

QUTIF

**Quantum Dynamics in Tailored
Intense Fields**

International Conference

4th September- 7th September 2017

Venue:
Physikzentrum Bad Honnef
Hauptstraße 5
53604 Bad Honnef



Monday, 4th September 2017

08:45 – 09:00	Welcome
09:00 – 09:35	Marc Vrakking <i>The many benefits of high-harmonic radiation</i>
09:35 – 10:00	Matthias Wollenhaupt <i>Three-dimensional control of photoelectron distributions bichromatic polarization-tailored laser pulses</i>
10:00 – 10:25	Ulf Saalman <i>The non-adiabatic channel in ultra-short high-frequency laser pulses</i>
10:25 – 10:50	Coffee Break
10:50 – 11:25	Jian Wu <i>Ultrafast photoelectron emission clocked by orthogonally polarized two-color laser fields</i>
11:25 – 11:50	Sebastian Eckart <i>Strong-field ionization with counterrotating circularly polarized two-color laser fields</i>
11:50 – 12:15	Nicolas Eicke <i>Trajectory-free ionization times in strong field ionization</i>
12:30 – 14:00	Lunch
14:00 – 14:35	André Bandrauk <i>High order harmonic generation with ultrashort bichromatic circularly polarized pulses is shown to occur efficiently in molecules by recollision</i>
14:35 – 15:00	Azra Gazibegović - Busuladžić <i>Strong-field ionization of molecules by bicircular laser fields</i>

15:00 – 15:25	Alvaro Jiménez-Galán <i>Attosecond recorder of the polarizations state of light</i>
15:25 – 16:00	Coffee Break
16:00 – 16:35	Oren Cohen <i>Selection Rules in High Harmonic Generation</i>
16:35 – 17:00	Nickolai Zhavoronkov <i>Control of attosecond light polarization in two-color bi-circular field</i>
17:00 – 17:25	Yinyu Zhang <i>Single-shot, real-time, carrier-envelope phase measurement based on stereographic-above-threshold ionization at short-wave infrared wavelength</i>
18:30 – 19:45	Dinner
19:45	Poster Session

Invited talks: 30 + 5 minutes

Other talks: 20 + 5 minutes

Tuesday, 5th September 2017

09:00 – 09:35 Alfred Leitenstorfer
Subcycle quantum physics of light and matter

09:35 – 10:00 Claus Ropers
Coherent Control and Quantum State Reconstruction of Free-Electron Beams in Ultrafast Electron Microscopy

10:00 – 10:25 Tristan Müller
Towards ultra long-range ab-initio calculations

10:25 – 10:50 Coffee Break

10:50 – 11:25 Mette Gaarde
Theory of high harmonic generation in solids: band structure, orientation dependence and emission time

11:25 – 12:00 Johan Mauritsson
Opto-optical phase modulation of extreme ultraviolet light pulses

12:00 – 12:15 Young Scientists Meeting

12:30 – 13:30 Lunch

13:30 – 19:00 Excursion/ Free time

19:00 Conference Dinner

Invited talks: 30 + 5 minutes

Other talks: 20 + 5 minutes

Wednesday, 6th September 2017

09:00 – 09:35 Péter Dombi
Nanoplasmonic near-field probing with ultrafast photoemission

09:35 – 10:00 Jonas Wätzel
Tunable high harmonic pulses from nanorings swirled by optical vortices

10:00 – 10:25 Arohi Jain
Attosecond-streaking spectroscopy on a liquid-water microjet

10:25 – 10:50 Coffee Break

10:50 – 11:25 Eleftherios Goulielmakis
Exploring the ultrafast frontiers of condensed phase physics with synthesized light fields

11:25 – 11:50 Matthias Kling
Attosecond physics at the nanoscale

11:50 – 12:15 Arne Senftleben
Intermediate state dependence of femtosecond photoelectron circular dichroism

12:30 – 13:30 Lunch

14:00 – 14:35 Pascal Salières
Attosecond photoionization dynamics close to Fano resonances

14:35 – 15:00 Jochen Küpper
Imaging molecules through strong-field-ionization and electron rescattering

15:00 – 15:25 Victor Despré
Ultrafast non-adiabatic relaxation of the naphthalene molecule after inner-valence ionization by a short XUV pulse

Thursday, 7th September 2017

15:25 – 16:00 Coffee Break

16:00 – 16:25 Stefanie Gräfe
Circular dichroism in the angular distribution of strong-field ionization of asymmetric triatomic model molecules

16:25 – 16:50 Ihar Babushkin
Signatures of electron ionization dynamics by ionization

16:50 – 17:15 David Ayuso
Chiral high-harmonic spectroscopy

18:30 – 19:45 Dinner

19:45 Poster Session

Invited talks: 30 + 5 minutes

Other talks: 20 + 5 minutes

09:00 – 09:35 Lukas Gallmann
Attosecond inter- and intra-band dynamics in bulk solids

09:35 – 10:00 Oliver Mücke
Crystal-Symmetry-Controlled Polarization Effects in High-Harmonic Generation in Solids

10:00 – 10:25 Maksim Kunitski
Rotating rotationless: nonadiabatic alignment of the helium dimer

10:25 – 10:50 Coffee Break

10:50 – 11:25 Toru Morishita
Near-forward rescattering photoelectron holography in strong-field ionization: extraction of the phase of the scattering amplitude

11:25 – 11:50 Alejandro Saenz
Molecular hydrogen in ultrashort intense laser fields

11:50 – 12:15 Florian Oppermann
The helium hydride molecular ion (HeH^+) in strong laser fields

12:15 – 12:30 Conclusion

12:30 – 13:30 Lunch

14:00 Departure

Abstracts

The many benefits of high-harmonic radiation

Marc Vrakking

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Presently, the emergence of novel extreme ultra-violet (XUV)/X-ray sources such as sources based on High-Harmonic Generation (HHG) and free electron lasers like FLASH, FERMI and the LCLS, is causing a revolution in the ultrafast laser sciences. Using ultrafast XUV/X-ray sources, structural changes can be observed with spatial resolutions down to Angström-scales, with time-resolutions down to the femtosecond, or – in some cases – attosecond scale, and, using the dependence of XUV/X-ray core-level transitions on the atomic mass, often times with atomic specificity.

It is against this background that in the last few years, we have shifted a large part of our research efforts at the Max-Born Institute towards the further development of lab-scale XUV/X-ray sources, and towards the exploitation of ultrashort XUV/X-ray pulses in a range of novel spectroscopies. In my talk, I will present an overview of these research activities, and in doing so, will present an overview of the various ways that HHG techniques can be used to great advantage to study attosecond electronic and femtosecond structural dynamics. I will moreover sketch an outlook of further improvements in lab-scale XUV/X-ray sources that we may envision in the next few years, and the novel research opportunities that this will bring.

Three-dimensional control of photoelectron distributions bichromatic polarization-tailored laser pulses

Matthias Wollenhaupt

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Polarization-tailored bichromatic laser fields have emerged as new twist to steer ultrafast electron dynamics on their intrinsic timescales. Recently, we introduced a novel approach to the generation of polarization-tailored bichromatic fields, based on ultrafast pulse shaping applied to an octave-spanning CEP-stable white light supercontinuum (WLS) [1-2]. The setup provides full control over all bichromatic pulse parameters such as the frequency and amplitude ratio, the spectral phase profile (including CEP and relative phase) and the polarization state of both colors. Bichromatic pulse shaping opens up a new class of polarization-tailored waveforms with application to multi-path coherent control of ultrafast dynamics, generation and control of high harmonics and the design of polarization-sensitive two-color pump-probe experiments with phase-locked CEP-stable laser pulses at a broad range of excitation wavelengths. In our experiments we employ polarization-shaped bichromatic fields to study resonance-enhanced multi-photon ionization (REMPI) of atoms as a prototype scenario for multi-path coherent control. Three-dimensional detection of the photoelectron momentum distribution by photoelectron imaging

tomography provides detailed insights into the excitation and ionization dynamics. In addition, the generation of vortex-shaped photoelectron wave packets from single color REMPI of potassium atoms with sequences of two time-delayed CRCP few-cycle pulses is demonstrated [3].

- [1] S. Kerbstadt, L. Englert, T. Bayer, M. Wollenhaupt, "Ultrashort polarization-tailored bichromatic field", *J. Mod. Opt.*, published online (2016), DOI: 10.1080/09500340.2016.1271151
- [2] S. Kerbstadt, D. Timmer, L. Englert, T. Bayer, M. Wollenhaupt, "Ultrashort polarization-tailored bichromatic fields from a CEP-stable white light supercontinuum", *Optics Express*, accepted (2017)
- [3] D. Pengel, S. Kerbstadt, D. Johannmeyer, L. Englert, T. Bayer and M. Wollenhaupt, "Electron Vortices in Femtosecond Multiphoton Ionization", *Phys. Rev. Lett.* 118, 053003 (2017)

The non-adiabatic channel in ultra-short high-frequency laser pulses

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The dressing of states by strong high-frequency radiation modifies their energies and couplings. In the case of short pulses this, as will be shown, may drive non-adiabatic electron processes sensitive to the envelope-derivative of the laser pulse. As a consequence, a single pulse with two peaks in the derivative separated by the width of the single pulse acts in this regime like a traditional double pulse. The two ensuing non-adiabatic ionization bursts have slightly different ionization amplitudes. This difference is due to redistribution of continuum electron energy during the bursts, negligible in standard photo-ionization. Time-dependent perturbation theory based on a cycle-averaged Kramers-Henneberger gauge permits a detailed understanding of these unusual dynamical features in ultrafast electron dynamics.

Ultrafast photoelectron emission clocked by orthogonally polarized two-color laser fields

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Exposed to strong laser fields, electrons bound to atoms and molecules may release into the continuum directly by absorbing multiple photons, or via an intermediate state when its potential energy resonantly matches the photons. As compared to the photoelectron released directly into the continuum, it is recently predicted that there is a remarkable emission delay for the two-photon ionization of Helium when a resonant intermediate state is involved. It is understood as a time delay acquired during the transition from the resonant intermediate state to the continuum, which is denoted as the absorption time delay and related to the energy derivative of the phase that the electron acquired during the transition. As compared to the tunneling time delay or the relative time delay between the photoelectrons emitted from different initial states, this

absorption time delay on the resonant intermediate state is yet experimentally unobserved.

Here, we demonstrate experimental observation of the absorption time delay on the Freeman resonance states in strong-field multiphoton ionization of Argon atoms. In strong laser fields, the potential energy of the excited Rydberg state may be lifted to match the photons, i.e., Freeman resonance. By employing a phase-controlled orthogonal two-color (OTC) femtosecond laser pulse with comparable fundamental and second-harmonic field intensities, spatial- and energy-resolved photoelectron angular distributions (PADs) are measured as a function of the relative phase of the OTC field. The OTC field spatiotemporally steers the emission dynamics of the photoelectrons and meanwhile allows us to unambiguously distinguish the main and sideband peaks of the above-threshold ionization spectrum. The relative phase shift between the main and sideband peaks gradually decreases with increasing electron energy, and becomes zero for the fast electron which is mainly produced by the rescattering process. We observe a Freeman resonance delay of 140 ± 40 attoseconds between the photoelectrons emitted via the 4f and 5p Rydberg states of Argon when they are ac-Stark shifted to be resonant with the energy of multiple photons of the driving fields.

Strong-field ionization with counterrotating circularly polarized two-color laser fields

Sebastian Eckart

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We report on our results on studying single ionization of helium in counterrotating circular two-color (CRTC) laser fields (780 nm & 390 nm) with overall intensities of up to $8 \cdot 10^{14}$ W/cm². The three dimensional momenta of the fragments are recorded using Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) as experimental technique. We present our new results regarding single photoelectron interferences that are investigated in three dimensional momentum space. Further we discuss new insights regarding the low energy structure of the photoelectron in CRTC laser fields.

Trajectory-free ionization times in strong field ionization

Nicolas Eicke

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Using a purely quantum mechanical approach without trajectories, we are able to compute momentum-resolved ionization times of electrons released by strong-field ionization of atoms. For the attoclock setting, we show that the dominant emission angle corresponds well to instantaneous ionization at the maximum field if the precise value of the angle is determined appropriately and if the field intensity is small enough to avoid depletion of the bound state. Finally, we discuss applications to bicircular fields.

High order harmonic generation with ultrashort bichromatic circularly polarized pulses is shown to occur efficiently in molecules by recollision

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High Order Harmonic Generation with ultrashort bichromatic circularly polarized pulses is shown to occur efficiently in molecules by recollision [1] especially when the net time dependent electric field symmetry is coincident with molecular rotational symmetry [2]. HHG polarization is dominantly circular for specific frequencies and rotational symmetries as predicted by dynamical symmetry rules. Circular harmonics are then used to generate new circular attosecond pulses [3] which are sources for the creation of attosecond electronic currents and magnetic pulses in molecules [4].

[1] T Zuo, AD Bandrauk, J Nonlin, Opt Phys Mater 4, 533(1995)

[2] AD Bandrauk, F Mauger, KJ Yuan, J Phys B 49, 23LT01(2016)

[3] KJ Yuan, AD Bandrauk, Phys Rev Lett 110, 023003(2013)

[4] KJ Yuan, AD Bandrauk, Phys Rev A 88, 013417(2013)

Strong-field ionization of molecules by bicircular laser field

Azra Gazibegović – Busuladžić

University of Sarajevo, Zmaja od bosne 35, 71000 Sarajevo, Bosnia and Herzegovina

Exploration of strong-field ionization (and HHG) of atoms by bicircular laser fields showed that ATI atomic spectra exhibit many-fold reflectional, and rotational symmetries, depending on the field symmetry itself. Due to the fact that reflectional symmetry of the ATI spectra can be linked to the time inversion of the process, there is no reflectional symmetry in HATI spectra. Only few of these symmetries remain in molecular spectra case, depending on molecular orientation and type. Comparison of eventual remaining symmetries of ATI and HATI spectra for homonuclear and heteronuclear diatomic molecules, and some polyatomic molecules is going to be presented and theoretically explained. Some differences in HATI bicircular spectra due to the shape of the highest occupied molecular orbital can be noticed. Symmetries for HATI spectra for non-aligned and partially aligned molecules are going to be discussed.

Attosecond recorder of the polarization state of light

Alvaro Jiménez-Galán

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Attosecond science has made first steps towards time-resolved imaging of electron dynamics in the condensed phase. However, time-resolved probing of such fundamental condensed phase processes as magnetization dynamics, spin currents, or chiral interactions on the attosecond timescale is a dream that requires addressing

two major challenges: a) generation and b) complete characterization of chiral attosecond pulses. What's more, the first step cannot be made without the second: no claim of generating such pulses can be made without simultaneous measurement of their helicity, degree of polarization, and temporal profile, with no a priori assumptions. Indeed, state of the art experimental setups have now provided robust foundation for the generation of circularly polarized XUV light at individual frequencies and attosecond pulse trains. Yet, the second problem - that of characterizing isolated attosecond pulses, especially their degree of polarization, i.e. distinguishing polarized from partially polarized light - has so far remained unsolved.

Here we propose a complete solution to this problem. We put it on stringent test using ab initio simulations and demonstrate that our method lies fully within the current experimental capabilities.

Our solution is general and physically appealing - it relies on measuring the extrinsic 2D chirality induced in a spherically symmetric atom by a chiral attosecond pulse: the chiral properties of the attosecond pump pulse are mapped onto the attosecond electronic response and detected via left-right asymmetry in the photo-electron spectra induced by a non-chiral, linearly polarized infrared probe pulse. The asymmetry faithfully reproduces the complete polarization state of the attosecond pump pulse. We demonstrate robust reconstruction of partially polarized attosecond pulse from the ab-initio simulated measurements under typical experimental conditions.

Complete characterization of isolated attosecond pulses will enable time-resolved studies of chiral-sensitive light-matter interaction, from electron dynamics in chiral molecules to ultrafast magnetization and spin transport in condensed matter with unprecedented temporal resolution. We believe that we have developed the core tool necessary for this endeavor.

Selection rules in high harmonic generation

Oren Cohen

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High harmonic generation (HHG) is very sensitive to the presence of symmetries in the driving laser and nonlinear medium. Such dynamical symmetries are reflected in forbidden harmonics, polarization and other properties of the high harmonics. As such, dynamical symmetries and their associated selection rules are useful for HHG-based ultrafast spectroscopy of electronic state populations, molecular structures and configurations, as well as for producing high harmonics with special features. I will present a general group theory formalism for describing dynamical symmetries and selection rules in high harmonic generation [1]. Several novel selection rules will be presented, including a discrete elliptical symmetry that results with high harmonics, where all the harmonic orders have the same ellipticity. Experimentally, I will present utilization of the discrete rotational dynamical symmetry for generation of bright circularly polarized high harmonics [2,3] and polarization-fan high harmonics [4] as well as their applications for nano-scale magnetic imaging [5] and chiral discrimination.

[1] O. Neufeld, D. Podolsky and O. Cohen, arXiv: 1706.01087

[2] A. Fleischer, O. Kfir, T. Diskin, P. Sidorenko, and O. Cohen, Nature Photonics, 8, 543 (2014)

[3] O. Kfir et al., Nature Photonics 9, 99 (2015)

[4] A. Fleischer, O. Kfir, P. Sidorenko, and O. Cohen, J. Phys. B., 50, 034001 (2017)

[5] O. Kfir et al., arXiv:1706.07695

Control of attosecond light polarization in two-color bi-circular fields

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We present a way to control an ellipticity of attosecond pulse train produced through generation of high harmonics by coherently combining the fields from infrared laser and its second harmonic with counter-rotating circularly polarization. In such combination of the driving fields out of 35 fs pulses from Ti: sapphire laser system, the generated XUV -radiation consists of pairs of circularly polarized harmonics orders $3N \pm 1$ with harmonics order $3N$ forbidden. Herewith, the $(3N + 1)$ harmonics have polarization with helicity as fundamental beam, whereas the harmonics $(3N - 1)$ as second-harmonic beam. Although the generated high harmonics are chiral, the whole XUV-spectrum is non-chiral, because the overall chirality cancels since the pairs of counter-rotating harmonics have a similar intensity. We investigate the possibilities for creation of a helicity dependent filter to extract a group of harmonics with one definite helicity. Follow this way we will be able to generate chiral spectrum and produce attosecond pulse train and isolated attosecond pulses with circular polarization. The propensity rules responsible for the contrast between harmonic lines of the opposite helicity in the HHG spectra of noble gas were formulated. We use a specific property of Ne atoms, explained on basis of semiclassical model (SAE approximation), to asymmetry in emission of the left- and right-circularly polarized high harmonics and boost this asymmetry by exploring helicity sensitive phase-matching conditions in a thin gas cell. A proper combination of experimental parameters related to phase-matching, intensities of fundamental and second harmonic laser fields, and their mutual temporal position, allows us to influence the intensity of the $3N-1$ harmonics in a wide range. The generated XUV-spectra demonstrate the possibility to control the relative intensity of the harmonics $3N-1$ (polarized as second harmonic driving beam) compared to harmonics $3N+1$ (polarized as fundamental driving beam) from 1 down to 0.1. The corresponding train of attosecond pulses was estimated to be circularly polarized with ellipticity permanently tuned up to 0.7. The demonstrated helicity dependent filter gives a new possibility to tune the harmonics ellipticity in good controllable and predictable way. We also apply this approach for few-cycle driving pulses and succeed to produce chiral XUV continuum potentially suitable for generation of circularly polarized isolated attosecond pulses.

Single-shot, real-time, carrier-envelope phase measurement based on stereographic-above-threshold ionization at short-wave infrared wavelength

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The carrier-envelope phasemeter (CEPM), based on stereographic above-threshold ionization (stereo-ATI) measurement, has proven to be a precise, real-time, single-shot instrument for CEP measurement at laser wavelengths around 0.8 μm [1]. It also allows for simultaneous characterization of the pulse duration. Going to short-wave infrared (SWIR, 1.4-3 μm) and infrared (IR, 3-8 μm) driving laser wavelengths in strong-field physics is interesting from multiple perspectives, e.g. increasing the high-harmonic generation (HHG) cut-off and obtaining attosecond pulses with shorter pulse lengths. However, long wavelengths make the CEP measurement challenging as it results in a dramatic reduction of the yields of the high-energy back-scattered electrons that are utilized by the CEPM, due to larger electron wave packet spreading and higher return energies. Here, we propose and implement a CEPM based on the stereo-ATI of Xenon operating with few-cycle (10-15 fs) laser pulses at 1.8 μm . The achieved precision of single-shot CEP measurement is ~ 120 mrad and the pulse length measurement is calibrated by a standard frequency-resolved optical gating (FROG) measurement.

[1] T. Rathje et al., J. Phys. B: At. Mol. Opt. Phys. 45, 074003 (2012)

Subcycle quantum physics of light and matter

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Recent progress of an extreme time-domain approach to quantum physics is presented where electrons and photons are manipulated on subcycle optical scales. In the first two examples, phase-locked electric fields are exploited to control structural properties and transport phenomena in electronic systems.

A first experiment uses intense few-cycle transients centered at 25 THz for transient biasing of bulk GaAs with internal amplitudes beyond 10 MV/cm which are far beyond the stationary breakdown field. Synchronized few-femtosecond pulses covering near-infrared, visible and near-ultraviolet frequencies probe the spectral transmission change in the interband transition region with subcycle temporal resolution. We study the evolution of the electronic system from the Franz-Keldysh regime all the way into Wannier-Stark localization [1].

The second example demonstrates electronic transport over the few-nanometer gap of a plasmonic device on an attosecond time scale [2]. Broadband field enhancement allows us to induce tunneling in a single-electron regime working with single-cycle near-infrared bias transients of a total pulse energy in the pJ range. Ultrabroadband Er:fiber technology provides passively phase-locked pulses at multi-MHz repetition

rate that are ideal for future investigations of quantum transport phenomena at optical frequencies and elevated temperatures.

Finally, the first quantum measurements of electric fields will be introduced [3-5]. Here, the bound valence electrons of a semiconductor serve as a test charge for probing the electric field in a four-dimensional space-time volume via electro-optic sampling with few-femtosecond laser pulses. In a first step, we have directly analyzed the vacuum fluctuations of the electric field. Subsequently, we were able to generate squeezed vacua where the quantum noise is redistributed via local modulation of the co-propagating reference frame in a nonlinear emitter crystal. The shape of these temporal noise patterns is dictated by the uncertainty principle and relates to the formation of highly correlated photon states.

[1] C. Schmidt et al., submitted

[2] T. Rybka et al., Nature Photon. 10, 667 (2016)

[3] C. Riek, Science 350, 420 (2015)

[4] A. S. Moskalenko et al., Phys. Rev. Lett. 115, 263601 (2015)

[5] C. Riek et al., Nature 541, 376 (2017)

Coherent Control and Quantum State Reconstruction of Free-Electron Beams in Ultrafast Electron Microscopy

C. Ropers, K. E. Priebe, C. Rathje, A. Feist, S. V. Yalunin, S. Schäfer

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Ultrafast Transmission Electron Microscopy (UTEM) is an emerging technique to study structural and electronic dynamics on the nanoscale. Besides its use as an analytical tool with simultaneous femtosecond temporal and nanometer spatial resolution [1], UTEM also provides for a unique test bench to study quantum optics phenomena with free electrons.

This talk will discuss several examples of free-electron beams interacting with optical near-fields at nanostructures, emphasizing quantum coherent processes. Specifically, for swift electrons traversing intense optical near-fields [2,3], we observe multilevel Rabi-oscillations on a ladder of quantized free-electron states [4], and implement Ramsey-type dual interactions in polarization-controlled, spatially separated near-fields [5]. Employing phase-locked two-color fields, coherent control of free-electron states is demonstrated [6], and we introduce a scheme to characterize the quantum state of such phase-modulated free-electron states in terms of their density matrix or Wigner function [6]. Finally, we demonstrate various new possibilities in the coherent manipulation of the longitudinal and transverse degrees of freedom of free-electron wave functions, including the optical preparation of attosecond electron pulse trains [6].

[1] A. Feist et al., "Ultrafast transmission electron microscopy using a laser-driven field emitter: Femtosecond resolution with a high coherence electron beam", Ultramicroscopy 176, 63 (2017)

[2] B. Barwick, D. J. Flannigan, and A. H. Zewail, "Photon-induced near-field electron microscopy", Nature 462, 902 (2009)

[3] F. J. García de Abajo, Ana Asenjo-Garcia and Mathieu Kociak, "Multiphoton absorption and emission by interaction of swift electrons with evanescent light fields", *Nano Lett.* 10, 1859 (2010)

[4] A. Feist, K. E. Echternkamp, J. Schauss, S. V. Yalunin, S. Schäfer, and C. Ropers, "Quantum coherent optical phase modulation in an ultrafast transmission electron microscope", *Nature* 521, 200 (2015)

[5] K. E. Echternkamp, A. Feist, S. Schäfer, and C. Ropers, "Ramsey-type phase control of free electron beams", *Nature Phys.* 12, 1000 (2016)

[6] K. E. Priebe, C. Rathje, S. V. Yalunin, T. Hohage, A. Feist, S. Schäfer, and C. Ropers, "Attosecond Electron Pulse Trains and Quantum State Reconstruction in Ultrafast Transmission Electron Microscopy", arXiv 1706.03680 (2017)

Towards ultra long-range ab-initio calculations

T. Müller, S. Sharma, E.K.U. Gross, J.K. Dewhurst

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We propose a generalization of the Bloch state which involves an additional sum over a finer grid in reciprocal space around each k-point. This allows for ab-initio calculations of ultra long-range modulations in the density which may involve millions of unit cells but with an efficiency rivaling that of single unit cell. This is due to a new algorithm developed specifically for solving the particular eigenvalue problem that this ansatz requires. Thus physical effects on the micron length scale, which nevertheless depend on details of the electronic structure on nanometer length scales, can be computed exactly within density functional theory. Specifically, our method should be applicable to treat spatially dependent magnetic and electric fields extending far beyond the length scale of a single unit cell.

Theory of high harmonic generation in solids: band structure, orientation dependence and emission time

Mette Gaarde

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This talk present a general overview of our theory for high harmonic generation in solids, using a momentum space description of the generation process in terms of strong-field-driven electron dynamics on the band structure, including general predictions and comparisons to recent experimental results. We concentrate in particular on the connection between crystal symmetry, through its band structure, and the orientation dependence of the harmonic emission. We show that the orientation dependence of both the spectral yield and the sub-cycle time profile of the harmonic radiation can be understood in terms of the coupling strengths and relative curvatures of the valence band and the low-lying conduction bands. In particular, we show that in some systems this gives rise to a rapid shift of a quarter optical cycle in the timing of harmonics in the secondary plateau as the crystal is rotated relative to the laser polarization.

Opto-optical phase modulation of extreme ultraviolet light pulses

Johan Mauritsson

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We present an experimental and theoretical study of the recently demonstrated opto-optical modulator, a method for controlling the direction, duration and timing of extreme ultra-violet (XUV) pulses using infrared (IR) control pulses [1]. Coherent XUV light is used to promote an ensemble of atoms to a superposition of the ground state and a series of excited states and the IR pulse is used to control the phase of the light emitted by the excited atoms.

An ensemble of atoms exposed to a short, coherent light pulse will respond collectively and the excited atoms will act as oscillating dipoles. These dipoles may continue to oscillate coherently for a long time after the excitation pulse has passed, resulting in forward scattered light known as free induction decay (FID) [2,3]. This forward scattered light has the same spatial properties as the excitation pulse, but the phase is shifted by π . The overlap between the input and generated fields will yield the normal Lorentzian absorption spectrum observed in optical spectroscopy. For any other phase shift, there will be an asymmetric profile, as for example when an autoionizing resonance is excited [4]. By applying an IR control pulse after the excitation pulse we can control the phase of the dipole emitters. In the simplest case this happens because the IR pulse couples excited states, leading to Stark shifts.

If the delay controlled IR pulse is co-linear, but non-coaxial, with the XUV pulse a Stark induced phase gradient can be induced resulting in a precise control of the direction and timing of the xFID emission. If the induced phase gradient is non-linear, either because the intensity variation across the ensemble of dipoles is non-linear or because the Stark shift varies non-linearly with intensity, the spatial shape of the XUV emission can be controlled. We observe, for instance, that the direction of the xFID signal from the 2p state in helium is opposite to that for the higher np manifold at low IR probe intensities, as expected from the direction of the applied Stark shift. In addition the emission from the 2p state splits for higher IR probe intensities, emitting FID in two directions since the gradient of the Stark shift of this state changes sign with increasing IR intensity. This forms an effective XUV beam splitter. We also observe that by applying a non-linear intensity variation across the ensemble we can overlap spatially different parts of the XUV emission in the far-field where interference is observed, opening the door for "which way" interference experiments.

Applications for the opto-optical phase modulator include reducing the temporal jitter in optical-FEL pump-probe experiments, background free 2D spectroscopy in the XUV, and ultrafast 'which-way' interferometry.

[1] S. Bengtsson, et al., *Nature Photonics* 11, 252-258 (2017)

[2] R. G. Brewer and R. L. Shoemaker, *Phys. Rev. A* 6, 2001 (1972)

[3] F. A. Hopf, R. F. Shea, and M. O. Scully, *Phys. Rev. A* 7, 2105 (1973)

[4] C. Ott, et al., *Science* 340, 716-720 (2013)

Nanoplasmonic near-field probing with ultrafast photoemission

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Plasmonic enhancement of optical near-fields at nanostructures ensures the nanoscale localization of the energy of light. Although this phenomenon allowed pioneering applications in spectroscopy, photovoltaics and sensorics, the measurement of the maximum achievable nanoplasmonic field enhancement for a particular sample is still a challenging problem. Here, we present nanoplasmonic near-field measurement experiments with the help of photoemitted electrons induced by femtosecond laser pulses.

The method is experimentally demonstrated on different model systems involving nanoparticles exhibiting localized plasmons and thin films with controlled surface roughness supporting propagating surface plasmons. As a first step, ultrashort laser pulses excite propagating or localized plasmons, of which plasmonic near fields induce photoemission of electrons into vacuum surrounding the sample [1-3]. Photoelectron spectra are then measured with a time-of-flight spectrometer. Since the highest photoelectron energies originate from nanometric rescattering of the electrons from the metal surface, nanoscale near-fields can be measured by analyzing the the highest electron energies in the electron spectra. This way, field enhancement values between 21 and 51 for 4 different samples (supporting localized and/or propagating plasmons) were measured. These data are in very good agreement with the results of accurate finite-difference time-domain (FDTD) simulation of the given nanoplasmonic system performed without the use of fitting parameters.

Additionally, further analysis of results on thin films reveal details of coupling between propagating and localized plasmons. The demonstrated method, combined with more sophisticated electron imaging techniques can be extended for full characterization of nanoplasmonic near-fields promising nanometer lateral resolution. These results establish ultrafast plasmonic photoelectrons as versatile probes for nanoplasmonic near-fields enabling time-resolved studies, as well.

[1] P. Dombi et al., "Ultrafast strong-field photoemission from plasmonic nanoparticles," *Nano Lett.* 13, 674-678 (2013)

[2] S. M. Teichmann et al., "Strong-field plasmonic photoemission in the mid-IR at <1 GW/cm² intensity," *Sci. Rep.* 5, 7584 (2015)

[3] P. Rácz et al., "Measurement of nanoplasmonic field enhancement with ultrafast photoemission", *Nano Lett.* 17, 1181-1186 (2017)

Tunable high harmonic pulses from nanorings swirled by optical vortices

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The interaction of light carrying orbital angular momentum [1], also called optical vortices, with matter opens the door to exciting effects and mechanisms beyond the optical dipole selection rules. We demonstrated in the past that the transfer of OAM to charge density can be used to manipulate and steer the carrier dynamics. A direct consequence is the possibility to generate a sizable, directed photocurrent [2,3].

Here we report on the irradiation of intercalated nanorings by optical vortices which ignites a charge flow that emits coherent trains of high harmonic bursts. The frequencies and time structures are highly controllable by the topological charge of the driving vortex beam. As an analogy to synchrotron radiation, the polarization of emitted harmonics is also selectable by tuning to the appropriate emission angle with respect to the ring plane. As a demonstration of the fundamental quantum mechanical tunneling process, the non-equilibrium orbital magnetic moment triggered in a ring is translated to the smaller and larger attached rings leading, respectively, to high and low-frequency harmonic generation [4].

Our findings show that the associated frequencies of the emitted harmonics are tunable by simply changing the waist and/or the winding number of the optical vortex, without the need to increase the pulse intensity which, in return, could lead to material damage. These findings follow from a full-fledged two-dimensional quantum dynamic simulations for realistic material and laser parameters. The proposed setup is non-destructive as only short vortex pulses of moderate intensities are needed, and it offers a versatile tool for nanoscale optical and spectroscopic applications such as local, single beam pump-probe experiments.

[1] L. Allen et al., The orbital angular momentum of light and the transformation of Laguerre-Gaussian modes, *Phys. Rev. A* 45, 8185 (1992)

[2] J. Wätzel and J. Berakdar., Centrifugal photovoltaic and photogalvanic effects driven by structured light, *Sci. Rep.* 6, 21476 (2016)

[3] J. Wätzel et al., Optical vortex driven charge current loop and optomagnetism in fullerenes, *Carbon* 99, 439-443 (2016)

[4] J. Wätzel and J. Berakdar., to be submitted

Attosecond-streaking spectroscopy on a liquid-water microjet

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Attosecond-streaking experiments on gas- and liquid-phase water employing the liquid microjet are presented. The streaking traces are used to extract delays between various states, giving access to photoionization dynamics, transport and scattering processes in water.

Exploring the ultrafast frontiers of condensed phase physics with synthesized light fields

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Having the shortest optical^{1,2,3} and soft x-ray fields⁴ as a part of its repertoire, attosecond physics has recently opened new avenues for exploring ultrafast electronic processes in atoms^{5,6}, molecules⁷, surfaces⁸ and nanostructures⁹. I will discuss how recent advancements of the “ultrafast toolbox” allow, the exploration and control of fundamental electronic phenomena in condensed media. Electron motion in bulk media, driven by intense, precisely-sculpted, optical fields give rise to controllable electric currents, the frequency of which extends to the multi-Petahertz range⁹⁻¹⁰, advancing lightwave electronics¹⁰ to new realms of speed and precision. Coherent extreme ultraviolet radiation emerging by these coherent charge oscillations⁹ offers direct insight into structural and dynamical properties of the underlying medium, inaccessible by conventional solid-state spectroscopies. By endowing essential x-ray spectroscopies of solids with attosecond temporal resolution, optical half-cycle fields, combined with extreme ultraviolet pulses, offer, access into the attosecond dephasing of electronic excitation in solids opening the realm of soft x-ray excitonics¹¹. We anticipate these new capabilities to result in far reaching implications in fundamental and applied, electronic and photonic sciences.

- [1] Goulielmakis E. et al., Science 305, 1267 (2004)
- [2] Wirth A. et al., Science 334, 195 (2011)
- [3] Hassan M. Th et al., Nature 530,66 (2016)
- [4] Goulielmakis E. et al., Science 320, 1614 (2008)
- [5] Goulielmakis E. et al., Nature 466, 739 (2010)
- [6] Kienberger R. et al., Nature 427, 817 (2004) [6], Smirnova et. al, Nature 460,972(2009)
- [7] Cavalieri A L et al., Nature 449, 1029 (2007)
- [8] Krueger M et al. Nature 475, 78 (2011)
- [9] Luu T.T. et al., Nature 521, 498 (2015), Garg et .al., Nature 538, 359 (2016)
- [10] Goulielmakis E. et al., Science 317, 769 (2007)
- [11] Moulet A. et al., Science (2017) in print

Attosecond physics at the nanoscale

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One of the most fundamental processes in nature is light-induced photoemission of electrons. When an electron is photoemitted inside a solid, it can undergo elastic and inelastic collisions before escaping from the surface. The mean free path of both types of collisions depends on electron energy and typically lies in the nanometer scale for

sub-keV electron energies in most solids. Earlier work showed experimental and conceptual problems for determining the inelastic scattering mean free path, in particular for energies below 50 eV. Since the characteristic scattering time scale is in the attosecond regime, attosecond technology provides an opportunity to directly access electron scattering in the time domain. Such studies have, however, been limited to the investigation of metals or adlayer-covered metals, and accumulative charging of dielectrics prevented their implementation on dielectrics. We have overcome this problem by using nanoscopic dielectric targets in a continuous beam, replacing the nanoscopic solid for each laser shot [1]. Our studies reveal that the observed streaking delay for electrons emitted from SiO₂ nanoparticles is almost entirely determined by the inelastic scattering time [2]. Additional effects of the elastic scattering time strongly depend on the materials permittivity, and are found to cancel in the permittivity region of typical dielectric materials. Our results signify that experimentally accessible relative streaking delays serve as a direct clock for inelastic scattering times in dielectrics. The approach for clocking inelastic scattering times was demonstrated on SiO₂, but should be transferable to other dielectrics. Field localization at nanostructures illuminated with laser pulses of well-defined waveform enables spatialtemporal tailoring of the near-fields for sub-cycle control of electron dynamics at the nanoscale [3]. We have investigated modifications of fundamental ponderomotive recollision dynamics via multi-electron effects [3,4], studied non-ponderomotive effects in the electron emission from metallic nanotips [5], and implemented attosecond near-field sampling [6,7]. Field-propagation-induced shaping of near-fields results in size-dependent directional tuning of the electron emission from isolated nanospheres. We present recent progress towards an all-optical control of the electron emission direction and underlying many-particle physics in intense, femtosecond linearly and counter-rotating circularly polarized two-color fields tailoring the spatio-temporal near-fields of SiO₂ nanoparticles [8]. The many-body dynamics is modeled with trajectory-based Mean-field Mie Monte-Carlo (M₃C) simulations.

- [1] Q. Liu et al., “Attosecond streaking metrology with isolated nanotargets”, submitted.
- [2] L. Seiffert et al., “Attosecond chronoscopy of electron scattering in dielectric nanoparticles”, Nat. Phys. 13, 766-770 (2017)
- [3] M.F. Ciappina et al., “Attosecond physics at the nanoscale”, Rep. Progr. Phys. 80, 054401 (2017)
- [4] L. Seiffert et al., “Trapping field assisted backscattering in strong-field photoemission from dielectric nanospheres”, J. Mod. Opt. 64, 1096-1103 (2017)
- [5] J. Schötz et al., “Non-adiabatic ponderomotive effects in photoemission from nanotips in intense mid-infrared laser fields”, submitted
- [6] B. Förg et al. “Attosecond nanoscale near-field sampling”, Nat. Comm. 7, 11717 (2016)
- [7] J. Schötz et al., “Reconstruction of Nanoscale Near Fields by Attosecond Streaking”, IEEE J. Sel. Top. Quant. El. 23, 8700111 (2017)
- [8] Q. Liu et al., “All-optical spatio-temporal control of electron emission from SiO₂ nanospheres with two-color femtosecond laser pulses”, in Preparation

Intermediate state dependence of femtosecond photoelectron circular dichroism

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The intermediate state dependence of photoelectron circular dichroism (PECD) in resonance-enhanced multi-photon ionization of fenchone in the gas phase is experimentally studied. By scanning the excitation wavelength from 359 to 431 nm, we simultaneously excite a different number electronically distinct resonances. In the PECD experiment performed with a broadband femtosecond laser, their respective contributions to the photoelectron spectrum can be resolved. High-resolution spectroscopy allows us to identify two of the resonances as belonging to the B- and C-bands, which involve excitation to states with 3s and 3p Rydberg character, respectively. We observe a sign change in the PECD signal, depending on which electronic state is used as an intermediate, and are able to identify two differently behaving contributions within the C-band. Scanning the laser wavelength reveals a decrease of PECD magnitude with increasing photoelectron energy for the 3s state.

Attosecond photoionization dynamics close to Fano resonances

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Autoionization is a prototypical example of ultrafast photoemission dynamics governed by electron correlation. This dynamics is encoded in the scattering phase and amplitude of the released electron wavepacket (EWP). The recent progress in attosecond spectroscopy is making possible the spectral characterization of this resonant EWP. The phase variation across Fano resonances [1] was first measured in argon using the RABBIT technique [2], or attosecond streaking [3]. Then, in Gruson et al. [4], we fully characterized the EWP emitted through the sp²⁺ Fano resonance in helium, using spectrally-resolved RABBIT, so-called Rainbow RABBIT. This allowed reconstructing the complete autoionization dynamics, including the resonance buildup (Fig. 1). This evidenced how photoelectron wavepackets are born and morph into asymmetric Fano profiles. During this talk, I will review the recent studies performed in collaboration between CEA-Saclay and Lund University in attosecond photoionization spectroscopy.

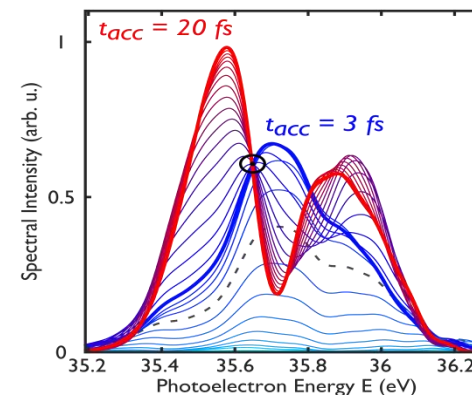


Figure 1. Experimental reconstruction of the buildup of the helium Fano 2s2p resonance using spectrally-resolved electron interferometry (from [4]). The photoelectron spectrum is plotted as a function of the upper temporal limit (accumulation time t_{acc}) used for the inverse Fourier transform of the temporal EWP. The evolution of the spectrum is shown for accumulation times t_{acc} between -10 fs and 20 fs and $\Delta t_{acc} = 1$ fs.

- [1] U. Fano, *Phys. Rev.* **124**, 1866 (1961)
- [2] M. Kotur, *et al.*, *Nat. Com.* **7**, 10566 (2016)
- [3] M. Sabbar, *et al.*, *Phys. Rev. Lett.* **115**, 133001 (2015)
- [4] V. Gruson, *et al.*, *Science* **354**, 6313 (2016)

Imaging molecules through strong-field-ionization and electron rescattering

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Strongly controlled molecules allow to image the structure and dynamics of individual species of complex molecules and molecular aggregates directly in the molecular frame. I will briefly present the state of the art of the spatial separation of molecular species, such as cluster sizes, structural isomers, or quantum states as well as their alignment and orientation. Then I will discuss details of the photoelectron angular distributions visible in the images obtained from these highly controlled samples and the implications for the recording of spatiotemporally atomically resolved molecular movies.

Ultrafast non-adiabatic relaxation of the naphthalene molecule after inner-valence ionization by a short XUV pulse

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XUV induced dynamics in molecules are nowadays accessible in experiment thanks to the development of attosecond laser pulses [1]. The simulation of such dynamics is, however, very challenging. With such photon energies, inner-valence electrons can be removed, which, due to the strong multi-electronic effects, populate a large multitude of electronic states [2]. Furthermore, a strong non-adiabatic coupling between the electronic and the nuclear degrees of freedom is expected [3]. To overcome these difficulties we developed a model that permits to treat the ultrafast non-adiabatic relaxation that occurs in the naphthalene molecule after the population of cationic eigenstates lying close to the double ionization threshold. The model is based on a vibronic-coupling Hamiltonian derived from the electronic potential energy surfaces obtain with the Algebraic Diagrammatic Construction scheme. It includes 23 cationic states and 25 normal vibrational modes. The model Hamiltonian was used to propagate nuclear wave packets on the coupled manifold of cationic states with the Multi Configuration Time Dependent Hartree method. Our results have permitted to interpret a recent experiment performed by the group of F. Lépine (Institute of Light and Matter, Lyon). Using time-resolved electron momentum imaging, the relaxation times of several cationic states of naphthalene have been directly observed. The measured relaxations times fall in the range of few tens of femtoseconds and, counter intuitively, increase with the increase of the energy of the states. Our simulations well reproduce this behavior and show that it is a result of the increasing density of states when approaching the double ionization threshold.

[1] F. Lépine, M. Y. Ivanov, M. J. J. Vrakking, Nature Photonics 8, 195-204 (2014)

[2] L. S. Cederbaum, W. Domcke, J. Schirmer, W. Von Niessen, Adv. Chem. Phys. 65, 115-159 (1986)

[3] A. Marciniak, V. Despré, T. Barillot, M. C. E. Galbraith, J. Klei, C.-H. Yang, C. T. L. Smeenk, V. Loriot, S. Nagaprasad Reddy, A. G. G. M. Tielens, S. Mahapatra, A. I. Kuleff, M. J. J. Vrakking, F. Lépine, Nature Communications 6, 7909 (2015)

Circular dichroism in the angular distribution of strong-field ionization of asymmetric triatomic model molecules

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The interaction of femtosecond intense, circularly polarized laser fields with chiral molecules features asymmetries in the photoelectron angular distribution. Here, we demonstrate using fully ab initio quantum dynamical simulations, how the electronic structure and symmetry of non-linear triatomic molecules are imprinted into the photoelectron momentum distribution following strong-field ionization. For this, we have implemented a two-dimensional model system consisting of an asymmetric

nuclear environment for a single active electron. We show how the photoelectron momentum distribution depends on the electronic structure of the model molecule and discuss the role of excited states and the Coulombic structure of the continuum for the photoelectron spectra.

Signatures of electron ionization dynamics by ionization

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Brunel radiation appears during ionization of electron and subsequent dynamics, and is independent on the return to the atomic core. Surprisingly, although the time scale of the ionization process is much faster than the period of the most of Brunel harmonics, the signatures of the ionization dynamics can be found in the Brunel harmonic spectrum.

Chiral high-harmonic spectroscopy

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High-harmonic generation is a nonlinear process that converts intense infrared radiation into high-frequency light. It can be understood as a sequence of three steps: tunnel ionization, laser-driven acceleration of the electron in the continuum, and recombination with the core resulting in the emission of harmonic light. Since there is a well-defined relationship between the duration of the electron round-trip and the energy released during recombination, the harmonic spectrum provides snapshots of the dynamics in the ion. Recently, the application of elliptically polarized fields has allowed to probe molecular chirality with sub-femtosecond time resolution [1], opening new directions in high-harmonic spectroscopy [2].

In general, the chiral response of a system increases with the ellipticity (chirality) of the driving field, usually maximizing for circularly polarized light. However, the harmonic signal quickly drops with ellipticity as the liberated electron misses recollision with the core. In this context, the use of two-color counter-rotating bi-circular fields [3] constitutes a promising tool for exploring time-resolved chiral dynamics as they allow recombination while maximizing the chiral response.

We have calculated the high-harmonic spectra of the chiral molecule propylene oxide generated by different configurations of intense chiral laser fields. The chiral response of the system arises due to the interplay between electric and magnetic effects. Our calculations have been performed using the method described in [4,5], including ab initio strong-field ionization rates [6] and photorecombination matrix elements [7]. The resulting harmonic dipole has been averaged on a Lebedev grid in order to account for the experimental condition of randomly oriented molecules. We will discuss the most relevant features arising in the spectra, comparing with recent experimental data

and showing that high-harmonic spectroscopy constitutes a promising tool for probing chiral dynamics with sub-femtosecond time resolution.

- [1] R. Cireasa et al Nat. Phys. 11, 654-8 (2015)
- [2] O. Smirnova et al, J. Phys. B 48, 234005 (2015)
- [3] D. B. Milošević et al, Phys. Rev. A 62, 011403 (2000)
- [4] O. Smirnova and M. Ivanov, Chap. 7 in Attosecond and XUV physics, Wiley (2013)
- [5] B. D. Bruner et al, Farad. Discuss. 194, 369-405, (2016)
- [6] M. Spanner et al, Phys. Rev. A 80, 063411 (2009)
- [7] D. Brambila, et al, J. Phys. B 48, 245101 (2015)

Attosecond inter- and intra-band dynamics in bulk solids

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We study the transient extreme ultraviolet (XUV) response of bulk solids pumped by near-infrared (NIR) light with peak intensities on the order of 10^{12} W/cm². Comparison of our attosecond transient absorption spectroscopy data with time-dependent density-functional theory (TD-DFT) and simple analytical models allows us to disentangle the contributions of inter- and intra-band electron dynamics in the observed response.

In thin films of polycrystalline diamond, several infrared photons are needed to bridge the bandgap. In these samples, we find the XUV transient absorption signal to be dominated by intra-band electron motion [1]. We identify the measured structures as a manifestation of the dynamical Franz-Keldysh effect driven at NIR frequency and observed well above the band gap of the material.

How does this picture change as we move from a large-bandgap dielectric to a material that can be resonantly driven by the NIR field? Extending our experiments and theoretical studies to the direct-bandgap semiconductor GaAs, we find that rather surprisingly the transient response to the resonant driving field is still governed by intra-band currents. Furthermore, we find that the intra-band dynamics enhances the yield of real excitations into the conduction band through inter-band transitions.

- [1] M. Lucchini, S. A. Sato, A. Ludwig, J. Herrmann, M. Volkov, L. Kasmi, Y. Shinohara, K. Yabana, L. Gallmann, U. Keller, Science 353, 916 (2016)

Crystal-Symmetry-Controlled Polarization Effects in High-Harmonic Generation in Solids

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Several recent works investigating high-order harmonic generation (HHG) in solids have further elucidated the fundamental differences in the underlying microscopic generation mechanism compared to HHG in gases [1]. Profound differences originating from the crystal symmetry can be observed in the polarization properties of the emitted higher harmonics. One striking manifestation was identified in collaboration with Rupert Huber's group (University of Regensburg) in HHG from GaSe, in which the temporal structure of the polarization and the carrier-envelope phase (CEP) slip of the high-harmonic waveforms can be controlled by the crystal symmetry [2]. A second intriguing example is the *anisotropic* ellipticity-dependence of HHG from solids, which was first observed in MgO [3] and interpreted with *real-space* trajectories in a 2D one-band model including scattering from neighboring atomic sites. We have revisited this phenomenon in collaboration with Angel Rubio's group (MPSD Hamburg). *Ab-initio* TDDFT simulations [4,5] and HHG experiments [6] revealed that the various higher-harmonic orders generated in solids (Si [4-6], MgO [5]) exhibit qualitatively different sensitivity to the driver pulse's ellipticity, resulting from a different response of intraband and interband dynamics [5]. By exploiting the driver ellipticity as new control knob, the electron wavepacket can be steered in *momentum space*, thereby it is even possible to extend the HHG cutoff for non-zero ellipticity by 30% [5]. Theoretically predicted [5] *circularly polarized* higher harmonics, whose helicities are determined by crystal symmetry, from single-color driver pulses have been observed in our HHG experiments [6]. Unprecedented opportunities are now emerging using tailored sub-cycle driver waveforms [7] and in combination with intense THz transients, that were recently used in strong-field experiments in condensed matter such as liquid water [8].

- [1] U. Huttner, M. Kira, and S. W. Koch, Laser Photonics Rev. **11**, 1700049 (2017)
- [2] F. Langer, M. Hohenleutner, U. Huttner, S. W. Koch, M. Kira, and R. Huber, Nature Photonics **11**, 227 (2017)
- [3] Y. S. You, D. A. Reis, and S. Ghimire, Nature Physics **12**, 345 (2017)
- [4] N. Tancogne-Dejean, O. D. Mücke, F. X. Kärtner, and A. Rubio, Phys. Rev. Lett. **118**, 087403 (2017)
- [5] N. Tancogne-Dejean, O. D. Mücke, F. X. Kärtner, and A. Rubio, "Ellipticity dependence of high-harmonic generation in solids originating from coupled intraband and interband dynamics," Nature Commun. in press (2017); also [arXiv:1706.02890](https://arxiv.org/abs/1706.02890).
- [6] N. Klemke, G. Di Sciacca, Y. Yang, G. M. Rossi, R. E. Mainz, N. Tancogne-Dejean, A. Rubio, F. X. Kärtner, and O. D. Mücke, "Circularly polarized high-order harmonics from solids driven by single-color infrared pulses," talk Wed_5:45pm, ATTO 2017, Xi'an, China, July 2-7, 2017
- [7] O. D. Mücke, S. Fang, G. Cirimi, G. M. Rossi, S.-H. Chia, H. Ye, Y. Yang, R. Mainz, C. Manzoni, P. Farinello, G. Cerullo, and F. X. Kärtner, IEEE J. Sel. Top. Quantum Electron. **21**, 8700712 (2015)

[8] L. Song, P. Zalden, X. Wu, H. Huang, O. D. Mücke, C. Bressler, and F. X. Kärtner, "Terahertz-induced optical birefringence in liquid water," paper STu3J.8, CLEO 2017, San Jose, CA, USA, May 14-19, 2017.

Rotating rotationless: nonadiabatic alignment of the helium dimer

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Quantum mechanically rotational and vibrational dynamics in molecules is time evolution of corresponding wave packets. Such dynamics can be periodic, as well-known rotational and vibrational revivals with wave packets consisting of many coherently prepared bound states. How would the rotational and vibrational dynamics look like in a molecular system with a single bound state? One of such extreme quantum systems is the helium dimer, where the two-body potential supports only one state.

We applied the nonadiabatic "kick" to the helium dimer by a femtosecond laser pulse (pump) and watched evolution of the system by Coulomb explosion imaging, which was initiated by the second much more intense delayed probe pulse. The observed time-dependent alignment of the helium dimer, as well as time evolution of the rovibronic wave packet, is going to be discussed in the talk.

Near-forward rescattering photoelectron holography in strong-field ionization: extraction of the phase of the scattering amplitude

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We revisit the concept of near-forward rescattering strong-field photoelectron holography introduced by Y. Huismans et al. [1]. The recently developed adiabatic theory [2] is used to show how the phase of the scattering amplitude for near-forward rescattering of an ionized electron by the parent ion is encoded in and can be read out from the corresponding interference pattern in photoelectron momentum distributions (PEMDs) produced in the ionization of atoms and molecules by intense laser pulses. A procedure to extract the phase is proposed. Its application to PEMDs obtained by solving the time-dependent Schrödinger equation for a model atom yields results in good agreement with scattering calculations [3]. Some illustrative results are presented for a finite range potential. This establishes a novel general approach to extracting structural information from strong-field observables capable of providing time-resolved imaging of ultrafast processes.

[1] Y. Huismans et al., Science 331, 61 (2011)

[2] O. I. Tolstikhin and T. Morishita, Phys. Rev. A 86, 043417 (2012)

[3] Y. Zhou, O. I. Tolstikhin, and Toru Morishita, Phys. Rev. Lett. 116, 173001 (2016)

Molecular hydrogen in ultrashort intense laser fields

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Molecular hydrogen remains the prototype molecular system for trying to understand the behaviour of molecules in ultrashort intense laser fields. Over the years, we have developed different levels of approximation describing this system. This talk will first provide an overview over the results obtained previously and then discuss our recent extension of a numerical approach for solving the time-dependent Schrödinger equation of both electrons (in full dimension) by including also a full quantum treatment of the vibrational motion. Among others, the isotope effect will be discussed.

The helium hydride molecular ion (HeH⁺) in strong laser fields

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The simple heteronuclear molecule HeH⁺ is studied both experimentally and theoretically. Ionization is the dominating process in strong 800 nm and 400 nm laser pulses. Future experiments will cover longer wavelengths and linearly polarized two-color-pulses, where dissociation is expected to play a more important role. In this case the different isotopologues (in particular HeD⁺) can be used to study the effect of nuclear masses on the dissociation process.

Posters

1. Xiaochun Gong (ECNU Shanghai)
Two-dimensional asymmetric dissociative ionization of H₂
2. Kang Lin (ECNU Shanghai)
Controlling electron-ion rescattering by bicircular two-color laser pulse
3. Wenbin Zhang (ECNU Shanghai)
Dissociative frustrated double ionization of H₂ steered by phase-controlled two-color femtosecond laser pulses
4. Carlos Granados (Uni Halle)
Ionization of Hydrogenic Atoms by Twisted Beams
5. Stefanie Kerbstadt (Uni Oldenburg)
CEP-dependent asymmetries in the photoelectron momentum distribution from multiphoton-ionization in polarization-shaped bichromatic fields
6. Ossama Kullie (Uni Kassel)
Tunneling time in attosecond experiments and the time-energy uncertainty relation
7. Birger Böning (Uni Jena)
Attosecond streaking with twisted X waves and intense infrared pulses
8. Willi Paufler (Uni Jena)
Strong field ionization with short twisted NIR pulses
9. Ingo Barth (MPI Halle)
Opposite photoelectron angular shifts induced by laser-dressed current-carrying orbitals in strong elliptically polarized pulses
10. Sajal Kumar Giri (MPIPKS Dresden)
Prediction of X-ray pulse properties for strong field induced photo-ionization
11. Christoph Leithold (Uni Jena)
Imaging Fourier Spectroscopy for Nonlinear Delay Measurements
12. Daniel Reich (Uni Kassel)
Towards Branching Ratio Control in Hot Photoassociation of Mg₂ - Theory, Numerics and Experiment
13. Timo Paschen (Uni Erlangen)
Tba
14. Christoph Jusko (Uni Hannover)
Spatiotemporal Characterization of Tailored Filaments
15. Martin Wagner (TU München)
Attosecond-streaking spectroscopy on a liquid-water microjet
16. Evangelos Karamatkos (DESY Hamburg)
Optimization of strong laser field-free alignment using tailored light fields
17. Andrea Eschenlohr (Uni Duisburg)
Femtosecond laser-induced spin dynamics in ultrathin Co/Cu (001) films
18. Ingmar Schubert (HU Berlin)
Calculation of strong-field ionisation yields and Keldysh adiabaticity times using the complex-scaling theory
19. Christian Markus Dietrich (Uni Hannover)
THz radiation by strong tailored fields in a doubly resonant cavity design
20. Lun Yue (Uni Jena)
Application of surface-hopping methods on strong-field-induced breakup of diatomics
21. Eric Mascarenhas (MPI Halle)
Ionization dynamics of current carrying p_± orbitals in OAM pulses
22. Sebastian Eckart (Uni Frankfurt)
Ring Currents in Single Atoms – Ultrafast Preparation and Detection using Strong Field Ionization
23. Daniel Trabert (Uni Frankfurt)
j-resolved measurement of spin polarized electrons produced by strong-field ionization
24. Bruno Schulz (HU Berlin)
HeH⁺ in intense laser fields -- influence of dipole moment and polarizability
25. Daniel Würzler (Uni Jena)
Velocity Map Imaging and Semi-classical analysis of Scattering Dynamics in Orthogonal Two-color Fields
26. Lennart Seiffert (Uni Rostock)
Attosecond Electron Scattering in Dielectrics
27. Kilian Fehre (Uni Frankfurt)
Multicoincidence Studies of Ionization of Chiral Molecules in Strong Laser Fields
28. Jinzhen Zhu (LMU München)
Freeman Resonance Enhanced Photo Electron Spectra
29. Sergej Neb (Uni Bielefeld)
Attosecond delays in the photoemission from the layered, centrosymmetric Bi₂Te₃ and non-centrosymmetric BiTeCl crystals
30. Simon Brennecke (Uni Hannover)
Non-dipole effects in strong-field ionization
31. Arohi Jain (ETH Zürich)
Attosecond-Streaking Spectroscopy on a Liquid-Water Microjet
32. Nicolas Rendler (Uni Freiburg)
Imaging electrons from dopant-induced helium nanoplasmas
33. Jeffrey Brown (Louisiana State University)
Modeling laser filamentation in the mid-infrared

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Schedule

Time	Sunday Sep 3rd	Monday Sep 4th	Tuesday Sep 5th	Wednesday Sep 6th	Thursday Sep 7th	Time
8:45		Welcome				
9:00		Vrakking	Leitenstorfer	Dombi	Gallmann	09:00
9:35		Wollenhaupt	Ropers	Wätzel	Mücke	09:35
10:00		Saalmann	Müller	Jain	Kunitski	10:00
10:25		Coffee	Coffee	Coffee	Coffee	10:25
10:50		Wu	Gaarde	Goulielmakis	Morishita	10:50
11:25		Eckart	Mauritsson	Kling	Saenz	11:25
11:50		Eicke		Senftleben	Oppermann	11:50
12:00			Young Sci. Meeting			
12:15					Conclusion	12:15
12:30		Lunch	Lunch	Lunch	Lunch	12:30
14:00		Bandrauk	Excursion/ Free Time	Salières	Departure	14:00
14:35		Gazibegović - Busuladžić		Küpper		14:35
15:00		Jiménez-Galán				
15:25		Coffee		Despré		15:00
16:00		Cohen		Coffee		15:25
16:35		Zhavoronkov		Gräfe		16:00
17:00	Arrival	Zhang		Babushkin		16:25
18:30				Ayuso		16:50
19:00	Dinner	Dinner	Dinner		18:30	
19:45		Posters	Conference Dinner	Posters		19:45

Internet Access:

Connect to the visible access point with SSID PBH and use the following password:

011235813 “the first 8 Fibonacci number (not digits)”
Encryption **WPA-2 personal**

For participants without laptops there is a computer room on the ground floor with 4 windows computers (2 English and 2 Deutsch), password **pbb**.

Things to remember on Check-out:

- Pay your bill for the drinks at the room next to the main entrance
- Return your key: Please return it to the key cabinet behind the reception desk in the foyer, preferably at the right spot
- On your day of departure, check out of your room before 9:00 am