

Magnetic Interfaces and Nanostructures Room 209 F W - Session MI+2D-WeA

Magnetic Interfaces and Nanostructures Oral Session

Moderators: Valeria Lauter, Oak Ridge National Laboratory, Hendrik Ohldag, Lawrence Berkeley National Laboratory

2:15pm **MI+2D-WeA-1 Probing Heterogeneity in 2D van der Waals Materials via Cryogenic STEM**, *Miaofang Chi, Joy Chao, Haoyang Ni*, Oak Ridge National Laboratory

INVITED

Quantum materials exhibit unique phenomena and functionalities that extend beyond classical physics. The use of 2D sheets and the construction of hetero- and moiré structures have emerged as promising approaches to inducing exotic quantum effects. However, studying these materials via cryogenic scanning transmission electron microscopy (STEM) and electron energy loss spectroscopy (EELS) has traditionally been limited by stage instability. Recent advancements in stage design by manufacturers now provide new opportunities for this research. In this talk, I will present our ongoing studies using atomic-scale cryogenic STEM and monochromated EELS to investigate the coupling between lattice and electronic structures in several representative 2D van der Waals materials relevant to magnetic storage and spintronic applications. One key example is the discovery of layer-number-dependent phase transitions in CrCl_3 during cooling. Another is the impact of defects and secondary phases on the magnetic structure evolution of $\text{Fe}_{5-x}\text{GeTe}_2$ (FGT-512). Additionally, we have mapped local excitons in moiré-structured MoTe_2 . These studies demonstrate that the electronic and magnetic properties of 2D materials can be tuned by controlling the layer number or engineering moiré structures. They also highlight the power of combining high-resolution cryogenic STEM imaging and spectroscopy to advance the understanding of quantum materials.[1]

[1] This work was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences, and Engineering Division and was performed at the Center for Nanophase Materials Sciences at ORNL.

2:45pm **MI+2D-WeA-3 Examining the influence of magnetic and electron beam probes on the topologically-protected edge states of 2D Bi_2Te_3 Nanoplates**, *Timothy Carlson¹, Swathi Kadaba*, Wake Forest University; *Gabriel Marcus*, Quoharent; *Motahhare Mirhosseini, David Carroll*, Wake Forest University

In this work well defined, stoichiometric two-dimensional (2D) nanoplates of the topological insulator, Bi_2Te_3 , were imaged using magnetic force microscopy (MFM), atomic force microscopy (AFM), and high resolution transmission electron microscopy (HRTEM) including techniques such as electron energy loss spectroscopy (EELS) and cross sectional TEM. Nanoplates with a diameter range of 0.5 to 1.5 μm and $\sim 6\text{-}15\text{nm}$ thick were supported on highly ordered pyrolytic graphite (HOPG) for the scanning probes and ultra thin, lacy TEM grids for the electron probes imaging and spectroscopy. In the case for the MFM experiments, the relative strength of the edge-fields were characterized by adjusting the lift heights resulting in a unique relationship between the magnetic probe and the nanoplates under observation. For the EELS experiments, the data was collected on the edges of the nanoplates and signatures indicative of edge channels was observed. We suggest in both cases time-reversal symmetry breaking in the Bi_2Te_3 nanoplate from the field of the magnetic cantilever and the high electron flux from the electron beam. These symmetry breaking interactions are believed to produce induced, topologically protected currents. The addition of an applied DC bias to the tip enabled the controlled filling of Landau levels by lowering or raising the fermi level. Previous studies suggest Bi_2Te_3 nanoplates of similar proportions to lie within the 3D topological insulator family and therefore harbor 2D surface states, however, based on the nature of the contrast seen in the MFM, electron energy loss spectroscopy (EELS), and our synthesis method we argue these nanoplates fall within the 2D topological insulator family. These studies reveal the existence of persistent currents in our 2D Bi_2Te_3 system at room temperature and point to MFM and EELS as powerful tools for probing such topologically protected quantum spin hall states.

3:00pm **MI+2D-WeA-4 Surface of Topological Weyl Semimetal $\text{PtBi}_{1.6}$** , *Zheng Gai*, Oak Ridge National Laboratory; *Dejia Kong*, Department of Chemistry, University of Virginia, Charlottesville, VA 22903; *Rongying Jin*, University of South Carolina, Columbia, SC 29208

PtBi_{2-x} (specifically $\text{PtBi}_{1.6}$) is a noncentrosymmetric Weyl semimetal that hosts topologically protected surface states, making it a fascinating platform for exploring exotic surface and bulk phenomena. The material naturally cleaves to reveal two distinct surface terminations: a buckled Bi1 surface with $3m$ symmetry, and a flat Bi2 surface with m symmetry. $\text{PtBi}_{1.6}$ also exhibits giant magnetoresistance, surface superconductivity, and evidence of robust quasiparticle interference patterns, making it a compelling candidate for applications in spintronics, quantum sensing, and topological quantum computing. However, several key questions remain open. One major challenge is understanding the role of surface states in transport phenomena—particularly whether they contribute to the large magnetoresistance observed at low temperatures. Our scanning tunneling microscopy (STM) studies reveal the presence of both Bi1 and Bi2 terminations upon cleaving, consistent with prior surface-sensitive spectroscopic studies. Detailed quasiparticle interference (QPI) analyses highlight contrasting behaviors on the two surface types, suggesting that the electronic structure and scattering mechanisms are highly termination-dependent. Additionally, we examine the impact of atomic-scale defects on the surface states, providing insight into their stability and resilience. These findings deepen our understanding of surface-bulk interplay in noncentrosymmetric topological systems and underscore the importance of surface engineering in future device applications.

The STM work of this research was conducted at the Center for Nanophase Materials Sciences, ORNL, which is a DOE Office of Science User Facility.

3:15pm **MI+2D-WeA-5 Visualizing Electronic and Magnetic Structure at Nanoscale for Spintronics**, *Jyoti Katoch*, Carnegie Mellon University, USA

INVITED

Topological semimetals, such as WTe_2 and TaIrTe_4 , have strong spin-orbit coupling, non-trivial band dispersion, and bulk and surface spin polarized states. A combination of intrinsic spin Hall effect and surface state driven efficient and unconventional spin current generation can be obtained in these systems for manipulating the magnetic order. However, the comprehensive understanding of electronic structure, which is directly responsible for charge to spin conversion, of these systems at mesoscopic scale remains critical missing. I will discuss our results on probing spatially resolved electronic structure of atomically thin layers of WTe_2 and TaIrTe_4 using nanoARPES. Moreover, recently, we reported the first experimental realization of field-free deterministic magnetic switching of a perpendicularly polarized van der Waals (vdW) magnet employing spin current with out-of-plane spin polarization in layered WTe_2 . We will discuss our efforts to utilize the photoemission electron microscopy (PEEM) paired with x-ray magnetic circular dichroism (XMCD) to obtain a spatially resolved view on the underlying mechanism of this magnetic switching behavior. Finally, I will discuss our experiments aimed at nanoscale imaging of magnetic structure of atomically thin films of a vdW antiferromagnet, i.e., CrSBr . Layered magnetic systems display highly intriguing properties, such as thickness-dependent magnetic ground state, electric field tunability, enhancement of interlayer AFM exchange coupling in the ultra-thin limit, and tunable magnon-magnon coupling, to name a few. We will report on experiments wherein we employ PEE) paired surface-sensitive XMCD/XMLD to perform layer-dependent domain imaging in mesoscopic sized samples of CrSBr . We will discuss detailed thickness, temperature, and externally applied magnetic field-dependent magnetic domain imaging of atomically thin samples of CrSBr .

4:15pm **MI+2D-WeA-9 Layered Systems for Spintronics and Quantum Sensing of Spin Dynamics**, *Simran Singh*, Carnegie Mellon University

INVITED

Low-dimensional systems and their atomically precise heterostructures are a modular material platform to study emergent spin and magnetism related phenomena. I will present our work(s) on exploring topological semimetals and layered magnets based low-dimensional heterostructures to realize novel spin-galvanic effects for electric field control of the magnetic order, demonstrate a new type of unidirectional magnetoresistance, and realize an unconventional form of anomalous Hall effect. First, I will discuss our experiments to employ spin-current with an out-of-plane spin polarization generated in a low-symmetry topological semimetal to deterministically switch and read the magnetic state(s) of perpendicularly polarized magnets. Secondly, I will discuss the experimental realization of unconventional form of anomalous Hall effect in a low-dimensional

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heterostructures, which is proportional to not only out-of-plane magnetization but also to in-plane magnetization component, potentially expanding the parameter space for designing dissipationless edge transport in low-dimensional systems. Furthermore, spin-defects can be engineered in low-dimensional systems – an appealing prospect for quantum sensing technologies. Time permitting, I will present our work aimed at utilizing designer spin defects embedded in a two-dimensional system to probe broadband spin dynamics.

4:45pm MI+2D-WeA-11 Surface Electronic Structure Comparison of Fe-Intercalated and 2H-TaS₂, *Dejia Kong, Sree Sourav Das, Jacob St. Martin*, University of Virginia, USA; *Peter Siegfried*, George Mason University; *Zhiqiang Mao, Seng Huat Lee*, The Pennsylvania State University; *Ian Harrison*, University of Virginia; *Nirmal Ghimire*, University of Notre Dame; *Mona Zebbarjadi*, University of Virginia, USA; **Zheng Gai**, Oak Ridge National Laboratory, USA; *Petra Reinke*, University of Virginia, USA

Anisotropic ferromagnetic phases can be introduced to transitional metal dichalcogenide (TMD) TaS₂ through intercalating Fe in the van der Waals (vdW) gap. By deviating from the commensurate values ($x = \frac{1}{4}$ or $\frac{1}{3}$), the crystalline structure as well as the magnetotransport properties of the TMD system can be tuned. For instance, Fe_{1/4}TaS₂ has a centrosymmetric 2×2 structure while Fe_{1/3}TaS₂ has a non-centrosymmetric $\sqrt{3} \times \sqrt{3}$ supercell structure. The magnetic Curie temperature of Fe_xTaS₂ also exhibits a strong dependence on Fe concentration. We evaluate Fe_{0.28}TaS₂ and 2H-TaS₂ samples using STM/Spectroscopy (STM/S) and density functional theory (DFT) to investigate the real-space intercalant electronic structure comparatively and the potential phase segregation between the two commensurate compounds. Fe_{0.28}TaS₂ shows a supercell at 77 K, whereas 2H-TaS₂ displays no apparent supercell at the same temperature. Fe vacancy defects and clusters are discovered in the intercalated surface, and their surrounding local density of states (LDOS) shows non-trivial differences at energies compared to the pristine Fe_{0.28}TaS₂ area, which is related to Fe orbitals contributions based on the DFT calculations.

The STM work of this research was conducted at the Center for Nanophase Materials Sciences, ORNL, which is a DOE Office of Science User Facility.

5:00pm MI+2D-WeA-12 Chirality, Surface Termination and Anti-ferromagnetic Alignment in Fe(III) Spin Crossover Salts, *Mohammad Zaid Zaz¹*, University of Nebraska-Lincoln; *Wai Kiat Chin, Arjun Subedi, Gauthami Viswan*, University of Nebraska - Lincoln; *Alpha T.N'Daiye*, Advanced Light Source, Lawrence Berkeley National Laboratory; *Alexander Wysocki*, University of Nebraska-Kearney; *Rebecca Lai, Peter A Dowben*, University of Nebraska - Lincoln

Switchable molecular materials based on 3d transition metal complexes are a rich platform for exploring phenomenon related to symmetry breaking which include chirality and surface termination. In certain di-nuclear species, magnetic ordering between different metal ions is also witnessed. We explore chirality, surface termination and anti-ferromagnetic alignment in an Fe(III) spin crossover complex namely [Fe(qsal)₂Ni(dmit)₂] where, qsal = N(8quinolyl)salicylaldimine, and dmit²⁻ = 1,3-dithiol-2-thione-4,5-dithiolato. We employ spatially resolved Fe-L_{3,2} edge X-ray absorption spectroscopy to probe the chiral signature at the Fe metal center. Surface termination is studied by complementary X-ray photoemission spectroscopy and energy dispersive X-ray spectroscopy. These are further complemented by inverse photoemission spectroscopy and Fe, Ni-L_{3,2} edge X-ray absorption spectroscopy. Finally, we investigate the anti-ferromagnetic alignment in this system by X-ray magnetic circular dichroism measurements at the Fe and Ni core.

5:15pm MI+2D-WeA-13 Impact of Nanoscale Curvature on the Structural and Magnetic Properties of Co/Pd Alloys, *Asma Qdemat, Asma Qdemat*, ORNL

Researchers have studied a lot about the properties of magnetic thin films grown on flat substrates. This is mostly because it's easy to make them and there are well established ways to process them. However, researchers have not studied enough about how nanoscale curvature affects them. This study aims to fill that gap by directly comparing the structure and magnetic behavior of Co/Pd alloy thin films deposited on flat versus curved surfaces.

In this contribution, we will present a detailed investigation of the effects of how nanoscale curvature affects the structural and magnetic properties of Co/Pd alloys deposited on flat silicon substrates and highly ordered monolayers of 50 nm and 200 nm SiO₂ nanospheres, using molecular beam epitaxy (MBE). We used a variety of advanced methods to study our

system, including magnetometry, X-ray reflectivity (XRR), polarized neutron reflectometry (PNR), and grazing-incidence small-angle neutron and X-ray scattering (GISANS/GISAXS) was employed to probe both depth-resolved and lateral properties.

Structural analysis via SEM and XRR revealed that films deposited on flat silicon maintained smoother, while those grown on curved nanospheres exhibited increased surface roughness and disrupted periodicity. In films on curved substrates, a parabolic scattering length density (SLD) model was necessary to capture the curvature-induced gradient in density profiles. GISAXS and GISANS confirmed these findings, showing less nanospheres ordering and greater lateral roughness, particularly in thicker films. Furthermore, magnetically, the nanoscale curvature significantly influenced anisotropy. SQUID measurements showed strong perpendicular magnetic anisotropy (PMA) in films on flat substrates, with square hysteresis loops and high remanence. In contrast, films on curved nanospheres had increased coercivity, reduced saturation magnetization, and a tilted magnetization axis, effects that were more pronounced in thinner films. These observations were further confirmed by PNR, which revealed that curvature changes the magnetic SLD profiles and increases the Co magnetic moment. This is likely due to strain and changes in the interfacial coupling.

Our findings show that nanoscale curvature is important in controlling how magnetic alloys behave. Curvature can reduce the uniformity of the structure and the magnetic properties. But it can also open up new ways to control local magnetic interactions by altering strain and anisotropy. These insights are very important for the development of flexible, conformal magnetic devices where precise control over magnetic anisotropy is required.

5:30pm MI+2D-WeA-14 Emergence of local magnetic moment in ternary TaWSe₂ single crystal via atomic clustering, *Jewook Park*, Oak Ridge National Laboratory

Ternary transition metal dichalcogenides (TMDs) provide a versatile platform to explore novel electronic and magnetic ground states via compositional substitution and local structural modulations. Using a combination of scanning tunneling microscopy and spectroscopy (STM/S), magnetic property measurements, and density functional theory (DFT) calculations, we analyze the emergence of local magnetic moments driven by the clustering of Ta atoms in ternary TaWSe₂ single crystals. STM topography reveals triangular clusters of Ta atoms embedded within W-rich regions of TaWSe₂. These clusters exhibit a consistent shape and an orderly arrangement throughout the surfaces. DFT calculations show that these Ta clusters induce local strain, giving rise to localized magnetic moments. The magnetic behavior is further corroborated by temperature-dependent magnetization measurements, which exhibit a magnetic transition near 50 K. This study offers a pathway to engineer magnetism in TMD systems with potential applications in spintronic and quantum materials.

2D Materials

Room 208 W - Session
2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM

2D Materials: Optoelectronics and Moire Excitons

Moderator: Daniel Yimam, Oak Ridge National Laboratory

8:00am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-1 Probing the Ultrafast Charge Dynamics and Exciton Emission from Single Atomic Defects in 2D Semiconductors by Lightwave-Driven STM**, *Laric Bobzien, Lysander Huberich, Jonas Allerbeck, Eve Ammerman, Nils Krane, Andres Ortega-Guerrero, Carlo Pignedoli, Oliver Gröning*, Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland; *Joshua A. Robinson*, The Pennsylvania State University; *Bruno Schuler*, Empa, Swiss Federal Laboratories for Materials Science and Technology, Switzerland **INVITED** Two-dimensional (2D) semiconductors provide an exciting platform to engineer atomic quantum systems in a robust, yet tunable solid-state system. This talk explores the intriguing physics of single point defects in transition metal dichalcogenide (TMD) monolayers, investigated through atomically resolved scanning probe microscopy.

We have determined the layer-dependent charge transfer lifetimes of selenium vacancies in WSe_2 on graphene substrates, spanning picosecond to nanosecond timescales [1]. By leveraging our recently developed lightwave-driven scanning tunneling microscope (THz-STM) [2,3], we could probe the ultrafast charge dynamics on the atomic scale. Time-domain sampling with a THz pump-THz probe scheme enabled capturing atomic-scale snapshots of transient Coulomb blockade, a hallmark of charge transport mediated by quantized defect states [4].

Moreover, the extended charge state lifetimes provided by hBN decoupling layers facilitated the local, electrical stimulation of excitonic emission from pristine MoS_2 and individual charged defects via STM luminescence (STML).

By combining the structural and electronic properties accessible by conventional scanning probe microscopy with the optical fingerprint from STML and the excited-state dynamics revealed through pump-probe THz-STM, we gain a comprehensive microscopic understanding of localized quantum states in low-dimensional materials.

References:

- [1] L. Bobzien et al. Phys. Rev. Lett. (accepted, arxiv: 2407.04508)
- [2] J. Allerbeck et al. ACS Photonics 10, 3888 (2023)
- [3] L. Bobzien et al. APL Mater. 12, 051110 (2024)
- [4] J. Allerbeck et al. arXiv:2412.13718 (2024)
- [5] L. Huberich et al. (in preparation)

8:30am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-3 Many-Body Effects on Excitons, Trions, and Defect-Bound States in 2D Materials**, *Kai Xiao, Taegwan Park, Alexander Poretzky*, Oak Ridge National Laboratory, USA; *Xufan Li*, Honda Research Institute; *Kyungnam Kang*, Oak Ridge National Laboratory, USA; *Austin Houston*, University of Tennessee, Knoxville; *Christopher Rouleau, David Geohagan*, Oak Ridge National Laboratory, USA Two-dimensional (2D) materials, particularly transition metal dichalcogenides (TMDs) exhibit strong many-body interactions due to reduced dielectric screening and spatial confinement. These interactions, involving electrons, holes, excitons, phonons, and plasmons, give rise to emergent phenomena distinct from their bulk counterparts. In this talk, I will present our recent investigations into the many-body effects on the optical properties and ultrafast excitonic dynamics of monolayer and bilayer TMDs. Specifically, we synthesized isotopically pure monolayer MoS_2 and highly defective WS_2 via nonequilibrium chemical vapor deposition, enabling a controlled study of isotope effects, defects, and background doping on excitonic behavior. Using ultrafast laser spectroscopy and temperature-dependent optical spectroscopy, we observed pronounced many-body interactions, including exciton-phonon and exciton-electron coupling, which significantly influence exciton energy, dynamics, and light-matter interactions in both monolayer and bilayer TMDs. These strong interactions give rise to novel quantum states and make 2D materials promising platforms for next-generation optoelectronics, quantum information technologies, and fundamental condensed matter physics.

Synthesis science was supported by the U.S. Dept. of Energy, Office of Science, Materials Science and Engineering Division. This work was performed at the Center for Nanophase Materials Sciences, which is a DOE Office of Science User Facility.

8:45am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-4 Proximity-Induced "Magic" Raman Bands in TERS Spectra of MoS_2 / WS_2 @ 1L h-BN-Capped Gold**, *Andrey Krayev*, HORIBA Scientific; *Pavel Valencia Acuna*, PNNL; *Ju-Hyun Jung*, Pohang University of Science and Technology (POSTECH), Republic of Korea; *Cheol-Joo Kim*, POSTECH, Republic of Korea; *Andrew Mannix*, Stanford University; *Eleonora Isotta*, Max Planck Institute for Sustainable Materials, Germany; *Chih-Feng Wang*, PNNL

Recently it was proposed to use the monolayer h-BN – capped gold substrates as an ideal platform for the gap mode TERS and TEPL imaging, that on the one hand, should preserve strong gap mode enhancement of Raman signal due to small thickness (0.3 nm) of the dielectric h-BN layer, and on the other hand preserve strong TEPL response due to de-coupling of 2D semiconductors from the metallic substrate. TERS data collected on mono- and a few-layer-thick crystals of MoS_2 and WS_2 on 1L-h-BN-capped gold show both the TERS and TEPL response, confirming the validity of the proposed approach.

In addition to the enhancement of both the PL and Raman signal, in the course of assessment of TERS/TEPL response of mono- and a few-layer-thick crystals of MoS_2 and WS_2 deposited on 1L h-BN-capped gold we observed in TERS spectra, completely unexpectedly, appearance of Raman bands at about 796 cm^{-1} and 76 cm^{-1} which are not normally observed in regular Raman spectra of h-BN or WS_2/MoS_2 . We can safely state that these "magic" bands belong to h-BN as they appear at the same spectral position in TERS spectra of both the monolayer MoS_2 and WS_2 deposited on the monolayer h-BN capped gold, moreover, the 796 cm^{-1} band often was the strongest band observed in TERS spectra, even stronger than A' mode from WS_2 or MoS_2 . Presence of the transition metal dichalcogenide (TMD) monolayer is mandatory for the appearance of these "magic" bands as they are absent outside of the monolayer TMDs in these samples. Literature search showed that similar (but not identical) phenomenon was observed earlier in h-BN encapsulated $\text{WSe}_2/\text{MoSe}_2$ and WS_2 . There have been several significant differences between our data and the earlier reported one: in our case we have not been able to observe the "magic bands" in MoSe_2 and WSe_2 @ 1L h-BN@Au, while WS_2 monolayers deposited on the same substrate as WSe_2 , showed expected response. More importantly, the excitation laser wavelength dependence in our case was completely different from what was reported earlier: in WS_2 -based samples we observed strong "magic" bands with excitation at 830 nm, 785nm, 594nm, but not 633nm, the wavelength closest to the A exciton in this material. This excitation profile is remarkably reminiscent of the excitation profile of the monolayer WS_2 in intimate contact with silver where we observed strong dip of the intensity of main A' mode in TERS spectra at 633nm excitation wavelength.

We will argue that intricate interaction between the tip-substrate gap plasmon, TMD excitons and most probably, normally mid-IR-active phonons in h-BN is responsible for the appearance of observed "magic" bands.

9:00am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-5 Correlated Excitons in TMDC Moiré Superlattice**, *Sufei Shi*, Carnegie Mellon University **INVITED**

In a strongly correlated electronic system, Coulomb interactions among electrons dominate over kinetic energy. Recently, two-dimensional (2D) moiré superlattices of van der Waals materials have emerged as a promising platform to study correlated physics and exotic quantum phases in 2D. In transition metal dichalcogenides (TMDCs) based moiré superlattices, the combination of large effective mass and strong moiré coupling renders the easier formation of flat bands and stronger electronic correlation, compared with graphene moiré superlattices. Meanwhile, the strong Coulomb interaction in 2D also leads to tightly bound excitons with large binding energy in TMDCs. In this talk, we will discuss how to use optical spectroscopy to investigate excitonic physics and strongly correlated phenomena in TMDC moiré superlattice, along with correlated exciton states arising from strong interactions.

9:30am **2D+AQS+EM+MI+MN+NS+QS+SS+TF-ThM-7 Sub-Stoichiometric Phases in 2D MoTe_2** , *Onyedikachi Alanwoko, Nirasha Rajapakse, Matthias Batzill*, University of South Florida

Atom vacancy formation in crystalline materials is energetically expensive. To lower the energy cost for non-stoichiometry, point defects can condense into energetically more favorable extended defects. Studies on Mo-dichalcogenides have shown that excess Mo is condensed into closed, triangular Mirror Twin Boundary (MTB) loops. These MTBs can form in high densities where the triangular loops connect and form a cross-hatched network of MTBs. Here we show through Scanning Tunneling Microscopy (STM) that periodically ordered MTB networks can obtain a homologous series of sub-stoichiometric MoTe_{2-x} phases. We systematically investigate

the preparation conditions (which include a variation of the growth temperature, Te-desorption by post-growth annealing, and vapor-deposited Mo), enabling the controlled synthesis of these new phases. The different phases require different synthesis procedures, and once formed, these phases appear thermally stable in vacuum. The ability to control and create these different phases of MoTe₂ and other two-dimensional (2D) materials is a promising way of realizing new electronic and chemical properties of 2D materials. Particularly promising is the observation that we can react MoTe₂ with dissimilar transition metals to create new doped or alloyed 2D materials with potentially desirable properties.

11:00am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-13 Microwave Imaging of Excitonic States and Fractional Chern Insulators in 2D Transition Metal Dichalcogenides, Zhurun Ji, SLAC National Accelerator Laboratory/ MIT INVITED

Nanoscale electrodynamics offers a unique perspective on states with bulk-edge correspondence or spatially dependent excitations. I will introduce our latest advancements in optically coupled microwave impedance microscopy, a technique that enhances our capability to explore electrodynamics at the nanometer scale. I will discuss our recent studies utilizing this technology to extract spectroscopic information on exciton excitations within transition metal dichalcogenide systems. Additionally, I will share our recent findings on probing topological and correlated electronic states, specifically the fractional Chern insulator states in twisted TMD bilayers.

11:30am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-15 Control and Properties of Single Dislocations in Van Der Waals Nanowires, Peter Sutter, Eli Sutter, University of Nebraska - Lincoln

Line defects (dislocations) not only govern the mechanical properties of crystalline solids but they can also produce distinct electronic, thermal, and topological effects. Identifying and accessing this functionality requires control over the placement and geometry of single dislocations embedded in a small host volume to maximize emerging effects. We have identified a synthetic route that enables the rational placement and tuning of dislocation in van der Waals nanowires, where the 2D/layered crystal structure limits the possible defect configurations and the nanowire architecture puts single dislocations in close proximity to the entire host volume.¹ While homogeneous layered nanowires carry individual screw dislocations, the synthesis of radial (core-shell) nanowire heterostructures transforms the defect into a mixed (helical) dislocation whose edge-to-screw ratio is continuously tunable via the core-shell lattice mismatch.

Such deterministic control over defects now enables the probing of functionality arising with single dislocations. For example, germanium sulfide van der Waals nanowires carrying single screw dislocations incorporate Eshelby twist and thus adopt a chiral twisted structure,² which for the first time allowed the identification of chirality effects in the photonic properties of a single nanostructure.³ Using cathodoluminescence spectroscopy, whispering gallery modes could be excited and probed to directly compare the photonics of chiral and achiral segments in single nanowires. The data show systematic shifts in energy, which with the help of simulations are assigned to chiral whispering gallery modes in wires hosting a single dislocation.

The ability to design nanomaterials containing individual dislocations with controlled geometry paves the way for identifying a broad range of functional properties of dislocations, with the potential to herald a paradigm shift from the traditional strategy of suppressing dislocations to embracing and harnessing them as core elements of new technologies.

1. P. Sutter, R.R. Unocic, and E. Sutter, *Journal of the American Chemical Society* 145, 20503 (2023); DOI: 10.1021/jacs.3c06469
2. P. Sutter, S. Wimer, and E. Sutter, *Nature* 570, 354 (2019); DOI: 10.1038/s41586-019-1147-x
3. P. Sutter, L. Khosravi-Khorashad, C.V. Ciobanu, and E. Sutter, *Materials Horizons* 10, 3830 (2023); DOI: 10.1039/D3MH00693J

11:45am 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-16 Two-Dimensional Keldysh Theory for Non-Resonant Strong-Field Ionization of Monolayer 2D Materials, Tsing-Hua Her, UNC Charlotte; Che-Hao Chang, NTHU, Taiwan; Kenan Darden, UNC Charlotte; Tsun-Hsu Chang, NTHU, Taiwan; Hsin-Yu Yao, NCCU, Taiwan

Over the past decade, a diverse array of intense light interactions with monolayer two-dimensional (2D) materials have been reported, including low- and high-order harmonic generation (HHG), multiphoton absorption, optical injection of spin and charge currents, terahertz generation, and

laser-induced dielectric breakdown. These processes are all initiated by interband transition of electrons induced by light with photon energy smaller than the bandgap energy. Some modeling efforts based on perturbation theory were attempted but their discrepancies with experiments are at least one order of magnitude, highlighting the lack of theoretical understanding of strong-field ionization in monolayer 2D materials. In this work, we report a new formalism of strong-field ionization for monolayer two-dimensional semiconductors based on 2D Keldysh (KLD) theory [T. -H. Her et al., *Optica* 12, 538-545 (2025)]. We take this approach because the original Keldysh theory [L. V. Keldysh, *Soviet Physics JETP* 20, 8 (1965)] is the only theory that yields, for simplified band dispersion, analytical formulas for the cycle-averaged non-resonant ionization rate in bulk solids induced by a monochromatic electric field of arbitrary strength. It provides a smooth transition between multiphoton and tunneling ionization as what we now call the ‘Keldysh parameter’, γ , varies from $\gamma \gg 1$ to $\gamma \ll 1$. Due to their analyticity, Keldysh’s formulas are widely employed for qualitative modeling of strong-field ionization in bulk solids. In this presentation, we generalize Keldysh’s formulas to monolayer two-dimensional semiconductors. We derive closed-form formulas and their asymptotic forms for a two-band model with a Kane dispersion. We also derive selection rules related to the parity of multiphoton orders near the band edge. We validate our theory by comparing it to recent experiments and modeling of strong-field ionization in monolayer transition metal dichalcogenides (TMDs) with very good agreement (Figs. 1-3 of supplemental document) Specifically, our theory predicts a higher interband electron tunneling rate for 2D compared to 3D in the MIR frequency range, which successfully explains the 10x discrepancy (Fig. 3) between the experiment and modeling for HHG from monolayer TMDs [Liu et al., *Nature Phys* 13, 262–265 (2017)]. Considering the tremendous success of the original Keldysh theory in describing strong-field optical phenomena in atoms and solids, our 2D Keldysh theory is expected to find a wide range of applications in intense light-2D material interaction, such as optical limiting, multi-photon photodetection, THz generation through quantum interference, and photo-carrier doping for HHG.

12:00pm 2D+AQs+EM+MI+MN+NS+QS+SS+TF-ThM-17 Thickness Dependent Band Gap and Electrical Anisotropy of 2DSnSe, Marshall Frye, Jonathan Chin, Joshua Wahl, Jeremy Knight, Georgia Institute of Technology; Walter Smith, Purdue University; Dilara Sen, Samuel Kovach, Kenyon University; Frank Peiris, Kenyon College; Charles Paillard, University of Arkansas; Thomas Beechem, Purdue University; Anna Osterholm, Lauren Garten, Georgia Institute of Technology

2D SnSe presents unique opportunities for optoelectronics, and scalable microelectronics, but it is first critical to understand how the electrical and optical response change upon downscaling. Tailoring the band gap and electrical anisotropy of 2D monochalcogenides, like SnSe, has previously been shown but the mechanisms that drive the changes in band gap are still not understood. This study reveals how changes in bond length and structure drive the thickness dependences of band gap, carrier mobility and lifetime of SnSe thin films. Molecular beam epitaxy is used to deposit (2h00) oriented SnSe thin films with thicknesses ranging from 4 nm to 80 nm. The direct band gap increases from 1.4 eV at 80 nm to 1.9 eV at 4 nm, underscoring the potential of SnSe as a tunable and direct band gap material for thin film optoelectronics. Raman spectroscopy shows different simultaneously changes in the crystal structure and bonding occurring parallel versus perpendicular to the 2D plane with decreasing film thickness. TEM further supports the hypothesis that the increase in the band gap with reduced thickness is due to changes in crystal structure resulting in a contraction of the out-of-plane SnSe covalent bonds, while the in-plane bond length increases. In addition to the reduction in band gap, tracking the time dependent photoluminescence shows an increase in carrier lifetime with decreasing film thickness, while Hall measurements show a change in the carrier mobility with decreasing thickness. Overall, this work provides the critical missing insight needed to design these optically and electronically relevant 2D materials for scalability.

Actinides and Rare Earths

Room 207 A W - Session AC+MI-ThM

Superconductivity, Magnetism, Electron Correlation and Complex Behavior

Moderators: Krzysztof Gofryk, Idaho National Laboratory, James G. Tobin, University of Wisconsin-Oshkosh

8:00am AC+MI-ThM-1 Phase Transition and Magnetism in UTe_2 , Dominik Legut, VSB - Technical University of Ostrava, Czechia; **Alexander Shick,** Institute of Physics CAS, Prague, Czechia; **Urszula Wdowik,** VSB - Technical University of Ostrava, Czechia

INVITED

For the magnetic properties of UTe_2 the correlated band theory implemented as a combination of the relativistic density functional theory with exact diagonalization [DFT+U(ED)] of the Anderson impurity term with Coulomb repulsion U in the $5f$ shell needs to be applied. This allows us to determine the orbital to spin ratio as well as number of the uranium valence states in close correspondence with recent experiment (XANES, XMCD). The uranium atom $5f$ -shell ground state with 33% of f^2 and 58% of f^3 configurations is determined[1]. In contrast to the above, for the bonding in UTe_2 it is satisfactory to be modelled by DFT+U methodology. We theoretically determined the lattice contribution to the specific heat of UTe_2 over the measured temperatures ranging from 30 to 400 K as well as the the orthorhombic-to-tetragonal phase transition pressure of 3.8 GPa at room temperature in very good agreement with the recent experimental studies. Last, but not least we determined the Raman spectra that were compared with recent Raman scattering experiments as well.

[1] A. B. Shick, U. D. Wdowik, I. Halevy, and D. Legut, Spin and Orbital Magnetic Moments of UTe_2 induced by the external magnetic field, Scientific Reports **14**, 25337 (2024), <https://doi.org/10.1038/s41598-024-75321-4>.

[2] U. D. Wdowik, M. Valiska, A. Cabala, F. Borodavka, E. Samolova, and D. Legut, Raman spectroscopy and pressure-induced structural phase transition in UTe_2 , Frontiers of Physics, **20**, 014204 (2025), <https://doi.org/10.15302/frontphys.2025.014204>.

8:30am AC+MI-ThM-3 Single-ion Anisotropy Controls Magnetic Excitations in REMn_2Sn_6 (RE = Tb, Dy, Ho) Ferimagnetic Kagome Metals, Kelsey Collins, Michael Susner, Michael Newburger, Air Force Research Laboratory, Materials and Manufacturing Directorate, USA

The REMn_2Sn_6 family of materials, where RE is a generic rare earth trivalent cation, have attracted much interest due to their complex electronic and magnetic structures. This complexity arises from the coupling of highly anisotropic lanthanide ions spins with large spin-orbit couplings to the spins of the Mn atoms, which are arrayed in a kagome lattice, a lattice topology known to give rise to exotic topological phenomena. This interplay of magnetic anisotropy and electronic topology motivates investigation into the magnetic excitations of these materials, which unlike the ground state magnetic structures of this family have not been extensively studied. Herein, we use Brillouin light scattering to measure the magnon spectra of TbMn_2Sn_6 , DyMn_2Sn_6 , and HoMn_2Sn_6 , measuring the magnon frequency, lifetime, and intensity as a function of applied magnetic field and sample temperature. We find that the identity of the lanthanide effectively tunes the frequency of the magnon over a range from ~ 18 GHz (Tb) to ~ 5 GHz (Ho) at zero applied field. Despite the difference in the magnetic ground states between a canted easy-axis (Tb) and easy-cone (Dy, Ho) spin orientation, these three congeners respond remarkably similarly to a magnetic field. Quantitative fitting of the three magnon dispersions reveals that the tuning of the magnon frequencies stems primarily from the differing magnetic anisotropies of the lanthanide ions. This work demonstrates that the anisotropy of the trivalent ion controls not only the magnetic ground state, but also the magnetic excitations, in this family of topological magnets.

8:45am AC+MI-ThM-4 Suppression of the CDW State in UPT_2Si_2 by Ir Substitution; $5f$ States Into Bonding, Ladislav Havela, Charles University, Faculty of Mathematics and Physics, Czechia; **Volodymyr Buturlim,** Idaho National Laboratory; **Silvie Cerna,** Oleksandra Koloskova, Charles University, Faculty of Mathematics and Physics, Czechia; **Daniel Chaney,** ESRF, Grenoble, France; **Peter Minarik,** Charles University, Faculty of Mathematics and Physics, Czechia; **Mayerling Martinez Celis,** CRISMAT, University of Caen, France; **Dominik Legut,** Charles University, Faculty of Mathematics and Physics, Czechia

$5f$ states in light actinides adopt either an itinerant, i.e. bonding, nature, or they preserve their localized atomic character similar to free ions and they

stand aside from bonding. The large pool of known U intermetallics comprises mainly compounds with itinerant $5f$ states. One of exceptions is arguably UPT_2Si_2 , at which some features of $5f$ localization were identified [1,2]. One of its interesting features is the Charge Density Wave (CDW) with a propagation vector (0.42,0,0), developing below $T = 320$ K [3]. Importantly, practically identical CDW appears also in multiple rare-earth isotopes REPT_2Si_2 with localized (or empty) $4f$ states, all crystallizing in the tetragonal structure type CaBe_2Ge_2 [4]. While the CDW phenomenon is very interesting per se (one can discuss whether it is primarily due to phonon softening of Fermi surface nesting), one can also assume it as a sensitive indicator of the $5f$ localization. The only U-based sibling, ULr_2Si_2 , is undoubtedly an itinerant antiferromagnet and no CDW has been reported.

Here we describe results of the study of the pseudo-ternary system $\text{U}(\text{Pt}_{1-x}\text{Ir}_x)_2\text{Si}_2$. The γ coefficient of 32 mJ/mol K^2 of UPT_2Si_2 starts to increase for $x > 0.05$, reaching 100 mJ/mol K^2 for 20% Ir, which indicates that the localization with $5f$ states out of the Fermi level is suppressed already for low Ir concentrations. Variations of lattice parameters a, c are non-monotonous, but the unit cell volume tends to decrease, which is compatible with the progress in $5f$ bonding. The Néel temperature T_N of the AF order decreases towards 6 K in ULr_2Si_2 . The diffuse X-ray scattering experiment at ESRF, ID28 beamline, reveals that the CDW state, developing gradually below 400 K, is still present for $x = 0.05$, where γ is still rather low, 33 mJ/mol K^2 . Further CDW development will be revealed at a forthcoming experiment.

This work was supported by the Czech Science Foundation under the grant # 25-16339S.

[1] R.A. Steeman et al., J. Phys.: Condens. Matter **2**, 4059 (1990).

[2] R.A. Steeman et al., J. Magn. Magn. Mater. **76&77**, 435 (1988).

[3] J. Lee et al., Phys. Rev. B **102**, 041112(R) (2020).

[4] M. Falkowski et al., Phys. Rev. B **101**, 174110 (2020).

9:00am AC+MI-ThM-5 Revisiting Unconventional Superconductivity in Thorium-Doped UBe_{13} , Yusei Shimizu, The University of Tokyo, Japan; **Mitja Krnel,** Andreas Leithe-Jasper, Markus König, Ulrich Burkhardt, Nazar Zarembo, Thomas Lühmann, Manuel Brando, Eteri Svanidze, Max Planck Institute for Chemical Physics of Solids, Germany

INVITED

The uranium-based superconductors have attracted considerable interest because of their unusual superconducting (SC) and normal-state properties. Among them, UBe_{13} (cubic O_h^6 , space group #226) has attracted much attention as a promising candidate for spin triplet superconductivity since the early stage [1]. The strong sample dependence of this superconductivity [2,3] and the lack of understanding of its $5f$ electronic state make the unraveling of superconductivity in UBe_{13} even more difficult. In particular, the non-monotonic Th concentration dependence of T_{SC} in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ and occurrence of SC double transition of heat capacity with a small amount of thorium ($0.019 < x < 0.045$) [4-8] are quite anomalous properties, and understanding this multiple SC phase diagram is important for elucidating the true nature of uranium spin triplet superconductors.

In this study, we focus on the low-temperature physics on thorium-doped UBe_{13} and we revisit their unusual SC and normal-state properties. We have fabricated polycrystals of $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ ($x = 0.01, 0.015, 0.02, 0.03, 0.04, 0.05, 0.07$) in an arc furnace. We determined their lattice constants from x-ray powder diffraction. Previous studies have found double transition of superconductivity at $0.019 < x < 0.045$ in heat capacity [5-8]. In order to clarify whether this double SC transition is intrinsic, we have performed detailed EDS (Energy Dispersive X-ray Spectroscopy), low-temperature heat-capacity and electrical resistivity measurements for $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$. The EDS results show that the distribution of Th is uniform within the crystals and that there is no heterogeneous $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ composition within the experimental accuracy. Furthermore, the low-temperature heat capacity results for $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ show that for $x = 0.02, 0.03, 0.04$ a second transition occurs in the SC state, while for $x = 0.015, 0.05$ only one SC transition is observed, which is consistent with previous studies. In our presentation, we will discuss the detail of SC H - T - x phase diagram and non-Fermi-liquid behavior in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$.

[1] H. R. Ott et al., Phys. Rev. Lett. **50**, 1595 (1983).; H. R. Ott et al., Phys. Rev. Lett. **52**, 1915 (1984). [2] A. Amon et al., Sci. Rep. **8**, 10654 (2018). [3] H. M. Voltz et al., Philos. Mag. **98**, 2003 (2018). [4] J. L. Smith et al., Physica B **135**, 3 (1985). [5] H. R. Ott et al., Phys. Rev. B **31**, 1651(R) (1985). [6] R. H. Heffner et al., Phys. Rev. Lett. **65**, 2816 (1990). [7] F. Kromer et al., Phys. Rev. Lett. **81**, 4476 (1998). F. Kromer et al., Phys. Rev. B **62**, 12477 (2000). [8] Y. Shimizu et al., Phys. Rev. B **96**, 100505(R) (2017).

Thursday Afternoon, September 25, 2025

2D Materials

Room 208 W - Session 2D+AQS+MI+NS+QS+TF-ThA

2D Materials: Magnets and Topological Phenomena

Moderators: Rafik Addou, The University of Texas at Dallas, Zhurun Ji, MIT

2:15pm 2D+AQS+MI+NS+QS+TF-ThA-1 Non-Local Transport from Magnetic Topological Superconductivity in 2D Fe-Chalcogenides, *Kenneth Burch*, Boston College **INVITED**

Magneto-Chiral topological superconductivity is a rare phase long pursued for error-free quantum computation. Its 1D chiral modes possess topologically protected long-range coherence well beyond that of the Cooper pairs, which could be fruitful for quantum transduction and low-temperature spin transport. While evidence for such modes is mounting, unambiguous signatures, such as non-local transport via co-tunneling, remain elusive. I will describe our realization of 1D chiral hinge modes mediating the direct tunneling of electrons from source to drain in FeTe_{0.55}Se_{0.45}. Specifically, I will discuss our evidence that the non-local tunneling signatures are decoherence-free and emerge from this material's combination of surface magnetism, bulk topology, and superconductivity. Time remaining, I will discuss how these advances can be used for Majorana Circuits and future efforts in cryogenic spintronics

2:45pm 2D+AQS+MI+NS+QS+TF-ThA-3 Integer and Fractional Chern Insulators in moiré MoTe₂, *Yihang Zeng*, Purdue University **INVITED**

The fractional Chern insulator (FCI), a lattice analogue of the renowned fractional quantum Hall state, was theorized to exist without external magnetic field. FCI provides a pathway towards novel topologically ordered quantum phases that are useful for decoherence-free quantum computation. Two-dimensional (2D) moiré materials, featuring strong correlation, non-trivial band topology and unparalleled tunability, stands as an ideal platform for realizing FCI. In this talk, I will first present our innovative optoelectronic detection method, which is capable of detecting the chemical potential in arbitrary 2D materials. Employing this new technique, we successfully observed an FCI and integer Chern insulator in the zero magnetic field limit in MoTe₂-based moiré materials. We further study the FCI and ferromagnetism as a function of twist angle.

3:15pm 2D+AQS+MI+NS+QS+TF-ThA-5 Conducting Scanned Probe Investigations of the Bismuthine Termination of Intrinsic Topological Superlattice Bi₂-Bi₂Se₃, *Lakshan Don Manuwelge Don, Mysidia Leff, Md. Sakauat Hasan Sakib*, Miami University; *Seth Shields*, The Ohio State University; *Joseph Corbett*, Miami University

Topological materials, characterized by symmetry-protected electronic states and robust surface conduction, represent a frontier in quantum materials research. Their non-trivial band topology enables dissipationless edge states, spin-momentum locking, and resilience to disorder, making them strong candidates for spin-orbit torque devices, magnetic field sensors, and polarized light detectors, to name a few. These properties have positioned topological materials as important material of interest as development of scalable quantum technologies grows.

In this study, we explore the atomic and electronic properties of the bismuthine-terminated topological semimetal Bi₂-Bi₂Se₃ using scanning tunneling microscopy (STM) and conductive atomic force microscopy (C-AFM). Bi₂-Bi₂Se₃ is an intrinsic superlattice material *s* comprised of a Bi₂Se₃ quintuple layer (QL) slab and a 2D Bismuthine (Bi₂) layer separated by a van der Waals gaps. The topological surface state on the 001 orientation depends on the terminating layer, with two distinct possible topologically protected surface states.

The unique step heights between the Bi₂Se₃ QL and Bismuthine layer enable termination characterization through careful step height analysis. Atomically resolved STM measurements on a Bismuthine terminated step reveal a distinct honeycomb lattice, while scanning tunneling spectroscopy (STS) captures a Dirac cone in local density of states centered at the Fermi level, in excellent agreement with angle-resolved photoemission spectroscopy (ARPES).

Using C-AFM under ambient conditions, we investigate force-dependent I-V spectroscopy. Utilizing step height analysis, we find a bismuthine terminated step and perform point spectroscopy. At low applied forces, differential conductance (dI/dV) spectra reveal a Dirac cone, mirroring STM results and confirming the presence of topologically protected surface states even under ambient conditions! As mechanical force increases, we observe a transition in transport behavior, from quantum tunneling to Ohmic conduction. Additionally, a voltage and force-dependent crossover from direct tunneling to Fowler-Nordheim tunneling is identified.

Our findings revealing the atomic structure and Dirac cone of the bismuthine termination in the topological semimetal Bi₂-Bi₂Se₃. Interestingly these feature are observable even under ambient condition. We find no degradation with time, freshly grown sample versus those that have sat for months give the same results.

3:30pm 2D+AQS+MI+NS+QS+TF-ThA-6 Local Spectroscopy Study of Gate-controlled Energy Gap in Monolayer 1T'-WTe₂, *Tiancong Zhu*, Purdue University; *Zehao He*, University of California at Berkeley; *Michal Papaj*, University of Houston; *Samuel Stolz*, Department of Physics, University of California, Berkeley; *Tianye Wang*, *Canxun Zhang*, *Yan-Qi Wang*, *Joel Moore*, *Zi Qiang Qiu*, *Feng Wang*, *Michael Crommie*, University of California at Berkeley

The interplay between strong correlation and topology can lead to intriguing quantum phases of matter. In monolayer 1T'-WTe₂, the non-trivial topology gives rise to the quantum spin Hall insulator (QSHI) phase, characterized by helical 1D edge states surrounding the insulating 2D bulk. While experimental evidences support quantized conductance through the 1D helical edge states, the nature of the insulating bulk, whether attributed to spin-orbit coupling or strong correlation, remains under debate. Here, we employ scanning tunneling microscopy and spectroscopy (STM/S) on gate-tunable 1T'-WTe₂ devices to shed light on this problem. Our samples are fabricated using a combination of molecular beam epitaxy (MBE) and van der Waals (vdW) stacking technique, which allows us to synthesize high-quality monolayer 1T'-WTe₂ films on a gate tunable graphene field effective transistor supported by hBN. Gate-dependent STS reveals a substantial energy gap in 1T'-WTe₂ at its charge neutrality, which diminishes when the Fermi level is tuned into either the conduction or valence band. STS across the sample edges shows that the edge states persist at all gate voltages, while Fourier transform-STM measurement in the bulk further shows the evolution of the bulk band structure at different carrier densities. We will compare our experimental data with existing theoretical models, such as the SOC-induced gap and the proposed excitonic insulator phase, and suggest future experimental directions to further elucidate the origin of the energy gap.

3:45pm 2D+AQS+MI+NS+QS+TF-ThA-7 Exploring Moiré Magnetism in Twisted Two-Dimensional Magnets, *Liuyan Zhao*, University of Michigan **INVITED**

Moiré superlattice emerges from the interference between two mismatched atomic lattices, and it has led to tremendous success in designing and tailoring the electronic states in two-dimensional (2D) homo- and hetero-structures. Yet, the power of moiré superlattice in controlling the spin degree of freedom and thus modifying the magnetic states is much less explored. Only very recently after the development of 2D magnet research, there have been a few experimental attempts in realizing moiré magnetism in twisted 2D magnet homo-structures. In this talk, I will show our recent effort in studying magnetic phases in twisted double bilayer chromium triiodide (CrI₃) and progressive steps towards realizing moiré magnetism. Noting that bilayer CrI₃ is a layered antiferromagnet and that any homogeneous stacking of two bilayers necessarily produces zero magnetization, we have revealed, in twisted double bilayer CrI₃, an unexpected net magnetization showing up at intermediate twist angles and its accompanied noncollinear spin textures. I will show the optical spectroscopy signatures of this twist-induced magnetic phase, then discuss its dependence on twist angle, external magnetic field, and temperature.

Actinides and Rare Earths

Room 207 A W - Session AC+MI-ThA

Early Career and Rising Stars

Moderators: Krzysztof Gofryk, Idaho National Laboratory, *Evgeniya Tereshina-Chitrova*, Charles University, Prague, Czech Republic, *Itzhak Halevy*, Ben Gurion Uni. Be'er Sheva, *Edgar Buck*, PNNL

2:15pm AC+MI-ThA-1 Combinatorically Estimating the Orbital Occupancy of Actinides using an Entropic Approach, *Miles Beaux*, *Benjamin Heiner*, Los Alamos National Laboratory **INVITED**

Predicting material properties in *f*-block elements, especially actinides, is complicated by their complex electronic structures, such as multiconfigurational ground states and strong correlation effects. These structures arise from large electron degrees of freedom, posing challenges in modelling their behavior. A non-integer orbital occupancy representation describes the superposition mixing of multiple near-energy degenerate configurations. This representation generalizes by approximation to

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established ground states in elements with simpler electronic structures and enables an over-approximation of entropy for multiconfigurational ground state structures. A complementary combinatorial approach applies Hund's rule constraints to establish an under-approximation of entropy. Together, these methods bracket entropy limits, providing insights into electronic configurations that most significantly contribute to the multiconfigurational ground states of actinide elements to a low order approximation. Under an energy degeneracy assumption weighted by configuration permutations, calculations iteratively refine the contributing configurations, yielding low-order orbital occupancy estimates that align with experimental data and theoretical models. (LA-UR-25-22711)

2:45pm AC+MI-ThA-3 Applications of Scanning Tunneling Microscopy in Heavy Element Studies, Benjamin Heiner¹, Miles Beaux, Los Alamos National Laboratory

Scanning Tunneling Microscopy and Spectroscopy (STM/S) are powerful techniques for investigating atomic, molecular, and surface properties. At Los Alamos National Laboratory, a specialized instrument designed to contain and probe samples containing heavy elements (i.e. actinides) allows us to study of the most uncharacterized elements on the periodic table. This capability has facilitated new insights into the electronic structure of plutonium oxides, intermetallics, and complexes. Using temperature-resolved STS, we can directly and continuously measure the total density of states of these materials across the Fermi energy, addressing a critical gap in experimental plutonium data. These advancements provide valuable information for understanding the electronic behavior of plutonium, with implications for fundamental science and nuclear materials research. Additionally, our ongoing efforts aim to apply these techniques to molecular complexes containing a single actinide atom, enabling both STM imaging and localized STS probing of individual actinide atoms. LA-UR-25-22710

3:00pm AC+MI-ThA-4 Electronic Structure of Uranium-Based Ferromagnet UPS, Sabin Regmi, Idaho National Laboratory; *Alexei Fedorov,* Lawrence Berkeley National Laboratory; *Dariusz Kaczorowski,* Polish Academy of Sciences, Poland; *Peter Oppeneer,* Uppsala University, Sweden; *Krzysztof Gofryk,* Idaho National Laboratory

Strongly correlated *f*-electron systems often exhibit intriguing properties such as unconventional superconductivity and heavy fermion behaviors. Particularly in 5*f*-electron systems, the understanding of the relation between *f* electrons and observed physical properties has been a challenge due to their duality. Here, we present an angle-resolved photoemissions spectroscopy (ARPES) study of uranium-based ferromagnet UPS, supported by density-functional theory calculations. Measurements carried out at on and off-resonant photon energies suggest strong contribution from U 5*f* in the vicinity of the Fermi level and *c-f* hybridization. The results reveal the Fermi surface, underlying electronic structure in this system, and the nature of the 5*f* electrons in this ferromagnetic material. This work provides a valuable platform to advance the fundamental understanding of the 5*f* electronic structure in uranium-based and actinide materials in general.

***This work is supported by Idaho National Laboratory's laboratory directed research and development (LDRD) program and the US Department of energy (DOE) Basic Energy Sciences, Materials Sciences and Engineering Division.*

3:15pm AC+MI-ThA-5 The Plutonium Auto-reduction Reaction, Predicting Kinetics, and Assessing Impacts to Surface Science Measurements, Timothy Gorey, Daniel Rodriguez, Sarah Hernandez, Los Alamos National Laboratory

Plutonium is a fascinating and difficult material to measure in vacuum systems due to its auto-catalytic reduction of higher oxides into plutonium sesquioxide (Pu₂O₃). This "auto-reduction" reaction complicates surface science measurements aiming to understand higher oxides, because these layers, when exposed to vacuum as is required for many surface-sensitive techniques (e.g. X-ray Photoelectron and Auger Electron spectroscopies (XPS and AES), and Secondary Ion Mass Spectrometry (SIMS)) spontaneously converts into sesquioxide. This presentation will discuss an XPS-focused study into the nuances of high oxide (PuO₂) surface analysis and propose likely mechanistic origins for the auto-reduction reaction as well as methods to predict the chemical progression of the surface.

3:30pm AC+MI-ThA-6 Magnetic Properties of UP₂ Probed by High-Magnetic Field, Volodymyr Buturlim², Sabin Regmi, Idaho National Laboratory; *Rubi KM,* High Magnetic Field Laboratory, Los Alamos National Laboratory; *Dariusz Kaczorowski,* Polish Academy of Sciences, Poland; *Neil Harrison,* Los Alamos National Laboratory; *Krzysztof Gofryk,* Idaho National Laboratory

Due to its complex tetragonal crystal structure, with three distinct uranium sites, UP₂ stands out among other uranium dipnictides such as UAs₂, USB₂, and UBi₂. UP₂ exhibits antiferromagnetic ordering at ambient pressure with *T_N* = 204 K and an effective moment of $\mu_{\text{eff}} = 2.29 \mu_B/\text{U}$. The neutron scattering experiment indicates that the ordered moment is parallel to the [0 0 1] direction and equals 2.0 μ_B/U . There is, however, a lack of information regarding the magnetic properties of UP₂ in high magnetic fields, particularly concerning its magnetic phase diagram. Here we present detailed experimental and theoretical studies of the magnetic properties of oriented high-quality single crystals of UP₂. The measurements were performed at the High Magnetic Field Laboratory, Los Alamos National Laboratory, using pulsed magnetic fields up to 60 T. We will discuss details of the obtained phase diagram and its relationships to the localization/delocalization of 5*f*-electrons in this material.

3:45pm AC+MI-ThA-7 Properties of Carbon-Related Point Defects in Plutonium Oxides, Andrew Rowberg, Kyoung Eun Kweon, Scott Donald, Lawrence Livermore National Laboratory

Carbon is a ubiquitous impurity; therefore, investigating how it incorporates in materials is vital for understanding their properties, stability, and performance. Here, we evaluate the formation of carbon impurities in the most common stoichiometric plutonium oxides, PuO₂ and Pu₂O₃, which has not been systematically studied to date. We use hybrid density functional theory calculations to compute formation energies and other relevant properties of carbon species in various configurations. We find the stability of carbon defects to be strongly dependent on charge state and oxygen coordination environments. Accordingly, these properties can influence the phase evolution between PuO₂ and Pu₂O₃. We also evaluate the interactions between carbon and other defects present in these oxides.

4:00pm AC+MI-ThA-8 Vacancy-mediated Conduction Tunability in Epitaxial SmN, Kevin Vallejo, Volodymyr Buturlim, Zachery Cresswell, Brelon May, Brooke Campbell, Idaho National Laboratory; *Bobby Duersch,* University of Utah; *Krzysztof Gofryk,* Idaho National Laboratory

We establish the relationship between native N vacancies, introduced through varying growth parameters, and electronic properties of SmN thin films grown via molecular beam epitaxy grown on MgO(001). We show substrate temperature having a larger impact on V_{\$}_N_{\$} formation during growth than the ratio of Sm to N atoms. We observe a transition from insulating to conducting behavior of the film over a range of two orders of magnitude, from highly resistive to highly conductive. X-ray photoelectron spectroscopy and room temperature electrical transport results confirm the rapid degradation of the film despite the presence of capping layers. A ferromagnetic feature in the film is shown through low-temperature resistivity measurements to be the onset of ferromagnetic behavior. These promising results indicate a path forward in the epitaxy of versatile materials able to provide monolithic integration of different electronic behaviors without the associated strain brought about by heteroepitaxial integration of dissimilar materials. The integration between SmN and several transition metal nitride compounds has the potential to unlock new electronic and spintronic device architectures with low strain barriers.

¹ JVST Highlighted Talk

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² JVST Highlighted Talk

Actinides and Rare Earths

Room 207 A W - Session AC+MI-FrM

Spectroscopy, Spectrometry, 5f Behavior and Forensics

Moderators: Ladislav Havela, Charles University, Czech Republic, Gertrud Zwirnagl, Technical University Braunschweig, Alison Pugmire, LANL

8:15am AC+MI-FrM-1 Exploring the Surface Chemistry of Plutonium using ToF-SIMS, Sarah Hernandez, Los Alamos National Laboratory INVITED

Plutonium metal is highly reactive by immediately forming an oxide layer when exposed to air and quickly forming a hydride when exposed to hydrogen. The fundamental understanding of the impact of impurities and defects on the effect of oxidation and corrosion of Pu is limited in both experimental and theoretical studies. Time-of-Flight Secondary Ion Mass Spectroscopy (ToF-SIMS) is a unique surface science technique that is highly sensitive to the first 1-2 monolayers of the surface (<1nm) and can detect all isotopes (including hydrogen) at parts-per-million levels, which gives a comprehensive survey of surface constituents. This technique also provides a structural and reactivity, chemisorption versus physisorption, information and complements other surface science techniques, such as X-ray photoelectron spectroscopy (XPS). In general, ToF-SIMS may provide a more in-depth analysis of surface constituents that otherwise might not be detected or deconvolute from a complex XPS spectra. A newly installed ToF-SIMS nanoToF 3 at LANL uses a 30 kV Bi³⁺ liquid metal ion gun as the primary ion source and has a mass resolution of 12,000 ($\Delta m/m$), thus providing a new level of mass resolution and sensitivity on Pu surfaces that was not previously achieved. I will show recently collected ToF-SIMS results of hydrogen and oxygen gas reactions on alpha-Pu and 2 at. % Ga stabilized δ -Pu surfaces and how they compare with other.

8:45am AC+MI-FrM-3 HERFD vs XAS: The Case for Equivalence, J G Tobin, U. Wisconsin - Oshkosh

The advent of new, powerful, highly efficient, multi-component, X-ray monochromators used in the detection of tender x-rays has revolutionized spectroscopic investigations of the 5f electronic structure. All of the new experiments are, in essence, variants of X-ray Emission Spectroscopy (XES), where the improved monochromatized detection, applied to novel specific decay pathways, plays a key role. In HERFD (High Energy Resolution Fluorescence Detection) a type of Resonant Inelastic X-Ray Scattering (RIXS), the monochromatized XES detection allows the performance of a scattering experiment with vastly improved resolution. It is argued here that HERFD devolves into a higher resolution version of X-Ray Absorption Spectroscopy (XAS). It has been shown that the M_4 and M_5 spectra are essentially direct measurements of the j-specific ($5f_{5/2}$ and $5f_{7/2}$) Unoccupied Density of States (UDOS), which can be directly correlated with the UDOS from Inverse Photoelectron Spectroscopy (IPES) and Bremsstrahlung Isochromat Spectroscopy (BIS). [1-3] Furthermore, a remarkable level of agreement is achieved between a model based upon the UDOS of Th and a series of HERFD and IPES/BIS results with various 5f occupation levels. [4-6] Finally, the historical record of XAS will be examined, demonstrating the success of various resonant decay schemes as measures of the underlying XAS.

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2. J. G. Tobin, H. Ramanantoanina, C. Daul, S.-W. Yu, P. Roussel, S. Nowak, R. Alonso-Mori, T. Kroll, D. Nordlund, T.-C. Weng, D. Sokaras, "The Unoccupied Electronic Structure of Actinide Dioxides," *Phys. Rev. B* 105, 125129 (2022)
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9:00am AC+MI-FrM-4 Room Temperature H₂ Dosing on Polished α -Pu Surfaces with XPS, Daniel Rodriguez¹, Timothy Gorey, William Ponder, Alessandro Mazza, Raymond Atta-Fynn, Sarah Hernandez, Los Alamos National Laboratory

Plutonium (Pu) is a complex element with an interesting electronic structure, and it is also a material of great importance for both nuclear energy and security. To better understand its interaction with gases, surface analysis of the alpha (α) variant provides valuable insight when coupled with a technique such as X-ray photoelectron spectroscopy (XPS). Different core electron orbitals may be probed, and binding energies from emitted electrons provide information on the local chemical state, i.e., degree of oxidation, reduction, or carbonization within the α -Pu.

Here we investigated the effect of hydrogen (H₂) gas dosing of α -Pu surfaces, which reacts and forms plutonium hydride (PuH₂) at temperatures >100 °C. By slowing the kinetics at room temperature, we may witness H₂ dynamics on native α -Pu surfaces, and view how Pu materials such as oxidized and carbonized forms evolve with H₂ exposure. In addition, we present our findings from density functional theory (DFT) validating experimental observation. To provide an example, **Fig. 1** shows a plot of various Pu 4f spectra. In red, metal α -Pu is observed after having been sputtered to remove both surface contaminants and the native oxide layer. The defining metal feature in the $4f_{7/2}$ peak is seen at ~422.2 eV. Next, the sample was dosed with H₂ gas for 198 Langmuir (L) (blue line), and then the exposure was increased (green line) until reaching 396 L. A clear reduction in the signal's intensity is seen in both the 5/2 and 7/2 metal peaks. Secondly, the 7/2 satellite shows an increase in signal, which is indicative of surface passivation. Clearly, more is needed to know what these H₂ induced changes signify, and this presentation will show additional spectra from the O 1s, C 1s, and the Pu valence band, along with DFT to contextualize the ongoing mechanisms of H₂ with the α -Pu surface.

9:15am AC+MI-FrM-5 Ab Initio Modeling of Hydrogen Interaction with the Surface of α -Pu, Raymond Atta-Fynn, Sarah Hernandez, Los Alamos National Laboratory

Hydrogen (H) reacts strongly with plutonium (Pu) metal, with the reaction primarily initiated on the metal surface. However very little is known theoretically about H dynamics on the surface of the ambient temperature phase of Pu, namely α -Pu. In this regard, we carried out calculations on H interactions with the α -Pu(020) surface using density functional theory-based geometry optimizations and *ab initio* molecular dynamics at 300 K. Molecular H₂ dissociated spontaneously on the metal surface at room temperature, resulting in atomic H chemisorption. The energy barriers to diffusion of the chemisorbed H from the surface into the subsurface and bulk layers were modeled using accelerated *ab initio* molecular dynamics. The magnitudes of the energy barriers to H diffusion in relation to hydride formation will be discussed.

9:30am AC+MI-FrM-6 A Novel Lexan-Aerogel Detector for Fission Track Analysis for Advancing Nuclear Forensics, Itzhak Halevy, Rami Babayew, Yaacov Yehuda-Zada, Ben Gurion University Be'er Sheva, Israel; Galit Bar, Soreq Nuclear Research Center, Israel; Noam Elgad, Mark Last, Ben Gurion University Be'er Sheva, Israel; Jan Lorincik, Research Centre Řež, Czechia; Itzhak Orion, Ben Gurion University Be'er Sheva, Israel; Shay Dadon, Nuclear Research Center Negev, Israel; Aryeh M. Weiss, Bar Ilan University, Israel; Galit Katarivas Levy, Ben Gurion University Be'er Sheva, Israel

Fission track analysis is a technique employed in nuclear forensics to identify and examine fission isotopes. This technique is specific for small samples in the range of a few picograms or to analyze bigger samples and check for homogeneity.

In the old Lexan detector, the tracks are pretty close, and that limits much the ability to count the tracks and analyze the length of the tracks. The main target of the fission track is to locate the fission ions in between a lot of other isotopes. The located fission ions could be transferred to other techniques like ICP-MS for further analysis. Better separation between tracks and analysis could lead to showing the yield of fission products, which is specific to every fission isotope. The yield fission products are two humps on the graph that are equal in area. One hump is around A=95, 135; in the length of the track histogram, the two humps look different due to

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the difference in dE/dx of the different energies. The light elements hump looks narrow, and the heavy elements hump looks wide; still, the area of those humps is equal. We created a novel detector for fission track analysis with the Lexan-modified detector.

This innovative detector exhibits more dispersion of fission tracks. In this innovative approach, we adhered aerogel to the Lexan. The aerogel has a low absorption coefficient; hence, it does not substantially obstruct the fission products in the detector. The incorporation of aerogel modifies the geometric configuration, enlarges the dimensions of the fission track stars, and increases the separation between individual tracks, as seen in Fig. 1 in the supplement. A fission track star of a size of 150 microns can reach 350 microns with the aerogel configuration. Given that the fission products are distributed isotopically while the aerogel is two-dimensional, it is necessary to employ stereoscopic projection to facilitate their integration. An illustration of this enhancement of the fission track star is seen in Fig. 1, where the dimensions of the fission track star are greater and the tracks are widely spread. The newly developed analytical program, **Finder**, may utilize a 2D representation of the fission track star. Whether an actual star or a simulated star, of a fission track to conduct analysis and provide 3D evaluations, therefore illustrating the fission yield of the fission isotope. The analysis of the fission track star is shown in Fig. 2, supp. The fission track analysis of ^{235}U star in that software is depicted in Fig. 3 supp.

Fission track length before the detector and in it are shown in that figure of the fission track analysis of ^{235}U star.

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