HL 6: Ultrafast Phenoma I

Time: Monday 9:30–12:45 Location: POT 51

Invited Talk HL 6.1 Mon 9:30 POT 51

Optical Coherent Multidimensional Spectroscopy of Semiconductor Nanostructures — •Steven Cundiff — University of Michigan, Ann Arbor, Michigan, USA

Optical coherent multidimensional spectroscopy excels at removing the effects of inhomogeneity and revealing the details of coupling for resonant excitations. In semiconductors, the excitations are typically excitons. In nanostructures, inhomogeneity arises due to size and alloy fluctuations, while coupling can arise due to many-body interactions.

In quantum wells, coherent two-dimensional spectroscopy reveals the essential role of many-body interactions, both in terms of the nonlinear response of the exciton resonance and coupling between peaks. Disorder arises because of fluctuations in the well width, and 2D spectroscopy can monitor the migration of excitons among localization sites.

For very thin quantum wells, strongly localized, quantum-dot like states, are present. Using high spatial resolution, it is possible to isolate these states and observe many-body coupling that occurs due to carriers present in the quantum-well states.

Self-organized quantum dots display strong inhomogeneous broadening, which usually prevents the observation of coherent effects, such as Rabi oscillations, in an ensemble measurement. However, 2D spectroscopy allows an individual frequency group to be isolated, which in turn means that clear Rabi flopping can be observed. Furthermore, coherent control of the biexcitonic state is possible due to the broad bandwidth of the excitation pulses.

HL 6.2 Mon 10:00 POT 51

Localized High-Harmonic Generation in Semiconductor Nanostructures — •Murat Sivis[1,2], Marco Taucer[2], Kyle Johnston[2], Giulio Vampa[2], André Staudte[2], Andrei. Yu. Naumov[2], David. M. Villeneuve[2], Paul B. Corkum[2], and Claus Ropers[1] — [1]4th Physical Institute - Solids and Nanostructures, Georg-August University, Göttingen, Germany — [2]Joint Attosecond Science Laboratory, National Research Council of Canada and University of Ottawa, Ottawa, Canada.

High-harmonic generation (HHG) in solid-state systems, as recently demonstrated in semiconductors[1–3], enables the transfer of gas-phase attosecond spectroscopy techniques to condensed matter. In general, HHG is sensitive to the electronic structure of the generation medium and the local driving laser field. Both of these properties can be routinely tailored in solids by modifying the chemical composition and the microstructure. Here, we study HHG in nanostructured zinc oxide and silicon crystals. We use wavelength-selective microscopic imaging to characterize the harmonics (at 2 μm driving wavelength) and find enhanced emission in nanofabricated grating structures as well as in gallium-implanted patterns. Our results illustrate novel means to control HHG and to use the harmonic emission as a unique local probe to investigate structural, chemical or electronic dynamics in solid-state systems.

[1]S. Ghimire *et al.* Nat. Phys. **7**, 138-141 (2011).
[2]O. Schubert *et al.* Nat. Photon. **8**, 119-123 (2014).
[3]G. Vampa *et al.* Nature **522**, 462-464 (2015).

HL 6.3 Mon 10:15 POT 51

Atomistic modeling of exciton and charge dynamics in a ZnO nanocrystal — •Dirk Ziemann and Volkhard May — Institut für Physik, Newtonstr. 15, Humboldt Universität zu Berlin, D-12489 Berlin, Germany

There is an increasing interest in ZnO semiconductor materials due to, i. a., its wide band gap and large exciton binding energy. Especially ZnO nanostructures are promising candidates for modern optical and opto electronic devices. For a quantitative understanding of the processes and time scales in theses systems an atomistic description of realistic systems, which are tens of nanometers in size and consist of thousands of atoms, is needed.

Therefor, in this talk the dynamics of excitons and charges in a spherical ZnO nanocrystal (10.000 atoms) will be discussed. The dynamics are described by a stochastic Schrödinger equation taking the initial photoexcitation and the subsequent relaxtion via electron/exciton phonon interaction into acount. The electronic and phononic states are modeled by the density functional based tight binding (dftb) method, which deals with huge systems and is still able to consider, e. g., surface effects, ligands und defects on an atomistic level. Excitonic states are calculated with a configuration interaction (CI) approach.

HL 6.4 Mon 10:30 POT 51

Terahertz narrowband-pump broadband-probe spectroscopy of intersubband transitions in a wide single quantum well — •Johannes Schmidt, Stephan Winnerl, Emmanouil Dimakis, Harald Schneider, and Manfred Helm — Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

Electron relaxation dynamics of intersubband transitions (ISBT) in quantum wells can cover a broad range from ps to ns time scales, dependent on the transition energy compared to the LO-phonon energy [1]. Most of previous studies used various types of narrowband interband or intraband spectroscopy, where only one transition can be studied at a time. We have developed a setup that allows for narrowband THz pumping using the free electron laser FELBE together with broadband THz probing employing time-domain spectroscopy, and thus enabling us to record the full spectral relaxation dynamics under resonant pumping.

We study a 40 nm wide modulation doped single GaAs quantum well; this guarantees a homogeneous intensity over the active structure (which is not the case in multi quantum wells). To improve the sensitivity of the measurements, we modulate the electron density using proper gate voltages.

THz absorption of the 1-2 ISBT is seen at 2.15 THz and the 2-3 transition is observed at 3.4 THz. While pumping the 1-2 transition resonantly, a relaxation time of 1100 ps is probed. We will also report on the full probe spectra, on excitation of the 2-3 transition, and we are searching for pump-induced quantum coherent effects.

HL 6.5 Mon 10:45 POT 51

Phonon Wave Packet Emission Signatures in Four-Wave Mixing Micro-Spectroscopy of Single Quantum Dots — •Daniel Wigger[1], Tomasz Jakubczyk[2,3], Valentin Delmonte[2,3], Quentin Mermillod[2,3], Doris E. Reiter[1], Jacek Kasprzak[2,3], and Tilmann Kuhn[1] — [1]Institut für Festkörpertheorie, Universität Münster, Wilhelm-Klemm-Str. 10, 48149 Münster — [2]Univ. Grenoble Alpes, F-38000 Grenoble, France — [3]CNRS, Institut Néel, "Nanophysique et Semiconducteurs" Group, F-38000 Grenoble, France

Four-Wave Mixing (FWM) micro-spectroscopy is a powerful tool to investigate the level structure of single emitters. This knowledge is essential for the use of single emitters in quantum technological applications. We investigate single self-assembled Quantum Dots (QDs), which stand out as scalable solid state single emitters but suffer from their inevitable coupling to lattice vibrations. The optical excitation of the QD exciton on the ps timescale leads to the generation of travelling phonon wave packets [1]. This generation of phonons results in a loss of coherence in the excitonic subsystem. For ultrafast excitations this is directly reflected in a rapid drop of the FWM signal. We present a combined theoretical and experimental study on the temperature dependence of the corresponding FWM signals [2].

[1] Wigger et al., J. Phys.: Condens. Matter 26, 355802 (2014)
[2] Jakubczyk et al., ACS Photonics (2016)

Coffee Break

HL 6.6 Mon 11:30 POT 51

Ultrafast Extreme-UV ARPES Study of the Transition-Metal Dichalcogenide $MoSe_2$ — •Jan Heye Buss[1], Julian Maklar[1], Frederic Joucken[1], Yiming Xu[1], He Wang[1], Changhyun Ko[2], Sefaattin Tongay[2], Junqiao Wu[2], and Robert A. Kaindl[1] — [1]Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, USA — [2]Dept. of Materials Science and Engineering, University of California, Berkeley, USA

Semiconducting transition-metal dichalcogenides exhibit intriguing physical properties, including a large spin-orbit splitting, strong Coulomb interactions, and optical access to the valley degree of freedom. Important insight into the fundamental microscopic interactions can be obtained via studies of the momentum-resolved non-equilibrium

carrier dynamics. Here, we present time-resolved ARPES investigations of $MoSe_2$ crystals using high-repetition-rate extreme-UV femtosecond pulses, enabling us to track the electron dynamics within the full Brillouin zone with high sensitivity. After resonantly driving excitons at the K-point, the transient ARPES signals reveal a rapid time evolution governed by inter-valley scattering to the conduction band minimum on a 70-fs time scale. We will discuss the momentum-space dynamics as well as distinct temporal and spectral features that yield first evidence for the observation of excitons via angle-resolved photoemission spectroscopy.

HL 6.7 Mon 11:45 POT 51

Ultrafast electron transfer-induced CO_2 activation at a ZnO surface — •Lukas Gierster, Sesha Vempati, and Julia Stähler — Department of Physical Chemistry, Fritz-Haber-Institute of the Max-Planck-Society, Faradayweg 4-6, 14195 Berlin, Germany

Since many years, ZnO has been used as a catalyst which facilitates the conversion of carbon dioxide into valuable chemicals such as methanol [1]. CO_2, which is the most stable carbon oxide species, needs to be activated in order to start hydrogenation. Recent experimental and theoretical work showed that CO_2 adsorbs in a bent configuration with tridentate chemical binding on the ZnO (10-10) surface [1]. However, the energies of the frontier molecular orbitals of the adsorbed CO_2 molecules in the chemical reaction are still not known. We investigate this question using two-photon photoelectron spectroscopy (2PPE), which gives access to occupied and unoccupied electronic states and the dynamics therein. Static photoelectron spectroscopy shows that the work function of the surface increases considerably upon CO_2 adsorption possibly due to a (partial) reduction of the molecules and their dipole moment. A time-resolved pump-probe experiment suggests that electrons are injected from the ZnO substrate into the CO_2 molecules after above band gap photoexcitation of the substrate. The injected electrons populate the CO_2 LUMO, which subsequently shifts down in energy. This leads to a built up of an electronic state just below the Fermi energy within few picoseconds, which is likely to be related to the activated CO_2 molecules.

[1] K. Kotsis, et al., Z. Phys. Chem. 222, 891-915 (2008).

HL 6.8 Mon 12:00 POT 51

Complete Analysis of a transmission electron diffraction pattern of a molybdenum disulphide-graphite heterostructure — •Marlene Adrian, Arne Senftleben, Silvio Morgenstern, and Thomas Baumert — University of Kassel, Institute of Physics (CINSaT), D-34132 Kassel, Germany

The combination of various 2D layered materials in multilayer heterostructures arises great interest in the current science. Due to the large variety of electronic properties in the group of 2D layered materials, the combination opens a new pathway towards ultrasmall electronic devices. In this contribution we present a complete mathematical description of multilayer heterostructures and a full characterisation of their diffraction patterns. A 27 nm thick molybdenum disulphide-graphite heterostructure was produced and analysed with the methods presented. Additionally, the ultrafast lattice dynamics after optical excitation of the sample will be discussed.

HL 6.9 Mon 12:15 POT 51

Ultrafast transmission electron microscopy using laser-triggered field emitters — •Thomas Danz, Armin Feist, Nora Bach, Nara Rubiano da Silva, Marcel Möller, Sascha Schäfer, and Claus Ropers — IV. Physical Institute – Solids and Nanostructures, University of Göttingen, Germany

Ultrafast transmission electron microscopy (UTEM) is a promising approach to investigate ultrafast processes with nanometer spatial resolution [1]. Using the versatile imaging, diffraction, and spectroscopy capabilities of such an instrument, structural, electronic, and magnetic dynamics can be probed in a laser pump/electron probe scheme [2].

The pulsed electron source of the Göttingen UTEM project employs linear photoemission from a nanoscopic Schottky emitter, delivering highly coherent electron pulses with down to 200 fs pulse duration, 0.6 eV energy width, and sub-1 nm focused beam diameter [3]. We present first applications, as well as prospects and challenges of the UTEM in ultrafast electron imaging, diffraction, energy loss spectroscopy (EELS), and holography, and discuss approaches to reversibly drive electronic and structural phase transitions in inhomogeneous systems.

[1] A. H. Zewail, Science **328**, 187 (2010)
[2] A. Feist *et al.*, in preparation
[3] A. Feist *et al.*, arXiv:1611.05022

HL 6.10 Mon 12:30 POT 51

A combined approach of k.p-perturbation theory and semiconductor Bloch equations for the analysis of ultrafast photocurrents — •Reinold Podzimski[1], Huynh Thanh Duc[2], and Torsten Meier[1] — [1]Department Physik and Center of Optoelectronics and Photonics Paderborn (CeOPP), Universität Paderborn, Warburger Str. 100, D-33098 Paderborn, Germany; — [2]Ho Chi Minh City Institute of Physics, Vietnam Academy of Science and Technology, Mac Dinh Chi Street 1, District 1, Ho Chi Minh City, Vietnam;

In unbiased non-centrosymmetric semiconductors electronic currents can be generated directly by optical laser pulses. Combining k.p-perturbation theory with the semiconductor Bloch equations provides an approach to theoretically describe these photocurrents in bulk and quantum well systems [1,2]. We investigate shift and rectification currents in bulk GaAs and GaAs quantum well systems. The transparent approach of the semiconductor Bloch equations allows to identify the microscopic origin of several phenomena. Using a geodesic grid for the discretization of the **k**-space we are able to include the Coulomb interaction and to investigate excitonic contributions.

[1] R. Podzimski, H. T. Duc, and T. Meier, Proc. SPIE **97460W**, 9746 (2016).
[2] H. T. Duc, R. Podzimski, S. Priyadarshi, M. Bieler, and T. Meier, Phys. Rev. B **94**, 085305 (2016).