O 30: 2D Materials I: Electronic Structure

Time: Tuesday 10:30–13:15   Location: GER 37

O 30.1 Tue 10:30 GER 37

Controlling the charge density wave transition in single-layer $TiTe_{2x}Se_{2(1-x)}$ alloys by band gap engineering — ●Tommaso Antonelli, Andela Zivanovic, Chiara Bigi, Brendan Edwards, Akhil Rajan, and Phil King — University of St. Andrews

The narrow gap semiconductor monolayer (ML) $TiSe_2$ undergoes a controversial charge density wave transition [1, 2] whose nature is still poorly understood. Recently, a similar instability has been observed in the sister semimetallic compound ML-$TiTe_2$ showing a weaker CDW coupling as compared to the selenide compound [3, 4]. Using molecular beam epitaxy, we grow ML-$TiTe_{2x}Se_{2(1-x)}$ alloys to engineer the band gap in the normal phase and tune the CDW coupling in the system. Performing angle-resolved photoemission spectroscopy on the alloy films, we demonstrate the effective semiconductor to semimetal transition in the high-temperature phase band structure and observe the consequent modulation of the CDW critical temperature. Our experimental results are rationalised using a simple theoretical model bringing new insight on how to manipulate this collective phase at the 2D limit.

[1] P. Chen *et al.*, Nat. Commun. 6, 8943 (2015) [2] M. D. Watson *et al.*, 2D Materials 8, 015004, (2021) [3] P. Chen *et al.*, Nat. Commun., 8, 516 (2017) [4] T. Antonelli *et al.*, npj Quantum Mater. 7, 98 (2022)

O 30.2 Tue 10:45 GER 37

Charge density wave in 2H-$NbSe_2$ studied by combined STM/nc-AFM — ●Nikhil Seeja Sivakumar[1], Marion van Midden Mavric[1], Nora Huijgen[1], Umut Kamber[2], Daniel Wegner[1], Alexander Ako Khajetoorians[1], and Nadine Hauptmann[1] — [1]Institute for Molecules and Materials, Radboud University, Nijmegen, The Netherlands — [2]Department of Physics, Princeton University, USA

Van der Waals materials exhibit interesting quantum phases such as charge density waves (CDW) which originate from an electronic instability along with structural rearrangements. One example is 2H-$NbSe_2$ which exhibits an incommensurate CDW below 33 K. The origin of the CDW in 2H-$NbSe_2$ is currently understood to be driven by electron-phonon coupling [1]. To characterize the CDW with atomic scale resolution, Scanning Tunneling Microscopy (STM) is frequently used which is sensitive to the convolution of the geometric structure and the local density of states.

Here, we utilize combined low-temperature STM and non-contact Atomic Force Microscopy (STM/nc-AFM) based on the qPlus design to study the CDW at the surface of 2H-$NbSe_2$. We detail the distancedependent contrast of the atomic lattice and the CDW in current and frequency shift images. We further compare force spectroscopy data at selected positions of the CDW unit cell and analyze different force contributions.

[1] Flicker, F., van Wezel, J. Nat. Commun. 6, 7034 (2015).

O 30.3 Tue 11:00 GER 37

Circular dichroism in valence band photoemission from $CrGeTe_3$ — ●Honey Boban[1], Mohammed Qahosh[1], Xiao Hou[1], Magdalena Szczepanik[2], Ewa Partyka-Jankowska[2], Tomasz Sobol[2], Tom G Saunderson[3,4], Yuriy Mokrousov[3,4], Mathias Kläui[3], Claus Michael Schneider[1], and Lukasz Plucinski[1] — [1]PGI-6, FZ Jülich, Germany — [2]NSRC SOLARIS, Poland — [3]University of Mainz, Germany — [4]PGI-1, FZ Jülich, Germany

$CrGeTe_3$(CGT) is a layered ferromagnetic semiconductor with a bulk Curie temperature (T_c) of 63K[1] and an indirect bandgap of 0.4eV[2]. A previous band structure mapping study of CGT[1] revealed band renormalizations below and above T_c and resonance enhancements at the Cr 3p absorption edge. CGT contains large Te atoms (Z=52), therefore spin-momentum locked splittings, Weyl nodes, and anticrossings are expected due to a combination of ferromagnetism and spin-orbit coupling, as well as the related Berry curvature physics. We performed light-polarization-dependent and temperature-dependent angle-resolved photoemission (ARPES) measurements on surfaces of bulk CGT. We have observed renormalizations in the dichroic ARPES maps below and above T_c, which could be indirectly related to magnetic ordering, even though our technique most likely averages spin-up and spin-down domains. Circular dichroic ARPES maps contain an intrinsic contribution due to the orbital angular momentum through dipole selection rules and an extrinsic contribution due to the experimental geometry. Our current work focuses on disentangling the two effects. Ref: [1] PRB **101**, 205125(2020), [2] PRB **98**, 125127(2018)

O 30.4 Tue 11:15 GER 37

Investigation of phase-transitions in $MoTe_2$ using ToF-MM — ●O. Fedchenko[1], F. Diekmann[2], K. Medjanik[1], M. Kallmayer[3], S. Babenkov[1], D. Vasilyev[1], O. Tkach[1], M. Souliou[4], M. Frachet[4], M. Le Tacon[4], K. Rossnagel[2], A. Winkelmann[5], G. Schönhense[1], and H.-J. Elmers[1] — [1]JGU, Institut für Physik, Mainz, Germany — [2]Institut für Experimentelle und Angewandte Physik, CAU Kiel, Germany — [3]Surface Concept GmbH, Mainz — [4]KIT, IQMT, Karlsruhe, Germany — [5]ACMN, AGH University of Science and Technology, Krakow, Poland

$MoTe_2$ is unique among transition metal dichalcogenides (TMDCs) because it can be grown in both the semiconducting (2H) and semimetallic (1T') phase. The transitions between these phases can be controlled with temperature and doping concentration. Using time-of-flight momentum microscopy (ToF-MM), the thermally-induced inversion symmetry breaking in doped $Mo_{1-x}W_xTe_2$ is investigated. Pristine $MoTe_2$ and $Mo_{0.98}W_{0.02}Te_2$ take the 2H phase, which does not show any thermal induced phase transition. For increased W doping concentration we obtain the metastable monoclinic 1T' phase at 300K (RT), which then can be transformed to the orthorhombic T_d phase at T<170 K (LT). Raman and XRD measurements confirm the structural transitions.The T_d-phase exhibits topological states identified by Fermi arcs, up to binding energy of $E_B = 75$ meV.

O 30.5 Tue 11:30 GER 37

Geometry-induced spin-filtering in photoemission maps from WTe_2 surface states — Tristan Heider[1], Gustav Bihlmayer[2], Jakub Schusser[3,4], Frierich Reinert[4], Jan Minar[3], Stefan Blügel[2], Claus M. Schneider[1], and ●Lukasz Plucinski[1] — [1]PGI-6 Forschungszentrum Jülich — [2]PGI-1 Forschungszentrum Jülich — [3]University of West Bohemia, Pilsen, Czech Republic — [4]Experimentelle Physik VII, Universität Würzburg

WTe_2 is a semi-metallic quantum material that exhibits non-saturating magnetoresistance and potentially hosts Weyl type-II nodes [1]. Through laser-driven spin-polarized ARPES Fermi surface mapping, we demonstrate highly asymmetric spin textures of electrons photoemitted from the surface states of WTe_2 [2]. Such asymmetries are not present in the initial state spin textures, which are bound by time-reversal and crystal lattice mirror plane symmetries. The findings are reproduced qualitatively by theoretical modeling within the one-step model photoemission formalism, and a simple toy-model suggests that a similar effect shall be observed in other materials with low symmetry. Our spin-polarized maps, with detail comparable to the previous spin-integrated maps [3], have been measured using the newly developed high-resolution instrument at PGI-6 in Jülich that is based on a hemispherical analyzer with the scanning lens, an exchange-scattering spin detector, and a *cw* 6 eV laser.

[1] P. K. Das et. al. Electron. Struct. 1, 014003 (2019) and refs. therein. [2] T. Heider et al., arXiv:2210.10870 (2022). [3] F. Y. Bruno et al., Phys. Rev. B 94, 121112 (2016).

O 30.6 Tue 11:45 GER 37

Observing Duffing oscillations in 2D materials by ultrafast electron imaging — ●Alexander Schröder, Lina Hansen, and Sascha Schäfer — Institute of Physics, University of Oldenburg, Germany

Two-dimensional materials are an ideal test bed for probing non-linear mechanical resonances. With sample thicknesses down to the atomic scale, the nonlinear regime of mechanical resonances can be observed even for small actuation forces. Previously, the response of such nano- and micromechanical systems was probed by optical or electronic means, leaving details of the mechanical mode structures and their connection to atomic defects unresolved. Here, we present the nanoscale probing of nonlinear Duffing resonances in free-standing graphite membranes using large-angle convergent-beam diffraction in an ultrafast transmission electron microscope equipped with an event-based electron detector with nanosecond temporal resolution. The vibrational membrane modes are excited by a diode laser beam with a tunable

modulation rate and a series of resonances are observed in the MHz frequency range with high quality factors (up to 10^4). As expected for prototypical Duffing resonances, increasing the optical excitation strength yields a strong nonlinear hardening of the oscillator, with an increase in vibrational amplitude, an asymmetric broadening of the resonance line shape, bistable behavior, and a characteristic frequency-dependent phase shift. The novel detection scheme as proposed here is ideally suited for characterizing the behavior of ultra-small resonators difficult to address with other techniques.

O 30.7 Tue 12:00 GER 37

Limitations of supercell extrapolation for charged defects at surfaces & 2D materials — •Christoph Freysoldt[1], Anne Marie Z. Tan[2,3], Richard G. Hennig[2], and Jörg Neugebauer[1] — [1]Max-Planck-Institut für Eisenforschung, Max-Planck-Straße 1, 40237 Düsseldorf, Germany — [2]Department of Materials Science and Engineering, University of Florida, Gainesville, Florida 32611, USA — [3]School of Mechanical and Aerospace Engineering, Nanyang Technological University, Singapore 639798, Singapore

Ab initio calculations of defects often employ the repeated slab approach for its simplicity. Unfortunately, spurious interactions between the defect and its periodic images make it sometimes challenging to estimate the dilute limit, that is relevant for comparison to experiment, from computationally affordable system sizes. A simple and seemingly straightforward approach is to extrapolate from a series of finite supercell sizes to the infinite-size limit, assuming a smooth, polynomial dependence of the energy on inverse supercell size. By means of explicit density-functional theory supercell calculations and simplified electrostatic models we demonstrate that the dependence of the formation energy on supercell size can be more complex than is commonly assumed. We show that this complexity cannot be captured by the simple extrapolation approaches. In contrast, suitable correction schemes are shown to provide rapidly converging results even for 2D materials.

[1] C. Freysoldt, J. Neugebauer, A. M. Z. Tan, R. G. Hennig, Phys. Rev. B **105**, 014103 (2022).

O 30.8 Tue 12:15 GER 37

Excited state geometry relaxation of point defects in hexagonal boron nitride — •Alexander Kirchhoff, Thorsten Deilmann, and Michael Rohlfing — Westfälische Wilhelms-Universität Münster, Institut für Festkörpertheorie, Wilhelm-Klemm-Straße 10, 48149 Münster

The optical gap of pristine hexagonal boron nitride is above $5\,\mathrm{eV}$ and therefore in the deep UV range. Emitters in the optical regime can act as single photon sources and gained attraction due to their possible room temperature stability. However, their atomic composition is still elusive. Experiments combining spectroscopic methods to probe ground as well as excited state of defects in hBN have revealed the relevance of the dependence of optoelectronic properties on the nuclear geometry.

In our work, this dependence is studied within the framework of density functional theory and the GW/BSE approximation. We focus on carbon based defects in hBN and present a detailed analysis of their excitonic spectrum [1]. $C_B O_N$ for example has a defect bound exciton at $1.5\,\mathrm{eV}$. We obtain a Stokes shift of about $1\,\mathrm{eV}$ by calculating the energy surfaces of ground and excited state.

[1] Kirchhoff, Deilmann, Krüger and Rohlfing, Phys. Rev. B 106, 045118 (2022)

O 30.9 Tue 12:30 GER 37

Semiclassical theory of plasmons in inhomogeneous two-dimensional systems — •Tjacco Koskamp, Koen Reijnders, and Mikhail Katsnelson — Radboud University, Nijmegen, The Netherlands

We consider plasmons, collective oscillations of conduction electrons, in inhomogeneous two-dimensional systems with parabolic dispersion. In recent years, the quantum regime for plasmons has become experimen-

tally accessible. Although these systems can be studied numerically, this approach is limited to relatively small systems. Recently, a comprehensive analytical theory for plasmons based on the semiclassical (WKB) approximation was proposed [1]. It describes plasmons in inhomogeneous three-dimensional systems, and is applicable when the inhomogeneity, in e.g. the electron density, varies smoothly.

In this talk, we extend this theory to plasmons in inhomogeneous two-dimensional systems. We consider the equation of motion for the density matrix and take the electron-electron interaction into account through the RPA. We solve this system of equations self-consistently, carefully separating the in-plane and out-of-plane degrees of freedom. In this way, the effective classical Hamiltonian for plasmons is obtained. Subsequently, we apply this theory to a scattering experiment. We first construct the classical trajectories and then use the semiclassical phase shift to compute the differential scattering cross section.

[1] K.J.A. Reijnders, T. Tudorovskiy, M.I. Katsnelson, Ann. Phys. 446, 169116 (2022)

O 30.10 Tue 12:45 GER 37

Hot electron driven charge carrier dynamics at the MoS_2/gold interface: a sum frequency generation study — •Tao Yang, Erik Pollmann, Stephan Sleziona, Peter Kratzer, Marika Schleberger, Richard Kramer Campen, and Yujin Tong — Fakultät für Physik, Universität Duisburg-Essen, 47057 Duisburg, Germany

Monolayer transition metal dichalcogenides (TMDCs) have potential applications as optoelectronics and (photo)electrocatalysts due to their fascinating electronic and optical properties. Applying these extraordinary properties to build devices and catalysts, it is often necessary to bring TMDCs into contact with a metallic surface. However, the charge transfer process across the semiconductor-metal interface is still not clear. While the quench of photoluminescence spectra has been reported for monolayer TMDCs on gold, which was understood to be a charge transfer of excited electrons from TMDC monolayer into the metal, it's interesting to ask what happens when the charge carrier is excited from the metal side. Here we use an azimuthal-dependent sum frequency generation combined with a pump-probe scheme to study the effect of the selectively excited hot electrons by 800 nm pump laser on the behavior of excitons of monolayer MoS_2. Our results show that the hot electrons from the gold substrate can significantly suppress the formation of excitons.

O 30.11 Tue 13:00 GER 37

Structure and electronic-optical properties of polymeric carbon nitride — •Changbin Im, Björn Kirchhoff, and Timo Jacob — Ulm University, Institute of Electrochemistry, Ulm, Germany

Understanding of the electronic structure and optical properties of polymeric carbon nitrides (PCNs) is of key interest in order to exploit their photo-physical and photo-electrocatalytic capabilities. The numerous defects of typical PCNs, however, present a limitation to computational studies that aim to improve our fundamental understanding of this material class. Here, we demonstrate a systematic thermodynamic study and structural characterization of various heptazine based-PCN models from the viewpoint of electronic and optical properties obtained with hybrid density functional theory (DFT). Our calculations reveal the key structural factors (degree of condensation, interlayer distance, corrugation) that govern the formation of various PCN motifs and their optical properties.[1] We discuss how each of the key factors influences the electronic structure of PCNs. We also suggest the concept of exciton binding energy in PCNs and discuss the polaronic interactions between heptazine units due to the localized exciton nature. Furthermore, we disclose a failure of GGA in the description of correlated electrons that underestimates the energy of the conjugated electrons comparing hybrid DFT. From the results, we could not only deduce a more complete model for the diverse PCN motifs but could additionally derive fundamental trends for future material optimization.

[1] Im, C. et al. (2022). http://arxiv.org/abs/2208.02582 submitted.