HL 46: Ultra-fast phenomena

Time: Friday 9:30–12:45 Location: H31

HL 46.1 Fri 9:30 H31

Ultrafast x-ray scattering from laser-driven electronic systems — •Darya Gorelova[1,2], David A. Reis[3,4,5], and Robin Santra[1,2,6] — [1]Center for Free-Electron Laser Science, DESY, Notkestrasse 85, D-22607 Hamburg, Germany — [2]The Hamburg Centre for Ultrafast Imaging, University of Hamburg, Luruper Chaussee 149, D-22761 Hamburg, Germany — [3]PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, California 94025, USA — [4]Department of Applied Physics, Stanford University, Stanford, California 94305, USA — [5]Department of Photon Science, Stanford University, Stanford, California 94305, USA — [6]Department of Physics, University of Hamburg, Jungiusstrasse 9, D-20355 Hamburg, Germany

We theoretically study x-ray scattering from an electronic system in the presence of a laser field driving electron dynamics in this system [1]. We apply Floquet theory to describe a laser-driven electronic system, and then obtain the scattering probability of an arbitrary nonresonant x-ray pulse from such a system employing the framework of quantum electrodynamics. We apply our theory for a calculation of a diffraction signal from a driven crystal in the regime of high harmonic generation. We connect the properties of the energy- and time-resolved diffraction signals to the properties of electron dynamics driven by the pump pulse.

[1] Daria Popova-Gorelova, David A. Reis and Robin Santra, accepted to Phys. Rev. B, arXiv:1811.02246.

HL 46.2 Fri 9:45 H31

Transient birefringence and dichroism of ZnO studied by femtosecond time-resolved spectroscopic ellipsometry — •Oliver Herrfurth[1], Steffen Richter[2], Mateusz Rębarz[2], Shirly Espinoza[2], Joshua A. Leveillee[3], André Schleife[3], Jakob Andreasson[2,4], Marius Grundmann[1], and Rüdiger Schmidt-Grund[1] — [1]Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Leipzig — [2]ELI Beamlines, Institute of Physics, Czech Academy of Science, Prague, Czech Republic — [3]Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, USA — [4]Condensed Matter Physics, Department of Physics, Chalmers University of Technology, Gothenburg, Sweden

We report on UV-pump-induced transient birefringence and dichroism of an *m*-plane oriented ZnO thin film measured by femtosecond pump-probe spectroscopic ellipsometry in the spectral range from 340 nm to 690 nm (1.8 eV to 3.6 eV) with approximately 100 fs time-resolution. The pump-induced density of electron-hole pairs is estimated as $3 \times 10^{19}\mathrm{cm}^{-3}$. We determine the complex dielectric function tensor from measurements parallel and perpendicular to the crystal's optic axis using a transfer matrix algorithm. Comparison to the ZnO bandstructure and first-principles dielectric function (DF) calculations provides evidence for inter-valence-band transitions of "hot" charge carriers near the *M* point in the Brillouin zone. They are governed by selection rules for dipole transitions for perpendicular and parallel light polarization which cause the transient optical anisotropy.

HL 46.3 Fri 10:00 H31

Diffuse scattering in a laser controlled state above the melting threshold — •Tobias Zier, Sabrina Schuster, Eeuwe S. Zijstra, and Martin E. Garcia — Theoretische Physik, Universität Kassel, Heinrich-Plett-Str. 40, 34132 Kassel, Germany

With recent time-resolved x-ray diffraction techniques it became possible to measure, besides Bragg diffraction peaks, time-dependent diffuse scattering signals. Within these signals important information about the atomic motion after a femtosecond-laser excitation are included, which are mandatory to distinguish, e.g., coherent phonon motion from thermally squeezed one. Here, we analyze the diffuse scattering of bulk silicon in a laser-controlled state in the nonthermal melting region, by performing ab initio dynamics simulations using CHIVES (Code for Highly excIted Valence Electron Systems). Even though, nonthermal melting is a stochastic process we developed a control mechanism that enables us to drive the system above the nonthermal melting threshold into a stable state. Characteristic for this new state are different electronic properties compared to the ground state. Our results will give new insights into the atomic configuration within the control mechanism.

HL 46.4 Fri 10:15 H31

Transient absorption and population dynamics of laser-excited conjugated molecules from RT-TDDFT — •Jannis Krumland[1], Ana Valencia[1], Stefano Pittalis[2], Carlo Andrea Rozzi[2], and Caterina Cocchi[1] — [1]Physics Dept. and IRIS Adlershof, Humboldt-Universität zu Berlin, Germany — [2]CNR NANO, Modena, Italy

Pump-probe spectroscopy is a powerful tool to study the electronic and optical properties of materials. In transient absorption spectroscopy, the system is illuminated by two femtosecond laser pulses with the first one exciting the system and the second one probing its response at a given time delay. Real-time time-dependent density functional theory (RT-TDDFT) can be adopted for ab- initio simulations of these experiments [1]. In RT-TDDFT, the electron density is propagated in time, enabling the calculation of optical properties in the linear regime and beyond. We apply this method to investigate ethylene, benzene and thiophene molecules representing the building blocks of a variety of organic compounds. By applying laser pulses of increasing intensity in resonance with the gap transition, we analyze the dynamics of photo-excited electrons. From the transient absorption spectra, we identify the signatures of excitations due to the laser-induced population of initially unoccupied states. The underlying mechanisms of excited-state absorption are discussed in relation with benchmark calculations unraveling their excitations in the linear response [2].

[1] U. de Giovannini et al., ChemPhysChem 14 1363 (2013)
[2] J. Krumland et al., in preparation (2019)

HL 46.5 Fri 10:30 H31

Externally Controlled Lotka-Volterra Dynamics in a Linearly Polarized Polariton Fluid — •Matthias Pukrop and Stefan Schumacher — Department of Physics and CeOPP, Paderborn University, 33098 Paderborn, Germany

Coherent polariton fluids in semiconductor microcavities provide a platform for optical pattern formation and all-optical switching of patterns induced by a weak control beam. Recently, reversible on-demand switching was studied for a linearly polarized polariton system. Switching times and achievable gain were discussed based on detailed numerical simulations of the coupled light-field exciton dynamics obtained within a microscopic semiconductor theory [1]. Here we derive a simplified population competition model to gain more detailed insight into the switching mechanisms based on dynamical systems theory. The model we derived leads to an extended generalized Lotka-Volterra system for two competing populations controlled by an external source term. We will give an overview of both the existence and stability properties of possible steady states in the relevant parameter space, spanned by the strength of anisotropy and external control. We construct phase boundaries in representative regions of the non-trivial parameter space and characterize relevant bifurcations. The population competition model reproduces all key features of the switching processes observed in the full numerical simulations of the rather complex system and at the same time is simple enough for a fully analytical understanding of the underlying system dynamics.

[1] P. Lewandowski et al., Opt. Express 25, 31056-31063 (2017).

HL 46.6 Fri 10:45 H31

Ultrafast coherent 2D fluorescence micro-spectroscopy on semiconducting carbon nanotubes at room temperature — •Matthias Nuss, Alexandru Badalan, Jan-Hagen Krohn, Kerstin Müller, Donghai Li, Friedrich Schöppler, Tobias Hertel, and Tobias Brixner — Institut für Physikalische und Theoretische Chemie I, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Further developments in nano- and molecular electronics would benefit strongly from the possibility of the spatio-temporal evolution of molecular processes. Non-linear ultrafast techniques provide insights into energy transfer pathways, e.g., mediated via electronic coupling. A comprehensive way to observe these dynamics is ultrafast coherent 2D fluorescence micro-spectroscopy [1]. This method is a generalization of transient absorption spectroscopy with frequency resolution both for the pump and the probe step, combined with spatial resolution in an optical microscope. This provides the capability to observe, e.g., inhomogeneous line broading as well as the formation and an-

nihilation dynamics of excitons on the femtosecond timescale. Here, we utilize the third-order 2D signal for monitoring electronic coupling and energy transfer processes in semiconducting single-walled carbon nanotubes. To this end, an LCD-shaped four pulse sequence with 13 fs temporal encoding of each puls is focused through an NA = 1.4 objective and the fluorescence is detected as a function of inter-pulse time delays and phases.
[1] S. Goetz, et al., *Optics Express* 26, Nr. 4: 3915-25 (2018)

15 min. break

HL 46.7 Fri 11:15 H31

Non-equilibrium (Transient) Experiments on Zinc Oxide — •Shirly Espinoza[1], Steffen Richter[1], Oliver Herrfurth[2], Mateusz Rebarz[1], Stefan Zollner[3], Marius Grundman[2], Jakob Andreasson[1,4], and Rüdiger Schmidt-Grund[2] — [1]ELI Beamlines, Institute of Physics, Czech Academy of Sciences, Czech Republic — [2]Semiconductor Physics Group, Felix Bloch Institute for Solid State Physics, Leipzig, Germany — [3]New Mexico State University, Department of Physics, Las Cruces, NM, USA — [4]Condensed Matter Physics, Department of Physics, Chalmers University of Technology, Göteborg, Sweden

Zinc Oxide is a wide bandgap semiconductor considered for applications in electronics and optoelectronics devices. Its wide bandgap allows high power operation at high temperature; therefore, studies about its electron distribution at non-equilibrium values are crucial for the design of such device. Our work was done by pump-probe ellipsometry, a technique that allows the distinction between real and imaginary part of the dielectric function. We observed bleaching of the excitonic absorption at the band edge, and occurrence of intra-valence-band absorption during the very first picoseconds. Electron-phonon scattering causes thermalization, which creates non-thermal phonon distributions delaying the charge relaxation to tens of picosecond. The final heat dissipation happens in the scale of nanoseconds.

HL 46.8 Fri 11:30 H31

Ultrafast lattice dynamics in thin film black phosphorus — •Patrick-Nigel Hildebrandt[1,2], Daniela Zahn[1], Thomas Vasileiadis[1], Helene Seiler[1], Yingpeng Qi[1], and Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, Berlin, Germany — [2]Humboldt-Universität zu Berlin, Germany

We study the ultrafast lattice dynamics of thesemiconductor black phosphorus after photoexitation. Using femtosecond electron diffraction, we obtain a time-resolved picture of the electron-lattice equilibration and the subsequent relaxation of a non-thermal phonon population in an anisotropic material. Along the in-plane directions, *armchair* and *zigzag*, we measure an anisotropic Debye-Waller-Factor. The time-dependence of the average mean-square displacement reveals anisotropic phonon relaxation. This shows that the structural anisotropy of black phosphorus leads to anisotropic lattice dynamics, which is tentatively explained by different phonon density of states with *armchair*- or *zigzag*-polarization.

HL 46.9 Fri 11:45 H31

Ultrafast photo-induced spin polarized currents in nanostructers — •Michael Kraus, Dominik Schulze, Anna Dyrdal, and Jamal Berakdar — Martin-Luther Universität Halle-Wittenberg

We present theoretical predictions on the ultrafast generation of spin-polarized currents and pure spin currents by an appropriate nanostructuring of the sample and/or by modulating local fields produced by nearby plasmonic elements. It is shown how spin currents are steered by spatio-temporal tailoring of the driving photonic fields.

HL 46.10 Fri 12:00 H31

Modeling white light coherent 2D-spectroscopy on electrically pumped semiconductor nanostructures — •Aris Koulas-Simos[1,2], Mirco Kolarczik[1], Benjamin Lingnau[2], Bastian Herzog[1], Sophia Helmrich[1], Ulrike Woggon[1], Nina Owschimikow[1], and Kathy Lüdge[2] — [1]Institut für Optik und Atomare Physik, Technische Universität Berlin — [2]Institut für Theoretische Physik, Technische Universität Berlin

We investigate the coherent coupling mechanisms of an electrically pumped InAs/InGaAs quantum dot-in-a-well semiconductor optical amplifier. We track the phase evolution of the system using white light coherent 2D spectroscopy with collinear pulses and electronic frequency filtering. In the experiment, we observe signatures of the quantum dot ground and excited state and a coupling between them. Interestingly, we also observe the existence of crossed excitons, i.e. coupling of states with different dimensionality. Our observations can be described using a theoretical model based on the Maxwell Bloch equations, where we incorporate the local charge carrier and polarisation dynamics microscopically. Similar to a heterodyne experiment, we vary the phase of both the pump and probe pulse, which allows us to distinguish the different bands of the outcoming signal and to extract a 2D spectrum corresponding to the rephasing sideband.

HL 46.11 Fri 12:15 H31

Higher-order contributions and non-perturbative effects in the nonlinear optical absorption of direct-gap semiconductors — •Wolf-Rüdiger Hannes and Torsten Meier — Department of Physics and CeOPP, University of Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany

The semiconductor Bloch equations including the intraband acceleration [1] are used to study the nonlinear absorption of single or multiple light pulses by direct-gap semiconductors. The steady state response can be described analytically by multi-photon absorption coefficients, which are shown to agree well with numerical results for short pulses and/or finite dephasing and relaxation times. The dependencies on the light frequencies, band gap energy, initial populations, and the time delay between the pulses are analyzed. In third order we confirm some previously described aspects [2] such as the strong enhancement of two-photon absorption for non-degenerate light frequencies. In higher orders we discuss both multi-photon absorption and dispersive corrections to lower orders. Beyond the perturbative treatment of the Bloch equations we investigate the intensity-dependent optical absorption for a single incident pulse and in a pump-probe setup.
[1] H. T. Duc, T. Meier, and S. W. Koch, Phys. Rev. Lett. **95**, 086606 (2005).
[2] C. Aversa *et al.*, Phys. Rev. B **50**, 18073 (1994).

HL 46.12 Fri 12:30 H31

Bringing nonlinear stimulated emission to the infrared: From sapphire and fused silica to perovskites — •Thomas Winkler[1], Sean Bourelle[1], Thomas Baumert[2], and Felix Deschler[1] — [1]Cavendish Laboratory, University of Cambridge, United Kingdom — [2]Institut für Physik und CINSaT, Universität Kassel, Deutschland

While the stimulated emission of light was postulated over more than a hundred years ago and the laser has found its way into every laboratory, its nonlinear counterpart (i.e. two-photon stimulated emission) has only been observed in a handful of experiments so far. Therefore it was very surprising when we recently discovered the nonlinear amplification of an ultraviolet femtosecond laser pulse in a piece of optically excited sapphire (LADIE effect [1]). The effect holds high promises for laser technology, nonlinear microscopy and laser-spectroscopy as it provides e.g. a different set of selection rules. Here, we present extended studies showing the possibility of switching between two nonlinear amplification processes in fused silica, one being related to free carriers, whereas the other is related to the characteristic and self-trapped excitons. Furthermore, we discuss our recent studies to expand the nonlinear stimulated emission from the ultraviolet into the infrared regime. To that extend we utilize novel 2D and 3D perovskite materials, which showed great properties for optoelectronic devices. Having band gaps in visible spectrum and long carrier lifetimes, they are an ideal sample system to probe the two-photon stimulated emission in an ultrafast pump-probe experiment with tunable- and broadband infrared probe pulses. [1]T.Winkler *et al.* Nature Physics **14**, 74-79 (2018)