CPP 21: Two-dimensional Materials I (joint session HL/CPP)

Time: Tuesday 9:30–13:00 Location: H36

CPP 21.1 Tue 9:30 H36

Electronic Dipole Spin Resonance of 2D Semiconductor Spin Qubits — •Matthew Brooks and Guido Burkard — Universität Konstanz, Konstanz, Deutschland

Monolayer transition metal dichalcogenides (TMDs) offer a novel two-dimensional platform for semiconductor devices. One such application, whereby the added low dimensional crystal physics (i.e. optical spin selection rules) may prove TMDs a competitive candidate, is quantum dots as qubits. The band structure of TMD monolayers offers a number of different degrees of freedom and combinations thereof as potential qubit basis, primarily electron spin, valley isospin and the combination of the two due to the strong spin orbit coulping known as a Kramers qubit. Pure spin qubits in monolayer MoX_2 (where X = S or Se) have been shown to be achievable by energetically isolating a single valley and tuning to a spin degenerate regime within that valley by a combination of a sufficiently small quantum dot radius and large perpendicular magnetic field. Within such a TMD spin qubit, we theoretically induce and analyse single qubit rotations with an electric dipole spin resonance. We employ a rotating wave approximation within a time dependant Schrieffer-Wolf approximation to derive analytic expressions for the Rabi frequency of single qubit oscilations, and compare this result to more exact numerics, as to find optimal operational regimes.

CPP 21.2 Tue 9:45 H36

Metalorganic Vapour Phase Epitaxy (MOVPE) Technology for 2D Transition Metal Dichalcogenides (TMDC) — •Michael Heuken[1,2], Annika Grundmann[2], Dominik Andrzejewski[3], Tilmar Kümmell[3], Gerd Bacher[3], Holger Kalisch[2], and Andrei Vescan[2] — [1]AIXTRON SE, Dornkaulstr. 2, 52134 Herzogenrath, Germany — [2]Compound Semiconductor Technology, RWTH Aachen University, Sommerfeldstr. 18, 52074 Aachen, Germany — [3]Werkstoffe der Elektrotechnik and CENIDE, University Duisburg-Essen, Bismarckstr. 81, 47057 Duisburg, Germany

The 2D TMDC MoS_2 and WS_2 have raised strong interest due to their exceptional properties and prospects for micro- and optoelectronics. For fundamental material physics and the development of (opto)electronic devices, a reproducible deposition technology providing uniform layers of controlled thickness and purity is indispensable. MOVPE is perfectly suited for this task and can furthermore be scaled up to production with high volume and yield. Here, we report on the systematic investigation of 2D MoS_2 and WS_2 growth on sapphire using a hydride-free MOVPE process in a commercial AIXTRON multi-wafer MOVPE reactor. Metal hexacarbonyls (WCO and MCO) as well as DTBS are used as high-purity MO sources. The impact of the fundamental growth parameters is discussed, shedding light on nucleation and lateral 2D growth until full layer coalescence. Samples are characterized using Raman spectroscopy, SEM, AFM, PL, and reflectometry. Finally, a WS_2-based LED will be presented to demonstrate the applicability of 2D TMDC for optoelectronic devices.

CPP 21.3 Tue 10:00 H36

Interplay of bright and dark excitons in transition metal dichalcogenides — •Malte Selig[1], Dominik Christiansen[1], Florian Katsch[1], Ermin Malic[2], and Andreas Knorr[1] — [1]Nichtlineare Optik und Quantenelektronik, Institut für Theoretische Physik, Technische Universität Berlin, Deutschland — [2]Chalmers University of Technology, Department of Physics, Göteborg, Sweden

The extraordinarily strong Coulomb interaction in monolayers of transition metal dichalcogenides (TMDs) gives rise to the formation of tightly bound electron hole pairs, excitons, which dominate the optical and electronic properties of these ultrathin materials. In particular, a variety of dark exciton states occurs, including excitons with opposite spins of the constituent carriers [1] and excitons with momenta well above the radiative cone [2]. In this talk, we will address the impact of such dark exciton states to the optical properties of TMDs, including optical lineshape [2,3], luminescence properties [4] and intervalley relaxation. Additionally, we compare the influence of these states on the excitonic linewidth between mono- and bilayer samples [5].

[1] X.-X. Zhang et al., Nature Nanotechnology 12, 883 (2017)
[2] M. Selig et al., Nature Communications 13279 (2016)
[3] D. Christiansen et al., Physical Review Letters 119, 187402 (2017)
[4] M. Selig et al., 2D Materials 5 035017 (2018)
[5] A. Raja et al., Nano Letters, 18 (10), 6135 (2018)

CPP 21.4 Tue 10:15 H36

Many-body quantum Monte Carlo study of 2D materials: cohesion and band gap in single-layer phosphorene — •Tobias Frank[1], Rene Derian[2], Kamil Tokar[2], Lubos Mitas[3], Jaroslav Fabian[1], and Ivan Stich[2,4] — [1]University of Regensburg, Regensburg, Germany — [2]Slovak Academy of Sciences, Bratislava, Slovakia — [3]North Carolina State University, Raleigh, NC — [4]University of Ss. Cyril and Methodius, Trnava, Slovakia

Quantum Monte Carlo (QMC) is applied to obtain the fundamental (quasiparticle) electronic band gap, Δ_f, of a semiconducting 2D phosphorene. Similarly to other 2D materials, the electronic structure of phosphorene is strongly influenced by reduced screening, making it challenging to obtain reliable predictions by conventional methods. Using the recently uncovered universal scaling between the exciton binding energy and Δ_f, we predict the optical gap of about 1.7 eV that can be directly related to experiments. The QMC gaps agree with recent optical absorption and photoluminescence measurements. We also predict the cohesion of phosphorene to be only slightly smaller than that of the bulk crystal. Our investigations not only benchmark GW methods and experiments, but also open the field of 2D electronic structure to computationally intensive but highly predictive QMC methods which include many-body effects such as electronic correlations. We were supported by GRK Grant No. 1570, the International Doctorate Program Topological Insulators of the Elite Network of Bavaria, and DFG SFB 1277 (B07). We acknowledge the Gauss Centre for Supercomputing (www.gauss-centre.eu) for funding.

CPP 21.5 Tue 10:30 H36

Interlayer band-to-band tunneling in h-BN encapsulated MoS2-WSe2 heterojunction — •Phanish Chava[1,2], Vivek Mootheri[1], Himani Arora[1,2], Kenji Wantanbe[3], Takashi Taniguchi[3], Manfred Helm[1], and Artur Erbe[1] — [1]Helmholtz Zentrum Dresden-Rossendorf, Bautzner Landstrasse 400, 01328 Dresden, Germany — [2]TU Dresden, 01062 Dresden, Germany — [3]National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

Quantum mechanical band-to-band tunneling (BTBT) is a particular type of carrier injection mechanism which is responsible for the electronic transport in tunneling based devices like Esaki diode and Tunnel Field Effect Transistor (TFET). Atomically thin layers of transition metal dichalcogenides (TMDCs) are promising semiconducting materials for realizing such devices owing to their sharp interfaces. In this work, we demonstrate BTBT between the layers of molybdenum disulfide (MoS2) and tungsten diselenide (WSe2) in a MoS2-WSe2 heterojunction which is encapsulated with hexagonal boron nitride (h-BN) on the top and bottom. Also, we employ few-layer graphene as the contact material to the heterojunction thereby forming a 2D-2D van der Waals contact. We find that the device works as a p-TFET for for negative top gate voltages and an n-MOSFET for positive top gate voltages. The device exhibits Negative Transconductance (NTC) in the positive gate voltage regime, a minimum sub-threshold swing of about 170 mV / dec at 125 K and an ON-OFF ratio of about 10^6.

CPP 21.6 Tue 10:45 H36

Effects of the Fermi Level Energy on the Adsorption of O2 to Monolayer MoS2 — •Philip Klement[1], Christina Steinke[3], Sangam Chatterjee[1], Tim Wehling[3], and Martin Eickhoff[2] — [1]Institute of Experimental Physics I and Center for Materials Research, Justus Liebig University Giessen, D-35392 Giessen, Germany — [2]Institute of Solid State Physics, University of Bremen, D-28359 Bremen, Germany — [3]Institute for Theoretical Physics and Bremen Center for Computational Material Sciences, University of Bremen, D-28359 Bremen, Germany

Two-dimensional transition metal dichalcogenides possess large surface-to-volume ratios that make them ideal candidates for sensing applications such as detecting the surface adsorption of specific gas molecules. The resulting changes of the electrical and optical properties allow for detection and analysis of interaction mechanisms at the sensing interface. Specifically, we investigate the influence of O2 adsorption on monolayer MoS2 and the role of the Fermi level energy in

this process. We record the response in photoluminescence and transport properties of monolayer MoS2 upon O2 adsorption and the impact of external electric gating. We find an increase of the photoluminescence intensity and a reduction of the conductivity upon O2 adsorption, and show that the adsorption can be enhanced by an increase of the Fermi level energy. These results demonstrate that ionosorption of O2 on MoS2 by charge transfer only occurs if free carriers are available in the conduction band of MoS2. Furthermore, photoluminescence recording is rendered advantageous for sensing.

15 min. break

CPP 21.7 Tue 11:15 H36

Zeeman splitting and inverted polarization of biexciton emission in monolayer WS_2 — •Philipp Nagler[1], Mariana V. Ballottin[2], Anatolie A. Mitioglu[2], Mikhail V. Durnev[3], Takashi Taniguchi[4], Kenji Watanabe[4], Alexey Chernikov[1], Christian Schüller[1], Mikhail Glazov[3], Peter C. M. Christianen[2], and Tobias Korn[1] — [1]University of Regensburg — [2]High Field Magnet Laboratory, Nijmegen — [3]Ioffe Institute, St. Petersburg, Russia — [4]NIMS, Tsukuba, Japan

Monolayer TMDCs are an ideal testbed to study the physics of quasiparticles in the two-dimensional limit. Besides excitons, more complex many-body states such as trions and biexcitons can emerge due to the strong Coulomb interaction in these materials. Here, we shed light on the intricate many-body physics of biexcitons in monolayer WS_2 [1]. The encapsulation of the monolayer between two sheets of hBN significantly reduces the overall spectral broadening and allows us to observe biexciton emission with linewidths below 5 meV at 4 K. In magneto-PL experiments, we observe an inverted field-induced polarization, implying a preferential population of the high-energy peak in emission. We explain this unusual phenomenon by considering the evolution of the total energy of the biexciton complex in a magnetic field. Based on the experimental results and the developed model we are able to identify the momentum-space configuration of the optically dominant biexciton state of monolayer WS_2.

[1] P. Nagler et al., *Phys. Rev. Lett.* **121**, 057402 (2018)

CPP 21.8 Tue 11:30 H36

Electromagnetically induced transparency in second-harmonic generation from monolayer WSe2 — •Kai-Qiang Lin, Robert Martin, Sebastian Bange, and John Lupton — Institut für Experimentelle und Angewandte Physik, Universität Regensburg, 93053 Regensburg, Germany

Electromagnetically induced transparency (EIT) occurs in atomic systems and shows versatile applications in slow-light generation, gain without inversion and optical quantum-information processing. We demonstrate a cavity-free, atomic-like EIT effect in single-layer crystals of WSe2, probed by exploiting the intrinsic second-harmonic generation (SHG) arising from the breaking of inversion symmetry.[1] Under conditions of double resonance of the driving and radiated field with the fundamental transitions, the SHG spectrum bifurcates. The feature follows a pump-wavelength-dependent spectral anticrossing, accurately described by a ladder-type three-level model. Crucially, the SHG power-law exponent diverges from the canonical value of 2 to follow a Fano-like dispersion with wavelength. This dispersion is retained at room temperature, implying persistence of quantum interference. Our work opens up opportunities to exploit quantum nonlinear optics such as inversionless gain in the solid state.

[1]. K.-Q. Lin, S. Bange and J. M. Lupton, Nature Physics, in press. (preprint: https://arxiv.org/abs/1811.09479)

CPP 21.9 Tue 11:45 H36

Studying exciton-phonon interaction in a $MoSe_2$ monolayer by fluorescence-detected 2D electronic micro-spectroscopy — •Donghai Li[1], Chiara Trovatello[2], Stefano Dal Conte[2], Matthias Nuss[1], Giancarlo Soavi[3], Andrea Ferrari[3], Giulio Cerullo[2,4], and Tobias Brixner[1] — [1]Institut für Physikalische und Theoretische Chemie, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany — [2]Dipartimento di Fisica, Politecnico di Milano, Piazza Leonardo da Vinci 32, 20133 Milano, Italy — [3]Cambridge Graphene Centre, University of Cambridge, Cambridge, UK. — [4]Istituto di Fotonica e Nanotecnologie, CNR, Piazza Leonardo da Vinci 32, 20133 Milano, Italy

Monolayer transition metal dichalcogenides (TMDs) received extensive attention as they have extraordinary opto-electronic characteristic. Exciton-phonon interaction in TMD monolayers can result in sidebands in their absorption spectra, for some dark states are activated by exciton-phonon scattering [1]. However, in the absorption spectra, the phonon-induced sidebands are obscured by inhomogeneous broadening. Here, we study the excitonic physics of a $MoSe_2$ monolayer using fluorescence-based two-dimensional (2D) electronic spectroscopy. We confirm and extend the theoretical prediction [1] by our observations of sidebands and their quantum beating behavior within the 2D spectra. The analysis indicates a four-level energy structure with one dark state near the excited state and an additional, unpredicted, one near the ground state activated by exciton-phonon scattering.

[1] D. Christiansen et al., Phys. Rev. Lett. **119**, 187402 (2017)

CPP 21.10 Tue 12:00 H36

Transport and photoelectron spectroscopy of few-layer epitaxial WSe_2 — •Hiro Nakamura[1], Avaise Mohammed[1], Philipp Rosenzweig[1], Kathrin Müller[1], Peter Wochner[1], Armin Schulz[1], Mona Stadler[2], Michael Jetter[2], Peter Michler[2], Ulrich Starke[1], and Hidenori Takagi[1,3,4] — [1]Max Planck Institute for Solid State Research — [2]Institut für Halbleiteroptik und Funktionelle Grenzflächen, University of Stuttgart — [3]Department of Physics, University of Tokyo — [4]Institute for Functional Matter and Quantum Technologies, University of Stuttgart

Giant spin splitting of monolayer WSe_2 is an interesting element both for spintronics and topological phenomena. Here, we present electronic and structural properties of few-layer WSe_2 grown by hybrid pulsed-laser deposition. Angle-resolved, ultraviolet and X-ray photoelectron spectroscopy reveal the band structure of monolayer WSe_2 including strong spin-orbit splitting, as well as clarify the band alignment between WSe_2 and the underlying graphene/SiC substrate, indicating electron transfer from graphene to WSe_2. To access the valence bands in transport, hole-doping of few-layer WSe_2 has been performed using Nb as a dopant. The Hall effect and transport measurements confirmed the expected density of hole carriers, and associated metallic conduction down to low temperatures for optimal films. Synchrotron X-ray diffraction sheds further light on structure-property relationships, in particular strain effects in the WSe_2 layer.

CPP 21.11 Tue 12:15 H36

Excitation-Induced Transition from Direct to Indirect Band Gaps in Monolayer TMDs — •Daniel Erben[1], Alexander Steinhoff[1], Michael Lorke[1,2], Tim Wehling[1,2], Christopher Gies[1], and Frank Jahnke[1] — [1]Institute for Theoretical Physics, University of Bremen — [2]Bremen Center for Computational Materials Science, University of Bremen

Monolayers of transition metal dichalcogenides (TMDs) show exceptionally strong Coulomb interaction between charge carriers due to the small thickness and weak dielectric screening. Many-body interactions induced by excited charge carriers directly influence the electronic and optical properties in these materials. Strong many-particle renormalizations caused by the Coulomb interaction of the excited carriers will be discussed for MoS_2, $MoSe_2$, WS_2 and WSe_2. We solve the semiconductor Bloch equations on the full Brillouin zone using ab-initio band structures and interaction matrix elements.

Large excitation-dependent band-gap renormalizations are found. In all four materials, the conduction band Σ-valley exhibits a stronger shift to lower energies than the K-valley. As a result, all four TMDs show a tendency to become more indirect or even undergo a transition from a direct to indirect band gap with increasing excited carrier density.

For optical excitation of monolayer TMDs, we also study the connection between pump fluence and excited carrier density. The contributions of various many-body effects to a strong non-linearity are identified.

CPP 21.12 Tue 12:30 H36

Theory of Exciton-Exciton Coupling in Atomically Thin Transition Metal Dichalcogenides — •Florian Katsch, Malte Selig, and Andreas Knorr — Institut für Theoretische Physik, Nichtlineare Optik und Quantenelektronik, Technische Universität Berlin, 10623 Berlin, Germany

The valley-selective circular dichroism in monolayer transition metal dichalcogenides (TMDCs) and the subsequent fast valley depolarization due to intervalley Coulomb interactions enables the study of new many-body physics in these atomically thin materials. Here, a microscopic approach is presented to describe the optical response of monolayer TMDCs dominated by strongly correlated, bound electron-

hole pairs [1, 2]. The approach includes Hartree–Fock and correlation effects up to two excitonic excitations [3], as well as TMDC typical Coulomb intra- and intervalley coupling and exciton-phonon interactions [4]. The developed theory is applied to access the exciton dynamics in the coherent limit and contributes to the understanding of valley-selective pump-probe experiments [5, 6].

[1] A. L. Ivanov and H. Haug, *Phys. Rev. B* **48**, 1490 (1993).
[2] F. Katsch et. al., *Phys. Status Solidi B*, 1800185 (2018).
[3] V. M. Axt and A. Stahl, *Z. Phys. B* **93**, 2 (1994).
[4] M. Selig et. al., *2D Mater.* **5**, 035017 (2018).
[5] C. Mai et. al., *Nano Lett.* **14**, 202 (2013).
[6] R. Schmidt et. at., *Nano Lett.* **16**, 2945 (2016).

CPP 21.13 Tue 12:45 H36

Creation and optical spectroscopy of localized excitons in 2D MoS_2 — •Oleg Gridenco, Sven Mehrkens, Kathrin Sebald, Christian Tessarek, Martin Eickhoff, and Jürgen Gutowski — Institute of Solid State Physics, University of Bremen, Otto-Hahn-Allee 1, D-28359 Bremen, Germany

It is known that missing atoms in a semiconductor are exciton trapping sites, moreover, excitons can bind to impurity atoms or can be trapped in a potential well created by local strain or structural defects. In this context, structuring with a focused ion beam gives the opportunity for manipulation of 2D materials on the nanometer scale. In this study, we explore how focused gallium ion irradiation affects the intrinsic luminescence and vibrational properties of atomically thin MoS_2. Defects were introduced by scanning the Ga^+ ion probe over a certain area of the flake using a focused ion beam (FIB). The amount of defects was controlled by varying the Ga^+ ion dose starting from 2 x 10^{12} ions/cm^2 until the PL signal was completely vanished (2 x 10^{13} ions/cm^2). After Ga^+ ion irradiation, micro-photoluminescence measurements at T=4K show that the A exciton emission is suppressed and a new peak, a bound exciton (D) located at 1.75 eV, emerges. This broad peak is redshifted by ΔE$\sim$170 meV with respect to the neutral exciton X emission. Encapsulating monolayer MoS_2 into hBN drastically reduces the inhomogeneous contributions to the exciton linewidth [1]. The possibilities of tailoring optically active defect centers in 2D MoS_2 to even host single-photon emitters will be discussed.

[1] E. Courtade et al., Appl. Phys. Lett. **113**, 032106 (2018).