HL 48: Ultra-fast Phenomena

Time: Friday 9:30–12:00 Location: POT 361

HL 48.1 Fri 9:30 POT 361

Characterizing the nonadiabatic tunneling dynamics in solid-state high harmonic generation — •Ruixin Zuo[1], Xiaohong Song[2], Shuai Ben[2], Weifeng Yang[2], and Torsten Meier[1] — [1]Department of Physics, Paderborn University, Warburger Str. 100, D-33098 Paderborn, Germany — [2]School of Science, Hainan University, Hainan 570288, China

Tunneling is a fundamental quantum process which may result from light-matter interaction. High-harmonic radiation, as an ultrafast phenomenon initiated by the tunneling excitation, carries information about the electronic dynamics in the classically forbidden region. We introduce a quantum trajectory analysis to identify the underlying quantum dynamics governing solid-state high harmonics which improves the understanding of neighboring-atoms collisions [1]. It is revealed that properties of the electron-hole pairs when emerging in the classically allowed region like the group velocity and the relative displacement are dictated by the nonadibaticity of tunnel ionization and are crucial in determining the subsequent propagation and radiation. Furthermore, we show that the two-color high harmonic spectroscopy validates the quantum trajectory analysis and enables us to probe the tunneling dynamics in strongly driven solid-state systems.

[1] R. Zuo, A. Trautmann, G. Wang, W.-R. Hannnes, S. Yang, X. Song, T. Meier, M. Ciappina, H. T. Duc, and W. Yang, Neighboring atom collisions in solid-state high harmonic generation, Ultrafast Science **2021**, 9861923 (2021).

HL 48.2 Fri 9:45 POT 361

Compact, CEP-stable, few-cycle OPCPA for single attosecond pulse generation — •Bastian Manschwetus, Filippo Campi, Thomas Braatz, Sebastian Starosielec, Jan-Heye Buss, Michael Schulz, and Robert Riedel — Class 5 Photonics, Hamburg, Germany

Attosecond technology pathed the way for studying ultrafast electronic processes in atoms, molecules, solids, and complex many body systems. Carrier envelope phase (CEP) stabilization in the few-cycle regime is a key enabler of this technology. A crucial step forward is the demonstration of high-repetition rate optical parametric chirped-pulse amplifier (OPCPA) systems for the generation of phase-stable few-cycle, μJ-level driver pulses for high-harmonic-generation (HHG). In this work, we present an OPCPA providing white light seeded, actively controlled CEP stable, 9 fs pulses around 900 nm center wavelength with 30 μJ pulse energy as a high-harmonic driver for attosecond experiments at 200 kHz repetition rate.

HL 48.3 Fri 10:00 POT 361

Transient Optical Property Changes in Semimetals with Strong Electron-Phonon Coupling — •Fabian Thiemann[1], Germán Sciaini[2], Alexander Kassen[1], Tyler Lott[2], and Michael Horn-von Hoegen[1] — [1]University of Duisburg-Essen, Lotharstr. 1, 47057 Duisburg, Germany — [2]University of Waterloo, 200 University Avenue West, Waterloo, ON N2L 3G1, Canada

Group V semimetals, such as bismuth used in this study, are well known for their strong electron-phonon coupling due to the inherent Peierls distortion of their lattice. In particular, the displacively excited coherent A_{1g} phonon mode plays an important role in ultrafast studies on Bi. With knowledge of the excitation density we used a series of thin Bi films on Si(111) to reconstruct the transient dielectric function in an all optical pump-probe experiment. Though we only access the first half ps of dynamics, we separate the contributions from the excited carrier system and the coherent atomic motion. Our results suggest that we can describe the changes in optical properties caused by the non-equilibrium distribution of carriers still similar to a thermal dependency in equilibrium. In the regime of high excitation densities, we no longer can use the two-band Raman susceptibility approach to describe changes caused by the coherent phonons. We suggest that the light scattering is greatly enhanced by the excited carriers and that recently proposed band shifts in the time domain mainly define this part of changes of the dielectric function.

HL 48.4 Fri 10:15 POT 361

Ultrafast optical control of polariton energy in an organic semiconductor microcavity — •K.E. McGhee[1,2], M. Guizzardi[3], R. Jayaprakash[1], K. Georgiou[1,4], G. Cerullo[3,5], T. Jessewitsch[6], A. Zasedatelev[7], U. Scherf[6], T. Virgili[5], P.G. Lagoudakis[7], and D.G. Lidzey[1] — [1]University of Sheffield, UK — [2]Universität Leipzig, Germany — [3]Politecnico di Milano, Italy — [4]University of Cyprus, Cyprus — [5]Istituto di Fotonica e Nanotecnologia-CNR, Italy — [6]Universität Wuppertal, Germany — [7]University of Southampton, UK

Exciton-polariton condensates are of great interest due to their potential applications in polariton logic devices, nonlinear photonic integrated circuits and quantum simulators. In this work, we have fabricated polariton microcavities designed to allow the dynamic trapping and manipulation of such condensates using ultrashort laser pulses. Using transient pump-probe spectroscopy, we have saturated the electronic transition of a weakly-coupled dye in a cavity containing a second strongly-coupled dye. In doing so, we alter the cavity effective refractive index and therefore the position of the lower polariton branch. We demonstrate a maximum blueshift of this mode of 12 meV, dependent on the pump fluence, and show that this effect is ultrafast, with switch 'on' and 'off' times of less than 1 ps. Utilising this controllable energy shift, it should be possible to dynamically 'write' energy barriers into organic polariton microcavities without affecting the coupling strength. This would allow important studies on interacting polariton condensates for the development of next generation quantum devices.

HL 48.5 Fri 10:30 POT 361

Imaging nanoscale electron dynamics with extreme ultraviolet radiation — •Hung-Tzu Chang[1], Sergey Zayko[1], Jakob Hagen[1], Murat Sivis[1,2], and Claus Ropers[1,2] — [1]Max Planck Institute for Multidisciplinary Sciences, 37077 Goettingen, Germany — [2]4th Physical Institute, University of Goettingen, 37077 Goettingen, Germany

Non-equilibrium electronic processes such as electron thermalization, ballistic transport, and spin diffusion, occur at femtosecond time- and nanometer length-scales. While ultrafast spectroscopic methods can trace the temporal evolution of electronic states [1], capturing those dynamics in real space remains challenging. Here we demonstrate time-resolved coherent diffractive imaging using a table-top extreme ultraviolet (EUV) source based on high harmonic generation [2,3], where the sample is first excited by a near-infrared pump pulse and probed with core-level absorption induced by a time-delayed EUV pulse. The valence electron dynamics can be retrieved from the spatially resolved changes of the EUV field amplitude and phase exiting the sample after localized photoexcitation. This technique provides nanometer spatial resolution in addition to the time resolution and element specificity of femtosecond table-top core-level spectroscopy, and paves the way for further understanding of the mechanisms of photophysical processes in condensed matter.

[1] Chang et al., Phys. Rev. B 103(6), 064305 (2021). [2] Kfir et al., Sci. Adv. 3(12), eaao4641 (2017). [3] Zayko et al., Nat. Commun. 12(1), 6337 (2021)

15 min. break

HL 48.6 Fri 11:00 POT 361

Coherent-to-incoherent crossover of photoexcited electron-phonon dynamics in 2D materials — •Enrico Perfetto[1,2] and Gianluca Stefanucci[1,2] — [1]Dipartimento di Fisica, Universita' di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy — [2]INFN, Sezione di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy

We present a first principles nonequilibrium Green's function approach to describe the carrier and nuclear dynamics of 2D materials. Our scheme is based on the simultaneous propagation of the electronic and phononic degrees of freedom and includes the GW, Ehrenfest, and Fan-Migdal self-energies. The method scales linearly with the propagation time, and allows to describe in a conserving fashion the retarded Coulomb screening and the cooling dynamics of hot carriers via phonon emission. Numerical results are provided for a monolayer MoS2 photoexcited above the gap. The intra-valley scattering is responsible for an ultrafast carriers migration toward the band edges already during pumping. Intervalley scattering occurs on a longer timescale, of the order of a few hundreds of femtoseconds. At high carrier density

the energy exchange between electrons and phonons is very efficient, leading to a sizable increase of the lattice temperature within one picosecond. During this processes the electronic coherence is lost. The lattice coherence, instead, survives for much longer. Several hundreds of femtoseconds after the dephasing of the electronic polarization the nuclear displacements still exhibit undamped oscillations.

HL 48.7 Fri 11:15 POT 361

Coherent versus incoherent excitons: Stability, time-dependent ARPES spectrum and Floquet topological phases — •Gianluca Stefanucci[1,2] and Enrico Perfetto[1,2] — [1]Dipartimento di Fisica, Universita' di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy — [2]INFN, Sezione di Roma Tor Vergata, Via della Ricerca Scientifica 1, 00133 Rome, Italy

We consider a band insulator turning into a nonequilibrium (NEQ) exciton superfluid (XS) after resonant pumping. The NEQ-XS is characterized by self-sustained coherent oscillations of the superfluid condensate. We show that the ARPES spectrum from long probe pulses features a subgap excitonic sideband; it originates from the condensate-dressing of the conduction states and it has an intensity proportional to the excitonic wavefunction squared [PRB94, 245303 (2016); PRM3, 124601 (2019)]. Reducing the probe duration below the condensate period the ARPES signal becomes periodic in the impinging time of the probe [PRB101, 041201(R)(2020)]. The stability of the NEQ-XS as the conduction density grows is jeopardized by the increased screening efficiency of the looser excitons. Nonetheless, a proper nonequilibrium self-consistent treatment of screening indicates that NEQ-XS is stable up to relatively high densities [PRB102, 085203 (2020)]. We also show that a p-wave NEQ-XS at high enough density undergoes a Floquet topological transition [PRL125, 106401 (2020)]. Phonon-induced decoherence eventually transforms the XS phase into an incoherent exciton-polaron fluid. During this process the excitonic sideband broadens and red-shifts (Stokes shift) [PRB103, 245103 (2021)]

HL 48.8 Fri 11:30 POT 361

Intermolecular conical intersections in acceptor-donor-acceptor aggregated molecules — •Somayeh Souri[1], Katrin Winte[1], Antonietta De Sio[1], Christoph Lienau[1], Elena Mena-Osteritz[3], Peter Bäuerle[2], Teresa Kraus[3], Fulu Zheng[2], Mohamed Madjet[2], and Sergei Tretiak[4] — [1]University of Oldenburg, Germany — [2]University of Bremen, Germany — [3]Ulm University, Germany — [4]Los Alamos National Laboratory, USA

In molecules, strong coupling between electronic excitations and vibrational modes may result in conical intersections (CoIns) of multi-dimensional potential energy surfaces. At CoIns different electronic states cross and couple strongly via vibrational modes. This results in large nonadiabatic couplings violating the Born-Oppenheimer approximation. While CoIns are ubiquitous in molecules, not much is known about their occurrence and relevance for intermolecular ultrafast energy and charge transfer dynamics in the solid state. Here, we reveal the existence of intermolecular CoIns in thin films of aggregated acceptor-donor-acceptor-type oligomers by ultrafast two-dimensional electronic spectroscopy (2DES). The 2DES maps show a grid-like peak pattern at early times, followed by a rapid reduction of the peak spacings which transforms into a featureless map after only ˜40 fs. We take this abrupt change in the 2DES maps as clear evidence for the passage of the optically launched vibrational wave packet through the intermolecular CoIn in the thin films. Our results, which are confirmed by simulations, suggest new opportunities for guiding the coherent flow of energy and charge in solid-state nanostructures.

HL 48.9 Fri 11:45 POT 361

Potential signatures of hydrodynamic transport captured by THz high harmonic generation — •Tim Priessnitz[1], Thales de Oliveira[2], Liwen Feng[1,3], Min-Jae Kim[1,3], Sergey Kovalev[2], Bernhard Keimer[1], and Stefan Kaiser[1,3] — [1]Max-Planck Institute for Solid State Research, 70569 Stuttgart, Germany — [2]Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany — [3]Institute of Solid State and Materials Physics, Technical University Dresden, 01069 Dresden, Germany

Terahertz high harmonic generation (THz HHG) is a common property of nonlinear systems. Recently it has been used to investigate fundamental principles that govern transport and nonlinear dynamics in novel quantum materials like graphene or Dirac semimetals. However, these studies have not yet been successfully extended to low temperatures where hydrodynamic effects come into play. Optical phenomena such as THz HHG are proposed to provide a more efficient way to probe hydrodynamic effects than previously studied DC transport measurements, which identified the delafossite $PdCoO_2$ as a candidate to observe such properties. Here, we report on THz HHG in thin films of $PdCoO_2$ at low temperatures and we will discuss potential signatures of hydrodynamics, contributing to the ongoing puzzle of low-temperature origins of THz HHG.