A 11: Interaction with VUV and X-ray light I

Time: Monday 16:30–19:00 Location: V47.03

Invited Talk A 11.1 Mon 16:30 V47.03

Tracing ultrafast light-induced dynamics in small organic molecules — •Artem Rudenko — Max Planck Advanced Study Group at CFEL, Hamburg — Max Planck Institute for Nuclear Physics, Heidelberg

One of the main driving forces behind the development of novel intense short-pulsed light sources is a dream to "watch" chemical reactions in real time and to define properties of the transition states. The basic idea is that one short laser pulse (the 'pump') triggers the reaction, i.e., launches molecular wave packet, which is than "filmed" at different times by a second ('probe') pulse. The availability of ultrashort light bursts in a broad range of wavelengths, from the infrared and visible to XUV and X-ray domains opens exiting new possibilities for both, initiating different types of molecular dynamics and taking snapshots of molecular structure. In this contribution several illustrative examples of all optical, XUV-XUV and two-color pump-probe experiments on small organic molecules (e.g., CH4, C2H2, C2H4 etc.) will be presented, with a focus on mapping dissociation, isomerization, and H2/H3 elimination reactions. Current status and perspectives for different probe schemes, such as Coulomb explosion imaging, photoelectron diffraction and holography will be discussed.

Invited Talk A 11.2 Mon 17:00 V47.03

Rydberg atoms in strong laser fields — •Ulli Eichmann — Max-Born-Institute, D-12489 Berlin, Germany

The widely accepted and very successful concept of tunneling ionization of atoms in strong laser fields fails to describe the ionization of Rydberg atoms in these fields. This is despite the fact that the Keldysh-parameter, typically used to separate the tunneling from the multiphoton scenario, suggests just tunneling. Instead, one might be tempted to assume that the loosely bound Rydberg electron, which temporarily gains kinetic energy exceeding its binding energy by several orders of magnitude is easily removed from the atom. On the other hand, if one considers the loosely bound Rydberg electron as a free electron, absorption of photons is not possible.

We will present results on the ionization dynamics of Rydberg atoms in strong laser fields with intensities up to 5 10^{15}W/cm^2. We are able to prepare Rydberg atoms in a wide range of principal quantum numbers and more importantly, also in a wide range of angular momentum states. In the experiments we find a large percentage of Rydberg atoms surviving the very strong laser field. Moreover, direct position sensitive measurement of neutral atoms allows for the observation of deflection of surviving Rydberg atoms indicating unambiguously their interaction with the strong focused laser field. We will discuss the underlying physical mechanisms, particularly in the context of strong field stabilization of atoms.

A 11.3 Mon 17:30 V47.03

Multiwavelength anomalous diffraction at high x-ray intensity — •Sang-Kil Son[1], Henry N. Chapman[1,2], and Robin Santra[1,2] — [1]Center for Free-Electron Laser Science, DESY, Germany — [2]Department of Physics, University of Hamburg, Germany

The multi-wavelength anomalous diffraction (MAD) method is widely used in x-ray crystallography with synchrotron radiation to determine phase information by employing dispersion corrections from heavy atoms on coherent x-ray scattering. X-ray free-electron lasers (FELs) show promise for revealing the structure of single molecules or nanocrystals within femtoseconds, but the phase problem remains largely unsolved. Because of the extremely high fluence of FELs, samples experience severe and unavoidable electronic radiation damage, especially to heavy atoms, which hinders direct implementation of the MAD method with x-ray FELs. We propose a generalized version of the MAD phasing method at high x-ray intensity. We demonstrate the existence of a Karle–Hendrickson-type equation for the MAD method in the high-intensity regime and calculate relevant coefficients with electronic damage dynamics and accompanying changes of the dispersion correction. Here we present the xatom toolkit to simulate detailed electronic damage dynamics and discuss how the proposed method is applicable to the phase problem in femtosecond x-ray nanocrystallography.

A 11.4 Mon 17:45 V47.03

Enhanced nonlinear response of Ne^{8+} to intense ultrafast X-rays — •Arina Sytcheva[1], Stefan Pabst[1,2], Sang-Kil Son[1], and Robin Santra[1,2] — [1]Center for Free-Electron Laser Science, DESY, Germany — [2]Department of Physics, University of Hamburg, Germany

We report on the possible reasons for the discrepancy between the theoretical two-photon ionization cross section (TPICS), $\sim 4 \cdot 10^{-56}$ cm^{4}s, of Ne^{8+} obtained within the lowest nonvanishing order of perturbation theory (LOPT) for continuous light and the experimental value, 7×10^{-54} cm^{4}s, reported in [Phys. Rev. Lett. **106**, 083002 (2011)] at a photon energy of 1110 eV and pulse bandwidth of 11 eV. We consider Ne^{8+} exposed to coherent and chaotic ensembles of intense X-ray pulses using the time-dependent configuration-interaction singles (TDCIS) method and LOPT, respectively. A coherent ensemble of pulses of 11-eV bandwidth centered at 1110 eV yields a TPICS enhanced by a factor of ~ 1.25 with respect to the continuous-light TPICS, while a chaotic ensemble yields an enhancement factor of ~ 40. This enhancement is due to the presence of the one-photon $1s^2$–$1s4p$ resonance located at 1127 eV and the finite bandwidth of the X-ray pulse. Using the TDCIS approach, we also show that, for currently available radiation intensities, two-photon ionization of a $1s$ electron in neutral neon is much less probable than one-photon ionization of a valence electron.

A 11.5 Mon 18:00 V47.03

Resonance-Enhanced X-ray Multiple Ionization of Heavy Atoms at LCLS — •Benedikt Rudek[1,2], Daniel Rolles[1,3], Artem Rudenko[1,2], Sang-Kil Son[5], Lutz Foucar[1,2], Benjamin Erk[1,3], Sascha Epp[1,3], Robert Hartmann[4], Lothar Strüder[1,4], Robin Santra[5,6], Joachim Ullrich[1,3], and the CAMP collaboration[5,7] — [1]Max Planck Advanced Study Group at CFEL, Hamburg — [2]MPI für medizinische Forschung, Heidelberg — [3]MPI für Kernphysik, Heidelberg — [4]MPI Halbleiterlabor, München — [5]Center for Free-Electron Laser Science (CFEL), DESY, Hamburg — [6]Universität Hamburg — [7]LCLS, SLAC, Menlo Park

The interaction of ultra-intense X-rays with rare gas atoms was studied at LCLS, where series of inner shell ionizations yield unprecedentedly high charge states within a single shot. Xenon, in particular, was ionized up to 36+, which requires ionization energies far exceeding the photon energy. Combined experimental and theoretical analysis of ion charge state distributions and simultaneously recorded fluorescence spectra showed that resonant excitations are responsibele for the enhanced ionization. This resonantly enhanced X-ray multi-ionization process (REXMI) is predicted to boost ionization in certain ranges of photon energy and thus enhance radiation damage in the vicinity of heavy atoms.

A 11.6 Mon 18:15 V47.03

Terahertz streaking reveals chirped Auger electron emission — •Bernd Schütte[1,3], Sebastian Bauch[2], Ulrike Frühling[1], Marek Wieland[1], Michael Gensch[4,5], Elke Plönjes[4], Thomas Gaumnitz[1], Armin Azima[1], Michael Bonitz[2], and Markus Drescher[1] — [1]Universität Hamburg — [2]Christian-Albrechts-Universität, Kiel — [3]Max-Born-Institut, Berlin — [4]Deutsches Elektronen-Synchrotron DESY, Hamburg — [5]Helmholtz-Zentrum Dresden Rossendorf

We have investigated the Auger decay in xenon and krypton atoms in the presence of a strong terahertz field. The experiments were performed at two different light sources, namely the free-electron laser in Hamburg (FLASH) and a source of high-order harmonic generation (HHG). By measuring at different phases of the terahertz streaking field and from different observation directions, a time-dependent energetic chirp of the Auger electrons was observed. The origin of this behavior was found to be an energy transfer between the Auger electron and the earlier emitted photoelectron in the laser field. This interaction alters the Auger electron energy depending on its emission time with respect to the photoelectron. The experimentally obtained data agree well with theoretical calculations, thereby facilitating the proposed model. Thus, terahertz streaking proves to be an excellent tool for recording electron dynamics in the femtosecond range.

A 11.7 Mon 18:30 V47.03

Nuclear excitation and collective effects with coherent x-ray

light — Andre Junker, Christoph H. Keitel, and •Adriana Pálffy — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

With present and upcoming x-ray light sources such as the X-ray Free Electron Laser (XFEL), the direct interaction between nuclei and super-intense laser fields has become feasible, opening the new field of nuclear quantum optics [1]. One of the main difficulties with driving nuclear transitions arises from the weak coupling between nuclei and the electromagnetic field leading to very narrow bandwidths. In solid state targets that allow recoilless nuclear excitation and decay, the excitation caused by a single photon may be shared by a large number of nuclei, forming a collective excited state with a sometimes significantly larger decay width than that of a single nucleus.

Following this line, in this work we investigate theoretically the direct laser-nucleus interaction taking into account for the first time the line broadening due to collective effects. Furthermore, we update former estimates [2] considering the experimental advances of the XFEL regarding photon frequency, focus and intensity. Our results show an enhancement by several orders of magnitude of the excited state population compared to previously reported values [2].

[1] T. J. Bürvenich, J. Evers and C. H. Keitel, Phys. Rev. Lett. **96** 142501 (2006).

[2] A. Pálffy, J. Evers and C. H. Keitel, Phys. Rev. C **77** 044602 (2008), A. Pálffy, J. Mod. Opt. 55, 2603 (2008).

A 11.8 Mon 18:45 V47.03

Imaging electronic quantum motion with light — •Gopal Dixit[1], Oriol Vendrell[1], and Robin Santra[1,2] — [1]Center for Free-Electron Laser Science, DESY, Notkestrasse 85, D-22607 Hamburg, Germany — [2]Department of Physics, University of Hamburg, D-20355 Hamburg, Germany

Imaging the quantum motion of electrons not only in real time, but also in real space is essential to understand for example bond breaking and formation in molecules, and charge migration in peptides and biological systems. Time-resolved imaging (TRI) interrogates the unfolding electronic motion in such systems. We show that scattering patterns, obtained by X-ray TRI from an electronic wavepacket, encode spatial and temporal correlations that deviate from the common notion of the instantaneous electronic density as the key quantity being probed. X-ray TRI can be realized with the advent of novel light sources such as X-ray free electron lasers. Surprisingly, the patterns provide an unusually visual manifestation of the quantum nature of light. This quantum nature becomes central only for non-stationary electronic states. The illustrative example used here as a proof of principle lies in the time and energy range of interest corresponding to the dynamics of valence electrons in more complex molecular and biological systems.