DS 3: 2D semiconductors and van der Waals heterostructures I (joint session HL/DS)

Time: Monday 13:30–16:15 Location: H4

Invited Talk DS 3.1 Mon 13:30 H4

The role of chalcogen vacancies for atomic defect emission in MoS_2 — Elmar Mitterreiter[1], Bruno Schuler[2], Daniel Hernangómez-Pérez[3], Julian Klein[4], Jonathan Finley[1], Sivan Refaely-Abramson[3], Alexander Holleitner[1], Alexander Weber-Bargioni[5], and •Christoph Kastl[1] — [1]Walter Schottky Institute, TU Munich — [2]nanotech@surfaces Laboratory, Empa — [3]Department of Molecular Chemistry and Materials Science, Weizmann Institute of Science — [4]Massachusetts Institute of Technology — [5]Molecular Foundry, Lawrence Berkeley National Laboratory

The microscopic understanding of defect-related modifications in 2D materials requires correlation between atomic structure and resulting macroscopic electronic, optical or excitonic properties. Combing controlled defect engineering with optical spectroscopy as well as atomic imaging and ab-initio theory, we identify the optical signature of pristine chalcogen vacancies in single layer MoS_2. [1] Vacancies introduce a narrow optical emission, markedly different from previously observed broad luminescence bands. Comparing annealed vs. He-ion treated MoS_2, we establish that the recently discovered single-photon emitters in He-ion irradiated MoS_2 originate from chalcogen vacancies. Using focused ion beam irradiation, the latter can be created site-selectively [2] with a spatial precision better than 10 nm [3], which is important for a prospective integration of defect-based single photon emitters into quantum photonic circuits. [1] E. Mitterreiter et al., Nat. Commun. 12, 3822 (2021). [2] J. Klein et al. ACS Photonics 8, 669-677 (2021). [3] E. Mitterreiter et al., Nano Lett. 20, 4437 (2020).

DS 3.2 Mon 14:00 H4

Coherent light emission of exciton-polaritons in an atomically thin crystal at room temperature — •Hangyong Shan[1], Lukas Lackner[1], Bo Han[1], Evgeny Sedov[2], Falk Eilenberger[4], Sebastian Klembt[3], Sven Höfling[3], Alexey V. Kavokin[2], Christian Schneider[1], and Carlos Anton-Solanas[1] — [1]Institute of Physics, Carl von Ossietzky University, 26129 Oldenburg, Germany. — [2]School of Science, Westlake University, 310024 Hangzhou, People's Republic of China. — [3]Technische Physik, Universität Würzburg, D-97074 Würzburg, Am Hubland, Germany. — [4]Institute of Applied Physics, Abbe Center of Photonics, Friedrich Schiller University, 07745 Jena, Germany.

We experimentally study the coherence of exciton-polaritons in a Fabry-Perot microcavity loaded with an atomically thin WSe2 layer. Via Michelson interferometry, we capture clear evidence of increased spatial and temporal coherence of the emitted light from the spatially confined system ground-state. The coherence build-up is accompanied by a threshold-like behaviour of the emitted light intensity, which is a fingerprint of a polariton condensation effect. Valley-physics is manifested in the presence of an external magnetic field, which allows us to manipulate K and K* polaritons via the Valley-Zeeman-effect. Our findings are of high application relevance, as they confirm the possibility to use atomically thin crystals as simple and versatile components of coherent light-sources, and in valleytronic applications at room temperature.

DS 3.3 Mon 14:15 H4

Bosonic condensation of exciton-polaritons in an atomically thin crystal — •Carlos Anton-Solanas[1,2], Maximilian Waldherr[1], Martin Klaas[1], Holger Suchomel[1], Tristan H. Harder[1], Hui Cai[3], Evgeny Sedov[4], Sebastian Klembt[1], Alexey V. Kavokin[4], Sefaattin Tongay[5], Kenji Watanabe[6], Takashi Taniguchi[6], Sven Hoefling[1], and Christian Schneider[2] — [1]Univ. Wuerzburg, Germany — [2]Univ. Oldenburg, Germany — [3]Univ. California, USA — [4]Westlake Univ., China — [5]Arizona State Univ., USA — [6]Nat. Institute for Materials Science, Japan

Semiconducting monolayer crystals have emerged as a new platform for studies of tightly bound excitons and many-body excitations in ultimately thin materials. Their giant dipole coupling to optical fields makes them very appealing for (nano-) photonic devices, and for fundamental investigations in the framework of cavity quantum electrodynamics.

Our experiments demonstrate the strong light-matter coupling and, for the first time, the bosonic condensation of exciton-polaritons in an atomically thin layer of $MoSe_2$ coupled to a hybrid micro-cavity [1].

We demonstrate the emergence of long-range first-order spatial coherence, via interferometric $g^{(1)}(\tau)$ measures, and we have investigated the Zeeman splitting effects of condensed polaritons under strong magnetic fields.

[1] Anton-Solanas, C., Waldherr, M., Klaas, M. et al. Bosonic condensation of exciton polaritons in an atomically thin crystal. Nat. Mater. (2021).

DS 3.4 Mon 14:30 H4

Hybridization between monolayer transition-metal dichalcogenides and conjugated molecular adsorbants — •Jannis Krumland[1] and Caterina Cocchi[1,2] — [1]Humboldt-Universität zu Berlin — [2]Carl von Ossietzky Universität Oldenburg

We present a first-principles study on electronic hybridization in inorganic-oraganic interfaces composed of monolayer transition-metal dichalcogenides (TMDCs; molybdenum and tungsten disulfide and diselenide) and exemplary carbon-conjugated molecules such as pyrene and perylene. By means of band-structure unfolding techniques applied to hybrid density-functional theory calculations including spin-orbit coupling, we achieve an intuitive and clear description of electronic interaction between the inorganic and organic components of the heterostructures. From atom-projected band structures, we are able to rationalize the strong mixing between the valence states of the TMDC and the molecular orbitals. We additionally clarify why the highest occupied orbital couples with the TMDC bands only very weakly, regardless of the composition of the interface. The proposed analysis based on band structure unfolding lends itself for computationally efficient and yet reliable predictions of electronic interactions in more complex hybrid interfaces including larger molecules harvesting visible radiation.

DS 3.5 Mon 14:45 H4

Tunable Polymer/Air Bragg Optical Microcavity Configurations for Light-Matter Coupling with Transition-Metal Dichalcogenides and their Heterostructures — •Chirag Palekar[1,2], Stephan Reitzenstein[1], and Arash Rahimi-Iman[2] — [1]Present address: Institute of Solid State Physics, Technische Universität Berlin, D-10623 Berlin, Germany — [2]Faculty of Physics and Materials Sciences Center, Philipps-Universität Marburg, 35032 Marburg, Germany

Light-matter interactions (LMI) in semiconducting materials is being studied extensively with the help of optical microcavities. Specifically, tunable microcavities provide a versatile platform to control the LMI between the material excitation and cavity photons. Here, we explore a new resonator approach which can be employed to achieve microscopic photonic Fabry-Pérot (FP) cavities with mechanically- tunable resonator modes and polymer/air Bragg mirrors [1], directly on a chip or device substrate in combination with active materials. Moreover, our simulations based on the transfer matrix method show, compression-induced mode control of the air-Bragg cavities enables tuning between the weak and strong coupling regime. Using this unique cavity configurations, LMI experiments with 2D semiconductors such as transitionmetal dichalcogenides (TMDC) are very attractive. Additionally, incorporation of TMDC heterostructures in FP cavities will provide a platform to understand the new regimes of Dicke superradiance as well as Bose-Einstein condensation of Moiré exciton-polaritons. Ref.: [1] Phys. Status Solidi RRL 2021, 15, 2100182

15 min. break

DS 3.6 Mon 15:15 H4

Phonon-assisted exciton dissociation in transition metal dichalcogenides — •Raul Perea-Causin[1], Samuel Brem[1], and Ermin Malic[1,2] — [1]Chalmers University of Technology, Gothenburg, Sweden — [2]Philipps-Universität, Marburg, Germany

Monolayers of transition metal dichalcogenides (TMDs) have been established in the last years as promising materials for novel optoelectronic devices. However, the performance of such devices is often limited by the dissociation of tightly bound excitons into free electrons and holes. While previous studies have investigated tunneling at large electric fields, we focus in this work on phonon-assisted exciton dissociation that is expected to be the dominant mechanism at small fields.

We present a microscopic model based on the density matrix formalism providing access to time- and momentum-resolved exciton dynamics including phonon-assisted dissociation [1]. We track the pathway of excitons from optical excitation via thermalization to dissociation, identifying the main transitions and dissociation channels. Furthermore, we find intrinsic limits for the quantum efficiency and response time of a TMD-based photodetector and investigate their tunability with externally accessible knobs, such as excitation energy, substrate screening, temperature and strain.

Our work provides microscopic insights in fundamental mechanisms behind exciton dissociation and can serve as a guide for the optimization of TMD-based optoelectronic devices.

[1] R. Perea-Causin et al., Nanoscale 13, 1884 (2021)

DS 3.7 Mon 15:30 H4

Lattice Configurations of Self-Assembled Folded Graphene — •Lina Bockhorn, Johannes C. Rode, Lucas Gnörich, Pengfei Zuo, and Rolf J. Haug — Institut für Festkörperphysik, Leibniz Universität Hannover, 30167 Hannover, Germany

The stacking- and folding angle of 2D materials to 3D structures has emerged as an important, novel tuning parameter for the tailoring of optical, mechanical, electronic and magnetic properties. Therefore, it is highly desirable to gather insight into the mechanical formation of these structures on the nano-scale.

Here, we focus on the evolution of self-assembled folded graphene generated via atomic force microscopy technique, which could give a deep insight into its underlying growth energy [1, 2, 3]. The self-assembly process involves the folding-over of the graphene layer and the subsequent growth of a twisted graphene bilayer. We conclude, that these self-assembled structures move not only forward during the growth process but also appear to rotate and lock in at specific commensurate twist angles.

[1] J. C. Rode et al., Ann. Phys. 529, 1700025 (2017).
[2] J. C. Rode et al., 2D Mater. 6, 015021 (2018).
[3] L. Bockhorn et al., Appl. Phys. Lett. 118, 173101 (2021).

DS 3.8 Mon 15:45 H4

All-optical polarization and amplitude modulation of second harmonic generation in atomically thin semiconductors — •Sebastian Klimmer — Institute of Solid State Physics, Friedrich Schiller University Jena, Jena, Germany

Nonlinear optics is of paramount importance in several fields of science and technology. This is particularly true in the case of second harmonic generation (SHG), which is commonly used for frequency conversion, self-referencing of frequency combs, crystal characterization, sensing, and ultra-short pulse characterization. Large efforts have been devoted in the last years to realizing electrical and all-optical modulation of SHG in atomically thin materials, which are easy to integrate on photonic platforms and thus ideal for novel nano-photonic devices. Here, we propose a new approach to broadband all-optical modulation of SHG in 2D materials. Our concept is based only on symmetry considerations and thus is applicable to any material of the D3h symmetry group and with deep sub-wavelength thickness, such as all monolayer transition metal dichalcogenides. With this approach we demonstrate a 90° rotation of the polarization of the emitted SH on a time-scale limited only by the fundamental pulse duration. In addition, this ultrafast polarization switch can be immediately applied to realize all-optical SH amplitude modulation with depth of 100 %. Our results outperform any previous work on all-optical SHG modulation [1,2] in terms of modulation speed, modulation depth and SHG bandwidth.

[1] Taghinejad M. *et al.*, Small **16**, 1906347 (2020)
[2] Cheng Y. *et al.*, Nano Lett. **20**, 11 (2020) 8053-8058

DS 3.9 Mon 16:00 H4

Microscopic Theory of Exciton-Exciton Annihilation in Two-Dimensional Semiconductors — •Alexander Steinhoff, Matthias Florian, and Frank Jahnke — Institute for Theoretical Physics, University of Bremen, Bremen, Germany

Auger-like exciton-exciton annihilation (EEA) is considered the key fundamental limitation to quantum yield in devices based on excitons in two-dimensional (2d) materials. Since it is challenging to experimentally disentangle EEA from competing processes, guidance of a quantitative theory is highly desirable. The very nature of EEA requires a material-realistic description that is not available to date.

We present a many-body theory of EEA based on first-principle band structures and Coulomb interaction matrix elements that goes beyond an effective bosonic picture. Applying our theory to monolayer MoS_2 encapsulated in hexagonal BN, we obtain an EEA coefficient in the order of 10^{-3} cm^2s^{-1} at room temperature, suggesting that carrier losses are often dominated by other processes, such as defect-assisted scattering.

Our studies open a perspective to quantify the efficiency of intrinsic EEA processes in various 2d materials in the focus of modern materials research.