A 11: Attosecond Physics I

Zeit: Dienstag 14:00–16:00 Raum: VMP 6 HS-B

Fachvortrag A 11.1 Di 14:00 VMP 6 HS-B

Experimental molecular orbital tomography by attosecond electron wavepackets — •Stefan Haessler[1], Willem Boutu[1], Cecilia Giovanetti[2], Jeremie Caillat[2], Thierry Ruchon[1], Hamed Merdji[1], Pierre Breger[1], Bertrand Carre[1], Alfred Maquet[2], Richard Taieb[2], and Pascal Salieres[1] — [1]CEA-Saclay, IRAMIS, Service des Photons, Atomes et Molécules, 91191 Gif-sur-Yvette, France — [2]UPMC Université Paris 06, LCPMR, 11 rue Pierre et Marie Curie, 75231 Paris, France

The strong interaction of a molecule with a laser field frees by tunnel ionization an attosecond electron wavepacket that probes its bound state half a laser cycle later as it recollides with the core. The information is encoded in the attosecond XUV burst emitted during recombination. Complete characterization of this observable gives access to the transition dipole moment over a large momentum span. With suitable assumptions on the EWP, the retrieval of the highest occupied molecular orbital (HOMO) as a function of space and time becomes possible.

We measured amplitudes and phases of the recombination dipole as a function of alignment angle for N_2 and CO_2 molecules by characterizing their attosecond emission with the RABITT technique. With these data, we successfully perform tomographic reconstructions of the HOMOs with a reduced set of assumptions as compared to the first demonstration by Itatani et al. Potentials and limitations of this technique will be discussed based on the deviations of the experimentally determined orbitals from theoretical ones.

Fachvortrag A 11.2 Di 14:30 VMP 6 HS-B

Multi-orbital effects for high harmonic generation in N_2 — •Markus Gühr, Brian K. McFarland, Joseph P. Farrell, and Philip H. Bucksbaum — Stanford PULSE Institute, SLAC National Accelerator Lab, Menlo Park, CA 94025 and Physics Department, Stanford University, Stanford CA 94305, USA

High harmonic generation (HHG) proceeds in three steps. First, a part of the electron wave function tunnels out of the valence orbital. The liberated electron wave packet accelerates in the laser field and finally coherently recombines with the initially ionized orbital. This leads to emission of harmonics of the driving laser frequency. The harmonic spectrum contains information about the valence orbital [1]. For molecules, the highest occupied molecular orbital (HOMO) is generally thought to be responsible for ionization and recombination during HHG. Molecular electronic states energetically below the HOMO should contribute to HHG, but this behavior has not been observed previously. Utilizing molecular alignment, we obtain experimental evidence that the more deeply bound HOMO-1 with its π_u symmetry also contributes to HHG in N_2 [2]. Semi-classical simulations of the recombination process to the HOMO and HOMO-1 support our observation. This opens the route to imaging coherent superpositions of electronic orbitals.

[1] J. Itatani et al., Nature **432**, 867 (2004)
[2] B. K. McFarland, J. P. Farrell, P. H. Bucksbaum and M. Gühr, Science **322**, 1232 (2008)

A 11.3 Di 15:00 VMP 6 HS-B

Strong field Cooper minimum in High Harmonic Generation — •Markus Gühr, Joseph P. Farrell, Brian K. McFarland, and Philip H. Bucksbaum — Stanford PULSE Institute, SLAC National Accelerator Lab, Menlo Park, CA 94025 and Physics Department, Stanford University, Stanford CA 94305, USA

The so called Cooper minimum is a prominent feature in the VUV photoionization cross section. It results from a cancellation of the transition matrix element from the atomic ground state to the continuum states at certain photon energies and is accompanied by a π phase jump in the matrix element [1]. We observe the Cooper minimum in high harmonic generation (HHG) spectrum of argon. In addition to the spectral amplitude modulation we also observe a spectral phase jump of π around the Cooper minimum. The recombination step of HHG can be described by the inverse photoionization matrix element and we apply the photoionization theory to simulate our data. In the experiment, we observe a shift of the HHG Cooper minimum with respect to the photoionization Cooper minimum of about 5 eV. We explain this shift by a strong-field mixing of the continuum states during the recombination step in HHG. The results show the opportunity for controlled attosecond pulse shaping.

[1] J. W. Cooper, Physical Review **128**, 681 (1962)

A 11.4 Di 15:15 VMP 6 HS-B

Monitoring nuclear and electronic dynamics using high-order harmonic radiation — •Stefanie Gräfe[1], Daniil V. Kartashov[2], and Joachim Burgdörfer[1] — [1]Institut für Theoretische Physik, TU Wien, Österreich — [2]Institut für Photonik, TU Wien, Österreich

We investigate the possibility to simultaneously monitor nuclear and electronic dynamics in diatomic molecules using HHG. Applying a simple two-dimensional model analogously to the hydrogen molecular ion with one nuclear and one electronic coordinate, we demonstrate numerically the possibility to use high-order harmonic spectra as a dynamical structural probe, identifying the traces of nuclear and electronic dynamics in the HHG spectra.

The harmonic spectra contain rich information, e.g. peak broadening due to nuclear motion, the prominent feature of two-center interference minima showing directly the temporal change in the internuclear distance [Lein et al., Phys. Rev. A, 2002]. We can distinguish the contributions of different electronic states leading to additional interferences in high-harmonics radiation. The occurrence of interference structures is a result of inversion symmetry breaking due to localization dynamics; the electron being ionized from one state (or one potential well) may recombine to several other coherently populated states residing in both potential wells. As this process is only efficient in a region where the nuclear autocorrelation function is substantially different from zero, the information on the internuclear coordinate maps onto cross terms in a defined spectral energy region. Support by FWF-SFB ADLIS and the Lise-Meitner program, M1077-N16, is highly acknowledged.

A 11.5 Di 15:30 VMP 6 HS-B

Molecular orbital tomography using HHG and ATI — •Elmar van der Zwan, Ciprian Chirila, and Manfred Lein — Institute for Physics, University of Kassel, Germany

In 2004 a revolutionary method to perform tomographic imaging of molecular orbitals using high-harmonic generation (HHG) caught a lot of attention [1]. The method is based on the simplification that the returning electron in the three-step model can be modeled as a plane wave. We have shown that orbitals of arbitrary symmetry can be reconstructed if one uses extremely short laser pulses that ensure the continuum wave packet recombines from one side only [2]. Within the single-active-electron approximation, the challenge of the scheme lies in the accurate determination of the continuum wave packet. For a known orbital, an expression for the continuum wave packet in terms of classical trajectories is derived from the Lewenstein model. We propose an experimentally feasible method to determine the composition of the continuum wave packet using both HHG and angularly resolved above-threshold ionization (ATI) electrons. By incorporating the ATI electrons in the procedure, the instantaneous tunneling rates for electron emission parallel to the applied field can be measured. This avoids one of the major assumptions in the tomographic scheme, namely approximating the tunnel ionization of the molecule by that of the reference.

[1] Itatani *et al.* Nature **432**, 867-871 (2004)
[2] van der Zwan *et al.* Phys. Rev. A **78**, 033410 (2008)

A 11.6 Di 15:45 VMP 6 HS-B

Probing shake-up states in He by laser controlled rescattering of XUV photoelectrons — •Renate Pazourek[1], Johannes Feist[1], Stefan Nagele[1], Emil Persson[1], Lee A Collins[2], Barry I Schneider[3], and Joachim Burgdörfer[1] — [1]Inst. for Theor. Physics, Vienna University of Technology, Austria — [2]Theor. Division, Los Alamos National Laboratory, USA — [3]Physics Division, NSF, USA

Recent progress in the generation of ultrashort XUV pulses and few-cycle infrared pulses opens the possibility for various novel pump-probe setups for the study of ultrafast electronic dynamics. We present numerical ab initio simulations where we employ an XUV pulse to singly ionize a helium atom in the presence of an IR field which can induce rescattering of the ionized electron at the parent ion.

For XUV pulses with photon energies slightly above the first ionization potential ($I_1 \approx 24.6\,\mathrm{eV}$) one electron is ionized and the remaining

ion relaxes to the He^+ ground state. For photon energies $\hbar\omega > 65.4\,\mathrm{eV}$ a second channel, where the bound electron is excited into a shake-up state, opens. In this contribution we present velocity maps, i. e. angularly resolved momentum distributions of the singly ionized electrons, experimentally accessible by a velocity map imaging spectrometer. Insights into two-electron effects, a sine qua non for the shake-up process to occur, can be obtained. We compare our ab initio simulations (without approximations for inter-electron interactions) with single-active electron calculations in order to show to what extent electron-electron interactions account for the observed effects.