

Oral abstracts for atom2016

Sample fragmentation and delivery in single particle Coherent Diffractive Imaging experiments

Andreasson, Jakob

(ELI Beamlines, Institute of Physics, Prague, Czech Republic)

The presentation covers the interlinked topics of sample fragmentation and sample delivery in single particle Coherent Diffractive Imaging (CDI) experiments at X-ray Free Electron Laser (FEL) sources. Experimental results from ion Time of Flight mass spectrometry and ion drift measurements at the FLASH and LCLS FELs on bio-particles are reviewed together with supporting simulations. Emphasis is on the results from the first fragmentation experiments on bio-particles ranging between 15-250 nm in the hard X-ray regime. In these experiments ion drift measurements were combined with single particle X-ray diffraction to provide an extended picture of the interaction dynamics at X-rays intensities from 10^{16} to 10^{19} W/cm². CDI is used to determine the X-ray intensity on the sample and sample size on individual hits and through the kinetic energy distribution of the accelerated ions we can identify both the Coulomb and thermal part of the fragmentation for every single particle. Furthermore, an ultrafast hit finder for on-line data reduction at European XFEL that has been developed from the work on detecting the fastest protons from exploding bio-samples is introduced and we suggest a concept for a hit finder for bio-samples in a water jet. Finally the presentation introduces the concepts of aerosol sample delivery for single particle CDI, reviews recent advances and discusses remaining challenges.

Towards attosecond science at the nano-scale

Arnold, Cord

(Lund University, Faculty of Engineering, Atomic Physics, Sweden)

Numerical approaches to tunneling times in strong-field ionization

Bauke, Heiko

(MPI für Kernphysik, Theory Division, Heidelberg,)

Ultrafast optical tuning of exchange interaction and dynamical quantum phase transitions in spin systems

Berakdar, Jamal

(MLU Halle-Wittenberg, Physics, Theory, Halle, Germany)

The exchange interaction is the central microscopic quantity for the underlying mechanism of magnetically ordered systems [1]. As it is determined by the overlap of relevant orbitals that can be excited swiftly by appropriate laser pulses, one may envision controlling magnetic phases optically. Thermodynamic phases are however macroscopically established and a transition between them is inherently slow. Results of two studies will be presented related to the ultrafast modulation of the indirect exchange interaction in low-dimensional system, and to the onset of a new type of fast dynamical phase transitions in oxide-based helical spin systems when irradiated by fs, appropriately tailored pulses [2].

[1] J. M. D. Coey Magnetism and Magnetic Materials (Cambridge Uni Press, 2010).

[2] Azimi et al. Phys. Rev. B 94, 064423 (2016)

Attosecond soft x-ray spectroscopy in condensed matter

Biegert, Jens

(ICFO - The Institute of Photonic Sciences, Attoscience and Ultrafast Optics, Castelldefels, Spain)

Oral abstracts for atom2016

High harmonic Generation spectroscopy of quantum non-equilibrium thermodynamics

Bondar, Denys

(Princeton University, Department of Chemistry, Princeton, USA)

Attosecond condensed matter dynamics - unraveling the difference between semiconductors and dielectrics

Brabec, Thomas

(University of Ottawa, Department of Physics and Astronomy, Ottawa, Canada)

A new theoretical approach for accurate multi-electron dynamics in strong fields: the time-dependent two-particle reduced density matrix method

Brezinova, Iva

(Vienna University of Technology, Institute for Theoretical Physics, Vienna, Austria)

Strong field phenomena are often described within the single-active electron approximation which usually is able to pin down the essential physics of the underlying effects. Correlations, while omnipresent in multi-electron systems, are difficult to account for. Time-dependent density functional theory (TDDFT), for example, while applicable to large systems, faces the difficulty that electronic exchange and correlation functionals which include dynamical correlation and memory effects are largely unknown. Wavefunction based methods such as the multi-configurational time-dependent Hartree-Fock (MCTDHF), while in principle exact, scale exponentially with particle number and are thus limited to small systems.

In this talk a new approach will be presented which aims at bridging the gap between TDDFT and N-particle wavefunction based methods by using the two-particle reduced density matrix. The use of the wavefunction is completely avoided. We show first benchmark calculations for high-harmonic generation in multi-electron atoms and demonstrate the high accuracy of the new method. Marked differences between the fully correlated spectrum and the spectrum obtained from TDDFT and time-dependent Hartree-Fock are found.

The crossover from Multiphoton to tunnelling ionisation in laser-atom interaction.

Briggs, John

(Albert-Ludwigs University of Freiburg, Physikalisches Institut, Freiburg, Germany)

A simple theory is presented to describe the ionisation of a hydrogen atom by a moderately strong laser field. The ionisation is considered to occur in two steps. The first is the polarisation of the atom by the virtual absorption of one to several photons leading to an off-shell excited atomic state. This step is described in perturbation theory. The second step is the tunnelling of the virtually excited electron into the continuum. Depending upon the size of the Keldysh parameter, this theory describes a continuous crossover from predominant multi photon ionisation to the limit of predominant direct tunnelling out of the ground state.

Attosecond delays in valence band photoemission from metal

Castiglioni, Luca

(Universität Zürich, Physik-Institut, Zürich, Switzerland)

Oral abstracts for atom2016

Ultrafast tunneling states of a many-electron atom: From weak to strong fields

Chen, Yi-Jen

(Center for Free-Electron Science, DESY, Hamburg, Germany)

Strong-field probing of atomic and molecular dynamics

DiMauro, Lou

(The Ohio State University, The Ohio State University, Physics, Columbus, USA)

Multidimensional Attosecond Spectroscopy

Dudovich, Nirit

(Weizmann Institute of Science, Weizmann Institute of Science, Physics of complex systems, Rehovot, Israel)

Performing time-resolved measurements with attosecond precision is a significant challenge. Currently, two main approaches have been successfully demonstrated. The first approach, Attosecond Pump-Probe Spectroscopy, applies an attosecond pulse to initiate or probe a fast-evolving process. An alternative approach, Attosecond Self-Imaging, applies the attosecond production process, the HHG mechanism, to perform the measurement. HHG carries the potential of combining sub-Ångstrom spatial and attosecond temporal resolution of electronic structures and dynamics in molecules. This idea has generated an increasing number of experiments in both atomic and molecular systems that resolve the symmetry of the contributing orbitals and the inter-nuclear separation with high contrast.

Although attosecond spectroscopy holds great promise for both measurement and control of matter, the understanding and implementation of most processes pose significant challenges. The extreme nonlinear nature of the interaction offers numerous channels, strongly coupled by the strong laser field, in which electronic dynamics can evolve.

In the talk I will describe a new measurement scheme integrates the two main branches in attosecond spectroscopy – the pump-probe scheme with HHG spectroscopy. In this scheme the initial excitation is induced by an attosecond XUV pulse. Within one optical period an electron is ionized by the XUV pulse via photoionization, accelerated by the strong laser field, and re-collides with the parent ion, consequently emitting XUV radiation. Integration of attosecond pulses with recollision combines the simplicity of single XUV photon excitation with the accuracy provided by the recollision process. This new mechanism enables decoupling of ionization and propagation steps and control over initial energy conditions for the recollision process. Most importantly, the XUV excitation holds the potential for providing access to deeply located inner orbitals, thus allowing a wide range of multielectron phenomena.

Ultrafast high harmonic spectroscopy of dielectric breakdown in strongly correlated systems

Ferreira da Silva, Rui Emanuel

(Max Born Institut, Berlin, Germany)

We investigate theoretically the high harmonic generation process in the 1D Fermi-Hubbard model in the quantum tunneling regime with THz fields. This is the first theoretical study of High Harmonic generation in solids where electron correlation effects are explicitly taken into account. We observe a distinct mechanism for the harmonic emission from the one observed in semiconductors that relies on the production of doublon-hole pairs. An insulator-to-metal transition is observed and we can time this phase transition using the time profile of the harmonic emission. This opens the window for the investigation of ultrafast phase transitions in the condensed phase using high harmonic spectroscopy.

Oral abstracts for atom2016

Ultrafast nanophotonic phenomena in nanographenes

Garcia de Abajo, Javier

(ICFO - The Institute of Photonic Sciences, Nanophotonics Theory Group, Castelldefels (Barcelona), Spain)

Graphene exhibits unique electrical, optical, and thermal properties that are a continuous source of unexpected phenomena [1-4]. In this presentation, I will review recent advances in the field of graphene nanophotonics, including the following results obtained by my group: the design and realistic description of a new class of random metamaterials incorporating optical gain and displaying a varied photonic behaviour ranging from stable lasing to chaotic regimes [1]; a new strategy for molecular sensing that relies on the strong plasmon-driven nonlinearity of nanographenes [2]; a scenario in which radiative heat transfer is the fastest cooling mechanism, even beating relaxation to phonons [3]; and the generation of intense high harmonics from graphene, assisted by its plasmons [4]. These results constitute examples that extend ultrafast optical phenomena in new directions with strong potential for technological applications.

[1] A. Marini and F. J. García de Abajo, "Graphene-based active random metamaterial for cavity-free lasing," *Phys. Rev. Lett.* 116, 217401 (2016).

[2] R. Yu, J. D. Cox, and F. J. García de Abajo, "Nonlinear plasmonic sensing with nanographene," *Phys. Rev. Lett.* 117, 123904 (2016).

[3] R. Yu, A. Manjavacas, and F. J. García de Abajo, "Ultrafast radiative heat transfer," arXiv:1608.05767.

[4] J. D. Cox, A. Marini, and F. J. García de Abajo, "Plasmon-assisted high-harmonic generation in graphene," arXiv:1609.09794.

Assessment of the spatial resolution of Tip-Enhanced Raman Spectroscopy

Gräfe, Stefanie

(Friedrich-Schiller-Universität Jena, Institute for Physical Chemistry, Institut fuer Physikalische Chemie, Jena, Germany)

Experimental evidence of extremely high spatial resolution of tip-enhanced Raman scattering (TERS) has been recently demonstrated. In this talk, I will summarize several of these experimental results and present results of our full quantum chemical description (at the density functional level of theory) of the non-resonant chemical effects on the Raman spectrum of a molecule mapped by a tip, modeled as a single silver atom or a small silver cluster. Pronounced changes in the Raman pattern and its intensities can be seen, depending on the conformation of the nanoparticle–substrate system, concluding that the spatial resolution of the chemical contribution of TERS can be in the sub-nm range.

Physics with strong fields at needle tips and light-field driven currents in graphene

Hommelhoff, Peter

(Universität Erlangen-Nürnberg, Laser Physics, Erlangen,)

After a brief review of the hallmarks of attosecond physics observed at solids, we will show how our strong-field physics control over electrons in the near-field of needle tips can be utilized to obtain insight knowledge of the nanoscale near-field itself. This not only allows us to measure directly the magnitude of field enhancement effects but also its phase -- the latter in conjunction with an attosecond XUV drive pulses. Furthermore, we show that we can map the focal phase of broadband focused laser pulses with nanometer spatial and attosecond temporal resolution. With two-color driving fields we have observed quantum path interference in electron emission with a visibility of more than 98%. In the second part of the talk we will discuss strong-field excitations of electrons in the 2-dimensional material graphene. We observe an intriguing interplay between intraband and interband dynamics. Because of the Dirac-cone bandstructure, an apparent bandgap always exists that acts as a 50/50 beamsplitter: part of the optically driven electron wavefunction diabatically transitions to the conduction band, part of it adiabatically stays

Oral abstracts for atom2016

in the valence band. Within an optical cycle, this transition is crossed twice, so that effectively we have set up an electron matter wave interferometer, also known as Landau-Zener-Stückelberg interferometer. We will show that with the help of the carrier-envelope phase we can determine the relative phase between the two arms and can so control the conduction band population. Similarities to strong-field physics in atoms will be discussed.

A frequency comb transferred by surface plasmons

Kim, Seungchul

(Pusan National University, Pusan, Korea, Republic of)

Frequency comb, millions of narrow-linewidth optical modes referenced to an atomic clock, has shown remarkable potential in time/frequency metrology, atomic quantum physics and precision LIDARs. Applications have extended to coherent nonlinear Raman spectroscopy of molecules and quantum metrology for entangled atomic qubits.

Frequency comb will create novel possibilities in nano-photonics and plasmonics; however, its interrelation with surface plasmons is unexplored despite the important role that plasmonics play in ultrafast physics as well as nonlinear spectroscopy and quantum optics through the manipulation of light in a subwavelength scale. In this study, we investigated that frequency comb can be transformed to a form of plasmonic comb in plasmonic nanostructures and reverted to the original frequency comb without noticeable degradation of less than 6.51×10^{-19} in absolute frequency position, 2.92×10^{-19} in stability and 1 Hz in linewidth. The results indicate that the superior performance of a well-defined frequency comb can be applied to ultrafast nanoplasmonic spectroscopy, quantum metrology and subwavelength photonic circuits.

Attosecond streaking spectroscopy on nanostructures

Kling, Matthias

(Ludwig-Maximilians-Universität München, Max Planck Institute of Quantum Optics, Physics Department, Garching, Germany)

Ionisation and recombination of cluster in strong fields

Krishnamurthy, Manchikanti

(Tata Institute of Fundamental Research, Dept of Nuclear and Atomic Physics, Mumbai, India)

Intense ultrashort infrared light pulses are known for strong ionization of the inter gas clusters. While a large number of experimental features have been understood, charge resolved ions spectrometry has brought in some novel features. I will talk about some of the experiments that reveal some novel feature of ionization and charge reduction that occur in inert gas clusters.

Small atomic systems in He nanodroplets

Krishnan, Siva Rama

(Indian Institute of Technology Madras, Department of Physics, Chennai, India)

Recently, we have been involved in a series of collaborative experimental campaigns at the Elettra synchrotron to investigate He nanodroplets hosting small atomic systems with particular attention to photo-activating the He matrix and following the few-electron dynamics that ensue thereafter. Although, these host droplets have been hitherto hailed as ideal personalized cryostats for spectroscopy of dopants in or on them, our contrary approach has led to insights on the effects and interaction that environments have on small quantum systems immobilized by them. In particular, we demonstrate the role of the electronic structure of the host on atoms as well as the location of the dopant in electronic spectroscopy.

Oral abstracts for atom2016

While predictable charge-transfer between the host and dopant have been reaffirmed by our studies, the nature of Penning transfers and the associated photoelectron emission have been enunciated by imaging electrons and ions. Further, few-electron effects special to such weakly bound systems - distortions of doubly excited states and their Fano profiles, inter-atomic Coulomb decay and electron-transfer mediated decay - have been recently uncovered and will be reported in this talk.

(This work was performed in a joint collaboration between Uni. of Freiburg, Max Planck Institute fuer Kernphysik - Heidelberg and Indian Institute of Technology - Madras, India)

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- ii) A C LaForge et al., Physical Review A 93 (5), 050502
- iii) D Buchta et al., The Journal of chemical physics 139 (8), 084301
- iv) D Buchta et al., The Journal of Physical Chemistry A 117 (21), 4394-4403

Attosecond Dynamical Franz-Keldysh Effect

Lucchini, Matteo

(ETH Zürich, Institute of Quantum Electronics, Physics Department, Zurich, Switzerland)

Intense and short optical pulses give the possibility to manipulate electrons in dielectrics on a sub-femtosecond time scale with important implications for the future development of many relevant technological fields. The first step is to understand the role of inter- and intra-band transitions in fast electron excitation. We used attosecond transient absorption spectroscopy to study ultrafast dynamics induced by a few-femtosecond IR pulse in polycrystalline diamond. We observe sub-femtosecond transient features that fully recover after the interaction with the pump. Combining our results with theoretical models of increasing complexity we were able to identify the dynamical Franz-Keldysh effect (intra-band transitions) as the main mechanisms responsible for the observed features in the diamond transient optical response. This effect occurs in a largely unexplored transition regime between classical and quantum mechanical light-matter interaction.

Space-time imaging of collective and atomic-scale electron dynamics

Morimoto, Yuya

(Ludwig-Maximilians-Universität München, Faculty of Physics, Experimental Physics - Laser Physics, Garching, Germany)

The initial step in any light-matter interaction is motion of charges at the electromagnetic field cycles of the light. This involves collective carrier dynamics in conductive materials and atomic-scale charge displacements in dielectric materials. In this presentation, we summarize our recent advances in ultrafast electron diffraction toward imaging electronic motion in space and time. Single-electron pulses have no space charge and can therefore be compressed to a duration only limited by quantum effects. We recently succeeded in applying optically generated THz fields for pulse compression and metrology [1]. This produces electron pulses with stability and duration only limited by the quality of optical-field control. Electron microscopy with such pulses provides a space-time-vectorial map of collective electrodynamics in sub-wavelength structures such as metamaterial elements [2]. Atomic-scale charge motion in gas atoms was detected by taking advantage of laser-assisted electron scattering [3]. Simulations predict that such atomic-scale electronic motion can also be recorded in condensed matters, such as graphene, provided that the electron pulses are shorter than one optical cycle [4]. This should be achievable by combining THz compression with a second stage of optical compression; the latest results will be reported.

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Oral abstracts for atom2016

Imaging sub-Angstrom and femtosecond-scale atomic motion using self-referenced diffraction

Natan, Adi

(Stanford University / SLAC, PULSE Institute, Stanford, USA)

Time-resolved femtosecond x-ray diffraction patterns from laser-excited molecular iodine are used to create a movie of intramolecular motion with a temporal and spatial resolution of 30 fs and 0.3 Angstrom. This high fidelity is due to interference between the moving excitation and the unperturbed initial charge distribution. The initial state is used as the local oscillator for heterodyne amplification of the excited charge distribution to retrieve real-space de-novo movies of atomic motion on Angstrom and femtosecond scales. This x-ray interference has not been employed to image atomic motion in molecules before. Coherent vibrational motion and dispersion, dissociation, and rotational dephasing are all clearly visible in the data, thereby demonstrating the stunning sensitivity of heterodyne methods.

Theory of field induced non-equilibrium phase transitions in a Mott insulator

Oka, Takashi

(MPIPKS, Germany)

In the field of “strongly correlated electronics”, it is important to find a way, using external fields, to control various phases that emerges in correlated materials. The Mott insulator, i.e., half-filled lattice electrons whose motion is frozen by Coulomb repulsion, is a prototypical correlated insulator, and its non-equilibrium dynamics induced by electric fields are being studied extensively. In my talk, I will give a brief overview of this field and explain several mechanisms of non-equilibrium phase transitions induced by dc and ac-fields. (1) Interface Mott transition, (2) Doublon-hole pair creation by Landau-Zener tunneling (Fig.1(b)) equivalent to the Schwinger mechanism in high energy physics, (3) Keldysh crossover from Landau-Zener tunneling to multi-photon absorption, (4) Dynamical localization in strong laser fields (Fig. 1(c)). Many of these mechanisms can be displayed in a (F, ω) phase diagram.

Ultrafast Dynamics in the Insulator-to-Metal Phase Transition of Vanadium Dioxide Measured by Attosecond Transient Absorption Spectroscopy

Ott, Christian

(MPI für Kernphysik, Heidelberg)

The photoinduced insulator-to-metal phase transition in vanadium dioxide is experimentally investigated by attosecond transient absorption spectroscopy. The experimental setup incorporates sub-5-fs laser pulses with spectra ranging from below 600 to above 900 nm, acting as a pump pulse to excite the phase transition dynamics. Through the excitation from the vanadium 3p core level ($M_{2,3}$ edge at ~40 eV) these dynamics are accessed and probed in the extreme ultraviolet using attosecond pulses. The measured ultrafast changes cover a broad ~20-eV-wide spectral range, emphasizing core spectroscopic access and the importance of charge screening and electron correlation effects.

Laser-induced recollisions under the barrier: nonlocality in strong field phenomena

Popruzhenko, Sergei

(Nationalal Research Nuclear University MEPhI, Institute for Laser and Plasma Technologies, Theoretical Nuclear Physics, Moscow, Russian Federation)

By solving numerically the time-dependent Schrödinger equation we predict a noticeable, order-of-magnitude enhancement in the probability of nonlinear ionization of atoms in a strong linearly polarized laser field at energies around twice the ponderomotive energy where the transition from the direct to the rescattered electrons is expected. This enhancement forms a distinct low-energy plateau along the polarization direction, in contrast to an exponentially decaying spectrum predicted by the strong field approximation or by the three-step classical model of ionization. This enhancement has a physical origin similar to that of the low and very low energy structures and can be interpreted as a result of soft recollisions of the photoelectron with its parent ion. It is essential however that these recollisions can only

Oral abstracts for atom2016

be treated as appearing in complex space and time. As a result, they cannot be separated into the tunneling event and the subsequent classical motion. Instead, the entire quantum path determines the ionization probability making the process of ionization nonlocal. We identify parameters of atomic targets and laser fields suitable for observation of the low-energy plateau in strong field ionization. Possible manifestations of this effect in the generation of high order harmonics are also briefly discussed.

X-ray Emission Spectroscopy in ultra-fast regime

Pushkar, Yulia

(Purdue University, Department of Physics and Astronomy, West Lafayette, USA)

Electronic structure of metal centers in materials and proteins can be studied by X-ray emission spectroscopy. Dispersive detection of high resolution X-ray fluorescence is best suited for ultra-fast applications as instant snapshots of electronic structure are obtained with single pulse of X-rays.

In Photosystem II laser pump X-ray probe time resolved measurements allowed us to follow evolution of electronic structure in the oxygen evolving complex (Mn₄Ca – cluster) within few microseconds. DFT analysis of PS II was conducted to predict changes in the Gibbs energies of catalytic steps and search for structure – activity relationships. To aid interpretation of the Mn K β X-ray emission spectra of the Mn₄Ca cluster model Mn-compounds have been analyzed. Transition of these experiments to free electron laser require re-evaluation of the damage threshold. Our preliminary Mn K β emission spectroscopy data show that Mn ions in solution are subjected to multi-photon excitations / multiple ionizations within single 10-40 fs pulse of the LCLS source.

High Field Effects in Solid State Matter-based Accelerators and Light Sources

Rosenzweig, James

(UCLA, Particle Beam Physics Laboratory, Physics and Astronomy, Los Angeles, USA)

Diffraction imaging of nanoparticles and ultrafast nanoplasma dynamics

Rupp, Daniela

(Technische Universität Berlin, Institut für Optik und atomare Physik, Cluster und Nanoteilchen, Berlin, Germany)

Free-electron lasers and high-harmonic generation sources produce intense femtosecond pulses in the extreme ultraviolet (XUV) and x-ray regime. Thriving research fields have evolved around the opportunity to investigate transient states and ultrafast dynamics with both high spatial and temporal resolution, leading to a deeper understanding of intense x-ray matter interaction.

We have studied the interaction of intense XUV pulses with single nanoclusters in a combined diffractive imaging and spectroscopic approach. From the diffraction patterns we can extract size and shape of the nanoparticle as well as the XUV intensity the particle was irradiated with. This allows the analysis of the simultaneously measured ion spectra in unprecedented detail, uncovering a previously unnoticed energy redistribution process in the nanoplasma [1].

But also the nanoplasma formation itself that takes place during the interaction with the femtosecond pulse leaves distinct features in the diffraction images. An outer shell of the nanoplasma with drastically changed refraction can be observed using XUV wavelengths resonant to atomic and ionic states. The ability to map the spatial distribution of resonant charge states in combination with the developing capabilities of the light sources – towards attosecond pulse duration and coherent control – will enable the imaging of ultrafast electron dynamics with unprecedented spatial resolution on their natural time scale.

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Oral abstracts for atom2016

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Is there dynamic interference?

Saalmann, Ulf

(MPIPKS, Max Planck Institute for the Physics of Complex Systems (MPIPKS), Finite Systems, Dresden, Germany)

Dynamic interference has been predicted to occur for the photo-effect in intense laser pulses. Thereby, non-linear behavior does not emerge as higher-order processes (a.k.a. above-threshold ionization), but rather dynamical Stark shifts that induce a double-slit-like interference in time. In contrast to published claims, dynamic interference does not occur for ground state hydrogen. We show, however, that by using hydrogen in an excited state one indeed will be able to observe it. Although the process is of non-perturbative nature, we could fully disentangle system and pulse properties allowing for predictive statements about the visibility of the effect without performing any cumbersome numerical propagation.

Solid-state attosecond population dynamics

Schultze, Martin

(Max Planck Institute of Quantum Optics, Attosecond Physics, Garching, Germany)

Attosecond soft-X-ray pulses permit the time resolved investigation of multi-electron dynamics in many systems. As an example I will discuss recent experiments exploring the role of electronic correlations in the photoionization of atoms with sub-attosecond resolution and show how the extension of these capabilities towards attosecond solid state physics provides us with a time-domain understanding of the excitation of electrons across the band gap of semiconductors and dielectrics, the resulting band structure modifications and the energy exchange dynamics between light-field and solid.

Coherent optical control of free electron beams in an ultrafast transmission electron microscope

Schäfer, Sascha

(Georg-August-Universität Göttingen, IV. Physikalisches Institut, Göttingen, Germany)

The interaction of free electrons with light has long been studied in physics, both because of its fundamental character as well as its practical applications, such as in the generation of synchrotron radiation, the utilization of Compton scattering, and in free-electron laser science.

Ultrafast transmission electron microscopy (UTEM) [1,2] provides a novel experimental access to study the strong-field interaction of high-energy electron beams with nanoscale light fields. Specifically, when an electron pulse traverses an intense optical near-field, a coherent optical phase is imprinted on the electron's wave function. Within the electron energy spectrum, this phase modulation results in a phase-locked comb of photon side-bands [2,3], allowing to gain control on the longitudinal and transverse electron pulse properties by light [4].

I will discuss, several applications of this concept, including the spatial mapping of optical near-fields, Ramsey interferometry with free electron beams [5], multi-color synthesis of electron spectra [6], schemes for the generation of attosecond electron pulse trains [2], and the transverse optical phase modulation of electron beams [7].

Finally, I will give some outlook in a more general context on future developments in time-resolved electron microscopy, which may enable the spatio-temporal mapping of structural, electronic, spin and plasmonic degrees of freedom with nanometer-femtosecond resolution.

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Oral abstracts for atom2016

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Nanosystems in Ultrafast and Superstrong Fields: Attosecond Phenomena

Stockman, Mark

(Georgia State University, Center for Nano-Optics (CeNO), Physics and Astronomy, Atlanta, USA)

We present our latest results for a new class of phenomena in condensed matter nanooptics when a strong optical field $\sim 1\text{-}3\text{ V/\AA}$ changes a solid within optical cycle (1-10). Such a pulse drives ampere-scale currents in dielectrics and adiabatically controls their properties, including optical absorption and reflection, extreme UV absorption, and generation of high harmonics (11) in a non-perturbative manner on a 100-as temporal scale. Applied to a metal, such a pulse causes an instantaneous and, potentially, reversible change from the metallic to semimetallic properties. We will also discuss our latest theoretical results on graphene and other two-dimensional solids that in a strong ultrashort pulse field exhibit unique behavior related to their topological properties in the reciprocal space (12-15). These phenomena are the fastest processes in optics unfolding within a half period of light. They offer potential for petahertz-bandwidth signal processing, generation of high harmonics on a nanometer spatial scale, etc.

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Single-photon induced Collapse of Magnesium Complexes embedded in Helium Nanodroplets

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Single-photon induced Collapse of Magnesium Complexes embedded in Helium Nanodroplets

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There is evidence that Magnesium atoms in helium nanodroplets form a metastable network of single atoms surrounded by the layer of helium atoms. Resonant two-photon ionization spectroscopy of Mg atom doped helium droplets reveals a narrow peak blue-shifted relative to the atomic transition. For doping levels in between 2-20\,atoms an additional peak nearby shows up irrespective of cluster size. The optical spectra suggest that single magnesium atoms are dissolved within the droplets having an interatomic Mg-Mg distance of about 10 Å, i.e. a magnesium foam is formed. By scanning the laser wavelength close to the atomic and foam resonance photoemission probes the electronic structure of the Mg complexes after excitation. Instead of a single photoemission line complex photoelectron spectra are obtained when more than a single magnesium atom is present in the droplet but almost no pickup dependence is observed. The features in the electron spectra can be attributed to highly excited atoms indicating a collapse accompanied by a rapid heating of the metastable structure.

Ultrafast collective electron dynamics in simple metal clusters studied by angle resolved photoelectron spectroscopy

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Clusters and nanoparticles often have properties rather different to those of the corresponding bulk material, which is due to their large surface-to-volume ratio and in general to quantum size effects, the discretization of otherwise continuous densities of states. Especially the latter effect makes them highly interesting candidates for the study of few to many particle physics. Recently the use of angle-resolved photoelectron spectroscopy allowed obtaining direct information on the nature of the electronic single particle wavefunctions in these particles [1]. New results demonstrate that the angular distributions are very sensitive to the ultrafast multi-electron dynamics during the photoionization process. It turns out that these dynamics lead to a surprisingly simple and universal form of the angular distributions, which neither depend on the cluster size nor on the cluster material.

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Oral abstracts for atom2016

Attosecond streaking spectra with the classical Wigner method

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The novel method to simulate the attosecond streaking spectrograms of atoms and molecules is introduced. The method combines the classical Wigner method with perturbation theory with respect to the XUV light. The combination results in a two dimensional integral with respect to time, which is evaluated either directly or converted into a single dimensional integral by approximate treatment of quantum coherence. Comparison with direct solutions of the Schroedinger equation for several atoms shows that the method is capable of fast direct simulation of speaking spectra with errors in ionization delays in order of several attoseconds.