PCSI
Room Ballroom - Session PCSI-MoA2

Oxides I

Moderator: Jiefei Zhang, Argonne National Laboratory

4:30pm **PCSI-MoA2-31 Ferroelectric-Control of Magnetotransport in Ruddlesden-Popper Strontium Iridates**, ***Xia Hong***, University of Nebraska-Lincoln **INVITED**

The Ruddlesden-Popper 5*d* iridates $Sr_{n+1}Ir_nO_{3n+1}$ (SIO) represents a model system where the close competition between charge itinerancy, strong spin-orbit coupling, and onsite Coulomb energy can lead to a rich phase diagram. Nonvolatile voltage control of their electronic and magnetic properties is of intensive research interests for designing emergent quantum states and developing novel energy-efficient device applications, which is yet to be demonstrated. This talk discusses the tuning of magnetotransport properties of ultrathin SIO (n = 1, 2, ∞) films via interfacial ferroelectric $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) (Fig. 1a). Nonvolatile polarization control of metal-insulator transition (MIT) has been achieved in $SrIrO_3$ and $Sr_3Ir_2O_7$. Bulk $SrIrO_3$ is a correlated semimetal. In the 2D limit, $SrIrO_3$ thin films exhibit weak (anti)localization [1], which are modulated via PZT polarization reversal. Anomalous Hall and topological Hall effects are observed in PZT/$SrIrO_3$ close to the MIT, signaling emergent magnetism. All-oxide ferroelectric tunnel junctions (FTJs) composed of $LaNiO_3$ (LNO)/PZT/$Sr_3Ir_2O_7$ exhibit significantly enhanced electroresistance (ER) at room temperature (RT) compared with FTJs exploiting other correlated oxide electrodes (Fig. 1b), which can be attributed to polarization induced MIT in interfacial $Sr_3Ir_2O_7$ [2]. While single-layer Sr_2IrO_4 is a J-1/2 Mott insulator, the PZT/Sr_2IrO_4 heterostructure shows transport characteristic of a 2D correlated metal (Fig. 1c) due to the interfacial charge transfer effect [3]. The in-plane magnetoresistance (MR) and out-of-plane anisotropic MR show distinct temperature and magnetic field dependences for single-layer Sr_2IrO_4 and PZT/Sr_2IrO_4 (Fig. 1d), suggesting polarization controlled magnetic anisotropy and possible emergence of incipient ferromagnetism. These results showcase how charge-, lattice-, and symmetry-mediated interfacial coupling with ferroelectrics can be leveraged to induce emergent phenomena and design voltage-controlled functionalities in 5*d* iridates.

[1] L. Zhang et al, APL Materials **8**, 051108 (2020).

[2] Y. Zhang et al, Applied Physics Letters**125**, 102904 (2024).

[3] Y. Zhang et al, Advanced Physics Research **4**, 2400208 (2025).

5:00pm **PCSI-MoA2-37 UPGRADED: Observation of Field-Induced Interface Reactions in HfO_2-Based Ferroelectric Devices by HAXPES**, ***Takashi Onaya***, *Takahiro Nagata, Kazuhito Tsukagoshi, Toshihide Nabatame,* National Institute for Materials Science, Japan

Ferroelectric HfO_2-based materials are attractive for future nonvolatile memory devices due to their excellent CMOS compatibility and thickness scalability (<10 nm) [1]. However,it remains critical reliability issues in practical applications such as fatigue and hard breakdown during electric field cycling[2]. These endurance degradations were thought to be mainly caused by the formation of defects such as oxygen vacancies (V_O) in HfO_2-based films[3], butit has been unclear why V_O increases during field cycling. In this study, we investigated the interface reaction between the ferroelectric $Hf_xZr_{1-x}O_2$ (HZO) film and TiN electrode during field cycling by synchrotron hard X-ray photoelectron spectroscopy (HAXPES) to clarify the origin of endurance degradation.

Three different switching cycles, such as pristine for 10^0, wake-up for 10^4, and fatigue for 10^8, were applied to TiN/HZO (10 nm)/TiN ferroelectric capacitors. Then, chemical states of Ti at the HZO/TiN bottom-electrode interface were evaluated by synchrotron HAXPES with the X-ray energy of 8 keV after removal of TiN top-electrodes. In the pristine state, Ti-O (~4969 eV) and Ti-N peaks (~4966 eV) of Ti 1s spectra were observed, indicating that a TiO_xN_y interfacial-layer (IL) was formed at the HZO/TiN interface. In the wake-up state, no difference of Ti 1s spectra was observed between the pristine and wake-up states. Therefore, the HZO/TiN interface did not change during wake-up field cycling. In the fatigue state, on the other hand, the Ti-O and TiO_xN_y components increased compared to those of the pristine and wake-up states. This indicates that an oxygen-rich and thicker TiO_xN_y-IL formed at the HZO/TiN interface. Therefore, a field-induced interface reaction accompanied by oxygen diffusion from the HZO film to TiN electrodes should occur during fatigue field cycling, resulting in additional V_O formation in the HZO film. In conclusion, we clarified one of the origins of endurance degradation by evaluating field-induced interface reactions using synchrotron HAXPES.

[1] T. S. Böscke et al., Appl. Phys. Lett. **99**, 102903 (2011).

[2] M. Pešić et al., Adv. Funct. Mater. **26**, 4601 (2016).

[3] W. Hamouda et al., Appl. Phys. Lett. **120**, 202902 (2022).

5:15pm **PCSI-MoA2-40 Ferroelectric Control of Metallicity and Magnetism in Ultrathin Sr_2IrO_4 Films**, ***Yuanyuan Zhang***, *Qiuchen Wu, Yifei Hao, Peace Adegbite, Peter Dowben, Xia Hong,* University of Nebraska-Lincoln

The Ruddlesden-Popper 5*d* iridate Sr_2IrO_4 is an antiferromagnetic Mott insulator in which the electronic, magnetic, and structural states are strongly coupled. Achieving nonvolatile voltage control of its electronic and magnetic properties is of significant fundamental and technological interests. Here, we report the ferroelectric control of metallicity and magnetism in ultrathin Sr_2IrO_4 via interfacial ferroelectric $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) [1]. Reversing the polarization of PZT induces nonvolatile, reversible resistance modulation in Sr_2IrO_4. The conductance of PZT/Sr_2IrO_4 in both polarization states is predominantly above the quantum conductance of e^2/h and exhibits In(T) behavior, which is characteristic of a 2D weakly localized metal. This behavior is in sharp contrast to the 3D variable range hopping (VRH) behavior observed in single-layer Sr_2IrO_4, suggesting that interfacing with PZT induces a metal-insulator transition and dimensionality crossover. X-ray photoelectron spectroscopy reveals significant charge transfer at the interface, corroborated by Hall effect measurements, which can account for the enhanced metallicity.

In the single-layer sample, the in-plane MR exhibits low-field hysteresis associated with the antiferromagnetic spin-flip transition below 160 K, together with a large negative MR. In contrast, the heterostructure shows negligible low-field hysteresis with positive MR dominating down to 6 K. Below 6 K, the MR exhibits an opposite field-dependence compared with the single-layer sample, i.e., positive MR at low field followed by negative MR at high field, with a magnitude one order smaller. In addition, field-sweep-rate-dependent resistance dip is observed in the heterostructure, possibly associated with an incipient ferromagnetic state.

Switching the polarization of PZT modulates the out-of-plane anisotropic magnetoresistance R_{AMR}. For the polarization up state, unusual multi-level resistance pinning appears in R_{AMR} below 30 K, indicating polarization modulation of magnetic anisotropy. These results reveal the charge-meditated emergent phenomena at the ferroelectric/Sr_2IrO_4 heterointerface.

[1] Y. Zhang, Q. Wu, Y. Hao, et al., Adv. Phys. Res. 4, 2400208 (2025).

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