DS 7: 2D Materials 2 (joint session HL/CPP/DS)

Time: Monday 15:00–18:30 Location: H36

DS 7.1 Mon 15:00 H36

On-demand light emission from helium ion induced defects in atomically thin WS_2 — •Nina Pettinger, Ana Micevic, Alexander Hötger, Christoph Kastl, and Alexander Holleitner — TU Munich, Germany

Optically active defects created with a helium ion microscope (HIM) propose the possibility for structuring and tailoring quantum emitters on an atomistic scale [1]. We introduce the generation of positioned defects in encapsulated monolayer WS_2 with a HIM. The HIM induced defects exhibit sharp photoluminescence emission in the energy range of 1.55 to 1.79 eV.

[1] J. Klein and L. Sigl et al., ACS Photonics 8, 669 (2021).

DS 7.2 Mon 15:15 H36

Concept of an all-optical THz near-field microscope for flakes of 2D materials — •Ahmad-Reza Etemadi, Sebastian Matschy, Ahana Bhattacharya, and Martin Mittendorff — Department of physics, University of Duisburg-Essen, 47057 Duisburg, Germany

While THz spectroscopy is an excellent tool to investigate the free charge carriers in many semiconducting materials, the long-wavelength is an inherent feature linked to a large spot size in the millimeter range, and thus large samples are required. Small flakes of two-dimensional materials exfoliated from bulk crystals are usually much smaller than the spot size of a conventional THz spectrometer. The direct detection of the THz signal in the vicinity of the flake gains the phase and amplitude information with a higher spatial resolution. This is accessible by placing the sample directly on top of an electro-optic crystal. Sampling the THz field at the flake position gives access to the complex conductivity and thus the carrier density as well as the carrier mobility. A frequency-doubled fiber laser with a pulse duration of about 80 fs at 780 nm is exploited to generate and sample the THz field. GaSe, and ZnTe are employed as electro-optic crystals. Here we present the current state of the near-field microscope and the first measurements of the spatial resolution. The experimental results are accompanied by rigorous modeling of the THz propagation within the electro-optic crystal.

DS 7.3 Mon 15:30 H36

Ab initio description of valley-selective circular dichroism — •Maximilian Schebek[1], Yiming Pan[2], Cecilia Vona[1], Claudia Draxl[1], and Fabio Caruso[2] — [1]Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin, Germany — [2]Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, Kiel, Germany

By enabling control of valley degrees of freedom, valley-selective circular dichroism (VSCD) has become a key concept in valleytronics. In this work, we present an *ab initio* many-body theory of VSCD based on the Bethe-Salpeter equation. Our approach provides a new route to accurately predict the degree of valley polarization upon absorption of circularly polarized light. With the example of monolayer transition-metal dichalcogenides, we further show that valley excitons - bound electron-hole pairs formed at either the K or $\overline{\text{K}}$ valley upon absorption of circularly-polarized light - are chiral quasiparticles characterized by a finite orbital angular momentum (OAM). Beside governing the interaction with circularly polarized light, the OAM results in a finite magnetization of excitons, which in turn provides a route for the interaction of excitons with external magnetic fields and other spin-orbital degrees of freedom.

DS 7.4 Mon 15:45 H36

Dark and bright exciton dynamics probed by time-resolved photoluminescence in hBN-encapsulated $MoWSe_2$ monolayers — •Julian Schröer[1,3], Joanna Kutrowska-Girzycka[2], Leszek Bryja[2], Joanna Jadczak[2], and Jörg Debus[1] — [1]TU Dortmund, Experimentelle Physik 2, AG Debus — [2]Wroclaw University of Science and Technology, Department of Experimental Physics — [3]Universität Rostock, Institut für Physik, AG Korn: Zweidimensionale Kristalle und Heterostrukturen

Semiconducting monolayers of ternary $MoWSe_2$ alloys combine the unique properties of the binary transition metal dichalcogenide (TMDC) materials $MoSe_2$ and WSe_2. The alloying leads to, for example, brightening of the momentum- and spin-forbidden dark exciton states. Detailed studies on the dynamics of these brightened dark states are missing. We report on the exciton and trion formation lying in the 1-3 ps range, while the decay time approaches hundreds picoseconds. Additionally, strong dependences on the temperature and exciting laser light polarization are observed. In time-resolved and stationary photoluminescence measurements, we reveal the impact of the crystal disorder potential on the exciton properties. The polarization dynamics of the exciton and trion photoluminescence indicate possible contributions from chiral phonons as well as electrons and holes from different valleys of the Brillouin zone. Our work is a further step towards a deeper understanding of the dynamics of dark excitons in TMDC materials.

15 min. break

DS 7.5 Mon 16:15 H36

Signatures of a degenerate many-body state of interlayer excitons in a van der Waals heterostack — •Johannes Figueiredo[1], Lukas Sigl[1], Florian Sigger[1], Jonas Kiemle[1], Mirco Troue[1], Ursula Wurstbauer[2], and Alexander Holleitner[1] — [1]Walter-Schottky-Institut, Technical University of Munich — [2]Institute of Physics, Westfälische Wilhelms-Universität Münster

In atomistic van der Waals heterostacks of transition metal dichalcogenides, the reduced dimensionality and changing dielectric environement leads to the formation of stronlgy bound excitons. Optically generated interlayer excitons exhibit an additional spatial separation of the electron-hole pair with a reduced overlap of the electrons' and holes' wave-functions, evidenced through their long lifetimes. These long-lived, photogenerated composite bosons yield several signatures of a quantum degenerate many-body system at cryogenic temperatures. The emergence of this state is in accordance with theoretical predictions of a critical condensation temperature above 10K. We present new insights into the phase-diagram of such interlayer exciton ensembles. [1]

[1] L. Sigl *et al.*, Phys. Rev. Research 2, 042044(R) (2020)

DS 7.6 Mon 16:30 H36

exciton species in highly doped WS_2 monolayers — david tiede, •hossein ostovar, hendrik lambers, nihit saigal, and ursula wurstbauer — Institute of Physics, University of Münster, Münster, Germany

Semiconducting two-dimensional transition metal dichalcogenides such as WS_2 excel due to their exciton dominated light-matter interaction even at room temperature (RT) that is highly tunable by external stimuli such as doping, light excitation, dielectric environment, or strain [1]. In this work, an optimized field effect structure utilizing a polymer electrolyte top gate electrode is employed to study the evolution of the optical response in monolayer WS_2 at RT in dependence of doping by means of photoluminescence and spectroscopic imaging ellipsometry measurements. The huge geometrical gate capacitance enables capacitance spectroscopy of the conduction band as well as valence band edge yielding a gap energy of 2.6eV in agreement with the determination from the exciton Rydberg series. The gate allows the injection of large electron and hole densities exceeding $10^{14} cm^{-2}$, sufficient to enable the exciton Mott transition. The obtained doping dependent emission and absorption spectra also facilitate the identification of phonon activated, neutral and charged exciton species as well as dressed excitons in a fermi sea. We acknowledge financial support via DFG WU 637/7-1 and SPP2244. [1] U. Wurstbauer et al. J. Phys. D: Appl. Phys. 50, 173001 (2017).

DS 7.7 Mon 16:45 H36

Pump probe signatures of interlayer excitons in TMDC heterostructures — •Henry Mittenzwey, Manuel Katzer, Andreas Knorr, and Malte Selig — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

TMDC heterobilayers are promising candidates for novel optoelectronic applications, since they exhibit long-lived excitonic states with spatially separated electrons and holes located in different layers. The relaxation dynamics of these interlayer excitons and their interplay with intralayer excitons are still under investigation.

Here, we present a microscopic description for the phonon and tunneling induced formation and relaxation of intra- and interlayer excitons in a $MoSe_2/WSe_2$ stack. Based on the microscopic dynamics we calculate the pump probe signal for intra- and interlayer transition and their population dynamics including hot exciton bottleneck effects.

DS 7.8 Mon 17:00 H36

Angle- and polarization-resolved luminescence from suspended and hBN encapsulated $MoSe_2$ monolayers — •Bo Han[1], Sven Stephan[1], Joshua J.P. Thompson[2], Martin Esmann[1], Carlos Antón-Solanas[1], Hangyong Shan[1], Samuel Brem[3], Christoph lienau[1], Kenji Watanabe[4], Takashi Taniguchi[4], Martin Silies[1], Ermin Malic[2,3], and Christian Schneider[1] — [1]Carl von Ossietzky Universität, Oldenburg, Germany. — [2]Philipps Universität, Marburg, Germany. — [3]Chalmers University of Technology, Gothenburg, Sweden. — [4]National Institute for Materials Science, Tsukuba, Japan.

We apply combined angle- and polarization-resolved spectroscopy to explore the interplay of excitonic physics and phenomena arising from the commonly utilized encapsulation on the optical properties of atomically thin transition metal dichalcogenides. In our study, we probe $MoSe_2$ monolayers which are prepared in both a suspended and an encapsulated manner. We show that the hBN encapsulation significantly enhances the linear polarization of exciton PL at large emission angles. This degree of linear polarization of excitons can increase up to 17 % in the hBN encapsulated samples. As confirmed by finite-difference time-domain simulations, it can be directly connected to the optical anisotropy of the hBN layers. In comparison, the linear polarization at finite exciton momenta is significantly reduced in suspended $MoSe_2$ monolayer, and only becomes notable at cryogenic conditions. This phenomenon strongly suggest that the effect is rooted in the k-dependent anisotropic exchange coupling inherent in 2D excitons.

15 min. break

DS 7.9 Mon 17:30 H36

Photonic and Phononic Couplings in Hybrid High-Q Nanocavities with Encapsulated MoS2 Monolayer — •Chenjiang Qian[1], Viviana Villafañe[1], Pedro Soubelet[1], Alexander Hötger[1], Takashi Taniguchi[2], Kenji Watanabe[2], Nathan Wilson[1], Andreas Stier[1], Alexander Holleitner[1], and Jonathan Finley[1] — [1]Walter Schottky Institut and Physik Department, Am Coulombwall 4, 85748 Garching, Germany — [2]National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

Monolayer TMDs are ideal active materials for solid-state cQED. However, the direct cou-pling of TMDs to 0D nanocavities whilst preserving pristine excitonic properties and large cavi-ty-TMD overlap remains a challenge. Most commonly, non-encapsulated TMDs are stacked on top of prefabricated photonic structures using pick-and-place assembly. In this case, environ-mental disorders strongly perturb the excitonic properties. Whilst disorder can be mitigated by full hBN encapsulation, this approach moves the TMD away from the cavity field, thereby, trad-ing spatial coupling for homogeneous linewidth. Here, we integrate hBN/MoS2/hBN heterostruc-tures to Si3N4 nanobeams as hybrid nanocavities. Our approach solves the trade-off problem by making the unpatterned heterostructure a functional part of the cavity field. Therefore, the pris-tine excitonic quality, high cavity mode Q-factor $>$ 10000, and the strong cavity-MoS2 overlap are achieved simultaneously. We study the coupling of MoS2 excitons to the cavity optical and vibrational modes using PL and Raman spectroscopy, and novel coupling phenomena are ob-served based.

DS 7.10 Mon 17:45 H36

Terahertz free carrier absorption to modulate the optical properties of nanometer-thick van der Waals semiconductors — •Tommaso Venanzi[1,2], Malte Selig[3], Alexej Pashkin[2], Stephan Winnerl[2], Manuel Katzer[3], Himani Arora[2], Artur Erbe[2], Amalia Patane[4], Zakhar R. Kudrynskyi[4], Zakhar D. Kovalyuk[5], Leonetta Baldassarre[1], Andreas Knorr[3], Manfred Helm[2], and Harald Schneider[2] — [1]Sapienza University of Rome, 00185 Rome, Italy — [2]Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany — [3]Technical University Berlin, 10623 Berlin, Germany — [4]University of Nottingham, Nottingham NG7 2RD, UK — [5]The National Academy of Sciences of Ukraine, 58001 Chernivtsi, Ukraine

Free carriers in doped semiconductors absorb terahertz radiation when the frequency of the electromagnetic field is lower or comparable to the plasma frequency of the system. This phenomenon can be used to manipulate the optical response of the material. We present here the results of two different experiments performed at the infrared free-electron laser FELBE on atomically-thin van der Waals semiconductors. In MoSe2 monolayers, we observe a terahertz-induced redshift of the trion resonance. Terahertz absorption induces an average high momentum to the carriers and this momentum gets transferred during the trion formation, resulting in a net redshift in the absorption. In few-layer InSe, the terahertz pulses induce a transient quenching of the photoluminescence emission. In both cases, a microscopic study of the hot carrier distribution cooling is also presented.

DS 7.11 Mon 18:00 H36

Theory of Exciton-Phonon Interaction for Stationary State Experiments in Atomically Thin Semiconductors — •Manuel Katzer, Andreas Knorr, and Malte Selig — Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

Atomically thin semiconductors exhibit tightly bound electron hole pairs which stimulated exciton research in recent years [1]. While typical experimental techniques include the cw excitation of the material, only few is known theoretically about the related exciton dynamics and the formation of non-equilibrium steady states. Based on excitonic Boltzmann scattering equations, we demonstrate that the formation of such stationary states is also accompanied with the formation of phonon replica in the photoluminescence excitation spectrum [2], in agreement with available experiments [3]. So far, many studies focused on the understanding of exciton dynamics in the limit of weak excitation. Above this limit, we find both bosonic but also fermionic contributions to the thermalization, due to the co-bosonic nature of excitons. Based on a Heisenberg equation of motion ansatz [4], we discuss the first order of non-linear exciton-phonon interaction exceeding the classical Boltzmann scattering limit, in order to analyze the exciton thermalization at elevated excitation densities.
[1] Wang et al. RMP, 90, 021001 (2018). [2] Selig et al. arXiv:2201.03362 (2022). [3] Chow et al., Nano lett. 17, 1194 (2017); Shree et al. PRB 98, 035302 (2018). [4] Selig et al. PRR, 1, 022007 (2019); Katsch et al., PRL 124 25 257402 (2020).

DS 7.12 Mon 18:15 H36

Ultrafast control of spins in transition metal dichalcogenides — •Abhijeet Kumar[1], Denis Yagodkin[1], Douglas J. Bock[1], Nele Stetzuhn[1,2], Sviatoslav Kovalchuk[1], Alexey Melnikov[3], Peter Elliott[2], Sangeeta Sharma[2], Cornelius Gahl[1], and Kirill I. Bolotin[1] — [1]Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany — [2]Max-Born-Institut für Nichtlineare Optik und Kurzzeitspektroskopie, Max-Born Straße 2a, 12489 Berlin, Germany — [3]Institute for Physics, Martin Luther University Halle, 06120 Halle, Germany

Control and manipulation of the coupled spin/valley degrees of freedom in transition metal dichalcogenides (TMDs) are essential for their applications in spin/valleytronics. Here, we achieve ultrafast control of spins in TMDs via two distinct approaches, namely, proximity-coupling to another TMD and strain. First, we use a type-II heterostructure $MoS_2 - MoSe_2$ to enable directional optical pumping of spin-polarized carriers. We find that the photoexcited carriers conserve their spin for both tunneling directions across the interface. We observe dramatic differences in the spin/valley depolarization rates for electrons and holes, 30 and $<1\ ns^{-1}$, respectively, which relates to the disparity in the spin-orbit splitting in conduction and valence bands of TMDs. Second, by applying biaxial strain (exceeding 2%) in monolayer WSe_2, we evidence the hybridization of the conduction bands with the in-gap localized defects that brightens the lowest-lying dark excitons. This novel hybrid state exhibits unique spin/valley signatures which are strongly manipulated on picosecond-timescale by strain and doping.