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Interfacial magnetic coupling in ultrathin all-manganite La0.7Sr0.3MnO3-TbMnO3 superlattices
Y. F. Tian, O. I. Lebedev, V. V. Roddatis, W. N. Lin, J. F. Ding, S. J. Hu, S. S. Yan, and T. Wu

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Interface and dipolar anisotropies. Transition metal oxides with the film thickness, and the ground states in ultrathin films of spins in magnetic materials are well known to evolve with geometrically confined doping in such artificial low-dimensional systems.22–29 Epitaxial heterostructures as a result of expectation of emergent functionalities. The long-range collective behaviors become thinner, which has been ascribed to both extrinsic and intrinsic effects such as structural and compositional inhomogeneities, interfacial charge transfer, and strain-induced orbital reconstruction. In a flurry of recent studies, ultrathin manganite films were incorporated in epitaxial heterostructures as a result of expectation of emergent physics such as tailored interfacial magnetic coupling and geometrically confined doping in such artificial low-dimensional systems.30–32 As a result of the competing exchange interactions, TMO displays sinusoidal antiferromagnetic Mn\(^{3+}\) spin ordering at the Néel temperature \(T_N\) ~ 41 K, spiral Mn\(^{3+}\) spin ordering at the ferroelectric transition temperature \(T_{lock}\) ~ 28 K, and long range ferromagnetic Tb\(^{3+}\) spin ordering below \(T_{Tb}\) ~ 7 K. Orthorhombic-structured TMO is compatible with other perovskites, and epitaxial thin films have been fabricated.33,34 Recently, TMO-based heterostructures including rectifying junctions and manganite bilayers were investigated.35–37 The correlation effects involving multiple degrees of freedom are expected to be enhanced at interfaces in ultrathin heterostructures. However, so far there has been no report on the fabrication and characterization of TMO-based SLs.

In this work, we fabricated a series of epitaxial manganite SLs composed of ultrathin LSMO and TMO layers, and studied their magnetic properties. Remarkably, sizable low-temperature magnetism with hysteresis was stabilized in the SLs down to a LSMO layer thickness of two unit cells (u.c.), which is accompanied by drastically enhanced coercive field of more than 3000 Oe. Furthermore, exchange bias was observed in the SLs, which can be attributed to the strong magnetic coupling across the LSMO-TMO interface.

Epitaxial [m/n]x SLs (as schematically shown in Fig. 1(a)), consisting of m.u.c. of LSMO and n.u.c. of TMO, repeated x times, were grown on TiO\(_2\)-terminated (001) SrTiO\(_3\) (STO) single-crystal substrates using pulsed laser deposition (PLD) technique assisted by reflection high-energy electron diffraction (RHEED). The growth took place at 750°C under an oxygen pressure of 0.05 millibars. A laser fluence of ~1.5 J/cm\(^2\) was used.
used for the depositions, and the frequency of the excimer laser was 1 and 2 Hz for LSMO and TMO, respectively. After deposition, samples were first cooled down to 600°C under an oxygen pressure of 100 millibars, and then kept at 600°C for 1 h at 400 millibars oxygen before cooling down to room temperature.

The structure of SLs was studied by using a double corrected transmission electron microscope (TEM, JEM-ARM 200F, JEOL Ltd.) equipped with a high-angle annular dark-field (HAADF) and CENTURIO energy-dispersive X-ray spectroscopy (EDX) detector. Cross-section TEM sample was prepared using a focus ion beam instrument (FEI Helios 600 NanoLab DualBeam). The magnetic properties of the LSMO-TMO SLs were measured using a superconducting quantum interference device (SQUID) magnetometer. For the field-cooled (FC) and zero-field-cooled (ZFC) magnetization measurements, the sample was cooled down from 300 K to low temperature with and without the magnetic field, respectively. Both the ZFC and FC magnetization versus temperature (M-T) curves were measured during the warming process.

Figure 1(b) shows a typical result of in-situ RHEED intensity oscillation recorded during the growth of [2/2]10 LSMO-TMO SL. A clear oscillation of the specular reflection spot indicates a continuous layer-by-layer growth mode for both LSMO and TMO layers. However, the RHEED intensity generally decreases when the growth of TMO units starts indicating that compared to LSMO it is more difficult to maintain the layer-by-layer growth mode for TMO. The chemical composition and the SL structure were further confirmed for a thicker SL by the element sensitive second ion mass spectrometry (SIMS) measurements. As shown in Figure 2, the periodic oscillations of cation concentrations were clearly resolved, which are consistent with the designed SL structure. Furthermore, we did not observe any impurity elements above the detection level.

The results of TEM structural investigation of a typical LSMO-TMO SL are presented in Figure 3. The low magnification image in Figure 3(a) clearly shows that the SL is coherent and uniform without any precipitate of secondary phases in the examined area. The selected area electron diffraction (SAED) pattern in Figure 3(b) confirms the lattice structure and epitaxial growth. The epitaxial relationships were determined as [001]_{STO} || [001]_{LSMO} || [110]_{TMO} and [100]_{STO} || [100]_{LSMO} || [001]_{TMO}. Figure 3(c) shows the aberration-corrected high resolution HAADF-scanning transmission electron microscopy (STEM) image of the LSMO-TMO SL. The bright spots correspond to the positions of atomic columns and their brightness roughly corresponds to Z^2 with Z as the atomic number of chemical element. As a result, the HAADF-STEM technique allows us to distinguish the LSMO and TMO layers, and the deviation of layer thickness was found to be within 1 u.c. at the interfaces.

Figure 4(a) shows the magnetic hysteresis loops for various SLs measured at 10 K. As the most notable feature, all the SLs show hysteresis loops with finite remnant magnetic moments, including the SL with 2 u.c. of LSMO in the repeating unit, i.e., samples [2/2]_{10} and [2/4]_{32}. In this work,
we did not fabricate any SL with layers of single u.c. because of the cation mixing at the interfaces cannot be completely excluded. Assuming the magnetic signal solely comes from the LSMO layers, we re-plot the magnetization data in Figure 4(b) with a unit of $\mu_B$/u.c. and the saturation magnetization values of all the SLs are roughly at the same level. In general, the saturation magnetization of the SLs is lower than the value of 3.7 $\mu_B$/u.c. expected for bulk LSMO, which indicates suppression of collinear spin ordering in the LSMO-TMO SLs. Nevertheless, the stabilization of magnetic order with hysteresis in such ultrathin LSMO-TMO SLs is quite remarkable, considering that the critical LSMO layer thickness to exhibit ferromagnetism is 3 u.c. for LSMO single layers. Symmetry breaking, charge transfer, and spin frustration at the LSMO-substrate and LSMO-air interfaces are often cited to account for the suppressed magnetism in ultrathin LSMO films. At the LSMO-TMO interface, the Mn ions interact not only with each other like in the double-exchange LSMO but also with the spin sublattices in TMO. Particularly, in the [2/n] SLs, all the magnetic Mn ions in LSMO interface with the TMO layers. The proximity of TMO apparently plays the decisive role in stabilizing the magnetic order with hysteresis in such ultrathin LSMO-TMO SLs. Nevertheless, the stabilization of magnetic order with hysteresis in such ultrathin LSMO-TMO SLs can be obtained by examining the temperature dependence of various LSMO-TMO SLs can be obtained by examining the temperature dependence of various LSMO-TMO SLs.

Careful examinations of the hysteresis loops reveal the emergence of exchange bias $H_E$ together with a giant enhancement of coercivity $H_C$ in the LSMO-TMO SLs. Here, $H_C$ is defined as $H_C = |H_+ - H_-|/2$, where $H_+$ and $H_-$ denote the right and left coercivity field, respectively. As shown in Figure 4(d), among the SLs we measured the [4/8]$_{16}$ sample shows the highest exchange bias of $\sim 170$ Oe at 10 K. Its value of $H_C$ at 10 K is $\sim 3200$ Oe; in comparison, $H_C$ is only $\sim 90$ Oe for 8 u.c. LSMO thin films. A similar effect of coercivity enhancement was previously reported in other manganite-based heterostructures such as LSMO/La$_2$CuO$_4$ bilayer (705 Oe at 5 K) and LSMO/BiFeO$_3$ bilayer (850 Oe at 7 K). This observation of the enhanced magnetic anisotropy in LSMO-TMO SLs unambiguously underscores the significant exchange interaction and spin pinning across the LSMO-TMO interfaces.

Considering that the exchange bias is highest in the [4/8]$_{16}$ LSMO-TMO SL, we measured its temperature dependent magnetic properties. The data in Figure 5(a) reveal that the irreversible temperature decreases with increasing magnetic field, which are consistent with the existence of spin frustration. Figure 5(b) shows the magnetization vs. magnetic field data measured from 5 to 300 K. Both coercivity and exchange bias quickly decreases on increasing temperature, indicating weakened exchange coupling in the SLs due to thermal excitation.

More insights on the magnetic interactions in LSMO-TMO SLs can be obtained by examining the temperature dependence of $H_C$ and $H_E$ as shown in Figure 6. Remarkably, the exchange bias emerges near the $T_N$ of bulk TMO. In macroscopic TMO single crystals, sinusoidal antiferromagnetic Mn$^{3+}$ spin ordering occurs at $T_N \sim 41$ K, which however may not fully develop in ultrathin SLs with TMO layers of a few u.c. due to the symmetry breaking and nanoscale confinement. Nevertheless, the temperature dependence shown in Figure 6 suggests that the strong magnetic interaction and spin pinning only appear at low temperatures, and the onset temperature is intriguingly close to the $T_N$ of bulk TMO. In addition, as shown in Figure 6(a), $H_E$ shows the exponential temperature dependent decay following a phenomenological expression: $H_E = H_E^0 \exp\left(-T/T_0\right)$. This exponential temperature dependence has previously been observed in diverse systems such as manganite nanoparticles, bilayer, and superlattices, which can be ascribed to the competing magnetic interactions and spin frustration at the interfaces. In contrast, $H_C$ deviates from this exponential law at low temperatures (Figure 6(b)).
suggesting that $H_E$ and $H_C$ in the low-temperature regime do not share exactly the same origin.

It is insightful to compare the magnetic properties of LSMO-TMO SLs with those of bilayers. Since SLs contain multiple interfaces, much more than the single interface in bilayers, we expect stronger interfacial magnetic coupling in SLs. Indeed, in the LSMO (8 u.c.)-TMO (40 nm) bilayer, $H_E$ is 42 Oe and $H_C$ is 1200 Oe at 5 K.\textsuperscript{35} On the other hand, in the [4/8]$_{16}$ SL sample, both $H_E$ and $H_C$ are greatly enhanced, i.e., $H_E$ is found to be 320 Oe and $H_C$ is 3320 Oe at 5 K. The remarkable enhancement illustrates a stronger interfacial exchange interaction in the SLs compared to the bilayer counterparts. This clearly underscores ultrathin SLs as a powerful approach towards achieving effective exchange coupling in manganite heterostructures. The fact that the largest exchange bias and coercivity enhancement was observed in the [4/8]$_{16}$ SL suggests that a certain layer thickness of TMO is needed to achieve effective exchange coupling at the TMO-LSMO interfaces. On the other hand, thicker TMO layers are harder to grow in the layer-by-layer mode, and the rougher interfaces may result into the lower saturation magnetization. For example, after the saturation magnetization is normalized according to the total LSMO thickness, the value of the [4/4]$_5$ SL (2.93 $\mu_B$/u.c.) is higher than that of [4/8]$_{16}$ SL (2.25 $\mu_B$/u.c.). Furthermore, nanoscale domains were observed in TMO thin films,\textsuperscript{44} and their interactions with the magnetic domains in LSMO will be investigated in the future.\textsuperscript{45,46}

In summary, the emergence of significantly enhanced coercivity and exchange bias in all-manganite LSMO-TMO SLs underscores such unit-cell-controlled heterostructures as a viable route towards tailored artificial magnetic materials. It is quite remarkable that sizable magnetization with hysteresis can be stabilized in LSMO-TMO SLs with the layers being as thin as two u.c. The competing exchange coupling mechanisms within the spin sublattices are responsible for the observed rich magnetic behaviors. We believe that insights on the magnetic phenomena in such artificial heterostructures and advancements in designing interfacial coupling will lead to technological breakthroughs in exploring future magnetic materials.

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