A 6: Time-resolved spectroscopy

Time: Monday 17:00–19:00 Location: HS 20

Invited Talk A 6.1 Mon 17:00 HS 20

Phase-modulated harmonic light spectroscopy — •Lukas Bruder, Ulrich Bangert, Marcel Binz, Andreas Wituschek, Marcel Mudrich, and Frank Stienkemeier — Physikalisches Institut, Universität Freiburg

Coherent time-resolved spectroscopy is a powerful tool to study ultrafast dynamics in complex systems. It is highly desirable to extend these techniques to the XUV spectral range. However, demands on phase stability increase significantly when going to short wavelengths and advanced pulse manipulation in the XUV is challenging. We suggest an approach based on acousto-optical phase modulation of the driving/seed laser combined with harmonic lock-in detection. In this approach, demands on phase stability are drastically reduced and signals are efficiently isolated and amplified. We demonstrate this concept in a proof-of-principle study with second harmonic generation. The concept is characterized in a femtosecond pump-probe experiment measuring electronic wave packet dynamics in atomic systems. Our results show promise for an implementation in HHG and HGHG light sources.

Invited Talk A 6.2 Mon 17:30 HS 20

Amplitude and phase control of an atom's optical response — •Alexander Blättermann, Andreas Kaldun, Veit Stooss, Thomas Ding, Christian Ott, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

The optical response of an atom to ultrashort bursts of light gives rise to a characteristic fingerprint of its electronic system - the atomic absorption spectrum. Originating from the superposition of quantum states, we are able to control the optical response by acting on the very same quantum states with an intense laser pulse. Varying the timing and intensity of this laser pulse allows us to modify the amplitude and phase of the optical response anytime during the decay of the excited state. These degrees of freedom enable a variety of experimental schemes, such as a static pulse configuration with tunable intensity up to a dynamic configuration with tunable delay. While the former scheme granted control over the Fano-q parameter of a spectral line [1], the latter recently enabled us to observe the formation of a Fano resonance in time [2]. A third scheme even allows for an in situ characterization of the NIR pulse providing helpful information for data analysis [3]. Our experiments are supported by an analytic model of the laser-controlled dipole moment [4], which establishes a direct link between time and frequency domain.

[1] Science 340, 716 (2013)
[2] Science 354, 738 (2016)
[3] Opt. Lett. 40, 3464 (2015)
[4] J. Phys. B: At. Mol. Opt. Phys. 47, 124008 (2014)

A 6.3 Mon 18:00 HS 20

Using strong-field spectroscopy to observe the ultrafast buildup of a Fano resonance — •Veit Stooss[1], Alexander Blättermann[1], Andreas Kaldun[1], Stefan Donsa[2], Hui Wei[3], Renate Pazourek[2], Stefan Nagele[2], Christian Ott[1], Chii-Dong Lin[3], Joachim Burgdörfer[2], and Thomas Pfeifer[1] — [1]Max-Planck-Institut für Kernphysik, Heidelberg, Germany, EU — [2]Institute for Theoretical Physics, Vienna University of Technology, Vienna, Austria, EU — [3]Department of Physics, Kansas State University, Manhattan, USA

High-harmonic generation and the resulting attosecond light pulses in the extreme ultraviolet (XUV) spectral range enables access to the dynamics of electrons in atomic and molecular systems. The method of strong-field spectroscopy uses these pulses to excite bound electronic states, which are subsequently perturbed in their natural evolution with a high-intensity ultrashort near-infrared (NIR) pulse. By observing the thus modified XUV absorption spectrum one gains insights into such strong-field-driven dynamics. Here, we use this method to create a controllable temporal gate between excitation and termination of a bound-bound transition to experimentally observe the emergence of an asymmetric Fano absorption resonance in helium [1]. The results shown are in good agreement with *ab-initio* calculations as well as analytical models describing the buildup of a Fano resonance.

[1] Observing the ultrafast buildup of a Fano resonance in the time domain - Science, Vol. 354, Issue 6313, pp. 738-741 (2016)

A 6.4 Mon 18:15 HS 20

Laser-induced grating spectroscopy with absolute time information — •Jan Reislöhner, Christoph G. Leithold, and Adrian N. Pfeiffer — Institute of Optics and Quantum Electronics, Abbe Center of Photonics, Friedrich Schiller University, Max-Wien-Platz 1, 07743 Jena

The nonlinear response function of transparent dielectrics is studied in time and spectral domain. A laser-induced grating is generated in a thin sample with short laser pulses (~5 fs, 700 nm) and imaged with a 2f-2f imaging system. The individual beams (the two fundamental beams and two orders of self-diffraction) are blocked consecutively in the imaging system while varying the pulse delay. From the absolute phase of the interference gratings, the delay between the fundamental pulses and the nonlinear optical response can be determined with subcycle precision. However, the optical path difference in the optical system is usually not known and inhibits the measurement of the absolute grating phase.

In a first step, the information contained in relative phase measurements (intensity dependence, material dependence) is examined. In a second step, a method is discussed for retrieving the absolute grating phase by stepwise translation and rotation of each element of the optical system.

A 6.5 Mon 18:30 HS 20

Two-electron dynamics in Helium driven by intense XUV radiation — •Lennart Aufleger[1], Thomas Ding[1], Marc Rebholz[1], Maximilian Hartmann[1], Alexander Magunia[1], David Wachs[1], Veit Stooss[1], Paul Birk[1], Gergana Borisova[1], Andrew Attar[2], Thomas Gaumnitz[3], Zhi Heng Loh[4], Stefan Düsterer[5], Christian Ott[1], and Thomas Pfeifer[1] — [1]MPI für Kernphysik, Heidelberg, Germany — [2]UC Berkeley, Berkeley, USA — [3]ETHZ, Zürich, Switzerland — [4]NTU Singapore, Singapore — [5]DESY, Hamburg, Germany

The measurement of laser-induced strong-field dynamics in few-electron systems provides a way to investigate their correlated nature. Using a lab-based HHG source we have studied such correlations that are imprinted on the XUV absorption line shape and investigated them by modification with strong fields in the near infrared and visible (VIS) spectral region. [C. Ott et al., Science 340, 716 (2013)]. Here, we present an extension of the strong-field modification scheme to the XUV-only spectral region. Using intense and partially coherent light of a FEL source (FLASH@DESY), the transition into the doubly-excited state 2s2p in Helium at 60.15 eV was driven. This excitation scenario represents a most basic two level system with a cooperative excitation of two electrons. The variation of pulse intensities between 10^{12} W/cm^2 and 10^{14} W/cm^2 induces a change in the experimentally observed XUV absorption line shape. A transformation from a Fano to a Lorentzian line shape is also confirmed by few-level simulations that were carried out.

A 6.6 Mon 18:45 HS 20

Time-Dependent Strong-Field Effects in Argon and Nitrogen — •Paul Birk, Veit Stooss, Maximilian Hartmann, Alexander Blättermann, Kristina Meyer, Christan Ott, and Thomas Pfeifer — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Extreme ultraviolet (XUV) light pulses in the attosecond regime produced by high-harmonic generation of near-infrared (NIR) light are powerful tools to investigate intra-atomic and -molecular processes at ultra-high time resolution. With the help of an all-optical approach we gain access to bound-state dynamics and correlations between electronic states in atoms and molecules. In previous work we have investigated the laser-induced phase shifts and couplings of excited states in helium by measuring its perturbed polarization decay [1-2]. Based on this work on a two-electron system, we here study and compare time-dependent strong-field effects in argon and molecular nitrogen. Electronic and vibrational states of the target gases are excited by a weak XUV pulse and subsequently perturbed with a strong NIR pulse. We present first results and interpretations of XUV absorption spectra and their changes depending on both the time delay and intensity of the strong NIR pulse.

[1] Blättermann et. al., *J. Phys. B*, **47**, 124008 (2014)
[2] Blättermann et. al., *Optics Letters*, **40**, 3464 (2015)