HL 33: Optical Properties 2

Time: Thursday 15:00–18:00 Location: H32

HL 33.1 Thu 15:00 H32

Implementation of the Bethe-Salpeter Equation using Crystal Symmetries — •Jörn Stöhler[1,2], Dmitrii Nabok[1], Stefan Blügel[1], and Christoph Friedrich[1] — [1]Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich, Germany — [2]RWTH Aachen University, Germany

The Bethe-Salpeter equation (BSE) and *GW* approximation are two many-body perturbation theory techniques that together form the state-of-the-art method to include electron-hole interaction in periodic systems. The BSE has proven to be the most accurate tool to compute optical absorption for the valence and core energy region, as well as electron energy loss.

We have implemented the BSE in the SPEX code, a full-potential linearized augmented planewave (FLAPW) code that supports Green-function based methods including the *GW* approximation, optical spectra in the random phase approximation, and more. We use crystal symmetries to achieve a significant computational speedup for the construction and diagonalization of an effective electron-hole Hamiltonian in the Tamm-Dancoff approximation, from which we obtain symmetric exciton wavefunctions and energies.

Our code is parallelized and has been tested for various bulk, layered and monolayer semiconductors, among them LiF and MoS_2, and includes spin-orbit coupling. The results agree with available theoretical and experimental spectra from the literature.

HL 33.2 Thu 15:15 H32

The Berry dipole photovoltaic demon and the thermodynamics of photo-current generation within the optical gap of metals — Li-kun Shi[1], Oles Matsyshyn[2,1], Justin C. W. Song[2], and •Inti Sodemann Villadiego[3,1] — [1]Max-Planck-Institut fur Physik komplexer Systeme — [2]Division of Physics and Applied Physics, Nanyang Technological University, Singapore — [3]Institut fur Theoretische Physik, Universitat Leipzig

Berry phase driven photo-voltaic effects offer novel mechanisms that could allow to engineer a new generation of opto-electronic technologies.

We will show that there is a large class of bulk photovoltaic mechanisms that make possible to produce a net rectified photo-voltaic current even when the impinging radiation has a frequency that resides within the optical gap of the material, in contrast to previous claims. We will describe the thermodynamics of these in-gap rectification effects and show that most of these mechanisms are necessarily accompanied by a small but finite irreversible photon absorption in order for them to be consistent with the laws of thermodynamics. There is, however, one remarkable exception: the intra-band non-linear Hall effect arising from the anomalous velocity induced by the Berry curvature. This non-linear Hall effect allows to have a photovoltaic mechanisms whose maximum allowed efficiency can be 100% for the conversion of circularly polarized light onto electricity. More remarkably, because it is a reversible process, this same mechanism can be conversely used as a a highly efficient electrical amplifier of circularly polarized light.

HL 33.3 Thu 15:30 H32

Nonlinear photocurrents induced by terahertz radiation in twisted bilayer graphene — •Stefan Hubmann[1], Philipp Soul[1], Giorgio di Battista[2], Marcel Hild[1], Kenji Watanabe[3], Takashi Taniguchi[3], Dmitri Efetov[2], and Sergey Ganichev[1] — [1]Terahertz Center, University of Regensburg, 93040 Regensburg, Germany — [2]ICFO, Castelldefels, Barcelona 08860, Spain — [3]National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

We report on the observation of nonlinear photocurrent and photoconductivity in twisted bilayer graphene (tBLG) with twist angles below 1°. We show that excitation of the tBLG bulk causes a photocurrent, whose sign and magnitude are controlled by the orientation of the radiation electric field and the photon helicity. The developed theory shows that the current is formed by asymmetric scattering in gyrotropic tBLG. For the observed photocurrents, we demonstrate the emergence of pronounced oscillations upon variation of the gate voltage, which correlate with the oscillations of the sample resistance. These photocurrent oscillations originate in interband transitions between a multitude of subbands in tBLG. Furthermore, at higher radiation intensities, we detected a nonlinear intensity dependence of bulk photogalvanic current and photoconductivity. These nonlinear photoresponses are caused by the interplay between interband, intersubband, and intraband transition. This interplay is controlled by the Fermi level position with respect to the Moiré subbands. We show that the photosignals saturate with rising intensity, while contributions from different transitions differ in their respective saturation behavior.

HL 33.4 Thu 15:45 H32

Dielectric function of $CuBr_xI_{1-x}$ thin films — •E. Krüger[1], M. Seifert[2], M. Bar[1], S. Merker[3], P. Bischoff[1], H. Krautscheid[3], S. Botti[2], M. Grundmann[1], and C. Sturm[1] — [1]Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Germany — [2]Friedrich-Schiller-Universität Jena, Institut für Festkörpertheorie und -optik, Germany — [3]Universität Leipzig, Institut für Anorganische Chemie, Germany

Copper halides such as CuI and CuBr are promising p-type semiconductors for transparent optoelectronic devices, especially due to the recently proposed hole density tunability [1]. Here, we present the dielectric function of $CuBr_xI_{1-x}$ thin films ($0 \leq x \leq 1$) determined by spectroscopic ellipsometry in the spectral range from 0.7 eV to 6.5 eV at room temperature. The observed features in the dielectric function are attributed to various electronic transitions in the Brillouin zone [2]. Non-monotonic behavior is observed for the band gap energy as a function of alloy composition revealing a quadratic bowing parameter of 0.5 in good agreement with literature [3]. The spin-orbit splitting decreases linearly from 650 meV for CuI to 150 meV for CuBr. The experimental results are compared with DFT-calculated band structures for different alloy compositions. The effects of bond length mismatches, chemical disorder, and different contributions of metal and halogen atoms to the upper valence bands are discussed in detail.

[1] Yamada et al., Adv. Funct. Mater. **30**, 2003096 (2020)
[2] Krüger et al., APL **113**, 172102 (2018)
[3] M. Cardona, Phys. Rev. **113**, 69 (1963)

HL 33.5 Thu 16:00 H32

Neutralisation of detrimental effects on the Rydberg exciton absorption spectrum — •Katharina Brägelmann, Mariam Harati, Binod Panda, Julian Heckötter, and Marc Assmann — Experimentelle Physik II, Technische Universität Dortmund, 44225 Dortmund

We report on the neutralisation of charged impurities by excitation of Rydberg excitons with surprisingly small laser powers. Rydberg excitons are highly excited states in Cu_2O with principal quantum numbers of up to $n = 30$ [1] with extensions in μm range. The well-known theories propose an n^{-3} scaling for both oscillator strengths and linewidths of the excitons. Usually the highest states ($n = 16$ and above) show some deviation from those theories, as oscillator strengths are smaller and linewidths are wider than expected, which leads to an reduced absorption of these states. Those deviation are known to stem from the presence of charged impurities in the material [2]. Here, we show a way to increase the absorption and to minimize the deviations mentioned above. This increase of absorption happens when the system is pumped in an extremly narrow energy region around the band gap (less than 1 meV) with very small powers of only $0.1 - 10\,\mu$W. This effect indicates a 'purification' of the illuminated volume as the naturally charged impurities have less detrimental impact on the high excitonic states. This research contributes to a deeper understanding of impurity - exciton interactions.

[1]M. A. M. Versteegh et al., Phys. Rev. B 104, 245206 (2021).
[2]S. O. Krüger et al., Phys. Rev. B 101 (2020).

15 min. break

Invited Talk HL 33.6 Thu 16:30 H32

Ultrastrong light-matter coupling in materials — •Niclas S. Mueller[1,2], Eduardo B. Barros[3], Florian Schulz[4], Holger Lange[4], and Stephanie Reich[1] — [1]Department of Physics, Freie Universität Berlin, Berlin, Germany — [2]Present address: NanoPhotonics Centre, Cavendish Laboratory, University of Cambridge, United Kingdom — [3]Department of Physics, Universidade Federal do Ceara, Fortaleza, Ceara, Brazil — [4]Department of Physical Chemistry, University of Hamburg, Hamburg, Germany

Driven by the field of cavity quantum electrodynamics there is an ever-growing quest for systems with extreme light-matter coupling. In the regimes of ultra- and deep strong coupling the coupling strength becomes comparable to the bare excitation energy, leading to exotic phenomena like virtual photons in the ground state and the breakdown of the Purcell effect. Here, we discuss how ultrastrong coupling is systematically achieved in materials, without the need for external cavities. We introduce densely packed supercrystals of gold nanoparticles as an artificial material where the coupling strength can be tuned from ultra- to deep strong coupling. Using a unified theory of dipole-active material excitations, we show that light-matter coupling gets maximized in three-dimensional materials, setting an upper limit for the coupling strength in cavities. From a large set of experimental data, we identify phonons in ferroelectrics, excitons in molecular crystals, and plasmons in metallic supercrystals as excitations where light-matter coupling is so strong that it affects the material ground state, eventually leading to phase transitions and changing the mechanical properties.

HL 33.7 Thu 17:00 H32

Optical Characterization of Phase-Pure Wurtzite GaAs/II-VI Core/Shell Nanowires — •Mike Külkens, Marvin Marco Jansen, Detlev Grützmacher, and Alexander Pawlis — Peter-Grünberg-Institut (PGI-9), Forschungszentrum Jülich GmbH, Germany

Self-catalysed III/V semiconductor core/shell nanowires (NWs) grown by molecular beam epitaxy (MBE) provide enormous potential to develop miniaturized electronic and optoelectronic devices. Following our recent demonstration of WZ-phase-pure *GaAs* NW growth we investigated a novel type of hybrid NWs composed of a WZ-type GaAs core with various WZ-type II/VI-semiconductor shells. The shell provides excellent confinement and passivation of the GaAs core due to the large bandgap energy difference between the two materials and allows to tune the optical bandgap of the GaAs core by tensile strain within a range of several 100 meV.

Here we report on the structural and optical properties of WZ-phase-pure grown $GaAs/Zn_{1-x}Mg_xSe$ core/shell NWs. μ-PL investigations reveal the presence of tensile strain in the *GaAs* core induced by the $Zn_{1-x}Mg_xSe$ shell, which can be engineered via the magnesium concentration and the shell thickness. The measured redshift of the near-band emission from the GaAs core was verified by evaluation of the strain and its effect on the *GaAs* bandgap, using a hydrostatic strain model. The results presented here pave the way for applications of WZ-phase-pure *GaAs*/II-VI core/shell NWs for optoelectronic devices with tunable wavelength in the infrared spectral range.

HL 33.8 Thu 17:15 H32

ZnSe-Based Microdisk Resonators in Novel Supported Geometry — •Wilken Seemann[1], Christian Tessarek[1], Siqi Qiao[2], Nils von den Driesch[2], Alexander Pawlis[2], Gordon Callsen[1], and Jürgen Gutowski[1] — [1]Institute of Solid State Physics, University of Bremen, Germany — [2]Peter Grünberg Institute (PGI-9), Forschungszentrum Jülich, Germany

Microdisk resonators often suffer from thermal problems due to the limited contact of the underetched structure to the substrate. In order to circumvent this, ZnSe-based microdisks were fabricated in a supported geometry, i.e., in contact to the substrate over their whole bottom facet. This is achieved by growing a ZnSe:Cl quantum well (QW) encapsulated in ZnMgSe barriers on an AlAs underlayer. Oxidation of the underlayer to Al_2O_3 after disk fabrication increases the refractive index difference between the resonator and the substrate.

Scanning electron microscopy reveals a high structural quality of the fabricated microdisks. Micro-photoluminescence measurements show that the resulting resonators support high-Q resonances near the band edge emission of the ZnSe:Cl QW and a large number of whispering gallery modes on the defect emission band. The latter can be reproduced using a plane-wave model. Raman measurements of microdisks, as well as the as-grown ZnSe-based structure on the oxidized and non-oxidized AlAs underlayer, are used to analyze the mechanical properties of the disk and the influence of the oxidation process on the strain in the QW structure.

HL 33.9 Thu 17:30 H32

Enhancing directivity in optical waveguide antennas — •Henna Farheen[1], Lok-Yee Yan[2], Till Leuteritz[2], Siqi Qiao[2], Florian Spreyer[1], Christian Schlickriede[1], Viktor Quiring[1], Christof Eigner[1], Thomas Zentgraf[1], Stefan Linden[2], Viktor Myroshnychenko[1], and Jens Förstner[1] — [1]Paderborn University, Paderborn, Germany — [2]Universität Bonn, Bonn, Germany

We show the numerical and experimental realization of optimized broadband optical traveling-wave antennas made from low-loss dielectric materials. The antennas are composed of a director and reflector placed over a glass substrate and a dipole emitter located in the feed gap between them serves as an internal source of excitation. Our studies reveal that the highly directive nature of our antennas comes from two dominant guided TE modes excited in the waveguide-like director of the antenna, in addition to the leaky modes. Furthermore, our numerical results are in excellent agreement with the experimental measurements of the antennas that were fabricated using a two-step electron beam lithography. Compared to the previously studied plasmonic antennas for photon emission, our all-dielectric approach demonstrates a new class of highly directional, low loss, and broadband optical antennas.

[1]Farheen, Henna, et al. Optimization of optical waveguide antennas for directive emission of light. JOSA B 39.1 (2022): 83-91.

[2]Farheen, Henna, et al. Broadband optical Ta2O5 antennas for directional emission of light. Optics Express 30.11 (2022): 19288-19299.

HL 33.10 Thu 17:45 H32

First-principles study of momentum-forbidden excitons in bulk 2H-MoX2 (X= S, Se) — •Ravi Kaushik[1,2] and Sergey Artyukhin[1] — [1]Italian Institute of Technology, Genova, Italy — [2]University of Genova, Genova, Italy

Coulomb-bound electron-hole pairs (excitons) dominate the optical response of atomically thin transition metal dichalcogenide (TMD) semiconductors. While Mo-based TMDs monolayers have a direct gap, bulk MoS2 and MoSe2 possess an indirect gap, with momentum-forbidden lowest energy excitonic transitions. Here we study how the effects of translational symmetry breaking by thermal phonons and in scanning spectroscopies can lead to a violation of the usual optical selection rules.