

CPP 24: 2D Materials 5 (joint session HL/CPP/DS)

Time: Wednesday 9:30–12:00

Location: H36

CPP 24.1 Wed 9:30 H36

Dark exciton anti-funneling in monolayer transition metal dichalcogenides — ●ROBERTO ROSATI¹, ROBERT SCHMIDT², SAMUEL BREM¹, RAÜL PEREA-CAUSÍN³, IRIS NIEHUES⁴, JOHANNES KERN², JOHANN ADRIAN PREUSS², ROBERT SCHNEIDER², STEFFEN MICHAELIS DE VASCONCELLOS², RUDOLF BRATSCHITSCH², and ERMIN MALIC^{1,3} — ¹Philipps-Universität Marburg — ²University of Münster — ³Chalmers University of Technology — ⁴CIC nanoGUNE BRTA

Current nanoelectronics relies on transport. While charged carriers can be controlled by electric fields, atomically thin semiconductors are governed by excitons, which are neutral electron-hole pairs. Recently, strain engineering has been introduced to manipulate exciton diffusion [1] and propagation [2] in monolayer transition metal dichalcogenides. Strain-induced energy gradients give rise to exciton funneling up to a micrometer range. Combining spatiotemporal photoluminescence measurements with microscopic theory, here we track the way of excitons in time, space and energy. Surprisingly we find that in WS₂ excitons move away from high-strain regions, contrary to what we observe in MoSe₂ [2]. This anti-funneling behavior can be ascribed to dark excitons, whose strain-induced energy variations are opposite compared to bright excitons. Our findings open new possibilities to control transport in exciton-dominated materials.

[1] R. Rosati et al., 2D Mater. 8, 015030 (2021).

[2] R. Rosati, R. Schmidt et al., Nat. Commun. 12, 7221 (2021).

CPP 24.2 Wed 9:45 H36

Ultrafast nanoscopy of a Mott transition in twisted bilayer WSe₂ — ●SVENJA NERRETER¹, THOMAS SIDAY¹, FABIAN SANDNER¹, SAMUEL BREM^{2,3}, MARTIN ZIZLSPERGER¹, FELIX SCHIEGL¹, RAUL PEREA-CAUSIN³, MARKUS PLANKL¹, PHILIPP MERKL¹, FABIAN MOOSHAMMER^{1,4}, MARKUS A. HUBER¹, ERMIN MALIC^{2,3}, and RUPERT HUBER¹ — ¹Department of Physics and Regensburg Center for Ultrafast Nanoscopy, University of Regensburg, 93040 Regensburg — ²Department of Physics, Philipps-Universität Marburg, 35032 Marburg, Germany — ³Department of Physics, Chalmers University of Technology, 41296 Gothenburg, Sweden — ⁴Department of Physics, Columbia University, New York, NY 10027, USA

The density-driven transition of an exciton gas into a Fermi liquid of unbound electron-hole pairs has formed a compelling testing ground of many-body physics. Layered transition metal dichalcogenides feature advantageous conditions, yet nanoscale inhomogeneities have complicated quantitative studies of this elusive transition. Here, we use ultrafast polarization nanoscopy to trace optically bright and dark electron-hole pairs during an exciton Mott transition in a twisted homobilayer of WSe₂. At elevated densities, initially monomolecular recombination dynamics of optically dark excitons continuously evolve into the bimolecular recombination of unbound electron-hole pairs. We directly reveal how the Mott transition varies over nanometer length scales, evidencing strong spatial disorder in stacked monolayers and demonstrating the capabilities of our technique to resolve the local interplay of strong electronic correlations.

CPP 24.3 Wed 10:00 H36

Rashba excitons in the 2D Ruddlesden-Popper perovskite (BA)MAPI — ●PHILIPP MOSER¹, MARTIN SCHALK¹, ATSUSHIKO MIYATA², JOACHIM WOSNITZA², ANDREAS STIER¹, and JONATHAN FINLEY¹ — ¹Walter Schottky Institute, Garching, Germany — ²Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

Two-dimensional organic-inorganic perovskites have emerged as remarkable materials for energy conversion, optoelectronic and spintronic applications. Recently, the role of spin-orbit (SO) coupling and the resulting effects on the band-structure and dark/bright optical transitions has become a key topic of interest. The necessary structural inversion asymmetry for SO-coupling is predicted to stem from the organic cations comprising the crystals. As a result, dark excitons, red detuned from the bright exciton, have been discussed in this material system. Here, we investigate the exciton physics of the 2D Ruddlesden-Popper perovskite (BA)MAPI. By performing one-photon absorption, -PL and two-photon PLE spectroscopy, we investigate the optical transitions close to the R-point of the Brillouin zone and find distinct 2-photon transitions blue detuned from the ground state exciton that can be explained by a Rashba-split band-structure. Utiliz-

ing high-field magneto-spectroscopy up to B=60T, we determine that these absorption features are due to Wannier excitons. We determine the size and binding energy from the diamagnetic shift of the features and obtain evidence that 2D (BA)MAPI hosts strongly bound Rashba excitons.

CPP 24.4 Wed 10:15 H36

Ultrafast pseudospin quantum beats in multilayer WSe₂ and MoSe₂ — ●SIMON RAIBER¹, PAULO E. FARIA JUNIOR², DENNIS FALTER¹, SIMON FELDL¹, PETTER MARZENA¹, KENJI WATANABE³, TAKASHI TANIGUCHI⁴, JAROSLAV FABIAN², and CHRISTIAN SCHÜLLER¹ — ¹Institut für Experimentelle und Angewandte Physik, Universität Regensburg, D-93040 Regensburg, Germany — ²Institut für Theoretische Physik, Universität Regensburg, D-93040 Regensburg, Germany — ³Research Center for Functional Materials, NIMS, Tsukuba, Japan — ⁴International Center for Materials Nanoarchitectonics, NIMS, Tsukuba, Japan

We present investigations of excitonic transitions in mono- and multilayer WSe₂ and MoSe₂ materials by time-resolved Faraday ellipticity (TRFE) with in-plane magnetic fields, of up to B = 9 T. In monolayer samples, the measured TRFE time traces are almost independent of B, which confirms a close to zero in-plane exciton g factor, consistent with first-principles calculations. In stark contrast, we observe pronounced temporal oscillations in multilayer samples for B > 0. Remarkably, the extracted in-plane g factors are very close to reported out-of-plane exciton g factors of the materials, namely |g_{in}| = 3.1 +/- 0.2 and 2.5 +/- 0.2 for the 1s A excitons in WSe₂ and MoSe₂ multilayers, respectively. Our first-principles calculations nicely confirm the presence of a non-zero in-plane g for the multilayer samples. We propose that the oscillatory TRFE signal in the multilayer samples is caused by pseudospin quantum beats of excitons, which is a manifestation of spin- and pseudospin layer locking in the multilayer samples.

CPP 24.5 Wed 10:30 H36

Nonlinear Exciton Dynamics in Layered Heterostructures — ●VIPIN KRISHNA¹, XIAO CHEN², TARLAN HAMZAYEV¹, SILVANA BOTTI², and GIANCARLO SOAVI¹ — ¹Institute of Solid state Physics, Friedrich-Schiller-University, Jena — ²Institute of Theoretical Solid State Theory and Optics, Friedrich-Schiller-University, Jena

Transition-metal-dichalcogenides and related heterostructures (HS) are promising candidates for photonic and optoelectronic applications owing to strong light-matter coupling and electrically-tunable carrier dynamics. However, the presence of intense nonlinear effects such as Exciton-Exciton Annihilation (EEA) [1] limits the maximum realizable exciton-density, and is particularly efficient for interlayer-excitons (IL) due to their out-of-plane dipole nature [2]. In this work, we systematically study the onset of EEA in type-II WS₂/WSe₂ HS by steady-state and nonlinear time-resolved PL. We infer that in HS the generation rate is at least one order of magnitude larger for interlayer compared to intralayer-excitons for a given excitation fluence, as expected from the ultrafast interlayer-charge-transfer and consequent IL formation. However, we do not observe stronger EEA for interlayer compared to intralayer-excitons and observe that for HS the recombination dynamics are identical for both, suggesting that the EEA mechanism is dominated by the total excitonic-density via intra and interlayer-exciton interactions. Our work provides new insights on EEA mechanism, which is of paramount importance for optoelectronic-devices and study of excitonic-condensates with layered-materials. [1] Kuechle et. Al. J.OMX (2021), 12. [2] Sigl et. Al. Phys. Rev. B 105, 035417.

15 min. break

CPP 24.6 Wed 11:00 H36

Tunable exciton-polaritons emerging from WS₂ monolayer excitons in a photonic lattice at room temperature — ●LUKAS LACKNER¹, MARCO DUSEL², OLEG EGOROV³, BO HAN¹, HEIKO KNOPF³, FALK EILENBERGER³, CARLOS ANTON-SOLANAS¹, SVEN HÖFLING², and CHRISTIAN SCHNEIDER¹ — ¹University of Oldenburg, Oldenburg, Germany — ²University of Würzburg, Würzburg, Germany — ³Friedrich Schiller University Jena, Jena, Germany

The engineering of non-linear light-matter states in optical lattices has emerged as a key research strategy for the exploration of Hamilto-

nians in the spirit of ultrafast- and possibly quantum-simulation. It furthermore has revealed its potential to probe non-trivial topology phenomena. Excitons in atomically thin crystals have emerged as an ideal active medium for such purposes, since they couple strongly with light, and bear the potential to harness giant non-linearities and interactions.

In this work, we present an experiment conducted at room temperature in an open optical cavity of high quality, with an implemented one-dimensional photonic lattice. In our present work we integrate an atomically thin layer of WS_2 in such a device. We discuss the emergence and tunability of a lattice-band-structure in the tight-binding configuration at room temperature, fuelled by the emission from monolayer excitons[1].

References

- [1] L. Lackner *et al.*, *Nat Commun* **12**, 4933 (2021).

CPP 24.7 Wed 11:15 H36

Optical Spectroscopy of Colloidal Transition Metal Dichalcogenides — ●ANDRÉ PHILIPP FRAUENDORF¹, ANDRÉ NIEBUR², JENS HÜBNER¹, JANNIKA LAUTH^{2,3}, and MICHAEL OESTREICH¹ — ¹Institut für Festkörperphysik, Leibniz Universität Hannover — ²Institut für Physikalische Chemie und Elektrochemie, Leibniz Universität Hannover — ³Institut für Physikalische und Theoretische Chemie, Universität Tübingen

Atomically thin transition metal dichalcogenides (TMDs) are at the forefront of a new generation of two-dimensional semiconductor systems and experience an increasing research interest due to their unique optical properties. As an additional fabrication approach the wet-chemical synthesis has emerged as a promising method for the straightforward solution-processing of these materials.[1] Nevertheless, the optical properties of colloidal TMD mono- and few-layer structures have been sparsely studied.

Here, we demonstrate room-temperature micro-photoluminescence of colloidal TMD nanosheets. Both, mono- and multilayer photoluminescence are observed rendering these delicate structures fully competitive with conventionally fabricated TMDs.[1] In addition temperature-dependent transient absorption measurements are presented as a convincing technique for the exploration of the ultra-fast recombination dynamics of two-dimensional materials.[2]

[1] A. Frauendorf *et al.*, *J. Phys. Chem. C* **125**, 18841 (2021).

[2] A. Frauendorf *et al.*, Manuscript in preparation (2022).

CPP 24.8 Wed 11:30 H36

Capacitively and inductively coupled excitons in bilayer

MoS₂ — ●LUKAS SPONFELDNER¹, NADINE LEISGANG¹, SHIVANGI SHREE², IOANNIS PARADISANOS², KENJI WATANABE³, TAKASHI TANIGUCHI⁴, CEDRIC ROBERT², DELPHINE LAGARDE², ANDREA BALOCCHI², XAVIER MARIE², IANN C. GERBER², BERNHARD URBASZEK², and RICHARD J. WARBURTON¹ — ¹Department of Physics, University of Basel — ²Université de Toulouse, INSA-CNRS-UPS, LPCNO — ³Research Center for Functional Materials, National Institute for Materials Science — ⁴International Center for Materials Nanoarchitectonics, National Institute for Materials Science

Exciton-exciton couplings in semiconductors lead to a plethora of phenomena such as nonlinear optical effects and quantum condensation. Transition-metal dichalcogenides constitute a versatile platform to study these effects as the excitons are very robust and their couplings can be controlled by exploiting their spin and valley properties.

Here, we probe exciton-exciton couplings in gated-homobilayer MoS_2 . Using a driven-coupled oscillator model it is shown that the measured optical susceptibility reveals both the magnitude and the phase of the coupling constants. The interlayer excitons (IE) and intralayer B-excitons couple via a 0-phase (capacitive) coupling; the IE and the intralayer A-excitons couple via a π -phase (inductive) coupling. Using the IE as a sensor, the A-B intravalley exchange coupling is determined, a result which is also relevant for a monolayer. Finally, we realize a bright and highly tunable lowest-energy momentum-direct exciton at high electric fields.

CPP 24.9 Wed 11:45 H36

Controlling the non-linearity in two dimensional materials —

●MATHIAS FEDEROLF and SVEN HÖFLING — Technische Physik, Universität Würzburg, 97074 Würzburg, Germany

Recently Datta *et al.* [1] have shown that exciton-polaritons in bilayer MoS_2 experience a blueshift due to interacting with other exciton-polaritons. The observed blueshift is non-linear with respect to the laser power used for excitation. Due to the bilayer's nature interlayer-excitons can occur, which exhibit an out of plane dipole moment. Using an electric field along the out of plane axis those dipoles can be aligned and used to influence the exciton-exciton interaction. By using a varying electric field, we map the parameter space to gain deterministic control over the blueshift. Understanding and controlling the system allows us to tune the polariton-polariton interaction such that they can be used in future application *i.e.*, single-photon sources.

[1] Datta, Biswajit, *et al.* arXiv preprint arXiv:2110.13326 (2021).