A 11: Attosecond physics II

Time: Tuesday 14:00–16:00 Location: f107

Invited Talk A 11.1 Tue 14:00 f107

Probing electronic wavefunctions and chiral structure using all-optical attosecond interferometry — •Michael Krüger[1,2], Doron Azoury[1], Omer Kneller[1], Shaked Rozen[1], Barry D. Bruner[1], Alex Clergerie[3], Bernard Pons[3], Baptiste Fabre[3], Yann Mairesse[3], Oren Cohen[2], Olga Smirnova[4], and Nirit Dudovich[1] — [1]Department of Physics of Complex Systems, Weizmann Institute of Science, 76100 Rehovot, Israel — [2]Department of Physics and Solid State Institute, Technion, 32000 Haifa, Israel — [3]Université de Bordeaux, CNRS - CEA, CELIA, Talence, France — [4]Max-Born-Institut, 12489 Berlin, Germany

Phase retrieval of electronic wavefunctions generated by photoionization has been a longstanding challenge. Here we measure the time-reversed process of photoionization – photorecombination – in attosecond pulse generation. We demonstrate all-optical interferometry of two independent phase-locked attosecond light sources [1]. Our measurement enables us to directly determine the phase shift associated with electron scattering and with structural minima in atomic systems.

In a second study, we superimpose two attosecond light sources with perpendicular polarization, achieving direct time-domain polarization control [2]. We establish an extreme-ultraviolet lock-in detection scheme, allowing the isolation and amplification of weak chiral signals. We demonstrate our scheme by a phase-resolved measurement of magnetic circular dichroism.

[1] D. Azoury et al., Nature Photonics 13, 54 (2019).
[2] D. Azoury et al., Nature Photonics 13, 198 (2019).

A 11.2 Tue 14:30 f107

Attosecond pump-probe coincidence spectroscopy at 100 kHz — •Mikhail Osolodkov[1], Tobias Witting[1], Federico Furch[1], Felix Schell[1], Fabio Cavalcante[2], Carmen Menoni[2], Claus Peter Schulz[1], and Marc J.J. Vrakking[1] — [1]Max Born Institute, Berlin, Germany — [2]Department of Electrical and Computer Engineering, Colorado State University, Fort Collins, USA

Photoinduced coupled electron-nuclear dynamics in molecules typically proceeds on a femtosecond timescale which can be studied by attosecond pump-probe spectroscopy. Here we report on the setup of an extreme ultraviolet - near infrared (XUV-NIR) pump probe beamline operating at 100 kHz, which is combined with a reaction microscope, designed for high repetition rate operation [1]. The latter allows to measure the 3D momentum distributions of electrons and ions in coincidence and, thus, will enable detailed insights into photoinduced molecular processes. The beamline utilizes a high power noncollinear optical parametric chirped pulse amplification system (NOPCPA) operating at 100 kHz [2]. Both, XUV attosecond pulse trains (APTs), as well as isolated attosecond pulses (IAPs) can be generated through the high-order harmonic generation process (HHG) and delivered to the experiment together with synchronized NIR pulses. We present first results of pump-probe coincidence spectroscopy measurements, performed utilizing argon and molecular nitrogen as a target with XUV APTs and approximately 7 fs FWHM NIR pulses.

[1] Sascha Birkner, PhD thesis, Freien Universitat Berlin (2015)
[2] Federico J. Furch et al., Optics Letters 42, 2495-2498 (2017)

A 11.3 Tue 14:45 f107

Molecular Frame Studies of Channel-Resolved Laser-Driven Electron Recollision — Federico Branchi[1], Horst Rottke[1], Mark Mero[1], Marc J.J. Vrakking[1], Varun Makhija[2], and •Jochen Mikosch[1] — [1]Max-Born-Institut, Berlin, Germany — [2]University of Mary Washington, Fredericksburg, USA

When a molecule interacts with a strong, infrared laser field, a number of phase-locked attosecond processes can be initiated. From the perspective of transient probing of molecular structure, Laser-Induced Electron Diffraction (LIED) is of particular recent interest. In LIED, the tunnel-ionized electron wavepacket is accelerated and driven back to the parent molecule, where it rescatters elastically. With mid-infrared driving laser fields, where the achieved electron kinetic energies are high, bond lengths and angles of molecules can be extracted from the electron scattering images, by fitting the measured differential cross section with an independent atom model.

We are particularly interested in ionization channel-resolved studies, since LIED can be performed independently with two different continuum wavepackets, on the same molecule, at the same time. Such experiments are hence very powerful in testing the way in which structural information is retrieved from the data. Current measurements on 1,3-butadiene molecules are performed in a reaction microscope, which is coupled to a 100kHz repetition rate, mid-infrared OPCPA laser system. We will report on differences we found between the rescattering associated with ground and excited state ionization continuum and on extracting three-dimensional molecular frame information.

A 11.4 Tue 15:00 f107

Study of multiphoton transition in the continuum using RABBITT — •Divya Bharti, Farshad Shobeiry, Hemkumar Srinivas, Robert Moshammer, Thomas Pfeifer, and Anne Harth — Max Planck institut für Kernphysik, Heidelberg, Germany

Rapid advancement in the laser technology has made it possible to generate high power few-cycle Infrared (IR) pulses suitable for the generation of tabletop XUV radiation. These developments have made the study of attosecond physics accessible. Photoionization is a fundamental process in light-matter interaction. The photoelectrons released in this process carry information about the detection method as well as the electronic structure of the original atom.

We study single-photon ionization followed by multiphoton continuum-continuum (CC)-transitions in a RABBITT [1] like setup. RABBITT(Reconstruction of Attosecond beating by Interference of Two-Photon Transitions) is a widely used XUV-NIR pump-probe technique to study photoionization time delays. The presence of an IR (probe) field leads to the appearance of sidebands in the photoelectron spectra. These sidebands oscillate as the delay between XUV (pump) and IR field varies. We analyze the variation in the modulation of the sidebands with respect to e.g. the IR (probe) intensity or the probe frequency to analyze and study the effect of multiphoton transition. This technique will allow us to access the phases of CC-transition matrix elements [2].

[1] Muller, H. Appl Phys B (2002) 74
[2] Harth et al., Phys. Rev. A 99, 023410

A 11.5 Tue 15:15 f107

Static Coherent State Method: High-order Harmonic Generation in atomic and molecular systems in different gauges — •Mohammadreza Eidi and Jan-Michael Rost — Max Planck Institute for the Physics of Complex Systems - Dresden

We apply the static coherent states (SCS) method [1] to investigate high-order harmonic generation (HHG) in atomic and molecular systems, comparing results for different gauges. We present a case study of the Hydrogen molecular ion H2+ interacting with a few-cycle linearly polarized optical laser in full three-dimensional space. In the original implementation, coherent states are chosen randomly which requires a large number of them. Here, we employ a Gaussian optimization process technique to reduce the number of coherent states without sacrificing accuracy.

[1] Eidi, et.al. Applied Sciences, 8(8):1252, jul 2018.

A 11.6 Tue 15:30 f107

Imprinting orbital angular momentum onto a propagating matter wave — •Jonas Wätzel and Jamal Berakdar — Martin-Luther-Universität Halle-Wittenberg, Institut für Physik, Halle (Saale)

Recently, the transfer of orbital angular momentum (OAM) to a bound electron by absorbing photons of an optical vortex was proven experimentally [1]. In contrast, the vortex-matter interaction in the case of a propagating photoelectrons is debatable. Indeed, this amounts to transferring the phase information of a classical electromagnetic wave, defined within several hundreds of nanometers, to an ensemble of quantum particles with an extent of few angstroms. In this talk, I present experimental and numerical results of the two-color photoionization of an extended sample of He atoms. Surprisingly, we found that the carried OAM of the IR probe field can be transferred coherently to a photoelectron liberated by an XUV FEL laser pulse. Remarkably, the imprinted phase information survives the averaging over the ensemble of He atoms out to macroscopic distances, where the signal is observed. Our results point to the occurrence of non-dipolar transitions initiated by the IR vortex field, which are inaccessible by one-photon processes driven by conventional unstructured light waves. Based on

our predictions, new electron spectroscopy methods and qualitatively new analytical tools can be constructed.

[1] Schmiegelow, C. T. et al. "Transfer of optical orbital angular momentum to a bound electron" Nat. Commun. 7, 12998 (2016).

A 11.7 Tue 15:45 f107

Angular resolved photoemission of metal atoms embedded in helium nanodroplets in the MPI regime — •Bennet Krebs, Michael Zabel, Lev Kazak, and Josef Tiggesbäumker — Institut für Physik, Universität Rostock, Germany

Angular resolved photoelectron emission spectra of single metal atoms embedded in helium nanodroplets are measured, analyzed and compared to free atoms. A femtosecond laser system provides 110 fs, linear polarised laser pulses, which are used to ionize the atomic targets in the multiphoton regime (I$\approx 10^{13\cdots14}$W/cm^2) without ionizing the helium nanodroplet itself. Furthermore a time delay controlled two color setup with overlapping $2\omega/\omega$ (400 nm/800 nm) fields is used to probe the attosecond dynamics. For this we apply the highly sensitive Phase-of-the-Phase (PoP) method, which has been previously used to extract information about photoelectron trajectories. Compared to the anisotropic above-threshold-ionization (ATI) signals from free atoms a near isotropic emission is obtained for the embedded species. Furthermore, an enhancement of ATI signals and additional ATI orders can be observed. In the same vein we see a reduction of relative phase contrast. The impact of elastic scattering of the electrons with the surrounding helium environment will be discussed.