2D Materials
Room 304 - Session 2D-WeA
Surface-Sensitive Techniques for 2D Materials Characterization

Moderators: Qiong Ma, Boston College, **Dr. Chris Jozwiak**, Advanced Light Source, Lawrence Berkeley National Laboratory

2:15pm **2D-WeA-1 Exploring and Manipulating Charge Density Textures in Tas2 with NanoARPES***, **Chris Jozwiak**,* Advanced Light Source, Lawrence Berkeley National Laboratory **INVITED**

The ability to directly probe electronic structure in energy-momentum space with surface sensitivity makes ARPES a critical spectroscopic tool for studying 2D materials. Adding spatial resolution capabilities to its arsenal greatly expands the range of material systems and phenomena that can be impacted by nanoARPES. I will give an overview of this technique at the MAESTRO beam line at the Advanced Light Source, where we aim to maximize impact across the 10 um to 100 nm scales by enabling various *in operando* measurements. I will present measurements of 1T-TaS_2, a prototypical quasi-2D transition metal dichalcogenide that hosts a series of charge density wave phases and associated electronic phenomena. Although a classic target of ARPES experiments for decades, this scale of spatial resolution enables insights into the electronic textures in real- and momentum-space that form in this system. I will discuss these, with a particular focus on measurements of the metastable “hidden” state that can be created with electrical currents and ultrafast optical pluses.

2:45pm **2D-WeA-3 Visualization of Tunable Electronic Structure of Monolayer $TaIrTe_4$*, **Sandy Adhitia Ekahana**,* *Aalok Tiwari,* Carnegie Mellon University*; Souvik Sasmal,* Argonne National Laboratory*; Zefeng Cai, I-Hsuan Kao, Ravi Kumar Bandapelli,* Carnegie Mellon University*; Jian Tang,* Boston College*; Chenbo Min,* Carnegie Mellon University*; Tiema Qian,* UCLA*; Kenji Watanabe, Takashi Taniguchi,* NIMS (National Institute for Materials Science), Japan*; Ni Ni,* UCLA*; Qiong Ma,* Boston College*; Chris Jozwiak, Eli Rotenberg, Aaron Bostwick,* ALS-LBNL*; Simranjeet Singh, Noa Marom, Jyoti Katoch,* Carnegie Mellon University

The advent of *in operando* angle-resolved photoemission spectroscopy (ARPES) has unlocked new capabilities for dynamically tuning and visualizing electronic band structures. The success of this progress has been largely coupled by the recent advancements in the van der Waals heterostructure device stacking. Among the diverse transition metal dichalcogenide (TMDC) family, monolayer $TaIrTe_4$ is of particular interest due to previous transport measurements suggesting it hosts a dual quantum spin Hall state [1]. Here, we present *in operando* ARPES measurements on monolayer $TaIrTe_4$ devices, revealing an asymmetric electronic response to electrostatic gating. Hole doping via a negative back-gate voltage exhibits expected behavior by depleting the states near the Fermi level. Conversely, inducing electrons via positive gating does not result in the anticipated filling of the monolayer $TaIrTe_4$ upper band. Further intentional electron doping through *in situ* alkali surface dosing reveals further resistance to this Fermi level rigid shifts stems from an intrinsic band gap in monolayer $TaIrTe_4$, as suggested by our density functional theory (DFT) calculations utilizing the HSE hybrid functional [1]. We observe that this gap gradually closes as the electron concentration increases, pointing to strong electron interactions that dynamically modify the band structure. In conclusion, this study demonstrates that the conventional single-electron picture of rigid band shifting is insufficient to describe the complex band evolution observed during *in operando* ARPES.

[1] S. A. Ekahana *et al.*, "Visualization of tunable electronic structure of monolayer $TaIrTe_4$," *arXiv preprint*, arXiv:2601.11504 (2026)

3:00pm **2D-WeA-4 Substrate Driven Electronic Reconstruction in Bilayer Graphene***, **Aalok Tiwari**,* Carnegie Mellon University, USA, India*; Sandy Adhitia Ekahana, Souvik Sasmal,* Carnegie Mellon University, USA*; Liangtao Peng,* Washington University, St. Louis*; Pratik Saud,* Carnegie Mellon University, USA*; Syeda Faiza Rubab Sherazi,* University of Central Florida*; Ravi Kumar Bandapelli,* Carnegie Mellon University*; Duy Le,* University of Central Florida*; Rahul Rao,* Air Force Research Laboratory, Materials and Manufacturing Directorate, USA*; I-Hsuan Kao,* Carnegie Mellon University, USA*; Chris Jozwiak, Eli Rotenberg, Aaron Bostwick,* Advanced Light Source, Lawrence Berkeley National Laboratory*; Talat S Rahman,* University of Central Florida*; Simranjeet Singh,* Carnegie Mellon University, USA*; Shaffique Adam,* Washington University, St. Louis*; Jyoti Katoch,* Carnegie Mellon University, USA

The electronic and optical properties of two-dimensional van der Waals material systems are intimately tied to stacking configuration and sublattice arrangement. Interface engineering through substrate interactions, layer alignment and twist angle, offers a powerful route to design and manipulate electronic correlations. In this talk, I will use angle-resolved photoemission spectroscopy with micron sized spatial resolution (microARPES) to present direct evidence that substrate interactions and twist angle can dramatically modify the electronic band structure of bilayer graphene (BG). First, I will discuss our result of tuning the electronic structure of BG when interfaced with two distinct van der Waals substrates: hexagonal boron nitride (hBN), a wide-gap semiconductor, and α-$RuCl_3$, a Mott insulator, on the same sample. In the pristine heterostructure, differential charge transfer in the BG/hBN and BG/$RuCl_3$regions induces spatially modulated doping, forming an atomically sharp lateral p–p† junction. We observe a flattened valence band tip in BG on hBN, while BG on $RuCl_3$ exhibits strong hole doping. Upon electron doping via alkali-metal deposition, we access the conduction band in the heterostructure and observe that BG on hBN develops a ~0.4 eV band gap, while BG on $RuCl_3$ retains robust semimetallic in-gap states. These findings establish that substrate interactions modify BG's intrinsic ground state. Next, I will explore what occurs when graphene or BG is aligned with hBN at 0° or 30° along the high-symmetry Γ–K direction. At specific alignments, we observe flat bands away from the Fermi level whose dispersion is tunable via perpendicular electric field and strongly dependent on graphene–hBN relative orientation. These results demonstrate that substrate selection and layer alignment together provide a powerful basis for achieving correlated electronic phases in graphene-based heterostructures, opening pathways toward functional quantum devices in interface-tunable van der Waals systems.

3:15pm **2D-WeA-5 Interpreting Strain in X-Ray Photoelectron Spectroscopy Measurements of Two-Dimensional Materials***, **Muhidul Chaman**,* *Nargol Jalali, Michelle Becerra, Joy Roy, Joslin Prasanna,* The University of Texas at Dallas*; Paul Bagus,* The University of North Texas*; Rafik Addou, Cormac Toher, Kevin Brenner,* The University of Texas at Dallas

Strain is a common modification to two-dimensional (2D) materials. Their atomic thinness allows them to be strained from growth, interactions with substrates, and virtually all processing steps used to fabricate them into devices. As such, it is important to properly interpret strain in almost all characterizations of 2D materials. While the effects of strain are well understood for vibrational or valance electron spectroscopies, they are not well understood for core electron spectroscopies like X-ray photoelectron spectroscopy (XPS). In fact, the effects of strain on XPS measurements have been a longstanding challenge for many materials. This is partially due to the extreme difficulty in quantitatively computing core electron binding energies (BEs) compared to vibrational and electrical dispersions. This is problematic as the interpretation of XPS measurements requires reference peaks. As such, the lack of experimental and computational insights into the relationships between strain and core BEs significantly hinders the application of XPS measurements to 2D materials. We present the first XPS measurements that directly investigate the relationships between strain and core BEs in 2D (monolayer) MoS_2. These XPS measurements applied controlled strain by bending MoS_2 on a flexible substrate. The strain was then quantified using Raman spectroscopy and photoluminescence. We found the Mo $3d_{5/2}$ BE shifted by 0.19 eV and the S $2p_{3/2}$ BE also shifted by 0.11 eV in position per percentage of applied strain. These shifts are both significant (comparable to shifts from other physical and chemical phenomena) and counterintuitive to a simple electrostatic model that would predict opposite shifts. This suggested that there are changes in the core orbital distributions that offset the electrostatic effect. These shifts were investigated with density functional theory and proposed to arise from strain changing the bond distances, which changes the crystal and

ligand fields acting on the core orbitals. These measurements provide insights into the fundamental relationships between strain and core BEs and enable better interpretation of XPS measurements of 2D materials where strain is prolific.

3:30pm **2D-WeA-6 Electrical-XPS: A Novel Research Tool for Organic and Biological Materials***, **Hagai Cohen**,* The Weizmann Institute, Israel

XPSapplications to bio/organic systems encounter critical inherent challenges, in particular with those structural nuances that enrich functional diversity in biology. However, surprisingly effective answers can be gained by probing the electrical properties of specimens using an XPS-based technique, the chemically resolved electrical measurements (CREM). [1,2] A recent review focuses on this branch of electrical-XPS applications. [3]Challenges and their proposed answers are discussed, including specimens' limited stability, charging artifacts and, importantly, the typical XPS 'blindness' to key features of organic architectures. Among the unique capabilities demonstrated are the evaluation of monolayers' integrity, hydrogen-bond formation and structure-function relationships. Starting from small molecules up to relatively large supramolecular sugars and proteins, the CREM-XPS approach offers particularly attractive capabilities and a template for advanced characterization strategies.

References

1. I. Doron Mor et al., "Controlled Surface Charging as a Depth Profiling Probe for Mesoscopic Layers", *Nature* **406**, 382 (2000).
2. H. Cohen, "Chemically Resolved Electrical Measurements using X-ray Photoelectron Spectroscopy", *Appl. Phys. Lett.* **85**, 1271 (2004).
3. M. Miali et al., "Electrical XPS Meets Bio: In-Situ Chemo-Electrical Sensing and Activation of Organic Materials", *Phys. Chem. Chem. Phys.* **27,** 19591 (2025).

4:15pm **2D-WeA-9 Scanning Probe Microscopy of Engineered Bound States on Semiconductor Surfaces***, **Victor Brar**,* University of Wisconsin - Madison **INVITED**

I will present recent scanning probe microscopy (SPM) measurements of synthetic defects with deep in-gap energy levels in semiconducting 2D materials. It will be shown how charged impurities on the surface of WSe_2 can be manipulated to form artificial "nuclei" with deep, long-range two-dimensional Coulomb potentials that trap quasiparticles in hydrogenic-like orbits. These states persist to high quantum numbers, with binding energies exceeding 600 meV, such that the lowest level changes its occupation. Using multiple SPM modalities in combination with theoretical modeling, these experiments provide chemical insight into color centers in 2D materials and enable the creation of localized states with exotic properties.

4:45pm **2D-WeA-11 Interplay of Silver-Mean Quasiperiodicity and Weyl Semimetallicity in WTe_2***, **Daejin Eom**, So-Dam Sohn, Ja-Yong Koo, Chang-Youn Moon,* Korea Research Instutue of Standards and Science (KRISS), Republic of Korea

Quasiperiodic potential modulation in topological materials has attracted growing interest due to the intriguing interplay between the disorder effects associated with quasiperiodicity [1,2] and the topological invariance of these materials [3,4]. However, experimental studies on this topic remain scarce. Here, we employ the scanning tunneling microscopy (STM) to investigate the formation of an intercalation layer in WTe_2, a three-dimensional Weyl semimetal. The intercalant molecules are long-chain aliphatic *n*-alkanes dissolved in an epoxy resin. During thermal treatment, these molecules intercalate into the WTe_2 crystal and self-assemble into an atomically flat lamellar layer. Notably, the intercalation layer exhibits slight topographic variations in the STM images, characterized by two distinct heights, denoted as *L* and *H*. This variation forms a self-similar, quasiperiodic pattern, which is best described by the Pell sequence associated with the silver mean, although the coherence is occasionally disrupted by phason defects. Furthermore, the quasiperiodic potential modulation induced by the intercalation layer enhances the surface conductance near the Fermi level while preserving the semimetallic nature of WTe_2. This finding provides a direct experimental verification of theoretical predictions regarding the effects of quasiperiodic potential modulation on topological Weyl semimetals.

[1] C. Janot, *Quasicrystals: A Primer* (Oxford University Press, New York, 1994).[2] *Quasicrystals*, edited by T. Fujiwara and T. Ogawa (Springer, Berlin, 1990).[3] M. Z. Hasan, and C. L. Kane, Colloquium: Topological insulators, Rev. Mod. Phys. **82**, 3045 (2010).[4] X.-L. Qi and S.-C. Zhang, Topological insulators and superconductors, Rev. Mod. Phys. **83**, 1057 (2011).

5:00pm **2D-WeA-12 Emergent Superparamagnetism in TiS_2 Induced by Atomic-Scale Defects***, Thomas Pekarek, Patrick Keeney, Kyle Taylor, Cole Ratkus, Jason Haraldsen, **Paula Mariel Coelho**,* University of North Florida

Defect engineering has emerged as a powerful route to induce and control magnetism in otherwise nonmagnetic two-dimensional materials [1]. Titanium disulfide (TiS_2), a layered transition metal dichalcogenide traditionally studied for energy storage and electronic applications, is nonmagnetic in its pristine form, making it an ideal platform to explore defect-driven magnetic behavior [2]. Here, we report the observation of superparamagnetism in bulk 1T-TiS_2 arising from clusters of intrinsic defects. In a comprehensive scanning tunneling microscopy (STM), superconducting quantum interference device (SQUID) magnetometry, and density functional theory (DFT) study, we identify Ti adatoms as the primary source of local magnetic moments [3]. SQUID measurements reveal a pronounced paramagnetic response, and a Brillouin fit yields an effective spin of 4. This surprisingly large magnetic response is consistent with cooperative clustering of Ti adatoms forming superparamagnetic centers. Atomically resolved STM images show a high surface concentration of point defects and well-defined triangular clusters, while DFT calculations confirm that both isolated Ti adatoms and small Ti clusters generate localized magnetic moments and enhanced collective magnetic responses. These results establish TiS_2 as a defect-tunable magnetic material, highlighting its potential for future application in spintronics.

[1]P.M. Coelho, J. Phys.: Condens. Matter **36** 203001 (2024)

[2]P. J. Keeney et al Nanomaterials **15**, 1435 (2025)

[3]T.M. Pekarek et al., under review (2026)

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