HL 21: 2D Materials II: Electronic Structure, Excitations, etc. (joint session O/CPP/HL)

Time: Monday 15:00–18:15 Location: WIL C107

HL 21.1 Mon 15:00 WIL C107

Unfolding and analysis of a defect band structure using doped $MoSe_2$ and MoS_2 — •Stefan Rost, Christoph Friedrich, Irene Aguilera, Beata Kardynal, and Stefan Blügel — Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany

We investigate substitutional doping of chromium and phosphorus in $MoSe_2$ and MoS_2 monolayers, as they are promising candidates for single photon emission. The systems are characterized by density-functional-theory (DFT) studies of structural relaxation, projected density-of-states, and band structure, all calculated with the Jülich FLAPW code family (www.flapw.de). Different sizes of super-cells are necessary to calculate the doped system depending on the strength of interaction between the dopants. The super-cells contain between 3×3 and 5×5 monolayer unit cells. Compared to the pristine material, the band structure of the defect system is backfolded in **K**-space, which complicates a straightforward interpretation, in particular for low doping concentrations. We have implemented a method for unfolding the bands obtained from the super-cell calculation. The resulting band structure resembles the one of the pristine material, but it contains additional information about the defect system, which, in this sense, can be regarded as a perturbed host system. – The authors gratefully acknowledge the computing time granted through JARA-HPC on the supercomputer JURECA at Forschungszentrum Jülich, (project cjpgi10) as well as the support through „Integration of Molecular Components in Functional Macroscopic System“ initiative of VW Stiftung.

HL 21.2 Mon 15:15 WIL C107

Geometry, electronic structure, and bonding of single-domain h-BN on Pt(110) — •Marco Thaler[1], Dominik Steiner[1], Alexander Menzel[1], Florian Mittendorfer[2], and Erminald Bertel[1] — [1]Physikalische Chemie, Universität Innsbruck, Österreich — [2]Institut für Angewandte Physik, TU Wien, Österreich

Recently we reported single-domain growth of hexagonal Boron Nitride (h-BN) on Pt(110)[1]. This is a peculiar system, where the substrate adapts to the h-BN adlayer by forming a (1xn) missing-row reconstruction (n = 5 or 6). The bandstructure was investigated by angle-resolved UV photoemission (ARPES) and is very similar to that of a free-standing h-BN monolayer except for the appearance of umklapp bands reflecting the periodicity of the Moiré pattern and testifying for the perfect film quality. Binding energies agree with other h-BN/transition metal systems if referenced to the vacuum level. Additionally, we studied the local density of states (LDOS) for differently positioned atoms in the h-BN film by DFT calculations. For N atoms on top of Pt atoms a small LDOS maximum appears at the Fermi level, indicating a weak covalent contribution to the h-BN-Pt bonding. This, the mean h-BN-Pt(110) distance, and the workfunction change place the present system at the borderline between purely dispersive and chemisorptive bonding[2]

[1] Steiner, D., Mittendorfer, F., Bertel, E. ACS Nano 13, 7083-7090(2019)

[2] Bokdam, M., Brocks, G., Kelly, P. J. Phys. Rev. B 90, 085415(2014)

HL 21.3 Mon 15:30 WIL C107

Photoinduced band renormalization in the nodal-line semimetal ZrSiSe — •Gianmarco Gatti[1], Alberto Crepaldi[1], Nicolas Tancogne-Dejean[2], Michele Puppin[3], Angel Rubio[2], Majed Chergui[3], and Marco Grioni[1] — [1]Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland — [2]Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany — [3]Laboratory of Ultrafast Spectroscopy, ISIC, Ecole Polytechnique Fédérale de Lausanne (EPFL), CH-1015 Lausanne, Switzerland

ZrSiSe is a layered material that is classified as a nodal-line semimetal. Its low-energy bandstructure is composed by linearly dispersing surface and bulk bands whose crossing points creates a one-dimensional contour in the momentum space with vanishing density of states close to the Fermi level. We investigate its equilibrium and out-of-equilibrium electronic structure by the means of angle- and time-resolved photoemission spectroscopy and compare the results with ab initio calculations. Our analysis indicates that the dispersion of the Dirac quasiparticle can be correctly reproduced by density functional theory with the inclusion local and non-local electronic correlations. Moreover, we observe a transient renormalization of the bands velocity above the Fermi level under the photoexcitation produced by ultrashort infrared pulses. We associate the microscopic origin of this effect to the increased screening of the non-local Coulomb interaction due to the presence of high-energy and non-thermalized carriers.

HL 21.4 Mon 15:45 WIL C107

Micro-focus ARPES on a twisted graphene/hBN field-effect device — •Alfred Jones[1], Ryan Muzzio[2], Davide Curcio[1], Deepnarayan Biswas[1], Jill A. Miwa[1], Philip Hofmann[1], Simranjeet Singh[2], Chris Jozwiak[3], Eli Rotenberg[3], Aaron Bostwick[3], Roland J. Koch[3], Søren Ulstrup[1], and Jyoti Katoch[2] — [1]Aarhus University, Aarhus, Denmark — [2]Carnegie Mellon University, Pittsburgh, Pennsylvania, USA — [3]Advanced Light Source, E. O. Lawrence Berkeley National Laboratory, Berkeley, California, USA

Graphene/ hBN heterostructures are an ideal testing ground for functional 2D devices owing to the atomically smooth surface and weak screening offered by hBN. On top of the record mobilities this structure offers, controlling the twist angle between layers creates a superlattice effect from which exotic electronic properties can occur.

Here, I present a study of the ARPES spectrum of graphene integrated in a device architecture with a hBN dielectric and graphite gate electrode. Micron-scale ARPES based on an X-ray capillary was used to collect the Dirac spectrum at different gate-voltages. A clear, reversible doping effect up to $5 * 10^{12}$ cm^{-2} is observed, providing access to the doping dependent quasiparticle dynamics in graphene on hBN.

Simultaneous measurements of this gate-dependent Dirac cone dispersion and the electrical resistance of the device enables extraction of electronic mobility and lifetimes. Our approach thereby demonstrates the tantalizing prospect of combining electron transport measurements with a spectroscopic probe of the electronic structure.

HL 21.5 Mon 16:00 WIL C107

Final-State Effects in Photoemission from Black Phosphorus — •Charlotte E. Sanders[1], Irene Aguilera[2], Klara Volkaert[3], Deepnarayan Biswas[3], Marco Bianchi[3], and Philip Hofmann[3] — [1]Central Laser Facility, STFC Rutherford Appleton Laboratory, Harwell OX11 0QX, UK — [2]Institute of Energy Research - Photovoltaic, Forschungszentrum Jülich, D-52425 Jülich, Germany — [3]Department of Physics and Astronomy, Aarhus University, 8000-C Aarhus, Denmark

Intrinsically doped bulk black phosphorus, although a van der Waals layered crystal, has nontrivial interlayer interactions and out-of-plane dispersing (k_z) electronic states, with a direct bandgap at the Z point of the three-dimensional (3D) Brillouin zone. The material's 3D character is related to key properties such as the thickness dependence of the bandgap in thin films and the tunability of the bandgap by strain and electric field. Interestingly, studies from angle-resolved photoemission spectroscopy (ARPES) of the k_z dispersion reveal intensity modulations near the Fermi level that are difficult to interpret in terms of the valence band dispersion predicted by theory. They have been attributed to surface-resonant states [1,2]. However, on the basis of density functional theory calculations and ARPES data acquired across a broad photon energy range, we suggest here an alternative interpretation based on final-state effects. The results call attention to the meaning of the free-electron-like final-state assumption and to the limits of its applicability. [1] *PRB* **90** (2014) 085101. [2] *PRB* **93** (2016) 075207.

Invited Talk HL 21.6 Mon 16:15 WIL C107

Atomic scale neural circuitry capable of self-adaptation — •Brian Kiraly — Radboud University, Nijmegen, The Netherlands

Driven by the rise of artificial intelligence and its potential for reduced energy consumption, there have been expanded efforts directed toward investigating materials which can perform pattern recognition directly in hardware. This requires a step away from physical systems which show simple bistability, toward complex, stochastic systems, which are inherently tunable. At the moment, however, the state of the art in

neuromorphic computing still struggles with fundamental issues, such as scaling or adaptability, often referred to as on-chip learning. In this talk, I will show a ground-breaking example, in which as few as 7 cobalt dopants on the surface of anisotropic semiconducting black phosphorus [1,2] can be utilized for pattern recognition. For this demonstration, we illustrate that controlled coupling between Co atoms [3] leads to a stochastic system which is well described using the concept of a Boltzmann machine. Both probing and reading the system with a scanning tunneling microscope, I will demonstrate how we realize an atomic scale synaptic memory and how the stochastic dynamics adapt and learn, depending on the input stimulus. The main aspects of this work have been carried out in the Scanning Probe Microscopy department at Radboud University. [1] Kiraly, Knol, Volckaert, Biswas, Rudenko, et. al., Phys. Rev. Lett. 123, 216403 (2019). [2] Kiraly, Hauptmann, Rudenko, Katsnelson, Khajetoorians, Nano Lett. 17, 3607 (2017). [3] Kiraly, Rudenko, Weerdenburg, Wegner, Katsnelson, Khajetoorians, Nature Commun. 9, 3904, (2018).

HL 21.7 Mon 16:45 WIL C107

Diffusion of magnetic dopants in pristine and defected phosphorene — •Rohit Babar[1] and Mukul Kabir[1,2] — [1]Department of Physics, Indian Institute of Science Education and Research, Pune, India — [2]Centre for Energy Science, Indian Institute of Science Education and Research, Pune, India

The incorporation of transition metal atoms is a robust way to imprint magnetism in non-magnetic 2D materials. In this regard, phosphorene has emerged as a versatile host for spintronic applications. Combining first-principles calculations with kinetic Monte Carlo simulations, we study the binding, magnetism, and diffusion of TM (TM = Cr, Fe, Co) atoms in pristine and defected phosphorene. The TM migration is highly anisotropic and favorable along the zigzag axis due to the puckered structure of phosphorene. While TM adsorption on pristine surface induces a local moment, the TM diffusion is uncontrolled at room temperature. We further show that vacancy defects exert an attractive potential beyond 1 nm and act as trapping centers for magnetic dopants. Our findings will assist in overcoming the practical limitations of surface decoration in phosphorene.

HL 21.8 Mon 17:00 WIL C107

Ab-initio structural dynamics of laser-excited graphene — •Sergej Krylow[1], Felipe Valencia Hernandez[2], Bernd Bauerhenne[1], and Martin E. Garcia[1] — [1]University of Kassel, 34132 Kassel, Germany — [2]National University of Colombia, 111321 Bogota, Colombia

We calculate the response of graphene to an ultrafast laser pulse using ab initio density functional molecular dynamics simulations. Our results show a biexponential decay of the Bragg peak intenities of the (100) and (110) peaks. We are able to show that the fast decay is caused by the equilibration between the electrons and a few strongly coupled optical phonons (SCOPs). The slower decay can be attributed to the equilibration of the SCOPs to the other phonon modes. Furthermore, we analyze the decay pathways from the SCOPs to the other phonon modes.

HL 21.9 Mon 17:15 WIL C107

Tailoring the opto-electronic response of graphene nanoflakes by size and shape optimization — •Raquel Esteban-Puyuelo[1], Rajat Sonkar[2], Bhalchandra Pujari[2], Oscar Gråsäs[1], and Biplab Sanyal[1] — [1]Division of Materials Theory, Department of Physics and Astronomy, Uppsala University, Box-516, SE 75120, Sweden — [2]Centre for Modeling and Simulation, Savitribai Phule Pune University, Ganeshkhind, Pune 411007, India

The long spin-diffusion length, spin-lifetime and excellent optical absorption coefficient of graphene provide a promising platform for building opto-electronic devices as well as spin-based logic in the nanometer regime. We have used time-dependent density functional theory to study how the magnetic structure and optical properties of graphene nanoflakes depend on their size and shape. We optimize the magnetic ground state and the exchange coupling between the edges of the flakes to tailor the external fields needed to switch the magnetic ordering. Finally it's shown that the magnetic state alters the optical response of the flake leading to the possibility of opto-spintronic applications.

HL 21.10 Mon 17:30 WIL C107

Electronic and optical properties of two-dimensional magnets (CrI_3) and their effects on adjacent material (WSe_2/CrI_3) — •Marie-Christin Heissenbüttel, Michael Rohlfing, and Peter Krüger — Institut für Festkörpertheorie WWU, Münster, Deutschland

For the development of novel opto-electronic devices the access and manipulation of the spin degree of freedom is of fundamental importance. The recently discovered 2D magnets provide a great opportunity to study the delicate interplay of spin, orbital, charge and lattice degree of freedom and the manipulation of other non-magnetic 2D materials, as for example TMDCs, by proximity effects. In this talk I will demonstrate our results from ab-initio calculations for the magnetic monolayer of chromium triiodide (CrI_3). The electronic and optical properties are analyzed as well as the effects of magnetism on the optics of WSe_2 within the heterobilayer tungsten diselenide (WSe_2) on CrI_3 are shown. The investigation of CrI_3 by many body perturbation theory (MBPT) within the LDA+GdW approximation reveals a ferromagnetic and insulating behavior and the optical absorption obtained from the solution of the BSE shows large exciton binding energies up to 1.04 eV. Furthermore due to the impact of ferromagnetic proximity effects on the TMDC WSe_2 in the heterosystem WSe_2 / CrI_3 a lifting of the energy degeneracy in the K-valleys of WSe_2 is found. Both the electronic band gap at $\pm$K as well as the corresponding excitons in WSe_2 are affected.

HL 21.11 Mon 17:45 WIL C107

Engineering intrinsic π-magnetism in nanographenes — •Shantanu Mishra[1], Doreen Beyer[2], Kristjan Eimre[1], Shawulienu Kezilebieke[3], Reinhard Berger[2], Oliver Gröning[1], Peter Liljeroth[3], Carlo Pignedoli[1], Xinliang Feng[2], Pascal Ruffieux[1], and Roman Fasel[1] — [1]Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland — [2]Technical University of Dresden, Dresden, Germany — [3]Aalto University, Espoo, Finland

Among the multitude of properties realized in organic compounds due to the chemical versatility of carbon, magnetism remains one of the most challenging. The electronic structure of polycyclic aromatic hydrocarbons (nanographenes) depends critically on the topology of the underlying π-electron network, which provides a tunable platform to realize all-carbon magnetism at the nanoscale.

Combining rational design principles with on-surface synthesis, we engineer and probe emergent magnetism in elusive magnetic nanographenes, namely, Clar's goblet [1] and extended triangulenes [2], and their covalently-bonded assemblies. Our experimental approach follows low-temperature scanning tunneling microscopy and inelastic electron tunneling spectroscopy, with further insights provided by mean-field and many-body perturbation theory calculations.

[1] S. Mishra et al., Nature Nanotech. (in press)
[2] S. Mishra et al., J. Am. Chem. Soc. 141, 10621 (2019)

HL 21.12 Mon 18:00 WIL C107

Theoretical study on the magnetic structure of few-layer TMPS3 — •Tae Yun Kim[1,2,3] and Cheol-Hwan Park[1,2,3] — [1]Department of Physics and Astronomy, Seoul National University, Seoul 08826, Korea — [2]Gwanak-Gu Hoam-Ro 519 101-1205 — [3]Center for Theoretical Physics, Seoul National University, Seoul 08826, Korea

Transition metal phosphorus sulfides (TMPS3) are a family of the layered magnetic materials. Due to the layered structure and the rich spectrum of the magnetic structure [1], TMPS3 has been thought to be a good playground for testing two-dimensional magnetism in real world experiments [2]. It was found that the antiferromagnetic order in FePS3 remains down to the monolayer limit [3, 4], which was soon followed by the discovery of two-dimensional ferromagnetism in CrI3 [5] and Cr2Ge2Te6 [6]. More recently, the existence of the magnetic ordering in few-layer TMPS3 has been investigated in experiments [7, 8]. In this contribution, we present an in-depth investigation of the magnetic structure of few-layer TMPS3 based on the results of our first-principles calculations.

[1] R. Brec, Solid State Ionics 22, 3 (1986).
[2] K. S. Burch et al., Nature 563, 47-52 (2018).
[3] X. Wang et al., 2D Materials 3, 031009 (2016).
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[5] B. Huang et al., Nature 546, 270-273 (2017).
[6] C. Gong et al., Nature 546, 265-269 (2017).
[7] K. Kim et al., Nature Communications 10, 345 (2019).
[8] K. Kim et al., 2D Materials 6, 041001 (2019).