HL 59: 2D Semiconductors and van der Waals Heterostructures VI

The sesssion covers the physics of interlayer excitons and allied phenomena in van der Waals heterostructures.

Time: Friday 9:30–11:45 Location: H15

HL 59.1 Fri 9:30 H15

Interlayer excitons in electric and magnetic fields — •Thorsten Deilmann — Institute of Solid State Theory, University of Münster, Germany

Over the past years, more and more two-dimensional (2D) materials have been stacked to heterostructures. Due to the coupling interlayer excitations can form.

Here we report on a trilayer heterostructure of MoS_2 and WS_2. In contrast to bilayers, the coupling may lead to quadrupolar excitons. These can be clearly identified by the quadratic response of the corresponding excitons in our GW/BSE calculations in electric fields. In the second example, we report our studies of bulk CrSBr [1]. In a magnetic field the lowest bright excitations of the antiferromagnetically coupled CrSBr shift quadratically. Our calculations reveal the symmetry forbidden dark excitons and the increasing interlayer character. We develop a minimal model to explains this spin-dependent coupling. Despite its simplicity, the proposed model is generally applicable to any coupled 2D magnet.

[1] https://arxiv.org/abs/2403.20174

HL 59.2 Fri 9:45 H15

Signature of Rydberg-like states of interlayer excitons in MoSe2/WSe2 heterostructures — •Chirag Chandrakant Palekar[1], Paulo E. Faria Junior[2], Tobias Manthai[1], Maximilian Nagel[1], Bhabani Sankar Sahoo[1], Shachi Machchhar[1], Avijit Barua[1], Jaroslav Fabian[2], Barbara Rosa[1], and Stephan Reitzenstein[1] — [1]Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstrasse 36, 10623 Berlin, Germany — [2]Institute for Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany

The heterostructures of transition metal dichalcogenides (TMDCs), formed by the stacking of different TMDC monolayers (MLs), facilitates the formation of spatially indirect interlayer excitons (IX). Due to the higher binding energy, the excited excitonic states have typically been observed in TMDC monolayers for over a decade. Nevertheless, the Rydberg-like higher-order states of the IX in WSe2/MoSe2 HSs have thus far remained undetected due to their weak oscillator strength. In this work, we employ photoluminescence excitation (PLE) spectroscopy to identify the signatures of Rydberg-like higher order states of IX in WSe2/MoSe2 HSs. By examining HSs with varying twist angles, we are able to gain comprehensive insight into the twist angle dependence of such excited states of IX. We compare experimental and theoretical results on the twist angle-dependent behavior of observed states, providing a systematic investigation that advances current understanding of the optical and electronic properties of TMDC HS systems.

HL 59.3 Fri 10:00 H15

Twisted MoSe2 Homobilayer Behaving as a Heterobilayer — •Arka Karmakar — Institute of Experimental Physics, Faculty of Physics, University of Warsaw, 02-093 Warsaw, Poland

Heterostructures (HSs) formed by the transition-metal dichalcogenides (TMDCs) materials have shown great promise in next-generation (opto)electronic applications. Traditionally, at atomically closed proximity the charge transfer (CT) process dominates due to its fast timescale ($<$ 50 fs). In this talk, I introduce our latest work [1] on the ET process in a twisted molybdenum diselenide (MoSe2) homobilayer without any charge-blocking interlayer, i.e., in atomically closed proximity. We fabricated an unconventional homobilayer (i.e., HS) with a large twist angle (˜57°) by combining the chemical vapor deposition (CVD) and mechanical exfoliation (Exf.) techniques to fully exploit the lattice parameters mismatch and indirect/direct (CVD/Exf.) bandgap nature. These result in weakening the CT process and allowing the ET process to take over the carrier recombination channels. We employ a series of optical and electron spectroscopy techniques, complementing by the density functional theory (DFT) calculations, to describe a massive room temperature photoluminescence enhancement from the HS area due to an efficient ET process. Our results show that the electronically decoupled MoSe2 homobilayer is coupled by the ET process, mimicking a 'true' heterobilayer nature.

Reference: [1] A. Karmakar et al., "Twisted MoSe2 Homobilayer Behaving as a Heterobilayer", Nano Lett. 2024, 24, 31, 9459-9467.

HL 59.4 Fri 10:15 H15

Tailoring Interlayer Exciton Dyanamics of TMDC Heterostructures: From Bilayers to Trilayers embeded in Broadband DBR Cavities — •Bhabani Sankar Sahoo, Shachi Machchhar, Chirag Chandrakant Palekar, and Stephan Reitzenstein — Technische Universität Berlin, Berlin, Germany

The photoluminescence yield of interlayer excitons (IX) at room temperature remains limited due to electrons and holes residing in different layers. In this work we propose and implement a novel way to enhance the IX emission in transition metal dichalcogenide (TMDC) heterostructures. We first introduce an additional WSe2 on top of a heterobilayers (HBL) of WSe2/MoSe2 to form a heterotrilayer (HTL) with different twist angles which significantly boost the PL emission upto one order magnitude, attributed to improved overlap of the electronic wavefunctions and additional radiative pathways. Further embedding this HTL within a chirped multiresonant distributed Bragg reflector (DBR) microcavity provides strong exciton confinement, enhancing the radiative recombination rate and modifying the exciton lifetime. Compared to the HBL, the HTL displays a reduced exciton lifetime, indicative of stronger light-matter coupling within the cavity, and a pronounced increase in PL intensity due to enhanced photon density of states. Our study highlights the potential of tailored stacking and cavity integration to manipulate light-matter interactions in advanced TMDC heterostructure devices for next-generation optoelectronic applications.

HL 59.5 Fri 10:30 H15

Effect of the Direct-to-Indirect Bandgap Crossover on the Reverse Energy Transfer — •Gayatri Gayatri[1], Debashish Das[2], Natalia Zawadzka[1], Takashi Taniguchi[3], Kenji Watanabe[3], Adam Babiński[1], Saroj K. Nayak[2], Maciej R. Molas[1], and Arka Karmakr[1] — [1]University of Warsaw, Warsaw, Poland — [2]Indian Institute of Technology Bhubaneswar, Odisha, India — [3]National Institute for Materials Science, Ibaraki, Japan

Heterostructures (HSs) made by the vertical stacking of van der Waals monolayers (1Ls) have shown great potential in (opto)electronic devices. In the type-II transition metal dichalcogenide HSs, long-range energy transfer (ET) happens via the dipole-dipole coupling (Förster type). To investigate this, we studied HS made by the 1L tungsten disulfide (WS2) and 1L-5Ls molybdenum disulfide (MoS2), with hexagonal boron nitride (hBN) as a charge-blocking interlayer, using differential reflection contrast, photoluminescence (PL), and photoluminescence excitation. At room temperature, PL enhancement has been observed in the neutral exciton of WS2 in the WS2-hBN-1L MoS2 and WS2-hBN-2L MoS2 regions as compared to the isolated WS2 emission. This enhancement confirms an efficient ET from MoS2 B excitonic level to WS2 A excitonic level. As the number of MoS2 layers increases, bandgap changes from direct-to-indirect, promoting more immediate carrier scattering from the K-valley. Consequently, carrier population decreases and ET becomes less effective.

HL 59.6 Fri 10:45 H15

Interlayer Exciton Traps in TMD heterobilayers — •Thomas Klokkers[1,2], Mirco Troue[1,2], Johannes Figueiredo[1,2], Andreas Knorr[3], Ursula Wurstbauer[4], and Alexander Holleitner[1,2] — [1]Walter Schottky Institute, Technical University of Munich, Am Coulombwall 4a, 85748 Garching, Germany — [2]Munich Center for Quantum Science and Technology (MCQST), Schellingstr. 4, 80799 Munich, Germany — [3]Institute for Theoretical Physics, Nonlinear Optics and Quantum Electronics, Technical University of Berlin, 10623 Berlin, Germany — [4]Institute of Physics, Münster University, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany

Long-lived interlayer excitons in MoSe2/WSe2 heterostructures constitute a promising platform to explore many-body physics in a two-dimensional solid-state system. Introducing a trapping potential fa-

cilitates exploring the many-body regime by providing a platform to control the density of the interlayer exciton ensemble. Realizations of such exciton traps range from electrostatic strip gates to strain-induced potentials. In this work, we realize enhanced emission from interlayer excitons based on an optical trap. The laterally changed potential landscape allows the investigation of ensembles at high densities without the direct influence of temperature, excess charge carriers and laser-induced coherence.

15 min. break

HL 59.7 Fri 11:15 H15

Hybridized excitons in 2D van der Waals materials — •Andreas Stier — Walter Schottky Institut und TUM School of Natural Sciences, TU München, Garching, Deutschland

I will review our recent progress on magneto optical spectroscopy of atomically thin materials in magnetic fields up to 91 T with an emphasis on the spin-valley physics of neutral and charged excitons.

In monolayer (ML) semiconductors, magneto-absorption spectroscopy revealed the diamagnetic shifts of the exciton Rydberg states, which allowed the first direct experimental measure of the reduced mass and binding energy. Surprisingly, investigating the photoluminescence, we observe the emergence of a new excitonic peak, which we discuss in the framework of the theoretically predicted linear dispersing exciton branch originating from intervalley exchange interactions.

For heterostructures (HS) of a 2D semiconductor with graphene, we find a new multi-step proximity effect due to band folding in the HS, where we show that the spin-valley physics can be used to quantify interlayer hybridization. In HS from ML MoSe2 and the layered antiferromagnetic (AFM) semiconductor CrSBr, we show the formation of new exciton states depending on the twist angle. These excitons exhibit clear signatures of proximity coupling to the magnetic state of the AFM layer, such as hysteretic response to in- and out of plane B fields. We discuss these results in the framework of Ising-type spin-orbit proximity coupling.

HL 59.8 Fri 11:30 H15

Laterally extended states of interlayer excitons in reconstructed $MoSe_2/WSe_2$ heterostructures — •Johannes Figueiredo[1,2], Marten Richter[3], Mirco Troue[1,2], Jonas Kiemle[1,2], Hendrik Lambers[4], Torsten Stiehm[4], Ursula Wurstbauer[4], Andreas Knorr[3], and Alexander Holleitner[1,2] — [1]Walter Schottky Institute, TU Munich — [2]Munich Center for Quantum Science and Technology (MCQST) — [3]Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, TU Berlin — [4]Institute of Physics, Münster University

Heterostructures made from 2D transition-metal dichalcogenides are ideal platforms for exploring excitonic phenomena, including correlated moiré excitons and degenerate interlayer exciton ensembles. While atomic reconstruction is often assumed to localize excitons, we demonstrate that excitonic states in reconstructed $MoSe_2/WSe_2$ heterostructures can extend beyond the moiré periodicity [1]. Using real-space calculations, we provide lateral potential maps and corresponding excitonic wavefunctions for interlayer excitons in strain-relaxed heterostructures [2]. Cryogenic photoluminescence experiments corroborate the computed level structure and exciton relaxation dynamics. These findings align with recent coherence measurements on degenerate interlayer excitons and suggest potential many-body phenomena in dense, cold exciton ensembles [3].

[1] J. Figueiredo et al., arXiv:2411.19616 (2024) [2] M. Richter, PRB 109, 125308 (2024) [3] M. Troue and J. Figueiredo et al., PRL 131, 036902 (2023)