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Exchange coupling and coercivity enhancement in cuprate/manganite bilayers

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We report on the magnetic properties of cuprate/manganite bilayers composed of antiferromagnetic (AFM) La_2CuO_4 and ferromagnetic $\text{La}_{0.70}\text{Sr}_{0.30}\text{MnO}_3$. The temperature dependent magnetization data indicate an interfacial spin-glass state. Furthermore, the bilayer exhibits significant enhancement of coercivity compared to the $\text{La}_{0.70}\text{Sr}_{0.30}\text{MnO}_3$ single layer and reaches 705 Oe at 5 K. The dependence of coercivity on the AFM layer thickness indicates that the enhancement cannot be explained by the interfacial charge diffusion. Our results suggest that the exchange coupling between Mn and Cu spins and the magnetic frustration at the $\text{La}_2\text{CuO}_4/\text{La}_{0.70}\text{Sr}_{0.30}\text{MnO}_3$ interface must be considered to harness the properties of cuprate/manganite heterostructures. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4788719>]

Oxide heterostructures composed of ferromagnetic (FM) manganites and superconducting cuprates have attracted great attention for their importance in fundamental physics and potential applications.^{1–6} Interesting results have been observed in the cuprate/manganite heterostructures, such as spin injection,⁷ proximity effects,^{4–6,8,9} giant magnetoresistance,¹⁰ and interlayer magnetic coupling.^{2,6,11} Recently, the direct coupling between Mn and Cu spins at the interface was reported in superlattices of $\text{La}_{2/3}\text{Ca}_{1/3}\text{MnO}_3$ (LCMO) and $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO).^{2,12} It was found the coupling between Mn and Cu ions across the interface induces the orbital reconstruction in the interfacial CuO_2 plane, and as a result, the Cu ions gain ferromagnetic polarization.^{2,12} The magnetic coupling in such heterostructures can affect the superconducting critical temperature, and theoretical and experimental studies revealed either suppression or enhancement of superconductivity in YBCO/LCMO superlattices.^{6,13–15} In turn, the existence of the cuprate layer in the heterostructures can affect the magnetic properties of the manganite layer. There are a few reports on the exchange coupling in cuprate/manganite heterostructures. In a study on the magnetic properties of YBCO/ $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ superlattices,¹⁶ exchange bias was observed in the hysteresis loop and explained by the hole charge transfer from the YBCO layer to the $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ layer. A similar phenomenon was observed in YBCO/LCMO superlattices;¹¹ an AF order was suggested to form in the LCMO layer due to the interface disorder originated from factors including interdiffusion, stress, or oxygen stoichiometry. It should be noted that in these previous studies, the superconducting cuprates are optimally doped and the Cu ions in the CuO_2 planes have no long-range spin ordering. On the other hand, it is interesting to investigate the exchange coupling between the Mn and Cu ions in cuprate/manganite heterostructures with the cuprate layer itself possessing the long-range antiferromagnetic (AFM) order.

In this work, we studied the magnetic properties of cuprate/manganite bilayer composed by the parent material of

La-based high-temperature superconductors, La_2CuO_4 (LCO), and the prototypical FM manganite, $\text{La}_{0.70}\text{Sr}_{0.30}\text{MnO}_3$ (LSMO). It is well known that LCO is a two-dimensional Heisenberg antiferromagnet, and a weak inter-plane interaction gives rise to a three-dimensional long-range antiferromagnetism below the Néel temperature.^{17,18} Fig. 1(a) illustrates the ideal LCO/LSMO interface without any chemical and structural imperfection. It can be conceived immediately that the Mn-O-Cu coupling at the interface may cause the competition between the AFM and FM orders. Experimentally, we found that there exists strong Mn-O-Cu exchange coupling at the LCO/LSMO interface, which gives rise to a spin-glass state and enhanced coercivity.

LCO/LSMO bilayers were grown on atomically flat TiO_2 -terminated SrTiO_3 (001) substrates by pulsed laser deposition. The frequency of the excimer laser was 1 Hz and 3 Hz for LSMO and LCO, respectively. The fluency of the laser is 1.2 J/cm^2 . Both layers were grown at a temperature of

FIG. 1. (a) Idealized illustration of the atomic and spin configurations at the LCO/LSMO interface. (b) XRD θ - 2θ data of the bilayer. The stars (*) denote the peaks of STO substrate. The background picture is an atomic force microscopy image of $3 \mu\text{m} \times 3 \mu\text{m}$ for a single layer LSMO film. (c) RSM data around the (103) Bragg reflection of STO for the bilayer.

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750 °C under 20 Pa O₂ pressure. The reflection high energy electron diffraction (RHEED) was used to precisely grow 8 unit cell (~3 nm) LSMO, and the regular RHEED oscillation reveals a layer-by-layer growth mode. After the LCO layer was deposited on top of the LSMO layer, the samples were *in situ* annealed at 600 °C in 1 atm O₂ atmosphere for 1 h before cooling down to room temperature. For comparison, LCO and LSMO single layers were grown on SrTiO₃ (STO) substrates under the same conditions. For the field-cooled (FC) and zero-field-cooled (ZFC) magnetization measurements, the sample was cooled down from 350 K to the desired temperature with and without magnetic field, respectively. Both the ZFC and FC magnetization vs temperature (*M-T*) curves were measured on warming. The hysteresis loops were measured after the FC process from 350 K to the determined temperatures under a magnetic field of 5 kOe.

The x-ray diffraction (XRD) data in Fig. 1(b) show that the bilayer is c-axis orientated. No diffraction peak of LSMO was observed in the $\theta - 2\theta$ scan due to its ultrathin thickness. However, like suggested in several previous reports on LSMO single layers,^{19,20} the coherent layer-by-layer growth mode of 8 unit cell LSMO implies a fully strained state. The atomic force microscopy image of a 3 nm LSMO film in Fig. 1(b) shows a clear step-and-terrace surface, which reflects the layer-by-layer growth mode of LSMO. Fig. 1(c) shows the reciprocal space mapping (RSM) data of the bilayer around the STO (103) peak. It reveals that the lattice parameters of LCO are $a = 3.823$ Å and $c = 13.126$ Å. Considering that for the bulk LCO single crystals at room temperature, $a = 3.786$ Å and $c = 13.153$ Å,¹⁷ the LCO layer is under an in-plane tensile strain of 0.977%.

Figure 2(a) shows the magnetization vs. temperature (*M-T*) curves under 0.1 kOe for the bilayer of 40 nm LCO and 3 nm LSMO. Also shown in the figure are the data for

the reference 3 nm LSMO and 40 nm LCO single layers. The ferromagnetic transition temperature T_C determined from the differential of *M-T* curves for bilayer and LSMO film are 143 K and 181 K, respectively. The T_C of 8 unit cell LSMO is much lower than that of bulk single crystals (354 K),²¹ but it is among the range of reported values in literature.²² The origin of the decrease T_C in ultrathin LSMO films is still a topic of debates and out of the scope of this work. The Néel temperature T_N for the antiferromagnetic LCO layer cannot be determined due to its weak magnetic signal. Clearly, the magnetism of the bilayer is mainly contributed by the LSMO layer. But different from the single layer data, the ZFC *M-T* curve of the bilayer is featured with a pronounced peak at 74 K, which is accompanied by a bifurcation between the ZFC and FC curves. This phenomenon is often considered as the signature of spin-glass state due to spin frustration.^{23–26} As shown in Fig. 2(c), relationship between the peak temperature (T_p) of the ZFC *M-T* curves and the applied magnetic field appears to follow the Almeida-Thouless (AT) line, which is commonly observed in Ising spin systems.²³ The confirmation of a spin-glass state indicates the existence of spin frustration in the bilayer. From the fitting, we deduced a spin-glass freezing temperature T_f of 89.6 K. We also tried to further investigate the spin-glass state in the bilayer using *ac* susceptibility measurements. However, the signal of the bilayer was too small to achieve reliable data.

The hysteresis *M-H* loops for LCO/LSMO bilayers along with the LCO and LSMO reference single layers were measured after the FC process from 350 K under 5 kOe, and the data are shown in Fig. 2(d). It can be seen that compared to the LSMO single layer the bilayer shows a significant enhancement of the coercivity H_C from 120 Oe to 705 Oe. As expected, the magnetization of the LCO single layer is almost zero. Overall, the spin-glass state and H_C enhancement in

FIG. 2. (a) *M-T* data measured on the LCO/LSMO bilayer as well as on the LSMO and LCO single layers. The open and filled symbols represent the ZFC and FC curves, respectively. (b) *M-T* data of the LCO/LSMO bilayer sample measured under different magnetic fields, $H = 0.05, 0.1, 0.2, 0.3, 0.5$, and 1 kOe. The solid and dashed lines are the ZFC and FC curves, respectively. (c) AT line plotted for the field dependence of peak temperature T_p estimated from the ZFC *M-T* curves in (b). (d) *M-H* curves at 5 K for the LCO/LSMO, LSMO, and LCO samples.

Fig. 2 clearly suggests a strong exchange coupling between the AFM LCO layer and the FM LSMO layer. A similar enhancement of H_C was also reported for bilayers of AFM BiFeO₃ and LSMO.²⁷ For the LCO/LSMO bilayer, the AFM superexchange energy between nearest neighbour Cu spins in the CuO₂ plane, J_{AFM} , is ~ 133 meV,²⁸ while the FM double exchange coupling between the nearest neighbour Mn spins in LSMO, J_{FM} , is ~ 80 meV.²⁹ Furthermore, the antiferromagnetic interaction across the interface between Cu and Mn spins J_{int} was estimated to be ~ 40 meV.² Although it is difficult to elucidate the exact spin configuration of the interfacial ions, it is clear that the competition among the three exchange energies will cause a certain degree of frustration among the spin lattices; in particular, the Mn spins in LSMO near the interface will be disturbed by the AFM magnetic coupling to the Cu spins and develop the observed spin glass behavior.

It is noteworthy that no exchange bias was observed in the LCO/LSMO bilayers. In a previous study on YBCO/LSMO superlattices, it was proposed that hole transfer from the YBCO to the LSMO layer transforms a very thin LSMO layer into the overdoped AFM state, which in turn provides the magnetic pinning and exchange bias.¹⁶ The absence of exchange bias in the LCO/LSMO bilayers studied here suggests that charge transfer does not play a dominant role in the interfacial magnetic interaction. Another possible reason for the absence of exchange bias is that the Néel temperature T_N of the LCO film may be much higher than the T_C of the LSMO layer (143 K). The T_N of the bulk LCO can be as high as 320 K.²⁸ In the conventional model of exchange bias,³⁰ the T_N of the AFM layer should be lower than the T_C of the FM layer in order to allow the formation of interface coupling between the layers when cooling under an external magnetic field.

Figure 3(a) shows the M - H curves measured on the LCO/LSMO bilayer at different temperatures after the FC process. It can be seen that H_C monotonously decreases on increasing measurement temperature. As shown in Fig. 3(b), the temperature dependence of H_C follows the exponential law. A similar behavior was previously observed in manganite superlattices, and it was attributed to the magnetic frustration resulting from competing FM and AFM interactions.³¹ We also measured the temperature-dependent H_C of the reference 3 nm LSMO single layer. Within the whole temperature range, the H_C of the bilayer is much larger than that of the LSMO single layer.

Although our experiments cannot provide direct evidence on the possible chemical and charge diffusion at the LCO/LSMO interface, these effects may play important roles in the magnetic properties of bilayers. At the previously studied YBCO/LSMO interface, Ba and La interdiffusion was observed, which was proposed to enhance Mn⁴⁺ in the LSMO layer.³² In our bilayers, the cuprate LCO does not contain highly mobile Ba ions, but Sr may diffuse from the LSMO layer and be incorporated into the LCO layer. This is consistent with the slight decrease of T_C observed in the bilayer compared to the reference LSMO single layer (Fig. 2(a)). On the electronic aspect, massive hole transfer was observed in YBCO/LCMO superlattices.³³ In another study, it was proposed that the charge transfer at the YBCO/LSMO interface can induce unidirectional anisotropy in the LSMO layer.¹⁶ A recent theoretical work also suggested that the

FIG. 3. (a) M - H data measured on LCO/LSMO bilayer at different temperatures. (b) Temperature dependence of the coercivity. The line is fitting to the exponential law. The arrow marks the spin-glass freezing temperature T_f .

Fermi level of manganites, within a wide range of doping, lies above the chemical potential of cuprates, leading to electron transfer from the manganite to the cuprate.³⁴ The authors also speculated the possibility that the addition of electrons to AFM cuprates may lead to a superconducting state at the interface without introducing quenched disorder. But in our case, if the H_C enhancement is due to the charge diffusion, it should be independent of the AFM layer thickness. To test this scenario, we made LCO/LSMO bilayers with different LCO thicknesses while keeping the thickness of the LSMO layer fixed. In Fig. 4, it can be seen that when the AFM layer thickness decreases from 40 nm to 5 nm with the same thickness of the FM layer, the H_C decreases from

FIG. 4. M - H curves measured at 5 K on the LCO/LSMO bilayer samples with the LCO layer having different thicknesses.

705 to 358 Oe at 5 K. Since the charge transfer often happens on a length scale of nm,³³ the observed dependence of H_C enhancement on the AFM layer thickness suggests that the H_C enhancement is not originated from charge diffusion. Instead, the thickness dependence in Fig. 4 may be due to the fact that on increasing thickness the magnetocrystalline anisotropy of the AFM layer becomes larger, resulting in a stronger pinning force onto the FM spins in the LSMO layer.^{35,36}

In summary, we observed the definite evidence of the exchange coupling in the LCO/LSMO bilayers. At the interface, the magnetic coupling between Mn and Cu ions induces the frustration of FM ordering in LSMO layer, and as a result a spin-glass state and significant H_C enhancement were observed in the cuprate/manganite bilayer. Our data show that the H_C can be systematically tuned by varying the thickness of the AFM LCO layer, which may bear importance to the designs of oxide spintronic devices. Our data disfavor the scenario that charge transfer at the manganite/cuprate interface plays an important role in magnetic interactions although it likely exists. Furthermore, the observed magnetic characteristics of cuprate/manganite bilayers invite future theoretical studies to fully elucidate the profile of spin configurations at the interface.

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