A 28: Attosecond physics II

Time: Wednesday 10:30–12:45 Location: BAR 205

Invited Talk A 28.1 Wed 10:30 BAR 205

Quantum Interference Control of Free and Bound Electrons in Atoms and Molecules — •Thomas Pfeifer — Max-Planck Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg

Electrons are the fundamental building blocks of molecular and condensed-phase bonds and the motion of electrons (migration, localization, ionization, etc.) governs the course of chemical reactions and defines molecular potential-energy landscapes. In the valence shell of atoms and molecules, electron dynamics typically proceeds on few-femtosecond and faster time scales, such that extremely short pulses and controlled light electric fields are needed to measure and to control these scientifically relevant quantum-electronic processes.

In this talk, we shall discuss experiments and the physical description of electronic quantum control. It will be shown that by using very short light pulses with a stabilized carrier-envelope phase (CEP) – and thus a fully controlled temporal electric field evolution – it is possible to steer the process of ionization and electron localization in atoms and molecules.

Most importantly, we can identify the mechanisms of this type of control as fundamental scenarios of quantum control among energetically degenerate states. As theoretically established by Brumer and Shapiro a long time ago, quantum interference among different light-induced transition pathways in atoms and molecules is at the heart of electronic control experiments. The intuitive physical pictures arising from this traditional understanding of electronic control in combination with the possibilities opened up by modern CEP-stabilized laser light electric fields and attosecond pulses paves the way towards more comprehensive applications of electronic control in more complex systems.

A 28.2 Wed 11:00 BAR 205

Decoherence in Attosecond Photoionization — •Stefan Pabst[1,2], Loren Greenman[3], Phay J. Ho[4], David A. Mazziotti[4], and Robin Santra[1,2] — [1]Center for Free-Electron Laser Science, DESY, Notkestrasse 85, 22607 Hamburg, Germany — [2]Department of Physics,University of Hamburg, Jungiusstrasse 9, 20355 Hamburg, Germany — [3]Department of Chemistry and The James Franck Institute, The University of Chicago, Chicago, Illinois 60637, USA — [4]Argonne National Laboratory, Argonne, Illinois 60439, USA

The creation of superpositions of hole states via single-photon ionization using attosecond extreme-ultraviolet pulses is studied with the time-dependent configuration interaction singles (TDCIS) method. Specifically, the degree of coherence between hole states in atomic xenon is investigated. We find that interchannel coupling not only affects the hole populations, it also enhances the entanglement between the photoelectron and the remaining ion, thereby reducing the coherence within the ion. As a consequence, even if the spectral bandwidth of the ionizing pulse exceeds the energy splittings among the hole states involved, perfectly coherent hole wave packets cannot be formed. For sufficiently large spectral bandwidth, the coherence can only be increased by increasing the mean photon energy.

A 28.3 Wed 11:15 BAR 205

Time-resolved phase matching and macroscopic gating in few-cycle high-harmonic generation: Simulation and experimental results — •Michael Schönwald, Christian Ott, Philipp Raith, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, Heidelberg, Deutschland

High-harmonic generation (HHG) is a key technology for the direct exploration of atomic and molecular electron dynamics. This process is based on a macroscopically coherent response of a medium, i.e. the radiation produced by multiple atoms has to be phase matched. A time-domain simulation of phase matching will be presented, showing how phase matching can act as a temporal gate and thus allow HHG only during the leading edge of the driving laser pulse. The behaviour of the leading-edge gating mechanism will be analysed for different experimental conditions, such as peak intensity of the driving laser pulse, density of the gaseous generation medium and the distance between focus and generation medium. In our experiments, the leading-edge gate is observed by carrier-envelope phase (CEP) dependent measurements of HHG, including analysis of the half-cycle cut-off (HCO) emission. Experiments are carried out for various pressures and compared to the simulation. The HCO photon energy allows to extract the field strength of an individual half cycle of the driving laser pulse. In addition, the pressure-dependent total harmonic yield measured in the experiment is compared to the simulation for the leading-edge regime and yields a qualitative agreement, further confirming our understanding of the process that allows to generate isolated attosecond pulses.

A 28.4 Wed 11:30 BAR 205

Spectral reshaping of pulses propagating within a filament — •Emilia Schulz[1], Daniel S. Steingrube[1], Thomas Binhammer[2], Mette B. Gaarde[3], Arnaud Couairon[4], Uwe Morgner[1], and Milutin Kovacev[1] — [1]Leibniz Universität Hannover, Welfengarten 1 D-30167 Hannover, Germany; QUEST, Centre for Quantum Engineering and Space-Time Research, Hannover, Germany — [2]VENTEON Laser Technologies GmbH, D-30827 Garbsen, Germany — [3]Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA; PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA — [4]Centre de Physique Theorique, Ecole Polytechnique, CNRS, F-91128, Palaiseau, France

Filamentation of ultra-short laser pulses has become a versatile tool for ultra-fast applications as e.g. few-cycle-pulse generation. The complex spatio-temporal dynamics taking place during the filamentation process are so far only predicted from numerical simulations, and only a few efforts have been done in the experimental investigation of the evolution of propagating pulses within a filament. We present a cell design which is capable to realize a pressure gradient from 1000 mbar argon to a vacuum pressure of below 10^{-4} mbar over a distance of about 1 cm. This enables the investigation of filaments along their propagation direction. Self focusing and spectral broadening of the pulse during the filamentation process can now be investigated in detail. We present experimental results showing the influence of pressure and chirp on the spectral broadening. Numerical calculations show excellent agreement.

A 28.5 Wed 11:45 BAR 205

Spectrally resolved Maker Fringes in High-Order Harmonic Generation — •Christoph M. Heyl[1], Jens Güdde[2], Ulrich Höfer[2], and Anne L'Huillier[1] — [1]Department of Physics, Lund University, Lund, Sweden — [2]Fachbereich Physik und Zentrum für-Materialwissenschaften, Philipps-Universität, Marburg, Germany

We present theoretical and experimental studies of macroscopic interference effects in high-order harmonic generation (HHG) using 6 μJ, 45 fs laser pulses at 100 kHz repetition rate.

Every harmonic spectrum contains a fingerprint of both, microscopic and macroscopic effects. So far, however, mostly single atom effects have been emphasized. In a series of recent studies interference structures in HHG spectra have been interpreted as due to interferences between long and short electron trajectories which has been called quantum path interference (QPI). Here, we focus on the macroscopic effects and investigate how phase matching affects the harmonic spectrum. We show how the appearance of spectral fringes can be explained in terms of Maker fringes, due to the time dependent coherence length of the long trajectory. We explain further, how these spectral Maker fringes contain direct information about the time dependent phase matching conditions as well as about the intensity dependent phase of the microscopic dipole moment.

A 28.6 Wed 12:00 BAR 205

High-order harmonic generation by intensity spikes within filamention — •Daniel S. Steingrube[1], Emilia Schulz[1], Thomas Binhammer[2], Mette B. Gaarde[3], Arnaud Couairon[4], Uwe Morgner[1], and Milutin Kovacev[1] — [1]Leibniz Universität Hannover, Institut für Quantenoptik, Welfengarten 1, D-30167 Hannover, Germany; QUEST, Centre for Quantum Engineering and Space-Time Research, Hannover, Germany — [2]VENTEON Laser Technologies GmbH, D-30827 Garbsen, Germany — [3]Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA; PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA — [4]Centre de Physique Theorique, Ecole Polytechnique, CNRS, F-91128, Palaiseau, France

Few-cycle pulse generation using filamentation for pulse compression has been shown in a number of recent studies. These short and intense pulses can be applied for high-order harmonic generation (HHG) which is a prerequisite to the production of attosecond pulses.

We realized an experimental setup to combine both steps of pulse compression and HHG in a semi-infinite gas cell at atmospheric pressure where the critical power for filamentation is reached. The generation of high-order harmonics directly in the filament shows the occurrence of intensity spikes above the clamping intensity of filamentation. Our results confirm recent theoretical predictions and show a continuous harmonic spectrum in the XUV-spectral range promising for single attosecond pulse generation directly from a filament.

A 28.7 Wed 12:15 BAR 205

Attosecond Twin-Pulse Control by Generalized Kinetic Heterodyne Mixing — •Philipp Raith, Christian Ott, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany

Attosecond double pulses with defined spectral properties (e.g. spectral-phase difference between the pulses) are a useful tool for spectral interferometry measurements of attosecond quantum dynamics in atoms and molecules. For this purpose, we study the generation and manipulation of attosecond double-pulse (called twin pulse at identical intensities of the two pulses) production in high-order harmonic generation by a combination of two-color laser fields and carrier-envelope-phase (CEP) control methods. It is shown that both relative amplitude and phase of the double pulses can be independently set by making use of multidimensional parameter control methods. Two technical implementation routes are discussed: kinetic heterodyne mixing of a strong laser field and its second harmonic and split-spectrum phase-step control using a single spectrally coherent broad-band laser field. This multidimensional control scheme including the CEP can be considered a general approach for controlling electron dynamics for applications ranging from electron localization in molecules to attosecond pulse shaping.

A 28.8 Wed 12:30 BAR 205

Attosecond neutron scatterig from H2 and D2 — •C. Aris Dreismann[1], Evan Gray[2], and Tom Blach[2] — [1]Institute of Chemistry, TU Berlin, Germany — [2]Griffith University, School of Biomolecular and Physical Sciences, Brisbane, Australia

Quantum entanglement (QE) has emerged as the most emblematic feature of quantum mechanics. Nuclei and electrons in condensed matter and/or molecules are usually entangled, due to the prevailing interactions. However the "environment" of a microscopic system (e.g. a proton in a H2 molecule) may cause an ultrafast decoherence thus making atomic and/or nuclear QE effects not directly accessible to experiments. Applying neutron Compton scattering (NCS) in the energy transfer range of ca. 1-100 eV, the relevant proton-neutron scattering time lies in the sub-femtosecond time range. Results of current NCS experiments from H2 and D2 in the gas phase at T = 40 K are reported, showing that the measured Compton profiles (in the attosecond scattering range) reveal new features of quantum dynamics which contradict conventional theoretical expectations. The experimental NCS setup of ISIS, Rutherford Appleton Laboratory is shortly introduced. The theoretical frame of "attosecond scattering from open quantum systems" [1] is discussed, in connection with the new experimental findings.

[1] C. A. Chatzidimitriou-Dreismann and M. Krzystyniak, Laser Physics **20**, 990 (2010).