HL 9: 2D Materials and Heterostructures: Interlayer Excitons

Time: Monday 15:00–18:30 Location: EW 201

HL 9.1 Mon 15:00 EW 201

New interlayer excitons in 2D bilayers revealed under strong electric field — •Sviatoslav Kovalchuk[1], Kyrylo Greben[1], Abhijeet Kumar[1], Simon Pessel[1], Jan Soyka[2], Qing Cao[2], Kenji Watanabe[3], Takashi Taniguchi[3], Dominik Christiansen[4], Malte Selig[4], Andreas Knorr[4], Siegfried Eigler[2], and Kirill Bolotin[1] — [1]Physics Department, FU Berlin, Berlin — [2]Institute of Chemistry and Biochemistry, FU Berlin, Berlin — [3]National Institute for Materials Science, Tsukuba, Japan — [4]Physics Department, TU Berlin, Berlin

Excitons in bilayer transition metal dichalcogenides (2L-TMDs) are Coulomb-bound electron/hole pairs that can be viewed as broadly tunable analogs of atomic or molecular systems. Here, we study the properties of 2L-TMD excitons under a strong electric field. To overcome the field limit reached in previous experiments, we developed a new organic/inorganic molecular gating technique. This approach allows us to achieve an electric field strength of about 0.35 V/nm, more than a factor of two higher than achieved previously in purely solid-state gated devices. Under this field, inter- and intralayer excitons are brought into an energetic resonance, allowing us to discover new emergent properties of the resulting states. We detect a previously unseen interlayer exciton that only becomes visible at high field through hybridization with A exciton. Moreover, the system experiences an ultra-strong Stark splitting of $>$ 380 meV with exciton energies tunable over a large range of the optical spectrum, holding potential for optoelectronics.

HL 9.2 Mon 15:15 EW 201

Signatures of efficient intervalley scattering by acoustic phonons in WSe2/MoSe2 heterostructure — •Hendrik Lambers[1], Nihit Saigal[1], Jonas Kiemle[2], Alexander W. Holleitner[2], and Ursula Wurstbauer[1] — [1]Institute of Physics, University Münster, Germany — [2]Walter Schottky Institute and Physics Department, TU Munich, Germany

The ability to host excitonic phenomena and correlated phases gives rise to many recent studies on TMDC bilayers. In particular interlayer excitons in heterobilayers are promising candidates to form coherent many body states [1]. The role of exciton-phonon interaction for the thermalization process of these interlayer excitons and the dominant type of involved phonons are of ongoing interest [2]. We employ resonant Raman scattering at cryogenic temperatures to study the exciton phonon coupling in WSe2/MoSe2 heterostructures. The resonance profiles of degenerated WSe2 A1'/E' phonon modes are significantly affected by the assembly into heterobilayers. The profiles cannot be explained only by an incoming and an outgoing resonance of a first order Raman process. Our findings indicate a higher order Raman scattering process involving acoustic M or K point phonons of WSe2 and therefore strong interaction of acoustic phonons with electronic states, that results in efficient intervalley scattering of charge carriers. [1] M. Troue et al., Phys. Rev. Lett. 131, 036902 (2023). [2] M. Katzer et al., Phys. Rev B 108, L121102 (2023).

HL 9.3 Mon 15:30 EW 201

Emergent Trion-Phonon Coupling in Atomically Reconstructed $MoSe_2$-WSe_2 Heterobilayers — •P. Parzefall[1], S. Meier[1], Y. Zhumagulov[2], M. Dietl[1], N. Meier[1], J. Lichtenberger[1], M. Kempf[1], J. Holler[1], P. Nagler[1], K. Watanabe[3], T. Taniguchi[3], P. E. Faria JR.[2], J. Fabian[2], T. Korn[4], and C. Schüller[1] — [1]Institut für Exp. und Angewandte Physik, Uni Regensburg (UR), Germany — [2]Institut für Theo. Physik, UR, Germany — [3]NIMS, Tsukuba Ibaraki, Japan — [4]Institut für Physik, Uni Rostock, Germany

We report about low-frequency Raman studies on atomically-reconstructed $MoSe_2$-WSe_2 heterostructures (HS) at cryogenic temperatures. In these experiments, we tune a Ti:Sapphire laser into close resonance to the intralayer excitonic transitions of both, $MoSe_2$ and WSe_2, and compare the resulting Raman spectra to the well-known off-resonant case [1]. We detect a low-enery Raman mode, which is observable only at the intralayer trion (X^-) resonances. The energy of the emergent Raman mode is close to the interlayer shear mode (ISM) of the HS [1] but it exhibits different polarization selection rules and a lineshape, resembling a Fano-profile.

We interpret our results as follows: The X^- gains an interlayer character via its second electron, which is located in the Q valley and distributed over both layers, and which enables the coupling to the interlayer shear mode. [1] J. Holler et al., APL **117**, 013104 (2020) [2] S. Meier et al., Phys. Rev. Res. **5**, L032036 (2023)

HL 9.4 Mon 15:45 EW 201

Spin Relaxation of Interlayer Excitons in TMDC Heterostructures — •Henry Mittenzwey[1], Abhijeet Kumar[2], Kirill Bolotin[2], Malte Selig[1], and Andreas Knorr[1] — [1]Technische Universität Berlin, Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Hardenbergstraße 36, 10623 Berlin, Germany — [2]Department of Physics, Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

TMDC heterobilayers are promising candidates for new excitonic phases, since they exhibit long-lived excitonic states with spatially separated electrons and holes located in different atomically thin layers. The relaxation dynamics of these interlayer excitons after their optical excitation is still under investigation.

Based on Heisenberg equations of motion and a correlation expansion of many-body interactions, we discuss the temporally resolved spin dynamics of interlayer excitonic occupations in momentum space: Due to charge separation, interlayer excitons give rise to an out-of-plane self-induced electric field. This leads to Rashba spin-orbit interaction, which is absent in TMDC monolayers. By tuning the total excitonic interlayer occupation, the strength of the internal electric field can be varied and the Rashba spin relaxation can be enhanced or suppressed.

To address the experimental verification of these effects, we discuss, how spin relaxation dynamics of interlayer excitons can be detected in optical pump probe measurements.

HL 9.5 Mon 16:00 EW 201

Impact of hybrid exciton-exciton interactions on transport in 2D materials — •Daniel Erkensten[1], Samuel Brem[2], Raul Perea-Causin[1], Joakim Hagel[1], Fedele Tagarelli[3], Edoardo Lopriore[3], Ermin Malic[2,1], and Andras Kis[3] — [1]Chalmers University of Technology — [2]Philipps-Universität Marburg — [3]Ecole Polytechnique Fédérale de Lausanne

Transition-metal dichalcogenide bilayers host exciton states that hybridize via electron or hole tunneling, forming layer-hybridized excitons with large oscillator strengths and out-of-plane dipole moments. In this joint theory-experiment work, we combine microscopic many-particle theory with spatiotemporal measurements to investigate the role of hybrid exciton-exciton interactions in exciton transport in WSe_2 homobilayers [1,2]. The energetically lowest state in these structures is shown to be electrically tunable, transitioning from an intralayer-like to an interlayer-like ground state via the application of an out-of-plane electric field [1]. This finding leads to two intriguing interaction regimes for hybrid excitons: a low-dipole regime at small electric fields involving weakly interacting excitons which exhibit conventional diffusion, and a high-dipole regime at elevated electric fields governed by strongly interacting excitons and highly anomalous diffusion. Our work highlights the remarkable electrical tunability of hybrid exciton-exciton interactions, providing insights for future research in this evolving field.

[1] D.Erkensten et al. Nanoscale, 15, 11064-11071 (2023).
[2] F. Tagarelli, D. Erkensten et al. Nat. Photonics, 71, 615-621 (2023).

HL 9.6 Mon 16:15 EW 201

Manipulation of hybrid interlayer excitons in 2D materials — •M. Federolf, A. Davgol, E. Flanagan, M. Emmerling, V. Kaushik, and S. Höfling — Julius-Maximilians-Universität Würzburg, Lehrstuhl für Technische Physik

It has been shown, that in two layers of transition-metal dichalcogenides hybrid interlayer exciton can form by the coupling of the interlayer exciton with the intralayer exciton [1]. Here the electron in one layer interacts with a tunneling hole located betwen the layers. This configuration exhibits an out of plane dipole moment while maintaining a large oscillator strength due to the hybrid character. The degeneration of the dipole orientation can be lifted using an external electric field leading to a splitting into two distinct states [2,3].

Further, these hybrid interlayer excitons promise enhanced interaction with themselves compared to the A exciton. By introducing a higher density, the blueshift of the hybrid interlayer polaritons exceeds the blueshift of the A polaritons [4,5].

We try to combine those effects by introducing higher exciton densities while simultaneously applying an out of plane electric field and investigating the interaction of the different excitons.

References [1] Gerber, I.C., et al. Phys. Rev. B 99, 035443 (2019). [2] Lorchat, E., et al., Phys. Rev. Lett. 126, 037401 (2021). [3] Leisgang, N., et al., Nat Nano 15, 901*907 (2020). [4] Datta, B., et al., Nat Comm 13, 6341 (2022). [5] Louca, C., et al., Nat Comm 14, 3818 (2023).

15 min. break

HL 9.7 Mon 16:45 EW 201

Extended spatial coherence of interlayer excitons in $MoSe_2/WSe_2$ heterobilayers — •Johannes Figueiredo[1,2], Mirco Troue[1,2], Lukas Sigl[1,2], Christos Paspalides[1,2], Manuel Katzer[3], Malte Selig[3], Andreas Knorr[3], Ursula Wurstbauer[4], and Alexander Holleitner[1,2] — [1]Walter Schottky Institute, TU Munich — [2]MCQST — [3]Institute for Theoretical Physics, Nonlinear Optics and Quantum Electronics, TU Berlin — [4]Institute of Physics, Münster University

A heterobilayer arrangement of $MoSe_2/WSe_2$ monolayers allows the optical generation of long-lived electron-hole pairs known as interlayer excitons [1]. Consequently, those heterostructures present an ideal platform to study optically excited many-body phenomena of an interacting Bose gas at low bath temperatures [2]. Our work advances the study of coherent, photoluminescent interlayer exciton ensembles in $MoSe_2/WSe_2$ heterobilayers by utilizing a spatially resolved point-inversion Michelson-Morley interferometer. We report on the spatial coherence of a dense interlayer exciton ensemble for a wide range of temperatures and exciton densities. Below 10 K, we detect a spatial coherence length of interlayer excitons limited only by the lateral expansion of the exciton ensembles. Our research sheds new light on the physics of coherent exciton phases in the proposed heterostacks [3].

[1] L. Sigl *et al.*, Phys. Rev. Research 2, 042044(R) (2020) [2] M. Katzer *et al.*, Phys. Rev. B 108, L121102 (2023) [3] M. Troue and J. Figueiredo *et al.*, Phys. Rev. Lett. 131, 036902 (2023)

HL 9.8 Mon 17:00 EW 201

Theory of interlayer excitons dynamics in 2D TMDCs Heterolayers including reconstruction through strain and disorder — •Marten Richter — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, Berlin, Germany

After combining two TMDC monolayers to form a heterolayer small angles between the lattices lead to the formation of Moiré structures.

Recently it has been shown, that strain leads to a reconstruction of the domains formed by the Moiré structure. This contribution uses reconstructed strain fields with some disorder as a basis to calculate the potential landscape of electrons and holes forming interlayer excitons.

Opposed to the common solution of the center-of-mass exciton wave function in reciprocal space, here the wave function is solved in real space to accommodate disorder and to provide additional insights into the structure of exciton states. Using the obtained exciton basis exciton-phonon scattering elements and lineshapes are calculated after a polaron transformation, which allows the calculation of exciton relaxation and realistic optical signals such as linear absorption, PL.

HL 9.9 Mon 17:15 EW 201

The impact of twist angle and band alignment on the excitation of valley-polarized free charge carriers in WSe2/MoSe2 heterobilayers — •Jo Bertram[1], Marc Schütte[1], Frank Volmer[2], Manfred Ersfeld[1], Lars Rathmann[1], Kenji Watanabe[3], Takashi Taniguchi[4], Christoph Stampfer[1], Lutz Waldecker[1], and Bernd Beschoten[1] — [1]2nd Institute of Physics and JARA-FIT, RWTH Aachen University, Germany — [2]AMO GmbH, Advanced Microelectronic Center Aachen (AMICA), Aachen, Germany — [3]Research Center for Functional Materials, NIMS, Tsukuba, Japan — [4]International Center for Materials Nanoarchitectonics, NIMS, Tsukuba, Japan

Transition metal dichalcogenides (TMDs) are of interest in valley- and spintronics due to the fact that a valley polarization in these materials can be created by circularly polarized light [1]. Here, we show that the band alignment in heterostructures of different TMD monolayers enables new ways of transferring the valley polarization between the layers. In particular, we identify a transfer process in WSe2/MoSe2 heterostructures where the polarization from excitons in WSe2 is transferred to free charge carriers in MoSe2, exhibiting valley lifetimes that are strongly dependent on the twist angle [2]. Furthermore, we show that a variation in the doping of the TMDs as well as the addition of an hBN spacer layer significantly impacts the band alignment of the heterostructure, resulting in fundamentally different valley dynamics.

[1] Nature Reviews Materials 7, 449 (2022), [2] npj 2D Mater Appl 7, 58 (2023)

HL 9.10 Mon 17:30 EW 201

Investigating moiré interlayer excitons under the influence of atomic reconstructions — •Nils-Erik Schütte[1], Carl Emil Mørch Nielsen[2], Niclas Götting[1], Frederik Lohof[1], Gabriel Bester[2], and Christopher Gies[1] — [1]Institute for Theoretical Physics, University of Bremen — [2]Institute of Physical Chemistry, University of Hamburg

The moiré pattern which emerges due to a relative rotation between two monolayers of transition metal dichalcogenides (TMDs) features a long lattice period for small twist angles. The resulting band structure modulation acts as an effective potential for interlayer excitons (IXs). However, lattice reconstructions change the moiré potential which forms broader and deeper potential minima, realizing a periodic array of quantum wells for IXs. This serves as an implementation of the Bose-Hubbard (BH) model for the simulation of correlated excitonic states.

Expanding on previous results [1], we describe the correlated behavior of IXs in the moiré lattice with an extended BH model taking into account non-local interactions and a material realistic modelling of the dielectric screening. Considering interatomic forces, we discuss how the BH parameters and especially the excitonic wave functions are influenced by local atomic reconstructions. Furthermore, by solving the BH model with a sublattice mean-field description, we address the question in how far correlated states of moiré excitons emerge at different twist angles and integer as well as fractional lattice fillings.

[1] Götting et al., Phys. Rev. B 105, 165419 (2022)

HL 9.11 Mon 17:45 EW 201

Pressure Dependence of Intra- and Interlayer Excitons in 2H-MoS_2 Bilayers — •Jan-Hauke Graalmann[1], Paul Steeger[2], Robert Schmidt[2], Ilya Kupenko[3], Carmen Sanchez-Valle[3], Philipp Marauhn[1], Thorsten Deilmann[1], Steffen Michaelis de Vasconcellos[2], Michael Rohlfing[1], and Rudolf Bratschitsch[2] — [1]University of Münster, Institute of Solid State Theory, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany — [2]University of Münster, Institute of Physics and Center for Nanotechnology, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany — [3]University of Münster, Institute of Mineralogy, Corrensstr. 24, 48149 Münster

The optical spectrum of the MoS_2 bilayer changes under pressure as theoretical and experimental studies have shown [1].

Our computational investigations of the structural, electronic and optical properties are based on elasticity theory, DFT, GdW and the Bethe-Salpeter equation. The stress conditions in our diamond anvil cell experiment result in an effective shift of the excitation energy of the A exciton towards higher energies with increasing pressure. This shift corresponds strongly to the behavior of the direct band gap at the K point. Due to a growing valence band splitting for increasing pressures, the interlayer exciton shows a smaller shift. In total, the A-IL energy splitting decreases under pressure.

Furthermore, the theoretical reproduction of the experimental results shows a suppression of the transfer of hydrostatic pressure due to the interaction between the bilayer and the substrate.

[1] P. Steeger, J.-H. Graalmann et al., Nano Lett., 23, (2023)

HL 9.12 Mon 18:00 EW 201

Correlated Magnetism in Moiré Exciton-Polaritons — •Lukas Lackner[1], Johannes Scherzer[2], Bo Han[1], Jens-Christian Drawer[1], Christoph Bennenhei[1], Martin Esmann[1], Alexander Högele[2], and Christian Schneider[1] — [1]Carl von Ossietzky University Oldenburg, Germany — [2]Ludwig-Maximilians-Universität München, Germany

Due to the reduced dielectric screening in van der Waals materials, Coulomb interactions and many-body correlations are enhanced. This allows for a large variety of experimentally observables such as Wigner crystals, quantum Hall phases and Hubbard-correlated magnetic phases. In Moiré lattices, due to the periodic potential landscape and thus the reduced kinetic energy of confined quasi-particles, correlations display their fingerprints more prominently. In this work, we present the evolution of a correlation induced magnetic phase of Moiré confined exciton-polaritons based on a charge tunable MoSe2/WS2 van der Waals heterostructure in a spectrally tunable open microcavity. Approaching the Mott state of the triangular Moiré lattice at a filling of one electron per lattice site, Moiré exciton-polartions acquire a giant, nonlinear g-factor reflecting the emergence of correlated magnetism in our electron system.

HL 9.13 Mon 18:15 EW 201

Theoretical description of ultraviolet interlayer trions in bilayer 2H-WSe$_2$ — •Ruven Hübner[1], Alexander Steinhoff[1], and Matthias Florian[2] — [1]Institut für Theoretische Physik, Universität Bremen, Germany — [2]University of Michigan, Dept. of Electrical Engineering and Computer Science, Ann Arbor, MI, USA

For an extended period, interlayer excitons in bilayers of transition metal dichalcogenides (TMDs) have received a great deal of attention. Numerous studies have explored the influence of material combination, stacking configuration, twist angle and external electric fields. However, most such studies exclusively focus on excitons, formed near the band edge, confined to electronic states with a parabolic dispersion and a homogeneous interlayer character. More recently, experimental findings have unveiled ultraviolet excitons, involving high-lying and non-parabolic bands of TMD materials at approximately double the band gap energy [1]. These states open up the possibility of forming new kinds of trions, in which the additional charge carrier is provided by doping electrons near the band edge. In this talk we focus on such high-lying trions formed within a WSe$_2$ inversion symmetric homobilayer. We develop a theoretic framework, which classifies distinct trion species based on their dominant interlayer localization of each of the three constituent particles, starting from Wannierized electronic states based on a DFT calculation. We discuss the influence of doping density and use our results to explain very recent experiments.

[1] KQ. Lin et al., Nature Communications **12**, 5500 (2021)