A 1: Interaction with VUV and X-ray light (FEL) I

Time: Monday 10:30–13:00 Location: BAR 205

Invited Talk A 1.1 Mon 10:30 BAR 205

First results from multi-coincidence experiments at LCLS — •Daniel Rolles[1,2], Benedikt Rudek[1,3], Artem Rudenko[1,3], Benjamin Erk[1,3], Lutz Foucar[1,2], Sascha Epp[1,3], Ilme Schlichting[1,3], Lothar Strüder[1,4], Joachim Ullrich[1,2], and the CAMP collaboration[1] — [1]Max Planck Advanced Study Group at CFEL, Hamburg — [2]Max-Planck-Institut für medizinische Forschung, Heidelberg — [3]Max-Planck-Institut für Kernphysik, Heidelberg — [4]MPI Halbleiterlabor, München

The CFEL-ASG Multi-Purpose (CAMP) instrument [1] designed, built, and operated by the the Max Planck Advanced Study Group (ASG) at the Center for Free Electron Laser Science (CFEL) with its unique combination of large-area, single-photon counting pnCCD detectors and electron and ion spectrometers allows simultaneous detection of scattered and fluorescent photons, photoelectrons and photoions on a shot-by-shot basis. It was successfully commissioned at the LCLS AMO beamline in November 2009 and has since been used for fifteen experiments ranging from AMO, solid-state, surface, and plasma physics to material sciences, chemistry, and biology. Here, we focus on the experiments that took advantage of the multi-particle detection capabilities, namely electron, ion and fluorescence spectroscopy experiments on atoms, (laser-aligned) molecules, and clusters as well as diffractive imaging experiments on clusters and nanoparticles with simultaneous ion detection.

[1] L. Strüder et al., Nucl. Instr. Meth. Phys. Res. A 614 (2010) 483.

Invited Talk A 1.2 Mon 11:00 BAR 205

X-FEL induced multi-photon processes — •Bertold Krässig — Argonne National Laboratory, Argonne, IL 60439, USA

The opening of the world's first X-ray Free Electron Laser (X-FEL), the Linac Coherent Light Source (LCLS) at the SLAC National Accelerator Laboratory, marked an important milestone in the effort to probe matter at the length and time scales of atoms and molecules. Never before has it been possible to direct hundreds of x-ray photons (800 eV-8000 eV) onto every single atom at the focus of the x-ray beam during a single pulse lasting 200 fs or less. In this talk I will present results of some of the first experiments carried out at the LCLS with the goal of exploring multiphoton processes in atoms at short wavelengths. The atom of choice in these experiment was neon. We found the target atoms to become continually altered during a single x-ray pulse by absorbing multiple photons sequentially, in some cases removing all 8 valence electrons if the photon energy is below the 1s ionization threshold, or all 10 electrons if the photon energy is above the 1s ionization threshold of Ne^{9+}. At sufficiently high beam intensities and energies above the 1s ionization thresholds, both 1s electrons can be sequentially removed before Auger decay occurs. Such creation of a hollow atom leads to a temporary reduction in the probability for photoabsorption, and the reduction becomes the more prominent the longer the lifetime of the hollow atom state. Our results demonstrate how different, and in many ways complementary, X-FEL sources are as a research tool compared to traditional synchrotron radiation facilities.

Invited Talk A 1.3 Mon 11:30 BAR 205

X-ray femtochemistry: Mapping the electronic structure of molecules during chemical reactions with x-ray spectroscopy — •Philippe Wernet — Helmholtz-Zentrum Berlin für Materialien und Energie

We use ultrafast x-ray spectroscopy to map the electronic structure of molecules during chemical reactions in the gas phase and in solution. X-ray spectroscopy gives unique access to the electronic structure and hence allows for unprecedented insight into the transient states of atoms and molecules. Our recent results will be used to illustrate this: Femtosecond photoelectron spectroscopy with a high harmonic generation set up in the lab revealed new insight into the ultrafast dissociation of Br2 molecules in the gas phase. Femtosecond resonant inelastic x-ray scattering (RIXS) at the free electron laser LCLS for the first time allowed us to track both the occupied and the unoccupied valence orbitals of a molecule during dissociation. We mapped the electronic structure of Fe(CO)5 in real time during its dissociation to Fe(CO)4 and CO in solution in a symmetry-sensitive and element-selective way and locally at the Fe atom. With this and with our research on bonding and structure of water and molecules in the gas phase and in solution a perspective on chemical dynamics with ultrashort x-ray pulses from lab- and accelerator-driven short-pulse x-ray sources will be given.

A 1.4 Mon 12:00 BAR 205

Nonlinear atomic response to ultraintense and ultrashort x-ray pulses — Gilles Doumy[1,2], C. Roedig[1], •Sang-Kil Son[3], C. Blaga[1], A. DiChiara[1], A. Agostini[1], L.F. DiMauro[1], R. Santra[3,4], N. Berrah[5], C. Bostedt[6], J.D. Bozek[6], P.H. Bucksbaum[7], J. Cryan[7], L. Fang[5], S. Ghimire[7], M.J. Glownia[7], M. Hoener[5], E.P. Kanter[2], B. Krässig[2], M. Messerschmidt[6], D. Reis[7], N. Rohringer[8], and L. Young[2] — [1]The Ohio State University, USA — [2]Argonne National Laboratory, USA — [3]Center for Free-Electron Laser Science, DESY, Germany — [4]University of Hamburg, Germany — [5]Western Michigan University, USA — [6]Linac Coherent Light Source, SLAC National Accelerator Laboratory, USA — [7]PULSE Institute, SLAC National Accelerator Laboratory, USA — [8]Lawrence Livermore National Laboratory, USA

The nonlinear response of neon atoms to ultraintense, ultrashort x-ray pulses is investigated with the LCLS x-ray free-electron laser. The production of Ne^{9+} is observed at kilovolt x-ray photon energies below the absorption edge of the Ne^{8+} ground state and demonstrates a clear quadratic dependence on fluence. Theoretical analysis shows that the production is a combination of direct 2-photon ionization of the Ne^{8+} ground state and a high-order sequential process involving two 1-photon ionization processes via transient excited states on a time scale faster than the Auger decay. We find that the nonlinear 2-photon ionization cross section is orders of magnitude larger than expected.

A 1.5 Mon 12:15 BAR 205

Probing XUV FEL pulses with pump-probe autocorrelation — •Arne Senftleben[1], Moritz Kurka[1], Yuhai Jiang[1], Artem Rudenko[2], Oliver Herrwerth[3], Lutz Foucar[2], Kai-Uwe Kühnel[1], Matthias Kling[3], Stefan Düsterer[4], Claus-Dieter Schröter[1], Robert Moshammer[1], and Joachim Ullrich[1] — [1]Max-Planck-Institut für Kernphyik, Heidelberg — [2]Max-Planck Advanced Study Group at CFEL, Hamburg — [3]Max-Planck-Institut für Quantenoptik, Garching — [4]DESY, Hamburg

Using a split-mirror stage combined with a reaction microscope, autocorrelation traces of XUV pulses from the free-electron laser (FEL) at Hamburg were recorded for multiple ionization of atomic and molecular targets. Two characteristic time scales can be identified: the pulse-envelope duration and a substantially shorter coherence time. We conclude that the latter is a consequence of the internal pulse structure generated by the laser source. It is envisioned that this allows future pump-probe measurements with temporal resolution beneath the pulse duration. We reproduce and further characterise the FEL pulses using a partial-coherence model.

A 1.6 Mon 12:30 BAR 205

Novel light from high-order harmonic generation manipulated by XUV light — •Christian Buth[1], Markus C. Kohler[1], Joachim Ullrich[1,2], and Christoph H. Keitel[1] — [1]Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany — [2]Max-Planck Advanced Study Group at CFEL, 22607 Hamburg, Germany

We theoretically combine high harmonic generation (HHG) with resonant XUV excitation of a core electron into the transient valence vacancy that is created in the course of the HHG process: the first electron performs a HHG three-step process whereas, the second electron Rabi flops between the core and the transient valence vacancy. The modified HHG spectrum due to recombination with the valence and the core is determined and analyzed for krypton on the $3d \rightarrow 4p$ resonance in the ion in the light of the Free Electron Laser in Hamburg (FLASH). Our prediction offers novel prospects for nonlinear XUV physics, attosecond x rays, and tomographic imaging of core orbitals. — arXiv:1012.4930

A 1.7 Mon 12:45 BAR 205

Statistical modeling of FEL pulse shapes using a partial-coherence colored-noise approach — •Thomas Pfeifer[1], Yuhai

Jiang[1], Stefan Düsterer[2], Robert Moshammer[1] und Joachim Ullrich[1] — [1]Max-Planck Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany — [2]Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, 22605 Hamburg, Germany

Most free-electron lasers (FELs) producing intense pulses in the extreme ultraviolet (XUV) and x-ray spectral regions are currently operated in the self-amplified spontaneous emission (SASE) mode. Under these conditions, light emission starts from (quantum) noise and amplification of a light pulse occurs by a relativistic electron bunch oscillating in an undulator. This noisy origin of SASE FEL pulses results in dramatic fluctuations of pulse energy and pulse shape from shot to shot. Here, we present a simple numerical approach to model the pulse-shape statistics of such FEL pulses, even without specific knowledge of technical machine parameters such as electron bunch energy characteristics or undulator geometry. Only the measured average spectral shape and average pulse duration are required to produce sets of FEL pulse shapes for the exponential-growth ("linear") regime that fulfill statistical properties required by FEL theory. The modeled single-shot spectral shapes agree well with measured single-shot spectra. This method allows to include statistical variations of FEL pulse shapes into simulations of nonlinear FEL–matter interaction in pump–probe experiments. It will be shown that agreement of simulation results and experimental data is only achieved when pulse-shape statistics are accounted for.