HL 3: Focus Session: Electron-phonon interaction and ultrafast processes in semiconductors

Femtosecond pulsed lasers have recently emerged as powerful tools to probe the vibrational properties of bulk as well as nanostuctured semiconductors. The results of such experiments require an accurate theoretical modeling for their interpretation. While the tools for conducting such calculations start to emerge, their application to real systems remains challenging. At the ab-initio level the methods are computationally extremely demanding; while at the mesoscopic level, the electron (excitonic) - vibronic interaction requires careful parametrization. This focus session will give an overview of the present state of the art ab-initio and mesoscopic descriptions, with connection to modern experiments. (Organizers: Gabriel Bester, MPI Stuttgart, and Michael Oestreich, University of Hannover)

Time: Monday 9:30–14:00 Location: H13

Topical Talk HL 3.1 Mon 9:30 H13

Ultrafast processes in carbon nanotubes and quantum dots — •Ulrike Woggon — Institute of Optics and Atomic Physics, TU Berlin, Germany

Phonon-assisted ultrafast relaxation determines the femtosecond and picosecond dynamics of a great variety of semiconductor nanostructures. Famous examples are quantum dots, colloidal nanocrystals and semiconducting carbon nanotubes. Devices based on these nanostrcutures are thus ideally suited for high-speed telecommunication applications. In particular, QD-SOAs feature a great potential for ultrafast nonlinear signal processing. The understanding of the ultrafast scattering processes in nanotubes is the key for exploiting the huge application potential which nanotubes offer, e.g. for light-emitting and detecting nanoscale electronic devices. We investigate population and phase dynamics in In(Ga)As QDs by heterodyne ultrafast two-color pump-probe spectroscopy which yields complementary information on amplitude and phase dynamics. In a joint study of two-color pump-probe experiments and microscopic calculations based on the density matrix formalism, we extract, both experimentally and theoretically, picosecond carrier relaxation dynamics in single-walled carbon nantubes and ascribe it to the intraband scattering of excited carriers with acoustic phonons. The calculated picosecond relaxation times show a decrease for smaller tube diameters. Theoretical results of phonon-assisted scattering processes are in good agreement with the observed scattering and relaxation times.

HL 3.2 Mon 10:00 H13

Zero-point motion and temperature effects on the band gap renormalization of semiconductor nanoclusters — Peng Han and •Gabriel Bester — Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, 70569 Stuttgart, Germany.

Using the frozen-phonon approach based on *ab initio* density functional theory (DFT), we calculate the zero-point motion band gap renormalization and the temperature dependent of the band gap in semiconductor nanoclusters. Our method avoids the large computational costs (especially the slow convergence with respect to the number of unoccupied bands) associated with the calculation of the Fan-(self-energy-) and the Debye-Waller terms. A band gap reduction in silicon clusters with a magnitude of hundreds of meV induced by the quantum zero-point atomic motion is obtained; this quantum effect is found to increase with decreasing cluster size. Based on the Bose-Einstein distribution, we further study the temperature dependence of the band gap in semiconductor nanoclusters and find a reduction of 580 meV and 270 meV at room temperature for silicon clusters with radius of 9.6 and 11.9 Å, respectively. Furthermore, we find that the Si-H rotation and shear modes play an important role on the zero-point band gap renormalization of hydrogen passivated silicon clusters. By analyzing the mode dependence of the band gap reduction, we also see that the zero-point renormalization of the band gap is dominated by the optic-like vibrations with Γ_4 point group symmetry.

Topical Talk HL 3.3 Mon 10:15 H13

Quantum dots - artificial atoms, molecules or small pieces of bulk? Nonadiabatic molecular dynamics in the Kohn-Sham representation. — •Oleg Prezhdo[1], Heather Jaeger[1], Long Run[1], Amanda Neukirch[1], and Kim Hyeon-Deuk[2] — [1]University of Rochester, Rochester, NY, USA — [2]Kyoto University, Kyoto, Japan

Quantum dots (QD) are quasi-zero dimensional structures with a unique combination of solid-state and atom-like properties. Unlike bulk or atomic materials, QD properties can be modified continuously by changing QD shape and size. Often, the bulk and atomic viewpoints contradict each other. The atomic view suggests strong electron-hole and charge-phonon interactions, and slow energy relaxation due to mismatch between electronic energy gaps and phonon frequencies. The bulk view advocates that the kinetic energy of quantum confinement is greater than electron-hole interaction, charge-phonon coupling is weak, and relaxation through quasi-continuous bands is rapid. QDs exhibit new physical phenomena: The phonon bottleneck to electron energy relaxation and generation of multiple excitons can improve efficiencies of photovoltaic devices. Our state-of-the-art nonadiabatic molecular dynamics techniques, implemented within time-dependent density functional theory, allow us to model QDs at atomistic level and in time-domain, providing a unifying description of quantum dynamics on the nanoscale.

HL 3.4 Mon 10:45 H13

Ab initio molecular dynamics simulations of ultrafast melting of Si — •Tobias Zier, Eeuwe S. Zijlstra, and Martin E. Garcia — Theoretical Physics, University of Kassel, Germany

After an intense ultrashort-laser excitation of Si the crystalline structure disorders within several 100's of femtoseconds. This phenomenon is known as ultrafast melting. The underlying effect is the bond softening as a consequence of the laser-induced highly nonthermal state in which the electrons have a temperature of several 10 000 K, while the ions remain close to room temperature. Performing MD-Simulations for supercells with up to 640 atoms allowed us to follow the ionic motion after its excitation. Our results provide new insights in the first steps of nonthermal melting by showing that the ionic motion is dominated by different physical effects, dependent on the timescale, namely, acceleration, deceleration, and diffusion.

Topical Talk HL 3.5 Mon 11:00 H13

Out-of-equilibrium carrier dynamics in semiconductors: a novel approach — •Andrea Marini — Istituto di Struttura della Materia of the National Research Council, Via Salaria Km 29.3, I-00016 Monterotondo Stazione, Italy — European Theoretical Spectroscopy Facilities (ETSF)

In this talk I will present a novel approach based on the merging of Non-Equilibrium Green's function theory and Density Functional Theory to investigate the carrier dynamics following a pump excitation.

The case of bulk Silicon, a paradigmatic indirect gap semiconductor, is studied by using the Baym–Kadanoff equations. Both the electron–electron (e–e) and electron–phonon (e–p) self–energies are calculated fully *Ab–Initio* by using a semi–static GW approximation in the e–e case and a Fan self–energy in the e–p case. By using the generalized Baym–Kadanoff ansatz the two–time evolution is replaced by the only dynamics on the macroscopic time axis.

The enormous numerical difficulties connected with a real–time simulation of realistic systems is overcomed by using a completed collision approximation that further simplifies the memory effects connected to the time evolution. The carrier dynamics is shown to reduce in such a way to have stringent connections to the well–known equilibrium electron–electron and electron–phonon self–energies.

Lunch break

Topical Talk HL 3.6 Mon 12:30 H13

The role of phonons for exciton and biexciton generation in a quantum dot driven by adiabatic rapid passage — •Tilmann Kuhn — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität, 48149 Münster, Germany

The excitation of a quantum dot with chirped laser pulses provides a powerful tool for a robust preparation of quantum states. This process, known as adiabatic rapid passage, can be well understood using the concept of dressed states. Compared to an ideal few-level system interacting with a chirped light pulse, however, the dynamics in a quantum dot is strongly modified by the influence of acoustic phonons which may lead to dephasing and transitions between the dressed states and therefore deteriorate the adiabatic passage.

In this talk I will present a theoretical analysis of the role of phonons for the preparation of either exciton or biexciton states in a quantum dot. At low temperatures phonon emission leads to pronounced asymmetries in the exciton state preparation when changing the sign of the chirp. When using linearly polarized light, the fidelity of the exciton preparation may be additionally reduced by phonon-assisted biexciton generation. On the other hand, by suitably tailoring the pulse parameters the biexciton state can be selectively prepared by using either a two-photon resonance excitation or a two-color scheme.

HL 3.7 Mon 13:00 H13

Electron-phonon coupling in colloidal CdSe/CdS core-shell quantum dots — •Steffen Westerkamp, Andrei Schliwa, Amelie Biermann, and Christian Thomsen — Institut für Festkörperphysik, Technische Universität Berlin, Germany

Phonon frequencies and eigenvectors, and their coupling to excited electronic states are calculated for approximately spherical, CdSe/CdS core-shell quantum dots (QD).

The equilibrium atomic positions and phonon modes are obtained using an empirical force model. Electron and hole wavefunctions are calculated using the 8-band-kp envelope function method, thus taking into account band coupling, strain, and piezo/pyroelectric effects. These results are combined to determine the Huang-Rhys-factors thus reflecting the electron-phonon coupling for each phonon mode.

We vary size, core-shell thickness ratio, and the sharpness of the interface of the quantum dots and discuss the impact of interface- and surface-phonons.

Topical Talk HL 3.8 Mon 13:15 H13

Spin lifetime and electron-phonon interaction in graphene — •Guido Burkard — University of Konstanz, Germany

Graphene and carbon nanotubes represent interesting platforms for exploring the quantum coherence of localized electron spins in suitably tailored nanostructures [1]. Due to the low density of nuclear spins in graphene, one can expect long spin coherence times. However, as in semiconductor quantum dots, the spin lifetime in graphene quantum dots at low temperatures will be limited by the spin-orbit coupling and phonon emission processes. We present a theoretical study of the spin lifetime of single electrons as a function of the magnetic field in quantum dots formed by electrostatic confinement in gapped graphene [2] and in armchair graphene nanoribbons [3]. We show that spin relaxation processes can be suppressed in a phonon cavity such as in suspended carbon nanotubes, and instead a coherent strong coupling of a single spin and a single phonon mode becomes possible [4]. This coupling opens new possibilities for spin readout [5] and manipulation.

[1] B. Trauzettel, D. V. Bulaev, D. Loss, G. Burkard, Nature Phys. 3, 192 (2007). [2] P. R. Struck, G. Burkard, Phys. Rev. B 82, 125401 (2010). [3] M. Droth, G. Burkard, Phys. Rev. B 84, 155404 (2011). [4] A. Pályi, P. R. Struck, M. Rudner, K. Flensberg, G. Burkard, Phys. Rev. Lett. 108, 206811 (2012). [5] P. R. Struck, H. Wang, G. Burkard, arXiv:1212.1569 (2012).

HL 3.9 Mon 13:45 H13

Theory of optical emission in semiconductor nanostructures - selforganized, colloidal quantum dots and quantum wells — •Marten Richter, Uyen-Khanh Dang, Julia Kabuss, Alexander Carmele, Mario Schoth, Matthias-Rene Dachner, and Andreas Knorr — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Germany

The coupling of electrons and phonons in semiconductor nanostructures shows a strong impact on their quantum dynamics. In this talk, we discuss the electron–phonon assisted optical emission of quantum dots and quantum wells. We start with an analysis of the influence of acoustical and optical phonons on the fluorescence and Raman signals of selforganized quantum dots. In particular, frequency and time resolved spectra are analyzed. For quantum well intersubband transitions, longitudinal phonons cause inter- and intrasubband relaxation. We discuss these processes and their visibility in 2D coherent spectra. At the end, we give an outlook to the electron–phonon interaction in colloidal quantum dots compared to selforganized quantum dots, with a focus on the process of carrier multiplication.