O 83: Focus Session: Ultrafast Dynamics in Nanostructures II

Time: Thursday 15:00–17:30 Location: GER 38

Topical Talk O 83.1 Thu 15:00 GER 38

Imaging ultrafast electron dynamics in isolated nanoparticles — •Daniela Rupp — LFKP, ETH Zurich, Switzerland

Atomic clusters and nanodroplets are used as ideal model systems over all wavelength regimes for exploring the ultrafast physical processes underlying the formation and evolution of highly excited matter. Via single-pulse single-particle coherent diffractive imaging (CDI) with the intense femtosecond pulses from short-wavelength free electron lasers (X-FELs), it became possible to study single specimen in free flight.

In CDI, the elastically scattered light forms an interference pattern that encodes the particle's structure, allowing to investigate the morphology of fragile and short-lived specimen. Also light-induced dynamics after pulsed laser excitation can be visualized by time-resolved CDI. Even changes in the electronic properties were found to be imprinted in the CDI patterns, but their time-evolution could not be investigated with typical pulse durations of 100 femtoseconds.

We recently showed that, under favorable conditions, single helium nanodroplets can be also imaged with an intense HHG source, providing much shorter pulses. In time-resolved experiments, a moderately intense near-infrared (NIR) laser pulse, too weak to ionize helium nanodroplets, dramatically changed their scattering response in the extreme ultraviolet (XUV) regime. The ability to switch the optical properties of nanoscale matter within less than a femtosecond, and observe this temporally and spatially resolved, promise to impact a broad field of science from non-linear XUV optics to ultrafast material science.

O 83.2 Thu 15:30 GER 38

Sub-picosend Elastic Response of a supported Pd Nanoparticle Ensemble studied by Time-resolved X-ray Diffraction — Simon Chung[1], Thorben Eggert[2], •Vedran Vonk[1], Nastasia Mukharamova[1], Sebastian Matera[2], Roman Shayduk[3], Johannes Moeller[3], Nils Schewe[4], Eric Sauter[4], Christoph Schreuer[2], Anders Madsen[3], Karsten Reuter[2], and Andreas Stierle[1] — [1]CXNS - Centre for X-ray and Nanoscience, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [2]Theory Department, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6t, Berlin, 10587, Germany. — [3]European X-Ray Free-Electron Laser Facility GmbH, Holzkoppel 4, Schenefeld, 22869, Germany. — [4]Institute of Functional Interfaces (IFG), Karlsruher Institut fur Technologie (KIT), Hermann-von-Helmholtz-Platz 1, Eggenstein-Leopoldshafen, 76344, Germany.

We report on the results of a combined experimental and theoretical study of the structural response of a Pd nanoparticle ensemble upon fs-laser excitation. Time-resolved X-ray diffraction experiments, having a time resolution of approx. 100 fs, were performed at end-station MID of the European X-ray Free Electron Laser (XFEL). Molecular dynamics simulations were done taking explicitly into account the particle size distribution, as determined from SEM. The results are interepreted in terms of the dephasing of size-dependent elastic modes and picosecond energy dissipation of the nanoparticle ensemble towards the MgO single crystal support. An overview will be given about the experimental details, data analysis and molecular dynamics simulations.

O 83.3 Thu 15:45 GER 38

Description of High Garmonic Generation in Quantum Dots using a Tight-Binding approach — •Martin Thümmler[1], Alexander Croy[1], Stefanie Gräfe[1], and Ulf Peschel[2] — [1]Institute of Physical Chemistry, University of Jena — [2]Institute of Condensed Matter Theory and Optics, University of Jena

Recently, high harmonic generation (HHG) was experimentally observed in quantum dots showing striking differences compared to HHG in molecules and bulk materials. At the same time, the available theoretical tools (like semiconductor Bloch-equations and real-time density-functional theory (DFT)) have proven to be insufficient to describe the size dependence of the nonlinear response of these nanostructures. Here, we present a computationally feasible, real-space tight-binding method to account for the description of inter- and intraband harmonics, and ionization effects, while all model parameters are derived from DFT calculations. Inspired by recent experiments, we present first simulation results of HHG in 3D CdSe quantum dots for linear and elliptic polarization of the driving laser.

O 83.4 Thu 16:00 GER 38

Imaging of femtosecond spin dynamics at the nanoscale — •Sergey Zayko[1,4], Hung-Tzu Chang[1], Ofer Kfir[2], Timo Schmidt[3], Jakob Hagen[1], Michael Heigl[3], Murat Sivis[1,4], Manfred Albrecht[3], and Claus Ropers[1,4] — [1]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany — [2]School of Electrical Engineering, Tel Aviv University, Tel Aviv, Israel — [3]Experimental Physics IV, Institute for Physics, University of Augsburg, Germany — [4]IV Physical Institute, Solids and Nanostructures, Georg-August University

Femtosecond pulse durations combined with broad extreme-UV spectrum inherent to high harmonic generation (HHG) grant spectroscopic access to various material properties and phenomena including charge, spin and lattice dynamics. Here, we extend the applicability of HHG sources to direct real-space imaging of ultrafast processes. Specifically, we map femtosecond spin dynamics induces by optical excitations ferro- and ferrimagnetic materials. Our results demonstrate the first implementation of HHG radiation for element specific imaging as well as the first real-space imaging of femtosecond dynamics. Moreover, the achieved spatio-temporal resolution (down to 13.5 nm spatial and 15 fs temporal) is more than an order of magnitude higher than any magneto-optical imaging scheme reported so far, which would facilitate deeper fundamental understanding and, in particular, support application-oriented studies. Notably, the developed imaging scheme is directly applicable to a broad range of phenomena including transient absorption and structural phase transitions.

Topical Talk O 83.5 Thu 16:15 GER 38

Ultrafast coherent manipulation of free electrons via quantum interaction with shaped optical fields — •Giovanni Maria Vanacore — Department of Materials Science, University of Milano-Bicocca, Via Cozzi 55, 20126 Milano

The interaction between light and electrons can be exploited for generating radiation, or for controlling electron beams for dynamical investigation of materials, enabling new applications in quantum technologies and microscopy. In this contribution, I will describe an innovative method for coherent and versatile longitudinal/transverse manipulation of a free-electron wave function. Using appropriately shaped light fields in space and time, I will demonstrate how to modulate the energy, linear and orbital angular momenta, as well as spatial and temporal distributions of the electron wave function. The experiments have been performed in an ultrafast-TEM, where a pulsed electron beam was made to interact with shaped optical field generated via a spatial light modulator. The energy-momentum exchange resulting from such interaction was directly mapped via momentum-resolved ultrafast electron energy-loss spectroscopy. Our approach for arbitrary longitudinal/transverse electron modulation at the sub-fs timescale would pave the way to achieve unprecedented insights into non-equilibrium phenomena in advanced quantum materials, playing a decisive role in the rational design and engineering of future photonics and electronics application.

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O 83.6 Thu 16:45 GER 38

Structural Dynamics in Nanostructured Systems Probed by Ultrafast Transmission Electron Microscopy — •Nora Bach[1], Armin Feist[2], Marcel Möller[2], Claus Ropers[2], and Sascha Schäfer[1] — [1]Institute of Physics, University of Oldenburg, Germany — [2]Max Planck Institute for Multidisciplinary Sciences, Göttingen, Germany

One successful approach to investigate ultrafast nanoscale structural dynamics and to disentangle different excitation mechanisms in spatially inhomogeneous systems is based on local diffractive probing with nano-focused femtosecond electron pulses in an ultrafast transmission electron microscope (UTEM). Employing the Göttingen UTEM [1,2] in ultrafast convergent electron beam diffraction (U-CBED) mode [3], we study local dynamics in a multi-component model system consisting of a metal/semiconductor hybrid structure. Ultrashort optical excitation of a platinum stripe on a silicon membrane results in the generation of a multi-modal distortion wave propagating through the membrane. Pronounced lattice distortions are quantitatively tracked by U-CBED, and

experimental results are reproduced by numerical simulations demonstrating that a superposition of Lamb waves at resonance frequencies of the bilayer structure [4] governs the evolution of the displacement inhomogeneity within the depth of the membrane.

[1] A. Feist, N. Bach, et al., Ultramicroscopy 176, 63 (2017).
[2] N. Bach et al., Struct. Dynamics 6, 014301 (2019).
[3] A. Feist et al., Struct. Dynamics 5, 014302 (2018).
[4] N. Bach et al., Struct. Dynamics 8, 035101 (2021).

O 83.7 Thu 17:00 GER 38

Controlling and shaping the electron recoil and energy transfer via nearfield geometry — •Fatemeh Chahshouri[1] and Nahid Talebi[1,2] — [1]Institute of Experimental and Applied Physics, Kiel University, 24098 Kiel, Germany — [2]Kiel, Nano, Surface, and Interface Science, KiNSIS, Kiel University, 24098 Kiel, Germany

Inelastic interaction of free-electrons with optical near fields has recently attracted attention for manipulating and shaping free-electron wave packets. Understanding the nature and the dependence of the inelastic cross section on the polarization of the optical near-field modes of the nanostructures is important for both fundamental studies and the development of new applications in this field. Here, we investigate the effect of the laser-field polarization and nanostructures asymmetric geometry on shaping free-electrons and controlling the energy transfer mechanisms, but also tailoring the electron recoil. We show that, an oblique incident electric field improves the coupling efficiency for coherent control of the longitudinal and transversal phase modulation of an electron wave packet. We also demonstrate the possibility of tailoring the shape of the localized plasmons by incorporating specific arrangements of nanorods to enhance or hamper the transversal and longitudinal recoils of free-electrons. Our findings open up a route towards the spatial characterization of plasmonic near-fields for the coherent manipulation and control of slow electron beams for creating desired shapes of electron wave packets.

O 83.8 Thu 17:15 GER 38

Light-induced hexatic state in a layered quantum material — •Till Domröse[1], Thomas Danz[1], Sophie F. Schaible[2], Kai Rossnagel[3,4], Sergey V. Yalunin[1], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidiscipinary Sciences, Göttingen, Germany — [2]4th Physical Institute, University of Göttingen, Germany — [3]Institute of Experimental and Applied Physics, Kiel University, Germany — [4]Ruprecht Haensel Laboratory, DESY, Hamburg, Germany

Ultrafast transmission electron microscopy (UTEM) is a powerful technique for resolving non-equilibrium dynamics on the nanoscale, employing a stroboscopic laser pump/electron probe approach [1]. Here, we harness the high-coherence electron source of the Göttingen UTEM [2] in the investigation of a structural phase transition between two charge-density wave (CDW) phases in a layered material [3]. A three-dimensional reconstruction of the nascent CDW order by means of ultrafast tilt-series nanobeam diffraction allows us to identify a transient hexatic state in the early stages of the phase formation. As the optical excitation induces a loss of interlayer correlations, this two-dimensional intermediate is characterized by a high density of topological defects suppressing the translational symmetry, while the orientational order is weakly preserved. On longer timescales, defect recombination and the establishment of the equilibrium stacking sequence lead to the formation of the long-range ordered high-temperature CDW phase.

[1] A. H. Zewail, Science **328**, 187-193 (2010).
[2] A. Feist *et al.*, Ultramicroscopy **176**, 63-73 (2017).
[3] T. Domröse *et al.*, in Review, arXiv:2207.05571 [cond-mat.mtrl-sci]