DS 12: 2D Materials 4 (joint session HL/CPP/DS)

Time: Tuesday 9:30–12:00 Location: H36

Invited Talk DS 12.1 Tue 9:30 H36

Ultrafast all-optical modulation and frequency conversion in 2D materials — •Sebastian Klimmer[1], Artem Sinelnik[1,2], Isabelle Staude[1,2], and Giancarlo Soavi[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany — [2]Institute of Applied Physics, Friedrich Schiller University Jena, Jena, Germany

Large efforts have been devoted in the last years to realizing nonlinear integrated devices for frequency conversion, sensing, signal modulation and quantum optics. Two-dimensional (2D) materials, such as graphene and transition metal dichalcogenides (TMDs), provide distinct advantages in this respect thanks to their ease of integration on photonic platforms[1] and their atomically thin nature, which relaxes phase matching constraints and thus offers a practically unlimited bandwidth for nonlinear optical effects [2]. In this seminar I will present our recent results in the field of nonlinear optics with 2D materials, including ultra-broadband four-wave mixing in the telecom range, ultrafast all-optical modulation of second- and third-harmonic generation in TMDs[3] and graphene and ultrafast polarization-resolved second-harmonic spectroscopy to probe the valley degree of freedom in TMDs.

[1] He, J. *et al.*, Nano Lett. **21**, 7, 2709-2718 (2021)
[2] Trovatello, C. *et al.*, Nat. Photonics. **15**, 6-10 (2021)
[3] Klimmer, S. *et al.*, Nat. Photonics. **15**, 837-842 (2021)

DS 12.2 Tue 10:00 H36

strain tuning of exciton and trion dynamics in monolayer WSe2 at cryogenic temperatures — •zhao an[1], pedro soubelet[2], andreas v. stier[2], michael zopf[1], yaroslav zhumagullov[3], jaroslav fabian[3], paulo e. faria jonior[3], jonathan j. finley[2], and fei ding[1,4] — [1]Institut für Festkörperphysik, Leibniz Universität Hannover, Appelstraße 2, 30167 Hannover, Germany — [2]Walter Schottky Institut and Physik Department, Technische Universität München, 85748 Garching, Germany — [3]Institute for Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany — [4]Laboratorium fur Nano- und Quantenengineering, Leibniz Universität Hannover, Schneiderberg 39, 30167 Hannover, Germany

Transition metal dichalcogenides (TMD) receive increasing attention these years. In TMD monolayers, the light-matter interaction is driven by strong excitonic effects. Additional to neutral excitons, singlet/ triplet trions are observed, in which the additional charge is either in the same or opposite valley with respect to excitons. We apply dynamic strain at cryogenic temperatures to investigate the exciton dynamics of monolayer WSe2. Biaxial strain is electrically controlled via a piezoelectric actuator and transferred to the hBN/WSe2/hBN. We find that next to changes in the emission energy and intensity, the singlet-triplet trion fine structure is affected. Polarization-resolved PL spectroscopy reveals that biaxial strain alters the polarizations of trions, which is attributed to changes in the pumping of resident electrons and the intervalley scattering of excitons and electrons.

DS 12.3 Tue 10:15 H36

Optical nonlinearities in the excited carrier density of 2d TMDs — •Daniel Erben[1], Alexander Steinhoff[1], Michael Lorke[1,2], and Frank Jahnke[1] — [1]Institute for Theoretical Physics, University of Bremen — [2]Bremen Center for Computational Materials Science, University of Bremen

The prospects of using 2d transition metal dichalcogenides (TMDs) in future optoelectronic device application requires insight in the excitation dynamics of photoexcited charge carriers and the resulting optical nonlinearities. Utilizing ab-initio electronic-state calculations combined with many-body treatment of optical excitation, we calculate the excited carrier dynamics and the nonlinear absorption in MoS_2, $MoSe_2$, WS_2, and WSe_2 under various excitation conditions.
We find, that the increase of the carrier density with excitation strength deviates from a linear behaviour. Based on this, the validity range of a linear approximation for the excited carrier density as function of the pump fluence is determined. The use of a linear absorption coefficient of the unexcited system can significantly underestimate the achievable carrier density for strong pump fields. Furthermore, we study the excitation-induced many-body effects of excited charge carriers like band-gap renormalization, dephasing, screening, and scattering processes, that are mediated by the strong Coulomb interaction. Additional contributions to optical nonlinearities originate from phase space filling.

15 min. break

DS 12.4 Tue 10:45 H36

Second order coherence of a condensate of exciton-polaritons in an atomically thin crystal — •Jens-Christian Drawer[1], Hangyong Shan[1], Sven Höfling[2], Carlos Anton-Solanas[3], Martin Esmann[1], and Christian Schneider[1] — [1]Universität Oldenburg, Germany — [2]Universität Würzburg, Germany — [3]Universidad Autónoma de Madrid, Spain

We study the second order coherence of a condensate of exciton-polaritons emerging in a microcavity loaded with an atomically thin $MoSe_2$ crystal. Under cryogenic temperatures, angle-resolved PL and reflectivity measurements reveal the formation of two polariton resonances, as the hallmark of the strong coupling regime. The characteristic condensation threshold manifests via the nonlinear input-output-characteristics of the emission. In order to gain deeper information about the photon statistics emitted from the cavity, we perform the Hanbury Brown- and Twiss experiment as a function of the polariton occupation in the effective ground state. While the emission features a bunching effect below threshold, hinting at a thermal contribution of the polariton emission, above threshold the second order correlation transits towards $g^{(2)}(\tau = 0) = 1$, which is indicative for the formation of a coherent state in the quantum optical sense.

DS 12.5 Tue 11:00 H36

Theoretical description of moiré excitons in twisted $MoSe_2$ homobilayers — •Ruven Hübner[1], Malte Kremser[2], Viviana Villafañe[2], Marko M. Petrić[3], Matthias Florian[4], Alexander Steinhoff[1], Mackillo Kira[4], Nathan P. Wilson[2], Andreas V. Stier[2], Kai Muller[3], and Jonathan J. Finley[2] — [1]Institut für Theoretische Physik, Universität Bremen, Bremen, Germany — [2]Walter Schottky Institut and Physik Department, Technische Universität München, Garching, Germany — [3]Walter Schottky Institut and Department of Electrical and Computer Engineering, Technische Universität München, Garching, Germany — [4]University of Michigan, Dept. of Electrical Engineering and Computer Science, Ann Arbor, MI, USA

By introducing a twist between multiple monolayers of transition metal dichalcogenides we can observe superstructures with a new periodicity - namely the moiré lattice. Its size depends on the twist angle and therefore offers the possibility to modify properties like exciton energies as a function of the twist angle. We demonstrate, how DFT calculations of an untwisted $MoSe_2$ bilayer allow us to locally model the band variation inside the moiré unit cell at all dominant high symmetry points of the Brillouin zone. The resulting model provides access to arbitrary moiré potentials experienced by different exciton species and allows us to calculate their twist angle dependent spectra. For all twist angles we assign the lowest energy to interlayer excitons formed between the Γ- and K-valley. The twist angle dependent shift of 5 meV per degree for small angles is in good agreement with experiment.

DS 12.6 Tue 11:15 H36

Dielectric screening effects on the exciton binding-energy and exciton diffusion in a 2D material — •Lukas Gümbel, Philip Klement, and Sangam Chatterjee — Institute of Experimental Physics I and Center for Materials Research (ZfM/LaMa), Justus Liebig University Giessen, Heinrich-Buff-Ring 16, Giessen D-35392, Germany

Two-dimensional semiconductors have proven to be candidates for numerous applications in the field of optoelectronics. Especially transition-metal dichalcogenides such as WS_2 have attracted extensive research due to the direct band-gap emerging in the monolayer limit. The optoelectronic properties are dominated by tightly-bound excitons denoted as A, B, and C. As the electric field lines of the excitonic states extend into the surrounding material, the energy states are subject to dielectric screening effects. Here we show that stronger dielectric screening equally shifts the excitonic ground state energies

of the A-, B-, and C-excitons in WS_2 to lower energies. We find a shift of 20 meV in monolayers encapsulated in hBN and observe a non-hydrogenic Rydberg-series yielding a quasiparticle band-gap energy of 2.33 eV with an 1s excitonic binding energy of 0.30 eV. Additionally, we study exciton diffusion in different dielectric environments yielding a diffusion coefficient of 9 cm^2/s. These results complement the underlying theory and may pave the way to a deeper understanding of screening effects in various 2D-Materials.

DS 12.7 Tue 11:30 H36

Brightening of a dark monolayer semiconductor via strong light-matter coupling in a cavity — •Hangyong Shan[1], Ivan Iorsh[2], Bo Han[1], Falk Eilenberger[4], Martin Esmann[1], Sebastian Klembt[3], Sven Höfling[3], Carlos Antón-Solanas[1], Ivan A. Shelykh[2], and Christian Schneider[1] — [1]Oldenburg University, Oldenburg, Germany. — [2]St. Petersburg, Russia — [3]Universität Würzburg, Würzburg, Germany — [4]Friedrich Schiller University, Jena, Germany

We study the modification of the material properties via strong coupling and demonstrate an effective inversion of the excitonic band-ordering in a monolayer of WSe2 with spin-forbidden, optically dark ground state. In our experiments, we harness the strong light-matter coupling between cavity photon and the high energy, spin-allowed bright exciton, and thus creating two bright polaritonic modes in the optical bandgap with the lower polariton mode pushed below the WSe2 dark state. We demonstrate that in this regime the commonly observed luminescence quenching stemming from the fast relaxation to the dark ground state is prevented, which results in the brightening of this intrinsically dark material. We probe this effective brightening by temperature-dependent photoluminescence, and we find an excellent agreement with a theoretical model accounting for the inversion of the band ordering and phonon-assisted polariton relaxation.

DS 12.8 Tue 11:45 H36

Broadband pump-probe microscopy at 1.5 MHz repetition rate — •Devapriyo Mithun[1], Michael Frosz[2], and Giancarlo Soavi[1] — [1]Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany — [2]Max Planck Institute for the Science of Light, Erlangen, Germany

Ultrafast pump-probe spectroscopy is one of the most commonly used techniques to resolve photoinduced excited states dynamics: a pump pulse excites the system under investigation, which is then monitored by measuring the changes in the differential reflection ($\Delta R/R$) of a temporally delayed probe pulse. Here, we discuss the realization of a pump-probe setup, which exhibits high sensitivity operating with a temporal resolution of $\approx$ 100 fs and spatial resolution of $\approx$ 3 μm with 515 nm pump and a broadband probe spectrum in the range 650-1000 nm, generated with a photonic crystal fiber.

We modulate the pump pulse at 1.5 MHz using an acousto-optic modulator. By doing this, we achieve a sensitivity, defined as the minimum detectable $\Delta R/R$, of 10^{-7} at 10 ms integration time. Finally, we implemented a Fourier transform based interferometric detection scheme to achieve a fast measurement of $\Delta R/R$ over the entire broadband spectrum.

Our pump-probe setup provides a powerful tool for broadband pump-probe microscopy with high sensitivity and high temporal resolution, which is ideal for the study of nanostructures such as carbon nanotubes and layered materials.