2D Materials Technical Group
Room 303 - Session 2D+MI-TuA

2D Materials: Heterostructures, Twistronics, and Proximity Effects

Moderators: Francesca Tavazza, National Institute of Standard and Technology, **Suyang Xu**, Harvard University

2:20pm **2D+MI-TuA-1 Strategies for Controlling Structure and Magnetic Texture in 2D Magnets**, ***Frances Ross,*** MIT; *J. Klein,* MIT, USA **INVITED**

Electronic devices that incorporate two dimensional (2D) materials often require contacting or patterning the 2D layer for their fabrication. This is particularly important when we aim to build exciting new nanoscale magneto-electric devices based on 2D magnetic materials. CrSBr is such a 2D magnet that shows stability in air, giving this material a key advantage in practical device fabrication compared to other, less stable 2D magnets. Here we discuss strategies for controlling the structure and properties of CrSBr and related 2D magnets at the local, nanoscale level. We approach this via transmission electron microscopy, based on promising results for other 2D materials. We first discuss local control of structure. We find that electron beam irradiation in a scanning transmission electron microscope (STEM) induces a surprising structural change, where Cr atoms migrate into the van der Waals gap to create a new phase with layer direction (and, in theory, magnetization) perpendicular to the initial layers. The ability to modulate the magnetization direction deterministically is of great interest for quantum devices. Furthermore, since we find that defects in this material can be optically active and correlated with magnetic order and phases, the ability to use STEM to control individual defects will ultimately help to achieve precise control of the material's properties. We next discuss strategies for contacts. For other 2D materials, the 2D/3D contact resistance is known to improve when the contact layers have fewer grain boundaries. We therefore focus on epitaxial growth of metals and other 3D crystals onto the 2D surface. We show how *in situ* TEM imaging helps to clarify the growth mechanisms and interface structures formed during single crystal or heterostructured metal growth on graphene, hBN and transition metal dichalcogenides. We then explore how nucleation and epitaxy phenomena play out for pristine and patterned CrSBr and other 2D magnets. Overall, we conclude that atomic level structural and chemical modification are crucial for understanding properties and designing devices that use the exciting properties of the new 2D magnets. We suggest that rapidly advancing *in situ* TEM instrumentation promises exciting future opportunities where nanoscale growth and patterning create complex devices based on 2D materials.

3:00pm **2D+MI-TuA-3 Bidirectional Phonon Emission in van der Waals Heterojunctions During Ultrafast Charge Transfer**, ***Aditya Sood,*** Stanford University

Photoinduced charge transfer in van der Waals heterostructures occurs on ultrafast timescales of order 100 fs, despite the weak interlayer coupling and momentum mismatch. Little is understood about the microscopic mechanism behind this fast process and the role of the lattice in mediating it. Here, using femtosecond electron diffraction, we directly visualize lattice dynamics in photoexcited heterostructures of WSe_2/WS_2 monolayers. Following selective excitation of WSe_2, we measure unexpectedly concurrent heating of both WSe_2 and WS_2 on a 1 picosecond timescale, corresponding to an "apparent" interlayer thermal conductance that is >100x larger than that due to phonons alone. Using first-principles calculations, we identify a fast channel, involving an electronic state hybridized across the heterostructure, enabling phonon-assisted interlayer transfer of photoexcited electrons. Phonons are emitted in both layers on femtosecond timescales via this channel, consistent with the simultaneous lattice heating observed experimentally. Taken together, our work indicates strong electron-phonon coupling via layer-hybridized electronic states – a novel route to control energy transport across atomic junctions.

4:20pm **2D+MI-TuA-7 Understanding Structural, Chemical, and Number of Layer-Dependent Properties in 2D Lateral and Vertical Structures for Subsequent Optical Measurements**, *U. Kaiser,* ***Michael Mohn,*** University of Ulm, Germany **INVITED**

Properties of 2D materials can manifest at very different length scales. Charge density waves, magnetic ordering, inter- and intralayer excitons are studied also to understand their atomistic origin. Moreover, starting from exciting properties of low-twist angle graphene, twisted transition metal dichalcogenides are now explored, whereby the future of moiré superlattices is also dependent on reliable twist angle control. In addition, interfaces of transition metal dichalcogenide heterostructures such as Janus monolayers or lateral heterostructures have potential applications in optoelectronics, however very critical for carrier and exciton transport is that they are atomically sharp.

Here we use the low-voltage- spherical and chromatic aberration-corrected transmission electron microscope to measure and introduce structural and chemical variations in free-standing 2D materials on the atomic scale. In-situ and ex-situ optical measurements are performed and together with quantum-mechanical calculations their atomic-structure-based properties are understood.

We first report on advances in TEM sample preparation both for oxygen-sensitive TMDs as well as describe our sample platform to relate atomic defects in 2D materials produced by TEM with subsequent measurements in stacked devices. Then we describe studies on electron-beam-induced defects and observe the migration paths and associated property changes in a variety of single and few-layered free-standing structures of transition metal di-chalcogenides (TMDs) and transition metal phosphorus tri-chalcogenides (TMPTs). We also investigate the twist-angle-dependent moiré pattern formation in bilayers of TMDs by theoretical prediction-followed TEM experiments. From the comparison of monolayer, bi-layer and 2° twisted bilayer experimental images, we determine twist-angle-induced inhomogeneous stacking-related localized strain in the layers as well as the twist-angle-induced changes of the interlayer excitons located in the low-loss range of the EELS spectrum. We further report on the number of layer-dependent electronic properties of Pt-dichalcogenide family. We also show proof-of-principle experiments in which we transfer electron-exposed TMD flakes from a TEM grid to arbitrary substrates and measure the produced defects in photoluminescence and transport measurements. Moreover, the investigated lateral heterostructures show near-atomically sharp junctions with a typical extent of 3 nm for the covalently bonded $MoSe_2$-WSe_2 interface, determined by high-resolution transmission electron microscopy. This explains the considerably narrowed optical transition linewidth in the photoluminescence, reflectance contrast and Raman spectroscopy.

5:00pm **2D+MI-TuA-9 Determination of Band Offsets in Semiconductor Heterostructures (2D/3D) by Using XPS**, ***Mohamed Nejib Hedhili,*** *NG, B. Ooi,* King Abdullah University of Science and Technology, Saudi Arabia

Electrical and optical properties of heterojunction semiconductors are heavily influenced by the relative alignment of their energy band edges at the interface [1]. That is why the knowledge of this alignment is crucial for the design of heterostructure devices. In this regard, high-resolution X-ray photoemission spectroscopy (HR-XPS) has been shown to measure the valence band offset of heterojunction semiconductors quite accurately [2]. In this report, we present a study devoted to the characterization of 2D/ 3D heterojunction semiconductor materials using a myriad of techniques including HR-XPS, scanning transmission electron microscopy (STEM), atomic force microscopy (AFM), micro-Raman, absorbance, and microphotoluminescence spectroscopy.

The samples for this study were prepared by depositing an epitaxial GaN ($In_{0.15}Al_{0.85}N$) thin layer with molecular beam epitaxy (MBE) on chemically vapor deposition (CVD) grown single-layer (SL) MoS_2/c-sapphire (WSe_2/c-sapphire) substrates. The formation of SL of MoS_2 (WSe_2)was crucial to device properties and hence was confirmed by using both STEM and AFM techniques. HR-XPS analysis of samples was performed in two-steps to measure the valence band discontinuity for GaN ($In_{0.15}Al_{0.85}N$) / SL of MoS_2 (WSe_2) heterojunction interface. In first step, the core level binding energies with respect to the valence band maximum in both GaN ($In_{0.15}Al_{0.85}N$) and MoS_2 (WSe_2) bulk films were measured. Second, the subsequent measurements on the separation between Ga (In) and Mo (W) core levels for GaN ($In_{0.15}Al_{0.85}N$) thin layer grown SL-MoS_2 (SL-WSe_2)was measured. The valence band and conduction band offset values are determined.

The band alignment parameters determined here provide a route toward the integration of group III nitride semiconducting materials with transition metal dichalcogenides (TMDs) for designing and modeling their heterojunction-based electronic and optoelectronic devices.

References:

[1] U. Gnutzmann and K. Clausecker, Appl. Phys. 3, 9 (1974).

[2] J. C. Bernède, L. Cattin, P. Predeep, XPS Study of the Band Alignment at the Interface

ITO/CuI, Technology Letters 1 (1), 2 (2014).

5:20pm **2D+MI-TuA-10 Investigation of a Novel Layer-by-Layer Growth Methodology for Surface Metal-Organic Frameworks**, ***Nicholas Stucchi***, Clarkson University

The fabrication of highly ordered and crystalline surface-confined metal-organic frameworks (SURMOFs) has garnered interest in applications such as catalysis, gas storage, and gas separation. At present, the state of the art of SURMOF fabrication is a layer-by-layer (LBL) growth, wherein a functionalized substrate undergoes sequential immersions in solutions of the desired metal or ligand. This LBL strategy is commonly performed using gold substrates modified with carboxylate-terminated self-assembled monolayers (SAMs) which act as an initial nucleation site for the metal cluster. Careful control over deposition conditions and reaction times results in the formation of crystalline SURMOFs with tunable thicknesses. However, this technique is limited to substrates that can undergo the necessary SAM functionalization as well as being highly sensitive to the deposition conditions. I will discuss the development of a new LBL methodology that utilizes a pre-formed covalent-organic framework (COF) on the surface of highly oriented pyrolytic graphite (HOPG) as the template for SURMOF growth. The COF template should have the same geometry and a lattice parameter close to that of the desired SURMOF to minimize the strain of the first few layers. As such, COF-366-Zn was chosen as the template for UiO-67, which has a 3% lattice mismatch between the COF template and MOF. The COF was reacted with isonicotinic acid (INA) in which the pyridine axially coordinates to the zinc centers of the COF and the exposed carboxylate moieties of the INA serve as nucleation sites for the zirconium acetate clusters of UiO-67. The SURMOF was formed following several sequential reactions in the cluster and benezene-1,4-dicarboxylic acid (BDA) ligand solutions. The COF and initial INA binding will be characterized by scanning tunneling microscopy (STM) and Fourier-Transform Infrared (FTIR) spectroscopy. Atomic force microscopy (AFM) will be used to characterize the LBL growth of UiO-67. The crystallinity of the SURMOF will be determined using diffraction techniques.

Magnetic Interfaces and Nanostructures Division
Room 330 - Session MI-TuA

Topological Insulator Heterostructures

Moderators: Axel Enders, University of Nebraska-Lincoln, Germany, **Valeria Lauter**, Oak Ridge National Laboratory

2:20pm **MI-TuA-1 Evidence of Antiferromagnetic Coupling between Topological and Magnetic Insulators**, ***Leonid Rokhinson***, Purdue University **INVITED**

Exchange interaction between topological and magnetic insulators enables local control of topologically protected surface states by lithographically shaping magnetic materials. In previous works ferromagnetic exchange has been successfully realized. We report an experimental evidence of antiferromagnetic exchange between a topological insulator Bi2Se3 and a magnetic insulator EuSe. Spin-polarized neutron reflectometry reveals reduction of in-plane magnetic susceptibility up to 25K, well above the Neel temperature of the bulk EuSe. A combination of SQUID magnetometry and transport measurements indicates an antiferromagnetic interfacial exchange coupling that opens an energy gap in topological surface states. High temperature local control of topological surface states with zero net magnetization opens new opportunities for the design of electronic, spintronic and quantum computation devices, ranging from quantization of Hall conductance at zero fields to spatial localization of non-Abelian excitations in superconducting topological qubits.

3:00pm **MI-TuA-3 Infrared Magnetospectroscopy of Magnetic Topological Insulator Heterostructures**, ***Badih Assaf***, University of Notre Dame **INVITED**

Topological insulator (TI)-magnetic insulator(MI) heterostructures are employed to achieve efficient electrical switching of magnetization, owing to the spin-charge coupling enabled by the helical Dirac surface states of the TI. These surface states are however prone to the breaking of time-reversal symmetry by magnetism. A magnetic insulator deposited on the surface of a TI is thus hypothesized to induce a gapping of the Dirac surface states. This energy gap is challenging to measure using common surface spectroscopies since the surface is buried under the magnetic insulator. We develop TI-MI heterostructures based on the topological crystalline insulator $Pb_{1-x}Sn_xSe$ (x>0.2) and a magnetic insulator EuSe. The high mobility achieved in PbSnSe (>10000cm^2/Vs) allows us to measure the magnetic proximity induced gap using magnetooptical Landau level spectroscopy up to 17.5T at 4.5K. We find the upper bound of this gap to be close to 20meV. We confirm the coexistence of this gapped Dirac state with magnetism using neutron reflectometry. This result has important implications on the potential of TCIs for devices that rely on proximity between a magnet and a topological material, such as spintronic switching devices, since it is evident that the gap remains smaller than the Fermi energy and likely should not influence the spin-charge coupling characteristics of the TI.

4:20pm **MI-TuA-7 Topological States in the van der Waals Magnet $MnBi_2Te_4$: from 3D to 2D**, ***Hendrik Bentmann***, Wuerzburg University, Germany **INVITED**

The interplay of topology and magnetism is a route to spintronic applications based on dissipationless charge and spin transport. The van der Waals material MnBi2Te4 naturally combines strong spin-orbit interaction and local magnetic moments, opening a playground for the study of magnetic topological phenomena. In its 3D bulk phase, MnBi2Te4 forms an antiferromagnetic topological insulator [1]. Related topological states are also realized in the modular modular (Bi2Te3)n(MnBi2Te4) series, where the insertion of non-magnetic spacer layers yields modified magnetic properties [2,3]. In the main part of the talk, I will present more recent efforts to achieve 2D MnBi2Te4 layers using molecular beam epitaxy [4]. A particular focus will be on the realization of a single MnBi2Te4 monolayer, which shows robust 2D ferromagnetism below Tc = 14 K. As demonstrated by angle-resolved photoemission (ARPES), a monolayer MnBi2Te4 placed on Bi2Te3 induces a large magnetic gap in the topological surface state, promising realization of a robust quantum anomalous Hall state.

[1] M. M. Otrokov, I. I. Klimovskikh, HB, et al., Nature 576, 416 (2019).
[2] R. C. Vidal,..., HB, A. Isaeva, et int., Phys. Rev. X 9, 041065 (2019).
[3] R. C. Vidal, HB, et al., Phys. Rev. Lett. 126, 176403 (2021).
[4] P. Kagerer, C. Fornari,..., HB, F. Reinert et int., J. Appl. Phys. 128, 135303 (2020).

5:00pm **MI-TuA-9 Rashba-Type Splitting of the Au(110) Surface State: A Combined Inverse and Direct Photoemission Study**, ***Markus Donath***, *K. Ritter,* University of Münster, Germany; *K. Miyamoto, T. Okuda,* Hiroshima University, Japan

The Shockley surface state located at Y on the (1×2)-reconstructed Au(110) surface is predicted to exhibit a Rashba-type spin splitting. Previous photoemission experiments searched for this splitting but it could not be resolved yet. In order to uncover a possible splitting, the unoccupied surface state on Au(110) is examined with spin- and angle-resolved inverse photoemission, whereas Na-covered Au(110) allows for investigation of the now occupied surface state by means of spin- and angle-resolved direct photoemission [1]. Our data show clear spin splittings in the order of 100 meV with a sign reversal at Y in the surface state's in-plane spin components which is characteristic for a Rashba-type behavior. Furthermore, we deduce an effective mass of $m^* = (0.27 \pm 0.02)m_e$ and a Rashba parameter of $\alpha_R = (0.46 \pm 0.04)$ eVÅ from direct photoemission measurements.

[1] K.T. Ritter, K. Miyamoto, T. Okuda, and M. Donath, Phys. Rev. B **104**, L161101 (2021).

5:20pm **MI-TuA-10 Spin-Polarized Resonant Tunneling - a New Tool for Sensing and Manipulating Magnetism on the Atomic Scale**, ***Anika Schlenhoff***, Department of Physics, University of Hamburg, Germany **INVITED**

Atomic-scale magnetism as found in ultrathin films with non-collinear spin textures or in moiré structures of 2D-hybrid materials raise expectations for potential spintronic applications, demanding for atomic-scale, spin-sensitive, but yet robust techniques for sensing and manipulation. Spin-polarized image-potential states (sp-IPS) are unoccupied electronic states in the vacuum gap between a probe tip and a magnetic sample. They exhibit the same local spin quantization axis as the surface, even when it rotates on the atomic scale [1]. In a spin-polarized scanning tunneling microscopy (SP-STM) setup, spin-polarized electrons can tunnel resonantly from the magnetic tip via the sp-IPS into the surface, resulting in a magnetic image contrast mediated by these states.

Our SP-STM experiments on non-collinear spin textures in ultra-thin films demonstrate that the spin-polarized resonant electron tunneling via sp-IPS allows for atomic-scale spin-sensitive imaging in real space at tip-sample distances of up to 8 nm, providing a loophole from the hitherto existing dilemma of losing spatial resolution when increasing the tip-sample distance in a scanning probe setup [2]. Technically applicable to a variety of

material systems, our spin-polarized resonant tunneling studies on iron- and cobalt-intercalated graphene show that the IPS's sensitivity to the interlayer coupling of graphene to the metallic substrate and the resulting local IPS spin-polarization can be used for probing the graphene magnetism in the moiré unit cell [3].

When the electrons relax from the sp-IPS into the surface, a spin-transfer torque (STT) is exerted on the sample that can be exploited for thermally-assisted magnetization switching [4]. On ferromagnetic nano-islands, we observe IPS that are bound to the rim of the islands, causing a spatial modulation of the IPS electron spin-polarization above the uniformly magnetized nanoislands. As I will show, tunneling locally through the sp-IPS can be used to tune the spin-polarization of the resonant tunneling current and thus the STT for current-induced magnetization switching, using the sp-IPS as a spin-filter [5].

Utilizing the local spin-polarization of IPS via spin-polarized resonant tunneling, our approach qualifies for a spin-sensitive read-write technique with ultimate lateral resolution, potentially opening a pathway towards future technical applications.

[1] A. Schlenhoff *et al.*, Phys. Rev. Lett. **123**, 087202 (2019).

[2] A. Schlenhoff *et al.*, Appl. Phys. Lett. **116**, 122406 (2020).

[3] A. Schlenhoff *et al.*, in preparation.

[4] A. Schlenhoff *et al.*, Phys. Rev. Lett. **109**, 097602 (2012).

[5] A. Schlenhoff *et al.*, in preparation.

Magnetic Interfaces and Nanostructures Division
Room Ballroom A - Session MI-TuP

Magnetic Interfaces and Nanostructures Poster Session

MI-TuP-1 Microscopy with Momentum and Imaging Spin-Filter (Au/Ir), ***Marten Patt***, *M. Escher, N. Weber, T. Kuehn, M. Merkel,* FOCUS GmbH, Germany

Momentum Microscopy is a new technique in surface science, in which the momentum (or the real-space) distribution of photoelectrons is projected onto an image plane by using a photoemission electron microscope (PEEM) column. In case of momentum imaging the k_x-k_y plane can be energy-filtered by a double-hemispherical electrostatic analyzer (IDEA) to achieve a monochromatic momentum distribution. The ability of the method to map the complete angular distribution of photoelectrons is quite successfully used for photoemission orbital tomography (e.g. at the NanoESCA in Trieste [1]). An innovative extension of this technique is to use the monochromatic electron distribution behind the double-hemispherical analyzer for 2D imaging spin-filtering. Early experiments with a NanoESCA were performed with a W(100) single crystal as electron-mirror with spin-polarization dependent reflection [2], but only the step to a Au passivated Ir (100) single crystal with long-term stable scattering properties paved the way to a broadly based scientific useability.

We will show results from our two first commercial Au/Ir 2D Imaging Spin Filters. Pre-characterizations of the Au/Ir crystal were done with LEED and a Ferrum-Detector setup [3] to find optimal preparation conditions and scattering energies for a high single-point figure-of-merit (with Sherman function >60% and Reflectivity >1%) [4]. Spin-filtered images of magnetic domains show that along the diameter of the field of view (e.g. 36 µm) more than 100 separate image points can be resolved. This increases the effective 2D figure-of-merit of this spin-filter by nearly four orders of magnitude compared to single-channel spin-detectors.

MI-TuP-2 Investigating the Magnetic Properties of the Co-Tb Phase Diagram, ***Sydney Harrington***, *B. Wilfong,* United States Naval Academy*; D. Heiman,* Northeastern University*; M. Jamer,* United States Naval Academy

There has been recent interest in Co-Tb compounds for its potential use in various spintronic applications, since it has been shown to display spin-orbit torque while maintaining a low magnetic moment. While spintronic devices have been made from ferromagnetic materials, the low moment properties have been enticing for researchers since it will enable devices to become more efficient, perform at high speeds, and less corruptable. Compounds made from Co-Tb are especially interesting since Tb and Co combine a 4*f* and 3*d* orbitals, which allows for interesting magnetic coupling causing for a net magnetic moment or angular momentum to go to zero. The 3*d*-4*f* interactions are interesting to study since rare-earth elements normally have a large moment and large coercivity associated with hard ferromagnets and when a 3*d* transition element is added, these materials have an unpredictable magnetization properties.

The Co-Tb system has been shown to be advantageous in a variety of low-moment ferrimagnetic applications. In previous work, it was found that the Co-Tb system is ideal for potential spin-orbit torque with overall low-magnetization at room temperature. While various studies have shown the properties of the compound, there was not much known about the structure due to the amorphous nature in thin films. In our work, we have synthesized bulk ingots of Co-Tb in a variety of stoichiometric ratios and found that the Co_7Tb_2 is the phase responsible for the low-moment properties in various works. Through the structural and magnetic investigations of multi-phase ingots, we have determined the magnetic properties hexagonal and rhombohedral polymorphic phases of the Co_7Tb_2 structure and the effect of the other binary phases on the interfaces on the desired phase.

2D Materials Technical Group
Room 303 - Session 2D+EM+MI+NS+QS-WeM

2D Materials: Quantum and Symmetry-Protected States

Moderators: Thomas Michely, University of Cologne, Germany, **Frances Ross**, Massachusetts Institute of Technology

8:00am **2D+EM+MI+NS+QS-WeM-1 Semi-High Throughput Investigation of 2d Materials: Anomalous Quantum Confinement Effect and Spectral Properties**, ***Francesca Tavazza***, *K. Choudhary,* National Institute of Standard and Technology **INVITED**

Materials with van der Waals-bonding exhibit quantum confinement effect, in which the electronic bandgap of the three-dimensional (3D) form is lower than that of its two-dimensional (2D) counterpart. However, the possibility of an anomalous quantum confinement effect (AQCE) exists, where the bandgap trend is reversed. In this work, we computationally identify materials with AQCE. Using density functional theory (DFT), we compute ≈1000 OptB88vdW (semi-local functional), ≈50 HSE06 and ≈50 PBE0 (hybrid functional) bandgaps for bulk and their corresponding monolayers, in the JARVIS-DFT database. OptB88vdW identifies 65 AQCE materials, but the hybrid functionals only confirm such finding in 14 cases. Electronic structure analysis shows that AQCE is often characterized by the lowering of the conduction band in the monolayer and related changes in the p_z electronic orbital contribution. In addition to AQCE, the JARVIS-DFT contains IR and Raman spectra for many 2D materials. Properties of such spectra will be discussed as well.

8:40am **2D+EM+MI+NS+QS-WeM-3 Dry Patterning Chemically Sensitive Quantum Materials**, ***Joseph Benigno***, *Q. Zou, C. Cen, L. Li,* West Virginia University

Accurate, repeatable patterning of quantum material-based electronic devices is desirable for electrical transport measurements. However, the most common method, photolithography, can degrade, or even damage, chemically sensitive quantum materials during fabrication. Here we introduce a new dry-patterning method for device fabrication with lateral etching resolution down to ~30 µm. The new method utilizes a tabletop computer numerical control (CNC) router machine to gently etch patterns into thin films, leaving behind the desired device or devices on the substrate. We create Hall bars with conductive channel widths of 30, 60, and 120 µm from ~20 layer FeTe-capped superconducting single layer FeSe/$SrTiO_3$ systems. Transport measurements show the same zero resistance T_c of 10 K for the Van der Pauw (vdP) geometry and all Hall bar structures. However, the onset temperature T_{onset} is the largest at 28K for the vdP geometry, and decreases with the width of the Hall bar to 13K for the 60 µm device. Our method provides a new time-saving, cost-effective, and chemical-free strategy for fabrication of devices from quantum materials.

This research is supported by DOE DE-SC0021393.

9:00am **2D+EM+MI+NS+QS-WeM-4 Electron Transport and Charge Sensing in Strongly Coupled Quantum Dot Array in Silicon**, ***Fan Fei***, *J. Wyrick, P. Namboodiri, J. Fox,* NIST*; E. Khatami,* SJSU*; R. Silver,* NIST

Atomically precise donor-based quantum devices in silicon are fabricated using STM lithography, which has become a promising platform for solid state quantum computation and analog quantum simulation. Lattices of dopant-based quantum dots have unique advantages in simulating strongly correlated Fermionic systems of real atomic lattice sites because of their naturally occurring ion-cores which make them the Fermi-Hubbard sites in the Silicon Vacuum. Understanding electron transport and charge configuration in a smaller array is critical to using these arrays to simulate larger systems and explore various condensed matter physics phenomena such as superconductivity in the future. This talk will focus on the electron transport in the strongly coupled regime where the electrons delocalize across small N x N dot arrays. Numerical simulations for charge stability diagrams and transport properties show qualitatively agreement with our experiments. We apply rf reflectometry on a SLQD and use it as charge sensor for probing the electron configuration within the array.

9:20am **2D+EM+MI+NS+QS-WeM-5 Observation of the Layer Hall Effect in Topological Axion Antiferromagnet $MnBi_2Te_4$**, ***Suyang Xu***, Harvard University **INVITED**

While ferromagnets have been known and exploited for millennia, antiferromagnets were only discovered in the 1930s. The elusive nature indicates antiferromagnets' unique properties: At large scale, due to the absence of global magnetization, antiferromagnets may appear to behave like any non-magnetic material; At the microscopic level, however, the opposite alignment of spins forms a rich internal structure. In topological antiferromagnets, such an internal structure leads to a new possibility, where topology and Berry phase can acquire distinct spatial textures. We study this exciting possibility in an antiferromagnetic Axion insulator, even-layered $MnBi_2Te_4$ flakes. We report the observation of a new type of Hall effect, the layer Hall effect,where electrons from the top and bottom layers spontaneously deflect in opposite directions.

Reference:

A. Gao, et al."Layer Hall effect in a 2D topological axion antiferromagnet." *Nature* 595, 521 (2021).

11:00am **2D+EM+MI+NS+QS-WeM-10 Phonon Limited Mobility and Phonon Drag in h-BN Encapsulated Monolayer and AB-stacked Bilayer Graphene**, ***Vasili Perebeinos***, University at Buffalo

We report the electrical transport in h-BN encapsulated AB-stacked bilayer graphene theoretically and experimentally. Using the perturbation theory within the tight-binding model approach, we identify the dominant role of the shear phonon mode scattering on the carrier mobility in AB-stacked graphene bilayer at room temperature. The shear phonon mode is absent in free-standing monolayer graphene, which explains high mobilities in monolayer devices fabricated under similar conditions resulting in minimal Coulomb impurity scattering. At temperatures above 200K, the surface polar phonon scattering from the boron-nitride substrate contributes significantly to the experimental mobilities of 15,000 -20,000 cm^2/Vs at room temperature and carrier concentration n~10^{12} cm^2 reported here. A screened SPP potential for a dual gated bilayer and transferable tight-binding model allows us to predict mobility scaling with temperature and bandgap for both electrons and holes in agreement with the experiment Phys Rev. Lett. 128, 206602 (2022).

The resulting electron-SPP coupling is used to predict that, by exploiting the strong coupling of their electrons to surface polar phonons, van der Waals heterostructures can offer a suitable platform for phonon sensing, capable of resolving energy transfer at the single-phonon level. The geometry we consider is one in which a drag momentum is exerted on electrons in a graphene layer, by a single out-of-equilibrium phonon in a dielectric layer of hexagonal boron nitride, giving rise to a measurable induced voltage. Our numerical solution of the Boltzmann Transport Equation shows that this drag voltage can reach a level of a few hundred microvolts per phonon, well above experimental detection limits. Furthermore, we predict that drag voltage should be largely insensitive to the mobility of carriers in the graphene layer and increase the temperature up to at least 300 K, offering the potential of a versatile material platform for single-phonon sensing.

11:20am **2D+EM+MI+NS+QS-WeM-11 Exciton Physics at the Atomic Scale**, ***Daniel Gunlycke***, U.S. Naval Research Laboratory

Descriptions of excitons in pristine semiconducting crystals usually rely on the hydrogen model adopted for excitons. Owing to the weak screening in monolayer transition-metal dichalcogenides, however, the electron and hole separation in the strongest bound excitons is on the atomic scale, necessitating atomistic treatment. In this presentation, we present a minimalistic exciton model that accounts for the lattice and the spin-orbit and exchange interactions, thus making this model appropriate across the spectrum from Wannier to Frenkel excitons. Using this model, we show that the exciton lifetimes could be extended by transitioning the excitons into excitonic dark states. Longer exciton lifetimes could make these materials candidates for applications in energy management and quantum information processing.

11:40am **2D+EM+MI+NS+QS-WeM-12 Weyl Semimetals with Low-Symmetry Crystal Structure for Generating Out-of-Plane Oriented Spin Current**, ***Simranjeet Singh***, Carnegie Mellon University **INVITED**

Weyl semimetals (WSMs), such as WTe_2 and $MoTe_2$, host plethora of novel phenomena that are highly relevant for quantum spintronics, namely: Dirac type dispersion, strong spin-orbit coupling (SOC), Fermi arcs, and helical spin-momentum locked surface and bulk states. WSMsprovide a distinct opportunity to obtain highly efficient and unconventional charge to spin conversion owing to strong SOC, symmetry breaking, and these topology-based phenomena. On the other hand, spin-orbit torque (SOT) driven deterministic control of the magnetic state of a ferromagnet with perpendicular magnetic anisotropy is key to next generation spintronic applications including non-volatile, ultrafast, and energy efficient data

storage devices. But field-free deterministic SOT switching of perpendicular magnetization remains a challenge because it requires an out-of-plane oriented spin current, which is not allowed in conventional spin source materials such as heavy metals and topological insulators due to the system's symmetry. The exploitation of low-crystal symmetries WTe_2 and $MoTe_2$ offers a unique approach to achieve SOTs with unconventional forms[1]. In this work, I will discuss our experiments to realize field-free deterministic magnetic switching of a perpendicularly polarized van der Waals magnet employing an out-of-plane spin current generated in layered WTe_2, which is a quantum material with low-symmetry crystal structure[2]. I will also discuss our experiments aimed at achieving field-free SOT switching of semiconducting and insulating FMs using spin current in WSMs. Our work establishes transition metal dichalcogenides, with lower symmetry crystal structure, as an appealing spin source material for future spin-orbit torque related magnetic memory technologies.

[1]. MacNeill, D. *et al.* Control of spin–orbit torques through crystal symmetry in WTe_2/ferromagnet bilayers. *Nature Physics***13**, 300-305, (2017).

[2]. Kao, I-H *et al.* Deterministic switching of a perpendicularly polarized magnet using unconventional spin–orbit torques in WTe_2. Nature Materials (2022). https://doi.org/10.1038/s41563-022-01275-5

Actinides and Rare Earths Focus Topic
Room 318 - Session AC+LS+MI-WeM

Magnetism, Electron Correlation, and Superconductivity in the Actinides/Rare Earths

Moderators: James G. Tobin, University of Wisconsin-Oshkosh, **Ladislav Havela**, Charles University, Czech Republic, **Gertrud Zwicknagl**, Technical University Brauschweig, Germany

8:00am **AC+LS+MI-WeM-1 The Relation between Crystal Chemistry and Superconductivity in Actinide-Based Superconductors***, Eteri Svanidze,* Max Plank Institute, Dresden, Germany **INVITED**

Crystallographic features play an important role in the physical and chemical properties of a given solid-state material. In particular, actinide-based systems exhibit a wide range of properties – from unconventional superconductivity to peculiar magnetic orders. In this talk, I will highlight some of the old and new actinide-based superconductors, in which a delicate interplay between chemistry and physics is observed. A comprehensive characterization of properties of UBe_{13} has revealed a deep interrelation between the physical and chemical features. Notably, single crystals of this material tend of include many defects which have a dramatic effect on superconducting state [1]. Motivated by this issue, an alternative method of studying intrinsic properties is investigated [2-4]. By creating a micro-scale device, it is possible to measure intrinsic superconductivity of UBe_{13}, whichhas so far remained out of reach [4]. The properties of UBe_{13} are compare to those of other actinide-based superconductors – UTe_2 [5] and $Th_4Be_{33}Pt_{16}$ [6] – in which a strong coupling of lattice and superconducting properties is observed. By studying these systems, it is possible to expand the understanding of crystal chemistry of solid-state materials, while simultaneously providing an insight into which crystallographic parameters impact the physical properties of a given solid-state material.

[1] A. Amon *et al.*, "Tracking aluminium impurities in single crystals of the heavy-fermion superconductor UBe_{13},"*Sci. Rep.***8**, 10654 (2018)

[2] E. Svanidze *et al.*, "Revealing intrinsic properties of UBe_{13}", *in preparation* (2022)

[3] A. Amon *et al*., "Interplay of atomic interactions in the intermetallic semiconductor Be_5Pt", *Angew. Chem. Int. Ed.***58**, 2 (2019).

[4] I. Antonyshyn *et al*., "Micro-scale device - an alternative route for studying the intrinsic properties of solid-state materials: case of semiconducting TaGeIr", *Angew. Chem. Int. Ed.***59**, 2 (2020)

[5] E. Svanidze *et al.*, "Intrinsic crystal structure of UTe_2", *in preparation* (2022)

[6] P. Kozelj *et al.*, "A noncentrosymmetric cage superconductor $Th_4Be_{33}Pt_{16}$", *Sci. Rep.***11**, 22352 (2021)

8:40am **AC+LS+MI-WeM-3 Revealing The Beauty of Uranium Compounds:the UMB_4 (M=V, Cr, Fe, Co, Mo, W, Re, Os) and UFe_xSb_2 Cases***, Antonio Pereira Gonçalves,* Instituto Superior Técnico, Univ. Lisboa, Portugal **INVITED**

Uranium-based phases with d-metals show a vast and rich variety of physical properties, some of them unique, which are mainly driven by the large 5*f* spin-orbit coupling and the degree of delocalization of the 5*f* states. The understanding of such behaviors is critical for the advance of Solid State Sciences and can lead to the discovery of new phenomena and ground states. In this talk, examples of uranium compounds with unusual properties are presented, the focus being on UMB_4 materials and UFe_xSb_2 alloys.

UMB_4 (M=V, Cr, Fe, Co, Mo, W, Re, Os) compounds are quite notable, crystallizing in the $YCrB_4$-type (for V, Cr, Fe, Co) and $ThMoB_4$ type (for Mo, W, Re, Os) polymorphs. Both structures are closely related, containing the same number of atoms in the unit cell and similar coordination polyhedral, which allows the comparison of their properties as a function of the d-metal. Their electronic structure is influenced by a strong hybridization between the B-2*p* states and both the *d*-metals *d*-states and the uranium (5*f*) valence band states. This avoids the formation of narrow bands with high density of states at the Fermi level, even if the 5*f*-*d* hybridization is weak, i.e. for late d-metals. Hence, the explored UMB_4 compounds are weak Pauli paramagnets, with a density of states at Fermi level of only ~4 times higher than for α-U. Those crystalizing in the $ThMoB_4$ structure show anomalous lattice vibrations, in particular the presence of low-energy Einstein modes, which can lead to superconducting ground states.

UFe_xSb_2 alloys crystallize in the $HfCuSi_2$ type structure and present a remarkable ground state evolution with Fe concentration. $UFe_{0.5}Sb_2$ shows a non-Fermi liquid behavior, with a change from metallic to semiconductor-upon applying magnetic field. In contrast, $UFe_{0.7}Sb_2$ (and, in a minor scale, $UFeSb_2$), shows an increase in electrical resistivity with decreasing temperature under zero magnetic field, but for high magnetic fields a metallic behavior is established. First principles calculations indicated that in $UFeSb_2$ some bands cross the Fermi level, but they are relatively steep and, as a result, the density of states at Fermi level is low, which supports the semi-metallic character of this composition. In the case of $UFe_{0.5}Sb_2$, a higher concentration of bands at the Fermi level, some of them relatively flat, exist, which cause a sudden step-shaped drop of density of states just above Fermi level. The magnetic field application can raise the Fermi level into the pseudo-gap, which is probably the reason for the change from a metallic-like to a semiconductor-like behavior.

11:00am **AC+LS+MI-WeM-10 Uranium Hydrides Revisited***, Ladislav Havela,* Charles University, Faculty of Mathematics and Physics, Czechia*; D. Legut,* VSB Technical University Ostrava, Czechia*; J. Kolorenc,* Institute of Physics, Czech Academy of Sciences, Czechia

Uranium hydrides are not only materials important for understanding of fundamental aspects of actinides. They are also relevant for nuclear technologies as well as for specific hydrogen storage tasks, as e.g. storing of tritium in nuclear fusion devices. Electronic structure of uranium hydrides (α- and β-UH_3, UH_2) reflects two contradictory tendencies. One is a charge transfer from U towards H, the other is a stability of the *f* shell. The latter is illustrated also on the case of rare earth hydrides with anomalous rare earth elements, such as Yb or Eu, which surprisingly retain their lower valence state, i.e. with a higher *f*-occupancy. The transfer is thus realized in U by the 6*d* and 7*s* electrons, which become noticeably depleted, but the 5*f* occupancies remain high. The concomitant reduction of the 5*f*-6*d* hybridization contributes, together with the volume expansion, to pronounced ferromagnetism of the U hydrides with Curie temperatures far above 100 K. We compare individual hydrides described by the GGA+*U* calculations, yielding numbers on the charge transfer or spin and orbital moments, with relevant experimental data. Interesting feature obtained from calculations is that the two different U sites in β-UH_3 have the respective magnetizations non-collinear. The same calculations yield also phonon dispersion relations, which are used to determine the temperature dependence of phonon contribution to heat capacity. The agreement in the paramagnetic range is so good that it allows to determine both the magnetic entropy and changes of the Sommerfeld coefficient between the ground state and paramagnetic state, the latter being twice higher. The position of the U hydrides on the map of localization-delocalization dichotomy will be discussed.

The work was supported by the Czech Science Foundation under the grant No. 21-09766S.

11:20am **AC+LS+MI-WeM-11 Towards a Better Understanding of the Rkky Interaction in Ce- and Yb-Based Compounds: Anisotropies from Cef Effects and Fermi Surfaces**, ***Gertrud Zwicknagel,*** Institut für Mathematische Physik, Germany*; V. Zevin,* The Racah Institute of Physics, Israel

We calculate the influence of Crystalline Electric Field (CEF) effects and the Fermi surface topology on the indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) exchange interaction between local 4f moments in Ce- and Yb-compounds. Starting from a periodic Anderson model, we show that the anisotropy of the CEF ground state manifests itself in the anisotropy of the effective exchange coupling constant J(R,R') between two moments at sites R and R'. If spin-orbit effects among the conduction states can be neglected, the interaction between two Kramers doublets is isotropic in pseudo-spin space. Adopting the Stationary Phase approach, we derive the asymptotic form of the exchange constant for arbitrary Fermi surfaces. We evaluate the anisotropy of the RKKY interaction in tetragonal Ce- and Yb-122 compounds for realistic model bands. The influence of spin-orbit interaction among the conduction electrons is discussed.

11:40am **AC+LS+MI-WeM-12 Mechanically Forced Tuning of Interactions in Tetragonal 221 Intermetallics**, ***Petr Král,*** *J. Prchal,* Charles University, Czech Republic*; J. Kaštil,* Czech Academy of Sciences, Czech Republic*; D. Daisenberger,* Diamond Light Source, UK*; D. Staško,* Charles University, Czech Republic

Family of R_2T_2X intermetallics adopting the tetragonal Mo_2FeB_2-type crystal structure represents the group of anisotropic systems with potential to exhibit geometrical frustration effects due to the Shastry-Sutherland-like arrangement of *R*-atoms in the aa-planes alternated by usually non-magnetic *T*-*X* planes along the crystallographic *c*-axis. Crucial for resulting magnetic behavior are the interatomic distances affecting the exchange interactions between individual ions. Especially interesting may be the compounds close to the empirical boundary of crystal structure stability since, based on available data, the lowest observed unit-cell volume among Mo_2FeB_2-type materials is about $V_{f.u.,crit}$ = 200 Å^3.

The unique tool allowing to act directly on the interatomic distances without changes of chemical composition is mechanical pressure. Employing the high-pressure techniques, it is possible to influence all lattice-related physical properties including e.g. the magnetic interactions and electronic structure. Especially in the strongly anisotropic compounds, the hydrostatic pressure effect depends on the compressibility of individual lattice parameters. This is the case, when the uniaxial pressure may represent the opportunity to act selectively in chosen crystallographic directions and thus to provide the insight into the layout of interactions within the crystal lattice.

Herewith, we are presenting the results of systematical study of pressure impact on the R_2T_2X compounds. Special attention is paid to Ce_2Pd_2In considered to be one of the most interesting systems among R_2T_2X family. It represents the rare example of Ce-based ferromagnet, however the ground state is reached through the antiferromagnetic interphase. High sensitivity of magnetic behavior to hydrostatic pressure has been revealed showing the preference of antiferromagnetic phase at lower pressures followed by the Kondo-like anomaly formation and suppression of magnetic ordering at pressures around 4 GPa. Based on the results of high-pressure XRD experiment, the hydrostatic pressure acts more significantly within the basal plane. Indeed, different response to the uniaxial pressure applied along the *c*-axis was observed.

Compounds with the unit-cell volume closest to $V_{f.u.,crit}$ belongs to the series R_2Cu_2In. Among them the ferromagnets Dy_2Cu_2In (exhibiting the signs of spin-reorientation at lower temperatures) and Tm_2Cu_2In (with signs of spin-glass behavior) were investigated. Concerning the later one, the pressure was found to support the frustration effects responsible for the spin-glass properties as the unit-cell volume tends to reach the $V_{f.u.,crit}$.

12:00pm **AC+LS+MI-WeM-13 Effects of O_2 Growth Pressure on the Magnetization of $LaMnO_3$-$SrTiO_3$ Thin Films**, ***Ghadendra Bhandari,*** West Virginia University*; R. Trappen,* University of Waterloo*; N. Mottaghi, M. Holcomb,* West Virginia University

Magnetic properties of epitaxial $LaMnO_3$ thin films grown on $SrTiO_3$ substrate have been studied. Thin films are grown at various oxygen pressure using pulsed laser deposition and deposition has been monitored by reflection high energy electron diffraction (RHEED) to verify layer-by-layer growth. Bulk stoichiometric $LaMnO_3$ exhibits A-type antiferromagnetic order, whereas thin films show a ferromagnetic FM phase. Structure and magnetic properties have been characterized by X-ray diffractometry (XRD), X-ray reflectivity (XRR) and vibration sample magnetometry (VSM). The thin films are showing ferromagnetic (FM) phase. Depth dependent magnetization studied by polarized neutron beam reflectivity found that magnetization is higher near interfacial region and a magnetically dead region lies at the surface. We have observed c-lattice correlated magnetization, which is minimized at 30mTorr O_2 pressure.

Magnetic Interfaces and Nanostructures Division
Room 330 - Session MI-WeM

Spin Landscape I (Magnetic Structures in Real and Momentum Space)

Moderator: Mikel Holcomb, West Virginia University

8:00am **MI-WeM-1 Voltage Controlled Néel Vector Rotation in Zero Magnetic Field**, ***Christian Binek,*** *A. Mahmood,* University of Nebraska-Lincoln*; W. Echtenkamp,* University of Minnesota*; M. Street, J. Wang, S. Cao, T. Komesu, P. Dowben, P. Buragohain, H. Lu, A. Gruverman, A. Parthasaraty, S. Rakheja,* University of Nebraska-Lincoln*; J. Weaver, J. Lynn,* NIST-Gaithersburg **INVITED**

Voltage-controlled switching of remnant magnetic states paves the way towards ultra-low power and non-volatile spintronics. In this presentation, I report on a decade-long journey which took us from isothermal electric switching of exchange bias with the help of simultaneously applied electric and magnetic fields to pure voltage-controlled antiferromagnetic spintronics in zero magnetic field and at CMOS compatible temperatures. Nonvolatile Néel vector reorientation in the absence of an applied magnetic field, *H*, is demonstrated at CMOS compatible temperatures in prototype device structures which exploit the multi-functional properties of thin films of boron (B) doped Cr_2O_3. Boundary magnetization associated with the Néel vector orientation serves as state variable which is read via magnetoresistive detection in a Pt Hall bar adjacent to the B: Cr_2O_3 film. Switching of the Hall voltage between zero and non-zero values implies Néel vector rotation by 90-degrees in agreement with the observed voltage dependent contrast in magnetic force microscopy images. Piezo force microscopy data suggest the presence of polar nanoregions which give rise to uniform polarization in the presence of an applied electric field. The polarization is accompanied piezoelectric straining which, via magnetoelastic coupling, changes the magnetic anisotropy and thus the Néel vector orientation from out of plane to in-plane and back. B-doping enhances the Néel temperature, T_N, of pure chromia. Annealing of the device further increases the T_N-enhancement (up to 500K) at the interface between the Hall bar and the B: Cr_2O_3 surface via thermally activated B-diffusion. The diffusion mechanism is confirmed via cold neutron depth profiling measurements. Robust switching is demonstrated post-annealing for temperature as high as 400K. Theoretical modeling estimates switching speeds of about 100 ps making B: Cr_2O_3 a promising multifunctional single-phase material for energy efficient nonvolatile CMOS compatible memory applications.

We acknowledge financial support by NSF/EPSCoR RII Track-1: Emergent Quantum Materials and Technologies, OIA-2044049. The research was performed in part in the NNF: NNCI and the NCMN, supported by NSF under ECCS:2025208, and the NRI.

8:40am **MI-WeM-3 Discovering Magnetic Mechanisms in Room-Temperature Metallic Antiferromagnet Fe_3Ga_4**, ***Michelle Jamer,*** *B. Wilfong,* United States Naval Academy*; D. Baigutlin, O. Miroshkina, V. Buchelnikov, V. Sokolovskiy,* Chelyabinsk State University, Russian Federation*; G. Stephen, A. Friedman,* Laboratory for Physical Sciences*; R. Barua,* Virginia Commonwealth University*; B. Barbiellini,* LUT University, Finland*; D. Heiman,* Northeastern University **INVITED**

Recently, Fe_3Ga_4 has garnered much interest due to its unique magnetic structure which supports two unique magnetic transitions- one metamagnetic transition at low temperature to an antiferromagnetic helical spin structure (~70 K), and a second transition between the antiferromagnetic state to ferromagnetic state (~360 K). Due to the helical spin structure of the intermediate antiferromagnetic state, metallicity is not prohibited leading to the possibility of a room temperature metallic antiferromagnet, which is attractive for potential spintronic devices. Of particular interest, is fully understanding the magnetic phase diagram and the Fe-Fe coupling in the lattice which gives rise to the special helical ordering. In our work, we have prepared bulk ingots and single crystals of this compound to understand the magnetic coupling of Fe_3Ga_4 to determine the baseline properties. Through adding pressure to the system, we have found that we are able to change the transition temperatures of

the magnetic states- leading to an increase in the low temperature transition and a decrease in the high temperature transition. In these measurements, we have found that we are able to tune the high temperature transition to room temperature by adding pressure -while the structure retains its metallicity. A full discussion on the baseline Fe_3Ga_4 as well as its changes with pressure will be presented, including theoretical calculations supporting the magnetic structure as detected via magnetometry measurements.

11:00am **MI-WeM-10 Irradiative Control of FeRh's Metamagnetic Phase Change Under Three-Dimensional Spatial Confinement Interrogated by Polarized Neutron Scattering***, **Steven Bennett**,* Naval Research Laboratory **INVITED**

Phase change materials have been a staple for a wide array of memory technologies for many decades. The promise of antiferromagnetic electronics has pushed the envelope past using a straight forward resistive phase change, to the realm of high speed spin flipping, incommensurate spin density waves and magnonics which can propagate without a local magnetic anisotropy to hinder spin rotation. In this new memory paradigm we will need to develop a new set of materials and understanding of spin physics from which to build this next generation of high speed and low energy loss devices. In this seminar I will provide an overview of my teams recent discoveries on how the metamagnetic transition in FeRh, from antiferromagnetic to ferromagnetic ordering, can be controlled and triggered using low energy heavy and light ion irradiation [1][2], as well as joule heating in confined wire device geometries at high switching speeds. We interrogate the complexities of these spin systems using polarized neutron scattering, revealing highly localized effects of ion irradiation in the films and uncover a new effect for metamagnetic spin dynamics which could be pivotally important for modern antiferromagnetic spintronics.

[1] S.P. Bennett et. Al., Coatings, 11(6), 661, (2021)

[2] C. D. Cress et. Al., ACS Appl. Mater. Interfaces 13, 1, 836–847, (2021)

2D Materials Technical Group
Room 303 - Session 2D+EM+MI-WeA

2D Materials: Charge Density Waves, Magnetism, and Superconductivity

Moderators: An-Ping Li, Oak Ridge National Laboratory, **Xiaomeng Liu**, Princeton University

2:20pm **2D+EM+MI-WeA-1 Tunable Electronic Structure and Correlations in Quasi-Freestanding Monolayer Transition Metal Disulfides***, **Thomas Michely,*** Universität zu Köln, Germany **INVITED**

In situ reactive molecular beam epitaxy using single crystal graphene as a substrate enables to grow transition metal disulfides as quasi-free standing monolayers under well controlled conditions. Thereby access to their undisturbed electronic properties of as well as to those of their intrinsic defects is provided.

A non-invasive technique to shift the chemical potential in semiconducting transition metal disulfide layers like MoS_2 or WS_2 through p- and n-type doping of graphene is presented, while graphene remains a well-decoupled 2D substrate. These shifts induce giant band gap renormalizations, insulator-to-metal to insulator transitions and affects the metallic states in mirror twin boundaries.

Electronic correlations are known to be strong for dimensional reasons in transition metal dichalcogenide monolayers and often give rise to charge density waves and other competing electronic phases. The dependence of charge density waves on the environment and its layer dependence are investigated for several transition metal disulfides. We show that monolayers of VS_2 realize a CDW which stands out of our expectations. It displays a full CDW gap residing in the unoccupied states of monolayer VS_2 and the CDW induces a topological metal-metal (Lifshitz) transition. Non-linear coupling of transverse and longitudinal phonons is essential for the formation of the CDW and the full gap above the Fermi level.

Lastly we will focus to mirror twin boundaries in MoS_2 and investigate how to determine quantized polarization charges on these domain boundaries, whether they are subject to Fermi level pinning and how shifts in the Fermi level change screening of their line charge.

Contributions to this work by Clifford Murray, Camiel van Efferen, Wouter Jolie, Jeison Fischer, Timo Knispel, Joshua Hall, Stefan Kraus, Felix Huttmann, Carsten Busse, Niels Ehlen, Boris Senkovskiy, Martin Hell, Alexander Grüneis, Hannu-Pekka Komsa, Arkady Krasheninnikov, Jan Berges, Erik van Loon, Arne Schobert, Malte Rösner, Tim Wehling, Nico Rothenbach, Katharina Ollefs, Lucas Machado Arruda, Nick Brookes, Gunnar Schönhoff, Kurt Kummer, Heiko Wende, Philipp Weiß, Fabian Portner, and Achim Rosch are gratefully acknowledged.

3:00pm **2D+EM+MI-WeA-3 Dopants Modulated Interplay of Charge Density Wave and Superconductivity in 2D VdW Layered ZrTe3***, **Xiao Tong,*** . Center of Functional Nanomaterials, Brookhaven National Laboratory*; Y. Liu, Z. Hu,* Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory*; D. Leshchev,* National Synchrotron Light Source II, Brookhaven National Laboratory*; X. Zhu, H. Lei,* Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory*; E. Stavitski, K. Attenkofer,* National Synchrotron Light Source II, Brookhaven National Laboratory*; C. Petrovic,* Condensed Matter Physics and Materials Science, Department, Brookhaven National Laboratory

Two-dimensional transition metal trichalcogenides ZrTe3 holds atomic chains in the crystal structure give rise to quasi one-dimensional (quasi 1D) conduction, and features the charge density wav (CDW) below TCDW ¼ 63 K and filamentary superconductivity below 2 K, Here, we report that superconductivity(SC) is enhanced as the consequence of suppressed CDW for Hf dopped ZrTe3, in contrast, SC is suppressed as the consequence of enhanced CDW for the Se dopped ZrTe. Our XPS and Raman studies suggested the suppressed CDW is due to Hf caused disorder in Te2-Te3 atomic chains, while the enhanced CDW is due to Se induced enhanced electron-phonon coupling in unperturbed periodicity of the Te2-Te3 chains, respectively.

3:20pm **2D+EM+MI-WeA-4 Magnetic Order in a Coherent Kondo Lattice in 1T/1H $TaSe_2$ Heterostructures***, W. Wan, **Rishav Harsh**, P. Dreher, S. Sajan,* Donostia International Physics Center , Spain*; A. Menino, I. Errea,* Centro de Física de Materiales (CSIC-UPV-EHU), Spain*; F. de Juan, M. Ugeda,* Donostia International Physics Center , Spain

Kondo lattice systems are of fundamental importance for our understanding of quantum criticality and unconventional superconductivity. At the heart of their complexity lies the competition between the opposing forces of Kondo screening and magnetic interactions, which is revealed at very low temperatures as the moments start behaving coherently and eventually determines the fate of the ground state. While our understanding of Kondo lattices has traditionally relied on technically challenging strongly correlated bulk f-electron systems, new light is being shed on the problem thanks to heterostructures of 2D transition metal dichalcogenides, which realize a tunable Kondo lattice platform in a simple material. Here, we study the 1T/1H-$TaSe_2$ heterostructure by high-resolution Scanning Tunneling Spectroscopy at 300 mK, and show a well resolved splitting of the Kondo peak, which increases monotonically in a nonlinear fashion in the presence of an out-of-plane magnetic field. This behavior is unexpected for a fully screened Kondo lattice, and it originates instead from a ground state with residual magnetic order, consistent with a Kondo coupling much below the critical point in the Doniach phase diagram.

4:20pm **2D+EM+MI-WeA-7 Structural and Magnetic Properties of Ultrathin $Cr_{(1+\delta)}Te_2$ Films Grown by Van Der Waals Epitaxy***, **Kinga Lasek,** P. Coelho,* University of South Florida*; P. Gargiani , M. Valvidares,* ALBA Synchrotron Light Source, Spain*; K. Mohseni , H. Meyerheim , I. Kostanovskiy,* Max Planck Institute of Microstructure Physics, Germany*; K. Zberecki,* Warsaw University of Technology, Poland*; M. Batzill,* University of South Florida

Over the past years, researchers have proved that the layered structure of transition metal dichalcogenides (TMDs) enables the synthesis of novel materials. Specifically, introducing extra transition metal atoms into the vdW gap of the TMDs host lattice leads to various structural, electrical, and magnetic properties modulations. Exploring the latter, in particular, aligns with a recent search for ferromagnetic 2D materials.

In this talk, we will explore the epitaxial growth, structural, surface, and magnetic properties of a bi- to few-layer thick $Cr_{(1+\delta)}Te_2$ ($0 < \delta < 1$)films that represent a group of self-intercalated TMDs materials. These materials demonstratewell-known ferromagnetic ordering and exist in different compositional phases that vary by the amount of Cr intercalated (δ) between $CrTe_2$ layers.By detailed compositional and structural characterization, using scanning tunneling microscopy (STM), and high-resolution Rutherford backscattering (HR-RBS) we will show that the amount of self-intercalated Cr atoms can be controlled by post-growth annealing. Such modified films are characterized by an increased T_c up to 190K, a coercive field being reduced from 0.5 T to 0.3 T, and an isotropization of the magnetic anisotropy confirmed by XMCD measurements.

Finally, we will demonstrate that ultrathin vdW films can be prepared with the ultimate limit of a single self-intercalated layer by vdW epitaxy. These vdW materials maintain their ferromagnetic properties with desirable out-of-plane anisotropy and thus are potential ferromagnetic 2D materials that can be combined in vdW heterostructures by a bottom-up growth process.

4:40pm **2D+EM+MI-WeA-8 Transition Metal Silicates as a Platform for Robust Single Layer, Two-Dimensional Ferromagnetism***, **Nassar Doudin,** K. Saritas,* Yale University*; P. Shafer, A. T. N'Diaye,* Lawrence Berkeley National Laboratory (LBNL)*; S. Ismail-Beigi, E. Altman,* Yale University

Two-dimensional (2D) materials have received extensive attention and rapid development since the discovery of graphene in 2004.[1] Magnetism in two dimensions has long been at the heart of theoretical, experimental, and technological advances, where great efforts have been made to realize magnetism in 2D materials.[2] To date, 2D magnetic materials have been obtained via exfoliation from bulk samples; however, this makes it difficult to precisely control the thickness and domain size of the samples. Moreover, the most intensely studied 2D magnetic materials (e.g. CrI_3, $Cr_2Ge_2Te_6$) are unstable in air and are only ferromagnetic below ~50 K. Hence, exploring new 2D magnetic materials is of grand significance. Recently, vdW epitaxy techniques have fulfilled the growth of 2D vdW materials on metal substrates, such as 2D silica bilayers and related transition metal silicates which break inversion symmetry guaranteeing at least a piezoelectric response. The arrival of these 2D vdW materials opens up exciting opportunities for preparing 2D multiferroic materials. In this talk, we demonstrate air stable, single layer 2D $M_xSi_2O_5(OH)_y$ (M= Fe, Cr) structures consisting of a silica-capped metal oxide grown on Pd and Au substrates that exhibit ferromagnetic order at room temperature as measured by x-ray magnetic circular dichroism (XMCD) spectroscopy. Application of small magnetic fields proves that the observed magnetic structures follow a hysteretic behavior. SQUID magnetometry confirms these results and reveals high spin-polarization at room temperature with

in-plane magnetic anisotropy. The measurements are further supported by first-principles theoretical calculations which highlight approaches to stabilize the magnetic order. Thus $M_xSi_2O_5(OH)_y$ based 2D materials represent a new platform for single layer 2D ferromagnetism with potential multiferroic behavior.

1. Meyer, J. C. *et al.* The structure of suspended graphene sheets. *Nature***446**, 60–63 (2007).
2. K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. G. and A. A. F. Electric Field Effect in Atomically Thin Carbon Films. *Science.***306**, 666–669 (2004).

5:00pm **2D+EM+MI-WeA-9 Novel Materials for Quantum Computing Devices: Monolayer Topological Superconductors***, Yi-Ting Hsu,* University of Notre Dame **INVITED**

Atomically thin superconductors that possess 'topological properties' have attracted extensive attention since many of them are predicted to host exotic zero-energy quasi-particles called Majorana zero modes (MZMs). MZMs are proposed to be the building blocks in a type of fault-tolerant quantum computation scheme known as topological quantum computation (TQC). One of the pressing challenges in the field of TQC is to experimentally realize such a superconducting film and detect the existence of MZMs. In this talk, I will first discuss what a topological superconductor is and why it is a promising platform for a TQC device. I will then talk about candidate materials in the family of transition metal dichalcogenides. Finally, I will discuss our theory-experiment combined effort to systematically identify more candidates in two-dimensional material databases.

5:40pm **2D+EM+MI-WeA-11 Tuning Magnetism and Superconductivity in Single Layer FeSeTe by Chemical Pressure***, Basu Oli, Q. Zou, H. Zhang,* West Virginia University*; T. Shishidou, M. Weinert,* University of Wisconsin, Millwaukie*; L. Li,* West Virginia University

The interplay of topology, magnetism, and superconductivity in a single-layer FeX (X = S, Se, Te) epitaxially grown on $SrTiO_3$ (STO) substrate provides a model system for investigating a wide range of quantum phenomena. This work explores the impact of chemical pressure on magnetism and superconductivity in single-layer $FeSe_{1-x}Te_x$/STO grown by molecular beam epitaxy using *in-situ* angle-resolved photoemission spectroscopy and scanning tunneling microscopy/spectroscopy. We find that the Fermi surface consists of only an electron pocket at the M point, which decreases in size with increasing Te concentration and disappears completely for x > 0.75. At the Γ point, a hole pocket appears with x > 0.65, while the bands changes from parabolic to linear up to x = 0.9, where it reverts back to parabolic. Accompanying the changes in the band structure, the top of bands at the Γ also shift towards and then away from the Fermi level, indicative of a topological phase transition in $FeSe_{1-x}Te_x$/STO. At 4.3 K, while the FeTe films are non-superconducting, dI/dV tunneling spectra indicate the emergence of superconductivity when Se concentration is greater than 25%. Our spin spiral calculations indicate that the FeTe system exhibits long-range bi-collinear antiferromagnetic (AFM) order, which is tuned toward the checkerboard (CB) AFM fluctuations with the incorporation of Se. Our findings indicate that CB AFM fluctuations are critical for superconductivity in epitaxial single layer iron chalcogenide superconductors on STO.

This research is supported by DOE (DE-SC0017632).

6:00pm **2D+EM+MI-WeA-12 Peculiar Near-Contact Regime of Andreev Reflection at the Breakdown of a Tunnel Junction***, Petro Maksymovych, S. Song,* Oak Ridge National Laboratory*; J. Lado,* Aalto University, Finland*; W. Ko,* Oak Ridge National Laboratory

Recently we introduced non-contact Andreev reflection (NCAR) - a new experimental approach to quantify Andreev reflection in a tunable tunnel junction [1]. The technique utilizes the fundamental connection between the physics of the scattering process and the strength of exponential non-linearity of the tunneling current, and therefore adopts the tunneling current decay rate as the observable. NCAR simultaneously achieves spectroscopy of quasiparticle density of states, atomic-scale resolution and quantitative Andreev reflection, while avoiding the necessity to form invasive and mesoscale mechanical contacts.

One surprising observation of NCAR is that Andreev reflection does not have a monotonous dependence on tip-sample separation. In particular, the decay rate signature of Andreev reflection proceeds through a maximum just before the mechanical contact. In this talk, we will explore in detail the origin of this near-contact regime and its significance for the characterization of superconductivity. In particular, we will rationalize the observations within the accepted BKT model as well as accurate tight-binding simulations, revealing the fundamental connection between the tunneling barrier dependence of Andreev reflection and the order in perturbation theory responsible for the tunneling current. Furthermore, we will highlight the importance of higher order Andreev reflection for the measurement of unconventional superconductors. In general, tunneling and near-contact regimes will qualitatively differ from each other in the case of unconventional superconducting order parameters, enabling direct inference of their symmetry.

At the same time, we will demonstrate how quantitative comparison of statistical distributions of decay rate enables a complementary, probabilistic analysis of the Andreev reflection [2]. This purely informational approach is particularly important given the narrow parameter space that separates the now rich variety of techniques to directly probe superconductivity, enabling robust control over crossovers between non-contact, near-contact, multiple Andreev reflection as well as quasiparticle tunneling in atomic-scale junctions.

Research sponsored by Division of Materials Science and Engineering, Basic Energy Sciences, Office of Science, US Department of Energy. SPM experiments were carried out as part of a user project at the Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, a US Department of Energy Office of Science User Facility.

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Actinides and Rare Earths Focus Topic
Room 318 - Session AC+LS+MI-WeA
Chemistry and Physics of the Actinides/Rare Earths

Moderator: Krzysztof Gofryk, Idaho National Laboratory

2:20pm **AC+LS+MI-WeA-1** ***Novel Preorganized Ligands for Selective and Efficient Separation of f-Elements , Santa Jansone-Popova,*** Oak Ridge National Laboratory **INVITED**

Selective separation of f-elements (lanthanides and actinides) is challenging to realize due to their very similar properties. For example, lanthanides exist predominantly in 3+ oxidation state and their ionic radii decrease across the lanthanide series on average by only 0.01 Angstrom per unit increase in atomic number. Organic molecules (ligands) capable of magnifying these effects and selectively recognizing one element over another are extensively sought after. Implementation of such ligands in separating f-elements would result in substantial cost and energy savings. The neutral organic molecules decorated with at least four donor atoms (oxygens and/or nitrogens), designed to complex with metal ions and that do not undergo conformational change in the process, demonstrate exceptional size selectivity.[1-3] The innate rigidity and size of the cavity in addition to the electronic structure of ligands provide the means to control the selectivity across the trivalent f-element series by design. On the contrary, organic molecules with donor groups that are connected to freely rotating single bonds show high affinity for metal ions that are more Lewis acidic.[4] Superior performance of preorganized ligands in two immiscible solvent system in selectively separating adjacent lanthanides or lanthanides from actinides will be discussed in detail. More specifically, the chemistry of hydrophobic and hydrophilic bis-lactam-1,10-phenanthroline-based ligands having four donor atoms will be presented, focusing on the separation of adjacent lanthanides (e.g., Nd-Pr, Tb-Gd, Ho-Dy) and

Americium-Europium pair separation. Additionally, the contrasting performance of rigid ligands decorated with two to four donor atoms will be shown and compared to their non-preorganized counterparts.

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3:00pm **AC+LS+MI-WeA-3 Impact of Noncovalent Interactions on Actinide Structural Chemistry*, Karah E. Knope,*** Georgetown University*; J. Wacker,* Lawrence Berkeley National Lab*; M. Shore,* Georgetown University **INVITED**

Noncovalent interactions (NCIs) have received significant attention over the past several years. This interest is fueled by recent examples that have shown that these interactions, which exist beyond the first coordination sphere of a metal ion, can be used to isolate novel structural units, push chemical equilibria, and tune redox behavior. As illustrated by these examples, it has become clear that consideration of NCIs is important for understanding the overall chemical behavior of a metal ion. Taken together with our group's interest in the factors that govern actinide speciation and reactivity, our recent work has thus focused on the solution and solid-state structural chemistry of actinide complexes and clusters that form from acidic aqueous solution in the presence of heterocycles capable of N-H hydrogen bonding interactions. Specifically, we have examined the identity (i.e. composition, charge, coordination number) of actinide structural units that precipitate from monoanionic ligands systems including Br^{1-}, Cl^{1-}, and NO_3^{1-} with N-heterocycles ranging from pyridinium to terpyridinium. While bromide, chloride, and nitrate all form weak relatively weak complexes with the actinides (Th-Pu), clear effects of NCIs are present only in the chloride ligand system with chloride rich (e.g. $[Th(H_2O)_2Cl_6]^{2-}$) to chloride poor (e.g. $[Th(H_2O)_7Cl_2]^{2+}$) complexes having been observed. By contrast, speciation is much more limited in the Br^{1-} and NO_3^{1-} ligand systems. Presented here will be an overview of our experimental and computational efforts to understand the effects of NCIs on phase formation.

4:20pm **AC+LS+MI-WeA-7 XPS Characterization of a Pu-7at.%-Ga Alloy*, Paul Roussel,*** AWE, UK*; K. Graham, S. Hernandez, J. Joyce, T. Venhaus,* Los Alamos National Laboratory

Gallium is often used in small concentrations (<10 at. %) as an alloying element with plutonium to stabilize the face centred cubic phase.Most XPS analyses of these plutonium materials have focused on low concentration alloys (< 4 at. %) where it is difficult to measure the signal and effects of gallium.Here we report the analysis of a higher alloy concentration of $Pu_{0.93}Ga_{0.07}$ in the as received state, during sputter depth profiling and following oxidation either from the addition of Langmuir quantities of oxygen or the in situ environment of the ultra high vacuum system.Quantification of the spectra has been undertaken by comparison of the elastic peak intensity and this has allowed us to test the recently reported relative sensitivity factors for plutonium [1]. The higher alloying concentration has permitted quantification of gallium in both the metal and oxides and, for the first time, the gallium chemical state plot for both alloy and oxide has been determined along with those of plutonium and oxygen.In addition to this we highlight a novel method to quantify the C1s peak which overlaps with the $Pu5p_{1/2}$ photoemission line and demonstrate the effect of gallium on the in situ oxidation rate of this alloy.

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4:40pm **AC+LS+MI-WeA-8 Nanoscale Uranium Oxide: Correlating Colloidal Synthesis Pathways with Structure at the Atomic and Nanometer Length Scale*, Liane Moreau,*** Washington State University

Nanoscale uranium oxide is of interest for the development of advanced nuclear fuels and its importance in the corrosion of conventional fuel pellets and trends in uranium mobility within environmental systems. Knowledge of nanoscale properties of uranium oxide, however, are limited due to 1) a lack of synthetic methods capable of forming monodisperse actinide oxide nanoparticles and 2) the challenges associated with characterization of complex interfaces. Organic-phase colloidal approaches prove promising towards the synthesis of actinide oxide nanoparticles due to precedence for formation of highly monodisperse transition metal oxide nanoparticles, and the capability to synthesize particles in rigorously air- and water-free environments. Three systems will be discussed which exploit colloidal uranium oxide synthesis. The first explores oxidation kinetics in UO_2 nanoparticles. Synthesis is paired with X-ray diffraction and X-ray spectroscopy to compare nanoparticle oxidation trends with those observed in bulk counterparts under controlled oxidative environments. The second traces the formation mechanism of UO_2 nanoflowers and demonstrates the role of solvent interactions in anisotropic growth. The established growth patterns are particularly interesting as anisotropic growth in transition metal counterparts have traditionally depended on surface binding species and trace impurities rather than solvent chemistry. The third system investigates the deposition of uranium oxide onto iron nanoparticle cores, which serves as an analogue to environmental systems where uranium and plutonium have shown to preferentially bind to Fe-based minerals. The high-surface area particles provide a means to investigate the interface in atomistic detail. In describing the aforementioned systems, X-ray focused approaches and characterization method development will be discussed, which prove critical to resolving structural attributes and making connections between synthetic parameters and resulting atomic and nanoscale structure.

5:00pm **AC+LS+MI-WeA-9 Chemical Speciation Mapping of Spent Nuclear Fuel Using Soft X-Ray Spectromicroscopy at the Advanced Light Source*, Alexander Ditter,*** *D. Smiles, D. Lussier,* Lawrence Berkeley National Laboratory (LBNL)*; A. Altman,* Northwestern University*; M. Bachhav, L. He,* Idaho National Laboratory*; M. Mara,* Northwestern University*; S. Minasian,* Lawrence Berkeley National Laboratory (LBNL)*; C. Degueldre,* Lancaster, UK*; D. Shuh,* Lawrence Berkeley National Laboratory (LBNL)

A nuclear reactor is a complex environment, with high temperatures, large temperature gradients and a multitude of fission products in various states. Spent nuclear fuel offers a key window into this environment, but is an important object of study in its own right, particularly with regards to the release of radioisotopes into the environment. The oxidation of spent nuclear fuel is especially important as this can govern the transport and solubility of fission products. To study this process, a focused ion beam (FIB) was used to make thin sections of a low burnup spent fuel pellet at Idaho National Laboratory. These FIB sections were then measured at the O K-edge, U $N_{4,5}$-edges and Ce $M_{4,5}$-edges at the scanning transmission x-ray microscope (STXM) at Advanced Light Source (ALS) Beamline 11.0.2. O K-edge results were analyzed by a modified form of non-negative matrix factorization and revealed two main components, the bulk of the sample which is made up primarily of UO_2, and a thin (est. 8 nm) layer of UO_{2+x} on the surface of the FIB section. This oxidation occurred after FIB sectioning as a thin layer over the entire sample. Cerium measurements showed that the Ce (~ 0.4 wt. %) is at least predominantly in the 3+ oxidation state, though a small contribution of tetravalent cerium cannot be ruled out, representing the first known measurements of Ce oxidation state in spent nuclear fuel. This work lays the foundations for future STXM measurements of FIB sections of spent nuclear fuel which can be expanded to mixed-oxide, uranium nitride, or other advanced fuels.

5:20pm **AC+LS+MI-WeA-10 Structural, Thermodynamics, and the Electronic Properties of Al, Ga, In, and Tl Stabilized δ-Pu*, Sajib Barman,*** *S. Hernandez,* Los Alamos National Laboratory

The 5*f* electrons in plutonium show mystifying character contrary to other elements in the periodic table, where there are six solid state phases of plutonium that undergo five solid-state phase transformations in a relatively short temperature range. The low temperature α-phase is brittle and has a monoclinic structure where the high temperature δ-phase is ductile and has an FCC structure. From a metallurgical point of view, the ductile δ-Pu is mostly important due to its favorable elasticity compared to the brittle α-Pu. Therefore, stabilizing δ-Pu at room temperature can be done by alloying with Group IIIA elements (X = Al, Ga, In, and Tl). Nevertheless, the alloyed δ-Pu stability zones highly depend on the alloying content, where the stability zone decreases with increasing atomic number of the Group IIIA elements. In this work, we will present a systematic density functional theory investigation of the thermodynamic stability of alloyed δ-Pu with Group IIIA elements as a function of alloying content (3.125, 6.25, and 9.375 at. %). We have calculated that the δ-Pu-Ga alloys have the highest thermodynamic stability compared to the other Group IIIA alloys, where the stability trend follows Pu-Ga > Pu-Al > Pu-In > Pu-Tl at 0K. After volume optimization, the volume contracts for Pu-Ga and Pu-Al alloys, while the volume expands for Pu-In and Pu-Tl alloys with increasing alloy content. Finally, we will show the electronic structure analysis to

discuss the electronic interaction between Pu and the Group IIIA elements within the δ-matrix.

This work was carried out under Los Alamos National Laboratory's LDRD-DR project #20210001DR.

Magnetic Interfaces and Nanostructures Division
Room 330 - Session MI-WeA

Spin Landscape II (Magnetic Structures in Real and Momentum Space)

Moderator: Markus Donath, Muenster University, Germany

2:20pm **MI-WeA-1 Exploring Magnetic Reversal Behavior and Domain Structure in Perpendicular Anisotropy Layered Synthetic Antiferromagnets**, ***Olav Hellwig***, Chemnitz University of Technology and Helmholtz Zentrum Dresden-Rossendorf, Germany **INVITED**

In atomic antiferromagnets (AFMs) neither the magnetic field reversal behavior nor the magnetic domain structure are easily accessible. The reason for that is the usually very strong antiferromagnetic (AF) exchange interaction, yielding switching fields in the range of many Tesla and the even microscopically compensated magnetic moment without any significant stray fields to detect.

The situation is different in perpendicular magnetic anisotropy layered synthetic AFMs employing interlayer exchange coupled thin film multilayers with a significantly reduced AF-exchange interaction (based on the well-known Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction), which we use here to explore the magnetic reversal behavior and domain structure in layered type AFMs [1-7]. This model system also has the advantages of easy fabrication via conventional sputter deposition techniques and being compatible with smooth amorphous substrates, such as Si/SiO_2 wafers. Furthermore it is possible to tune the strength of the RKKY-type AF-interlayer exchange continuously via the individual thicknesses of the coupling layers (here are mostly Ru and Ir used).

The significantly reduced AF-exchange interaction strength becomes now also comparable to other magnetic energy terms, such as the anisotropy or demagnetization energy. This creates - to atomic AFMs unknown and so far unexplored - competitive magnetic energy landscapes. They lead to new reversal modes and domain structures, as we subsequently reduce the strength of the AF-interlayer exchange coupling from its dominating position [5] first below the perpendicular anisotropy energy [1-4,6,7] and second below the demagnetization energy of the layered AF system.

We will discuss the physics behind such new reversal modes and domain structures in perpendicular anisotropy layered synthetic AFMs and highlight potential applications in the arena of nanomagnetism and spintronics. Finally we will also point out in what respect such systems provide design aspects and opportunities beyond the scope of conventional atomic AFMs, in particular in the light of "Imperfectly Perfect Materials".

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3:00pm **MI-WeA-3 Influence of Underlayer Quality and Sputter Gas Pressure on Structural and Magnetic Properties of Co/Pt Multilayers**, ***Rico Ehrler***, *T. Uhlig, O. Hellwig,* Chemnitz University of Technology, Germany

Co/Pt multilayers (MLs) are standard systems for perpendicular anisotropy layered thin films. They are fully tunable via their layer thicknesses and number of repeats and can be easily fabricated using sputter deposition on amorphous substrates. Moreover, they were intensively investigated for their well-ordered stripe domain and bubble states [1,2] and as potential recording media [4]. Synthetic antiferromagnets with perpendicular anisotropy can also be based on these ML systems [5]. The growth conditions of the MLs are of particular importance for fine-tuning their structural and magnetic properties.

As was already shown in the literature, seed layers [4,6] and sputter gas pressure during deposition [1,3] are two very important factors that influence the structural as well as magnetic properties profoundly. The use of a specific underlayer is a common practice to define a crystalline texture for the ML on amorphous substrates. Pt and Pd are widely used as seed layer materials, sometimes in combination with additional, very thin adhesion layers to obtain a good bonding to the substrate [4,6].

For low sputter pressures, the ML is well-defined with sharp interfaces and a continuous, closed film that maintains a laterally continuous strong magnetic exchange. Under these conditions, well-ordered stripe domains and bubble states can form, which can be moved via external fields or currents laterally across the magnetic thin film [1,2]. At high deposition pressures, the MLs grow in a more isolated, granular fashion [1,3]. The layer structure shows a higher degree of disorder and a distinct reduction in the lateral exchange between adjacent grains. Such a microstructure is desirable for magnetic recording media, as it enables the static bit-wise storage of data, where every bit consists of many significantly smaller and magnetically isolated grains [1].

In this context, we will discuss the influence of the Pt seed on the structural and magnetic properties of the Co/Pt ML system, with emphasis on the impact of deposition pressure and adhesion layer. A systematic variation of the underlayer is presented for different ML deposition pressures, changing the characteristics from a continuous thin film to an isolated grain structure.

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3:20pm **MI-WeA-4 Thickness and Oxygen Growth Pressure Effects on Spontaneous Magnetization Reversal**, ***Mikel Barry Holcomb***, *G. Bhandari, N. Mottaghi, R. Trappen,* West Virginia University

Utilizing many techniques (bulk magnetometry, neutron reflectometry and resonant x-ray magnetic scattering), we have discovered and explored spontaneous magnetization reversal in complex oxide La0.7Sr0.3MnO3 thin films. The spontaneous magnetization reversal occurs at low applied fields and originates from the competition between different types of magnetic order. While the overall effect is observed across many sample thicknesses and oxygen growth pressures, these parameters affect the behavior systematically. Films were grown by pulsed laser deposition with reflection high energy electron diffraction to ensure layer-by-layer and high-quality growth.

4:20pm **MI-WeA-7 Thermally Induced Magnetic Order from Glassiness in Elemental Neodymium**, ***Daniel Wegner***, Radboud University, Netherlands **INVITED**

I will present results from our most recent spin-polarized scanning tunneling microscopy (SP-STM) study of single crystalline elemental neodymium (Nd) metal, which is a self-induced spin glass in its ground state [1]. Temperature in thermodynamics is synonymous with disorder, and responsible for ultimately destroying ordered phases. We found an unusual magnetic transition where, with increasing the temperature, long-range multi-Q magnetic order emerges from the glassy state [2]. Using temperature-dependent SP-STM, we characterized the local Q order in the spin-Q glass phase and quantified the emergence of long-range multi-Q order with increasing temperature. We developed two distinct analysis tools, which enable the quantification of the glass transition temperature, based on measured magnetization images. We compared these observations with atomic spin dynamics simulations, which reproduce the qualitative observation of a phase transition from a low-temperature spin glass phase to an intermediate ordered multi-Q phase. These simulations trace the origin of the unexpected high temperature order in weakened frustration driven by temperature-dependent sublattice correlations. Our findings constitute an example of order from disorder and provide a rich platform to study magnetization dynamics in a self-induced spin glass.

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Khajetoorians, *Nature Physics* (accepted), arXiv:2109.04815 (2022).

5:00pm **MI-WeA-9 Designing Antiferromagnetic Domain Landscapes via Focused Ion Beam Irradiation***, **Fabian Samad,*** Helmholtz-Zentrum Dresden - Rossendorf, Germany*; G. Hlawacek,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; L. Koch,* Technische Universität Chemnitz, Germany*; X. Xu,* Helmholtz Zentrum Dresden-Rossendorf, Germany*; O. Hellwig,* Helmholtz-Zentrum Dresden - Rossendorf, Germany

Layered thin film synthetic antiferromagnets (SAFs) are highly promising candidates for future technological applications, particularly in nanomagnetism and spintronics [1]. Their magnetic properties can be easily tuned by changing the individual layer-thicknesses and also further manipulated post-deposition by applying magnetic fields or electric currents [2]. Antiferromagnetic domains in SAFs are of specific interest, as they possess strongly desirable properties, such as high stability against external magnetic fields, and large domain wall velocities [3]. However, due to the absence of a net magnetization, the deterministic creation of microscopic SAF domains is challenging [2,4].

In this project, we show that ion beam irradiation can be used to 'write' complex microscopic SAF domain patterns [5]. For this, we employ a nanometer-focused He+ ion beam, which intermixes the layer interfaces, thus changing the local magnetic properties in a controlled manner. We report highly tunable magnetic and magnetoresistive behavior of the various SAF domain patterns in the presence of externally applied magnetic fields and electric currents.

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Magnetic Interfaces and Nanostructures Division
Room 330 - Session MI+2D+TF-ThM

Quantum Materials (2D)

Moderator: Zheng Gai, Oak Ridge National Laboratory

8:00am **MI+2D+TF-ThM-1 Exploration of Two Surfaces Observed in Weyl Semimetal $BaMnSb_2$, *Zheng Gai,*** *Q. Zou,* Oak Ridge Natinal Laboratory*; S. Huang,* University of South Carolina*; W. Ko, M. Fu,* Oak Ridge Natinal Laboratory*; Y. Yang, K. Zhao,* Louisiana State University*; S. Crittenden,* University of South Carolina*; E. Plummer,* Louisiana State University*; R. Jin,* University of South Carolina

Single crystalline $BaMnSb_2$ is considered as a 3D Weyl semimetal with the 2D electronic structure containing Dirac cones from the Sb sheet. The unique surface electronic structure can be probed by techniques such as angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy/spectroscopy (STM/S). However, these techniques require an in-depth understanding of the surface structure and electronic properties. We report experimental investigation of low-temperature cleaved $BaMnSb_2$ surfaces using STM/S and low energy electron diffraction (LEED). By natural cleavage, we find two terminations: one is Ba1 (above the orthorhombic distorted Sb sheet) and another Sb2 (at the surface of the Sb/Mn/Sb sandwich layer). Both terminations show the 2 × 1 surface reconstructions, with drastically different morphologies and electronic properties, however. The reconstructed structures, defect types and nature of the electronic structures of the two terminations are extensively studied. The quasiparticle interference (QPI) analysis also reveals that the surface-projected electronic band structures strongly depend on the surface termination. The existence of defects can greatly modify the local density of states to create electronic phase separations on the surface in the order of tens of nm scale. Our observation on the atomic structures of the terminations and the corresponding electronic structures provides critical information towards an understanding of topological properties of $BaMnSb_2$.

8:20am **MI+2D+TF-ThM-2 Properties of Mn_3Sn Films Grown on Sapphire Substrates Using Molecular Beam Epitaxy*, Sneha Upadhyay,*** Ohio University*; T. Erickson, D. Ingram,* Ohio University*; K. Sun,* The University of Michigan , Ann Arbor*; A. Smith,* Ohio University

The kagome antiferromagnet Mn_3Sn is a fascinating material because it's one of the rare antiferromagnets that exhibits large anomalous Hall and Nernst effects. This opens a new area of research using functional antiferromagnets[1], but for future device applications, it requires fabricating high-quality thin films. There are reports of the controlled growth of Mn_3Sn on substrates like *m*-plane sapphire,[2] Pt/Al_2O_3 (0001)[3], and others using sputtering growth, but this often can result in polycrystalline films. In this work, we investigate the growth of Mn_3Sn films on *c-plane* sapphire substrates using molecular beam epitaxy. Effusion cells are used for Mn and Sn sources which are calibrated using a quartz crystal thickness monitor. The growth is monitored *in-situ* using reflection high energy electron diffraction and *ex-situ* measurements are carried out using X-ray diffraction, Rutherford backscattering, and cross-sectional scanning transmission electron microscopy. The samples are grown at 500 ± 9°C and 416 ± 9 °C with an Mn: Sn atomic flux ratio of 3.2: 1 on *c-plane* sapphire substrates for 60 mins. We observe streaky RHEED patterns at both temperatures indicating high-quality crystalline growth with 2 different orientations, (0001) and (11-20), which are also backed up by the XRD spectra. STEM verifies ~3:1 Mn to Sn stoichiometry but also reveals discontiguous films. After optimizing the growth conditions, the next phase of the study is to begin *in-situ* scanning tunneling microscopy and spin-polarized STM studies of the structural, electronic, and magnetic properties of the *as-grown* Mn_3Sn surfaces, and in this presentation, we plan to present initial results.

The authors acknowledge support from the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering under Award No. DE-FG02-06ER46317. The authors would like to thank Dr. Stinaff and his students for back coating the sapphire substrates.

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8:40am **MI+2D+TF-ThM-3 Interfacial Magnetism in Oxide Heterostructures*, Alex Demkov,*** The University of Texas at Austin **INVITED**

New functionalities and unexpected electronic structures can emerge in artificially engineered complex oxide heterointerfaces due to the coupling of multiple physical properties such as ferroelectricity, ferromagnetism, conductivity, charge transfer, etc. Here, we discuss heterointerfaces involving perovskite oxides $SrTiO_3$, $LaAlO_3$, and $BaTiO_3$, as well as a rock salt ferromagnetic semiconductor EuO. Combining theoretical analysis, experimental growth, and characterization techniques with atomic level resolution, we highlight some of these intriguing emergent interfacial phenomena. We consider several means of creating a two-dimension carrier gas, through band offset engineering, polarization doping, and oxygen vacancy doping. In addition, we also discuss ways of manipulating these electron/hole gases and their potential applications in new electronic devices.

Starting from the pioneering discovery of a 2DEG at the LAO/STO interface, interfaces of transition metal oxides have been at the center of many theoretical and experimental studies. This two-dimensional carrier gas is rather different from the one occurring in semiconductor heterostructures. Its origins and mechanisms of spatial localization are rather complex. Thanks to modern experimental and theoretical techniques, one can identify which of these mechanisms are actually present at a given interface. In some cases, we can also tailor the choice of specific materials where one mechanism dominates, allowing us to study that particular mechanism in more detail.

Owing to relatively large band gaps and the presence of occupied as well as empty ***d*** and***f*** states, the oxide interfacial 2DEG shows a higher degree of spatial confinement.It is a unique physical system where, in the span of just about a nanometer, one can create strong interactions between multiple order parameters leading to phenomena that in the bulk are either very small or forbidden by symmetry. Besides the fundamental physics interest, these multi-functional interfaces may one day be utilized for technological applications.

The work has been supported by the Air Force Office of Scientific Research under grants FA9550-18-1-0053 and FA9550-12-10494, the Texas Advanced Computing Center, and by the National Science Foundation under grants IRES–1358111.

9:20am **MI+2D+TF-ThM-5 Epitaxial 2D Van Der Waals Magnets*, Roland Kawakami,*** Ohio State University **INVITED**

In this talk, I will discuss our latest advances on the epitaxial growth of 2D van der Waals (vdW) magnets and their integration with topological insulators (TI). This work is motivated by the realization of topological phases such as the quantum anomalous Hall effect and highly efficient spin-orbit torque produced by TIs. We have focused on integrating 2D magnets $MnSe_2$, Fe_3GeTe_2 (FGT) and $CrGeTe_3$ (CGT) with TIs Bi_2Se_3 and Bi_2Te_3.

Our initial studies of $MnSe_2$ growth on Bi_2Se_3 showed a tendency for the interdiffusion of Mn into the Bi_2Se_3. While this was initially undesirable, this ultimately led to the synthesis of $MnBi_2Se_4$ (MBS), a new magnetic topological insulator. This is a vdW material with septuple layer (Se-Bi-Se-Mn-Se-Bi-Se) base units, similar to its cousin $MnBi_2Te_4$ (MBT). However, the vdW phase is not the thermodynamically stable phase and bulk crystals do not exist, so the epitaxial stabilization of MBS creates the opportunity to explore the magnetic and topological properties of this material. We find that MBS is a layered antiferromagnet, similar to MBT, but a difference is that the magnetic moments lie in the plane of the film. Angle resolved photoemission experiments show the presence of a topological surface state with Dirac dispersion.

For bilayers of 2D magnets and TIs, we have focused on FGT and CGT films. A key step has been the optimization of FGT by studying its growth on Ge(111) substrates. Here, we learned that kinetics play an important role in the epitaxial growth. By varying the deposition rate, we control the formation or suppression of an initial tellurium-deficient non-van der Waals phase (Fe3Ge2) prior to realizing epitaxial growth of the vdW FGT phase. Using cross-sectional scanning transmission electron microscopy and scanning tunneling microscopy, we optimize the FGT films to have atomically smooth surfaces and abrupt interfaces with the Ge(111) substrate. The magnetic properties of our high quality material are confirmed through magneto-optic, magnetotransport, and spin-polarized

STM studies. Importantly, this demonstrates how the interplay of energetics and kinetics can help tune the re-evaporation rate of chalcogen atoms and interdiffusion from the underlayer. Utilizing these insights, we have developed the growth of FGT and CGT on Bi_2Te_3 for the integration of 2D magnets with TIs.

11:00am **MI+2D+TF-ThM-10 Hybrid Superconductor-Semiconductor Device***, **Sergey Frolov**,* University of Pittsburgh **INVITED**

Research into the generation, confirmation and manipulation of Majorana zero modes has brought heightened interest to hybrid materials systems made of fairly conventional components such as s-wave superconductor metals and semiconductors. The game in this arena is to carefully tailor the properties of an electronic device to make possible the observation of exotic physics such as topological superconductivity. The main lesson that we have learned is that the path to any plausible discovery lies through painstakingly careful and very deep understanding of the materials properties and device fabrication conditions. This being true for such a standard set of materials, the lesson certainly applies to more exotic compounds.

In this talk I will briefly summarize the status of the search for Majorana modes in superconductor-semiconductor hybrid devices. I will also highlight other unusual phenomena that arise in these systems such as higher order Josephson effects, time-reversal symmetry broken Josephson current phase relations. I will also show how these materials can be used to build quantum circuits with enhanced functionality, not necessarily arising from topological protection.

11:40am **MI+2D+TF-ThM-12 Magnetotransport in Graphene/$Pb_{0.24}Sn_{0.76}Te$ Heterostructures: Finding a Way to Avoid Catastrophe***, G. Stephen,* Laboratory for Physical Sciences*; I. Naumov,* Howard University*; N. Blumenschein, L. Sun, **Jennifer DeMell**,* Laboratory for Physical Sciences*; S. Shirodkar, P. Dev,* Howard University*; P. Taylor,* Army Research Laboratory*; J. Robinson, P. Campbell,* Naval Research Laboratory*; A. Hanbicki, A. Friedman,* Laboratory for Physical Sciences

While heterostructures are ubiquitous tools to enable new physics and device functionalities, the palette of available materials has never been richer. Combinations of two emerging material classes, 2D materials and topological materials, are particularly promising because of the wide range of possible permutations that are easily accessible. Individually, both graphene and $Pb_{0.24}Sn_{0.76}Te$ (PST) are widely investigated for spintronic applications because graphene's high carrier mobility and PST's topologically protected surface states are attractive platforms for spin transport. Here, we combine monolayer graphene with PST and demonstrate a hybrid system with properties enhanced relative to the constituent parts. Using magnetotransport measurements, we find carrier mobilities up to 20,000 cm^2/Vs and a magnetoresistance approaching 100%, greater than either material alone. We also establish there are two distinct transport channels and determine a lower bound on the spin relaxation time of 4.5 ps. The results can be explained using the polar catastrophe model, whereby a high mobility interface state results from a reconfiguration of charge due to a polar/non-polar interface interaction. Our results suggest that proximity induced interfaces states with hybrid properties can be added to the still growing list of remarkable behaviors in these novel materials.

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