DS 16: 2D semiconductors and van der Waals heterostructures III (joint session HL/DS)

Time: Tuesday 9:30–13:00 Location: POT 81

Invited Talk DS 16.1 Tue 9:30 POT 81

Radiative Lifetime and Fine Structure of Excitons in Transition Metal Dichalcogenide Monolayers — •Xavier Marie — Université de Toulouse, LPCNO, INSA-CNRS-UPS, Toulouse, France

Optical properties of atomically thin transition metal dichalcogenides are controlled by robust excitons characterized by a very large oscillator strength [1,2,3]. Encapsulation of monolayers such as MoSe2 in hexagonal boron nitride (hBN) yields narrow optical transitions approaching the homogeneous exciton linewidth [4,5]. We demonstrate that the exciton radiative rate in these van der Waals heterostructures can be tailored by a simple change of the hBN encapsulation layer thickness as a consequence of the Purcell effect [6].

The time-resolved photoluminescence measurements together with cw reflectivity and photoluminescence experiments show that the neutral exciton spontaneous emission time can be tuned by one order of magnitude depending on the thickness of the surrounding hBN layers.

I will also discuss recent results on the fine structure of excitons in MoSe2 and MoS2 monolayers.

[1] G. Wang et al, Rev. Mod. Phys. 90, 021001 (2018) [2] D. Lagarde et al, PRL 112, 047401 (2014) [3] C. Robert et al, PRB 93, 205423 (2016) [4] F. Cadiz et al, PRX 7, 021026 (2017) [5] G. Wang et al, PRL 119, 047401 (2017) [6] H.H. Fang et al, PRL 123, 067401 (2019)

DS 16.2 Tue 10:00 POT 81

Control of Excitons in Monolayer Transitionmetal Dichalcogenides via Extrinsic Dielectric Screening — •Frederik Otto, Philip Klement, and Sangam Chatterjee — Institute of Experimental Physics I, Justus-Liebig-University Giessen, Heinrich-Buff-Ring 16, D-35392 Giessen, Germany

Electric field lines between charges in freestanding monolayers of transition metal dichalcogenides (TMDs) can extend freely outside the material's perimeters due to the absence of screening carriers on top and on the bottom of the sample. Similarly, field lines extending into a close dielectric, *e.g.*, the substrate, are subject to extrinsic screening effects. This effectively weakens the Coulomb interaction of charge carriers and therefore, iduces changes to the exciton binding energy and electrical band gap.

In order to obtain knowledge about the interaction range and the effect of the static dielectric constant of the surrounding dielectric environment on A and B-excitons of the K-point, we investigated monolayer samples WSe_2 supported on either TiO_2 (high static dielectric constant) or SiO_2 (low static dielectric constant) and samples encapsulated between the two substrate materials.

DS 16.3 Tue 10:15 POT 81

Revealing the quantum nature of excitons in encapsulated monolayers by optical dispersion measurements — •Lorenz Maximilian Schneider[1], Shanece S. Esdaille[2], Daniel A. Rhodes[2], Katayun Barmak[3], James C. Hone[2], and Arash Rahimi-Iman[1] — [1]Faculty of Physics and Materials Sciences Center, Philipps-Universität Marburg, Marburg, 35032, Germany — [2]Department of Mechanical Engineering, Columbia University, New York, NY 10027, USA — [3]Department of Applied Physics and Applied Mathematics, Columbia University, New York, NY 10027

The dispersion of excitons in TMDC monolayers has been a topic of several recent theoretical studies and is the base of a true understanding of the physics of such systems. Nonetheless, the theoretical papers have contradicting predictions ranging from the formation of Dirac cones or the formation of a second linear exciton branch around zero in-plane momentum to calculations that just expect ordinary degeneracy-lifted parabolic dispersions. Here, we employ Fourier-space spectroscopy to directly measure the dispersion of the A-exciton in a high-quality h-BN encapsulated monolayer system. A remarkably strong dispersion with 2 meV shifts in within the light cone, respectively an effective mass of 7E-4 m_e can be deduced. Models based on exchange interaction and exciton–polariton formation due to the large oscillator strength are discussed in order to understand the phenomenon clearly observed in PL and reflection spectra [1]. Futhermore, density and temperature dependent behaviour is presented.

[1] L.M. Schneider et al., Optics Express (in press)

DS 16.4 Tue 10:30 POT 81

Light-driven capacitive charge injection in 0D-2D hybrid nanostructures — •Ilka Kriegel[1], Michele Ghini[1], Liberato Manna[1], Nicholas J. Borys[2], and P. James Schuck[3] — [1]Department of Nanochemistry, Istituto Italiano di Tecnologia, Italy — [2]Department of Physics, Montana State University, Bozeman, MT, USA — [3]Department of Mechanical Engineering, Columbia University, New York, NY, USA

In this work we reveal that the coupling of indium tin oxide (ITO) nanocrystals to monolayer MoS2 results in a light-driven charge-injection scheme that quasi-permanently dopes monolayer MoS2 to extents competing with electrostatic doping. The cooperative electronic properties of such novel 0D 2D hybrids display efficient and permanent charge separation after light absorption. The electrons are stored in the nanocrystals, while the holes are transferred to the 2D material accumulating in regions of initially enhanced n-type doping and preferentially along edges and grain boundaries. Charge separation over distances up to 40 μm away from the local (micron sized) optical excitation spot are observed. Notably, carrier injection follows a capacitor-like behavior with capacitance values in the femto Farad range leading to the photo-charging of a model capacitor. An average optically induced photodoping of each nanocrystal with more than 40 carriers is extracted. These studies present a foundational building block for next-generation light-driven energy storage devices. Additionally, the remote-control of local charge density opens prospects for contactless and optically driven 2D material electronics.

30 min. break

DS 16.5 Tue 11:15 POT 81

Broken adiabaticity induced by Lifshitz transition in MoS_2 and WS_2 single layers — •Dino Novko — Institute of Physics, Zagreb, Croatia — Donostia International Physics Center (DIPC), San Sebastián, Spain

The breakdown of the adiabatic Born-Oppenheimer approximation is striking dynamical phenomenon, however, it occurs only in a handful of layered materials. Here I show that adiabaticity breaks down in doped single-layer transition metal dichalcogenides in a quite intriguing manner. Namely, significant nonadiabatic coupling, which acts on frequencies of the Raman-active modes, is prompted by a Lifshitz transition due to depopulation and population of multiple valence and conduction valleys, respectively. The outset of the latter event is shown to be dictated by the interplay of highly non-local electron-electron interaction and spin-orbit coupling. In addition, intense electron-hole pair scatterings due to electron-phonon coupling are inducing phonon linewidth modifications as a function of doping. Comprehending these intricate dynamical effects turns out to be a key for mastering characterization of electron doping in two-dimensional nano-devices by means of Raman spectroscopy.

DS 16.6 Tue 11:30 POT 81

Transient Valley Grating Spectroscopy on WSe_2 — •Julian Wagner, Henning Kuhn, Robin Bernhardt, Jingyi Zhu, and Paul van Loosdrecht — Universität zu Köln, II. Physikalisches Institut, D-50937 Köln, Germany

The absence of space inversion symmetry combined with strong spin-orbit interactions and time-reversal in monolayer transition metal dichalcogenides lead to the emergence of a new quantum degree of freedom, the valley pseudospin. This valley degree of freedom can be manipulated making use of the selection rules for light matter interaction, i.e. one can for instance create pure pseudospin up or down states. These states, however, will decay due to intervalley scattering of the excitonic states involved. The mechanisms leading to intervalley scattering are currently intensely debated, partially due to the lack of experiments directly addressing this.

A direct technique to address the intervalley scattering is Transient Valley Grating Spectroscopy, which is a novel 4-wave mixing approach similar to spin grating techniques. Using this approach, we investigated the valley pseudospin dynamics in monolayer WSe_2. This allows for a direct determination of the intervalley scattering rate. Its temperature dependence shows that the dominant valley depolarization process is optical-phonon mediated intervalley scattering.

DS 16.7 Tue 11:45 POT 81

Exciton diffusion in WS_2 monolayers with suppressed disorder — •Koloman Wagner[1], Jonas Zipfel[1], Marvin Kulig[1], Raül Perea-Causín[2], Samuel Brem[2], Jonas D. Ziegler[1], Roberto Rosati[2], Takashi Taniguchi[3], Kenji Watanabe[3], Mikhail M. Glazov[4], Ermin Malic[2], and Alexey Chernikov[1] — [1]Department of Physics, University of Regensburg, Regensburg, Germany — [2]Department of Physics, Chalmers University of Technology, Gothenburg, Sweden — [3]National Institute for Materials Science, Tsukuba, Ibaraki, Japan — [4]Ioffe Institute, St. Petersburg, Russia

Excitons are known to dominate optical properties of semiconducting transition metal dichalcogenides (TMDCs) both for monolayers and heterostructures. While excitons can also propagate across large distances due to the two-dimensional nature of the system, their behavior remains highly sensitive to local environmental inhomogeneities. In our work, we take advantage of material encapsulation in high-quality hexagonal boron nitride to study inherent exciton propagation unobscured by disorder. Using spatially- and time-resolved photoluminescence microscopy we find highly efficient linear diffusion and pronounced non-linear phenomena. In order to explain our findings we employ a combination of numerical and analytical approaches, discuss the role of dark states as well as non-radiative exciton-exciton scattering and present a mechanism for rapid diffusion facilitated by free electron-hole plasma.

DS 16.8 Tue 12:00 POT 81

Chemical Trend of Transition-Metal Doping in WSe_2 — •Dan Han[1,2,3], Shiyou Chen[1], and Mao-Hua Du[2] — [1]East China Normal University, Shanghai, China — [2]Oak Ridge National Laboratory, Oak Ridge, USA — [3]Ludwig-Maximilians-Universität München, Munich, Germany

Transition-metal dichalcogenides (TMDs) are promising nanoscale materials with a wide range of applications. Chemical doping is a powerful tool for tailoring the physical and chemical properties of TMDs for targeted functionalities. As an important TMD, WSe_2 has great potential for applications in FET and CMOS technologies. However, precise control over the type and density of free carriers remains challenging. We performed first-principles calculations to study intrinsic defects and transition-metal (TM) dopants in WSe_2. Our results show that TM doping can effectively control the Fermi level in WSe_2 with no significant compensation by intrinsic defects. Nb and Ta are effective p-type dopants capable of generating a high free hole density in WSe_2. While n-type doping is possible by Re and Cu, the doping efficiency is reduced due to the lower attainable dopant concentration and higher ionization energies. The chemical trend in the attainable concentration of various substitutional TM dopants in WSe_2 is largely determined by the competition between the dopant incorporation in WSe_2 and the formation of the secondary phase $TMSe_2$. Such a competition is strongly affected by the different crystal environments of the TM ion in $TMSe_2$ and WSe_2.

DS 16.9 Tue 12:15 POT 81

Interlayer excitons in pristine bilayer MoS2 with strong oscillator strength — •Etienne Lorchat[1], Maximilian Waldherr[1], Sefaattin Tongay[2], Takashi Taniguchi[3], Kenji Watanabe[3], Christian Schneider[1], and Sven Höfling[1] — [1]Technische Physik and Wilhelm Conrad RöntgenResearch Center for Complex Material Systems, Physikalisches Institut, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany — [2]School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, Arizona 85287, United States — [3]National Institute for Materials Science, Tsukuba, Ibaraki 305-0044, Japan

In the high density limit, the interaction between electrons in a 2D electron gas (2DEG) and excitons in Transition Metal Dichalcogenide (TMD) are predicted to yield a new, unconventional superconductive phase via excitonic cooper pairing. However, this interaction only gets notable if exciton aquires an out of plane dipole to interact with the 2DEG as well as an in-plane dipole to be easily excited with light. The commonly studied excitons in TMD monolayers or heterobilayers present solely one type of the dipole. Among the various approach, we will present here the hybrid interlayer exciton in MoS2. Using field-dependent photoluminescence spectroscopy as well as reflectivity we will demonstrate the emergence of an excitonic resonance, which combines a static dipole moment with a significant oscillator strength.

DS 16.10 Tue 12:30 POT 81

Optoelectronic transport in van der Waals heterostructures of Weyl semimetal $MoTe_2$ — •Maanwinder Partap Singh[1,2], Jonas Kiemle[1,2], Alexander Holleitner[1,2], and Christoph Kastl[1,2] — [1]Walter Schottky Institut, Technical University of Munich, Am Coulombwall 4, 85748 Garching, Germany — [2]Munich Center for Quantum Science and Technology (MCQST), Schellingstraße 4, D-80799 Munich, Germany

Unlike topological insulators which are topologically protected by the bulk band gap, Weyl semimetal's topological protection comes from the fact that their 3D Weyl nodes are separated in momentum space. $MoTe_2$ belongs to the family of transition metal dichalcogenides, and it crystallizes in several structures. At room temperature, it exists as either hexagonal (2H, a semiconducting phase) or monoclinic (1T', a metallic phase) structure. Upon cooling, the monoclinic phase undergoes a transition at 240 K into an orthorhombic phase known as T_d phase, which breaks inversion symmetry and results in a type II Weyl semimetal phase. Here, we study the optoelectronic properties of $MoTe_2$ as function of temperature and layer number using photocurrent and photoconductance spectroscopy. In particular, we investigate the ultrafast electron dynamics using an on-chip Terahertz spectroscopy to disentangle hot electron currents and photogalvanic effects.

DS 16.11 Tue 12:45 POT 81

Surface Ripplocations in van der Waals materials: Structural & Electronic Properties — •James McHugh[1], Pavlos Mouratidis[1], Kenny Jolley[1], and Patrick Briddon[2] — [1]Dept. of Chemistry, Loughborough University — [2]School of Engineering, Newcastle University

Ripplocations are a new class of defect, unique to layered solids, which are characterised by the accommodation of extra material at sharp, localised folds. Dislocations in three-dimensional materials arise from the balance of strain and disregistry. In contrast, anisotropic van der Waals materials may completely alleviate strain through out-of-plane buckling.

We have conducted first-principles and analytical investigation of the properties of surface ripplocations on van der Waals layers. Analytical expressions for the formation energy, height and width of ripplocations are derived and compared to ab-initio simulations, showing that surface ripplocations readily form epitaxially on van der Waals materials.

The accommodation of extra material across a ripple is considered in terms of a Frenkel-Kontorova model, where it is found that ripplocations exhibit a "double kink" structure owing to the interplay of curvature and disregistry across the defect. Additionally, it is found that ripplocations induce a reduction in the band gap of layered semiconductors, with PDOS simulations identifying this change with the transition metal atoms across the curved regions. In combination with their high mobility this suggests the possibility of using ripplocations as a defect engineering platform.