Exchange bias in $La_{0.7}Sr_{0.3}CrO_3/La_{0.7}Sr_{0.3}MnO_3/La_{0.7}Sr_{0.3}CrO_3$ heterostructures

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In the recent past, heterostructures of magnetic oxide thin films have attracted a great deal of research excitement due to very interesting physical properties such as antiferromagnetic interlayer coupling, tunable exchange-bias, interfacial driven magnetic properties and high mobility electron gas across the interfaces. In this work, we report on the comprehensive magnetic properties observed from the heterostructures of (2 unit cells) $La_{0.7}Sr_{0.3}CrO_3$/(8 unit cells) $La_{0.7}Sr_{0.3}MnO_3$/(2 unit cells) $La_{0.7}Sr_{0.3}CrO_3$, which are epitaxially deposited on $SrTiO_3$ substrate by plasma-assisted oxide molecular beam epitaxy. Using SQUID magnetometer, the magnetic properties are studied when the magnetic field was applied both in plane and out of plane. The Curie temperature of this structure is found to be at 290 K. Most significantly, at 2 K, we observed a complete up/down shift (along magnetization axis) of hysteresis loop when the sample was cooled under a magnetic field of ± 5000 Oe in the in plane configuration. We believe that the strong antiferromagnetic (super) exchange coupling of Mn-Cr across the two interfaces is responsible for the observed exchange bias. We will present and discuss our in-detailed experimental findings collected on this heterostructure as a function of temperature and magnetic field.

I. INTRODUCTION

Transition metal oxides (TMOs) are a group compounds that have been of interest due to their interesting properties that make them prospective candidates for engineering applications and technologies. TMO heterostructures portray interesting physical phenomena often not seen in the bulk form such as interactions of atoms at the interface and across the boundary, reduced site coordination of lattice atoms at interface, and a break in translational symmetry of crystal potential.[1] TMO thin films, heterostructures with thicknesses ranging from micro- to nano-meters, display interfacial interactions that can lead to effects such as antiferromagnetic interlayer coupling, tunable exchange bias, thickness dependent phase transitions, and high

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mobility of two-dimensional electron gas across interfaces.[2-4] These interesting phenomenon come from the interactions occurring with lattice, charge , orbitals and spins that give a sensitivity to strain, electric and magnetic fields, as well as chemical doping.[5] Moreover, with these interesting properties, including magnetoresistance and half-metallic properties, TMO thin films can be used in applications such as spintronics, magnetic sensors, and magnetic random-access memory [1,6]. However, with the reconstruction of the structure occurring at the interface, a strong thickness dependent property arises thus leading to a surge in researchers trying to find ways to control these properties.

Particularly, $LaSrMnO_3$, or LSMO films are of interest as they exhibit these interesting properties mentioned before such as colossal magnetoresistance and half-metallicity. LSMO can be coupled with other TMOs such as $LaSrCrO_3$. The tri-layer thin film $La_{0.7}Sr_{0.3}CrO_3/La_{0.7}Sr_{0.3}MnO_3/La_{0.7}Sr_{0.3}CrO_3$ (LSCO/LSMO/LSCO) consists of an anti-ferromagnetic (AF) surface layer—where Cr and Mn ions exhibit an AF exchange interaction—situated between two robust ferromagnetic (FM) layers of LSCO that allow the LSMO layer to retain its ferromagnetic properties.[3] Along with the FM and AF exchange coupling are the effects of enhanced coercivity and asymmetry in magnetization reversal processes.[7-9] Many of these exchange bias effects have occurred in other thin film systems.[10-13] In this work, we will be presenting on the magnetic properties and the exchange bias as a function of temperature and magnetic field on the thin film $La_{0.7}Sr_{0.3}CrO_3/La_{0.7}Sr_{0.3}MnO_3/La_{0.7}Sr_{0.3}CrO_3$ on a $SrTiO_3$ (STO) substrate.

II. EXPERIMENTAL DETAILS

A. Heterostructure synthesis

The thin film made up of two layers of $La_{0.7}Sr_{0.3}CrO_3$ (LSCO) and a layer of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) were synthesized by plasma-assisted oxide molecular beam epitaxy. The thickness of the LSCO layers were 2 unit cells thick and with LSMO 8 unit cells in thickness. The layering of the thin film consisted of the LSMO layer sandwiched between LSCO layers epitaxially deposited in a (001) orientation of a $SrTiO_3$ substrate at a growth temperature of 800 °C. The LSCO and LSMO layers were grown in $3x10^{-6}$ Torr atomic oxygen from the plasma source. The sample was cooled down at a rate of 5 °C/min in $5x10^{-6}$ Torr oxygen to ensure that full oxidation occurred. The dimensions of the heterostructure are approximately 3.928 mm by 3.868 mm.

B. Magnetic measurements

Magnetic properties were measured with Quantum Design's MPMS 3 Superconducting Quantum Interference Device (SQUID) and Vibrating Sample Magnetometer (VSM). Temperature dependent measurements were done with a temperature range of 2-400 K with a measuring field of 500 Oe for zero-field cool (ZFC) and field cool (FC) and an applied field of 200

Oe for FC. Isothermal magnetization was measured between 2-350 K and at ± 5000 Oe in different configurations where the magnetic field was applied in plane, 90° rotated in plane, and out of plane. For in plane measurements, the thin film was mounted on a flat quartz rod; for out of plane sample was inserted and held in place in a plastic straw provided by Quantum Design. The resulting magnetic measurements have a small diamagnetic signal arising from the STO substrate. In separate measurements the diamagnetic signal has been subtracted for data seen in Fig. 1(a) and 3(a) confirming that the diamagnetic contribution does not significantly affect the overall magnetic behavior we observe for LSCO/LSMO/LSCO. Therefore, figures containing magnetic data are not refined with diamagnetic subtraction, however, an accurate representation of the magnetic behavior of LSCO/LSMO/LSCO is observed nonetheless.

III. RESULTS AND DISCUSSION

To begin magnetic measurements, the sample is heated to 400 K to the paramagnetic phase of the thin film as the expected Curie temperature (T_C) is 290 K. Temperature dependent (M-T) measurements are seen in Figure 1(a), where T_C can be estimated from the derivative of magnetic moment with respect to temperature. In both ZFC and FC curves, we see an unexpected spike in the curves at approximately 54 K. The significance of this spike seemingly stems from adsorbed oxygen on the surface sample while being cooled as explained in T. Dubroca *et al.* This is represented as a paramagnetic to antiferromagnetic transition occurring at approximately between 52-55 K.[14] In the M-T plot (Fig. 1(a)), it is confirmed that the sample magnetization is field dependent at a field cooling of 200 Oe. Additionally, the magnetic moment increases upon cooling with the application of an external field for FC compared to no field.

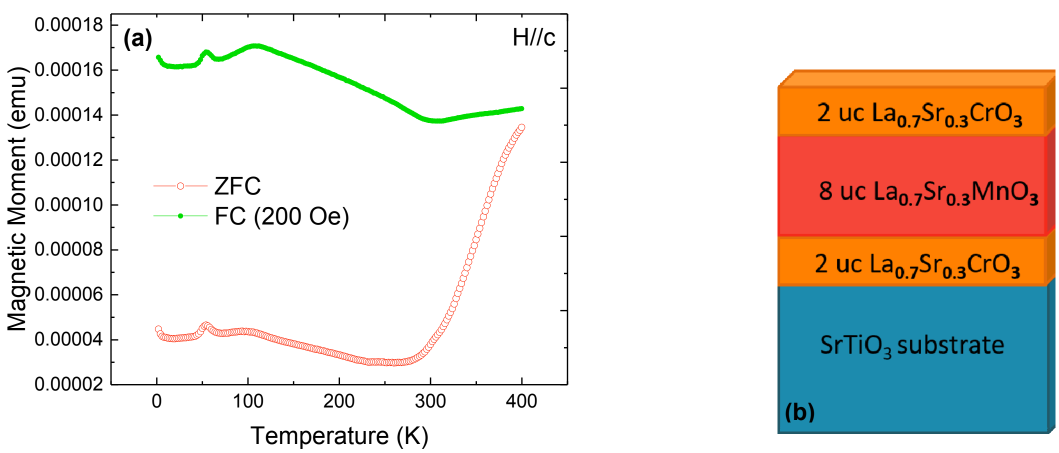

FIG. 1. (a) ZFC and FC (H = 200 Oe) curves measured at a field of 500 Oe for in plane (H//c) measurements between 2-400 K. (b) A schematic of the LSCO/LSMO/LSCO heterostructure on $SrTiO_3$ substrate is shown.

The next set of measurements are magnetic moment versus magnetic field (M-H), are between 2 to 350 K with no applied external field. An interesting phenomena occurs with in plane data particularly for hysteresis loops above the T_C measured at

300 and 350 K. Asymmetry and a broad hysteresis loop is observed for these temperatures and can be seen in Figure 2(a) and (b), suggesting an exchange bias effect possibly arising from the different magnetic phases of LSCO/LSMO/LSCO. Furthermore, for these M-H curves, the hysteresis loops do not completely saturate. In order to see if full saturation can be achieved and to rule out any artifact(s), test measurements are completed at 300 K at 10,000 Oe. It is observed that the loop still does not fully saturate with the application of set field and even more a possible training effect is noticed on the exchange bias additionally ruling out any artifact occurring with the measurement. The training effect is the gradual and monotonous degradation of the exchange bias shift along the field axis upon cycles that the system is going through with consecutive hysteresis loops at a fixed temperature, such as seen here at 300 K.[15] In Figure 2(c), an overall decrease in hysteresis loop is visible when the second run of the loop is measured. When comparing the coercive field (H_c) of each sweep to determine the exact units the loops have shifted the first sweep's left-most H_c to be -4267.85 Oe and the second sweep's H_c = -2748.93. The right-most H_c for both sweeps are relatively close at 1201.13 Oe and 1318.42 Oe, for the first and second sweeps, respectively. Figure 2(d) displays the configuration of 90° in plane for temperatures at 2 and 300 K. A diamagnetic signal is seen in the hysteresis loop at 300 K, however, a magnetic phase transition is still evident.

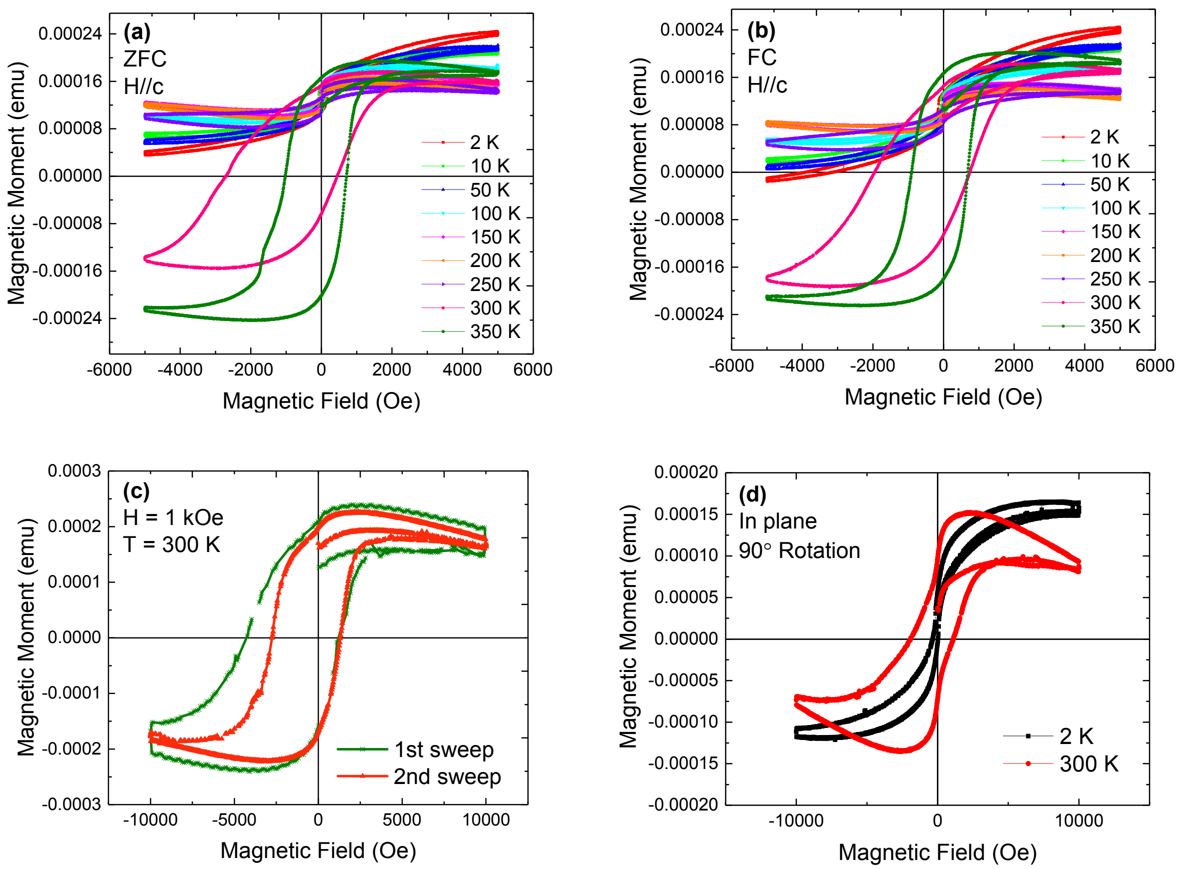

FIG. 2. In plane magnetic moment versus magnetic field for temperatures at 2-350 K for (a) ZFC and (b) FC. (c) M-H testing at 300 K with an applied magnetic field of 1 T. (d) M-H hysteresis loops at 2 and 300 K for in plane 90° rotation configuration.

In Figure 3(a), the 90° in plane rotation FC data is shown at 2 K with ± 0.5 T field. The observation is that there is a clear shift in the hysteresis loops for (+) 5,000 Oe and (-) 5,000 Oe. Moreover, it can be inferred that Figure 3(a)(b) shows the field dependence of LSCO/LSMO/LSCO for out of plane and in plane configurations, with a predominant upward shift on the magnetic moment axis for the in plane configuration at (+) 5000 Oe (Fig 3(a)). Furthermore, in magnetic field dependent measurements, saturation of the moment does not occur even at the maximum applied magnetic field 10,000 Oe. Magnetic anisotropy is also observed from comparing the hysteresis loop shapes of the three configurations at which the measurements were done. Additionally, the magnetic easy axis is found to be along the in plane configuration.

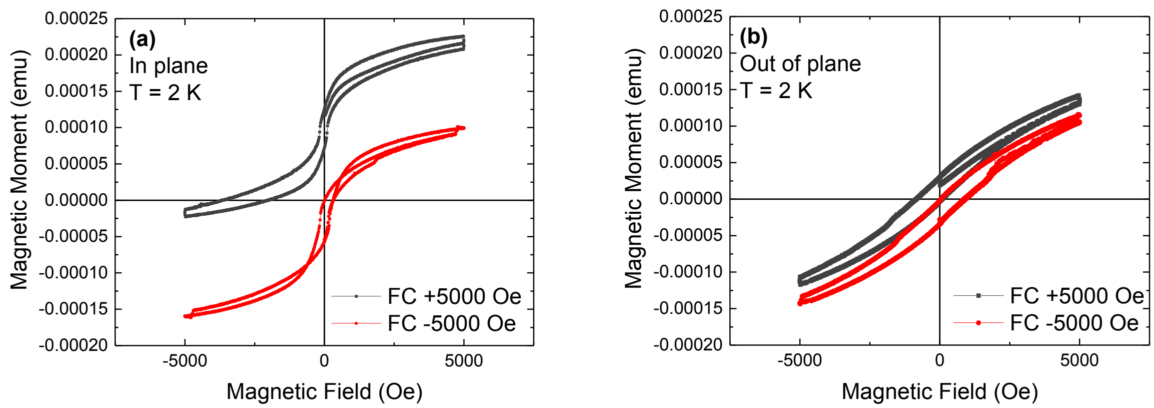

FIG. 3. M-H FC loops at ± 5,000 Oe for (a) in plane and (b) out of plane configurations at T = 2 K showing vertical (a) and horizontal (b) shifts of the magnetic hysteresis loops.

IV. CONCLUSION

The magnetic properties of LSCO/LSMO/LSCO deposited on STO have shown effects such as anti-ferromagnetic exchange coupling, exchange bias observed through asymmetry and a (+) shift on the magnetic moment axis, as well as an indication of magnetic anisotropy. A possible training effect has also been observed at 10,000 Oe and will need more investigation to completely comprehend the cyclability of the thin film. Additionally, it is believed that the strong anti-ferromagnetic (super) exchange coupling of Mn-Cr across the interfaces is responsible for the exchange bias observation.

The training effect phenomena observed at 300 K may be worth investigating in order to determine the number of cycles this thin film will go through until returning back to the original cycle. Future work may include taking steps toward completing structural characterization methods. Several prospective measurements will take place with aberration corrected high-angle annular dark-field scanning, transmission electron microscopy (TEM), electron energy loss spectroscopy (EELS), and energy dispersive x-ray spectroscopy chemical mapping in order to confirm unit cell layer thicknesses of the thin film and the valence states of elements in the layers.

V. ACKNOWLEDGMENTS

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