HL 22: Ultrafast Phenomena I (Joint session of HL and O, organized by HL)

Time: Tuesday 9:30–13:15 Location: H10

HL 22.1 Tue 9:30 H10

Sub-cycle quantum interference in solid-state high-harmonic generation — •Matthias Hohenleutner[1], Fabian Langer[1], Olaf Schubert[1], Matthias Knorr[1], Christoph Lange[1], Ulrich Huttner[2], Stephan W. Koch[2], Mackillo Kira[2], and Rupert Huber[1] — [1]University of Regensburg, 93040 Regensburg, Germany — [2]University of Marburg, 35032 Marburg, Germany

Utilizing intense light pulses to control electron motion in atoms and molecules has opened up spectacular new routes in ultrafast and attosecond photonics such as high-harmonic generation (HHG). The recent discovery of HHG in solids combines ultrafast quantum control with complex condensed matter systems. We employ intense, phase-controlled multi-THz waveforms to drive HHG in bulk gallium selenide. Non-resonantly driven interband polarization and simultaneous intraband carrier acceleration throughout the entire Brillouin zone result in the emission of extremely broadband, phase-locked high-harmonics (HH). More importantly yet, sub-cycle time- and field-resolution allows us to directly trace the underlying electron dynamics with precise temporal correlation to the driving waveform. Remarkably, the HH are emitted as a unipolar train of ultrashort, nearly unchirped few-femtosecond bursts, which coincide precisely with the driving field maxima. These features unravel a novel quantum interference of multiple interband excitation paths during HHG, as explained by our microscopic quantum theory. The first direct time domain study of HH from solids paves the way towards a full quantum picture of the underlying mechanisms and sparks hope for solid-state sub-femtosecond sources.

HL 22.2 Tue 9:45 H10

Lattice dynamics of optically excited few-layer graphite — Christian Gerbig, Silvio Morgenstern, Marlene Adrian, Cristian Sarpe, •Arne Senftleben, and Thomas Baumert — Institut für Physik und CINSaT, Universität Kassel, Heinrich-Plett-Straße 40, 34132 Kassel

Time-resolved diffraction with femtosecond electron pulses has become a promising technique to directly provide insights into photo induced primary dynamics at the atomic level in molecules and solids. Ultrashort pulse duration as well as extensive spatial coherence are desired, however, space charge effects complicate the bunching of multiple electrons in a single pulse. We experimentally investigate the interplay between spatial and temporal aspects of resolution limits in ultrafast electron diffraction (UED) on our highly compact transmission electron diffractometer. To that end, the initial source size and charge density of electron bunches are systematically manipulated and the resulting bunch properties at the sample position are fully characterized in terms of lateral coherence, temporal width and diffracted intensity. We obtain electron pulse durations down to 120 fs and transversal coherence lengths up to 20 nm. Instrumental impacts on the effective signal yield in diffraction and electron pulse brightness are discussed as well. The performance of our compact UED setup at selected electron pulse conditions is finally demonstrated in a time-resolved study of lattice heating in few-layer graphite after optical excitation. During the heating process, we observe shearing modes and acoustic breathing modes.

HL 22.3 Tue 10:00 H10

Ultrafast carrier dynamics in bulk MoS_2 studied by transient absorption spectroscopy — •Tim Völzer, Matthias Lütgens, Franziska Fennel, and Stefan Lochbrunner — Institut für Physik, Universität Rostock, Albert-Einstein-Str. 23, 18059 Rostock

Transistion metal dichalcogenides feature unique electronic and optical properties. In addition, they are characterised by a layered structure, allowing the preparation of atomically thin crystals. As a representative of those materials, molybdenum disulfide is suggested as a promising candidate for optoelectronic and photocatalytic applications. To characterise the optical excitation and its dynamics we performed both static transmission and time-resolved pump-probe spectroscopy.

Two absorption bands, labelled A and B, are observed in the static absorption spectrum which are attributed to direct transitions at the K point of the Brillouin zone. The lower electronic transition is excited within the femtosecond absorption measurements and the dynamics is followed by a white light continuum covering the whole visible range. A decrease of absorption for the A and B transition is observed whereas an increase of absorption is present for lower energies. This signature is indicative for a band renormalization induced by the population in the first excited band. The subsequent signal decay shows two distinct time dependencies, an exponential sub picosecond contribution caused by carrier-carrier and carrier-phonon scattering and a strongly non-exponential component for longer times. The latter is attributed to electron-hole recombination via defect states and shows an acceleration with increasing excitation density.

HL 22.4 Tue 10:15 H10

Internal structure and ultrafast dynamics of excitons in monolayer WSe_2 — •Christoph Pöllmann[1], Philipp Steinleitner[1], Ursula Leierseder[1], Philipp Nagler[1], Gerd Plechinger[1], Michael Porer[1], Rudolph Bratschitsch[2], Christian Schüller[1], Tobias Korn[1], and Rupert Huber[1] — [1]Department of Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Institute of Physics, University of Münster, 48149 Münster, Germany

Atomically thin transition metal dichalcogenides (TMDCs) promise groundbreaking optoelectronic applications and devices due to their direct bandgap in the optical range. As a consequence of the layered structure, unusually strongly bound excitons can exist even at room temperature, dominating the optical and electronical properties. In order to exploit the full potential of this new material system, key open questions regarding the excitons in TMDC monolayers have to be answered. Here we report the first direct observation of the intraexcitonic 1s-2p transition via time resolved pump/THz probe studies, tracing both optically bright and dark exciton states. Beside quantitative information about transition energies, oscillator strengths, linewidths and many-body effects a record fast radiative decay of bright excitons with a time constant of only 150 fs can be revealed.

HL 22.5 Tue 10:30 H10

Auger Recombination and Charge Transfer of CdSe/CdS Core/Shell Quantum Dot/Quantum Rods — •Mona Rafipoor[1,2], Jan Niehaus[3], Holger Lange[1,2], and Horst Weller[1,2,3] — [1]Universität Hamburg, Institut für Physikalische Chemie, Grindelallee 117, 20146 Hamburg — [2]The Hamburg Centre for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany — [3]CAN GmbH Grindelallee 117 20146 Hamburg

We investigated clustered CdSe/CdS quantum dots/quantum rods (QDQRs) by time-resolved optical spectroscopy.

Power dependent measurements were conducted by excitation with different intensities and recording time traces of the photoluminescence (PL) decay simultaneously with the PL intensity. Increasing the excitation power generally increases the average exciton population per QDQR. We observe signatures of exciton-exciton interactions starting with exciton populations of one. This is tentatively assigned to an interaction between the excitons across the QDQRs. The non-monotonic increase of the PL intensity with excitation power in this regime supports the idea of a new, non-radiative multiexciton decay across the cluster.

To get more Information about this kind of interaction we apply Transient Absorption spectroscopy. By varying the pump wavelength, we are able to specifically excite core and shell and follow the subsequent relaxation.

HL 22.6 Tue 10:45 H10

The role of intervalley scattering and phonon softening in the ultrafast carrier dynamics of PbTe — Prashant Padmanabhan, Kestutis Budzinauskas, Kiran H. Prabhakara, and •Paul H. M. van Loosdrecht — Physics Institute 2, University of Cologne, 50937 Cologne, Germany

PbTe is a leading thermoelectric material, notable for its low thermal conductivity and unusually large carrier mobility at very low doping levels. Here, we report on ultrafast pump-probe experiments on PbTe that shed light on the ultrafast relaxation of highly excited carriers. By employing time-resolved differential reflectivity measurements, we probe the dynamics of electron-electron and electron-phonon interactions on the femtosecond time-scale. Additionally, the use of a super-continuum probe pulse allows us to investigate the wavelength

dependence of the carrier relaxation dynamics. Our results suggest that phonon mediated intervalley scattering involving the band gap between the Γ and K points plays a significant role in carrier cooling. Further, the temperature dependence of these dynamics point to anomalous behavior that may be the result of phonon softening, recently reported to be a key element in the unique thermal properties of PbTe.

HL 22.7 Tue 11:00 H10

Theory of the coherent A_{1g} phonon decay in antimony — •Sergej Krylow, Eeuwe S. Zijlstra, and Martin E. Garcia — Universität Kassel, Theoretische Physik II, Heinrich-Plett-Straße 40, 34132 Kassel,Germany

The fluence-dependent decay paths of the femtosecond laser-excited coherent A_{1g} phonon mode in antimony are investigated by means of electronic-temperature-dependent density-functional-theory molecular-dynamics simulations, which yield an exponentially decaying coherent phonon, comparable to experiments. Additional calculations of the phonon dispersion indicate that third order decay processes occur mainly within the acoustic and fourth order processes within the optical phonon branches. This can also be seen in the subsequent analysis of the molecular dynamics simulations, which reveal that the third and fourth order phonon contributions to the decay can be influenced by means of the applied fluence.

30 min. Coffee Break

Invited Talk HL 22.8 Tue 11:45 H10

Coherent Quantum Dynamics of Excitons in Atomically Thin Semiconductors — •xiaoqin li — Univ. of Texas-Austin, Austin, USA

The transitional metal dichalcogenides (TMDs) are an emerging class of atomically thin semiconductors with tightly-bound excitons and charged excitons (i.e. trions). A fundamental property of these quasiparticles is quantum decoherence time, which reflects irreversible quantum dissipation arising from system (excitons) and bath (vacuum and other quasiparticles) interaction and determines the timescale during which excitons can be coherently manipulated. Dephasing time is also equivalent to the intrinsic homogeneous linewidth of exciton resonances. In addition, excitons in TMDs are localized at the corners of the Brillouin zone and provide a convenient way to optically manipulate the valley degree of freedom. Direct measurement of valley coherence time is challenging because it corresponds to non-radiative coherence between two degenerate states. Using ultrafast multi-dimensional optical spectroscopy, we investigate the intrinsic homogeneous linewidth of excitons, exciton valley coherence as well as coupling between excitons and trions. Our studies reveal coherent exciton dynamics on the order of ~ 100 fs in monolayer TMDs. Decoherence time is not only an important parameter for characterizing fundamental properties of excitons, but also serves as a sensitive probe for exciton-exciton and exciton-phonon interaction.

HL 22.9 Tue 12:15 H10

Ultrafast Pump-Probe Analysis of Exciton-Polariton Propagation — Jan Lohrenz[1], Stephan Melzer[1], Claudia Ruppert[1], Ilya Akimov[1], •Matthias Reichelt[2], Alexander Trautmann[2], Torsten Meier[2], and Markus Betz[1] — [1]Experimentelle Physik 2, TU Dortmund, Otto-Hahn-Str. 4, 44221 Dortmund, Germany — [2]Department of Physics and CeOPP, University of Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany

In bulk CdZnTe light is significantly slowed down in a spectral range close to the lower exciton-polariton branch. [1] Here, we investigate nonlinear optical signatures in a pump-probe experiment with femtosecond laser pulses. Experimental data and model simulations for the optical response of a nonlinearly driven two-level system suggest that the excitation induced dephasing [2] and slow light propagation lead to unexpected features in the differential transmission spectra. [3]

[1] T. Godde, I. A. Akimov, D. R. Yakovlev, H. Mariette, and M. Bayer, Phys. Rev. B **82**, 115332 (2010).
[2] H. Wang, K. Ferrio, D.G. Steel, Y.Z. Hu, R. Binder, and S.W. Koch, Phys. Rev. Lett. **71**, 1261 (1993).
[3] J. Lohrenz, S. Melzer, C. Ruppert, I.A. Akimov, M. Reichelt, A. Trautmann, T. Meier, M. Betz, to be published.

HL 22.10 Tue 12:30 H10

Dynamics of exciton-polariton condensates in semiconductor microcavities with periodic potentials — •Xuekai Ma[1], Stefan Schumacher[1], and Oleg Egorov[2] — [1]Physics Department, Universität Paderborn, Warburger Strasse 100, 33098 Paderborn, Germany — [2]Institute of Condensed Matter Theory and Solid State Optics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany

Exciton-polaritons are quasi-particles made of quantum well (QW) excitons coupled to cavity photons. They have very small effective mass ($10^{-4} m_e$) and lifetimes on the tens of picoseconds scale. Due to their photonic properties, they can be excited by light and probed, respectively. Due to their excitonic properties, nonlinearity is introduced into this system at elevated densities. Polaritons, which are composite bosons, can undergo a condensation process (with similarities to Bose-Einstein condensation) under incoherent excitation. Many features in the dynamics of polariton condensates can be described by a modified Gross-Piteavskii equation (GPE). Here, we study the nonlinear dynamics of polariton condensates in periodic potentials. In the presence of a periodic potential, a band structure including a band-gap can be obtained. We show that polariton condensates can occupy and switch between different energy states by changing the pump excitation intensity and shape. Our simulation results agree very well with recent experimental results.

HL 22.11 Tue 12:45 H10

Nonlinear terahertz quantum control of Landau-quantized electrons — •Thomas Maag[1], Andreas Bayer[1], Sebastian Baierl[1], Matthias Hohenleutner[1], Tobias Korn[1], Christian Schüller[1], Dominique Bougeard[1], Christoph Lange[1], Rupert Huber[1], Martin Mootz[2], Stephan W. Koch[2], and Mackillo Kira[2] — [1]Department of Physics, University of Regensburg, 93053 Regensburg, Germany — [2]Department of Physics, Philipps-University Marburg, 35032 Marburg, Germany

Controlling superpositions of many-body electronic quantum states in solids is impeded by rapid dephasing through inter-particle scattering. However, Walter Kohn found in 1961 that the cyclotron resonance of Landau-quantized electrons in a two-dimensional electron gas (2DEG) is immune to Coulomb forces. This protection warrants long coherence times and makes the system interesting for quantum control, but explicitly excludes nonlinearities. Here, we demonstrate how intense light pulses in the terahertz (THz) spectral range can induce well-controlled electronic many-body correlations in a magnetically biased 2DEG and tailor a distinctly anharmonic response. Coherent ladder climbing up to the 6th rung yields population inversion and abruptly increases dephasing. Strikingly, 2D THz spectroscopy reveals distinct multi-wave mixing signatures, which our quantum theory explains through Coulomb interactions between electrons and the positively charged ionic background. These many-body dynamics demonstrate how internal degrees of freedom of solid state quantum systems enable coherent nonlinear interactions for future ultrafast quantum information processing.

HL 22.12 Tue 13:00 H10

Ultrafast nonlinear response of GaAs under high pressures — •Johannes M. Braun[1,2], Jan F. Schmidt[3], Denis V. Seletskiy[3], Harald Schneider[1], Manfred Helm[1,2], and Alexej Pashkin[1,3] — [1]Helmholtz-Zentrum Dresden-Rossendorf, Germany — [2]TU Dresden, Germany — [3]Department of Physics and Center for Applied Photonics, University of Konstanz, Germany

Applying hydrostatic pressure leads to dramatic changes in the band structure of semiconductors. In particular, it enables a continuous tuning of the bandgap energy. Here we study the nonlinear response of bulk gallium arsenide (GaAs) in the vicinity of its bandgap. The optical pump-probe experiment is performed in a non-collinear reflection geometry at pressures up to 3 GPa generated inside a diamond anvil cell. By increasing pressure we observe pronounced slowing down of the relaxation dynamics of photoexcited charge carriers: the time constant of the dominating relaxation process increases from about 10 ps at ambient pressure to 35 ps above 0.7 GPa. These time scales are by an order of magnitude shorter than the recombination time determined using optical pump - THz probe spectroscopy. Thus, the fast dynamics observed in the optical pump-probe measurements is governed by the cooling of hot electron distribution and not by the recombination process. Furthermore, at pressures above 2 GPa the bandgap energy of GaAs is above the excitation spectrum of our experiment. The sample becomes transparent for the femtosecond pulses leading to a transient pump-probe signal with a negative sign due to the third order nonlinear response of GaAs.