HL 78: 2D Materials beyond Graphene: Dynamics and Excitation

Time: Thursday 10:30–13:30 Location: S054

Invited Talk HL 78.1 Thu 10:30 S054

Spin- and Pseudospin-Polarized Excited States in bulk WSe_2 — Roman Bertoni[1], Christopher Nicholson[1], Lutz Waldecker[1], Michele Puppin[1], Claude Monney[2], Cephise Cacho[3], Hannes Huebener[4], Umberto De Giovannini[4], Angel Rubio[4], Martin Wolf[1], and •Ralph Ernstorfer[1] — [1]Fritz-Haber-Institut der MPG, Berlin, DE — [2]University of Zurich, Zurich, CH — [3]Rutherford Appleton Laboratory, Didcot, UK — [4]University of the Basque Country, San Sebastian, ES

The peculiar electronic structure of layered semiconducting transition metal dichalgogenides (TMDC) like WSe_2 gives rise to internal quantum degrees of freedom of the electrons in addition to the spin, namely valley and layer pseudospins. Employing XUV-based time- and angle-resolved photoemission spectroscopy (trARPES) with resonant excitation of excitonic transitions, we observe circular dichroism in the excited state population in the K valleys of the topmost trilayer of bulk WSe_2. Such spin-, valley and layer-polarized excitations are a manifestation of broken site symmetry in an inversion-symmetric crystal. The valley- and layer-resolved view on excited state dynamics provided by trARPES is complemented by the investigation of electron-lattice coupling in multilayer WSe_2 with femtosecond electron diffraction. Latter studies reveal the dynamics of global energy transfer from electronic to vibrational degrees of freedom in TMDCs subsequent to excitonic as well as interband excitation.

HL 78.2 Thu 11:00 S054

Exciton dynamics in two-dimensional materials with strong spin-orbit interaction: $MoSe_2$ versus WSe_2 — •Daniel Schmidt[1], Tillmann Godde[2], Johannes Schmutzler[1], Marc Assmann[1], Jörg Debus[1], Freddie Withers[3], Osvaldo Del Pozo-Zamudio[2], Konstantin S. Novoselov[3], Andre Geim[3], Manfred Bayer[1], and Alexander Tartakovskii[2] — [1]Experimentelle Physik 2, Technische Universität Dortmund, D-44221 Dortmund, Germany — [2]Department of Physics and Astronomy, University of Sheffield, Sheffield S3 7RH, UK — [3]School of Physics and Astronomy, University of Manchester, Oxford Road, Manchester M13 9PL, UK

Monolayers of semiconducting transition metal dichalcogenides such as MoS_2, WS_2, $MoSe_2$ and WSe_2 have attracted considerable attention following the discovery of the indirect-to-direct bandgap transition from bulk to monolayer material and the coupling of spin and valley degrees of freedom in atomically thin layers. An important characteristic of these compounds is the strong spin-orbit interaction, which leads to a splitting between dark and bright exciton sub-bands. A detailed understanding of dark and bright exciton dynamics and non-radiative processes is important for light emitting applications as has been demonstrated in other systems such as phosphorescent organic light emitting diodes. We measure time-integrated and -resolved PL in monolayers of $MoSe_2$ and WSe_2 in a wide range of temperatures from 10 to 300K and gain insights into the exciton and trion dynamics. Our study reveals similar carrier dynamics for both materials, whereas pronounced differences have been observed for the overall PL intensities.

HL 78.3 Thu 11:15 S054

Electron dynamics in eiptaxial single layer MoS_2 — •Antonija Grubisic-Cabo[1], Jill A. Miwa[1], Signe S. Gronborg[1], Jonathon M. Riley[2], Jens C. Johannsen[3], Cephise Cacho[4], Oliver Alexander[4], Richard T. Chapman[4], Emma Springate[4], Marco Grioni[3], Jeppe V. Lauritsen[1], Phil D. C. King[2], Philip Hofmann[1], and Soren Ulstrup[1] — [1]Aarhus University, DK — [2]University of St. Andrews, UK — [3]Ecole Polytechnique Federale de Lausanne, CH — [4]CLF, STFC Rutherford Appleton Laboratory, UK

The current understanding of the optical properties and excited carrier dynamics in single-layer and few-layer transition metal dichalcogenides relies largely on a series of photoluminescence and differential absorption measurements. Since excitons dominate the optical response, the dynamics of free carriers cannot be studied directly. Here, we use time- and angle-resolved photoemission spectroscopy to directly measure free carriers in epitaxial single layer MoS_2 grown on either Au(111) or on graphene. For MoS_2/Au(111) we determine an ultrafast (50 fs) extraction of excited free carriers via the metal and ascertain a direct quasiparticle band gap of 1.95 eV. The observed quasiparticle gap is significantly smaller than the theoretically estimated value for free-standing MoS_2. This can be explained by a strong renormalisation of the band gap. For MoS_2 on graphene, we find indications of induced band shifts that lead to a time-dependence of the electronic structure.

HL 78.4 Thu 11:30 S054

Understanding optical properties of atomically thin semiconductors from a many-body perspective — •Matthias Drüppel, Thorsten Deilmann, Peter Krüger, and Michael Rohlfing — Institut für Festkörpertheorie, Westfälische Wilhelms-Universität, 48149 Münster, Germany

Transition metal dichalcogenides (TMDCs) open the door to a fascinating, fast growing field of two dimensional atomically thin semiconductors. This increases the demand for novel theoretical techniques, which allow to reliably calculate the optical properties in large systems, the inclusion of substrates or even correlation between more than two particles.

We take the state of the art approach of DFT $\rightarrow$ GW $\rightarrow$ Bethe-Salpeter equation (BSE) and apply the efficient LDA+GdW [1] method. This enables us to describe many-body electronic excitations at moderate numerical cost, being able to treat systems of up to 100 atoms. In the LDA+GdW approximation the quasiparticle self-energy corrections result from the difference between the correct semiconducting screening and hypothetical metallic screening.

Our results show how the optical properties of TMDCs are modified in different situations, e.g. when the screening of the substrate is taken into account, in the presence of vacancies, or when three excited particles form a correlated trion state.

[1] M. Rohlfing, Phys. Rev. B. **82**, 205127 (2010)

HL 78.5 Thu 11:45 S054

Scanning tunneling light emission from single layer MoS_2 — •Christian Lotze, Nils Krane, Julia Läger, Gaël Reecht, and Katharina J. Franke — FU Berlin, FB Physik, Arnimallee 14, 14195 Berlin

Transition-metal dichalcogenides form a group of interesting 2d materials. Among them, the semiconductor MoS_2 has attracted great interest, because it has been shown that it turns from an indirect-gap into a direct-gap semiconductor when reduced to a single layer [1]. As such, potential applications involve its usage for instance as light emitting device.

Here, we present a combined low temperature scanning tunneling (STM) and light emission (LE) study on MoS_2/Au(111). The STM geometry allows to locally inject electrons or holes with the tip into the single layer MoS_2. Inelastically tunneling electrons and holes can give rise to emission of photons [2,3]. Here, we detect and analyze spectrally resolved the electroluminescence from the MoS_2 monolayer on Au(111). We correlate these LE spectra with the electronic structure, that we obtained from scanning tunneling spectroscopy. Moreover we will look into the spatial variation of the LE signals and the role of defect sites.

[1] Mak *et al.*, PRL 105, 136805 (2010)
[2] Berndt *et al.*, PRL 67, 3796 (1991)
[3] Hoffmann *et al.*, Phys. Rev. Lett. 93, 076102 (2004)

HL 78.6 Thu 12:00 S054

Ultrafast photocurrent dynamics in monolayer MoS2 — Eric Parzinger, Anna Vernickel, Alexander Holleitner, and •Ursula Wurstbauer — Walter Schotty Institut and Physik Department, Technical University of Munich, Germany

Atomically thin semiconducting transition metal dichalcogenides such as MoS_2 are emergent materials for optical and electronic circuits. For possible high-frequency applications, we investigate the ultrafast photocurrents in monolayer MoS_2 on a picosecond time scale utilizing a recently developed pump-probe spectroscopy [1-3]. The observed photocurrent contains three major contributions [4]. An initial ultrafast response of $\leq$ 5ps is followed by an exponential decay within a few hundreds of picoseconds. The third very slow contribution peaks at around 1.5 ns. This slow part dominates the overall time-integrated photocurrent intensity. We discuss the impact of laser-induced heating,

the one of built-in fields at metal contacts, and the role of trap states. We acknowledge the financial support by the ERC-grant NanoREAL, the DFG excellence cluster Nanosystems Initiative Munich (NIM), and BaCaTec. [1] L. Prechtel, et al. Nature Communications 3, 646 (2012). [2] A. Brenneis, et al. Nature Nanotechnology 10, 135 (2015). [3] C. Kastl, et al. Nature Communications 6, 6617 (2015). [4] E. Parzinger et al. (2016).

HL 78.7 Thu 12:15 S054

Electronic excitations in transition metal dichalcogenides under the influence of dielectric environments — •Malte Rösner[1], Alexander Steinhoff[2], Roelof Groenewald[3], Frank Jahnke[2], Stephan Haas[3], Christopher Gies[2], and Tim O. Wehling[1] — [1]Institut für Theoretische Physik and Bremen Center for Computational Materials Science, Universität Bremen, Bremen, Germany — [2]Institut für Theoretische Physik, Universität Bremen, Bremen, Germany — [3]Department of Physics and Astronomy, University of Southern California, Los Angeles, CA, USA

We present a material-realistic approach to describe electronic interaction effects in transition metal dichalcogenides. On the basis of the Wannier function continuum electrostatics (WFCE) method [1], we are able to include the effects of the dielectric environment, which could serve as a promising tuning knob to control 2D material's properties. We study the doping dependence and the influence of different types of dielectric environments to electronic and plasmonic properties. We find electronic band structure and plasmon dispersion changes on the eV scale.

[1] M. Rösner et al., Phys. Rev. B 92, 085102 (2015)

HL 78.8 Thu 12:30 S054

Optoelectronic properties of sub-nanometer WS2 and TiS3 investigated by scanning near-field optical microscopy and nano-FTIR spectroscopy by using synchrotron radiation — •P. Patoka[1], G. Ulrich[1], A. Nguyen[2], A. Lipatov[3], A. Sinitskii[3], P. Hermann[4], B. Kästner[4], A. Hoehl[4], L. Bartels[2], P. Dowben[5], G. Ulm[4], and E. Rühl[1] — [1]Physikalische Chemie, Freie Universität Berlin, Germany — [2]Dept. of Chemistry, Univ. of California Riverside, U.S.A. — [3]Dept. of Chemistry, Univ. of Nebraska-Lincoln, U.S.A. — [4]Physikalisch-Technische Bundesanstalt (PTB), Germany — [5]Dept. of Physics and Astronomy, Univ. of Nebraska-Lincoln, U.S.A.

Among the 2D electronic materials that have received increased attention recently are the transition metal dichalcogenides (TMD). These materials, especially below nanometer thickness, exhibit promising optoelectronic properties for applications in low-dimension electronic circuits. The combined use of scattering-type near-field optical microscopy and the broadband synchrotron radiation source MLS (PTB, Berlin) allows for the highly sensitive spectromicroscopic characterization of such 2D semiconductors with a spatial resolution below 30 nm. We will present recent results on near-field imaging and nano-FTIR spectroscopy in mid-infrared regime down to monolayer thick TMD structures. Investigated are optical responses of WS2, such as its interaction with the optical phonon mode of the SiO2 substrate. We will also show evidence for high charge accumulation at the edges of the TiS3 structures revealed by optical mapping using tunable CO2 laser.

HL 78.9 Thu 12:45 S054

Investigations on the Phonon Spectrum of $TiSe_2$ in the CDW Phase — •Roland Hott, Rolf Heid, and Frank Weber — Karlsruhe Institute of Technology, Institute of Solid State Physics, P.O.B. 3640, D-76021 Karlsruhe, Germany

We report recent results of our investigations on the Charge Density Wave (CDW) phase transition in $TiSe_2$, performed both experimentally by means of high resolution Inelastic X-ray Scattering (IXS) as well as theoretically by Density Functional Theory (DFT) based ab-initio phonon calculations [1].

We extended our calculations to the case of Cu-doping where we found a huge hardening of the CDW-related soft phonon due to strong chemical bonding of the Cu atoms to the $TiSe_2$ host lattice. Moreover, we investigated the phonon spectrum of $TiSe_2$ in the CDW phase where we find the expected stabilisation of the lattice. Nevertheless, we still obtain here a sizeable electron-phonon coupling for the phonons which derive from the soft phonons of the normal (CDW-undistorted) phase.

[1] F. Weber, S. Rosenkranz, J.-P. Castellan, R. Osborn, G. Karapetrov, R. Hott, R. Heid, K.-P. Bohnen, A. Alatas, PRL **107**, 266401 (2011)

HL 78.10 Thu 13:00 S054

Ultrafast carrier multiplication in 1*T*-$TiSe_2$ — •Stephan Michael[1], Steffen Eich[1], Henry C. Kapteyn[2], Margaret M. Murnane[2], Michael Bauer[3], Kai Rossnagel[3], Hans Christian Schneider[1], Martin Aeschlimann[1], and Stefan Mathias[4] — [1]University of Kaiserslautern — [2]JILA, University of Colorado and NIST — [3]University of Kiel — [4]University of Göttingen

1*T*-$TiSe_2$ is a transition metal dichalcogenide, which has a charge density wave (CDW) state below a temperature of around 200 K, which may be due to an excitonic insulator mechanism and/or an Jahn-Teller effect. We studied optically excited carrier dynamics on ultrashort timescales in the CDW phase using time-resolved ARPES measurements and an effective two-band model including carrier-carrier Coulomb scattering. In the framework of this model we analyze the ultrafast response of this material to optical excitation, which is seen in the experiment. We show that carrier multiplication in the form of impact ionization is the most satisfactory explanation for the ultrafast redistribution of spectral weight observed in the ARPES measurements.

HL 78.11 Thu 13:15 S054

Charge density wave kinetics in 1T-TaS_2 monitored by ultrafast LEED — •Simon Schweda[1], Gero Storeck[1], Sebastian Schramm[1], Max Gulde[1], Kai Rossnagel[2], Sascha Schäfer[1], and Claus Ropers[1] — [1]IV. Physikalisches Institut, Universität Göttingen, D-37077 Göttingen — [2]Institut für Experimentelle und Angewandte Physik, Universität Kiel, D-24098 Kiel

We developed an ultrafast low-energy electron diffraction (ULEED) setup for the study of time-resolved structural dynamics at surfaces, extending our previous approach operating on ultrathin films in transmission [1]. A laser-driven nanometric needle emitter provides well-collimated electron pulses with durations of few tens of ps at the sample for electron energies in the range of 50-200 eV.

In a first application of this technique, we investigate optically induced transitions between charge density wave (CDW) phases [2] at a single-crystalline 1T-TaS_2 surface. In particular, the recovery of the nearly commensurate (NC) room-temperature phase after laser-excitation to the incommensurate (IC) phase is resolved in the time-domain. We find a strong dependence of the formation time on the energy density deposited. Furthermore, at higher optical fluences, the appearance of metastable NC antiphase domains is observed, caused by a rapid quench after optical excitation. Our results demonstrate the potential of ULEED for the study of complex ultrafast structural and electronic processes at surfaces.

[1] M. Gulde et al., Science 345, 200 (2014)
[2] M. Eichberger et al., Nature 468, 799 (2010)