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Tuning Metal-Insulator Transitions in Ultra-Thin Correlated Materials

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14. ABSTRACT
During the final year of the grant, we have demonstrated reversible tunability of coupled phases transitions, including a perpendicular ferromagnetic transition, a metal insulator transition, an optical transition and a structural phase transition, by electrolytic gating of SrIrO3/(La,Sr)MnO3 superlattices. All of these effects are found to exist at room temperature. In addition, we have combined strongly spin-orbit coupled SrIrO3 with CoFeB to find that we can manipulate the magnetic moments in the ferromagnetic CoFeB layer via microwave charge current in the SrIrO3 layer. The versatility of the SrIrO3 layer for a wide range of functionalities make it a promising material for applications of interest to the Air Force.

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The *main research objective* of the proposed program has been (i) to develop a class of 4d and 5d transition metal oxides on the verge of an MIT and (ii) to determine the extent to which its functional properties can be tuned by an external parameter.

During the *final year* of the grant, we have demonstrated reversible tunability of coupled phases transitions, including a perpendicular ferromagnetic transition, a metal insulator transition, an optical transition and a structural phase transition, by electrolytic gating of $SrIrO_3$/(La,Sr)MnO_3 superlattices. All of these effects are found to exist at room temperature. In addition, we have combined strongly spin-orbit coupled $SrIrO_3$ with CoFeB to find that we can manipulate the magnetic moments in the ferromagnetic CoFeB layer via microwave charge current in the $SrIrO_3$ layer. The versatility of the $SrIrO_3$ layer for a wide range of functionalities make it a promising material for electronic applications of interest to the Air Force.

• *Reversible Coupled Phase Transformations at Room Temperature in (La,Sr)MnO_3/$SrIrO_3$ superlattices.* One of the important results of this past year has been the demonstration of *reversibility* in the coupled phase transformations of perpendicular ferromagnetism, electrical transport, optical transparency and structure. Last year we had found that charge transfer from the iridate to the manganite, along with oxygen octahedral rotations at the interface, gives rise to interfacial perpendicular ferromagnetism in our superlattices and that this charge-based mechanism translated into electronic and optical phase transitions. However more recently we have demonstrated reversibility at room temperature and directly correlated it with a reversible structural transition associated with the dual ion transfer of oxygen and hydrogen ions. These superlattice materials with perpendicular magnetic anisotropy (PMA) may enable spin-torque-driven magnetic memory and logic devices.

More specifically we have performed electrolytic gating measurements of superlattices comprised of alternating unit-cells of $La_{0.2}Sr_{0.8}MnO_3$ and $SrIrO_3$

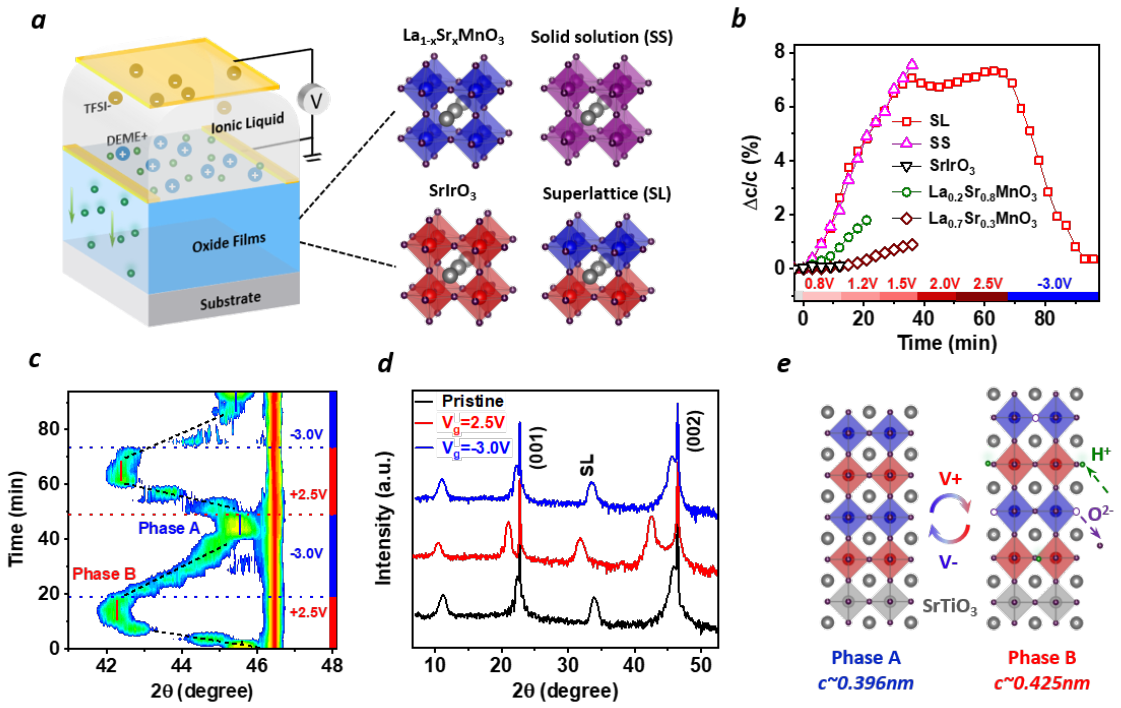

Figure 1. (a) Schematic of ionic liquid gating that induces ion transfer between oxides and ionic liquid. We studied manganate ($La_{0.2}Sr_{0.8}MnO_3$ and $La_{0.7}Sr_{0.3}MnO_3$) and iridate ($SrIrO_3$) films, solid solution ($Sr(Mn_{0.5}Ir_{0.5})O_3$) films and superlattices ($[(La_{0.2}Sr_{0.8}MnO_3)_1(SrIrO_3)_1]_{20}$) on $SrTiO_3$ substrates. (b) Modulation of out-of-plane lattice constant (Δc/c) during voltage cycling (with incremental positive voltages to +2.5 V and -3.0 V) for different samples. The lattice constant was extracted from in-situ measurements of the (002) peak position. (c) *In-situ* X-ray diffraction results of the superlattices around the (002) peak during ILG in repeated voltage cycling (sequence of +2.5 V, -3.0 V, +2.5 V, -3.0 V), showing a reversible electric-field-controlled transformation between two phases of the superlattices. (d) Full-range X-ray diffraction of a superlattice in as-grown pristine state, positively gated state and reversibly gated state. (e) Schematic of the reversible phase transformation mediated by dual-ion (both H^+ and O^{2-}) transfer.

($[(La_{0.2}Sr_{0.8}MnO_3)_1(SrIrO_3)_1]_{20}$), as well as single-phase films of $La_{0.2}Sr_{0.8}MnO_3$, $La_{0.7}Sr_{0.3}MnO_3$ and $SrIrO_3$. The ILG induces non-volatile reversible changes in the crystal structure from the original phase A to a new phase B. Phase B has up to a 7% lattice expansion along the (001) direction compared to phase A (Figure 1).

Through secondary ion mass spectrometry (SIMS), we confirm the transfer of both hydrogen and oxygen ions. SIMS results of the superlattice in phase B (red) reveal a significantly higher concentration of hydrogen than that of phase A (blue), confirming the incorporation of hydrogen ions in phase B. At the same time, there is evidence for oxygen ion extraction (oxygen vacancy creation) in phase B. By thermally annealing both phase A and B samples in $^{18}O_2$ gas, we find that the phase B sample after thermal annealing in $^{18}O_2$ shows a dramatic increase in ^{18}O signal throughout the entire sample, while the phase A sample only shows an increase at the surface (Figure 2). The extra incorporation of ^{18}O indicates that ILG also creates oxygen vacancies in phase B. It is noted that the SIMS results are consistent with polarized neutron reflectivity results, revealing the accumulation of oxygen vacancies and hydrogen ions in phase B.

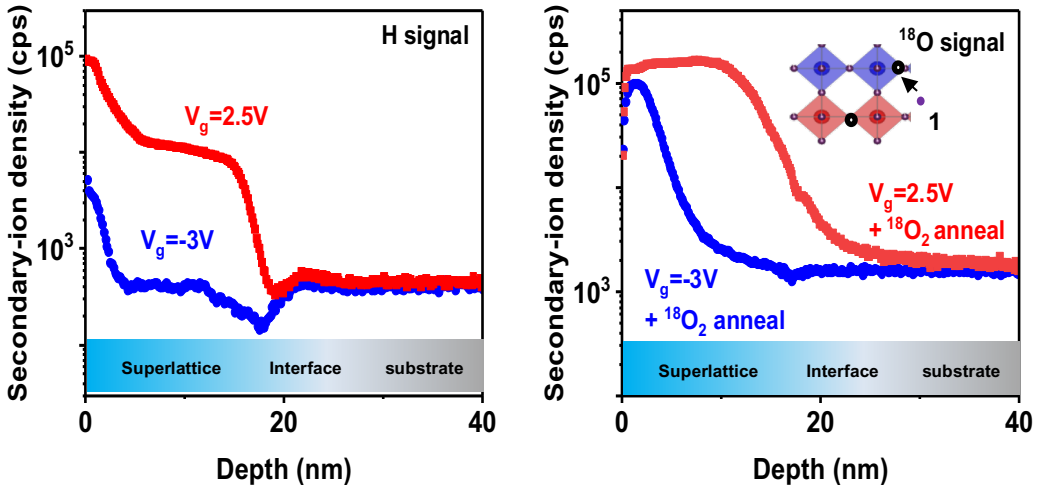

Figure 2. (a) Depth profiles of H and O signals in the two phases of the superlattices, by using secondary-ion mass spectrometry. The H signal was obtained in phase A (red) and B (blue) of the superlattices under ILG. The Ti signal from the substrate indicates the interface position. (b) Depth profiles of ^{18}O signals in the two phases of the superlattices after thermal annealing in $^{18}O_2$. To measure ^{18}O signal, the superlattices were first stabilized in phase A and B under ILG, and then thermally annealed in $^{18}O_2$ gas.

The large structural changes induced by ILG should be accompanied by changes in cation valence and oxygen bonding as measured by x-ray absorption spectroscopy at the Mn L-edge, Ir L-edge and O K-edges. Indeed the Mn and Ir absorption peaks show a shift to lower energies corresponding to decreases in Mn and Ir valence respectively.

Although it is challenging to precisely quantify the local concentration of ions, we note that the magnitude of lattice expansion (~7%) and valence change are comparable to the highest reported values in singe-phase perovskite TMOs (e.g. $H_xSrCoO_{3-\delta}$ with large transfer of hydrogen ($x \sim 1$) and oxygen ($\delta \sim 0.5$) ions). Strikingly the large structural and chemical changes are accompanied by a concurrent metal-insulator transition, suppression of perpendicular ferromagnetism and enhancement of optical transparency. These phenomena are not otherwise observed in the individual constituent oxides or their solid solutions. Our findings reveal a hitherto-unexplored strategy to develop complex oxides, in which the electrolyte-based ionic control can provide extensive tunability that is crucial for electronic/spintronic, energy and environmental applications.

• *Low Energy Magnetization Switching by Spin Hall Oxide $SrIrO_3$*

We have also recently demonstrated that $SrIrO_3$ is an effective spin-to-charge conversion material that enables us to control the magnetic moment of a neighboring ferromagnet. More specifically, we have found that $SrIrO_3$ can exhibit a spin Hall angle (i.e., the ratio of charge to spin current) as high as 1.6. To date, spin Hall angles of about 0.5 and magnetization switching of a neighboring ferromagnetic $SrRuO_3$ layer have been verified in $SrIrO_3$-based bilayers. In our study, we have demonstrated much more efficient magnetization switching of an adjacent ferromagnetic CoFeB layer. We also show for the first time in a $SrIrO_3$-based system that a unidirectional magnetoresistance can be measured and is indicative of a current-induced spin accumulation at the interface that is coupled to

the electrical conductivity. Together these results indicate a route to controlling resistance and detecting magnetization in a two-terminal geometry.

Publications

- M.J. Veit, Di Yi, Y. Suzuki, "Near Room Temperature Quantum Spin Hall Effect in a Perovskite Oxide Heterostructure," under review at *Science* (2019).

- Di Yi, Yujia Wang, Olaf M. J. van't Erve, Hongtao Yuan, Michael J. Veit, Alpha T. N'Diaye, Padraic Shafer, Elke Arenholz, Yongseong Choi, Pu Yu, Berend T. Jonker and Yuri Suzuki, "Electric Field Control of Coincident Magnetic, Electronic and Structural Transition in $SrMnO_3/SrIrO_3$ Superlattices," under review at *Nature Communications*.

Talks

- "Control of Emergent Magnetic Properties in Layered Superlattices," Di Yi (*invited talk*), 2018 European Materials Research Society Meeting, Warsaw, Poland, September 2018.
- "Controlling emergent magnetic properties in iridate-based superlattices", Di Yi (*invited talk*), NIST Center for Neutron Research, Gaithersburg, MD, October 2018.
- "Emergent Magnetic Phenomena via Oxide Epitaxy," Di Yi (*invited talk*), Physics Colloquium, Department of Physics, University of Arkansas, Fayetteville, AR, February 2019.
- "Electrical control of phase transitions through ion transfer in oxide superlattices", Di Yi, Yujia Wang, Hongtao Yuan, Olaf M. J. van't Erve, Michael J. Veit, Purnima Balakrishnan, Yongseong Choi, Jian Liu, Alpha T. N'Diaye, Padraic Shafer, Elke Arenholz, Berend T. Jonker, Pu Yu and Yuri Suzuki, American Physical Society Meeting, Boston, MA, March 2019.
- "Spintronic Functionality in Complex Oxides: Emergent Magnetic Phenomena and Electric Field Control," Di Yi (*invited talk*), Department of Materials Science and Engineering, North Carolina State University, Raleigh, NC, April 2019.

Significant Department of Defense Collaborations

In the past year, we have collaborated with Drs. Brandon Howe, Michael McConney, Krishnamurthy Mahalingham of AFRL and Michael Osofsky and Berry Jonker of NRL. The AFRL collaboration has focused on structural characterization of our manganite/iridate superlattices by transmission electron microscopy and electron energy loss spectroscopy. In addition, we have also provided ferromagnetic resonance characterization of low loss spinel ferrite thin films synthesized at AFRL for magnetoelastic applications.