Thin Films
Room 317 - Session TF-WeA

Far from Equilibrium Thin Films and Processes

Moderators: Dr. Saeed Almishal, Pennsylvania State University, **Prof. Lauren Garten**, Georgia Institute of Technology

2:15pm **TF-WeA-1 Frustration and Function in Single Crystal Compositionally Complex Oxides**, ***Zac Ward***, Oak Ridge National Laboratory **INVITED**

High entropy materials open vast compositional phase spaces inaccessible in conventional binary and ternary systems. Stabilizing single-crystal films comprising five or more principal cations in increasingly complex and reduced-symmetry crystal structures is creating new opportunities for discovering unexpected and exotic phenomena. Recent work has demonstrated that these systems can exhibit unique combinations of structural, thermal, and chemical robustness alongside continuously tunable electronic, magnetic, and optical properties relevant to next-generation computing and sensing architectures. The ultrashort quench times intrinsic to laser ablation are ideally suited to trapping these compositions as homogeneous, randomly mixed solid solutions. Here we discuss ongoing efforts using nonequilibrium pulsed laser deposition to synthesize perovskite, spinel, and Ruddlesden–Popper single crystal high entropy oxides. Observed functionalities are interpreted through many-body effects driven by inherent frustrations among coupled spin, charge, and lattice order parameters. Finally, we describe how these science drivers are motivating the development of autonomous synthesis platforms integrated with multimodal characterization and AI/ML-guided closed-loop optimization. These capabilities aim to accelerate the construction of reproducible defect–structure–function maps and to deliver experimentally grounded datasets essential for data-driven discovery.

2:45pm **TF-WeA-3 Designing Altermagnetism and Topological States in Ultrathin Multiferroic $BiFeO_3$**, ***Lucas Caretta***, Brown University **INVITED**

Magnetoelectric multiferroics - materials in which electric polarization and magnetic order are intrinsically coupled - offer a platform for ultralow-power switching, nonvolatile memory, and energy-efficient transduction. Yet practical deployment demands ultrathin films down to the atomic limit, where both orders typically degrade. Maintaining both order parameters at the thinnest scales in complex oxides remains a tremendous challenge, as uncompensated bound charge drives nanoscale depolarization in most ferroelectrics, while off-stoichiometry, reduced anisotropy, and charge transfer can produce magnetic dead layers in ultrathin oxides at substrate interfaces. Here, we realize a metastable multiferroic phase of $BiFeO_3$ (BFO) that not only sustains both order parameters at room temperature with no dead layer in the four unit cell, ultrathin limit, but also exhibits signatures of emergent altermagnetism. This second-order, thickness-driven phase transition gives rise to new multiferroic topological textures, where electric and magnetic symmetries become intertwined in previously unobserved ways. Additionally, we show pathways to design altermagnetism and topology at thicker length scales, where at domain boundaries, we uncover a rich hierarchy of noncollinear topological magnetoelectric structures, including polar bi-merons, polar vertices coupled to magnetic cycloid disclinations, and topological cycloidal knots, whose stability and dynamics are intimately linked to the underlying crystal symmetry. These findings establish a pathway to stabilize multi-order topology at device-relevant thicknesses and reveal emergent magnetic symmetry. Collectively, they reframe scaling limits in oxide multiferroics and open the door to emergent topological phenomena and ultralow-energy functionalities in correlated quantum materials.

3:15pm **TF-WeA-5 Chemical Disorder in Improper Ferroelectrics**, ***Billy Yang***, *Jeffrey Hodgson, David Sanchez, Sai Venkata Gayathri Ayyagari, Saeed S. I. Almishal, Nasim Alem, Jon-Paul Maria,* The Pennsylvania State University

Among rare-earth oxide derivatives, hexagonal manganites h-$RMnO_3$ (where R = Sc, Y, Dy-Lu) stand out as particularly interesting for exploring their geometrically driven improper ferroelectricity by exhibiting a polar phase with a space group of $P6_3cm$ below the Curie temperatures of approximately 1250 K. These materials further demonstrate multiferroic behavior below 100 K, with antiferromagnetic order occurring in the Mn sublattice. In this study, we report the synthesis of a high-entropy rare-earth manganite ($Y_{0.167}Gd_{0.167}Dy_{0.167}Ho_{0.167}Er_{0.167}Yb_{0.167}MnO_3$) in both bulk and thin-film forms, with X-ray diffraction (XRD) confirming that both forms stabilized as a hexagonal single-phase structure. Thin films with a targeted thickness of 80-100 nm were deposited by pulsed laser deposition (PLD) onto YSZ substrates and conductive bottom electrodes of Pt and ITO films for ferroelectric testing. Polarization-electric field measurements confirmed robust ferroelectric hysteresis loops at room temperature with a remanent polarization of ~2 μC/cm^2. Structural characterization via scanning transmission electron microscopy (STEM) reveals a polar structure accompanied by the rare-earth displacements and captures the domain structures. Preliminary magnetic measurements indicate non-negligible saturation magnetization values both in-plane and out-of-plane below 50 K. Furthermore, we investigate the influence of the growth atmosphere and discuss the associated defect chemistry in relation to the improper ferroelectricity. Ultimately, this work provides a detailed discussion of potential A-sites (e.g., Gd and Eu, which are traditionally orthorhombic-stabilizing species) and B-site disorder (e.g., Fe and Ga, for potential magnetic ordering tuning), and presents the first thorough investigation of improper ferroelectricity in a complex high-entropy system. These results demonstrate that the geometrically driven improper ferroelectricity remains remarkably robust against extreme local chemical disorder and defects, leading to a new design principle for novel multiferroic materials.

3:30pm **TF-WeA-6 Magnetic Exchange Bias Control in Kinetically-Arrested High-Entropy Oxide Heterostructures**, ***Joseph Petruska***, The Pennsylvania State University; *Timothy Charlton,* Oak Ridge National Laboratory; *John Heron,* University of Michigan; *Jon-Paul Maria, Saeed Almishal,* The Pennsylvania State University

High-entropy oxide (HEO) thin films grown under far-from-equilibrium conditions uniquely combine extreme chemical disorder with exceptional crystalline coherence. In this talk, we demonstrate predictive control of this anomalous crystalline state through the kinetic arrest of metastable macrostates. Specifically, aliovalent cation substitution, tightly controlled substrate temperature, and high adatom kinetic energy during growth enable deterministic programming of the out-of-plane lattice parameter in coherent rock salt HEOs while preserving in-plane epitaxial pinning to MgO. Using this approach, multilayer heterostructures sustaining lattice strains exceeding 5% can be stabilized, where the defect chemistry is dominated by trivalent cations compensated by cation vacancies, producing valence interfaces across which the Co valence evolves from predominantly Co^{2+} to a mixed Co^{2+}/Co^{3+} state. These chemically disordered yet coherent valence interfaces give rise to emergent magnetic phenomena that we interrogate using magnetic hysteresis measurements and polarized neutron reflectometry.We will highlight two exemplar pseudomorphic heterostructures:500°C (Sc,Mg,Co,Ni,Cu,Zn)O/300°C(Sc,Mg,Co,Ni,Cu,Zn)O(JSc/JSc)and500°C(Sc,Mg,Co,Ni,Cu,Zn)O/300°C(Cr,Mg,Co,Ni,Cu,Zn)O(JSc/JCr) where the abrupt, exceptionally large coherent strain and valence interface generates emergent magnetic macrostates inaccessible in equilibrium analogs.

4:15pm **TF-WeA-9 Probing Disorder and Metastability in Complex Oxide Thin Films with X-ray Absorption Spectroscopy**, ***Christina Rost***, Virginia Tech **INVITED**

X-ray absorption spectroscopy (XAS) has become an indispensable tool for understanding materials systems that exist far from equilibrium, particularly high entropy oxide and metastable thin films where conventional diffraction techniques often fail to capture the complexity of the local atomic structure. In these systems, functional behavior is frequently governed not by the average crystal structure, but by distributions in local coordination environments, oxidation states, and short-range chemical ordering that emerge from configurational disorder, strain, and kinetic stabilization during synthesis. XAS provides element-specific sensitivity to local bonding, coordination geometry, and electronic structure, enabling direct interrogation of chemically distinct local environments even in highly disordered or nanostructured films.Our work leverages synchrotron-based XAS to investigate the local structural evolution of compositionally complex oxide thin films synthesized under far-from-equilibrium growth conditions using pulsed laser deposition. These studies focus on understanding how compositional complexity, strain, and metastability influence local bonding environments and electronic structure in functional oxide systems. XAS enables direct probing of non-equilibrium valence states, local distortions, and chemically distinct coordination environments that are inaccessible through conventional crystallography alone. By combining XANES and EXAFS measurements with complementary electrical, ferroelectric, magnetic, and structural characterization, this approach establishes experimentally grounded links between local disorder and emergent functional behavior. These studies demonstrate that XAS is uniquely positioned to resolve the local structural

complexity that underpins next-generation functional oxide thin films designed through disorder, metastability, and non-equilibrium synthesis.

4:45pm **TF-WeA-11 Epitaxial Strain Engineering of Correlated Electronic States and Crystal Structure in Thin Film Nickelates**, ***Abigail Jiang***, Harvard University; *Maria Bambrick-Santoyo,* Massachusetts Institute of Technology; *Lopa Bhatt,* Cornell University; *Kyeong-Yoon Baek,* Harvard University; *Yi-Feng Zhao,* Arizona State University; *Dan Ferenc Segedin, Ari Turkiewicz, Jenna Hatmin, Grace Pan,* Harvard University; *Suchismita Sarker,* Cornell University; *Donald Walko,* Argonne National Lab; *Charles Brooks,* Harvard University; *Berit Goodge,* Max Planck Institute for Chemical Physics of Solids, Germany; *David Muller,* Cornell University; *Hua Zhou,* Argonne National Lab; *Antia Botana,* Arizona State University; *Julia Mundy,* Harvard University

Since the discovery of high-temperature superconductivity in cuprates, there have been long-standing efforts to understand and engineer the underlying electronic and crystal structure in superconducting materials. A new family of high-temperature superconductors was recently discovered in the *n=2,3* Ruddlesden-Popper (RP) nickelates [1,2]. When bulk crystals of these nickelates are placed under large hydrostatic pressures, superconductivity is stabilized concomitant with suppression of correlated density wave states, and also concomitant with crystallographic symmetry raising [1,2]. Thus far, compressive epitaxial strain has been demonstrated to mimic effects of pressure in *n=2* RP nickelates, leading to ambient-pressure superconductivity and equivalent structural changes in compressively-strained thin films [3,4]. However, ambient-pressure superconductivity and other analogous effects of strain have not yet been demonstrated in the *n=3* compound. Here, we use reactive oxide molecular beam epitaxy (MBE) to synthesize a series of *n=3* RP nickelate ($La_4Ni_3O_{10}$) thin films across a wide range of epitaxial strains [5]. Combining electronic transport, picoscale electron microscopy, and synchrotron X-ray diffraction techniques, we develop a strain-dependent electronic and structural phase diagram for *n=3*. Critically, we identify a structural distortion and corresponding crystal symmetry not present in *n=3* (or *n=2*) bulk crystals, highlighting key distinctions within the RP nickelate family towards engineering more robust superconducting materials, and demonstrating the unique capability of epitaxial synthesis to stabilize new metastable states in complex oxides.

[1] Sun, H. et al. *Nature* **621**, 493-498 (2023)

[2] Zhu, Y. et al. *Nature* **631**, 531-536 (2024)

[3] Ko, E.K. et al. *Nature* **638**, 935-940 (2025)

[4] Bhatt, L., Abarca Morales, E., Jiang, A.Y. et al. *Nature* **653**, 76-82 (2026)

[5] Jiang, A.Y.*, Bambrick-Santoyo, M.* et al. Submitted

5:00pm **TF-WeA-12 Optimizing Interfaces in High Entropy Carbides for Ultra High Hardness**, ***Nestor Marquez Rios***, *Tainara de Carvalho, Jon-Paul Maria,* The Pennsylvania State University

High chemical disorder compositions like high entropy transition metal carbides display potential for materials with high mechanical properties, high stability in extreme conditions and low synthesizability cost compared to the gold standard C-BN and diamond. These materials have shown the ability to impinge dislocation nucleation and dislocation motion at the nanoscale due to a non-uniform energy landscape; hardening the materials. A second mechanism that enhances hardness in materials is the interface addition in a multilayer system to impinge dislocation motion at the microscale. To tackle these two mechanisms, we have synthesized carbide films with high chemical disorder, (Ti Zr Hf Nb Ta) C, and multilayer them with TiC using 2 bipolar high-power impulse magnetron sputtering (HiPIMS) with methane gas as a carbon source.To determine the effect that synthesis parameters have on thin film crystalline structure, morphology and hardness we have being studying the effect reactive gas flow, temperature, HiPIMS conditions and interface quality and density in 600 nm thin films varying superlattice periods. X-ray diffraction (XRD), Scanning electron microscopy (SEM), Transmission electron microscopy (TEM) and Hardnes nanoindentation is employed to characterize the synthesized films.With XRD we established distinct superlattices with extremely sharp interfaces and with SEM we have being able to capture the ideal morphology to maximize hardness and minimize the possible morphology error in the hardness measurements. TEM Analysis is also used to study with higher resolution interface characteristics. We find these carbides superlattices display higher hardness values for films synthesized with smooth interfaceswith value of 35 GPa vs 30 GPa at sharp interface, and optimized methane flow. The morphology of the film is affected by the carbon content and showed that at stoichiometric carbide the films are smooth without columnar structure, with an increased grain density which in turn increases the dislocation motion resistance. Further development of the synthesis processmight lead to new hardened carbide structures with unexplored hardness values.

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