

Tailoring exchange bias in half-metallic $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ thin films for spin-valve applications.

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Abstract

We have utilized the antiferromagnetic nature and structural/chemical compatibility of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ with highly spin polarized $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ to prepare epitaxial exchange bias couples. A robust exchange bias (EB) shift of magnetization hysteresis with associated interfacial exchange energy $J \approx 0.13 \text{ erg/cm}^2$ at 10 K along with enhanced coercivity are reported. The EB effect was engineered to bring coercivity contrast between $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ and cobalt films in $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$ magnetic tunnel junctions.

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Spin-polarized transport in oxide based half-metals is a topic of intensive research these days because of its potential for application in spintronics devices. Some of the most investigated half-metallic oxides include CrO_2 , Fe_3O_4 , double perovskites like $\text{Sr}_2\text{FeMoO}_6$ and manganites $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ (LSMO) for certain doping. In particular, the manganite with stoichiometry $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ has been investigated widely for magnetic tunnel junctions (MTJs)[1], for which it is necessary to have a significant difference in the coercive fields of the two ferromagnetic electrode in order to have a step-like switching of magnetization and tunneling conductance. Typically, exchange bias (EB) phenomenon between a ferromagnet (FM)/antiferromagnet (AF) bilayer is used to engineer the coercivity in MTJs. The EB manifests itself as a shift of the magnetization loop along the field axis by a field H_{ex} and the coercive field H_C is enhanced. While mechanism of exchange bias since its discovery 40 years ago by Meiklejohn and Bean[2] is still not fully understood, it is widely believed to arise from the exchange coupling between spins at the interface between FM and AF layers.

It has been a long standing problem to find a suitable exchange bias antiferromagnet for $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO(FM)), while a positive exchange bias is seen in $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrRuO}_3$ bilayer where SrRuO_3 is a ferromagnet with $T_C \approx 150$ K[3].

Interestingly, $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ for $x = 0.55$ is an A-type antiferromagnet with pseudo-two-dimensional structure consisting of double-exchange controlled metallic FM planes aligned antiferromagnetically along the c-axis. Clearly, $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ (LSMO(AF)) is a potential candidate for exchange biasing $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ due to its good lattice and resistivity matching with the latter. In our previous studies of $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ thin films we have shown that it can be grown on SrTiO_3 (STO) substrate epitaxially. Such films undergo a Néel transition at $T_N \approx 220$ K[4, 5].

In this letter we show clear signatures of EB in LSMO(AF)/LSMO(FM) bilayers. The exchange bias effect has been used to control coercivity mismatch between LSMO(FM) and cobalt films and in LSMO(AF)/LSMO(FM)/STO/Co MTJ structures deposited on (100) STO substrates.

LSMO(AF)(80nm)/LSMO(FM)(50 nm)/STO(3nm) structure was prepared by pulsed laser deposition (PLD) method on (001) STO. All the layers were grown at 700°C in oxygen pressure of 0.4 mbar. The details of thin film preparation are reported elsewhere[4, 6]. Later on a 50 nm thick Co thin film was deposited at 150°C on some of the LSMO(AF)/LSMO(FM)/STO structure by e-

beam evaporation. The abbreviation Sample-B and Sample-T has been used in the manuscript to represent heterostructure $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3$ and $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$ respectively. The exchange bias was established through magnetization measurements carried out in a superconducting quantum interference device (SQUID) based magnetometer (Quantum Design MPMS-XL5).

Figure 1(a, b) shows the temperature dependence of field-cooled (FC) and zero-field-cooled (ZFC) magnetization $M(T)$ measured in a in-plane field of 1500 Oe for Sample-B (Fig. 1(a)) and Sample-T (Fig. 1(b)). The magnetic moment of the antiferromagnet LSMO(AF) ($M(5\text{ K}) \approx 32\text{ emu/cc}$) is usually an order of magnitude smaller than the moment of LSMO(FM) ($M(5\text{ K}) \approx 462\text{ emu/cc}$)[4, 6]. Therefore, the signature of AF transition at $T_N \approx 220\text{ K}$ of the 45/55 composition is buried in the strong $M(T)$ response of the ferromagnetic LSMO(FM) layer. The magnetic moment for the Sample-T is double compared to the moment of the cobalt-free sample as seen in Fig. 1(b). This difference can be attributed to the higher magnetic moment of the Co, which is extracted by subtracting the moments of Sample-T and Sample-B. The result of this subtraction is shown in the inset of Fig. 1(b). The moment of the Co film is relatively temperature independent in the range of 5 to 350

K as seen in the inset. A splitting of the FC and ZFC branches of $M(T)$ curves can also be seen below 50 K in Fig. 1(a,b) as indicated by arrows. Such splitting appears to be due to the onset of exchange interaction between uncompensated spins of LSMO(FM) and LSMO(AF) at the interface. This can be viewed as a blocking temperature T_B (≈ 50 K) of the bilayer system.

Magnetization loops at 10 K for Sample-B and Sample-T measured after cooling in zero field are shown in Fig. 2(a) and 2(b) respectively. The measurement was done after saturating the sample at 2000 Oe in-plane field. A shift in the hysteresis loop by ≈ 83 Oe towards the positive field direction is clearly seen in Fig. 2(a). The coercive (H_C) and exchange (H_{ex}) fields determined from switching fields H_C^+ and H_C^- according to $H_C = (H_C^+ - H_C^-)/2$ and $H_{ex} = (H_C^+ + H_C^-)/2$ are 60 and 83 Oe respectively. The shift of the loop seen even on zero-field cooling (ZFC) indicates an antiferromagnetic coupling between uncompensated spins at the LSMO(AF)/LSMO(FM) interface[3, 7].

In order to establish exchange bias effect, Sample B and T are cooled from 350 to 10 K with an in-plane field of 2000 Oe followed by measurement of hysteresis loops by sweeping the field over the range ± 2000 Oe. A shift in the hysteresis loop for the bilayer was

observed depending on the direction of reference field used for cooling as shown in Fig. 3 (Panels a & b). For ± 2000 Oe an exchange bias shift $H_{ex} \approx \pm 68$ Oe is calculated from the M-H loops. Also the coercive field H_C is enhanced by ≈ 26 Oe as compared to the H_C of the ZFC case. The exchange field H_{ex} was also found to decrease rapidly to zero for temperature above ≈ 50 K. The EB effect seen here is strikingly different from what is reported in the case of $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrRuO}_3$ bilayers[3] where the M(H) loop shifts in the same direction as that of the biasing field with an $H_{ex} \approx 98$ Oe. Although the EB in our case is slightly smaller (≈ 68 Oe), we believe the LSMO(AF) is better suited than SRO for exchange bias purposes because of the later's ferromagnetic character and a large uniaxial magnetocrystalline anisotropy. For the trilayer sample-T (Panels c & d of Fig. 3) the coercive field H_C of cobalt remains fixed at ≈ 42 Oe where as the H_C of LSMO(FM) can be shifted back and forth by exchange bias-controlled pinning field. This difference in the coercivity results in a distinct step in the M-H loop, thus making it a very interesting system from application point of view. Moreover the coercivity of LSMO(FM) can be further engineered by tuning the field used for cooling and the relative thickness of the FM and AF layers. Here it is important to point out that the coercive field of thin epitaxial LSMO(FM)

films deposited on (100)STO is comparable to the H_C of Cobalt[6]. Hence exchange bias of LSMO(FM)/Co bilayer with LSMO(AF) is necessary to realize coercivity contrast. Also the biasing direction can be reset by warming the sample back up to room temperature and cooling again with the field reoriented.

The negative H_{ex} with respect to reference cooling direction can be understood by ferromagnetic coupling between uncompensated moments in the (001) plane of LSMO(AF) and the moments of LSMO(FM) so that the latter are frozen in the direction of the applied field and therefore a bigger force or stronger external field in opposite direction is required to overcome this coupling. It is well known that for FM/AF bilayer the magnitude of H_{ex} is given by[7]; $|H_{ex}| = J/M_F t_F$, where J is the FM-AF interfacial exchange coupling energy, M_F and t_F are the magnetization per unit volume and thickness of the FM layer respectively. Substituting $M_F \approx 400$ emu/cc extracted from the data of Fig. 3(a), $|H_{ex}| \approx 68$ Oe , and $t_F \approx 50$ nm, the exchange coupling energy J comes out to be ≈ 0.13 erg/cm² at 10 K. The value of J for this bilayer system is comparable to J of conventional metallic bilayers like NiMn/FeNi ($J \approx 0.27$ erg/cm²)[7, 8], Co/IrMn ($J \approx 0.12$ erg/cm²)[9] and MnPt/FeNi ($J \approx 0.03$ erg/cm²) [10] etc.

The EB effect also results in enhancement of coercivity of LSMO(FM) layer which can be used for bringing coercivity contrast between LSMO(FM) and cobalt in LSMO/STO/Co tunnel junctions. A typical field dependence of magnetization and resistance for a LSMO(AF)/LSMO(FM)/STO/Co tunnel junction measured at 30 K is shown in the Fig. 4(a) and (b) respectively[11]. A step-like inverse TMR results because of the antiparallel configuration between LSMO(FM) and Co layers in the field range of 50-150 Oe. The resistance step also found to match well with the magnetization step as shown in Fig. 4(a). The TMR of these junctions is $\approx 6\%$ at 10 K. De Teresa *et al.*[12] have reported a strong bias dependent TMR in LSMO/STO/Co with a peak value of $\approx 30\%$ at 10 K in junctions of much smaller area. We are currently investigating the bias and area dependence of magnetoresistance in our junctions.

In summary we have demonstrated that antiferromagnetic $\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3$ can provide a robust exchange bias to FM $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$. The advantage of this material are its identical chemistry and close lattice match with the ferromagnetic LSMO(FM). The direction of exchange bias is shown to be controlled by the field used for cooling. The shift in the hysteresis loop was also used to bring coercivity contrast in

$\text{La}_{0.45}\text{Sr}_{0.55}\text{MnO}_3/\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$ heterostructures. Such shift has been used to realize sharp magnetic field controlled switching of conductance in tunnel junctions made of $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3/\text{SrTiO}_3/\text{Co}$.

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- [11] 200 nm Y_2O_3 was deposited through a shadow mask by e-beam evaporation on Sample-T leaving a $25 \times 4000 \mu\text{m}^2$ channel uncovered. Cross stripe of 50 nm thick and 25 μm wide cobalt film were deposited by e-beam evaporation to create junctions of area $25 \times 25 \mu\text{m}^2$. Base contact was made by Ar^+ ion milling of SrTiO_3 layer at the end of sample. These stripes were finally covered with 150 nm thick Ag-capping layer.
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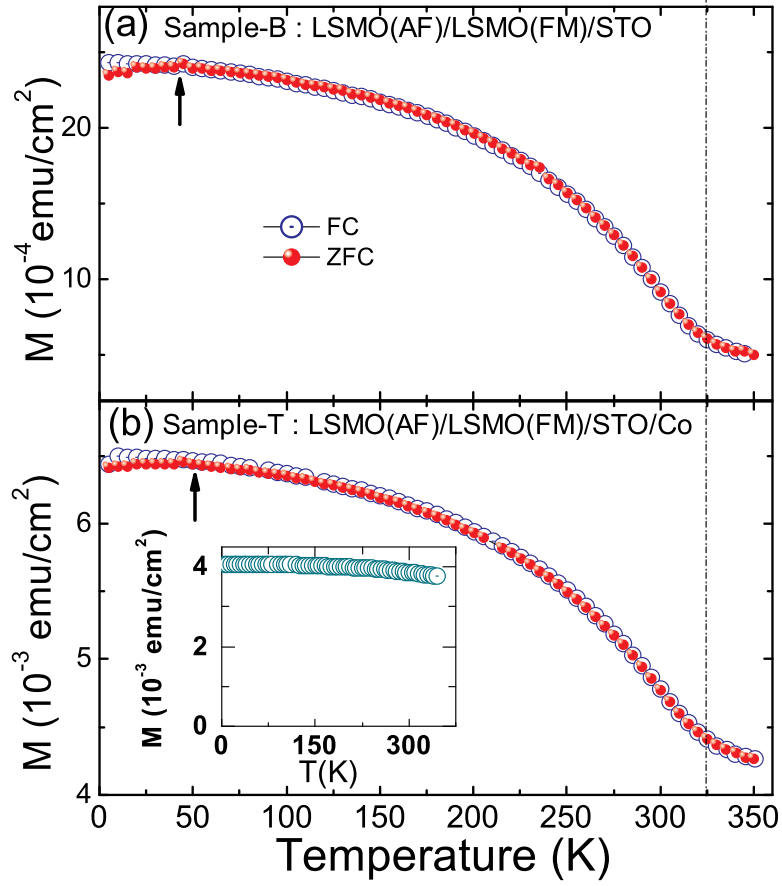


FIG. 1: Temperature dependence of field-cooled (FC) (open circle) and zero-field-cooled (ZFC)(closed circle) magnetization of Sample-B (a) and Sample-T (b) measured in 1500 Oe applied in the plane of the film. The inset in (b) shows the temperature dependence of FC magnetization of the cobalt layer determined by subtracting the FC magnetization values of sample-B and Sample-T.

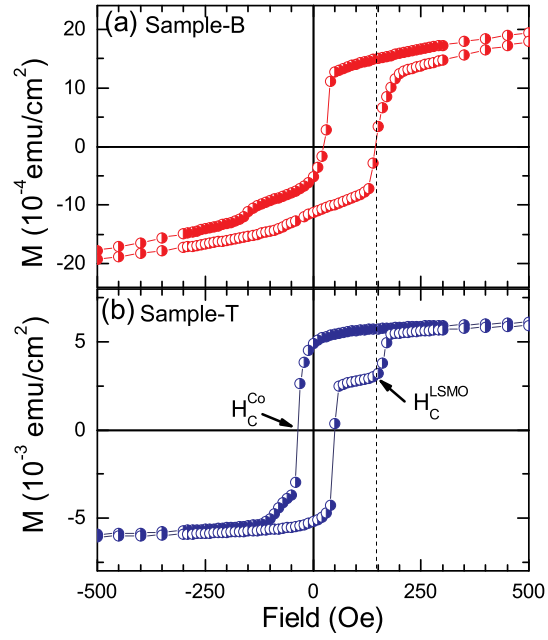


FIG. 2: Field dependence of in-plane magnetization of Sample-B (a) and Sample-T (b) measured at 10 K after cooling the sample in zero-field.

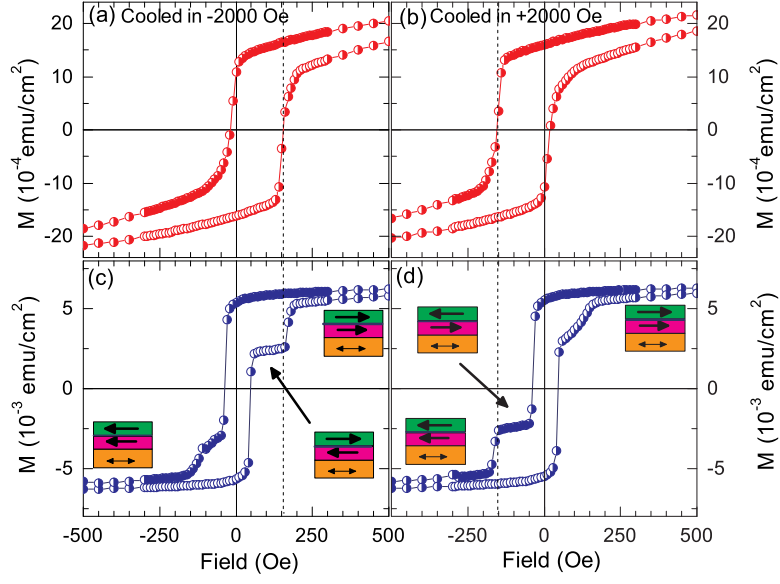


FIG. 3: Field dependence of magnetization of Sample-B (a & b) and Sample-T (c & d) measured at 10 K after cooling the sample in ± 2000 Oe field. The field axis in case of Sample-T can be divided into three zones according to relative orientation of magnetization of each layers as shown in boxes.

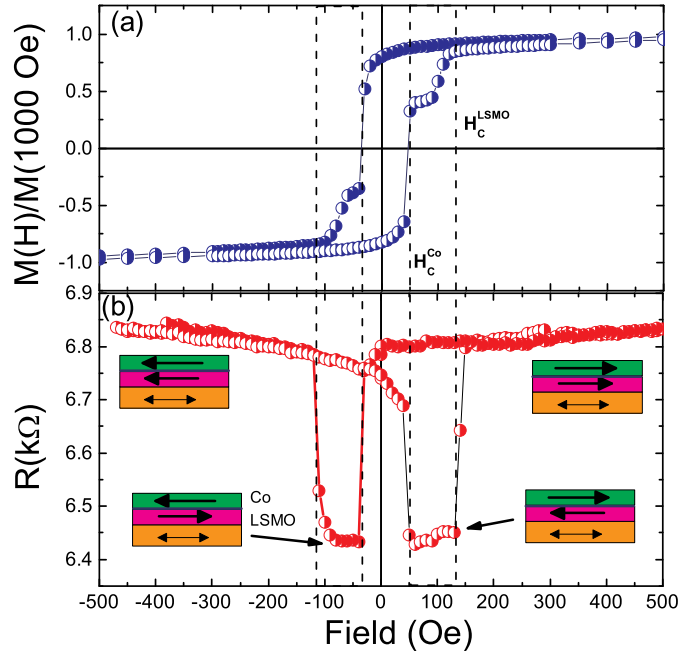


FIG. 4: Field dependence of (a) magnetization and (b) resistance of a LSMO(AF)(80nm)/LSMO(FM)(50nm)/STO(3nm)/Co(50nm) tunnel junction measured at 30 K. Low resistance state between H_c^{Co} and H_c^{LSMO} is due to inverse TMR observed in LSMO/STO/Co tunnel junctions.