Deutsche Forschungsgemeinschaft

QUTIF

Quantum Dynamics in Tailored Intense Fields

Final Colloquium

28th August – 1st September 2022

Venue:
Physikzentrum Bad Honnef
Hauptstraße 5
53604 Bad Honnef

Physikzentrum Bad Honnef

Leibniz Universität Hannover

Sunday, 28th August 2022

From 16:00	Registration
From 18:30	Dinner

Monday, 29th August 2022

10:45 – 11:00	Welcome
11:00 – 11:35	Louis DiMauro *Turning the table on strong field physics using attosecond pulses*
11:35 – 12:00	Jochen Mikosch *Channel-resolved laser-driven electron rescattering in the molecular frame*
12:00 – 12:30	Arnaud Rouzée *Strong laser-induced field-free alignment of molecules by tailored laser pulses*
12:30 – 14:00	Lunch
14:00 – 14:35	Hrvoje Petek *PetaHertz optoelectronics*
14:35 – 15:00	Walter Pfeiffer *Local field effects versus tunneling delays in EUV photoemission from BiTeCl*
15:00 – 15:30	Andrea Eschenlohr *Identification and optimization of femtosecond spin transfer at interfaces*
15:30 – 16:00	Coffee Break
16:00 – 16:25	Matthias Wollenhaupt *Photoelectron vortices*
16:25 – 16:50	Anne Harth *Multi-sideband RABBITT and the phase of continuum transitions*
16:50 – 17:15	Thomas Kubail Kalousdian *Time and angular resolved photoelectron spectra of highly-excited Rydberg states of hydrogen*
18:30	Dinner

Tuesday, 30th August 2022

09:00 – 09:35	Giulio Cerullo *Ultrafast charge transfer in heterostructures of two-dimensional materials*
09:35 – 10:00	Cristian Medina *Probing dynamics of helium nanoplasmas using extreme light sources*
10:00 – 10:30	Sascha Schäfer *Coherent interactions of strong optical near fields with free electrons*
10:30 – 11:00	Coffee break
11:00 – 11:35	Lars Bojer Madsen *Nondipole effects in atoms and solids in intense infrared laser pulses*
11:35 – 12:00	Victor Despré *Ab-initio simulations of ultrafast electron-nuclear dynamics: Paving the way to attochemistry*
12:00 – 12:30	Gerhard G. Paulus, Manfred Lein *Fragmentation of the helium hydride ion by intense light*
12:30 – 14:00	Lunch
14:30	Guided tour at Bundeskanzler-Adenauer-Haus
18:30	Dinner
20:00	Poster session

Wednesday, 31st August 2022

09:00 – 09:35	Carlos Hernández-García *Novel nonlinear physics driven by ultrafast structured laser pulses*
09:35 – 10:00	Andrey Yachmenev *Breaking of the nuclear spin symmetry in chiral super-rotors*
10:00 – 10:30	Nicola Mayer *HHG spectroscopy using bicircular fields in achiral and chiral media*
10:30 – 11:00	Coffee break
11:00 – 11:35	Eleftherios Goulielmakis *Attosecond nanooptics*
11:35 – 12:00	Jan Michael Rost *Multi-harmonic pulses interacting with small-gapped materials*
12:00 – 12:30	Miguel Angel Silva Toledo *Strong-field coherent control of isolated attosecond pulses up to the water window range*
12:30 – 14:00	Lunch
14:10 – 14:35	Ihar Babushkin *Extracting details of electron dynamics using the lowest-order harmonic response in strong tailored fields*
14:35 – 15:00	Ronak Narendra Shah *Complete temporal characterization of few cycle pulses for the investigation of nanostructures*
15:00 – 15:25	Yinyu Zhang *Carrier-Envelope phase (CEP) measurement and CEP-dependent strong-field ionization at long wavelengths*
15:30 – 16:00	Coffee break
16:00 – 16:25	Adrian Pfeiffer *Transient absorption and dispersion in the deep UV using a miniature beamline*
16:25 – 16:50	Sebastian Eckart *Amplitude, phase and entanglement in strong field ionization*
16:50 – 17:20	Lennart Seiffert, Philip Dienstbier *Analysis and control of nearfield-driven electron dynamics and charge-interactions at nanostructures - A summary*
18:30	Dinner

Thursday, 1st September 2022

09:00 – 09:35	Thomas Pfeifer *Measuring & steering one, two, and many electrons in combinations of intense NIR & XUV fields*
09:35 – 10:00	Mikhail Ivanov *All-optical Stückelberg spectroscopy of strongly driven Rydberg states*
10:00 – 10:25	Ulrike Frühling *Control of electron wave packets close to the continuum threshold using THz fields*
10:30 – 11:00	Coffee break
11:00 – 11:35	Jamal Berakdar *Structured laser fields for sub-wavelength dynamics*
11:35 – 12:30	Discussion / Perspectives / Conclusion
12:30 – 14:00	Lunch

Turning the table on strong field physics using attosecond pulses

Louis DiMauro (The Ohio State University)

An atom or molecule interacting with an intense, ultrafast laser pulse is a fundamental problem in modern physics. At intensities that are approximately one-tenth an atomic unit of field (50 V/A) the physics is well described by a semi-classical 3-step model where an (1) electron tunnel ionizes, (2) propagates under the strong-field and (3) rescatters with its parent core. The consequence of this physics has opened the areas of attosecond science and spatial-temporal molecular imaging. In addition, basic questions in quantum physics, like delays in photoionization and tunneling times, could be addressed in the laboratory. In a typical strong field experiment, the exponential rate of tunnel ionization step fixes the release phase of the electron wave packet (EWP) at the extreme of the laser's electric field. In this talk, we will review the basics of strong field physics in the context of the semi-classical model. I will then introduce a new approach that allows for more precise studies of the strong field processes. This method, dubbed Quantum Trajectory Simulator (QTS), mimics the 3-step model by replacing the tunneling step with single-photon ionization by an attosecond XUV pulse. A phase-locked intense low-frequency field then drives the EWP mirroring steps (2) and (3) but with little or no ionization. In essence, we are studying strong field physics in the laboratory with a degree of flexibility usually afforded to theory. We will present both experimental and theoretical results demonstrating the viability of this approach as a powerful tool for studying rescattering physics in a more diverse set of conditions than those produced in a normal strong field experiment.

Channel-resolved laser-driven electron rescattering in the molecular-frame

Jochen Mikosch (University of Kassel / Max Born Institute Berlin)

(Federico Branchi, Felix Schell, Tilmann Ehrlich, Mark Mero, Horst Rottke, Serguei Patchkovskii, Varun Makhija, Marc J. J. Vrakking, Jochen Mikosch)

QUTIF PI: Mikosch

A series of reaction microscope experiments on strong-field ionization and laser-driven electron rescattering of the asymmetric top molecule 1,3-butadiene is presented. Importantly, by virtue of the ion-electron coincidence detection, these experiments separate the ground-state (D0) and first excited state (D1) ionization channel. By analyzing coherent rotational wavepacket evolution we extract the polar and azimuthal angle-resolved molecular frame ionization and rescattering probability. By extracting the differential scattering cross section (DCS) for near- to mid-infrared wavelengths we explore the role of different continuum wavepackets for molecular structure determination. By measuring the ellipticity dependence of the rescattering probability under different conditions we explore the role of different quantum orbits. A multi-faceted picture of molecular effects in strong-field ionization and laser-induced electron diffraction ensues.

Strong laser-induced field-free alignment of molecules by tailored laser pulses

Arnaud Rouzée (Max Born Institute Berlin)

(Terry Mullins, Evangelos T. Karamatskos, Joss Wiese, Jolijn Onvlee, Andrey Yachmenev, Sebastian Trippel, Phillipp Stammer, Marc J. J. Vrakking, Jochen Küpper, Arnaud Rouzée)

QUTIF PIs: Küpper [1], **Rouzée** [2] – [1] University of Hamburg / DESY Hamburg, [2] Max Born Institute Berlin

Fundamental processes that form the basis for chemical reactions occur at very short time scales. To date, our understanding of these processes has been limited by a lack of appropriate tools for probing structure with atomic resolution on such fast time scales. The recent development of free electron laser facilities providing ultrashort X-ray pulses and ultrashort MeV electron beam technologies has enabled the imaging of molecular structure and dynamics by means of X-ray and electron diffraction. In addition, laser-based imaging techniques such as laser-induced electron diffraction are promising routes towards imaging ultrafast molecular dynamics.

In atomic-resolution diffractive imaging experiments performed in a randomly oriented molecular ensemble, most of the structural information contained in the diffraction patterns is washed out. For gas-phase molecules, it is therefore essential to "fix the molecule in space" in order to maximize the information content of imaging experiments. This can be achieved using laser-induced molecular alignment techniques, in which a first laser pulse is used to align the molecule along a specific direction. When an ultrashort laser pulse is used, with pulse duration shorter than the rotational period of the molecule, the interaction leads to a rotational wavepacket that rephases periodically after the laser pulse has ended leading to a transient alignment of the molecule in field-free condition. Laser-induced field-free alignment of molecules has been intensively investigated in the past 20 years, both for linear and asymmetric-top molecules. However, it remains quite challenging to achieve experimentally a high degree of alignment required for diffractive imaging experiments.

In this talk, we show that by combining rotational quantum-state selection devices with tailored laser pulses, an exquisite control of the alignment of molecules can be achieved experimentally for both linear and asymmetric-top molecules. We will present the results of two experiments performed in OCS and indole, in which we have achieved an unprecedented degree of 1-D and 3-D field-free alignment.

PetaHertz optoelectronics

Hrvoje Petek (University of Pittsburgh)

We address the question: can we select and control the classical vs. quantum response of matter by design of the interacting field? This is the question of PHz (1015 Hz) optoelectronics. To address this question, we examine the transition between the low-field quantum and high-field classical electron emission from a metal single crystal Ag(111) and semiconductor $SnSe_2$ surfaces. To transfer electrons from the Fermi level of a solid to the vacuum, one must promote them to above the work function Φ~2-6 eV, which defines the vacuum level, I_p. In Einsteinian photoemission this follows single photon absorption when the photon energy, $\hbar\omega > \Phi$, and hence is proportional to I. Here, however, we are concerned with high field interactions for $\hbar\omega < \Phi$, where photoemission of electrons occurs by multiphoton absorption as a perturbative quantum mechanical interaction. One can understand the optical field as generating a ladder of Floquet of quasi-energy states such that absorption of n quanta, $n\hbar\omega > \Phi$, enables multiphoton-photoemission (mPP) to occur as an $\vec{E}^{2n}$process [1]. Due to the high fields involved in typically n=2-4 excitations, however, one may expect nonperturbative field-induced effects also to contribute to photoelectron spectra. This may involve field-induced splitting and distortion of electronic bands as the Rabi frequency of the optical interaction exceeds the electronic polarization dephasing rates [2]. We will examine the onset of field emission in competition with mPP as a means of generating and controlling PHz optoelectronic currents.

This is a collaboration with Prof. Shijing Tan, University of Science and Technology of China, and Prof. Min Fang, Wuhan University, and their research groups.

[1] Reutzel, M., Li, A., & Petek, H. Coherent Two-Dimensional Multiphoton Photoelectron Spectroscopy of Metal Surfaces. Phys. Rev. X 9, 011044 (2019).

[2] Reutzel, M., Li, A., Wang, Z., & Petek, H. Coherent multidimensional photoelectron spectroscopy of ultrafast quasiparticle dressing by light. Nature Commun. 11, 2230 (2020).

Local field effects versus tunneling delays in EUV photoemission from BiTeCl

Walter Pfeiffer (Bielefeld University)

(S. Neb, A. Gebauer, W. Enns, J. H. Dil, V. M. Silkin, E. E. Krasovskii, P. M. Echenique, U. Heinzmann, W. Pfeiffer)

QUTIF PI: W. Pfeiffer

Attosecond time-resolved photoemission based on the photoelectron streaking in a time-correlated strong IR field allows investigating temporal delays in the photoemission from different initial states. Discrepancies between experimental observations and existing theoretical models advance our understanding of mechanisms that determine the photoemission kinematics and, for example, allowed recently the identification of an intra-atomic delay as significant contribution to the total photoemission delay [1].
Here we report on attosecond time-resolved photoemission from the layered and non-centrosymmetric crystal BiTeCl. The lack of inversion symmetry allows studying relative photoemission delays for differently terminated but well-defined and inert surfaces with identical bulk properties. The comparison with results from classical and quantum mechanical electron trajectory calculations results in a significant discrepancy for both terminated surfaces. The modeling includes the experimentally observed MFP (≈ 3.5 Å), the screened electron-hole interaction using a Yukawa potential, intra-atomic corrections within the Eisenbud-Wigner-Smith theory, and the dynamic atomic-scale screening of the IR field for both surface terminations. The discrepancy between model and experiment indicates a missing contribution in the model. Here we discuss the impact of local streaking fields within the solid and evanescent photoemission states on the electron emission dynamics. Whereas local streaking effects have only a minor effect on photoemission delays evanescent inverse LEED states can substantially reduce the emission delay.

[1] 1. F. Siek, S. Neb, P. Bartz, M. Hensen, C. Strüber, S. Fiechter, M. Torrent-Sucarrat, V. M. Silkin, E. E. Krasovskii, N. M. Kabachnik, S. Fritzsche, R. D. Muiño, P. M. Echenique, A. K. Kazansky, N. Müller, W. Pfeiffer, U. Heinzmann, Science, 357 (2017).

Identification and optimization of femtosecond spin transfer at interfaces

Andrea Eschenlohr (University of Duisburg-Essen)

QUTIF PIs: Eschenlohr [1], Sharma [2] – [1] University of Duisburg-Essen, [2] Max Born Institute Berlin

In order to exploit and control the transient, non-equilibrium state of condensed matter after optical excitation, the key challenge is to identify and separate the different competing microscopic processes involving the charge, spin and lattice degrees of freedom on their intrinsic timescales. At interfaces, transport processes further become important, and the interface properties are expected to modify the spectrum of excitations as well. We have analyzed the microscopic processes underlying light-induced dynamics at interfaces by combining femtosecond time-resolved second harmonic generation spectroscopy [1, 2] and ab-initio time-dependent density functional theory [3-10]: At Co/Cu(001) interfaces, we find that the ultrafast spin dynamics originates from spin-dependent charge transfer as well as back-transfer between Co and Cu. This femtosecond spin transfer competes with dissipation of spin angular momentum mediated by spin-orbit coupling already on timescales below 100 fs [11]. We further determine the transient spin injection efficiencies at different ferromagnet/metal interfaces and show how to optimize the spin injection through varying the substrates, pump fluences, and pulse durations [12].

[1] J. Chen et al., Appl. Phys. Lett. 110, 092407 (2017).
[2] A. Eschenlohr, J. Phys.: Condens. Matter. 33, 013001 (2020).
[3] S. Shallcross et al., Appl. Phys. Lett. 120, 032403 (2022).
[4] J. K. Dewhurst et al., Appl. Phys. Lett. 120, 042401 (2022).
[5] S. Sharma et al., Appl. Phys. Lett. 120, 062409 (2022).
[6] J. K. Dewhurst et al., Phys. Rev. B 104, 054438 (2021).
[7] Q. Z. Li et al., Phys. Rev. B 103, L081102 (2021).
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[9] S. Shallcross et al., arXiv:2207.11461 (2022).
[10] S. Sharma, P. Elliott, S. Shallcross, Optica, accepted; available at arXiv:2203.14286 (2022).
[11] J. Chen et al., Phys. Rev. Lett. 122, 067202 (2019).
[12] P. Elliott et al., arXiv:2206.00498v1 (2022).

Photoelectron vortices

Matthias Wollenhaupt (University of Oldenburg)

QUTIF PI: Wollenhaupt

Photoelectron vortices with c_6 rotational symmetry generated by multiphoton ionization (MPI) of potassium atoms with a single-color sequence of counterrotating circularly polarized femtosecond laser pulses have been experimentally demonstrated for the first time in Physical Review Letters 118 in 2017 [1]. Subsequently, CEP stable polarization-tailored bichromatic femtosecond laser fields have been used to create and manipulate coherent superpositions of angular momentum wave packets to produce photoelectron vortices with c_1, c_5 and c_7 rotational symmetry [2] [3]. As a first application, the ultrafast dynamics of Rydberg and spin–orbit wave packets have been imaged using bichromatic polarization-tailored schemes [4] [5]. A novel dynamic quantum state holography scheme has been introduced by superimposing a probe and a reference electron wave packet [6]. We have extended the bichromatic scheme to trichromatic triple-pulse sequences to study the phases from the MPI dynamics imprinted in the resulting photoelectron hologram [7] [8]. Very recently, three-dimensional photoelectron wave packet holography based on polarization-tailored trichromatic pulse sequences has been used to create c_2 and c_4 rotational symmetric electron vortices to determine continuum phases in atomic MPI [9]. The experiments have been complemented by two numerical studies in which we calculated the time evolution of the electron wave function in atomic and molecular MPI to analyze the dynamics of the bound-state and the vortex formation [10] [11]. Some of our results on photoelectron vortices obtained in the QUTIF program have been reviewed in Advances in Physics: X in 2019, Progress in Ultrafast Intense Laser Science XV in 2020 and Frontiers in Physics 9 in 2021 [12] [13] [14].

[1] Phys. Rev. Lett. 118, 053003 (2017).
[2] Nature Communications 10, 658 (2019).
[3] J. Phys. B 54 164002 (2021).
[4] Phys. Rev. A 99, 013406 (2019).
[5] New J. Phys. 21, 033001 (2019).
[6] Phys. Rev. A 101, 013430 (2020).
[7] Phys. Rev. A 104, 052805 (2021).
[8] Phys. Rev. A 105, 053113 (2022).
[9] Journal of Physics B, accepted, 2022.
[10] Phys. Rev. A 102, 013104 (2020).
[11] Front. Chem. 10, 899461 (2022).
[12] Advances in Physics: X, 4:1 (2019).
[13] Prog. In Ultrafast Intense Laser Science XV, 136
[14] Front. Phys. 9, 675258 (2021).

Multi-sideband RABBITT and the phase of continuum transitions

Anne Harth (Aalen University / MPIK Heidelberg)

(A. Harth, D. Schmidt, D. Bharti, H. Srinivas, F. Shobeiry, S. Dorra, R. Moshammer, T. Pfeifer, K. Hamilton, K. Bartschat)

QUTIF PI: Harth

Attosecond pulses allow the observation of ultrafast electron dynamics in a variety of different systems. One method to measure such ultrafast dynamics is based on the reconstruction of attosecond beating by the interference of two-photon transitions (also called RABBITT). It is an interferometric technique and involves, as the name suggests, two photon absorption steps: An extreme ultraviolet (XUV) attosecond pulse train ionizes a target by single-photon absorption, thereby creating main peaks in the photoelectron kinetic energy spectrum (first step). The presence of a temporally and spatially overlapped near-infrared (NIR) field creates one sideband (SB) in between these main peaks (second step).
The SB intensity oscillates sinusoidally as the delay between the XUV and the NIR beams is varied and a phase is extracted from this oscillation. A so-called asymptotic approximation considers the extracted phase as a sum of the Wigner phase linked to a single-photon ionization process and the continuum-continuum phase associated with further single-photon transitions in the continuum.
We are interested in this second step, the continuum dipole transitions, and developed a Multi-SB RABBITT method, where multiple SBs appear in between the main peaks. We present first angular resolved experimental and theoretical results of a three-SB RABBITT experiment on Argon and extend the asymptotic approximation to multi-SB RABBITT schemes. The strength of the multi-SB RABBITT method is its ability to focus on the continuum transitions only, independently of the ionization step and the chirp of the XUV beam.

Time and angular resolved photoelectron spectra of highly-excited Rydberg states of hydrogen

Thomas Kubail Kalousdian (Max Born Institute Berlin)

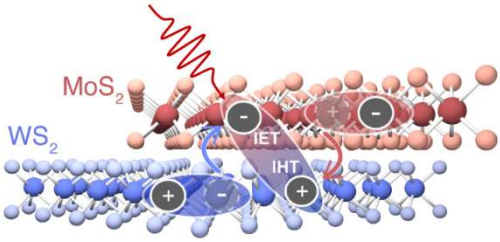
QUTIF PI: Vrakking

The ultrafast dynamics resulting from the excitation of high-lying electronic states in the vicinity of the first ionization potential of molecular hydrogen are investigated by time-resolved photoelectron and photoion spectroscopy. Femtosecond extreme-ultraviolet (XUV) pulses are used to launch a wavepacket on the electronic excited states of the molecule. Following the ionization of the excited molecules by a time-delayed ionizing near-infrared (NIR) laser pulse, the resulting wavepacket dynamics is mapped in the photoelectron momentum distributions that are recorded using a velocity map imaging spectrometer. Pronounced oscillations are observed in the yield and in the angular distribution of the vibrationally-resolved photoelectron spectrum. The quantum beat frequencies extracted by Fourier analysis of the time-resolved photoelectron spectrum are used to assign the pairs of Rydberg states. We find that most prominent features can be attributed to a small number of low-n (n=3,4) states. This allows for a detailed analysis of the dynamics between few pairs of states, even if the density of states in this spectral range is large. A full description of dynamics necessitates a description of the Rydberg states beyond the Born-Oppenheimer approximation, and indicates coupling between electronic angular momentum and the rotational angular momentum of the molecular core.

Ultrafast charge transfer in heterostructures of two-dimensional materials

Giulio Cerullo (Polytechnic University of Milan)

(Oleg Dogadov, Veronica Policht, Stefano Dal Conte, Giulio Cerullo)



Heterostructures (HS) of two-dimensional materials offer unlimited possibilities to design new materials for applications to optoelectronics and photonics. In such HS the electronic structure of the individual layers is well retained because of the weak interlayer van der Waals coupling. Nevertheless, new physical properties and functionalities arise beyond those of their constituent blocks, depending on the type and the stacking sequence of layers. In this presentation we use high time resolution ultrafast transient absorption (TA) and two-dimensional electronic spectroscopy (2DES) to resolve the interlayer charge scattering processes in HS.
We first study a $WSe_2/MoSe_2$ HS, which displays type II band alignment with a staggered gap, where the valence band maximum and the conduction band minimum are in the same layer. By two-colour pump-probe spectroscopy, we selectively photogenerate intralayer excitons in $MoSe_2$ and observe hole injection in WSe_2 on the sub-picosecond timescale, leading to the formation of interlayer excitons (ILX). The temperature dependence of the build-up and decay of interlayer excitons provide insights into the layer coupling mechanisms [1]. By tuning into the ILX emission band, we observe a signal which grows in on a picosecond timescale, significantly slower than the interlayer charge transfer process. This suggests that photoexcited carriers are not instantaneously converted into the ILX following interlayer scattering, but that rather intermediate scattering processes take place. We then perform 2DES, a method with both high frequency and temporal resolution, on a large-area WS_2/MoS_2 HS where we unambiguously time resolve both interlayer hole and electron transfer with 34 ± 14 and 69 ± 9 fs time constants, respectively [2]. We simultaneously

resolve additional optoelectronic processes including band gap renormalization and intralayer exciton coupling.
Finally, we investigate a graphene/WS_2 HS where, for excitation well below the bandgap of WS_2, we observe the characteristic signal of the A and B excitons of WS_2, indicating ultrafast charge transfer from graphene to the semiconductor [3]. The nonlinear excitation fluence dependence of the TA signal reveals that the underlying mechanism is hot electron/hole transfer, whereby a tail the hot Fermi-Dirac carrier distribution in graphene tunnels through the Schottky barrier. Hot electron transfer is promising for the development of broadband and efficient low-dimensional photodetectors.

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[3] C. Trovatello et al., npj 2D Mater Appl 6, 24 (2022).

Probing dynamics of helium nanoplasmas using extreme light sources

Cristian Medina (University of Freiburg)

(C. Medina, D. Schomas, M. Debatin, L. Ltaif, S. Krishnan, S. Mandal, S. Toleikis, C. Passow, N. Ekanayake, B. Erk, A. Tul Noor, E. Klimešová, Z. Hoque, J. Andreasson,)M. Krikunova, R. Moshammer, T. Pfeifer, A: Heidenreich, F. Stienkemeier, M. Mudrich)

QUTIF PI: Stienkemeier

We report 3 different pump-probe experiments of helium (He) nanodroplets (droplet size ranging from $5*10^4$ to 10^6 atoms) doped with a small krypton (Kr), Argon (Ar) or xenon (Xe) cluster. An extreme ultraviolet (XUV) or X-ray pulses overlapped to a strong-field near-infrared laser (NIR) pulse, were used either as pump or probe-pulse. The experiments were carried out at Flash-1 and CAMP end-stations at DESY and the ELI-beamlines instituted at Prague. The nanoplasma formation begins with a collective resonance in the droplet. This starts an ionization avalanche resulting in a Coulomb explosion. An ion time-of-flight (TOF) and electron velocity map imaging (eVMI) were recorded. Same as diffraction images correlated to their eVMI of pure He and Kr doped clusters.
We report the time-dependent results describing the dynamics of the plasma formation based on the ion yields and ignition probability of the system up to the nanosecond range. Molecular dynamics simulation showed that many Kr atoms are impact-ionized by the photoelectrons, the activation process of the Kr cluster core takes a picosecond (ps) or more, resulting in a broad pump-probe signal. The simulations reproduce the gross features of the broad experimental pump-probe signal with a signal maximum around 1~ps.

Coherent interactions of strong optical near fields with free electrons

Sascha Schäfer (University of Oldenburg)

QUTIF PIs: Groß [1], Lienau [1], Ropers [2], Schäfer [1] – [1] University of Oldenburg, [2] MPI-NAT Göttingen

Our joint project addresses the imaging of strong-field processes in nanosystems using the coherent interaction with ultrashort electron pulses and their manipulation by tailored light fields.

The work in the Lienau group (Univ. Oldenburg) has focused on implementing [1,2] and theoretically analyzing [3,4] an ultrafast electron microscope operating with slow electrons (~ 100 eV) in a point-projection geometry (UPEM) [5]. In contrast to commonly employed swift electrons, slow electrons increase the field-electron interaction time and thus enhance the near-field interaction, offering access to the optical excitations of single, small nanoparticles. This comes at the expense of a much increased phase mismatch between the electron and the optical near-field, rendering it experimentally challenging to harness the favorable properties of slow electrons. We report first observation of time-resolved diffraction of slow electrons by optical near fields in an UPEM. Free electron probe pulses with kinetic energies as low as 80 eV and a duration of 50 fs interact with the near-field of a nanometer-sized Yagi-Uda antenna. We observe electron energy gain and loss due to interaction with longitudinal near-field components and a sideways deflection of the passing electrons by transverse field components. The resulting transient diffraction patterns give access to the vectorial near-field components and the electron group velocity dispersion.

In the Ropers (Univ. Göttingen) and the Schäfer group (Univ. Oldenburg) high-energy electron pulses in ultrafast transmission electron microscopes [6,7] were utilized to study and exploit electron-light interactions mediated by nanostructured matter [8,9].

The Ropers Group focused on exploring the possibilities of using intense light fields to optically shape free-electron beams [10-12] and conversely, quantitatively study the properties of complex optical near fields at the nanoscale [13,14]. In particular, using time-harmonic optical phase-modulation of high-energy free-electron wavefunctions at the interface of an electron transparent membrane, attosecond electron pulse trains could be produced [9,12], and the high coherence of the employed ultrashort electron pulses enabled the implementation of a coherent inelastic electron beam splitter [11]. Furthermore, new schemes employ optical cavities and phase-matching for strongly enhanced electron light interactions [10]. The required quantitative understanding of nanoscale optical near-fields was investigated by combining complex plasmonic [13,14] or dielectric structures [10] with chiral light [13] or mode-specific optical excitation [10,14] broadening the technique of photon-induced near-field electron microscopy (PINEM). Finally, quantum optics concepts like strong coupling and optical coherence transfer using free electrons were theoretically explored [15,16], connecting to future work.

In the Schäfer group, a new ultrafast transmission electron microscope was constructed featuring broadband optical sample excitation. As a first example, it was demonstrated that

swift electrons launch an ultrashort light pulse in a silica fiber tip, giving rise to the emission of coherent broadband cathodoluminescence light with a frequency comb spectrum [17]. The experimental results are compared to a numerical model, describing the induced light field as a superposition of guided whispering gallery modes. Second, using the Oldenburg UTEM instrument, the coupling of femtosecond electron pulses to Fabry Perot modes in $MoSe_2$ thin films is demonstrated [18]. Inelastic electron light scattering with strongly chirped broadband optical pulses (200 nm FWHM bandwidth) allows for a precise energy-dependent mapping of coupling strengths and for the identification of material-specific scattering channels including resonances associated with self-hybridized exciton-polaritons. The spatial resolution achievable with this technique is analyzed and potential extensions for coupling electrons to individual exciton-polariton quantum states are discussed.

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Nondipole effects in atoms and solids in intense infrared laser pulses

Lars Bojer Madsen (Aarhus University)

As the wavelength of the intense driving laser pulse increases, magnetic-field effects beyond the electron-dipole approximation become important. In this talk, a nondipole theory [1] is discussed and applied to above threshold ionization (ATI), where a nondipole-induced shift of the energy-conserving ATI rings is identified [2]. It is shown how these shifts cause the ATI peaks to disappear, and how they can be made reappear if the shift of the ATI rings is accounted for [3]. In the adiabatic regime, it is shown how nondipole effects can be considered for the initial conditions used in two- and three-step models [4]. Finally, beyond-electric-dipole effects in intraband high-order harmonic generation in solids are discussed [5].

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Ab-initio simulations of ultrafast electron-nuclear dynamics: Paving the way to attochemistry

Victor Despré (Heidelberg University)

(Victor Despré, Alexander I. Kuleff)

QUTIF PI: Kuleff

The advent of attosecond physics allowed the observation and manipulation of dynamic processes occurring within the intrinsic time scale of the charge motion in atoms and molecules. This has opened the door to the realization of the dream of attochemistry, namely to control chemical reactions through the manipulation of the pure electron dynamics taking place in the first instants after the excitation of the system.
Thereby, the existence of long-lasting electronic coherences in molecular systems is the first key prerequisite to its realization. Furthermore, understating the mechanism leading to or preventing the loss of coherence is necessary for its development.
The first measurement of decoherence and revival in attosecond charge migration will be presented. This dynamics occurs after excitation of silane (SiH_4) by an IR pulse. Simulations treating quantum mechanically both the electronic and nuclear degrees of freedom permitting the interpretation of the experimental results will be discussed. Using these simulations, the behavior of the coherence and the possibility to conserve coherence trough conical intersection will be rationalized.
The second key prerequisite of the realization of attochemistry is the understanding of how charge migration can impact the reactivity of a molecular system. Recently, an XUV-pump IR-probe experiment performed on adenine has demonstrated a sub 3 fs delay in its dicationic signal. It will be shown, using multielectron wave-packet propagation, that this delay is due to a correlation-driven charge migration occurring in the correlation band region of the molecule that stabilizes the system. The stabilization is due to the delocalization of the created hole and its change from σ to π character. The generality of the observed dynamics makes correlation bands a promising playground for the exploration of the possibilities offered by attochemistry.

Fragmentation of the helium hydride ion by intense light

Gerhard G. Paulus (University of Jena)
Manfred Lein (Leibniz University Hannover)

QUTIF PIs: Gräfe [1], Lein [2], Paulus [1] – [1] University of Jena, [2] Leibniz University Hannover

The helium hydride ion HeH^+, being the simplest polar molecule in nature, is extremely asymmetric and may be viewed as a helium atom with an attached proton. In our combined experimental and theoretical project, we investigate the fragmentation of HeH^+ by various types of intense laser pulses and we shine light on a number of fundamental mechanisms that are difficult to observe clearly in other molecules. In the experiment, we measure kinetic-energy-release spectra using a coincidence detection setup that allows us to distinguish dissociation, single ionization and double ionization. Quantum mechanical Born-Oppenheimer and non-Born-Oppenheimer simulations as well as classical trajectory simulations are used to interpret the measurements and to investigate possibilities for subcycle control of the laser-induced dynamics. Unless the molecule is directly ionized by an exceedingly high laser intensity or the wavelength is too short to couple vibrational states, we find that strong-field dynamics of HeH^+ is generally initiated by vibrational excitation rather than electronic excitation thanks to the permanent dipole [1]. Whether the subsequent fragmentation is dominated by ionization or dissociation depends on the wavelength, which we varied between 400 and 4000 nm [2]. At long wavelength, ionization is suppressed and HeH^+ is a neat example of a microscopic anharmonic oscillator. At intermediate wavelength, dissociation and ionization compete and our quantum mechanical calculations show that the process can be controlled very efficiently when using a two-color field with controlled delay [3]. We have also compared the dynamics of different isotopologues and found dramatic differences in the dynamics due to the mass-sensitive dipole moment of the system. Furthermore, by comparison between experiment and classical simulations, we could confirm the presence of a polarizability-enhanced ionization mechanism in which the molecule is dynamically aligned during the pulse and thus ionizes more efficiently [4].

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Novel nonlinear physics driven by ultrafast structured laser pulses

Carlos Hernández-García (University of Salamanca)

The degree of control we have achieved over the manipulation of light is truly amazing. In particular, nowadays we can create ultrashort attosecond pulses (with durations of trillionths of a second), of very high frequencies (up to the soft X-rays), and with increasingly complex spatial structures thanks to our ability to harness their angular momentum. It's already a decade since the first and pioneering experiments of high harmonic generation (HHG) driven by laser pulses with different angular momentum properties, which have allowed us to, for example, achieve circularly polarized attosecond pulses [1], extreme-ultraviolet (EUV) harmonic vortices with high topological charges [2], time-varying OAM pulses [3], or EUV vector-vortex beams [4].

In this talk we will review some of the works that have triggered the field of ultrafast structured EUV pulses during the last decade. We will focus not only in the ability to tailor the angular momentum properties of high harmonics and attosecond pulses, but also on how through the angular momentum of the driving beam we can harness the frequency and divergence properties of the harmonic radiation [5].

On the other hand, by harnessing ultrafast structured laser pulses, it has been recently proposed the possibility to generate intense, ultrafast magnetic pulses isolated from the electric field (yet obeying Maxwell equations!) [6]. This idea has opened the possibility to explore ultrafast magnetism induced solely by an ultrafast magnetic field. We will show that when circularly polarized magnetic pulses are irradiated into magnetic materials, the magnetization response is chiral and, most importantly, nonlinear [7]. This introduces a novel way to perform all-optical switching at femtosecond or even sub-femtosecond timescales by using moderately intense (hundreds of Tesla) magnetic field pulses. Thus, structured laser pulses offer an appealing alternative to study magnetization dynamics at the attosecond timescale, where a complete understanding of the electronic and spin interactions remains unexplored.

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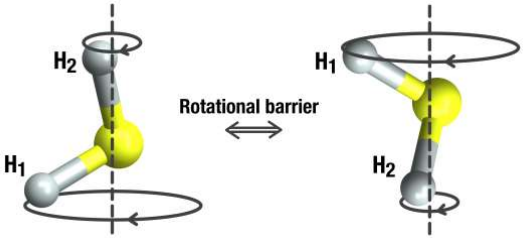
Breaking of the nuclear spin symmetry in chiral super-rotors

Andrey Yachmenev (CFEL Hamburg)

QUTIF PIs: Trippel, Yachmenev

Chirality is conventionally associated with a chemical or optical property of a molecule being in either of its two enantiomeric (mirror-image) forms. In a more general sense, chirality is determined by the time and space inversion (PT) symmetry of the system. That said, it is possible to induce and modulate chirality in statically non-chiral molecules. For example, by forcing a molecule to rotate coherently in one direction, i.e., to possess a well-defined helicity, we can create a chiral entity. Hydrogen sulfide (H_2S) is an excellent example: at high rotational excitation, it forms well separated near degenerate rotational cluster states where the molecule undergoes stable rotation around one of its S-H bonds in a clockwise or anti-clockwise manner [1, 2]. The equivalent hydrogen nuclei are put into orbits that are physically very different, isolating nuclei into permutationally non-overlapping areas that are separated by a high kinetic energy barrier.



In addition to the permutational isolation of equivalent nuclei, another factor contributing to symmetry breakdown is the presence of two identical nuclei in quite different environments. Two hydrogen nuclei revolving in a laboratory frame around different circles will encounter different magnetic fields, resulting in distinct nuclear spin polarisations.

I will present the accurate variational simulations of the nuclear spin hyperfine interactions in the rotational cluster states of H_2S and demonstrate how the nuclear spin symmetry breaking (orthopara transitions) leads to a strong polarization of hydrogen nuclei' spins that has the opposite sign for two equivalent enantiomeric forms, thus leading to the formation of nuclear spin diastereomers.

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HHG spectroscopy using bicircular fields in achiral and chiral media

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QUTIF PI: Smirnova
QUTIF PIs: Ivanov, Zhavoronkov

Bicircular fields are widely used in attoscience due to their intriguing properties, e.g. they can be used to produce circularly polarized XUV light via HHG. In this contribution we discuss two examples of HHG spectroscopy using these particular fields as drivers for the process. In the first example, we use bicircular fields in argon to elucidate the role of excited states in the HHG process. By introducing a time-delay between the fundamental and the second harmonic, we are able to image extremely long trajectories in HHG using the inherent spin-orbit clock of the ion core and show that Rydberg states can play a prominent role in the strong-field process. In the second example, we use tightly-focused, orbital angular momentum carrying, bicircular fields to create a chiral vortex, i.e. light that is chiral locally with azimuthally varying handedness. The spatial distribution of light's handedness is described by a highly tunable topological charge. Using chiral vortices to drive HHG, we are able to easily distinguish molecular enantiomers of opposite handedness and infer few percents of enantiomeric excess in isotropic mixtures.

Attosecond nanooptics

Eleftherios Goulielmakis (University of Rostock)

Having the shortest optical and soft x-ray fields as a part of its repertoire, attosecond physics has opened up new avenues for exploring ultrafast electronic processes in atoms, molecules, surfaces and nanostructures. We will discuss how recent advancements of the "ultrafast science toolbox" allow us to unite attosecond metrology and nanooptics. Real-time measurements of the field-induced emission of electrons from nanostructured metals enable the generation and control of attosecond electron pulses. These new tools will enable new ways of study of dynamic phenomena in solids with picometer spatial and attosecond temporal resolution.

Multi-harmonic pulses interacting with small-gapped materials

Jan Michael Rost (MPIPKS Dresden)

QUTIF PIs: Rost, Saalmann

Through the presence of Dirac points graphene and small-gapped 2D materials exhibit an intricate response to intense laser light. This response is particularly sensitive to multi-harmonic pulses with asymmetric intra-cycle pulse shapes. We develop a general picture for the electronic response enabling us to formulate pulse shapes which lead to large valley polarization, that is, a large difference of excitation between the two domains around the two Dirac points in the Brillouin zone, at moderate total excitation. We also elucidate the different effect dephasing has on excitation and valley polarization.

Strong-field coherent control of isolated attosecond pulses up to the water window range

Miguel Angel Silva Toledo (DESY Hamburg)

QUTIF PIs: Kärtner [1], Rubio [2] – [1] University of Hamburg / DESY Hamburg, [2] MPSD Hamburg / DESY Hamburg

The generation of isolated attosecond pulses (IAPs) covering the extreme ultraviolet (XUV) spectral range has allowed real-time tracking of electron motion following light-matter interaction [1]. The spectral extension of IAPs into the water window region (~ 280 – 530 eV), which covers the K-edges of C, N, and O, has the potential to reveal charge and energy transfer mechanisms underlying most chemical reactions and biological transformations with attosecond resolution and element specificity [2-3]. Yet, the current generation of water window, high-harmonic generation (HHG)-based IAP sources is hampered by low photon-flux and limited control of pulse parameters (e.g., central energy, bandwidth, and pulse duration). Overcoming these technological limitations will enable increasingly sophisticated attosecond experiments.

Here, we show the use of tailored, sub-cycle infrared pulses (i.e., shorter than their main oscillation period) [4], to generate amplitude- and phase-controlled IAPs in the XUV and soft X-ray range [5]. The IAPs are directly generated via HHG and without additional gating techniques. Sub-cycle pulse synthesis is realized by coherently combining near-infrared (0.65 – 1.1 μm, 6 fs, 150 μJ) and infrared (1.2- 2.2 μm, 8 fs, 600 μJ) pulses. By manipulating the relative phase among the constituent pulses and the overall carrier-envelope phase (CEP), we tune the central energy, bandwidth, and duration of the IAPs in the spectral range covering ~30 – 200 eV. Ar and Ne at low backing pressures (<1 bar) are used to drive HHG. Attosecond streaking measurements characterize the temporal profile of the generated IAPs and yield durations from ~80 to 240 attoseconds. Moreover, by focusing tighter and employing Ne and He at higher backing pressures (up to 10 bar), preliminary observations suggest the possibility of obtaining tunable IAPs in the water window region (up to ~450 eV). At present, experiments are underway to explore the possibility of increasing the IAP generation efficiency in the water window region through synthesized infrared waveforms and extending the spectral coverage all the way up to the oxygen K-edge.

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Extracting details of electron dynamics using the lowest-order harmonic response in strong tailored fields.

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(I. Babushkin, U. Morgner)

QUTIF PIs: Babushkin, Morgner

We show that observing the lowest frequency harmonics such as 0^{th}, 3^{rd}, 5^{th} in response of gases, solids and nanostructures to strong multicolor tailored fields gives a possibility to extract the details of electron dynamics during and after the optical excitation. In particular, the 0^{th} harmonics lies in the terahertz (THz) range, that is, has sub-millimeter wavelength; nevertheless, light with such long wavelength is able to provide important details about the angstrom-scale dynamics of electrons during ionization [1], delivering thus an imaging tool with extremely deep subwavelength resolution. More generally, the shape of the electronic wavepacket appearing during the ionization can be reconstructed by observing the polarization of the lowest order harmonics [1], giving thereby rise to "strong-field ellipsometry". Going beyond gases, we observe that in asymmetric metallic nanostructures, reshaping of the electronic wavepackets as it leaves the surface [2] also leaves the fingerprints in the lowest order harmonics, in particular in the 0^{th} harmonic. In solids, the 0^{th} harmonic is especially useful to detect the topological state [3]. We furthermore showed new possibilities for imaging of attosecond-scale atomic response by combining quantum light with strong tailored fields. Namely, in the presence of the strong optical field, single-photon wavepackets co-propagating with the driving pulse can change their shape [4]. Strong driving fields "amplify" the interaction between single photons located at different frequencies, well separated from the pump frequencies to the level of making such interactions observable at realistic propagation distances. The reshaped single photons wavepackets contain the information about attosecond-scale nonlinear response of the medium. Experimental possibilities for the intracavity generation of strong three color tailored pump pulses [5,6] via intracavity enhancement and nondegenreate optical parametric oscillation are discussed. We also shortly discuss some interesting peculiarities of the action of strong tailored ultrashort pulses in resonant media [7,8], as well as explore the generation of nontrivial THz waveshapes with strong tailored two-color fields [9] and generation of harmonics and isolated attosecond pulses [10] using strong THz fields and weak optical fields.

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Complete temporal characterization of few cycle pulses for the investigation of nanostructures

Ronak Narendra Shah (University of Freiburg)

QUTIF PI: Sansone

We show a measurement scheme to directly measure the electric field of laser pulses. In our method, the process of isolated attosecond pulse generation is used as a temporal tip which scans over the pulse to be measured. Using this technique, we are able to measure the complete polarization state of the laser pulse which paves way for future phase and polarisation sensitive measurements.

Carrier-Envelope phase (CEP) measurement and CEP-dependent strong-field ionization at long wavelengths

Yinyu Zhang (University of Jena)

(Y. Zhang, D. Zille, D. Hoff, P. Wustelt, A. M. Sayler, D. B. Milošević, M. Kübel, G. G. Paulus)

QUTIF PI: Paulus

Intense laser-matter interactions are generally determined by the instantaneous electric field of the laser pulse. When the laser pulses consist of only a few of optical cycles (few-cycle pulses), the waveform of the field can be tailored by the phase shift between the maxima of the envelope and peak of the oscillation, which is known as the "carrier-envelope phase" (CEP) or the "absolute phase". Controlling the CEP in light-matter interactions with few-cycle pulses has a profound impact on many applications in investigating the field-driven processes and attosecond physics. Our group has invented an instrument for measuring the CEP, the so-called carrier-envelope phasemeter (CEPM), which is based on the measurement of high-order photoelectron emission in opposing directions along the polarization axis of the laser field. This method has proven to be a robust, precise, real-time, and single-shot CEP and pulse length measurement technique in the visible spectral regime. As increasing the driving laser wavelength has many advantages from multiple perspectives in strong-field physics and attosecond physics, there is a high demand for the precise characterization of the CEP at long wavelengths.

In this talk, we will firstly present our achievements of expanding the single-shot CEP measurement to 1800 nm and the first CEP measurement at 3200 nm with a xenon-CEPM. The measurements are validated with simulations. In the theoretical analysis, we found a significant phase-averaging effect due to the spatial distribution of the CEP in focused few-cycle pulsed beams, which is much more complex than the well-known Gouy phase for monochromatic beams. We will demonstrate the impact of this effect in order to obtain an accurate characterization of CEP-dependent light-matter interactions. Next, we will present the measurement of CEP-dependent photoelectron spectra of caesium at 3200 nm, which could be a more appropriate target for a CEPM operating at mid-infrared wavelengths. In particular, we observed an anomalous high-order CEP-dependence in caesium, which will briefly be discussed at the end of this talk.

Transient absorption and dispersion in the deep UV using a miniature beamline

Adrian Pfeiffer (University of Jena)

QUTIF PI: A. Pfeiffer

In this project, spectroscopy with close-to-collinear beam geometry is investigated, using wide band gap dielectrics both for the generation of deep UV pulses and as probe samples. By superimposing two VIS-IR pulses, a multifaceted structure of temporal and spatial harmonics is created, which allows the generation and separation of pulses in the deep UV without having to rely on spectral filters or polarizers.
Surprisingly, the deep UV pulses consists of double pulses. A new method for the generation of ultrashort pulses by harmonic concatenation is introduced. DUV pulses with a duration of 1.5 fs are synthesized. A new technique is developed for the temporal characterization of the weak broadband DUV pulses, which is based on delay scans of the cross-phase modulation with intense VIS-IR pulses. The DUV pulses are used in a new, further developed variant of transient absorption spectroscopy, called transient absorption and dispersion spectroscopy (TADS), in which double pulses are used as probe pulses. For example, a 250 nm thick, free-standing diamond membrane with a band gap in the DUV is used as a sample.

Amplitude, Phase and Entanglement in Strong Field Ionization

Sebastian Eckart (Goethe University Frankfurt)

QUTIF PI: Dörner

We report on three different experiments with highly intense tailored light fields. All experiments are done using cold-target recoil-ion momentum spectroscopy (COLTRIMS) reaction microscopes and femtosecond laser pulses with tailored intense fields. In the first part of the talk, we show that co-rotating circular two-color (CoRTC) and counter-rotating circular two-color (CoRTC) laser fields (780 nm & 390 nm) can be used to access non-adiabatic offset in momentum space. These offsets affect the amplitude of the final electronic wave function in momentum space representation. Secondly, the technique of Holographic Angular Streaking of Electrons (HASE) is presented. HASE enables the retrieval of Wigner time delays in strong field ionization. Wigner time delays are a property of the final electronic wave function's phase in momentum space representation. For the strong-field ionization of molecular hydrogen, the Wigner time delays vary on an attosecond time scale as a function of the electron's emission direction with respect to the molecular axis. Finally, we report on a recent experiment, that shows that spatially separated entangled atoms can be prepared on femtosecond time scales using femtosecond laser pulses. A probe pulse is used to prove that the prepared atoms are entangled in an internal degree of freedom (the magnetic quantum number of the electrons), and not only classically correlated.

Analysis and control of nearfield-driven electron dynamics and charge-interactions at nanostructures - A summary

Lennart Seiffert (University of Rostock)
Philip Dienstbier (University of Erlangen-Nürnberg)

QUTIF PIs: Fennel [1], **Hommelhoff** [2], **Kling** [3] – [1] University of Rostock, [2] University of Erlangen-Nürnberg, [3] LMU München

The interaction of intense laser fields with nanostructures extends well-established strong-field mechanisms from atomic and molecular systems [1] by plasmonic effects and the rich electronic properties of solid-state systems. The fundamental differences arise from the enhanced and localized nearfields at the surface of nanostructures as well as collisional and collective effects, which modify the electron emission and propagation [2]. Combining these unique properties with nano-structuring capabilities is a promising route to light-driven electronics [3]. Here, we present a summary of our experimental and theoretical results on the dynamics of single electrons and the interaction between multiple charges in the nearfield of nanostructures, more precisely, dielectric and metallic nanospheres and needle tips. At nanospheres, nearfield-driven elastic backscattering of electrons was confirmed using carrier-envelope phase-dependent few-cycle pulses [4] revealing spectral cutoff-energies strongly enhanced compared to atomic targets. Semi-classical trajectory simulations revealed that this enhancement can be attributed to the nearfield-enhancement, a trapping field formed by charge separation at the surface [5] and subsequent Coulomb explosion of the emitted electron bunches. At high intensities, charge interaction dominates the tunneling process and results in a material-independent cut-off scaling for dielectrics [6] or sub-cycle metallization at even higher intensities [7]. Field propagation effects at nanospheres with sizes comparable to the optical wavelength introduce emission directionalities which could be actively tuned via either varying the particle size [8] or all optically by utilizing two-color fields [9]. Finally, attosecond streaking measurements at silica nanospheres enabled clocking of the collisional electron transport within the material [10]. Using a two-color laser field, we demonstrated coherent control of the yield emitted from a needle tip in the perturbative regime at needle tips. By tuning the relative phase between both colors, the emitted current could be almost fully modulated within a wide range of fundamental wavelengths [11-13]. The high visibility could be explained by the interference of two quantum pathways for tungsten tips, whereas, for gold tips, a third emission pathway is

required [14]. Within the strong field regime, we demonstrated the additional control of electron trajectories. Relative phase-resolved photoelectron spectra contained two signatures: an optimal phase depending on energy and a modulation of the high-energy cut-off. Trajectory and quantum simulations allowed us to identify the cut-off modulation with trajectory modifications encoding the second harmonic field strength and relate the optimal phase to the width of the instantaneous rate, which yields an emission duration of 710 ±30 attoseconds for out parameters [15].

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[3] Krausz, F. & Stockman, M. I. Attosecond metrology: from electron capture to future signal processing. Nat. Phot. 8, 205-213 (2014).
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[9] Liu, Q., et al. All-optical spatio-temporal control of electron emission from SiO2 nanospheres with femtosecond two-color laser fields. New. J. Phys. 21, 073011 (2019).
[10] Seiffert, L., et al. Attosecond chronoscopy of electron scattering in dielectric nanoparticles. Nat. Phys. 13, 766–770 (2017).
[11] Förster, M., et al. Two-color coherent control of femtosecond above-threshold photoemission from a tungsten nanotip. Phys. Rev. Lett. 117, 217601 (2016).
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[13] Li, A., et al. Quantum Interference Visibility Spectroscopy in Two-Color Photoemission from Tungsten Needle Tips. Phys. Rev. Lett. 126, 137403 (2021).
[14] Dienstbier, P., Paschen, T. & Hommelhoff, P. Two-color coherent control in photoemission from gold needle tips. J. Phys. B 54, 134002 (2021).
[15] Dienstbier, P. et al. manuscript in preparation.

Measuring & steering one, two, and many electrons in combinations of intense NIR & XUV fields

Thomas Pfeifer (MPIK Heidelberg)

Light-matter interaction at low intensity (single-photon absorption, photoemission, etc.) is a well-understood fundamental process and is thus already commonly used as a characterization tool, e.g. in sensing and diagnostics devices applied to small and complex systems. On the other hand, the physics of intense laser fields that are strongly driving matter leaves lots of room for exploration, learning and application. Part of the challenge is due to the ultrashort (attosecond) time scales on which these processes take place, as well as the emerging dynamical complexity of such processes. Starting already at the level of a single electron after ionizing an atom, strong-field and recollision physics were discovered to describe high-harmonic generation (HHG), above-threshold ionization (ATI) and nonsequential double ionization (NSDI). But lasers that strongly interact with matter typically move more than just one electron…

Current research aims to understand, and then to steer/employ, the fundamental physical mechanisms of increasingly complex systems, in which the single-active electron approximation is immediately violated or will eventually break down as the intensity of the interacting light fields is increased. It is thus of pivotal importance to uncover the mechanistic building blocks of two, or more, electrons interacting with intense laser fields.

Here, we will discuss some recent experiments conducted by the MPIK groups and cooperators exploring the physics of few- to many-electron quantum systems in mixtures of intense fields in the near infrared (NIR) and the extreme ultraviolet (XUV). A combination of light sources and detection techniques is employed for different scientific directions, including:

- high-repetition-rate (up to 100 MHz) intense laser sources for tomographic imaging of ATI electrons at lowest appearance intensities,
- the study of isolated continuum-continuum phase shifts and coincidence imaging of attosecond-controlled entangled electrons with reaction microscopes in two-color fields,
- NIR driven dynamics of state-specifically (XUV) prepared singly and doubly excited states by tunable free-electron laser (FEL) pulses,
- correlated and collective electron dynamics in He clusters and droplets interacting with NIR lasers, XUV synchrotron and FEL light,
- the combination of HHG and tunable FEL pulses for transient-absorption spectroscopy of state-selected atomic tunneling dynamics in (pre-)dissociating oxygen molecules,
- the steering of multiple electrons by modifying the effective electronic exchange interaction in molecules with intense IR fields.

Moving from few to many-electron physics in intense laser fields opens exciting perspectives and research directions for the future, with far-field visions ranging from the preparation and exploration of exotic quantum states of matter to laser-directed chemistry and molecular-scale quantum computing programmed by femto-to-attosecond-shaped light fields.

All-optical Stückelberg spectroscopy of strongly driven Rydberg states

Mikhail Ivanov (Max Born Institute Berlin)

The AC Stark shift of electronic levels is ubiquitous in the interaction of intense light fields with atoms and molecules. As the light intensity changes on the rising and falling edges of a femtosecond laser pulse, it shifts the Rydberg states in and out of multiphoton resonances with the ground state. The two resonant pathways for transient excitation arising at the leading and the trailing edges of the pulse generate Young's type interference, generally referred to as the Stückelberg oscillations. Here we report the observation of the Stückelberg oscillations in the intensity of the coherent free-induction decay following resonant multiphoton excitation. Moreover, combining the experimental results with accurate numerical simulations and a simple model, we use the Stückelberg oscillations to recover the population dynamics of strongly driven Rydberg states inside the laser pulse by all-optical measurements after the end of the pulse. We demonstrate the potential of this spectroscopy to characterize lifetimes of Rydberg states dressed by laser fields with strengths far exceeding the Coulomb field between the Rydberg electron and the core.

Control of electron wave packets close to the continuum threshold using THz fields

Ulrike Frühling (DESY Hamburg)

(Simon Brennecke [1)], Martin Ranke [2),3)], Anastasios Dimitriou [2),3),4)], Sophie Walther [2),3)], Mark J. Prandolini [2)], Manfred Lein [1)], Ulrike Frühling [2),3),5)] – [1)] Leibniz University Hannover, [2)] University of Hamburg, [3)] CUI Hamburg, [4)] NCSR Demokritos Athens, [5)] DESY Hamburg)

We investigate the dynamics of low-energy electrons in intense CEP stable terahertz light fields. Femtosecond NIR laser pulses are used to create electron wave packets trough multiphoton absorption in xenon at well-defined phases of the THz field. The photoelectron momentum distributions are recorded with a velocity map imaging spectrometer for different NIR/THz time delays. We observe signatures of various regimes of dynamics, ranging from recollision-free acceleration to coherent electron-ion scattering induced by the THz field. The measurements are confirmed by three-dimensional time-dependent Schrödinger equation simulations. A classical trajectory model allows us to identify scattering phenomena analogous to strong-field photoelectron holography and high-order above threshold ionization.

Structured laser fields for sub-wavelength dynamics

Jamal Berakdar (University of Halle-Wittenberg)

(Jonas Wätzel, Dominik Schulz, Benjamin Schwager, Jamal Berakdar)

The orbital phase and polarization textures of a propagating electromagnetic wave are not diffraction limited and dictate the field's spatial intensity profile [1]. Experimentally, such laser pulses have been realized in a wide range of frequencies spanning the XUV to the THz regimes and were used for a wide range of applications such as particles' trapping and spinning as well as optical communication and imaging. In this contribution, examples are discussed on how to use structured pulses or a combination thereof to drive and control the quantum dynamics on the sub-wavelength scale and to induce optical transitions and/or laser-dressed states which are not accessible for homogeneous fields [2].

[1] A. E. Siegman, Lasers (Uni. Sci. Books, 1986).
[2] J. Wätzel et al. Adv. Quantum Tech. 2, 1800096 (2019); PRL 128, 157205 (2022); Nat.Phot. 14, 554 (2020).

Poster session

01. Azizi, Sajjad
    Photoionization time delay in 2D model systems
02. Bharti, Divya
    Three sideband RABBITT measurements in Argon and Helium
03. Brennecke, Simon
    Control of electron wave packets close to the continuum threshold using near-single-cycle THz waveforms
04. Dubois, Jonathan
    Stark shift of circularly polarized light
05. Eckart, Sebastian
    Measurement of the attoclock-offset angle and the Wigner time delay in a single experiment
06. Eckart, Sebastian
    Nonadiabatic strong field ionization of atomic hydrogen
07. Habibovic, Dino
    Strong-field ionization of atoms beyond dipole approximation
08. Hergert, Germann
    Photon-Induced Near-Field Interaction in Ultrafast Point-Projection Electron Microscopy
09. Herzig, Elisabeth Anne
    Single cycle electron e mission from metal nanotips
10. Kim, Doyeong
    Transient absorption and dispersion using a miniature beamline
11. Köhnke, Darius
    Trichromatic shaper-based quantum state holography
12. Kubullek, Maximilian
    Tunable isolated attosecond pulse generation by tailored sub-cycle synthesized waveforms
13. Liewehr, Benjamin
    Signatures and scaling of injection harmonics in solids
14. Mhatre, Saurabh
    Mass-ratio dependent dissociation dynamics of HeH^+ isotopologues in intense laser fields
15. Oppermann, Florian
    Stationary initial states for quantum and classical simulations

16. Paulus, Gerhard G.
    Experimental study of the laser-induced ionization of heavy metal and metalloid ions: Au^+ and Si^+ in intense and sculpted femtosecond laser fields.
17. Saalmann, Ulf
    Measuring time-delays and denoising photo-electron spectra
18. Schmidt, Daniel
    Surprising similarities between one-sideband and three-sideband RABBITT
19. Shvetsov-Shilovskiy, Nikolay
    Transfer learning for retrieval of the internuclear distance in a molecule from photoelectron momentum distributions
20. Srinivas, Hemkumar
    RABBIT measurements with a reaction microscope
21. Trippel, Sebastian
    Picosecond pulse-shaping for strong three-dimensional field-free alignment of generic asymmetric-top molecules
22. Winter, Paul
    Bicircular attoclock with molecules
23. Vesalimahmoud, Newsha
    Second harmonic spectroscopy of Cu(001) surfaces
24. Müller, Niklas
    Fast electrons coupled to localized material resonances
25. Sen, Arnab
    Intensity- and wavelength dependence of strong field ionization and fragmentation dynamics of Argon dimer

Schedule

Time	Monday 29.08.2022	Tuesday 30.08.2022	Wednesday 31.08.2022	Thursday 01.09.2022
09:00		Cerullo	Hernández-García	Pfeifer
09:35		Medina	Yachmenev	Ivanov
10:00		Schäfer	Mayer	Frühling
10:30	(10:45)Welcome	Coffee		
11:00	DiMauro	Madsen	Goulielmakis	Berakdar
11:35	Mikosch	Despré	Rost	Discussion / Conclusion
12:00	Rouzeé	Paulus/Lein	Silva Toledo	
12:30	Lunch			
14:00	Petek	Free time / excursion	(14:10)Babushkin	
14:35	Pfeiffer, W.		Shah	
15:00	Eschenlohr		Zhang	
15:30	Coffee		Coffee	
16:00	Wollenhaupt		Pfeiffer, A.	
16:25	Harth		Eckart	
16:50	Kalousdian(17:15)		Seiffert/Dienstbier	
17:20				
18:30	Dinner			
20:00		Posters		

30+5 minutes
25+5 minutes
20+5 minutes

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.