38. EAS -Tagung "Extreme Atomic Systems"

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– list of abstracts –

Lennart Aufleger, MPI für Kernphysik Heidelberg

Two-electron dynamics in Helium driven by intense XUV radiation

The measurement of laser-induced strong-field dynamics in few-electron systems provides a way to investigate their correlated nature. Using a lab-based HHG source we have studied such correlations that are imprinted on the XUV absorption line shape and investigated them by modification with strong fields in the near infrared and visible (VIS) spectral region. [C. Ott et al., Science 340, 716 (2013)]. Here, we present an extension of the strong-field modification scheme to the XUV-only spectral region. Using intense and partially coherent light of a FEL source (FLASH@DESY), the transition into the doubly- excited state 2s2p in Helium at 60.15 eV was driven. This excitation scenario represents a most basic two level system with a cooperative excitation of two electrons. The variation of pulse intensities between 1012 W/cm^2 and 1014 W/cm^2 induces a change in the experimentally observed XUV absorption line shape. A transformation from a Fano to a Lorentzian line shape is also confirmed by few-level simulations that were carried out.

Panagiotis Giannakeas, Purdue University

Few-body physics for homo and heteronuclear systems

The Recent developments on the three-body physics of homo- and heteronuclear ultracold atoms will be discussed. More specifically, in the case of the homonuclear three-body collisions the regime of atom-dimer formation will be presented emphasising on the corresponding universal aspects. In particular, we show that beyond the zero-range models the atom-dimer resonant features are mainly associated with the excited Efimov states whereas the ground state does not approach the atom-dimer threshold.
In the case of heteronuclear three-body systems a detailed study of the three-body recombination processes will be discussed focusing on the recent experimental realizations by Ulmanis *et al* (Phys. Rev. Lett. 117, 153201 (2016)).
For the system of Li-Cs-Cs where the intraspecies possess positive and finite scattering length we observe that in the branch of negative interspecies scattering lengths the corresponding enhancements in the recombination rates are solely associated with excited Efimov states and exhibit a rather universal character with respect to the short range physics.

Nikolay Golubev, Universität Heidelberg

Coupled electron-nuclear dynamics after outer-valence ionization of propiolic acid

Since the early days of theoretical quantum mechanics, the aim of scientific efforts has been to treat coupled movement of electrons and nuclei. However, due to the fact that until very recently it was experimentally unattainable to observe ultrafast electronic motion, an emphasis was given to examine a non-adiabatic dynamics of nuclear wave packets.
In the last years, the rapidly developing attosecond-pulse techniques have given us the unique tool for direct studying and eventually controlling electronic motion. Consequently, theoretical methods where the dynamics of electrons and nuclei is treated explicitly are required. We propose a methodology allowing to trace coupled electron-nuclear dynamics in real time and space. We demonstrate on the molecule of propiolic acid how one can com-

pute the ultrafast charge migration triggered by removing an electron from an outer-valence molecular orbital, taking into account the coupling to the nuclear motion, and analyze it. We would like to emphasize that the presented scheme offers very promising perspectives for studying of a chemical reaction. In particular, it allows to describe a charge-directed reactivity phenomenon where one can predetermine the outcome of the chemical reaction by localizing the charge on desired side of the molecule.

Jiri Hofbrucker, Helmholtz Institute Jena

Relativistic calculations of the non-resonant two-photon K-shell ionization of neutral atoms

The non-resonant two-photon one-electron K-shell ionization of neutral atoms is studied within the framework of relativistic second-order perturbation theory and independent particle approximation. The importance of relativistic and screening effects in the total as well as differential cross sections is investigated. Our results show that, at near two-photon ionization threshold energies, the account for the screening effects of the remaining electrons leads to occurrence of an unexpected minimum in the total two-photon ionization cross section [1] and to elliptical dichroism in the photoelectron angular distribution. For ionization of heavy atoms, relativistic effects result in a significant decrease of the total cross section [2], and in distortion of the angular distribution into forward direction.

[1]J. Hofbrucker, A. V. Volotka, S. Fritzsche, Phys. Rev. A **94**, 063412 (2016).
[2]J. Hofbrucker, A. V. Volotka, S. Fritzsche, submitted to Nucl. Instr. Meth. Phys. Res.

Vladimir Yerokhin, St. Petersburg Polytechnical University

The g factor of the bound electron in H-like and Li-like ions for determination of fundamental constants and tests of QED

Measurements of the bound-electron g factor in light hydrogen-like ions, combined with sophisticated ab initio QED calculations provided one of the most stringent tests of QED and currently the best determination of the electron mass. In future, such experiments and calculations may give us access to an improved determination of the fine-structure constant and lay the foundation for precision tests of the Standard Model. In my talk I will discuss the present status and recent progress of theory of the bound-electron g factor of H-like and Li-like ions and perspectives for determination of the fine-structure constant from measurements of g factors of light few-electron ions [1].

[1] V. A. Yerokhin, E. Berseneva, Z. Harman, I. I. Tupitsyn, and C. H. Keitel, g Factor of Light Ions for an Improved Determination of the Fine-Structure Constant, Phys. Rev. Lett. **116**, 100801 (2016)

Nikolay Kabachnik, DESY Hamburg

Circular dichroism in multiphoton ionization of resonantly excited He^+ Ions.

Theoretical interpretation of the experiment, in which intense, circularly polarized extreme-ultraviolet (XUV) and near-infrared (NIR) laser pulses are combined to double-ionize atomic helium via the oriented intermediate $He^+(3p)$resonance state, is considered. In the experiment, applying angle-resolved electron spectroscopy, a large photon helicity dependence of the spectrum and the angular distribution of the electrons ejected from the resonance by NIR multiphoton absorption, were revealed. The measured circular dichroism is unexpectedly found to vary strongly as a function of the NIR intensity. The experimental data are well described by theoretical modeling. Possible reasons for strong intensity dependence is discussed.

Gregor Kastirke, Universität Frankfurt
The new COLTRIMS-Reaction microscope designed for European XFEL measures ultrafast dissociation of CH_3Cl

A newly designed COLTRIMS-setup was especially built for experiments at the European XFEL at the Small Quantum Systems instrument of the SASE3 Beamline. In order to investigate Ultrafast Dissociation in CH_3Cl we performed an experiment at the SOLEIL Synchrotron, France using this new setup. By taking coincidence measurements of ions and electrons, we exploit the energy of an Auger electron while the dissociation process is taking place. With this information, snapshots of the temporal evolution of a decaying orbital of a molecule fragmenting into separated atoms are obtained.

Victor Kimberg, KTH Royal Institute of Technology, Stockholm
X-ray Raman scattering in strong fields

X-ray Raman scattering is a well-established powerful tool for study of the electronic structure and nuclear dynamics in molecules, condensed phase, and surfaces. Traditional x-ray scattering spectroscopies are based on both radiative and nonradiative emission, such as non-resonant x-ray emission, resonant inelastic x-ray scattering (RIXS), Auger and resonant Auger spectroscopy. Recent invention of x-ray free-electron lasers providing ultrashort x-ray pulses with enormous photon flux allows to bring a qualitatively new aspect to x-ray spectroscopic techniques x-ray nonlinearity. I would like to overview our resent theoretical results on spectroscopy of x-ray induced processes in molecules in strong x-ray fields. I will start discussing stimulated emission and lasing the first x-ray non-linear effect already observed in atoms and predicted in molecules [1]. I will continue with more sophisticated all x-ray pump-probe schemes including stimulated RIXS spectroscopy with coherent and stochastic light sources [2]. Finally, I will briefly present recent experimental attempts on stimulated RIXS in molecules [3] and address experimental feasibility for high-resolution nonlinear x-ray spectroscopy.

Maciej Kosicki, Nicolaus Copernicus University Toruń
The isotopic substitution in reactions of ultracold alkali dimers: A statistical approach

Ultracold molecules have been extensively studied in recent years because of wide range of potential applications in quantum information theory, simulations of quantum gases or high-precision spectroscopy. Using statistical assumptions we have obtained theoretical predictions for possibilities of cooling polar molecules by their collisions with atoms. For this purposes, we have studied low-energy reactive collisions corresponded to reactions of the isotopic substitution in ultracold NaK molecules. Our research include statistical distributions of products - so far, only states of reactants were measured in experiments.

Alexander Kuleff, Universität Heidelberg
Sub-femtosecond charge migration in the valence shell initiated by core ionization

After ionization of a valence electron, the created hole can migrate ultrafast from one end of the molecule to another. Due to the advent of attosecond pulse techniques, the measuring and understanding of charge migration has become a central topic in attosecond science. Here, we pose the hitherto unconsidered question whether ionizing a core electron will also lead to charge migration. It is found that the created hole in the core stays put, but in response to this hole interesting electron dynamics takes place which can lead to intense charge migration in the valence shell. This migration is typically faster than that after ion-

ization of a valence electron and transpires on a shorter time scale than the natural decay of the core hole by the Auger process, making the subject very challenging to attosecond science.

Hannes Lindenblatt, MPI für Kernphysik
Multiphoton double ionisation of Neon studied at FLASH2

The ionisation of Neon was studied by an XUV pump-probe experiment at the new Reaction-Microscope endstation at FLASH2. The variable gap undulators provide the opportunity to quickly change the central wavelength of the FEL photons. Combined with the Online Photoionisation Spectrometer (OPIS) for single-shot monitoring of the wavelength, this allows sampling of processes sensitive to the photon energy at the high intensity provided by a free-electron laser.
The production of doubly charged Ne^{2+} yield is analysed as a function of the photon intensity and energy around the Ne^{+} $2s^2 2p^5 \rightarrow 2s2p^6$ transition energy of 26.9 eV.

Valentin Link, Technische Universität Dresden
Applying open-sytem methods to strongly correlated Bose-gases

Felix Mackenroth, mpipks Dresden
Using chirped pulses to control high-energy laser-ion acceleration

We present the novel ion acceleration mechanism "Chirped-Standing-Wave Acceleration". This mechanism is based on locking the electrons of a thin plasma layer to the moving nodes of a standing wave formed by reflecting a chirped laser pulse from a mirror behind the thin layer. The resulting longitudinal charge separation field between the displaced electrons and the residual ions then accelerates the latter. Since the plasma layer is stabilized by the standing wave, the formation of plasma instabilities is suppressed. Furthermore, the experimentally tunable laser chirp provides a versatile tool for manipulating the resulting ion beam in terms of maximum particle energy, particle number and spectral distribution. Through this scheme, proton beams, with energy spectra peaked around 100 MeV, were theoretically shown to be feasible for pulse energies at the level of 10 J.

Lukas Medisauskas, mpipks Dresden
Kramers-Henneberger atom at low frequencies

Transformation to a frame of reference that moves with a laser-driven electron is a well-known way to study the dynamics of atoms in strong laser field. In this reference frame, the stationary electron experiences a time-dependent potential of the atom. The cycle-averaged effective potential that the electron experiences even in extremely intense and high frequency fields still supports bound states, and is often referred to as the Kramers-Henneberger atom. Although the Kramers-Henneberger atoms can be defined rigorously only in the limit of high frequency fields, recently there were speculations on its existence at low laser frequencies. In this work, we develop an efficient numerical method to solve the full Floquet problem in the Kramers-Henneberger reference frame that allows treating hundreds of photon orders. Dynamics driven by short low-frequency laser pulses is studied explicitly using two-timescale formulation. Hence, the existence of the Kramers-Henneberger atom in the low frequency laser field is discussed.

Severin Meister, MPI für Kernphysik Heidelberg
Resonance, -enhanced ICD in Neon dimers

Interatomic Coulombic Decay (ICD) is an efficient relaxation mechanism in weakly bound environments. The released ICD electron has a distinct energy and could trigger successive processes. A first step towards the control of ICD, is to intentionally switch it on and off. For this, we employed the 2s–2p transition at 26.9eV (46.1nm) in Neon dimers.
In a two-photon absorption process, the first photon creates a 2p outer-valence vacancy, while the second photon resonantly drives the transition $Ne^+(2p^{-1}) \rightarrow Ne^+(2s^{-1})$ within the same atom. The following relaxation leads to the emission of a 2p ICD electron in the neighboring Neon atom. Finally, the two $Ne^+(2p^{-1})$ ions Coulomb explode.
As one of the first experiments at FLASH2, we used the variable-gap undulators to quickly tune the photon wavelength. By scanning the resonance, we found an enhancement of a factor of 2.5 for coincident $Ne^+(2p^{-1})$+$Ne^+(2p^{-1})$ ions. Momentum-resolved coincidence measurements were performed with a reaction microscope (FLASH-REMI).

Robert Müller, Physikalisch-Technische Bundesanstalt
Electronic bridge processes for the excitation of atomic nuclei

The energy structure of atomic nuclei has been studied already for decades. Out of the studied nuclei especially the thorium isotope ^{229}Th turned out to be particularly interesting. The reason for this is that this isotope has a first excited state only a few eV above the ground state, which is orders of magnitudes smaller than the excitation energies found up to now in other nuclei. There is a lot of effort spent to develop a so called nuclear clock based on this transition between the ground state of the ^{229}Th nucleus and its first excited state. Moreover even the idea of a nuclear laser has been proposed. For all these investigations it is necessary to excite the nucleus in a controlled way. A direct photoexcitation seems possible but turns out to be very difficult due to the extremely narrow linewidth of nuclear states. A much more promising scenario for the excitation of atomic nuclei is provided by so called electronic bridge processes. In these processes the nucleus is excited by energy that is transferred to it from the electron shell, which is much more likely than the direct photoexcitation due to the large overlap between the nucleus and the atomic electrons. In this contribution we will introduce the most promising electronic bridge processes for the excitation of atomic nuclei and we will present results for the nuclear excitation by electron transition (NEET). We will focus here on proposing different electron transition schemes that could be used to populate the ^{229}Th nucleus in doubly ionized thorium.

Natalia Oreshkina, MPI für Kernphysik Heidelberg
Variation of the fundamental constants in simple ions

Numerous few-electron atomic systems are considered, which can be effectively used for observing a potential variation of the fine-structure constant α and the electron-proton mass ratio m_e/m_p. We examine magnetic dipole transitions between hyperfine-splitting components in heavy highly charged H-like and Li-like ions with observably high sensitivity to a variation of α and m_e/m_p. The experimental spectra of the proposed systems consist of only one, strong line, which simplifies significantly the data analysis and shortens the necessary measurement time. Furthermore, we discuss that the contribution of possible systematic effects is much lower than in other systems proposed for such purposes.

Anton Peshkov, Helmholtz Institute Jena
Photoexcitation of atoms by Laguerre-Gaussian beamis

With the recent experimental advances in optics, it is possible to produce Laguerre-Gaussian (LG) light beams with a non-zero projection of the orbital angular momentum (OAM). During the last few years, it was shown that the OAM may affect the fundamental light-matter interaction processes. In a recent experiment, in particular, it was demonstrated for an atom placed on the axis of the incident LG light beam that the sublevel population of excited atomic states is determined by the beams OAM [1]. Following this experiment, we investigate theoretically the sublevel population of atoms with an arbitrary position with regard to the axis of the beam. We show that the sublevel population may vary significantly when the atoms are moved away from the beam axis. The population of the excited atoms is also found sensitive to the polarization, radial index, as well as the OAM of the incident LG beam; these effects can be observed experimentally by measuring the angular distribution of the subsequent fluorescence radiation.
[1] C. T. Schmiegelow et al., Nat. Commun. 7, 12998 (2016).
[2] A. A. Peshkov et al., Phys. Scr. 91, 064001 (2016).

Thomas Pfeifer, MPI für Kernphysik Heidelberg
X-ray diffractive imaging of intense-laser-driven dynamics in C60

Marc Rebholz, MPI für Kernphysik Heidelberg
XUV-pumpXUV-probe transient absorption experiments in small halogenated hydrocarbons

We present preliminary results of a XUV-pumpXUV-probe transient absorption experiment at the free-electron laser FLASH in Hamburg. The goal of our experiment is to determine how charge-rearrangement dynamics influence the dissociation of a molecule. The systems of investigation were small halogenated hydrocarbon molecules containing two iodine sites. We resonantly excited the 4d -¿ ?* transition with the first XUV pulse. This induces a breaking of one C-I bond. Shortly thereafter the exact same transition was probed with the second XUV pulse to investigate how the modified electronic environment can be accessed via the absorption spectrum of the dissociating molecule.

Ulf Saalmann, mpipks Dresden
Dynamic interference revisited

Georg Schmid, MPI für Kernphysik Heidelberg
Fragmentation Dynamics of Argon Dimers studied by XUV-IR Experiments at FLASH

XUV-IR pump-probe experiments on Argon dimers (Ar_2) were performed at the free-electron laser in Hamburg (FLASH).
Different XUV ($\hbar\omega = 27$eV) multiphoton ionization pathways such as interatomic coulombic decay (ICD), charge transfer, or frustrated multiple ionization could be identified by measuring the kinetic-energy release of the Coulomb-exploded dimer fragments using a reaction microscope. By applying an intense IR probe pulse ($I_{\mathrm{IR}} \sim 10^{14}$ W/cm^2), we were able to follow the dynamics of those fragmentation pathways in real time. Amongst other things, we could deduce an average lifetime of excited $\mathrm{Ar}^{2+}(3\mathrm{p}^{-3}nl)-\mathrm{Ar}$ states which decay via a fast charge transfer mechanism to $\mathrm{Ar}^{+}(3\mathrm{p}^{-2}nl)-\mathrm{Ar}^{+}(3\mathrm{p}^{-1})$.

Frans Schotsch, MPI für Kernphysik Heidelberg
Towards time-resolved single-photon double ionization of atoms

Double ionization by a single photon is one of the most fundamental processes involving electron correlation. In an XUV-IR pump-probe experiment we aim to investigate the ejection dynamics in a time-resolved manner, using the interferometric RABBIT technique as a clock. A reaction microscope allows for a kinematically complete detection of all reaction products in coincidence.

Bernd Schütte, Max-Born-Institut Berlin
Real-time observation of cluster charging in strong fields

Strong-field ionization of solid-density targets is fundamentally different from strong-field ionization of atoms, and can result in highly efficient absorption of laser energy. In order to understand fundamental strong-field phenomena, the investigation of isolated nanoparticles is advantageous, as energy is not dissipated into the environment, allowing one to focus on the primary laser-matter coupling mechanisms.
Here we trace the charging of Ar and Xe nanoparticles directly in the time domain by applying the recently developed ionization ignition method [1]. Seed electron generation by an intense XUV pulse allows us to temporally control the heating and ionization induced by a 1.5-ps NIR pulse, whose intensity ($I = 1.5 \times 10^{13}$ W/cm^2) is not sufficient to ionize neutral clusters. Surprisingly, we find that highly charged ions up to Xe^{15+} are produced. The average ion charge state increases exponentially during the rising edge of the NIR pulse, which is the first real-time observation of ionization avalanching. The experimental results will be compared with molecular dynamics calculations.
Our method provides new perspectives for the time-resolved investigation of strong-field phenomena in nanostructures, liquids and solids. It could e.g. be used to record the ablation of material in real time, which is relevant for practical applications such as laser machining.
[1] B. Schütte *et al.*, Phys. Rev. Lett. **116**, 033001 (2016)..

Andrey Surzhykov, Physikalisch-Technische Bundesanstalt
Scattering of twisted electrons

Jan Thiede, Universität Marburg
Triple ionization in strong laser fields

Mikhail Volkov, ETH Zürich
Attosecond dynamical Franz-Keldysh effect in dielectric nanofilms

We have applied attosecond transient absorption spectroscopy together with time-dependent density functional theory calculations to investigate ultrafast electron dynamics in a diamond nanofilm under the influence of a near-infrared (NIR) few-cycle laser pulse. The transient material response was probed with extreme-ultraviolet (XUV) attosecond pulses via valence-to-conduction band transitions. We observed transient features that oscillate at twice the NIR center frequency and fully recover after the interaction. Full multi-scale calculations reproduce the experimental results. By reducing the complexity of our theoretical analysis with an orbital decomposition method and by comparison with a simplified two-band parabolic model we were able to pinpoint the physical origin of observations as the dynamical Franz-Keldysh effect, which we observed for the first time in the close to petahertz domain.

Valentin Walther, mpipks Dresden
Nonlinear optics with semiconductor Rydberg-excitons

Recent decades have produced lots of research on exciton polaritons whose interactions are weak and (practically) local. The striking observation of Rydberg excitations in semiconductors has sparked a flurry of interest, because it might add some exaggerated Rydberg properties to the world of exciton physics. In this talk, I will discuss how these properties define the optical features of such systems. Starting with a few experimental parameters, we develop an atomistic multiple-level model of two-dimensional semiconductor excitons, including electromagnetic screening effects and additional modifications due to the band structure. Unlike ground state excitons, Rydberg excitons interact via dipole forces over large distances. We calculate excitonic pair interactions incorporating line crossings and state mixing. These long-ranged Rydberg interactions give rise to giant nonlocal optical nonlinearities, which we capture semi-analytically in a cluster expansion for low excitation densities. Finally, I will address the transverse photonic dynamics of a Rydberg-excited semiconductor cavity, where we expect an interesting competition of coherent and lossy effects.

Miriam Weller, Universität Frankfurt
Imaging the temporal evolution of molecular orbitals during ultrafast dissociation

We investigate the temporal evolution of molecular frame angular distributions of Auger electrons emitted during ultrafast dissociation of HCl following a resonant single photon excitation. The electron emission pattern changes its shape from that of a molecular σ-orbital to that of an atomic p-state as the system evolves from a molecule into two separated atoms. The talk will briefly introduce the topic of ultrafast dissociation, the measurement technique and present the results obtained.

Matthias Wollenhaupt, Universität Oldenburg
Bichromatic control of multi-photon ionization

Ultrashort bichromatic laser fields have emerged as an efficient tool to steer coherent electron dynamics. The beauty of bichromatic fields lies in their capability to disentangle different quantum pathways via frequency mixing and selection rules. Here we employ polarization-tailored bichromatic fields from a 4f polarization pulse shaper [1] to study resonance-enhanced multi-photon ionization of atoms as a prototype scenario for multi-pathway coherent control. Three-dimensional detection of the photoelectron momentum distribution by photoelectron imaging tomography provides detailed insights into the excitation and ionization dynamics. We present results of current experiments on the photoionization of potassium atoms with orthogonal linearly and counter-rotating circularly polarized bichromatic fields. In addition, the generation of vortex-shaped photoelectron wave packets from multi-photon ionization of potassium atoms with sequences of two time-delayed, counterrotating circularly polarized femtosecond laser pulses is demonstrated.
[1] S. Kerbstadt, L. Englert, T. Bayer, M. Wollenhaupt, J. Mod. Opt., accepted (2016)