PCSI
Room Keahou I - Session PCSI-MoA3
Magnetic Materials (2D, Monolayers, & Heterostructures

Moderator: Alex Demkov, The University of Texas

5:20pm **PCSI-MoA3-41 UPGRADED: Molecular Beam Epitaxy Growth and Stoichiometry-Induced Ferromagnetism in Altermagnetic Candidate MnTe**, ***Matthew Brahlek***, Oak Ridge National Laboratory, USA

The field of spintronics has seen much excitement around predictions of altermagnetism due to potential applications related to the associated novel spin-moment textures. MnTe stands out as a prominent altermagnetic candidate because of its layered A-type antiferromagnetic structure, high Néel temperature (approximately 310 K), and semiconducting properties. However, critical challenges remain regarding (1) how to synthesize high-quality, well-controlled samples and (2) understanding the fundamental properties of this material.

Our study presents two key findings. The first is that on lattice-matched InP(111), slight differences in the InP surface can be used to effectively select different polymorphs of MnTe when grown by molecular beam epitaxy (MBE). More specifically, the termination of the InP (111) substrate seems to be critical factor. In termination ((111)A)) triggers the nucleation of the NiAs structure, the candidate altermagnetic structure, whereas termination with P ((111)B stabilizes the ZnS structure, which is also antiferromagnetic and has a relatively large bandgap of ~2 eV.

Regarding the properties of MnTe in the NiAs structure, we reveal that the electronic and magnetic properties are influenced by the natural stoichiometry of MnTe [1]. Electronic transport measurements and in situ angle-resolved photoemission spectroscopy reveal that the films are inherently metallic, with the Fermi level situated in the valence band. The band structure aligns well with first-principles calculations for altermagnetic spin-splitting. Neutron diffraction confirms the antiferromagnetic nature of the film with planar anisotropy, while polarized neutron reflectometry indicates weak ferromagnetism, attributed to a slight Mn-richness inherent to the MBE-grown samples. Combined with the anomalous Hall effect, this work demonstrates that the electronic properties are significantly impacted by the weak ferromagnetism.

Overall, this research highlights potential mechanisms for investigating and ultimately controlling altermagnetic properties, thus paving the way for diverse spintronic applications.

[1] Michael Chilcote, Alessandro R Mazza, Qiangsheng Lu, Isaiah Gray, Qi Tian, Qinwen Deng, Duncan Moseley, An-Hsi Chen, Jason Lapano, Jason S Gardner, Gyula Eres, T Zac Ward, Erxi Feng, Huibo Cao, Valeria Lauter, Michael A McGuire, Raphael Hermann, David Parker, Myung-Geun Han, Asghar Kayani, Gaurab Rimal, Liang Wu, Timothy R Charlton, Robert G Moore, Matthew Brahlek, Advanced Functional Materials 2405829 (2024) 10.1002/adfm.202405829 [https://doi.org/10.1002/adfm.202405829]

5:40pm **PCSI-MoA3-45 Imaging and Writing Chiral Antiferromagnetic Domains in the 2D Triangular Antiferromagnet $Co_{1/3}NbS_2$**, ***Scott Crooker***, Los Alamos National Laboratory

The family of intercalated niobium and tantalum dichalcogenides, $M_{1/3}NbS_2$ and $M_{1/3}TaS_2$ (where the *3d* magnetic atom M= V, Cr, Mn, Fe, Co, Ni), are van der Waals materials that host a wide range of fascinating magnetic properties.$Co_{1/3}NbS_2$ is an antiferromagnet (AFM) that features layers of Co spins on a 2D triangular lattice (see Fig.1) -- an archetypal frustrated network that can lead to complex magnetic topologies. Despite its vanishing net magnetization, $Co_{1/3}NbS_2$ was recently shown to exhibit a giant anomalous Hall effect [1], suggesting a nontrivial AFM order and potential for AFM electronic and spintronic devices. Recent neutron diffraction studies point to a non-coplanar 3Q AFM order with scalar spin chirality [2]. In contrast to conventional collinear AFM order, this (and certain other) complex AFM spin configurations can allow for off-diagonal elements the conductivity, σ_{xy}, which in turn generates anomalous and topological Hall effects in transport studies.

Crucially, $\sigma_{xy}(w)$ are frequency-dependent, and at optical frequencies they generate Kerr rotation and magnetic circular dichroism (MCD).Thus, the full power of optical methods, including spectroscopy and spatially-resolved imaging, can now be applied to investigate the complex AFM order in $Co_{1/3}NbS_2$.Here we show [3], using light spanning infrared to ultraviolet (1-3 eV), that MCD is a powerful and incisive probe of chiral 3Q AFM order in these van der Waals magnets. Measurements at different photon energies are compared with DFT calculations. Scanning MCD microscopy is used to directly image chiral AFM domains, and also, to demonstrate optical writing of chiral 3Q domains.These studies suggest routes to AFM spintronic devices based on $Co_{1/3}NbS_2$ and related 2D magnets.

Fig. 1 a) Structure of $Co_{1/3}NbS_2$, with 2D triangular lattices of Co spins. b) MCD vs. *B, T* shows onset of giant MCD and hysteretic AFM switching below the AFM ordering temperature T_{Neel} ≈28.5 K.c) Direct optical imaging of chiral AFM domains. d) Optical writing of chiral AFM domains.

[1] N.J. Ghimire *et al*, Nat. Comm. **9**, 3280 (2018); G. Tenasini *et al*, Phys. Rev. Res. **2**, 023051 (2020).

[2] H. Takagi *et al*, Nat. Phys. **19**, 961 (2023).

[3] E. Kirstein *et al*, submitted.

5:45pm **PCSI-MoA3-46 Electrostatic Extension of Magnetic Proximity Effect in $La_{0.7}Sr_{0.4}Mn_{O3}$**, *Qianqian Lan, Michael Schnedler, Lars Freter,* Forschungszentrum Jülich GmbH, Germany*; Chuanshou Wang,* Southern University of Science and Technology, China*; Kurt Fischer,* National Institute of Technology, Japan*; Rafal E. Dunin-Borkowski,* ***Philipp Ebert***, Forschungszentrum Jülich GmbH, Germany

Many fascinating magnetic effects emerge at interfaces between layers with different magnetic orders. Interface confinement is intimately related to the magnetic proximity effect, which typically has a spatial extent of only a few atomic layers. This short extent is due to the underlying physical coupling mechanisms, such as the exchange interaction, the Dzyaloshinsky-Moriya interaction, interface states, rehybridization, and reconstruction, all of which are highly localized. We use off-axis electron holography to reveal an exceptionally long-range magnetic proximity effect reaching ∼40 nm at a ferromagnetic (FM)/ paramagnetic (PM) interface in $La_{0.7}Sr_{0.3}MnO_3$ (LSMO). This wide extent arises from carrier diffusion and drift across the interface, which changes the Mn^{3+}/Mn^{4+} ratio and thereby the density of magnetic moments and local Curie temperature. We determine the carrier concentration dependence of the Curie temperature and unravel the physical mechanism of the electrostatic extension of magnetic proximity effects, fundamentally reshaping our understanding of micromagnetism in perovskites.

5:50pm **PCSI-MoA3-47 Toward a First-Principles Theory of Rare-Earth Ions in Crystals**, *Yongbin Lee, Zhenhua Ning,* Ames National Laboratory*; R. Flint,* Ames Laboratory*; R.J. McQueeney,* Ames National Laboratory & Iowa State University*; I.I. Mazin,* George Mason University*;* ***Liqin Ke***, Ames National Laboratory

Density functional theory (DFT), including its extensions designed to treat strongly correlated localized electron systems such as DFT+U and DFT+dynamical mean field theory, has proven exceedingly useful in studying the magnetic properties of solids. However, materials with rare earths (R) have remained a notable exception. The most vital rare-earth magnetic properties, such as magnetocrystalline anisotropy (MA), have been notoriously elusive due to the ubiquitous self-interaction error present in nearly all available DFT flavors. In this work [1], we show explicitly how the orbital dependence of self-interaction error may contradict Hund's rules and plague MA calculations, and how analyzing DFT metastable states that respect Hund's rules can alleviate the problem. We systematically investigate and discuss several rare-earth-containing families, RCo_5, $R_2Fe_{14}B$, RFe_{12}, and RMn_6Sn_6, to benchmark the MA calculations in DFT+U. For all compounds we investigated, we found that our methodology reproduces the magnetic easy-axis, easy-plane, and non-trivial easy-cone anisotropies in full agreement with low-temperature experimental measurements. Besides the fully-numerical ab initio approach, we further illustrate an efficient semi-analytical perturbation method that treats the crystal field as a perturbation in the limit of large spin-orbit coupling. This approach evaluates the rare-earth anisotropy by assessing the dependence of crystal-field energy on spin-quantization axis rotation using 4f crystal-field levels obtained from non-spin-orbit calculations. Our analytical method provides a quantitative microscopic understanding of the factors that control MA and can be used for predicting new high-MA materials. Finally, in addition to bulk materials, we explore the potential of utilizing rare-earth MA in 2D materials.

[1] Y Lee, Z Ning, R Flint, R J McQueeney, I I Mazin, L Ke, arXiv:2407.10067

+ Author for correspondence: liqinke@ameslab.gov

5:55pm **PCSI-MoA3-48 Defect Mediated Helical Phase Reorientation by Uniaxial Stress**, *Tae-Hoon Kim, Haijun Zhao, Liqin Ke,* Ames National Laboratory*; **Lin Zhou**,* Iowa State University

Chiral magnetism has attracted extensive research attention due to its fundamental science and technological importance. In chiral magnet domains, the competition between exchange and Dzyaloshinskii–Moriya interaction (DMI) causes the spins to wind periodically on a plane either perpendicularly (helix) or with a canting angle (cone) along a specific direction, defined as a propagation vector (Q) [1]. Strain engineering enables precise control of nanoscale magnetism while minimizing energy consumption [2]. However, the spatial evolution of strain-induced spin rearrangement, critical for deterministic control of chiral magnetic structures, remains unclear.

In this study, we utilize in-situ Lorentz transmission electron microscopy to manipulate and monitor the reorientation of the helical phase under quantitatively applied uniaxial tensile stress. Our results demonstrate that the Q vector direction of the helical phase can be tuned using external stress. The underlying mechanisms that govern the spin reorientation are magnetic defect mediated, which involve either “break-and-reconnect” events, or dislocation gliding and annihilation within the helices. Simulations prove that the strain-induced anisotropic DMI plays a significant role in driving the reorientation of the helical phase. Our findings provide valuable insights into energy-efficient manipulation of magnetic nanophase for information technology.

[1] Schoenherr, P. et al. Topological domain walls in helimagnets. Nat. Phys. **14**, 465 (2018).

[2] Wang, J. Mechanical Control of Magnetic Order: From Phase Transition to Skyrmions. Annu. Rev. Mater. Res. **49**, 1 (2019).

Author Index

Bold page numbers indicate presenter

— B —
Brahlek, Matthew: PCSI-MoA3-41, **1**
— C —
Crooker, Scott: PCSI-MoA3-45, **1**
— D —
Dunin-Borkowski, Rafal E.: PCSI-MoA3-46, 1
— E —
Ebert, Philipp: PCSI-MoA3-46, **1**
— F —
Fischer, Kurt: PCSI-MoA3-46, 1
Flint, R.: PCSI-MoA3-47, 1
Freter, Lars: PCSI-MoA3-46, 1
— K —
Ke, Liqin: PCSI-MoA3-47, **1**; PCSI-MoA3-48, 2
Kim, Tae-Hoon: PCSI-MoA3-48, 2
— L —
Lan, Qianqian: PCSI-MoA3-46, 1
Lee, Yongbin: PCSI-MoA3-47, 1
— M —
Mazin, I.I.: PCSI-MoA3-47, 1
McQueeney, R.J.: PCSI-MoA3-47, 1
— N —
Ning, Zhenhua: PCSI-MoA3-47, 1
— S —
Schnedler, Michael: PCSI-MoA3-46, 1
— W —
Wang, Chuanshou: PCSI-MoA3-46, 1
— Z —
Zhao, Haijun: PCSI-MoA3-48, 2
Zhou, Lin: PCSI-MoA3-48, **2**