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CARL VON OSSIETZKY universität OLDENBURG

QUTIF

Quantum Dynamics in Tailored Intense Fields

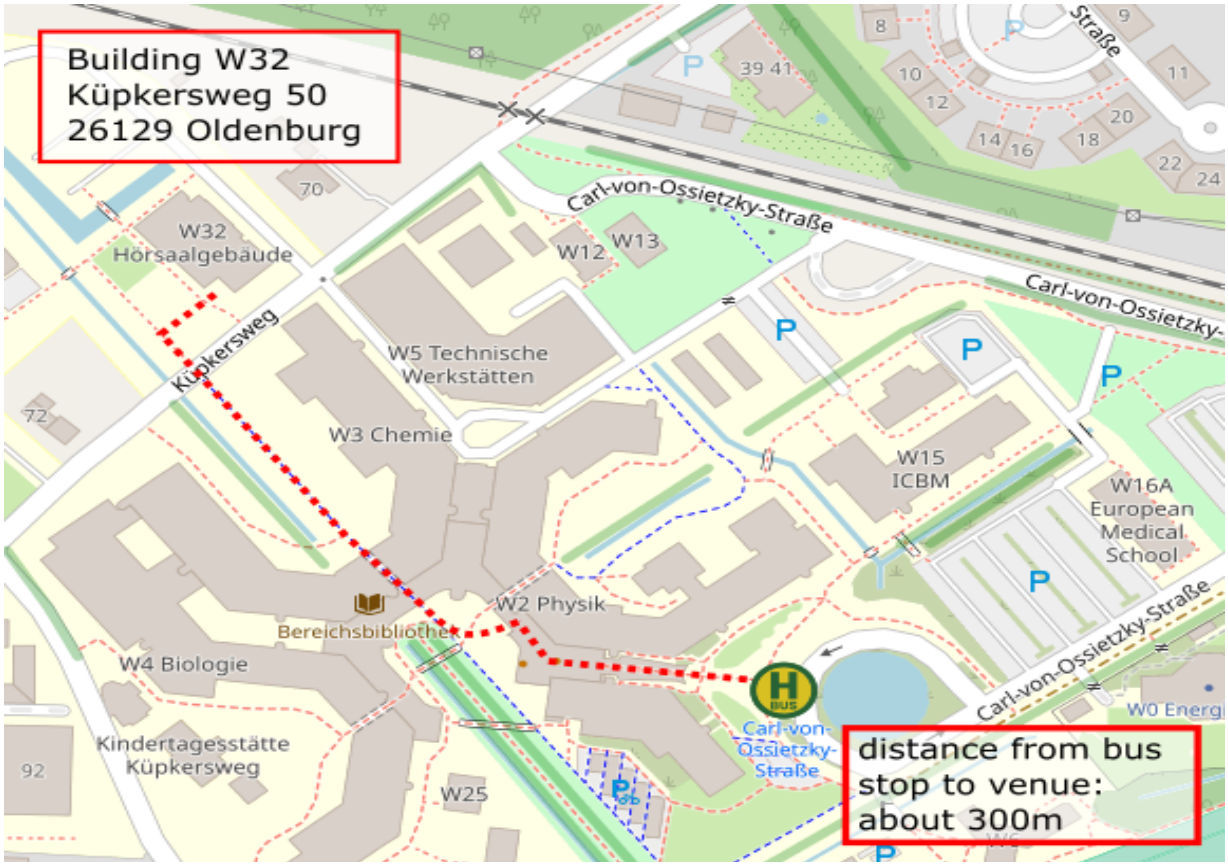
Ultrafast Nano-Optics

Ultrafast Nanoscale Dynamics

ULTRA

Annual Meeting, Oldenburg
25-27 February 2019

Venue:
Campus Wechloy, Building W32
Carl-von-Ossietzky Universität Oldenburg
Küpkersweg 50
26129 Oldenburg



VWG bus ticket is valid on 25th-27th February 2019
Bus 306 runs every 15 min. (:07, :22, :37, :52)
Taxi: 0441-25025, -61061, -63063, -24024

WLAN: QUTIF (no password)
travel time university / main station:
by bus: 13-15 min by taxi: 8-10 min

Monday, 25 February 2019

12:00-13:50	***Registration + refreshments***		
13:50*-14:00*	***Welcome***		
14:00-14:20	LEIN, Manfred *(University of Hannover)* *Overview of the QUTIF Priority Programme*	*Chair: Sascha Schäfer*	1
14:20-15:00	SANTRA, Robin *(DESY, University of Hamburg)* *Molecules at high X-ray intensity: Challenges for theory (invited)*		2
15:00-15:30	***Coffee Break***		
15:30-15:50	ESCHENLOHR, Andrea *(University of Duisburg-Essen)* *Identification and control of ultrafast spin dynamics by femtosecond laser pulses*	*Chair: Christoph Lienau*	3
15:50-16:10	REISLOEHNER, Jan *(University of Jena)* *Characterization of weak ultrashort deep UV pulses using cross-phase modulation scans*		4
16:10-16:30	FENNEL, Thomas *(University of Rostock)* *Coherent control of near-field driven photoemission*		5
16:30-17:00	***Young Scientists' Meeting / Coffee Break***		
17:00-18:20	***Poster Session***		
18:20-19:00	***Transfer to OLs Brauerei*** *(bus departure at bus stop: 6:37 pm)*		
19:00-22:00	***Brewery Visit and Dinner,*** *OLs Brauhaus, Stau 25/27, Entry: Rosenstraße, 26122 Oldenburg*		

Tuesday, 26 February 2019

09:00-09:40	CERULLO, Giulio *(Politecnico di Milano)* *Ultrafast carrier and spin dynamics in two-dimensional semiconductors (invited)*	*Chair: Matthias Wollenhaupt*	6
09:40-10:00	SAALMANN, Ulf *(Max-Planck-Institut PKS Dresden)* *Straightening fluctuating pulses with artificial neural networks*		7
10:00-10:20	OPPERMANN, Florian (*University of Hannover)* *Ionization and dissociation of HeH^+ at long wavelengths*		8
10:20-10:40	BABUSHKIN, Ihar *(University of Hannover)* *Accessing dynamics of optical tunnelling through terahertz emission*		9
10:40-11:10	***Coffee Break***		
11:10-11:30	KULEFF, Alexander *(University of Heidelberg)* *Charge-directed reactivity and its control by short laser pulses*	*Chair: Petra Groß*	10
11:30-11:50	TALLURI, Madhu Trivikram *(Max Born Institute)* *Towards the strong field dissociation of rovibration state-selected H_2^+*		11
11:50-12:10	KARAMATSKOS, Evangelos *(DESY)* *Imaging experiments in the molecular frame*		12
12:10-12:30	JIMÉNEZ GÁLAN, Álvaro *(Max Born Institute)* *Topological strong field physics on sub-laser cycle timescale*		13
12:30-14:00	***Lunch***		
14:00-14:20	ORDOÑEZ, Andrés *(Max Born Institute)* *A unified description of chiral effects within the electric-dipole approximation*	*Chair: Andrea Eschenlohr*	14
14:20-14:40	TRIPPEL, Sebastian *(DESY)* *Creating, imaging, and controlling dynamic chirality induced by molecular rotations: experimental approaches*		15
14:40-15:00	YACHMENEV, Andrey *(DESY)* *Creating, imaging, and controlling dynamic chirality induced by molecular rotations: theoretical approaches and simulations*		16
15:00-15:30	***Coffee Break***		
15:30-16:10	RUPP, Daniela *(Max Born Institute)* *Electron dynamics in single nanodroplets with an intense high harmonic source (invited)*	*Chair: Armin Feist*	17
16:10-16:30	AYUSO, David *(Max Born Institute)* *Ultrafast imaging of molecular chirality: achieving ultimate efficiency in high harmonic generation*		18
16:30-18:20	***Poster Session***		
18:20-19:00	***Transfer to Conference Dinner*** *(bus departure at bus stop: 6:37 pm)*		
19:00-22:00	***Conference Dinner,*** *Restaurant 'Caldero', Markt 23, 26122 Oldenburg*		

Wednesday, 27 February 2019

09:00-09:40	BRIDA, Daniele *(Universitè du Luxembourg)* *Single-cycle autocorrelation in attosecond coherent nanotransport (invited)*	*Chair: Thomas Fennel*	19
09:40-10:00	GEBAUER, Andreas *(University of Bielefeld)* *Equivalence of RABBITT and streaking delays in attosecond time-resolved photoemission at solid surfaces*		20
10:00-10:20	LIU, Qingcao *(LMU)* *All-optical spatio-temporal control of electron emission from SiO_2 nanospheres with femtosecond two-color laser fields*		21
10:20-10:50	***Coffee Break***		
10:50-11:10	AHMADI, Sayed Hamed *(University of Freiburg)* *Time-resolved petahertz spectroscopy*	*Chair: Daniele Brida*	22
11:10-11:30	HEIDENREICH, Andreas *(University of the Basque Country)* *Dopant-induced ignition of helium nanoplasmas*		23
11:30-11:50	WOLLENHAUPT, Matthias *(University of Oldenburg)* *Control of 3D free electron wave packets by bichromatic fields*		24
11:50-12:10	MÜCKE, Oliver (DESY) *SOLSTICE: novel opportunities for advanced quantum materials*		25
12:10-12:30	***Conclusion***		
12:30-13:40	***Lunch and Departure***		
13:00	***Lab Tour***		

Poster Presentations

Perturbative and strong-field control of photoemission from needle tips with a two-color field	Philip Dienstbier
Bichromatic quantum phase measurement of multiple Rydberg dynamics	Kevin Eickhoff
Photoemission time vs. streaking delay in attosecond time-resolved solid state photoemission	Andreas Gebauer
Ultrafast point-projection electron microscopy	Germann Hergert
Phase of continuum-continuum couplings	Anne Harth
Optimization of field-free alignment of molecules for imaging experiments	Evangelos Karamatskos
Odd electron wave packets from cycloidal ultrashort laser fields	Stefanie Kerbstadt
Strong-field dissociation of state-selected H_2^+ (v, J)	Thomas Kubail Kalousdian
Advances with attosecond electron pulse trains in ultrafast transmission electron microscopy	Thomas Rittmann
High-order above-threshold photoemission from nanotips controlled with two-color laser fields	Lennart Seiffert
Petahertz field reconstruction for the investigation of electronic dynamics in nanostructures	Ronak Narendra Shah
Creating, imaging, and controlling dynamic chirality induced by molecular rotations: exp. approaches	Sebastian Trippel
Fragmentation of HeH^+ by intense ultrashort laser pulses	Philipp Wustelt
Laser-dressed current-carrying orbitals in bicircular fields	Xiaosong Zhu

Abstracts

Monday, 25th February 2019

1 ***Overview of the QUTIF Priority Programme***
M. Lein
14:00 - 14:20

2 ***Molecules at high x-ray intensity: Challenges for theory*** *(invited)*
R. Santra
14:20 - 15:00

One of the key opportunities offered by the development of x-ray free-electron lasers is the determination, at atomic resolution, of the three-dimensional structure of biologically relevant macromolecules. The basic idea underlying molecular imaging using x-ray free-electron lasers is the "diffract-and-destroy" concept: If one uses an x-ray pulse that is sufficiently short (on the order of femtoseconds), then in a single shot an x-ray scattering pattern may be obtained that is practically unaffected by atomic displacements triggered by ionization events during the x-ray pulse. What cannot be eliminated in this way is the impact of the electronic damage on the x-ray scattering patterns. Theory, therefore, plays an important role in the development of this new imaging technique: A quantitative understanding is required of the damage processes occurring during the exposure of a molecule to an ultraintense, ultrafast x-ray pulse. In this talk, I will present progress we have made in order to address this challenge. One tool we have developed, XMDYN, is a molecular-dynamics code that utilizes ab-initio atomic electronic-structure information, computed on the fly, within a Monte-Carlo framework. XMDYN has been successfully tested through experiments at LCLS and SACLA. XMDYN is part of a powerful start-to-end simulation framework for single-particle imaging at the European XFEL. Recently, we have taken first steps towards a full ab-initio framework for simulating high-intensity x-ray–matter interactions. Our new XMOLECULE software solves the polyatomic quantum-mechanical electronic-structure problem for every electronic state arising during the exposure of a molecule to a strong x-ray pulse. From this information, electronic transition rates (such as Auger decay rates) are computed on the fly, and the associated rate equations are integrated utilizing a Monte-Carlo method. XMOLECULE played a key role in a recent LCLS experiment on iodomethane, in which hard x-rays focused to a peak intensity exceeding 10^{19} W/cm^2 produced the highest charge states ever formed using light. Not only did XMOLECULE correctly predict the charge-state distribution observed, but it also helped identify a new molecular ionization enhancement mechanism based on intramolecular charge transfer.

3 ***Identification and control of ultrafast spin dynamics by femtosecond laser pulses***
A. Eschenlohr, S. Sharma
15:30 - 15:50

Our project analyzes femtosecond spin dynamics in ferromagnetic solids and at interfaces driven by excitation with intense laser pulses in a close collaboration between experiment and ab-initio theory. We will briefly review our results of the first funding period, in particular the identification of optically induced spin (back-)transfer and spin-orbit coupling mediated ultrafast demagnetization at ferromagnet/paramagnet interfaces, and outline the next steps in achieving control of femtosecond spin dynamics by pulse tailoring.

4 ***Characterization of weak ultrashort deep UV pulses using cross-phase modulation scans***
J. Reisloehner, Ch. Leithold, A.N. Pfeiffer
15:50 - 16:10

Temporal pulse characterization methods can most often not be applied to UV pulses due to the lack of suitable nonlinear crystals and very low pulse energies. Here, a method is introduced for the characterization of two unknown and independent laser pulses. The applicability is broad, but the method is especially useful for pulses in the deep UV, because pulse energies on the picojoule-scale suffice. The basis is a spectral analysis of the two interfering UV pulses, while one of the pulses is phase shifted by an unknown VIS-IR pulse via cross-phase modulation. The pulse retrieval is analytic and the fidelity can be checked by comparing the complex-valued data trace with the retrieved trace.

5 ***Coherent control of near-field driven photoemission***
T. Fennel
16:10 - 16:30

Intense and well-defined ultrashort laser pulses have opened intriguing routes to access ultrafast electron dynamics in various forms of matter. Irrespective of the particular target, the ability to control the laser electric field waveform on a sub-cycle basis or the delay between spectral components of multi-color fields with attosecond resolution is key to observing and steering electronic motion in real time. The dominant processes occurring in nanoscale systems, however, depend strongly on the laser-driven near-fields and can vary fundamentally for different target classes. In this talk I will discuss this aspect for the example of two-color photoemission from dielectric nanospheres and metallic nanotips.

For the example of nanospheres, the opportunities arising from wavelength dependent near-field profiles that result from field propagation will be discussed [1]. The propagation effect is shown to enable directional control of the strong-field photoemission via phase-dependent two-color excitation [2]. In the second part, results on the two-color strong field photoemission from sharp metallic nanotips will be presented. Semiclassical and TDSE simulations, performed for a simplified tip model, show pronounced phase dependent signals with amplitudes similar to recent measurements [3]. The comparison of quantum and semiclassical models is shown to enable one to discriminate the individual impacts of the ionization rate and the electron trajectory on the two color effect found in the electron spectra [4].

1. Süßmann et al., *Field propagation induced directionality of carrier-envelope phase controlled photoemission from nanospheres*, Nat. Commun. **6**, 7944 (2015)
2. Liu et al., in preparation
3. M. Förster et al., *Two-Color Coherent Control of Femtosecond Above-Threshold Photoemission from a Tungsten Nanotip*, Phys. Rev. Lett. **117**, 217601 (2016)
4. Seiffert et al., *High-order above-threshold photoemission from nanotips controlled with two-color laser fields*, J. Phys. B **51**, 134001 (2018)

Tuesday, 26th February 2019

6 ***Ultrafast carrier and spin dynamics in two-dimensional semiconductors*** *(invited)*
G. Cerullo
09:00 - 09:40

Transition Metal Dichalcogenides (TMDs) are emerging materials for electronic, spintronic and quantum information processing applications. When thinned to a single layer (1L), they become direct bandgap two-dimensional semiconductors, with optical response dominated by tightly bound (up to 0.5 eV binding energy) excitons, due to the strong quantum confinement and the reduced Coulomb screening. In addiion, the large spin-orbit coupling lifts the spin degeneracy of the valence and the conduction bands; due to the lack of inversion symmetry, spin and valley indexes are locked and valley-polarized carriers can be excited by circularly polarized light. Finally, layers of different TMDs can be easily integrated vertically by mechanical stacking, forming van der Waals heterostructures with rich physics. In this talk we present results on the non-equilibrium optical response of 1L TMDs investigated by a variety of ultrafast optical spectroscopy techniques. We first study exciton dynamics of 1L-MoS_2 by broadband femtosecond transient absorption, showing that the non-equilibrium optical response is dominated by the renormalisation of both band gap and exciton binding energies caused by photo-excited charge carriers. We then use two-colour helicity-resolved pump-probe spectroscopy in order to disentangle the intervalley and intravalley spin-flip processes of electrons in the conduction band of 1L-WS_2, time resolving the formation of the dark excitons which are responsible for photoluminescence quenching in this material. Finally, we study heterostructures of WSe_2 and $MoSe_2$ and time resolve the sub-picosecond build-up time of the interlayer exciton formed upon injection of a hole from $MoSe_2$ to WSe_2, as well as its temperature-dependent lifetime.

7 ***Straightening fluctuating pulses with artificial neural networks***
U. Saalmann, S. Kumar Giri, J.M. Rost
09:40 - 10:00

Unseeded free-electron laser machines create strongly fluctuating pulses that change from shot to shot. Non-linear processes driven by those pulses depend critically on the actual fluctuation pattern, which is typically not of genuine interest. Here we discuss an attempt to "straighten" those pulses a posteriori. By means of artificial neural networks we map photo-electron spectra from fluctuating pulses to those from idealized ones.

8 ***Ionization and dissociation of HeH^+ at long wavelengths***
F. Oppermann
10:00 - 10:20

The helium hydride molecular ion is one of the simplest heteronuclear benchmark systems in the research area of strong-field laser matter interaction. At infrared wavelengths, fragmentation without electronic excitation (dissociation) and ionization followed by Coulomb explosion (ionization) are of similar importance. In an ω-2ω laser pulse, the dominating fragmentation pathway can be efficiently controlled by changing the relative delay of the two pulses on a sub-cycle timescale. The key ingredient is the asymmetry of the molecule and the kinematically complete measurement of the fragments. We present results from quantum mechanical simulations and an outlook for planned experiments.

9 ***Accessing dynamics of optical tunneling through terahertz emission***
I. Babushkin
10:20 - 10:40

Terahertz and higher order Brunel harmonics appear during ionization of electron and subsequent acceleration, 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 lowest Brunel harmonics, the signatures of the ionization dynamics can be still found in the Brunel harmonic spectrum.

10 ***Charge-directed reactivity and its control by short laser pulses***
A. Kuleff
11:10 - 11:30

Exposing molecules to ultrashort laser pulses can trigger pure electron dynamics in the excited or ionized system. In the case of ionization, these dynamics may manifest as an ultrafast migration of the initially created hole-charge throughout the ionized molecule and were termed charge migration. Charge migration can be solely driven by electron correlation and appeared to be a rich phenomenon with many facets that are rather characteristic of the molecule studied. Importantly, due to the coupling between the electronic and the nuclear motion, the control over the pure electron dynamics offers the extremely interesting possibility to steer the succeeding chemical reactivity by predetermining the reaction outcome at a very early stage. This is the paradigm of the emerging field of “attochemistry”. It will be shown how by appropriately tailored ultrashort laser pulses one can control the ultrafast charge migration in experimentally interesting molecules. Full quantum electron-nuclear dynamics calculations of the charge dynamics initiated by ionization will be presented and the possibilities to realize the dream of attochemistry, namely achieving control of chemical reactions by manipulation of electron coherences, will be discussed.

11 ***Towards the strong field dissociation of rovibration state-selected H_2^+***
M.T. Talluri, T.K. Kalousdian, N. Zhavoronkov, M.J.J. Vrakking
11:30 - 11:50

The Hydrogen molecular ion is a benchmark system for understanding the strong-field interaction of diatomic molecules. Novel light-induced phenomena like bond-softening [1, 2] and bond-hardening [3] were observed in the presence of intense laser fields with peak intensities beyond 10^{12} W/cm2. These observations were successfully explained in the Floquet formalism via formation of an avoided crossing between the adiabatic dressed potential surfaces of 1sσ and 2pσ states. Nonetheless, the role of molecular rotation has been overlooked in the strong-field dissociation studies. Theoretically, it has been shown that the molecular rotation, with respect to the laser polarization axis, provides an additional degree of freedom. This creates a light-induced conical intersection (LICI), where the two potential surfaces are degenerate–as can naturally occur in polyatomic molecules [4, 5]. The LICIs can be further explored by studying the dissociation of rovibration state-selected H_2^+ using photo-fragment angular distribution (PAD) measurements. This will provide insight into the geometric aspects of the potential energy surfaces near CIs, i.e. Berry phase effect on dissociating nuclear wavepackets [6]. For the unambiguous observation of these novel features, we are developing an experiment which involves the multiple stages: creation of narrow-band XUV source, at 75-80nm with $\delta E \sim$ 3meV; production of state-selected $H_2^+(v,J)$, using the pulse-field-ionization (PFI-ZEKE) method; building a beam-line for a two-color experiments probing strong-field dissociation; and a velocity-map imaging (VMI) detector for the PAD studies.

1. P. Backusbaum et al., PRL, 64, 1883 (1990).
2. K. Sandig, H. Figger, and T. Hänsch, PRL, 85, 4876 (2000).
3. A. Zavriyev et al., PRL, 70, 1077 (1981).
4. M. Sindelka, N. Moiseyev, and L.S. Cederbaum, JPB, 44, 045603 (2011).
5. A. Natan et al., PRL, 116, 143004 (2016).
6. F. Bouakline, JPCL, 9, 2271 (2018).

12 ***Imaging experiments in the molecular frame***
E. Karamatskos, G. Goldsztejn, S. Raabe, P. Stammer, T. Mullins, A.Trabattoni, R.R. Johansen, K. Dlugolecki, H. Stapelfeldt, S. Trippel, M.J.J. Vrakking, J. Küpper, A. Rouzée
11:50 - 12:10

Imaging the ultrafast dynamics of molecules requires experimental methods that offer at the same time atomic spatial and ideally sub-femtosecond temporal resolution. The possibility to prepare cold, controlled molecular samples in the gas phase, combined with elaborate methods to fix the molecules in space, are important prerequisites to image molecular dynamics directly in the molecule-fixed frame. Combined with the aforementioned prerequisites, the laser-induced electron diffraction (LIED) method has come up as a strong candidate towards recording the molecular movie. By employing the LIED method, the equilibrium molecular structure of OCS could be retrieved with atomic resolution. Molecular-frame angularly-resolved photoelectron spectra (MF-ARPES) show clear signatures of a dependence of the ionization and rescattering dynamics on the molecular frame. Furthermore, strong-field photoelectron holography was observed, yielding different interference patterns, depending on the orientation of the molecules with respect to the laser polarization of the ionizing field. A discussion of our recent experimental results will be presented.

13 ***Topological strong field physics on sub-laser cycle timescale***
Á. Jiménez Galán, R. Silva, B. Amorim, O. Smirnova, M. Ivanov
12:10 - 12:30

Quantum materials encompass a rich variety of systems with fascinating features. One of them is the topological phase transition, upon which an insulator becomes conducting, supporting robust currents around the insulator's edges [1,2]. "Protected" by the topological invariants of the bulk, the chiral edge states are robust to perturbations, making them appealing for applications, for example in dissipationless devices or topologically robust semiconductors. However, and surprisingly, the ultrafast dynamics of non-equilibrium electronic response to intense optical fields in these materials has remained virtually unexplored. Yet, understanding these dynamics is not only fundamentally interesting. It is also crucial for light-wave electronics in topological materials.

Attosecond science has made major progress in understanding ultrafast electron dynamics in solids [3,4]. Yet, so far it has mostly focused on the role of the band structure. The role of the topological properties, such as the Berry curvature and the topological invariants of condensed matter systems, on the attosecond dynamics of electronic response has been hardly explored. Does the highly non-equilibrium electron dynamics in the bulk, driven by a strong laser field, encode the topological properties on the sub-laser cycle time-scale? How do the Berry curvature and the Chern number affect the first step in the nonlinear response – the field-driven injection of electrons across the bandgap?

In this presentation, I will answer these questions using the paradigmatic example of the topological insulator, the Haldane system [5]. I will show how the topological state of the system controls its attosecond, highly non-equilibrium electronic response to strong low-frequency laser fields, in bulk. Topological effects can be identified on the directionality and the attosecond timing of an electron current injected into the conduction band by the oscillating electric field of an intense light pulse.

I will further show that the highly nonlinear optical response to strong fields, the high harmonic emission, displays topologically-dependent attosecond delays, and that the helicities of the emitted harmonics can record the phase diagram of the system and its topological invariants [6].

1. M.Z. Hasan and C.L. Kane, "*Colloquium: Topological insulators*", Rev. Mod. Phys. 82 3045 (2010)
2. X.L. Qi and S.C. Zhang, "*Topological insulators and semiconductors*", Rev. Mod. Phys. 83 1057 (2011)
3. G. Vampa et al., "*Linking high harmonics from gases and solids*", Nature 522 462 (2015)
4. S.Y. Kruchinin et al., "*Colloquium: Strong-field phenomena in periodic systems*", Rev. Mod. Phys. 90 021002 (2018)
5. F.D.M. Haldane, "*Model for a quantum Hall effect without Landau levels*", Phys. Rev. Lett. 61 2015 (1988)
6. R.E.F. Silva et al., "*Topological strong field physics on sub-laser cycle time scale*", arXiv:1806.11232.

14 ***A unified description of chiral effects within the electric-dipole approximation***
A. Ordoñez, O. Smirnova
14:00 - 14:20

Unlike standard circular dichroism, which relies on the magnetic-dipole interaction and yields a tiny chiral response, effects relying purely on the electric-dipole interaction consistently yield a huge chiral response. Based on our recent work [1], we provide a unified and very compact formalism for the description of several methods of chiral discrimination based on electric-dipole interaction, including photoelectron circular dichroism (PECD) [2], enantiosensitive microwave spectroscopy [3], photoexcitation circular dichroism [1], and photoelectron photoexcitation circular dichroism [1]. We show that, although the physics underlying the emergence of the chiral response is evidently different in all these methods, the pseudoscalar observable responsible for the chiral signal can always be presented in a standard form: a triple product of three vectors. Beyond offering a unified picture of the above mentioned effects and a generalization of all effects to arbitrary polarizations of the laser field [4], we present new compact formulas derived using our formalism describing the effect of pre-alignment of the molecular ensemble for PECD [5]. Furthermore, we show that a Berry-curvature-like field is responsible for PECD in isotropic samples [5].

1. S. Beaulieu et al., *Photoexcitation circular dichroism in chiral molecules*, Nature Physics 14, 484-489 (2018)
2. B. Ritchie, *Theory of the angular distribution of photoelectrons ejected from optically active molecules and molecular negative ions,* Physical Review A 13, 1411 (1976)
3. Patterson et al., *Enantiomer-specific detection of chiral molecules via microwave spectroscopy*, Nature 497, 475 (2013)
4. A.F. Ordonez and O. Smirnova, *Generalized perspective on chiral measurements without magnetic interactions*, arXiv:1802.06540 (2018)
5. A.F. Ordonez and O. Smirnova, *Propensity rules in photoelectron circular dichroism in chiral molecules II: General picture*, arXiv:1806.09050 (2018)

15 ***Creating, imaging, and controlling dynamic chirality induced by molecular rotations: experimental approaches***
S. Trippel
14:20 - 14:40

Our ideas to experimentally investigate rotationally induced chirality (RIC) will be presented. These include the basic principles, the planned implementation of a pulse shaper in our existing control-laser system, and simulations concerning novel velocity map imaging spectrometer which will allow us to directly verify RIC. Furthermore, we will present our results on the field-free 3D alignment and mixed field orientation of indole, an asymmetric top molecule, by shaped laser pulses. The degree of alignment and orientation is quantified and analyzed by the comparison of the observed rotational revival structure with theoretical predictions.

16 ***Creating, imaging, and controlling dynamic chirality induced by molecular rotations: theoretical approaches and simulations***
A. Yachmenev
14:40-15:00

We will present results of our recent simulations of the effect of rotationally induced chirality (RIC). This phenomenon emerges when a rigid symmetric molecule spins so fast around one of its bonds that it loses its symmetry. The clock-wise and anti-clock-wise rotating molecules form two distinct mirror versions of the molecule; in this sense the chirality is dynamically induced through extreme rotational excitations. The RIC effect is closely connected to the effect of rotational energy clustering exhibited by a class of polyatomic molecules with strong local mode vibrational character. A typical local-mode molecule is PH_3: it exhibits small couplings between different vibrational modes and bond angles very close to 90 degrees. Recently, in our group we have performed first proof-of-principle theoretical investigation of the RIC in PH_3, where we have shown that sufficient amount of molecules can be excited into rotational cluster states using the optical centrifuge. We have demonstrated that application of a strong dc electric field during the centrifuge pulse favors one rotating enantiomeric form over the other, creating dynamically chiral molecules with permanently oriented rotational angular momentum. Our main theoretical endeavors are now directed into optimizing the laser pulse parameters to increase the efficiency of rotational excitation and simulations of techniques for imaging of chirality, such photoelectron circular dichroism. Essential efforts of the theoretical research is also devoted to finding local-mode molecular candidates best suitable for experiment.

17 ***Diffraction imaging of ultrafast electron dynamics in single nanodroplets with an intense high harmonic source*** *(invited)*
D. Rupp, J. Jordan, K. Kolatzki, B. Kruse, B. Langbehn, N. Monserud, M. Sauppe, B. Schütte, R. Tanyag, A. Ulmer, M.J.J. Vrakking, J. Zimmermann, T. Möller, T. Fennel, A. Rouzee
15:30 - 16:10

Via diffraction imaging of single nanoparticles in free flight with XUV and X-ray free-electron lasers (FELs), we can explore light-induced excitation and ionization dynamics on the nanoscale with both high spatial and temporal resolution. From the measured diffraction patterns, formed by the interference of elastically scattered photons, the nanoparticle's structure can be determined. This allows to study such fragile and short-lived specimen as superfluid helium nanodroplets. In pump-probe configurations, structural changes, like e.g. ultrafast melting, can be induced and observed. Even faster electron dynamics also change the scattering response and can therefore in principle be traced by diffraction imaging.

By tuning the FEL wavelength to electronic resonances of the particle's material, we were able to trace characteristic features of the formation and evolution of a nanoplasma in the diffraction images. With the 100 femtosecond FEL pulses, it was however not possible to resolve the dynamics in time. While sub-femtosecond pulses are currently under development at FELs, already today, isolated attosecond XUV pulses and attosecond pulse trains can be generated with high-harmonic-generation (HHG) sources, however being typically weaker than FELs by orders of magnitude. In a favorable combination of experimental parameters, we could recently demonstrate single-particle single-shot diffractive imaging of individual helium nanodroplets with high-intensity HHG pulses (Rupp et al., Nature Communications 8, 493 (2017)). In a subsequent time-resolved approach using 35 fs IR and XUV pulses, we observed an ultrafast bleaching effect when both pulses overlapped. The results of these experiments, combined with the rapid development of today's XUV and X-ray lasers, provide a promising pathway to map excitation and charge separation processes in nanoscale matter on their natural time scale.

18 ***Ultrafast imaging of molecular chirality: achieving ultimate efficiency in high harmonic generation***
D. Ayuso
16:10 - 16:30

High harmonic generation records the electronic response to light in atoms, molecules and solids with sub-femtosecond temporal resolution. In turn, this response encodes properties of the interrogated quantum system, exposing such intriguing phenomena as topological phases of matter, magnetism and chirality. Since the XIX-th century, chiral response to light has been thought to require the interplay of the electric and magnetic components of the light field. Recent studies demonstrated that it can be triggered exclusively by the electric field, with efficiency surpassing the accepted standards by orders of magnitude. Here we show how the microscopic chiral response during high harmonic generation in randomly oriented chiral molecules can be mapped into a giant background free macroscopic chiral signal of the medium, discriminating between left-handed and right-handed molecules at the level of 200% of chiral response, manipulated selectively based on the molecular handedness, and fully characterized on the attosecond time scale. Our findings open the way to a new, efficient, ultrafast probe of chiral structure and dynamics applicable to gases, liquids and solids.

arXiv:1809.01632

Wednesday, 27th February 2019

19 ***Single-cycle autocorrelation in attosecond coherent nanotransport*** *(invited)*
D. Brida
09:00 - 09:40

We performed interferometric autocorrelations that exploit the electric currents coherently driven by single-cycle pulses at the gap of a single nanodevice. These measurements are performed in an extremely nonlinear regime that is achieved even at the minute pulse energies of few pJ.

20 ***Equivalence of RABBITT and streaking delays in attosecond time-resolved photoemission at solid surfaces***
A. Gebauer, W. Enns, B. Stadtmüller, M. Aeschlimann, W. Pfeiffer
09:40 - 10:00

The availability of attosecond extreme ultraviolet (EUV) pulses enabled access to electron dynamics at solid surfaces on their natural time scales. Via attosecond-time-resolved photoelectron spectroscopy the dynamics of the photoelectric effect in solid state systems can be investigated. We provide a comparison of delay information accessible by the two most important techniques attosecond streaking spectroscopy and Reconstruction of Attosecond Beating By Interference of Two-photon Transitions (RABBITT) at solid surfaces, respectively. The analysis is based on simulated time-resolved photoemission spectra obtained by solving the time-dependent Schrödinger equation in a single active electron approximation. The model potentials used here have been successfully applied to explain experimental data recently. We show a continuous transition from the few-cycle RABBITT regime to the streaking regime as two special cases of laser-assisted photoemission. The absolute delay times obtained by both methods agree with each other within the uncertainty limits and coincide with the classical time-of-flight for an electron propagating from the emitter atom to the bulk-vacuum interface for kinetic energies >10eV.

21 ***All-optical spatio-temporal control of electron emission from SiO_2 nanospheres with femtosecond two-color laser fields***
Q. Liu, Zherebtsov, L. Seiffert, M. Gallei, T. Fennel, M.F. Kling
10:00 - 10:20

Field localization by nanostructures illuminated with laser pulses of well-defined waveform enables spatio-temporal tailoring of the near-fields for sub-cycle control of electron dynamics at the nanoscale. Here, we apply intense linearly polarized two-color laser pulses for all-optical control of the electron emission from SiO_2 nanoparticles. For the size regime where light propagation effects become important we demonstrate the possibility to control the preferential emission angle of the highest energy electrons by varying the relative phase of the two-color field. Trajectory based semi-classical simulations show that for the investigated nanoparticle size range the directional switching of the fastest electrons can be attributed to the two-color control of electron trajectories, while the accompanied modification of the spatial distribution of the ionization rate on the nanoparticle surface has only a minor effect.

22 ***Time-resolved petahertz spectroscopy***
S.H. Ahmadi
10:50 - 11:10

Subcycle sensitivity in ultrafast spectroscopy requires precise knowledge of the optical field that induces the light-matter interactions at the fundamental level. It implies the knowledge of the time-dependent direction and magnitude of the electric filed E(t) for the entire duration of the pulse. Here, the complete reconstruction of the electric field of Petahertz (visible–infrared) pulses with energy as low as a few tens of nanojoules is demonstrated. The technique combines extreme ultraviolet interferometry with the generation of isolated attosecond pulses in order to reconstruct the instantaneous electric field vector direction and magnitude with an arbitrary time-dependent polarization state. This approach reveals the possibility to characterize completely the electric field of the pulses typically used in visible pump–probe spectroscopy. It also offers the possibility to measure the small differences imprinted on the electric field of a weak probe pulse interacting with a material with an ongoing electron dynamic.

23 ***Dopant-induced ignition of helium nanoplasmas***
A. Heidenreich, M. Mudrich, D. Schomas
11:10 - 11:30

Helium (He) nanodroplets irradiated by intense near-infrared laser pulses form a nanoplasma by avalanche-like electron impact ionizations even at lower laser intensities where He is not directly field ionized, provided that the droplets contain a few dopant atoms which provide seed electrons for the electron impact ionization avalanche. Using molecular dynamics simulations, we elucidate the mechanism which induces ionization avalanches, termed ignition, in He nanodroplets doped with small calcium (Ca_n), xenon (Xe_n) and mixed calcium-xenon (Ca_mXe_n) clusters. We find that the partial loss of seed electrons from the activated droplets starkly assists ignition, as the Coulomb barrier for ionization of He is lowered by the electric field of the dopant cations, and this deshielding of the cation charges enhances their electric field. In addition, the dopant ions assist the acceleration of the seed electrons ("powered flyby") by the laser field, supporting electron impact ionizations of He and also causing electron loss by catapulting electrons away.

24 ***Control of 3D free electron wave packets by bichromatic fields***
M. Wollenhaupt
11:30 - 11:50

Three-dimensional free electron wave packets with arbitrary rotational symmetry are created by multiphoton ionization of atoms with polarization-tailored laser fields and manipulated using optical phases such as the CEP and relative phases. In the experiment, we combine advanced supercontinuum pulse shaping with high-resolution photoelectron tomography. We employ a 4f polarization pulse shaper to sculpture bichromatic fields from a CEP-stable over-octave spanning white light supercontinuum by spectral amplitude and phase modulation. The experimental results show that multiphoton ionization of potassium atoms with a single-color sequence of counterrotating circularly polarized (CRCP) femtosecond laser pulses produces vortex-shaped photoelectron momentum distributions with C_6 or C_8 rotational symmetry. In contrast, bichromatic CEP-stable polarization-tailored counter- and corotating (COCP) femtosecond laser pulses generate C_7 rotationally symmetric and asymmetric momentum distributions. Our results reveal that in the multiphoton regime the symmetry of the free electron wave packets is not fully determined by the field symmetry, but completely described by the quantum interference of states with different angular momenta.

25 ***SOLSTICE: novel opportunities for advanced quantum materials***
O. Mücke, N. Klemke, H. Huang, N. Tancogne-Dejean, A. Rubio, and F.X. Kärtner
11:50 - 12:10

Lightwave-driven electronic dynamics occurring on sub-optical-cycle time scales in condensed matter is a fascinating frontier of attosecond science originally studied in atoms and molecules. Within the SOLSTICE-2 project, we aim to study solids irradiated by strong THz and tailored IR CEP-stable optical waveforms. In particular, investigating high-harmonic generation (HHG), we want to elucidate in greater depth the physical similarities and differences compared to the corresponding process in atomic and molecular gases. To this end, we want to synthesize the perfect wave for atomic-like HHG from 2D materials and compare it to the gas-phase counterpart. Furthermore, new opportunities of THz-dressing-based symmetry control can be explored. We plan to extend HHG from semiconductors and insulators to more complex solids such as strongly correlated materials and topological insulators. Our research thus aims to break new ground in combining strong-field attoscience and Mott-Hubbard physics. Polarization-state-resolved high-harmonic spectroscopy [1] will open up new avenues in ultrafast spectroscopy of quantum materials.

1. N. Klemke *et al.*, "*Polarization-state-resolved high-harmonic spectroscopy of solids*," arXiv:1805.10453 [physics.optics].