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High sensitivity low field magnetically gated resistive switching in $\text{CoFe}_2\text{O}_4/\text{La}_{0.66}\text{Sr}_{0.34}\text{MnO}_3$ heterostructure

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The phenomenon of resistive switching (RS) has been demonstrated in several non-magnetic and some magnetic oxide systems, however the “magnetic” aspect of magnetic oxides has not been emphasized especially in terms of low field tunability. In our work, we examined the $\text{CoFe}_2\text{O}_4/\text{La}_{0.66}\text{Sr}_{0.34}\text{MnO}_3$ all-magnetic oxide interface system for RS and discovered a very sharp (bipolar) transition at room temperature that can be gated with high sensitivity by low magnetic fields (~ 0 – 100 mT). By using a number of characterizations, we show that this is an interface effect, which may open up interesting directions for manipulation of the RS phenomenon. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4707373>]

Resistive switching (RS) is an interesting and intriguing phenomenon which has been intensely researched during the past decade in the context of non-volatile memory applications.^{1–3} This phenomenon has been demonstrated and analyzed in several non-magnetic and magnetic oxide systems; however, hardly any work is reported thus far on low magnetic field tuning of RS at room temperature. There is one previous report by Das *et al.*⁴ on low field magnetic tuning of RS in very thick (83 nm) sol-gel deposited NiO films but only at low temperature [Supplementary text #1 (Ref. 20)]. Our device configuration is distinctly different and involves pulsed laser deposition (PLD) grown ultrathin (few nm) CoFe_2O_4 (CFO) film on $\text{La}_{0.66}\text{Sr}_{0.34}\text{MnO}_3$ (LSMO), emphasizing interesting interface physics. CFO is a ferrimagnetic insulator which is known for its very high magnetostriction coefficient⁵ ($\lambda \sim 800 \times 10^{-6}$) and a very efficient spin filtering effect⁶ at room temperature. $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x = 0.34$, as determined by Rutherford backscattering) is a ferromagnetic conductor at room temperature due to large bandwidth and nearly linear Mn^{3+} -O- Mn^{4+} bond. Small changes in the Mn-O-Mn bond properties at an interface can, however, change the transport and magnetic properties significantly.^{7,8}

The heterostructures [Fig. 1(a)] were grown by PLD [KrF excimer laser, $\lambda = 248$ nm, 20 ns pulses] at repetition rate 10 Hz, energy density ~ 2 J/cm² which consisted of 100 nm LSMO as bottom contact on LaAlO_3 [LAO (100)],

followed by patterned 20 nm LSMO, and a few nm CFO (2, 4, and 6 nm) film for achieving a fresh high quality interface. The deposition conditions for both LSMO and CFO layers were (a) substrate temperature = 700 °C, (b) oxygen pressure during deposition = 100 mTorr, and (c) oxygen pressure during cooling = 400 Torr. The current perpendicular to plane (CPP) transport was studied across these interfaces by sweeping voltage in cycle (sweep sequence -5 V \rightarrow 0 V \rightarrow 5 V \rightarrow 0 V \rightarrow -5 V) with a sweep rate of 1.0 V/s and a step of 0.05 V. In the CPP transport, the top contact for the I-V measurements is indium. Measurements were also performed in magnetic field, with magnetic field applied parallel to film plane.

The transmission electron microscopy (TEM) cross section image [Fig. 1(b)] of a typical CFO/LSMO heterostructure shows parallel lattice planes, thickness uniformity, and high degree of surface smoothness [Fig. S1(a)]. The Rutherford backscattering spectrometry (RBS) [Supplementary Text #2, Fig. S1(b)²⁰] brings out the composition of the phases in the films and the sharp nature of interfaces (used for fitting) consistent with the TEM images. Fig. S1(c) shows atomic force microscopy (AFM) topographic image [root mean square (RMS) roughness of ~ 0.4 nm] and concurrently acquired conducting-AFM (CAFM) image [Fig. 1(c)] at negative sample bias of -3.5 V. The spatial distribution of current, measured by CAFM in CPP configuration, is uniform throughout the interface with a nanoscale modulation of current near grain boundaries.^{9,10} Given the $\sim 9.6\%$ lattice mismatch between CFO and LSMO, highly oriented grain boundary configurations are indeed expected in the CFO film.

Fig. 1(d) shows typical I-V curves obtained for different CFO thickness values. A large RS is seen towards positive polarity (LSMO +ve terminal) at a threshold voltage

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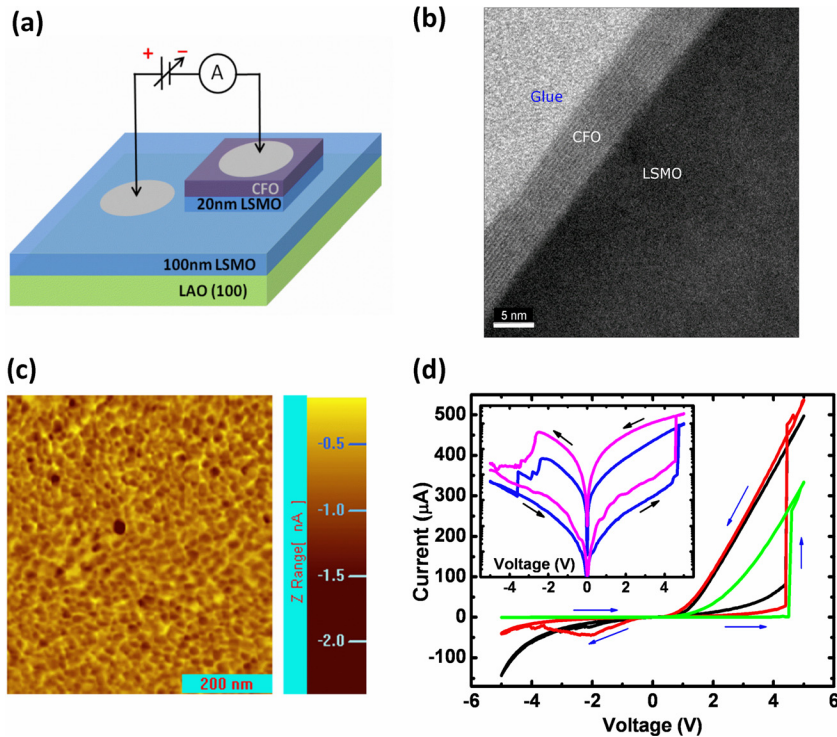


FIG. 1. (a) Device structure: comprised of a single crystal (001) LAO substrate, a 100 nm LSMO film, and freshly grown heterostructure of LSMO (20 nm, as bottom layer) and CFO of different thicknesses (2 nm, 4 nm, and 6 nm, as top layers). (b) Cross sectional TEM image of CFO (4 nm)/LSMO at higher resolution. (c) CAFM image (500 nm \times 500 nm) of CFO (4 nm)/LSMO case. (d) Typical I-V curves noted across LSMO-CFO interface in CPP mode for different CFO thicknesses (Black 2 nm, Red 4 nm, and Green 6 nm) in the absence of magnetic field. Inset shows lateral area dependence of resistive switching (Magenta curve 2 mm \times 3 mm and Blue 500 μ m \times 500 μ m) (current on log scale).

~ 4.45 V (± 0.05) for all the CFO thicknesses (2 nm, 4 nm, and 6 nm). A reverse transition occurs from low resistance state (LRS) to high resistance state (HRS) in the negative bias cycle, though it is not as sharp as the positive cycle. Interestingly, independence of RS switching voltage on CFO layer thickness implies that this is a purely interfacial phenomenon. The inset to Fig. 1(d) shows RS curves for two different device areas, namely 2 mm \times 3 mm and 500 μ m \times 500 μ m. It is clearly observed that the resistance values for the HRS and LRS increase with decreasing lateral area which is completely consistent with the interfacial type resistive switching.^{11,12} The diode-like rectifying characteristics observed here are commonly seen in p-n junction interfaces of doped manganites.^{13–15} The higher current values in LRS state can be attributed to the transport of current through CFO grain boundaries as evident from CAFM image discussed above. The mechanism for such rectification is the formation of Schottky barrier which is seen at the metal-manganite interfaces, particularly in the case of the Pr_{0.7}Ca_{0.3}MnO₃ (PCMO).¹⁶ Sawa *et al.*¹⁷ found similar rectifying character only after depositing a few unit cells of insulating Sm_{0.7}Ca_{0.3}MnO₃ (SCMO) on LSMO.

Fig. 2(a) shows the RS curve for 2 nm CFO as a function of magnetic field applied up to 72 mT in steps. The switching voltage is seen to reduce substantially with increasing magnetic field for all the cases of CFO thickness [Figs. S2(a) and S2(b)] at fairly low fields and that too without losing the sharpness of the transition. Most remarkably, the switching voltage shift depends rather strongly on the CFO thickness in the presence of magnetic field [Fig. 2(b)], whereas RS is independent of CFO thickness in the absence of magnetic field. The observed magnetic field induced switching voltage shift at 72 mT is ~ 2 V, ~ 1.4 V, and ~ 0.7 V for devices with the CFO layer thickness values of 2, 4, and 6 nm, respectively. This represents very high sensitivity at fairly low fields and that too at room temperature. The reversal of magnetic field

direction does not seem to be affecting the trend of RS voltage shift [Fig. S2(c)]. It is useful to point out that the threshold switching voltage in PCMO (itself acting as the RS layer) was found to decrease gradually with magnetic field at lower temperature (20 K) (Ref. 18, Figure 4) but at much higher field values than observed herein, which was attributed to the collapse of the charge ordering state under magnetic field (~ 4 T).¹⁸ Clearly, the mechanism in our case, which involves a large bandwidth manganite interfaced with high-resistivity CFO, is very different as discussed later. It is useful to state here that magnetization measurements [Supplementary Text #3, Figs. S3(a)–S3(d)]²⁰ revealed anti-ferromagnetic coupling between LSMO and CFO.

The RS phenomenon was also studied by changing reverse bias voltage (V_{rb}) from 0 V, -1 V, -2 V... to -7 V before the positive sweep [Fig. 2(c)]. It was observed that the threshold voltage (V_{th}) required to switch the device from HRS to LRS increases with increasing negative bias. This observation implies that the entity governing the RS is influenced not just by the polarity of applied electric field but also by its strength. This suggests the role of oxygen vacancies in the modification of Schottky barrier leading to RS and also rules out possible role of Joule heating effect because in that case the reduction of V_{rb} would have made the device to switch at higher V_{th} .^{11,19}

Another related important observation is the increase of device resistance in HRS with increasing magnetic field [Fig. 2(d)]. Positive magneto-resistance (MR) has been previously reported for LSMO in contact with Nb-doped SrTiO₃ (Nb-STO) where the electronic interactions at the interface of the junction are suggested to be responsible.¹⁵ On the other hand, in LRS, the resistance is seen to decrease with increasing magnetic field (negative MR), a behavior of the bulk LSMO. This observation implies that in HRS, the interface defines and controls the transport, while in LRS, the bulk of LSMO controls the transport.

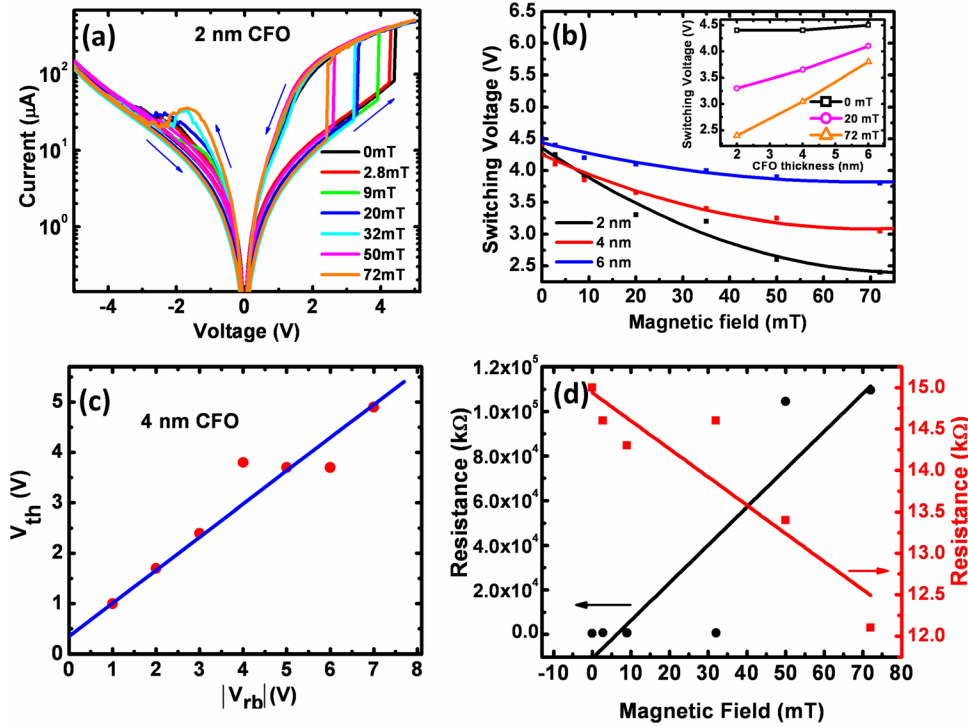


FIG. 2. (a) I-V characteristics of the sample of CFO thicknesses 2 nm under different magnetic fields, showing a systematic change with magnetic field. (b) Effect of magnetic field on switching voltage for different CFO thicknesses plotted together (curve fitted). Inset: the switching voltage as a function of the CFO thickness for different magnetic fields. (c) Resistive switching voltage (V_{th}) as a function of reverse bias voltage (V_{rb}). (d) Resistance plotted as a function of magnetic field in HRS (Black solid circle) and LRS (Red square).

We discuss one possible scenario which explains the above observations reasonably well. In the LSMO/CFO device, bulk LSMO away from interface is metallic at room temperature while the interfacial LSMO and the CFO layer are expected to contribute significantly to the device resistance. As suggested by the weak CFO thickness dependence of the switching voltage, the LSMO layer near the interface dominates the resistance in the HRS [Fig. 3(a)]. Under forward bias, the voltage will be primarily shared by the CFO and the interfacial LSMO layer. As external voltage increases, voltage drop across the interfacial LSMO layer

will increase reaching a threshold value which will push the positively charged oxygen vacancies into the CFO layer [Fig. 3(b)]. Exclusion of oxygen vacancies from the interfacial LSMO will increase its metallicity, thereby quenching the insulating interfacial barrier leading to switching from the HRS to the LRS [Fig. 3(c)]. Similarly, when device is reverse biased, the oxygen vacancies must revert back into the interfacial LSMO layer, eventually making it insulating and switching the device back again from the LRS to HRS.

If V is the external applied voltage at which the device switches from HRS to LRS in the absence of magnetic field,

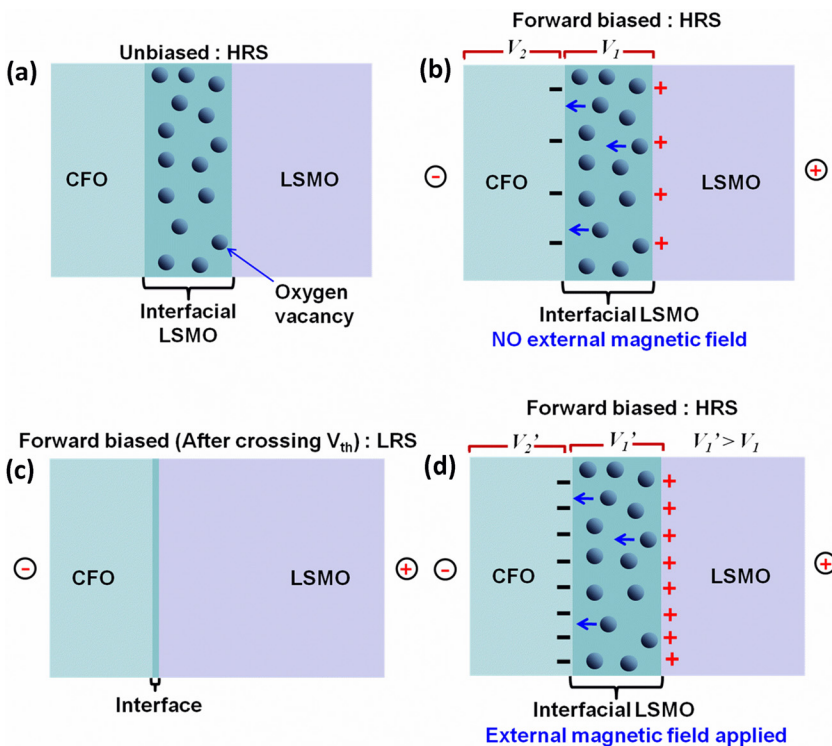


FIG. 3. Schematic showing (a) unbiased interfacial barrier formed at LSMO/CFO interface as a result of oxygen vacancies in HRS, (b) under forward bias before switching, (c) after switching from HRS to LRS, and (d) HRS in the presence of external magnetic field.

the voltage drops across the interfacial LSMO and CFO will be V_1 and V_2 , respectively, in proportion to their resistance values, such that $V = V_1 + V_2$. Thus, V_1 is the actual voltage essential to push oxygen vacancies into CFO and switch the device from HRS to LRS. Remembering that the HRS resistance of the interfacial LSMO layer is increased by the magnetic field (positive MR), the relative voltage drop across it must increase [Fig. 3(d)]. Since the minimum essential voltage for switching of the LSMO interfacial layer is fixed, the device will now switch from HRS to LRS at lesser applied voltage in the presence of magnetic field, as observed here. The key element in this argument is the possible origin of the positive MR of the interfacial LSMO layer which forms a junction with the lower resistivity metallic LSMO. A clue in this respect can be obtained from the work of Jin *et al.*¹⁵ on the Nb:STO/LSMO interface wherein the authors discuss how the relative spin polarization of the electrons around the Fermi level in each region of the junction determines how its MR is affected. In the LSMO system, the t_{2g} spin up band is full and e_g spin up band is partially filled. The t_{2g} spin down band is higher in energy and hence unfilled. Now, in the Schottky junction formation process, electrons spill over into the interfacial LSMO and initially fill the partially filled e_g band followed by partial filling of t_{2g} spin down band leading to band bending until the Fermi levels match. In the CPP transport process, it is the spin scattering of spin up electrons by the t_{2g} spin down electrons in this region that can lead to positive MR.¹⁵

In conclusion, we report an extremely sharp RS at room temperature in CFO/LSMO all-oxide magnetic heterostructure which is magnetically tunable with high sensitivity at fairly low magnetic fields. Remarkably, the switching does not show significant CFO thickness dependence in the absence of magnetic field but a strong CFO thickness dependence under applied magnetic field. We have discussed possible scenarios which can accommodate these observations reasonably well. We believe that the results reported in this paper add a “magnetic” dimension to the research on resistive switching.

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²⁰See supplementary material at <http://dx.doi.org/10.1063/1.4707373> for text #1 to 3 and figures from S1 to S3.