch. There are two FEL lines currently in operation at FERMI: FEL-1 is based on a single HGHG stage, producing VUV-XUV pulses in the 20 - 100 nm wavelength range, while FEL-2 is a two-stage HGHG cascade, operating in the soft-X-ray range, up to the carbon 1s edge.

During 2018 we performed a dedicated experiment, demonstrating the potential of the Echo-Enabled Harmonic Generation (EEHG) scheme to cover most of the spectral range of FERMI in a single stage. For this purpose, the FERMI FEL-2 line was temporarily modified. We report the first evidence of substantial gain in an EEHG soft-X ray Free Electron Laser. The experiment focused on harmonics 36 (~7.3nm) and 45 (5.8 nm) of an external UV laser and clearly demonstrated the expected EEHG capability of generating powerful and coherent FEL pulses, with strongly reduced sensitivity to electron-beam and external lasers imperfections when compared to HGHG [2].

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Time-resolved Photoionization and Fragmentation Dynamics of Fullerenes Studied with FELs

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ABSTRACT

Photo-induced responses of fullerenes subject to femtosecond free electron laser (FEL) provide critical understanding of the fundamental mechanisms [1,2] that drive the conversion of photons into chemical and kinetic energy on ultrafast timescales. We have carried out time-resolved measurements and calculations to investigate the behavior of fullerenes ionization using short x-ray photons. The experiment used the LCLS fresh slice x-ray pump and x-ray probe technique [3]. The time-resolved experiment is interpreted with Molecular Dynamics Modeling [4,5] to expose, from a fundamental point of view, the physical and chemical processes and their time evolution. The work provides information on radiation damage relevant to bio-molecules.

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Ultrafast Buildup of Magneto-electric Excitations by Polarization-structured Laser Pulses

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ABSTACT

Experiments demonstrated the feasibility of laser pulses that have spatially structured phases, as well as a spatially structured polarization distribution, both in the near and the far field [1,2]. For instance, optical vortices having a structured phase are nowadays routinely generated and exploited for a range of applications. While the light-matter interaction with optical vortices have been well studied, the distinctive features of the interaction of quantum matter with vector beams, meaning fields with spatially inhomogeneous polarization states, has been addressed only recently [3,4,5]. In the near field or in waveguides such photonic beams constitute skyrmionic structures with well-defined topological properties [2,6]. It is our aim to investigate the role of topology of short pulses of such beams while interacting with quantum matter, in particular quantum matter with a non-trivial topology. For instance, by contrasting the vector-beam excitations of C_{60} with C_{120}; note, latter has a toroidal structure and thus a non-simply-connected topology. This contribution will be focused on our results for the response of atomic and low-dimensional quantum structures to the irradiation with radially or azimuthally polarized cylindrical vector beams. Important classes of excitations can be identified: Radially polarized vector beams drive radially breathing charge-density oscillations via electric-type quantum transitions. Azimuthally polarized vector beams do not affect the charge at all but trigger, via a magnetic vector potential, a dynamic Aharonov-Bohm effect, meaning a vector-potential-driven oscillating magnetic moment. In contrast to vortex beams, no unidirectional currents are generated. Atoms driven by a radially polarized vector beam exhibit angular-momentum-conserving quadrupole transitions tunable by a static magnetic field, while when excited with an azimuthally polarized beam, different final-state magnetic sublevels can be accessed. A combination of vector beam with a spatially homogeneous laser pulses render possible the generation of toroidal moments, i.e. charge and current distributions that are coupled in such a way that the far-zone electric and magnetic field components vanish while the vector potential remains finite and gauge invariant. This is exciting in so far, as such a vector potential can be utilized to tweak the phase of quantum systems without exerting any forces. Also the penetration depth is markedly different from conventional electromagnetic fields. Examples and possible applications will be presented at the conference.

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Complete Reconstruction of Bound and Unbound Electronic Wavefunctions in Two-photon Double Ionization

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ABSTRACT

Using intense, linearly and circularly polarized XUV pulses from FEL FERMI [1], we realized the first Complete Experiment [2] in an ionic system. Photo-electron Angular Distribution (PAD) and cross sections were acquired for photon energies between 40 and 60eV, and with a fine scan in the 56.2-56.8eV range, corresponding to the energy of the auto-ionizing states of Ne II (fig.1a). In a 2-photon process, PADs can be described on the base of the Legendre polynomials by the relation: $I(\theta) = I_0/4\pi[1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)]$. The validity of the theoretical model is verified by the agreement with the β_N parameters (Fig. 1b), and the ratio of ionization of the different peaks. In the Cooper-Zare model, [3], the dynamics of each PI step is described by two absolute values of the single electron (*2p-εs* and *2p-εd*) ionization amplitudes and their relative phase. These quantum-mechanical parameters can be linked to the experimental data, thus determining a restricted allowed space of the PI amplitudes (fig. 1C). The intersection of the allowed spaces defines the solution.

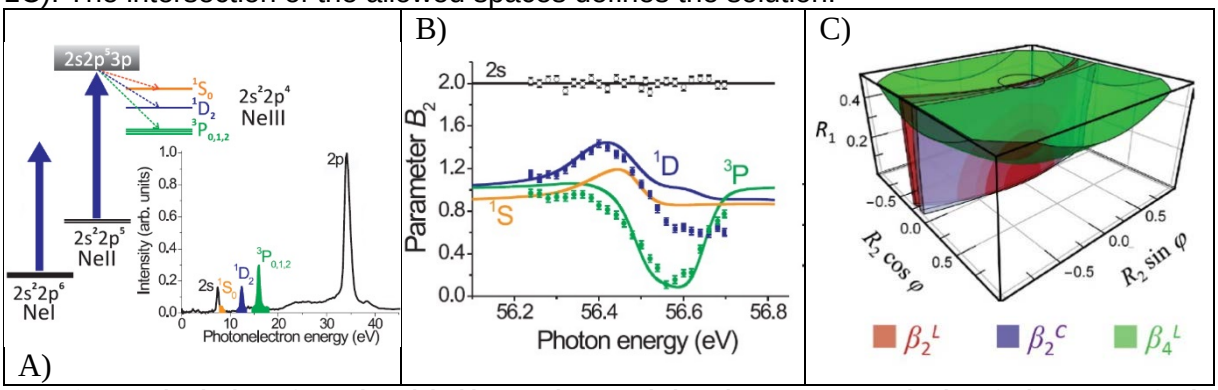

Fig. 1 a) Energy level scheme for singly and doubly ionized neon and photoelectron spectrum. The three final ionic states for the doubly ionized neon $^3P_{0,1,2}$, $^1D_2$, $^1S_0$ and the corresponding photoelectron peaks are indicated by green, blue and orange, respectively. B) Experimental (squares) and calculated (lines) β_2 parameter around the resonance of the auto-ionizing states for linearly polarized FEL. Colors as in previous figure. C) Complete experiment on PI of Ne^+ $2p^5$ ^2P to the Ne^{++} $2p^4$ ^3P state at the photon energy 53eV. Allowed sets of parameters R_1, $R_2\cos\varphi$, $R_2\sin\varphi$ extracted from the values of $\beta_2(C)$ (blue), $\beta_2(L)$ (red), $\beta_4(L)$ (green) for the ^3P peak.

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Nonsequential Two-photon Ionization of Inner-shell Electrons in Neutral Atoms

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ABSTRACT

Nonsequential two-photon ionization is a fundamental nonlinear process which became an important benchmark for studying the interaction of intense light with matter. The study of nonlinear high-order processes has been driven by the recent development of free-electron lasers [1], intense-light sources operating in the ultraviolet and x-ray energy domains. With today's high photon energies and beam intensities of free electron lasers, even two-photon ionization of the deep *K*-shell electron of medium and heavy elements has become possible. In recent years, free-electron lasers have already been used to detect the nonsequential two-photon *K*-shell ionization of neutral Ge, Cu, and Zr atoms [2-4].

In this contribution, we present our studies of the two-photon direct ionization of the *K*-shell electrons in neutral atoms. In particular, the importance of relativistic and screening effects in the total two-photon ionization cross section [5] as well as in the photoelectron angular distribution [6] is thoroughly investigated. We show that the relativistic effects significantly decrease the total cross section, while the screening effects of the inactive electrons lead to the occurrence of an unexpected minimum in the total cross section, where also the photoelectron distribution undergoes considerable changes.

Furthermore, we predict the existence of a so-called *nonlinear Cooper minimum*, i.e. photon energy at which the dominant ionization channel vanishes. It appears that the Cooper minimum is always present when an intermediate virtual electron state is sandwiched between two neighboring resonances with the same angular momentum. The interest in Cooper minima is sparked by its effect on the observables. The suppression of the dominant channel is imprinted not only in the total or angle-differential cross sections but also in the polarization of the subsequent fluorescence in forms of the clear peaks (or troughs). Moreover, near a nonlinear Cooper minimum, there are two positions of maximum elliptical dichroism in the photoelectron angular distributions [7]. This remarkable phenomenon of the maximum elliptical dichroism could be in particular used as a sensitive tool for analyzing the polarization state of photon beams produced by free-electron lasers as well as a precise tool for understanding nonsequential many-photon ionization.

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POSTERS SESSION

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A Theoretical Study of Time Delays in Above-threshold ω-2ω Ionization

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ABSTRACT

An experiment in atomic ionization by two-color lasers by Zipp *et al.*[1] has revealed that a pump-probe scheme can be used to characterize time delays in the emission of electrons in the above-threshold ionization regime for visible frequency of the pump and its first harmonic as a probe. To shed more light about the ionization processes of the mentioned experiment, in this work we perform a theoretical analysis of the time delays in Ar ionization by a two-color laser [Ti:Sapphire laser (800 nm) together with the first harmonic (400 nm)] for a typical ω−2ω configuration [2]. We perform simulations with the time dependent Schödinger equation[3] and compare these results with the strong-field and Coulomb-Volkov approximations. We find that the strong assumption of additive time delays adopted in streaking or RABBITT techniques needs to be revisited when the atomic system is subject to ω−2ω fields [4]. We show that time delays depend on the definition used to extract them from electron momentum distributions by considering the asymmetry in a particular direction of the electron emission or by integrating in hemispheres, and also considering a single energy at the multiphoton peaks or integrating over energy around the multiphoton peaks. Besides, we also find a large discrepancy between the results predicted by the strong-field approximation and the numerical solutions of the time dependent Schrödinger equation even at the highest simulated energies. With the help of the Coulomb-Volkov approximation, we see that the contribution of the long-range potential effects is almost negligible at intermediate to high emission energies with respect to the strong field approximation. We have also compared with *ab initio* simulations using short-range Yukawa potentials.

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Intracycle Interference in Angle-resolved Spectrum in Laser-assisted XUV Ionization

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ABSTRACT

In laser-assisted XUV photoemission (LAPE), the XUV and optical laser overlap in space and time. When the XUV pulse is longer than the laser period, a photoelectron is emitted into the optical dressing field where one or more quanta of energy can be absorbed and emitted. As a result, the photoelectron spectrum shows sidebands (SBs) separated in energy whose intensity depends on the XUV photon energy and dressing field strength[1].

The purpose of this work is to theoretically study the ionization of the hydrogen atom due to an XUV pulse in the presence of an IR laser with both fields linearly polarized in the same direction by means of a very simple semiclassical model. We show how the energy distribution of photoelectrons can be explained through the interference among electron trajectories. In this work, we analyze the angle-resolved photoelectron emission in LAPE as a function of the laser intensity for different XUV pulse durations. Within the semiclassical model[2] the electron energy spectrum can be explained in terms of different kind of interferences depending of which electron trajectories are involved: (i) The intercycle interference, which stems from trajectories released at different optical cycles, and (ii) the intracycle interference, which stems from trajectories within the same optical cycle[3]. The inter- and intracycle interferences are responsible for the formation and modulation of SBs, respectively. The intercycle interference gives rise to characteristic SBs separated by the laser photon energy in the PE spectrum for all emission angles. However, for perpendicular emission, electron trajectories born at the first and second halfcycles (within the same optical cycle) interfere destructively, resulting in SBs separated by twice the laser photon energy[4].

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Propagation of Extreme Ultraviolet Free-Electron-Laser Pulses through a Dense Sample of Helium Ions: Superfluorescence, Free-induction Decay, and Four-wave Mixing

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ABSTRACT

We have studied experimentally and numerically the propagation of intense free-electron laser pulses at a central wavelength of 24.3 nm (photon energy 51.0 eV) through a dense ($2x10^{23}$ atoms m^{-3}) sample of helium gas [1]. Effectively all atoms along the laser path are ionised, and the wavelength is such that the 1s and 4p states of He^+ are resonantly coupled. The experiments and numerical simulations suggest that the following phenomena occur:

- **superfluoresence:** Intense, highly directional emission was observed experimentally at wavelengths of 469 nm (4p-3s), 164 nm (3s-2p) and 30.4 nm (2p-1s). The emission at 469 nm showed pulse widths and delays characteristic of superfluorescence. Comparison with simulations suggest that the 3s-2p and 2p-1s decays are *yoked* superfluorescence, which occurs due to transfer of the coherence imparted by the initial excitation
- **free-induction decay:** Intense emission was also observed at the resonant wavelength of 24.3 nm (4s-1p), and at 25.4 nm (3s-1p). Comparison with simulations suggests that this is free-induction decay, although the time dependence was not observed experimentally.
- **four-wave mixing:** The numerical simulations also suggest that four-wave mixing should occur between the various fields present.

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Photoionization of H_2 using the Molecular R-Matrix with Time Approach

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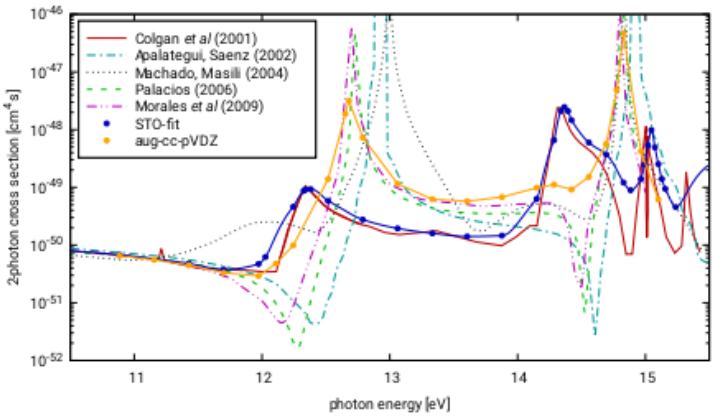
ABSTRACT

In our contribution we study one-, two- and four-photon photoionization of the hydrogen molecule by monochromatic pulses using the recent extension of the originally atomic-only R-matrix with time dependence (RMT) method [1] to molecules [2]. The molecular RMT combines the accuracy of the time propagation provided by RMT and the ab initio target desciption by the stationary molecular R-matrix package UKRmol+ [3], enabling correct description of multi-electron effects and electron correlation in the response of the target to the external field.

This work validates the UKRmol+/RMT combination on several processes well studied in the past. The generalized multi-photon cross sections for photoionization of H_2 by a weak, linearly polarized field obtained using the UKRmol+/RMT suites are compared to results from the Floquet R-matrix method [4], to those of a direct solution of the Schrödinger equation in DVR basis with ECS boundary conditions [5] and other approaches.

The RMT approach is very general and allows the study of a range of processes from the perturbative to highly non-linear regimes. Detailed understanding of the molecular RMT models for multi-photon ionization is a prerequisite for its application to processes such as strong-field ionization and high-harmonic generation.



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Ionization-induced Dissociation of Methane, Irradiated by 800 nm and 400 nm Laser Fields

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ABSTRACT

Irradiation of methane by intense laser fields leads to the production of several molecular ions CH_n^{m+} ($m = 1, 2$ with $n = 0 - 4$) and atomic/molecular hydrogen fragments such as H_n^{m+} ($m = 0, 1$ with $n = 1, 2$) [1]. For nanosecond pulses [2] the CH_4^+ ion was absent as a result of its dissociation into CH_3^+ + H. On the other hand, for 800 nm/8 fs pulses [3] all but the CH_4^+ and CH_3^+ ions disappeared, indicating that methane did not have enough time to dissociate. In the present work, carried out at the SQS laboratory, we include irradiation at 400 nm.

A typical TOF spectrum is presented in figure 1A) where the unstable methane trication is absent. On the other hand, the H_2^+ and H^+ ions, arising from coulomb explosion pathways, are present. For both laser wavelengths, the same dissociation trend, attributed to a step-wise dissociation mechanism, is observed. According to the field assisted dissociation or FAD model [4] the CH_3^+ ions have the highest dissociation threshold, acting as a bottleneck for the production of CH_2^+, CH^+, and C^+ ions. This pattern is confirmed by our experimental findings to be independent of the laser wavelength. However, a striking difference is observed in the case of H^+ ions where dissociative ionization induced by the 400 nm field results in ions with significantly lower kinetic energy especially at higher laser intensities [cf. figure 1B)].

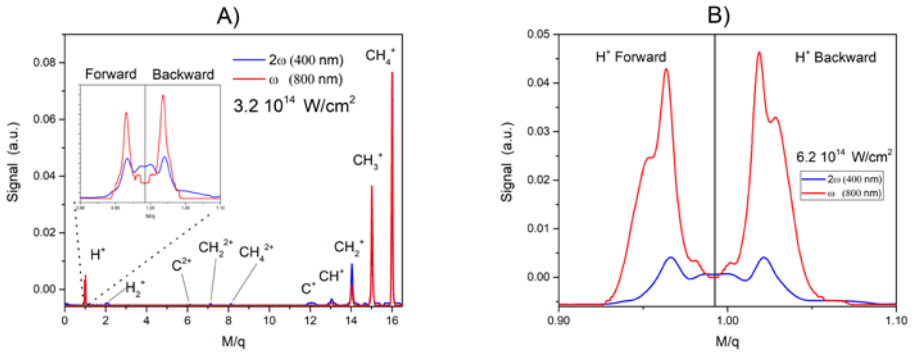

Fig. 1 A) A representative TOF spectrum for ionization induced dissociation of methane irradiated by 800 and 400 nm light at an intensity of approximately 3.2×10^{14} W/cm^2. **B)** Peaks of H^+ fragments corresponding to comparable intensities of the fundamental (800 nm) and the second harmonic (400 nm) at an intensity of 6.2×10^{14} W/cm^2.

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Laser Assisted Photoionization of Argon Atoms

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ABSTRACT

The laser-assisted XUV photoelectric effect (LAPE) occurs when extreme ultraviolet (XUV) and infrared (IR) lasers overlap in space and time. In this case, the photoelectron spectrum is factored into two contributions: one that accounts for the intracycle (during one IR cycle time interval) electron emission and the second one describing the intercycle interference (due to the emission at different IR cycles) [1,2,3]. This intercycle interference is the responsible of the well-know "sidebands" (Sbs) in the photoelectron spectrum separated at energy values given by

$$E_n = n\omega_L + \omega_X - I_p - U_p$$

where $\omega_{X(L)}$ is the frecuency of XUV(IR) pulse, I_p is the ionization energy, U_p the ponderomotive energy and n is the number of absorbed or emitted IR photons.

In the present work we consider the Argon photoionization from the subshells 3s, 3p0 and 3p1 due to a short XUV pulse assisted by an IR laser both linearly polarized. Within the continuum-distorted wave Strong Field Approximation (SFA) supported by the ab initio solution of the time-dependent Schrödinger equation (TDSE) we study the angle-resolved momentum distribution of photoelectron for the case that both fields are linearly polarized. We analyze and characterize the modulation of both factors, the intra- and intercycle contributions, on the spectrum.

In particular, we study several geometrical arrangements between the polarization vectors and the emission direction, that give rise to selective photoelectron sideband energies in the the ionization spectrum, as in the case of perpendicular emission where only odd sidebands are observed.

During the conference we will theoretically analyze the selection rules that governs the allowed photoelectron energies, showing that the intra-half-cycle factor [1,3] destructively interfere to cancel some sidebands peaks.

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Time-resolved Orbital Imaging by Photoelectrons

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ABSTRACT

We demonstrate the imaging of time-dependent valence electron densities following ionization. We employ a sequence of three laser pulses to prepare a wave packet in the spin-orbit split ground state of Ar^+ and record the resulting variations of the charge density using strong-field ionization on a femtosecond time scale. This approach will allow for imaging electron dynamics within molecules following ionization, such as charge migration and charge transfer.

Attosecond technology has provided a wealth of imaging approaches that provide snapshots of the electronic or nuclear structure of molecules with (sub-) femtosecond time resolution. Orbital imaging by photoelectrons has remained restricted to static electron densities, mostly due to contamination of the time-dependent signal with photoelectrons produced in the probe pulse [1].

Here, we present a pump-probe scheme that overcomes previous limitations of laser-based orbital imaging [2]. Our approach avoids contamination of the probe pulse signal by using an additional mid-IR streaking field [3] to tag the electrons emitted in the probe pulse. Their delay-dependent three-dimensional momentum distribution is recorded using Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS), probing the transient valence electron density in the ion. Orbital images are obtained by calculating normalized differences between the delay-dependent and delay-averaged photoelectron momentum spectra [3].

A spin-orbit wave packet in the argon cation is tracked and imaged using strong-field ionization probability of Ar^+, and consequently the recorded yield of Ar^{2+} ions. The maximum yield is obtained for electronic anti-revivals when the $m = 0$ orbital is occupied by two electrons. At the revivals, when only one electron occupies the $m = 0$ orbital, double ionization is suppressed. In the momentum space plane perpendicular to the laser polarization, a narrow distribution around the origin is obtained as a signature of the $m = 0$ orbital. As the wave packet evolves, the donut-shaped $|m| = 1$ orbital is revealed.

Moreover, we show that the longitudinal momentum distribution exhibits a delay dependence which carries information on the sub-cycle ionization dynamics of the coherent electron wave packet. We will discuss the reconstruction of the spatial electron density and future applications to electron dynamics in molecules.

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X-ray-induced Electronic and Structural Dynamics in CH_2BrI Molecules

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ABSTRACT

X-ray free-electron lasers (XFELs) opened the way to the investigation of unexplored dynamics and optical phenomena in the X-ray spectral region. At the XFEL facility SACLA in Japan, we have investigated the interaction of XFEL pulses with isolated molecules containing heavy iodine atoms, iodomethane (CH_3I) [1], diiodomethane (CH_2I_2) [2], and 5-iodouracil ($C_4H_3IN_2O_2$) [3]. Furthermore, we have identified different electronic decay pathways by X-ray pump–near infrared (NIR) probe measurements [4].

In this study, we performed multiple coincidence ion momentum spectroscopy of bromoiodomethane (CH_2BrI) molecules irradiated by a 5.5 keV intense XFEL pulse generated by SACLA. After XFEL irradiation, the molecules are multiply ionized via cycles of inner-shell photoionization of the iodine atom and subsequent Auger cascades, and further explode violently due to Coulomb repulsive forces. Since those processes occur competitively, the momenta of the released ions and their correlations carry information about the dynamical behavior of molecules.

We also performed time-resolved ion yield measurements for CH_2BrI molecules using the XFEL-pump–NIR laser-probe technique. The resulting partial ion yields as a function of the NIR laser pulse delay for each fragment show different temporal structures. On this basis, we have extracted information on the dynamics associated with the electronic decay processes. Thus, we characterized the interaction between electron and molecular dynamics induced by the XFEL radiation.

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Ionization of Kr II in the Region of Autoionizing Rydberg States

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ABSTRACT

Angle-resolved electron spectrometry in resonant photoionization of positively charged ions attracts more and more attention after the advent of FELs generating high intensity photon beams in the XUV wavelength range. The high density of singly charged ions in this case enables for example the investigation of sequential two-photon double ionization [1,2]. The photoelectron spectra and angular distributions (PADs) in the region of Rydberg autoionizing states of argon ions were recently studied both experimentally at FLASH [3] and theoretically [3,4] providing interesting results. In argon, the relativistic effects are not as strong as in heavier noble gases and it was possible to perform calculations within the non-relativistic R-matrix approach with corrections to experimental values of the fine-structure energy splitting.

Here we perform calculations of a similar process in krypton using the semi-relativistic R-matrix approach. Photoionization of Kr II is calculated in the region of Rydberg series of the autoionizing states Kr^+ ($4p^4\ ^3P_2$ ns, nd) converging to the ionization thresholds Kr^{++} ($4p^4\ ^3P_{1,0}$) as well as of other Rydberg series converging to the thresholds Kr^{++} ($4p^4\ ^1D$) and ($4p^4\ ^1S$). In contrast to [5], we concentrate in addition to the photoabsorption also on the photoelectron spectra and PADs. The low-energy resonance region below the Kr^{++} ($4p^4\ ^1D$) threshold is analyzed in detail, providing information on dominating ionization channels and PADs with accounting for alignment of the intermediate $Kr^+(4p^5\ ^2P_{3/2})$ ionic state.

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Nonlinear Cooper Minimum as a Precise Tool for Understanding Multi-photon Ionization

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ABSTRACT

The advances and increasing number of free electron lasers (FELs) revolutionized the exploration of inner-shell electron dynamics and nonlinear light-matter interaction by delivering intense extreme-ultraviolet and x-ray beams. However, the accuracy of the extracted cross sections is bound by the systematic uncertainty in the beam parameters which determine the absolute intensity. This causes large experimental uncertainties which in return do not allow to resolve differences between various theoretical approaches[1].

We suggest alternative measurable quantities which promise more accurate comparison of experiment with theory. It is generally expected that the properties of multiphoton ionization are dominantly determined by ionization channel with the highest angular momentum. Here, we show that this rule is broken not only at photon energies matching intermediate electronic level resonances, but also at a nonlinear equivalent of Cooper minimum, where the dominant ionization channel drops to zero. We further suggest that measuring observables such as photon-ion or fluorescence polarization, photoelectron or Auger electron distributions will accurately determine the position of these *nonlinear Cooper minima* and will allow for comparison of experimental measurements with theory at hitherto unreachable precision. Measurements of fluorescence or angular distributions of escaping electrons as a characteristic of multiphoton ionization are well established, and hence, measurements of these properties at the nonlinear Cooper minimum could readily find applications beyond fundamental importance, as it could also serve for applied fields such as spectroscopy of atoms and molecules[2].

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Attosecond Delays in Photoionization Studied with Seeded FEL at FERMI

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ABSTRACT

The Invention of probing methods on a time scale less than femtoseconds has led to the age of attosecond physics [1], and numerous such ultrafast phenomena currently are being investigated. Photoemission is one of those processes that has received a lot of interest, especially concerning the photoemission delay between photon absorption and photoelectron emission known as the Eisenbud-Wigner-Smith (EWS) delay [2].

We are presenting here a new method for measuring the EWS delay. In this method, we use short wavelength EUV light, consisting of phase-locked fundamental (ω) and second harmonic (2ω) pulses. It requires extremely accurate phase control (few attoseconds). Such fine control is available from the Italian free-electron laser (FEL), FERMI [3]. We report as a demonstration of the new method the EWS delay difference between one-and two-photon ionization of atomic Ne.

The target processes are the photoionizations of Ne by one or two photon(s):

$$Ne + 2\omega \rightarrow Ne^+ + e^-, \qquad Ne + \omega + \omega \rightarrow Ne^+ + e^- \text{ (non-resonant)}.$$

Because these electrons emitted by the two different pathways interfere with each other, the electron angular distribution is correlated to the difference of the unique phase shifts. The phase shift difference can be extracted by scanning the optical phase difference between ω and 2ω FEL pulses. The EWS delay τ is defined as the derivative of phase shift η with respect to the photoelectron kinetic energy E: $\tau = \hbar \cdot \partial\eta/\partial E$ [2]. We have measured the phase shift differences at several photon energies and found their slope to estimate the delay difference.

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