A 3: Attosecond pulses and high harmonics (jointly with K and Q)

Zeit: Montag 16:30–18:15 Raum: 3C

Hauptvortrag A 3.1 Mo 16:30 3C

Coherent Control with Shaped Attosecond Soft-X-Rays: Techniques and Application — •Thomas Pfeifer — Departments of Chemistry and Physics, University of California & Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

Recent progress in ultrafast laser technology enables the generation of soft-x-ray pulses down to $\sim$ 100 attoseconds in duration, thus allowing access to the unexplored realm of electron dynamics. On the other hand, coherent control of matter with shaped laser fields has reached its maturity with regard to controlling the *relative motion of atoms* (vibrations/phonons, phase transitions, molecular reactions and rotation). However, due to the lack of laser pulse shaping techniques in the soft-x-ray spectral region, coherent control has so far had only limited capability of controlling the *electronic wavefunction* directly, which is of fundamental importance to physics (multi-electronic correlation) and chemistry (bonding dynamics). In this talk, it will be shown how Coherent Control can be transferred to, and combined with Attosecond Science towards the goal of gaining comprehensive mastery of matter on the quantum scale. Experimental results, theoretical concepts, and simulations demonstrate the feasibility of using a) multicolor laser fields, b) phase-shaped laser pulses, and c) medium control in high-harmonic generation to enable shaping of pulses and pulse trains in the attosecond soft-x-ray domain. Also, the first experimental application of shaped coherent soft-x-rays towards the optimal control of electronic quantum processes (dissociative photoionization of SF_6) will be presented.

A 3.2 Mo 17:00 3C

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

Recently a method to perform tomographic imaging of molecular orbitals using high-harmonic generation has been proposed [1]. The method is based on the simplification that the returning electron in the three-step model can be modeled as a plane wave. 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. We compare two different forms for the reconstruction, and introduce an error-reduction algorithm that can be used to optimize the results. One of the challenges of the scheme lies in the accurate determination of the continuum wave packet. We determine the continuum wave packet in the Lewenstein model, assuming that the molecular orbital is known, and compare this with various methods to determine the continuum wave packet without knowledge of the orbital.

[1] J. Itatani, J. Levesque, D. Zeidler, H. Niikura, H. Pépin, J.C. Kieffer, P.B. Corkum and D.M. Villeneuve. *Tomographic imaging of molecular orbitals.* Nature 432, 867-871 (2004)

A 3.3 Mo 17:15 3C

Dressing and high-order harmonic generation in small molecules — •Ciprian Chirila and Manfred Lein — Universität Kassel, Institute of Physics, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

The strong-field approximation was recently extended to take into account the effect of vibrational motion and laser-induced coupling of the Born-Oppenheimer states on high-harmonic generation in molecules. We present detailed calculations of the harmonic spectra in H_2 and D_2 for long laser wavelengths (2000 nm), comparing the effects of the dressing to the case of 800 nm. The main effect of dressing is an overall reduction of the harmonic generation and, at long wavelengths, a non-negligible change in the ratio of harmonic signals from different isotopes.

A 3.4 Mo 17:30 3C

Extended Strong-Field Approximation including Collectivity — •Michael Ruggenthaler and Dieter Bauer — Max-Planck-Institut für Kernphysik, Heidelberg

High-order harmonic generation may often be treated within a single active electron picture using the so-called strong-field approximation (SFA) [1] to propagate the initial wave-function. However, if collective phenomena are to be included the standard SFA treatment will not suffice. Although the SFA may be extended via an extra term in the Hamiltonian accounting for the collective behavior of the multi-electron system, there is no straightforward definition of this term.

The problem can be reformulated in time-dependent density-functional theory [2,3] where the so-called Kohn-Sham orbitals are propagated such that total single-particle density is the same as those of the interacting system. The linear response of the density to the external potential gives rise to a time-dependent Hartree-exchange-correlation potential which then can be used within the framework of the extended SFA.

[1] W. Becker et al, Phys. Rev. A 56, 645 (1996)
[2] E. Runge and E. K. U. Gross, Phys. Rev. Lett. 52, 997 (1984)
[3] M. A. L. Marques et al, Time Dependent Density Functional Theory, Lect. Notes Phys. 706 (Springer, Heidelberg, 2006)

A 3.5 Mo 17:45 3C

Formation of Amplitude and Phase during High Harmonic Generation — •Markus Gühr, Brian K. McFarland, Joe P. Farrell, and Philip H. Bucksbaum — Stanford PULSE Center, Stanford University and SLAC, California, USA

High Harmonics of a laser field are generated during the interaction of an intense laser pulse with an atomic or molecular gas. The amplitude and phase of the harmonics contain information about the generation process and the symmetry of the electronic wave functions of the involved atoms or molecules [1].

We measure the amplitude of high harmonics generated in N_2 and Ar. Furthermore, we obtain the relative phase between the harmonics from N_2 and Ar by interferometric measurements on mixtures of the two gases. We observe phase jumps at the 33rd and the 25th harmonic. The first is accompanied by an amplitude minimum in Ar and attributed to a Cooper minimum. The second is accompanied by an amplitude minimum and a linewidth broadening in N_2. It results from the symmetry of the N_2 highest occupied molecular orbital.

The discussed phenomena have important implications for the amplitude and phase of attosecond pulses generated via high harmonic generation.

[1] M. Gühr, B. K. McFarland, J. P. Farrell and P. H. Bucksbaum, J. Phys. B: At. Mol. Opt. Phys., 40, 3745-3755 (2007)

A 3.6 Mo 18:00 3C

Enhancement of high-order harmonic generation by rare gas mixtures — •Mirko Prijatelj, Tobias Vockerodt, Daniel Steingrube, Uwe Morgner, and Milutin Kovacev — Institut für Quantenoptik, Leibniz Universität Hannover

We study the enhancement of high-order harmonic generation (HHG) by rare gas mixtures. Our experiment confirms recent results mixing He and Xe atoms. The harmonics from Xe atoms enhance the observed yield from He atoms by about two orders of magnitude. Moreover the cut-off position is extended compared to the spectrum of pure He atoms. We report on the experimental parameter sensitivity of the enhancement process and show first results which indicate that the atomic state structure is an important prerequisite. Our investigation extends as well towards experimental conditions suited for low-energy pump pulses as for example mode-locked Ti:sapphire femtosecond oscillator pulses with MHz repetition rates. These conditions are interesting for generating harmonics either intracavity or directly from a femtosecond oscillator. This experimental approach promises to lead to a joint frontier of precision spectroscopy and ultrafast science by extending frequency comb technology into the XUV spectral region.