HL 51: 2D Materials: Stacking and heterostructures (joint session O/HL/TT)

Time: Friday 9:30–12:30 Location: HSZ/0401

HL 51.1 Fri 9:30 HSZ/0401

A new fabrication method for metal intercalated epitaxial graphene devices — •Marc Bothe[1], Stefan Wundrack[1,2], Marcelo Jaime[1], Klaus Pierz[1], Frank Hohls[1], Rainer Stosch[1], Hans Werner Schumacher[1], Andrey Bakin[2], and Teresa Tschirner[1] — [1]Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany — [2]Institut für Halbleitertechnik, TU Braunschweig, Hans-Sommer-Str. 66, 38106 Braunschweig, Germany

Epitaxial graphene grown on silicon carbide is a promising platform to achieve metal intercalation. It enables the creation of two-dimensional metal layers that are encapsulated and protected by graphene. However, the use of metal intercalated graphene samples for lithographic device fabrication presents two critical challenges. First, solvents used in the lithography process lead to the deintercalation of the metal atoms. Second, the presence of lattice defects in the graphene - necessary for the intercalation process - compromises the structural and electronic integrity of the device. We present a novel fabrication method that solves these problems. In our approach, the graphene devices are first pre-structured using lithography and subsequently intercalated. This is made possible by a spatial separation on the sample between the device structures and the intercalation origin and by intercalation channels that can guide the intercalation front reliably to the devices. We demonstrate this method on gallium intercalated epitaxial graphene Hall bars that exhibit superconducting behaviour.

HL 51.2 Fri 9:45 HSZ/0401

Anisotropic Strain Observation in Naturally Occurring Buckling on Twisted Bilayer Graphene: A Nano-Raman Study — •Gustavo Soares[1], Rafael R. Barreto[1], Rafael Nadas[1], Kenji Watanabe[2], Takashi Taniguchi[2], Leonardo C. Campos[1], Luiz G. Cancado[1], and Angelo Malachias[1] — [1]Physics Department, Federal University of Minas Gerais, Belo Horizonte, Minas Gerais, 31270-901, Brazil — [2]National Institute for Materials Science, Tsukuba, Ibaraki, 3050047, Japan

This work investigates naturally occurring buckling and its relation with anisotropic strain relaxation in twisted bilayer graphene (tBG). In the tBG system a twist angle is imposed to influence graphene structural and electronic properties. Such condition directly implies in the occurrence of biaxial in-plane strain, with usual observation of buckled/wrinkled localized regions where the tBG morphology is no longer planar. Using tip-enhanced Raman spectroscopy, we conducted high-resolution mapping to analyze variations in Raman bands associated with twist angle variation and strain effects. Our findings reveal that localized strain gradients, modulated by twist angle variations, induce deterministic buckling in graphene layers, evidencing distinct uniaxial and biaxial strain regions. Finite element modeling further supports these observations, showing that buckling can store elastic energy sufficient to overcome usual tBG-substrate adhesion forces.

HL 51.3 Fri 10:00 HSZ/0401

Kirkendall Voids in Monolayer $Mo_xTa_yS_2$ Alloys on Au(111) — Kai Mehlich[1], Thais Chagas[1], Francis H. Davies[2,3], Alessia Bardazzi[1], Catherine Grover[1], Arkady V. Krasheninnikov[3], and •Carsten Busse[1] — [1]Department Physik, Universität Siegen, Walter-Flex-Straße 3, 57072 Siegen, Germany — [2]Ion Beam Physics and Materials Research, Helmholtz-Zentrum Dresden-Rossendorf, Bautzner Landstraße 400, 01328 Dresden, Germany — [3]Department of Physics and Astronomy, University of Exeter, Stocker Road, Exeter, EX4 4QL United Kingdom

During the growth of monolayer MoS_2-TaS_2 heterostructures, we observe the Kirkendall effect: Diffusing vacancies agglomerate in MoS_2, forming what are commonly termed Kirkendall voids - here manifesting as holes in the 2D layer. This phenomenon has not been previously reported in systems with reduced dimensionality.

We prepare the lateral heterostructures by reactive molecular beam epitaxy on chemically inert and weakly interacting Au(111). First, compact MoS_2 cores are grown. In a second step, TaS_2 is added in a sulfur-rich environment at elevated temperatures which promotes diffusion at the interface. The resulting heterostructures exhibit characteristic 2D Kirkendall holes surrounded by a $Mo_xTa_yS_2$ alloy region. These findings reveal defect-mediated processes in low-dimensional systems and open new avenues for designing 2D lateral heterostructures with intricate morphologies.

HL 51.4 Fri 10:15 HSZ/0401

Designing 2D Non-van der Waals Heterostructures — •Anastasiia Nihei[1,2], Tom Barnowsky[1,2], and Rico Friedrich[1,2] — [1]TU Dresden — [2]Helmholtz-Zentrum Dresden-Rossendorf

Heterostructure interfaces created by stacking two-dimensional (2D) materials offer a pathway to realizing advanced electronic and magnetic functionalities at the nanoscale. In this work, we present a high-throughput screening of non-van der Waals (non-vdW) 2D heterostructures; including sandwich-like stacking. Non-van der Waals 2D materials can be obtained by both top-down exfoliation as well as bottom-up growth approaches of non-layered bulk crystals [1]. Our approach uses the AFLOW-Hetbuilder - a newly developed tool for stacking 2D systems based on the coincidence lattice algorithm [2-4].

We investigate interfacial binding effects across a wide range of heterobilayers [5] and sandwich-like systems, analyzing their structural, electronic, and magnetic characteristics such as hybrid interface bands and robust magnetic coupling. Furthermore, we compare the binding energetics of non-vdW and conventional vdW heterostructures. The tunable interfacial properties of non-vdW 2D heterostructures provide a versatile platform for advanced functionalities with potential applications in electronics, spintronics, and the energy sector [5].

[1] R. Friedrich *et al.*, Nano Lett. **22**, 989, (2022).
[2] D.S. Koda *et al.*, J. Phys. Chem. C **120**, 10895, (2016).
[3] https://zenodo.org/record/4721346 .
[4] S. Divilov *et al.*, High Entropy Alloys Mater. **3**, 178 (2025).
[5] A. Nihei, *et al.*, arXiv DOI: 10.48550/arXiv.2503.12209 (2025).

HL 51.5 Fri 10:30 HSZ/0401

Second-Order Nonlinear Imaging for Probing 2D van der Waals Structures — •Tao Yang[1], Ben John[1], Kyoung P. Lee[2], Nasim Mirzajani[1], Martin Wolf[1], Xiaoqin Li[2], Martin Thaemer[1], Alexandar Paarmann[1], Niclas S. Mueller[3], and Alexander P. Fellows[1] — [1]Fritz Haber Institute, Berlin, Germany — [2]University of Texas at Austin, Austin, USA — [3]Freie Universitaet Berlin, Berlin, Germany

Twisted and stacked multi-layer architectures offer new opportunities for tailoring the electronic and optical properties of two-dimensional (2D) van der Waals materials. Reliable determination of crystal structure, stacking sequence, and twist angle is therefore crucial. Second-order nonlinear optical microscopy, including second-harmonic and sum-frequency generation (SHG and SFG, respectively), provides high sensitivity to crystal symmetry and orientation in non-centrosymmetric lattices. Recently, we employed heterodyne-detected vibrational SFG microscopy to probe the local structure of hexagonal Boron Nitride (hBN) monolayers with sub-micron resolution. By employing our developed azimuthal-scanning approach, we fully resolved the crystallographic structure and edge termination in monolayer flakes. Here, we extend these measurements to multi-layer hBN structures, using the same azimuthal-scanning approach in a newly developed SHG microscope to gain insight into their different stacking configurations. Our results highlight second-order nonlinear microscopy as a powerful tool for quantitative structural analysis in 2D materials and for future studies of moiré and twisted heterostructures.

HL 51.6 Fri 10:45 HSZ/0401

Moiré-Driven Electronic Modulations in Weakly Coupled h-BN/Graphite — •Fábio J. R. Costa[1,2,7], Daniel Arribas[2], Thiago G. L. Brito[2], Tin S. Cheng[3], Jonathan Bradford[3], Amelia Thompson[3], Alex Saywell[3], Christopher J. Mellor[3], Peter H. Beton[3], Sergey V. Novikov[3], Juliette Plo[4], Bernard Gil[4], Guillaume Cassabois[4,5], Luiz F. Zagonel[1], Klaus Kuhnke[2], Klaus Kern[2,6], and Anna Rosławska[2] — [1]University of Campinas, Brazil — [2]Max-Planck-Institut für Festkörperforschung, Germany — [3]University of Nottingham, UK — [4]Laboratoire Charles Coulomb, France — [5]Institut Universitaire de France, France — [6]EPFL, Lausanne, Switzerland — [7]Current address: Université de Strasbourg, CNRS, IPCMS, Strasbourg, France

Van der Waals materials and their heterostructures offer exciting opportunities for next-generation nanophotonic and electronic technolo-

gies. Their electronic and optical properties can be modulated by moiré superlattices that emerge when mismatched layers are stacked together. Hexagonal boron nitride (h-BN) is a key platform in this context, yet the influence of moiré patterns on weakly interacting h-BN interfaces remains underexplored. Here[1], we use scanning tunneling microscopy to resolve large moiré superlattices in h-BN/graphite and uncover pronounced nanoscale modulations of the electronic landscape, despite their weak interlayer interactions. These findings position moiré engineering in h-BN as a powerful tool to tailor local functionalities in van der Waals heterostructures. Ref.: 1. Fábio J. R. Costa *et al.* ACS Nano 2025 19 (40), 35528-35538

HL 51.7 Fri 11:00 HSZ/0401

μ-ARPES study on the fine electronic structure of misfit layer compound (PbSe)1.16(TiSe2)2 — •Haruki Muramatsu[1], Natsuki Mitsuishi[2,3], Teppei Ueno[4], Kenichi Ozawa[5], Kaya Kobayashi[4,6], and Kyoko Ishizaka[1,2] — [1]Dept. of Appl. Phys. & QPEC, Univ. of Tokyo — [2]CEMS, RIKEN — [3]Grad. Sch. Sci., Nagoya Univ. — [4]Dept. of Physics, Okayama Univ. — [5]KEK-PF — [6]RIES, Hokkaido Univ.

Misfit layered compounds have been attracting significant attention due to their lattice mismatches and resultant two-dimensional electronic structures reminiscent of van der Waals heterostructures. One such compound, (PbSe)1.16(TiSe2)2, consists of alternating stacking of PbSe monolayers (NaCl-type, four-fold symmetry) and TiSe2 bilayers (CdI2-type, three-fold symmetry). To elucidate its electronic structure, we performed μ-ARPES measurements by carefully distinguishing the cleavage surface terminations. In the presentation, we will discuss the novel electronic states reflecting the natural incommensurate heterostructure as well as charge density wave in the buried TiSe2 bilayer.

HL 51.8 Fri 11:15 HSZ/0401

Twisted $NbSe_2$ heterostructures — •Alexander Bäder[1,2], Clara Pfister[3,4], Tobias Wichmann[1,5], Laura Pätzold[3,4], Tim O. Wehling[3,4], and Felix Lüpke[1,2] — [1]Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich — [2]II. Physikalisches Institut, Universität zu Köln — [3]I. Institute of Theoretical Physics, U Hamburg — [4]The Hamburg Centre for Ultrafast Imaging — [5]Institut für Experimentalphysik IV A, RWTH Aachen

We fabricated monolayers (MLs) of the van der Waals material $NbSe_2$ that are rotated with respect to the underlying bulk $NbSe_2$ using our developed ultra-high vacuum (UHV) stacking technique. These heterostructures were characterized through the use of scanning tunneling microscopy and spectroscopy at a base temperature of 1.4 K. The MLs realize a variety of twist angles relative to the underlying bulk lattice, with the lowest observed twist angle being 10°. The MLs display the coexistence of charge density waves (CDW) and moiré effects: In the absence of strain, the MLs develop a 3×3 CDW, however the application of strain through interfacial disorder results in a 2×2 CDW state, supported by theoretical calculations. Compared to isolated MLs the superconducting order parameter is enhanced due to the proximity effect from the underlying bulk.

HL 51.9 Fri 11:30 HSZ/0401

band structure and charge density wave in a natural van der Waals heterostructure 4Hb-TaSe2 — •Fumihiko Kimura[1], Tomoki Maeda[2], Natsuki Mitsuishi[3,4], Kazuki Okada[2], Takuya Nomoto[5], Kenichi Ozawa[6], Masahiro Naritsuka[3], Tetsuo Hanaguri[3], Shunsuke Kitou[7], Yuiga Nakamura[8], Taka-hisa Arima[3,7], Takao Sasagawa[2], and Kyoko Ishizaka[1,3] — [1]Quantum-Phase Electronics Center & Department of Applied Physics, The University of Tokyo — [2]Laboratory for Materials and Structures, Science Tokyo — [3]RIKEN CEMS — [4]Graduate School of Science, Nagoya University — [5]Department of Physics, Tokyo Metropolitan University — [6]Institute of Materials Structure Science, High energy Accelerator Research Organization (KEK) — [7]Department of Advanced Materials Science, The University of Tokyo — [8]Japan Synchrotron Radiation Research Institute (JASRI), SPring-8

4Hb-TaSe2 is a natural van der Waals heterostructure charge density wave (CDW) material consisting of alternative stacking of monolayer 1T-TaSe2 (Mott insulator with Star-of-David CDW) and monolayer 2H-TaSe2 (superconductor with 3 by 3 CDW). Although transport measurements have suggested the existence of multiple CDWs, the details of the CDW and electronic structures remain unclear. In this study, we directly observed the electronic structure of 4Hb-TaSe2 by micro-focused angle-resolved photoemission spectroscopy with careful selection of the surface terminations. We discuss the temperature dependent electronic structure with comparison to the CDW structures obtained by scanning tunneling microscopy and X-ray diffraction.

HL 51.10 Fri 11:45 HSZ/0401

Coexisting charge density waves in twisted NbSe2 bilayers — •Christopher Tat Shun Cheung[1], Zachary A. H. Goodwin[2], Yixuan Han[3], Jiong Lu[3], Arash A. Mostofi[1], and Johannes Lischner[1] — [1]Departments of Physics and Materials and the Thomas Young Center for Theory and Simulation of Materials, Imperial College London, London SW7 2AZ, U.K. — [2]Institute for Functional Intelligent Materials, National University of Singapore, Singapore 117544, Singapore — [3]Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, United Kingdom

Twisted bilayers of semiconducting transition metal dichalcogenide (TMD) monolayers have been studied extensively. In contrast, twisted bilayers composed of metallic monolayers, such as NbSe2, remain less understood.

Monolayer NbSe2 can host different types of charge density waves (CDWs), in which the Nb atoms move away from their high-symmetry positions. In twisted bilayer NbSe2, identifying CDWs in relaxed structures is challenging because atomic relaxations occur both because of CDW formation and also because of the moiré pattern.

We have carried out large-scale first-principles calculations using density functional theory to study the moiré relaxations and CDWs in twisted bilayer NbSe2. We have developed methods for revealing the CDWs, and for locally classifying the type of CDW. We find that different types of CDWs coexist in the moiré unit cell due to the interactions with strain induced by moiré relaxations [1].

[1] Cheung et al, Nano Lett. 2024, 24, 12088-12094.

HL 51.11 Fri 12:00 HSZ/0401

MicroARPES studies of contact doping of monolayer transition metal dichalcogenides by RuCl3 — •Thomas Nielsen[1], Edvard Solbrekken[1], Alfred J. H. Jones[1], Zhihao Zhiang[1], Chakradar Sahoo[1], Kenji Watanabe[2], Takashi Taniguchi[2], Jill A. Miwa[1], Søren Ulstrup[1], Christian Overby[1], and Christian V-B. Fokdal[1] — [1]Aarhus University, Denmark — [2]National Institute for Materials Science, Japan

Placing van der Waals materials into contact with α-$RuCl_3$ has recently emerged as a method of modulating their electronic structures. The proximity to α-$RuCl_3$ has been observed to produce strong hole doping in the van der Waals material, and this has been established as a method to create better electrical contacts in transistor devices based on monolayer transition metal dichalcogenides (TMDs). Here, we use the microARPES endstation at the ASTRID2 synchrotron light source at Aarhus University in Denmark to study the valence bands and core levels of semiconducting monolayer TMDs in proximity to α-$RuCl_3$. We observe a large valence-band shift of $0.7-0.8$eV indicating strong hole doping. α-$RuCl_3$ is highly sensitive to the temperatures and chemicals typically used in the dry-transfer fabrication procedures of van der Waals heterostructures. How the degradation of α-$RuCl_3$ affects the proximity-induced doping of the TMD is discussed based on the ARPES measurements.

HL 51.12 Fri 12:15 HSZ/0401

STEM Investigation of Entropy Forbidden Ordering in CVD Grown WSe_2-$MoSe_2$ Alloys — •Max Bergmann[1], Matvei Kislitsyn[1], Julian Picker[2], Jürgen Belz[1], Robin Günkel[1], Badrosadat Ojaghi Dogahe[1], Shamail Ahmed[1], Andrey Turchanin[2], and Kerstin Volz[1] — [1]mar.quest | Marburg Center for Quantum Materials and Sustainable Technologies, Philipps-Universität Marburg, 35032 Marburg, Germany — [2]Faculty of Chemistry and Earth Sciences, Friedrich-Schiller-Universität, 07743 Jena, Germany

2D transition metal dichalcogenides have gained significant interest due to their optoelectronic properties, which can be tailored by structural variation. However, controllable production, namely growth, of such tailored structures still remains a key challenge towards large-scale production. In this study, we show lateral heterostructures of 2D $MoSe_2$ and WSe_2, grown on a SiO_2 TEM grid by chemical vapor deposition, that at the interface show a highly ordered structure of W and Mo atoms at the TMD positions, as observed by scanning transmission electron microscopy. This is in contrast to *ab initio* calculations, which ascribe unordered alloys as the preferred configuration, since entropy is the main driving force compared to formation enthalpy. We link this unexpected phenomenon to the initial nucleation of the material on a

clean $MoSe_2$ crystal edge, present before the alloy growth, together with special precursor chemistry. Furthermore, we show *ab initio* results in conjunction with the special quasirandom structure method on the bandstructure and optical properties of this structure.