

MO 1: Attosecond Science I (joint session A/MO)

Time: Monday 10:30–12:30

Location: K 1.011

Invited Talk

MO 1.1 Mon 10:30 K 1.011

Phase measurement and control with attosecond self-probing spectroscopy — ●MICHAEL KRÜGER — Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

Attosecond spectroscopy is based steering electron dynamics by the electric field waveform of a strong laser field. High-harmonic generation (HHG), the mechanism underlying the production of attosecond pulses in the extreme ultraviolet (XUV), provides an in-built spectroscopic pump-probe measurement with extremely high spatial and temporal accuracy. The amplitude and phase of the emitted XUV radiation encodes all parts of the light-matter interaction in the recollision process, including ionization, propagation and photo-recombination. Here we present two applications of attosecond self-probing spectroscopy. In the first experiment, we initiate HHG with an XUV pulse instead of tunneling ionization, enabling us to measure and control the XUV photo-ionization dynamics in the presence of a strong infrared field in amplitude and phase [1]. In the second experiment, we compare HHG from two atomic species using linear XUV interferometry and extract the absolute difference in the photo-recombination dipole phase with high spatial resolution [2]. Our method gives access to hitherto inaccessible phase information, enabling attosecond control of HHG and tomographic reconstruction of the electronic structure of matter.

[1] D. Azoury et al., Nat. Comm. 8, 1453 (2017). [2] D. Azoury et al., manuscript in preparation (2017).

Invited Talk

MO 1.2 Mon 11:00 K 1.011

Molecular Orbital Imprint in Laser-Driven Electron Recollision — FELIX SCHELL, TIMM BREDTMANN, CLAUS PETER SCHULZ, SERGUEI PATCHKOVSKII, MARC VRAKING, and ●JOCHEN MIKOSCH — Max-Born-Institute, Max-Born-Strasse 2A, 12489 Berlin

Electrons released by strong-field ionization from atoms, molecules, or in solids can be accelerated in the oscillating laser field and driven back to their ion core. The ensuing interaction, phase-locked to the optical cycle, initiates the central processes underlying attosecond science. A key long-standing assumption regards the returning electron wavepacket as a plane wave. Here we study laser-induced electron rescattering associated with two different ionization continua in the same, spatially aligned, polyatomic molecule [1]. We show by experiment and theory that the electron return probability is in fact molecular-frame dependent and carries structural information on the ionized orbital. Pronounced deviations of the returning wavepacket from plane-wave character have to be accounted for in analyzing attosecond experiments based on strong laser fields.

[1] F. Schell, T. Bredtmann, C.P. Schulz, S. Patchkovskii, M.J.J. Vrakking, and J. Mikosch (submitted)

MO 1.3 Mon 11:30 K 1.011

Valley-resolved Electronic Coherences in Silicon Observed by Attosecond Transient Absorption Spectroscopy — ●MICHAEL ZÜRCH¹, PETER M. KRAUS¹, HUNG-TZU CHANG¹, SCOTT K. CUSHING¹, DANIEL M. NEUMARK^{1,2}, and STEPHEN R. LEONE^{1,2,3} — ¹Department of Chemistry, University of California, Berkeley, CA 94720, USA — ²Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA — ³Department of Physics, University of California, Berkeley, CA 94720, USA

Understanding the absorption of light and subsequent carrier dynamics in semiconductors plays a crucial role for optimizing next-generation photonic devices for increasingly faster performance. Here, attosecond transient absorption spectroscopy is employed for studying electronic coherences in single crystalline silicon during excitation by an intense 5-fs optical pulse. Transient absorption changes in the conduction band (CB) of silicon are monitored by an attosecond pulse at the silicon L-edge. In a frequency-over-energy Fourier analysis of the recorded transient absorption in comparison to the band structure coherences are identified. The data suggests that the optical pulse can coherently couple the valence band (VB) and CB at various critical points of the band structure. The time domain measurement allows measuring lifetimes of these coherences as well as their sequence of generation. The results provide insight into complex couplings between bands that take place during excitation with broadband ultrashort laser pulses, an effect that should be general for most semiconductor materials.

MO 1.4 Mon 11:45 K 1.011

Light-Field-Driven Landau-Zener-Stückelberg interferometry — ●TAKUYA HIGUCHI and PETER HOMMELHOFF — Department Physik, Friedrich-Alexander-Universität Erlangen-Nürnberg (FAU), 91058 Erlangen

When electrons are placed under an optical field which is stronger than the internal field inside of a matter, their dynamics turn into non-perturbative regime. Recently, we have observed a transition from perturbative to non-perturbative electron dynamics in graphene [1]. The electron dynamics in the non-perturbative regime are well described by the light-field-driven dynamics. In particular, this field-driven dynamics has the same form as repeated Landau-Zener transitions and resultant quantum-path interference around the touching points of graphene's electron bands, known as Landau-Zener-Stückelberg (LZS) interference.

Such Landau-Zener like dynamics are found in various physical systems. Therefore, a comprehensive understanding of this light-field-driven LZS interferometry can provide a general prescription for formulating strong-field dynamics. The purpose of this presentation is to clarify the relations between the parameters involved in this light-field-driven LZS interferometry. For example, we clarify the condition defining the perturbative and the non-perturbative regimes. Relations with strong-field physics in atomic gaseous systems, such as the non-adiabaticity parameter and the intensity parameter, will be discussed.

[1] T. Higuchi, C. Heide, K. Ullmann, H. B. Weber, and P. Hommelhoff, Nature 550, 224 (2017).

MO 1.5 Mon 12:00 K 1.011

Direct observation of a core-hole spin-orbit wave packet using strong-field spectroscopy — ●ALEXANDER BLÄTTERMANN, MAXIMILIAN HARTMANN, PAUL BIRK, VEIT STOOSS, GERGANNA BORISOVA, CHRISTIAN OTT, and THOMAS PFEIFER — Max-Planck-Institut für Kernphysik, Heidelberg, Deutschland

Spin-orbit coupling and its dynamics play an important role from atomic physics through chemical reactions of molecules up to magnetic properties in material science. Thus far, experimental studies were limited to spin-orbit dynamics in the valence shell of atoms [1]. Here, we present the time-domain observation of ultrafast coherent dynamics among 4d-core-hole states in xenon. The wave packet is created by an extreme-ultraviolet (XUV) light pulse, which lifts a 4d electron to the 6p shell. The natural time-scale of 2.1 fs is dictated by the intrinsically strong fine-structure splitting among the two possible core-hole configurations. Strong-field spectroscopy, i.e., probing the XUV-induced dynamics by means of an intense near-infrared laser pulse [2], allows us to observe these fast wave packet dynamics via a multiphoton-induced coupling of the studied states. These results open a route to site-selective and element-specific studies of coherent electron dynamics in larger systems such as polyatomic molecules and complex solids, where core electrons provide local probes of the electronic structure and dynamics.

[1] Nature 466, 739 (2010)

[2] Science 354, 738 (2016)

MO 1.6 Mon 12:15 K 1.011

Observation of coherent spin-orbit wave packet dynamics in strong-field generated xenon ions — ●MAXIMILIAN HARTMANN, ALEXANDER BLÄTTERMANN, PAUL BIRK, VEIT STOOSS, GERGANNA BORISOVA, CHRISTIAN OTT, and THOMAS PFEIFER — Max-Planck-Institut für Kernphysik, Heidelberg, Deutschland

Removing the p-shell valence electron of noble gas atoms generates ions, whose ground state forms a doublet due to spin-orbit interaction. As shown theoretically for neon and xenon [1] and experimentally for krypton [2], both ionic state configurations can be populated in a partially coherent way by means of strong-field ionization, which will give rise to coherent dynamics of the thus created wave packet. For the case of xenon, the fine structure splitting reaches 1.3 eV, which leads to dynamics on a time scale of 3 fs.

Here, we present the observation of coherent wave-packet dynamics in field-ionized xenon generated by intense sub-2 cycle laser pulses and probed by means of attosecond transient absorption spectroscopy. Our results constitute a promising step towards understanding the coher-

ent response of multi-electron systems exposed to strong laser fields –
especially in the regime of sub-cycle field-induced ionization.

[1] Phys. Rev. A 79, 053402 (2009)

[2] Nature 466, 739 (2010)