A 12: Interaction with strong or short laser pulses III

Time: Tuesday 10:30–12:30 Location: V47.03

Invited Talk A 12.1 Tue 10:30 V47.03

Two-color photoionization studies at XUV and X-ray Free Electron Lasers — •Michael Meyer — European XFEL GmbH, Albert-Einstein-Ring 19, D-22761 Hamburg, Germany

The combination of intense femtosecond X-ray and NIR pulses produced by Free Electron Lasers (FEL) and synchronized optical lasers, respectively, offers various new opportunities to investigate the dynamics of atomic photoionization. Some recent results obtained at the XUV-FEL FLASH in Hamburg and the first X-ray FEL, the LCLS in Stanford, will be presented. In the experiments at FLASH, the optical dressing field gives rise to the so-called two-color Above Threshold Ionization, which could be studied for the first time in a regime free from unwanted interference effects. For resonant excitations, e.g. 3d –> 5p in atomic Kr, the NIR field causes a strong modification of the decay dynamics, which was experimentally investigated via the intensity-dependent shift of the resonance position and via the competition between resonant and direct Auger processes. Recent experiments at LCLS have taken advantage of the very short (2-5 fs) pulse duration, which coincides with the lifetime of the Ne 1s core hole and with the temporal width of one optical cycle of the NIR (800 nm) dressing laser. As a direct consequence, the angle-resolved KLL Auger spectra reveal strong intensity modulations induced by sub-cycle interferences, i.e. by the coherent emission of electrons produced during one cycle of the superimposed optical field.

A 12.2 Tue 11:00 V47.03

Attosecond Two-Electron Dynamics in Non-Sequential Double Ionization of Argon using Ultra-short Laser Pulses — •Nicolas Camus[1], Bettina Fischer[1], Manuel Kremer[1], Vandana Sharma[1], Artem Rudenko[1,2], Boris Bergues[3], Mathias Kübel[3], Nora G. Johnson[3], Mathias F. Kling[3], Thomas Pfeifer[1], Joachim Ullrich[1,2], and Robert Moshammer[1] — [1]Max-Planck-Institut für Kernphysik, Heidelberg — [2]Max-Planck Advanced Study Group at CFEL, Hamburg — [3]Max-Planck-Institut für Quantenoptik, Garching

We report on a kinematically complete experiment on non-sequential double ionization (NSDI) of argon using CEP characerized 6 fs laser pulses in combination with a reaction microscope. In NSDI the first ionized electron is driven back to the parent ion causing ionization of the second electron. In this study we choose a rather low intensity where the energy of the re-colliding electron is too small for impact ionization but large enough to excite the remaining Ar^+ ion. Loosing almost all of its kinetic energy during re-collision, the re-colliding electron gets recaptured such that a doubly-excited state is formed. Within a classical simulation we demonstrate that this highly excited complex has lost memory of its creation. We consistently relate measured momentum differences between the two electrons to time differences and, thus, follow the ionization of the doubly excited state in the laser field on a sub-fs time scale. A striking agreement between experimental and simulated distributions is observed with a most likely time difference between the two electrons for leaving the atom of 200 ± 100 as.

A 12.3 Tue 11:15 V47.03

Noisy pulses enhance temporal resolution in pump–probe spectroscopy — •Kristina Meyer, Christian Ott, Philipp Raith, Andreas Kaldun, Yuhai Jiang, Arne Senftleben, Moritz Kurka, Robert Moshammer, Joachim Ullrich, and Thomas Pfeifer — Max-Planck Institut für Kernphysik, Heidelberg

Observing dynamical processes relies on the availability of timing-controlled events (e.g. laser pulses) that are shorter in duration than the typical time scale of the dynamics to be measured. The production of ultra-short pulses is nowadays typically based on the exceptional coherence properties of laser light, hiding the fact that noisy, only partially coherent, light sources could have benefits. The development of Free-Electron Laser (FEL) sources has led to a reevaluation of temporal noise since self-amplified spontaneous emission (SASE) FEL pulse shapes vary widely from shot to shot. Here, we show that such noisy electric fields can be used to resolve temporal features of quantum dynamics with high precision that would be inaccessible using fully coherent pulses of the same average pulse duration. Our concept of noise-enhanced pump–probe spectroscopy is demonstrated for the example of a recently performed experiment investigating the two-photon double ionization of D_2 [1]. The method enables the measurement of dynamics on time scales that are shorter than the average pulse duration by more than a factor of 10. This finding does not only have important consequences for FEL science, but can lead to general paradigm shifts in ultrafast physics, including attosecond spectroscopy.

[1] Y. H. Jiang et al., Phys. Rev. A 81, 051402 (2010)

A 12.4 Tue 11:30 V47.03

Time-Resolved Photoelectron Diffraction in Laser-Aligned Molecules at Free-Electron Lasers — •Denis Anielski[1,2], Alaa Al-Shemmary[3], Rebecca Boll[1,2], Lauge Christensen[4], Sankar De[4], Siarhei Dziarzhytski[3], Benjamin Erk[1,2], Jochen Küpper[5,6], Terence Mullins[5], Harald Redlin[3], Daniel Rolles[1,7], Artem Rudenko[1,2], Kirsten Schnorr[2], Henrik Stapelfeldt[4], Stephan Stern[5,6], Sebastian Trippel[5], and Joachim Ullrich[1,2] — [1]Max Planck Advanced Study Group, CFEL, Hamburg — [2]MPI für Kernphysik, Heidelberg — [3]DESY, Hamburg — [4]University of Aarhus, Denmark — [5]CFEL, DESY, Hamburg — [6]Universität Hamburg — [7]MPI für medizinische Forschung, Heidelberg

The possibility to obtain femtosecond time-resolved information on single molecules with Angstrom spatial resolution is one of the driving forces for the development of short-pulse VUV and X-ray sources such as FELs. Time-resolved photoelectron diffraction allows to investigate fundamental chemical reaction dynamics in small to medium-sized organic molecules. At FLASH, OCS molecules were adiabatically aligned using a ns Nd:YAG laser ($\langle\cos^2(\theta)\rangle_{2D} = 0.82$), dissociated with a fs Ti:Sa laser and then photoionized by the FEL for different delays between Ti:Sa and FEL. A velocity map imaging spectrometer recorded the angular distributions of the photoelectrons. By comparing these distributions with multiple scattering calculations, the changing geometric structure of the molecule can be made visible.

A 12.5 Tue 11:45 V47.03

Photoelectron Diffraction in Laser-Aligned p-Fluorophenylacetylene (p-FAB) at LCLS — •Rebecca Boll[1,2], Denis Anielski[1,2], Christoph Bostedt[3], Lauge Christensen[4], Ryan Coffee[3], Sankar De[4], Sascha Epp[1,2], Benjamin Erk[1,2], Lutz Foucar[1,5], Jochen Küpper[6,7], Daniel Rolles[1,5], Arnaud Rouzee[8], Benedikt Rudek[1,2], Artem Rudenko[1,2], Henrik Stapelfeldt[4], Stephan Stern[6], Sebastian Trippel[6], and Joachim Ullrich[1,2] — [1]Max Planck Advanced Study Group at CFEL, Hamburg, Germany — [2]Max Planck Institute for Nuclear Physics, Heidelberg, Germany — [3]SLAC National Accelerator Laboratory, Stanford, USA — [4]Aarhus University, Denmark — [5]Max Planck Institute for Medical Research, Heidelberg, Germany — [6]CFEL, Hamburg, Germany — [7]University of Hamburg, Germany — [8]Max Born Institute, Berlin, Germany

Photoelectron diffraction patterns of adiabatically laser-aligned and mixed-field oriented p-FAB molecules were measured using femtosecond FEL-pulses at different photon energies at LCLS. Ions and electrons were recorded simultaneously in a double-sided velocity map imaging spectrometer installed in the CFEL-ASG MultiPurpose (CAMP) chamber. From the obtained angular distributions of the photoelectrons that are emitted and scattered within a single molecule, it is possible to retrieve information on the molecular structure on an Angstrom length scale.

A 12.6 Tue 12:00 V47.03

Coulomb explosion imaging of small organic molecules at LCLS — •Benjamin Erk[1,2], Artem Rudenko[1,2], Daniel Rolles[1,3], Benedikt Rudek[1,2], Lutz Foucar[1,3], Sascha Epp[1,2], Max Cryle[3], Ilme Schlichting[1,3], Arnaud Rouzee[4], Axel Hundertmark[4], Tatiana Marchenko[5], Mark Simon[5], Christoph Bostedt[6], Sebastian Schorb[6], Kiyoshi Ueda[7], Claus Dieter Schroeter[2], and Joachim Ullrich[1,2] — [1]Max-Planck ASG at CFEL, DESY, Hamburg, Germany — [2]MPI für Kernphysik, Heidelberg, Germany — [3]MPI für Medizinische Forschung, Heidelberg, Germany — [4]MBI, Berlin, Germany — [5]LCPMR, Paris, France — [6]LCLS, SLAC National Accelerator Laboratory, Menlo Park, USA — [7]IMRAM, Tohoku University, Sendai, Japan

Fragmentation of small organic molecules by intense few-fs soft X-ray

FEL-pulses has been studied using Coulomb explosion imaging. To increase and localize X-ray absorption, we studied methylselenol and ethylselenol compounds containing one high-Z atom, selenium. The experiment was conducted in the CFEL-ASG Multi-Purpose (CAMP) end station installed at the AMO beamline of the LCLS at Stanford. By measuring kinetic energies and emission angles of few ionic fragments in coincidence as a function of FEL intensity, we study sequential multi-photon absorption and reconstruct fragmentation pathways as well as molecular geometry at the moment of explosion. The results yield unique information on structural rearrangement and charge redistribution in the molecule, which has direct implications for radiation damage induced by intense X-ray pulses.

A 12.7 Tue 12:15 V47.03

Molecular dynamics studied with XUV pump–probe experiments — Yuhai Jiang[1], •Arne Senftleben[1], Moritz Kurka[1], Artem Rudenko[2], Kirsten Schnorr[1], Georg Schmid[1], Kristina Meyer[1], Thomas Pfeifer[1], Lutz Foucar[2], Oliver Herrwerth[3], Matthias Kübel[3], Matthias Kling[3], Joachim Ullrich[1], Claus-Dieter Schröter[1], and Robert Moshammer[1] — [1]Max-Planck-Insitut f. Kernphysik, Heidelberg — [2]Max-Planck Advanced Study Group, Hamburh — [3]Max-Planck-Institut f. Quantenoptik, Garching

Present free-electron lasers deliver pulses of extreme ultra-violet (XUV) radiation as short tens of femtoseconds. This enables to study molecular dynamics in real time using the pump–probe technique: Here, one pulse ionizes the molecule and initiates changes to its structure, while a time-delayed second pulsed induced Coulomb explosion. By measuring the momenta of the resulting fragment ions with a reaction microscope, we can reconstruct the molecular structure at the time of the second pulse. Through variation of the time-delay from shot to shot, a "molecular movie" is taken. We will show results from our recent investigations of dissociating diatomic molecules (D_2, N_2, I_2) and isomerization reactions in acetylene and ethylene.