## 2D Materials Technical Group
## Room 303 - Session 2D+AS+EM-ThA

## 2D Materials: Electron Microscopy and Photoemission Spectroscopy

**Moderators: Keun Su Kim**, Yonsei University, Republic of Korea, **Dmitry Kireev**, University of Texas at Austin

2:20pm **2D+AS+EM-ThA-1 Periodic Lattice Displacements in Low Dimensional Materials*, Robert Hovden,*** University of Michigan **INVITED**

Dramatic electronic changes are associated with periodic lattice displacements [1] where structure, even in 2D materials, requires higher dimensional measurement using scanning / transmission electron microscopy. In particular charge density waves are an emergent periodic modulation of the electron density that permeates a crystal with strong electron-lattice coupling. Strong evidence suggests that transformative correlated electron behavior may exist only in unrealized clean 2D materials such as 1T-$TaS_2$. Unfortunately, experiment and theory suggest that extrinsic disorder in free standing 2D layers impedes correlation-driven quantum behavior. Here we demonstrate a new route to realizing fragile 2D quantum states through epitaxial polytype engineering of van der Waals materials. The isolation of truly 2D charge density waves (CDWs) between metallic layers stabilizes commensurate long-range order and lifts the coupling between neighboring CDW layers to restore mirror symmetries via interlayer CDW twinning. The twinned-commensurate (tC-) CDW reported herein has a single metal–insulator phase transition at ~350 K as measured structurally and electronically [2]. Fast in-situ transmission electron microscopy and scanned nanobeam diffraction map the formation of tC-CDWs. This work introduces epitaxial polytype engineering of van der Waals materials to access latent 2D ground states distinct from conventional 2D fabrication.

Here we show the critical temperature for spatially-coherent, commensurate (C-) CDW in 1T-$TaS_2$ can be raised to well above room temperature (~150 K above the expected transition) by synthesizing clean interleaved 2D polytypic heterostructures. This stabilizes a collective insulating ground state (i.e. C-CDW) not expected to exist at room temperature. We show the formation of these spatially coherent states occurs when 2D CDWs are confined between metallic prismatic polytypes. At the same time, interleaving disables interlayer coupling between CDWs. This raises the critical temperature of the C-CDW and forms out-of-plane twinned commensurate (tC) CDWs as revealed by scanned nanobeam electron diffraction. These results demonstrate polytype engineering as a route to isolating 2D collective quantum states in a well-defined extrinsic environment with identical chemistry but distinct band structure.

[1] *Nature and evolution of incommensurate charge order in manganites visualized with cryogenic STEM,* I. El Baggari et al. **Proc. Natl. Acad. Sci. U.S.A. 115**, 1445 (2018)

[2] *Two-dimensional charge order stabilized in clean polytype heterostructures*, S. H. Sung et al. **Nature Communications**, **13** 413 (2022)

3:00pm **2D+AS+EM-ThA-3 Engineering of Nanoscale Heterogenous Transition Metal Dichalcogenide-Au Interfaces*, Alex Boehm,*** Sandia National Laboratories*; J. Fonseca,* Naval Research Laboratory*; K. Thuermer, J. Sugar,* Sandia National Laboratories*; J. Robinson,* Naval Research Laboratory*; T. Ohta,* Sandia National Laboratories

2-D transition metal dichalcogenides (TMDs) have recently garnered much attention owing to their extraordinary physical, chemical, electrical, and optical properties. However, early material and device studies have revealed that these properties can be greatly impacted by extrinsic factors such as substrate interactions, mechanical strain, and charge transfer. Thus, a careful understanding of the nuanced interactions between TMDs and other materials is critical for high performance devices. Of particular importance are the interfaces with metallic contacts. Here, one barrier are the spatial nonuniformities recently reported at these types of interfaces. Uncovering the impact of these heterogeneities on TMD properties and establishing strategies to control TMD-metal interfaces could enable an array of engineering pathways for future applications. In this work, we find that the electronic structures of mechanically exfoliated TMD-Au interfaces exhibit pronounced heterogeneity arising from microstructure of the supporting metal. Pertinent for device applications these electronic structure variations indicate fluctuating doping levels and Schottky barrier height across the junction. We examined the electronic structures of WS2 and WSe2 at high spatial resolution via photoemission electron microscopy (PEEM) revealing key differences in work function and binding energies of the occupied states. Furthermore, the inherent role of the underlying Au microstructure on the TMD electronic structure is revealed by electron backscatter diffraction (EBSD) and scanning tunneling microscopy (STM). Finally, simple processing methods are employed to fabricate homogenous TMD-Au interfaces while also tuning the electronic properties of the TMDs. Our findings illustrate that the electronic properties of TMDs are greatly impacted by metal interface interaction and provide a means to engineer these important junctions.

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3:20pm **2D+AS+EM-ThA-4 Advanced Laboratory-Based Momentum Microscopy and PEEM Analysis*, Stefan Böttcher Böttcher,*** SPECS Surface Nano Analysis GmbH, Germany*; D. Singh, T. Conard,* IMEC, Belgium*; M. Wietstruck,* SPECS Surface Nano Analysis GmbH, Germany*; P. van der Heide,* IMEC, Belgium*; A. Thissen,* SPECS Surface Nano Analysis GmbH, Belgium

Momentum Microscopy is a new technology for comprehensive surface analysis, providing high energy and angular resolved band structure mapping combined with advanced surface imaging capability. Extending this technology with laboratory-based instrumentation opens the possibility for detailed studies of new materials under well controlled environments. The combination of a PEEM lens for surface microscopy and momentum microscopy, allows for small spot analysis in ARPES and chemical sensitive surface mapping. In a joint project between IMEC and SPECS the possibilities for laboratory-based momentum microscopy, laser ARPES and x-ray spectroscopy and microscopy are evaluated in the framework of semi-industrial environment. We present a status report at the intersection between fundamental and applied research in surface science. We focus on the use of ARPES characterization in novel materials close to applied research and the functionality of x-ray analysis in PEEM and spectroscopy for chemical analysis.

3:40pm **2D+AS+EM-ThA-5 Epitaxial Growth and Electronic States of Ultrathin Bi (111) Films on Insb (111)B: Evidence of Inversion Symmetry Breaking via Film-Substrate Interactions*, Hadass S. Inbar,*** *J. Dong, A. Engel, C. Dempsey, Y. Chang,* University of California Santa Barbara*; A. Fedorov,* Advanced Light Source, Lawrence Berkeley National Laboratory*; C. Palmstrom,* University of California Santa Barbara

Quantum size effects in bismuth films have been the focus of the scientific community for decades. The spin–split Rashba surface states and large mass anisotropy in surface state valleys have made Bi films a promising system for future applications in spintronics and valleytronics. Moreover, in the field of topological materials, the Bi (111) bilayer (BL) is predicted to behave as a quantum Hall spin insulator[1]. Along the Bi (111) step edges, 1D helical modes were observed[2], an ingredient in one proposed platform to construct Majorana zero modes[3]. However, the synthesis of continuous ultrathin (<6 BL) Bi (111) epitaxial films on semiconducting substrates has remained a materials challenge. We report a study of ultrathin large-area Bi (111) layers grown on InSb (111)B substrates by molecular beam epitaxy and in-vacuo transferred for scanning tunneling microscopy and synchrotron-based angle-resolved photoemission spectroscopy. We show that large-area single-domain ultrathin Bi films can be stabilized through strong film-substrate interactions. Our study follows the evolution of tensile strain in the films, which is predicted to lead to a semimetallic to semiconducting transition. With decreasing film thickness from 13 to 1 BL, we quantify the confinement-induced shifts in the bulk band structure and trace the quantum well energy levels with a phase shift accumulation model. Significant substrate-film interactions breaking inversion symmetry affect the surface state dispersion leading to a surface state degeneracy which allows us to assign the topological order in Bi(111) thin films. The findings of this study offer a new route for epitaxial growth and integration of band-engineered Bi films with III-V substrates.

[1] Murakami, S. (2006). Quantum spin Hall effect and enhanced magnetic response by spin-orbit coupling. Physical Review Letters, 97(23), 236805.

[2] Drozdov, I. K., Alexandradinata, A., Jeon, S., Nadj-Perge, S., Ji, H., Cava, R. J., ... & Yazdani, A. (2014). One-dimensional topological edge states of bismuth bilayers. Nature Physics, 10(9), 664-669.

[3] Jäck, B., Xie, Y., Li, J., Jeon, S., Bernevig, B. A., & Yazdani, A. (2019). Observation of a Majorana zero mode in a topologically protected edge channel. Science, 364(6447), 1255-1259.

4:00pm **2D+AS+EM-ThA-6 Band Modulations: Revealing Moiré Effects in Twisted Bilayer 2D Materials**, ***Ryan Muzzio***, Carnegie Mellon University*; A. Jones, P. Majchrzak,* Aarhus University, Denmark*; H. Martins, S. Singh,* Carnegie Mellon University*; C. Jozwiak, A. Bostwick, E. Rotenberg,* Lawrence Berkeley National Laboratory*; P. Hofmann,* Aarhus University, Denmark*; S. Ulstrup,* aarhus University, Denmark*; J. Katoch,* Carnegie Mellon University

Two dimensional (2D) materials are a wonderful template to explore novel quantum phenomena in the ultra thin limit.They can be exfoliated to the desired thickness, stacked with other 2D flakes, and be integrated in device fabrication for electrical measurement.The addition of a twist angle between stacked 2D flakes produces a moiré lattice which can lead to drastic changes in their physical properties.For the case of bilayer graphene, introducing a ~1.1 degree rotation (the magic angle) leads to a low temperature superconducting state[1].This remarkable transport result has been explained via band structure theory and experiment[2,3,4] of the hybridization of the out-of-plane Pi orbitals of the graphene layers which forma weakly dispersing state at the fermi level.Beyond graphene, twist-angle dependent bilayer transition metal dichalcogenides (TMDCs) also display extraordinary novel moiré physics[5,6].In this presentation, we will discuss our ongoing analysis of nano- and micro-focused angle resolved photoemission spectroscopy (ARPES) performed on twisted bilayer graphene and TMDCs systems placed on hBN.We demonstrate, over a wide range of twist angles, the effect of the moiré lattice and proximity effects on the band structure by investigating the effective masses, band positionings, and location of the moiré bands across four TMDC heterobilayers.Our work demonstrates the tunability of the electronic properties in twisted 2D bilayers and the power of ARPES to provide a momentum-resolved view of their electronic structure.

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