Spacerless metal-manganite pseudo-spin-valve structure

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We fabricated pseudo-spin-valves by using $La_{0.7}Sr_{0.3}MnO_3$ and $Co_{33}Fe_{67}$ as ferromagnetic electrodes. A natural interface layer present between metal and manganite layers eliminated the need of depositing any nonmagnetic spacers. The magnetic layers were decoupled from each other, and the structure exhibited a positive magnetoresistive behavior. Direct comparison between magnetic and transport measurements concluded the occurrence of giant magnetoresistive effect in such a spacerless metal-oxide pseudo-spin-valve structure. The results have implications for a simple route to fabricate oxide-based spintronic devices. © *2008 American Institute of Physics*.
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I. INTRODUCTION

Investigations on magnetic multilayered structures have been fueled by their potentials in spintronics applications.[1] The first spintronic devices include giant magnetoresistive (GMR) multilayers[2] and magnetic tunnel junctions (MTJs).[3] In these structures, ferromagnetic electrodes are separated by nonmagnetic spacers, which are either conductors (in GMR devices) or insulating barriers (in MTJs). Electric current flowing through one magnetic layer is imposed with an asymmetric spin population. High and low resistive states can be achieved, as the spin-polarized charge carriers flow or tunnel through the spacer, and are subsequently sampled by the neighboring ferromagnetic layers; the resistive states depend on the relative magnetization orientation of the magnetic electrodes.

While various approaches have been adopted to boost the performances of such devices,[4–8] much less attention was paid on the physical structure of the magnetoresistive multilayers. Practical structures are dominated by the so-called "spin-valve" geometry, consisting of one nonmagnetic spacer layer sandwiched between two ferromagnetic electrodes. The choice of the barrier thickness is a delicate balance between two competing effects that can influence the functionality of such devices. Magnetically, the spacer should be sufficiently thick to decouple the two ferromagnetic electrodes. For a spacer thickness larger than ~3 nm, interlayer exchange coupling between magnetic layers is minimal,[9] and the main causes of coupling through the spacers in such a situation are pinholes[10] and magnetostatic ("orange-peel") coupling.[11] Magnetic coupling effect could generally be suppressed by using thick spacer layers. On the other hand, thin spacers enable significant portions of the (spin-polarized) charge carriers to commute to the neighboring magnetic layer and then sampled, thus enhancing the observed GMR effect. Typical spacer thicknesses are within a few nanometers for both GMR devices or MTJs for observable behavior. This imposes stringent requirements on the preparation of individual layers, for example, in terms of layer thicknesses and interfacial roughness control.

There are recent reports, which suggested that the spacer layers, under specific circumstances, can be formed naturally at the interface of two ferromagnets. In such reports, either one or both of the magnetic layers are oxides. For example, Abad *et al.*[12] suggested that the surface dead layer of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) can be employed as the barrier layer for magnetotransport devices. Based on this, Ruotolo *et al.*[13] prepared LSMO\$Ni_{80}Fe_{20}$ bilayers by sputtering and demonstrated a magnetoresistance of 12% at 4.2 K. On the other hand, Singh *et al.*[14] demonstrated high magnetoresistance in all-oxide Fe_3O_4\LSMO bilayers. Due to the large lattice mismatch between Fe_3O_4 and LSMO (6.7%), they claimed that the interface was structurally disordered. This resulted in the destruction of double exchange coupling between Mn^{3+} and Mn^{4+} ions at the interfacial LSMO layer, and the (magnetically) dead interface has facilitated the decoupling between the two ferromagnetic layers.

The above reports suggested that magnetoresistive effect can be obtained even without the deposition of artificial spacer layers. Nonmagnetic natural barriers, formed between two ferromagnetic layers under well-controlled deposition conditions, can be employed to decouple the ferromagnetic layers while allowing efficient spin transport of charge carriers. Here, we report our study on the magnetic and magnetotransport properties of LSMO\$Co_{33}Fe_{67}$ (CoFe) spacerless pseudo-spin-valve structure prepared by pulsed laser deposition. We demonstrate that clear magnetoresistive behavior can be observed up to 150 K in such a system. The structure provides a simple alternative for typical GMR or tunneling magnetoresistance (TMR) multilayers, which generally require delicate control of layer growth thicknesses and conditions to achieve desired properties.

II. EXPERIMENT

LSMO\CoFe junctions were deposited on $LaAlO_3$ (LAO) (001) substrates using pulsed laser deposition method. Instead of using LSMO as bottom electrodes, plain

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$LaNiO_3$ (LNO) films of thickness 100 nm were deposited on LAO substrates. Being a nonmagnetic conductor[15] [resistivity ~80 $\mu\Omega$ cm at 10 K, as measured from a single epitaxial film of thickness 100 nm grown on LAO (001) substrate], its resistivity was an order of magnitude lower than that of LSMO at the same temperature. The use of LNO as the bottom electrode allows a more uniform current flow across the pseudo-spin-valve junction, and should help to reduce the contribution of anisotropic magnetoresistance[16] (AMR) from the bottom electrodes (if any).

The LNO layers were grown at a substrate temperature of 650 °C under an oxygen pressure of 150 mTorr, with a laser fluence of 3.6 J cm^{-2}. 200 μm squares of LSMO, 50 nm in thickness, were then deposited on the LNO layer by using a thin stainless steel shadow mask. The deposition conditions were identical to that of the LNO electrode layer. To ensure the oxygen stoichiometry of the layers, the samples were naturally cooled in an ambient oxygen environment of 150 mTorr to room temperature after the deposition of each individual layer. Finally, 50 nm of CoFe was deposited, in the absence of ambient oxygen gas and without heating the substrate, on top of the LSMO squares. The deposition rates for LNO and CoFe were ~5 nm/min, and 20 nm/min for LSMO.

To investigate the magnetotransport properties of the junctions, electrical connections were made on the CoFe electrodes and the bottom LNO films by ultrasound wire bonding. In such a configuration, the measurements were in the so-called "current-perpendicular-to-plane" geometry for GMR studies.[2] Four-point measurement technique was used to probe the resistances of the junctions, thus minimizing the potential resistance contributions from the LNO layer. A constant current of 10 mA was applied across the film during the measurement. The magnetic properties of the structure were characterized by measuring a trilayer plain film sample with a vibrating sample magnetometer. In both the magnetic and transport measurements, magnetic field was applied along the plane of the films.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the x-ray diffraction profile of a LNO\LSMO\CoFe trilayer thin-film sample. Due to the close lattice parameters of the layers, their peaks were not distinguishable from each other in the θ-2θ scan. ϕ-scans of the LSMO and LNO (202) planes (data not shown) indicated cube-on-cube growth of the layers on LAO substrates. The perovskite structures of LNO and LSMO, with similar lattice parameters (a_{LSMO}=3.87 Å and a_{LNO}=3.84 Å for pseudocubic structure), ensured epitaxial growth of LSMO on top of the LNO electrode layer. On the other hand, no diffraction peak corresponding to the CoFe layer was observed, which implies its polycrystalline nature resulting from the growth at room temperature.

The hysteresis loops of the sample, measured at various temperatures, are illustrated in Fig. 2. Double-coercivity behavior was observed in all of the measurements up to 200 K. As the temperature increased, coercivities of the layers gradually decreased. At 100 K, the coercivity values matched

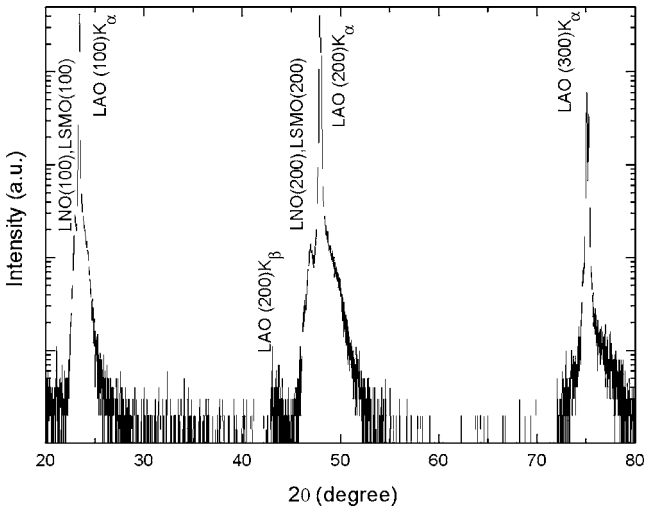

FIG. 1. X-ray diffraction θ-2θ scan pattern of a LSMO\LNO\Co\Fe trilayer film deposited on a LAO (001) substrate.

well with those of the single LSMO (110 Oe) and CoFe (19 Oe) films (inset of Fig. 2), which were deposited under identical conditions as the corresponding layers in trilayer structure. We therefore suggest that the LSMO and CoFe were the hard and soft layers of the pseudo-spin-valve, respectively. At all temperatures up to 100 K, the LSMO layer clearly showed a higher coercivity compared to the CoFe in our samples. The occurrence of such a double-coercivity behavior suggested that the two magnetic layers were magnetically decoupled from one another, which is an important prerequisite for the observation of the GMR effect.

A direct comparison between the hysteresis loop of the plane trilayer film, and the magnetoresistance behavior of a LNO\LSMO\CoFe junction, both measured at 50 K, is shown in Fig. 3. The MR ratio is defined as $[100\times(R_{ap}$

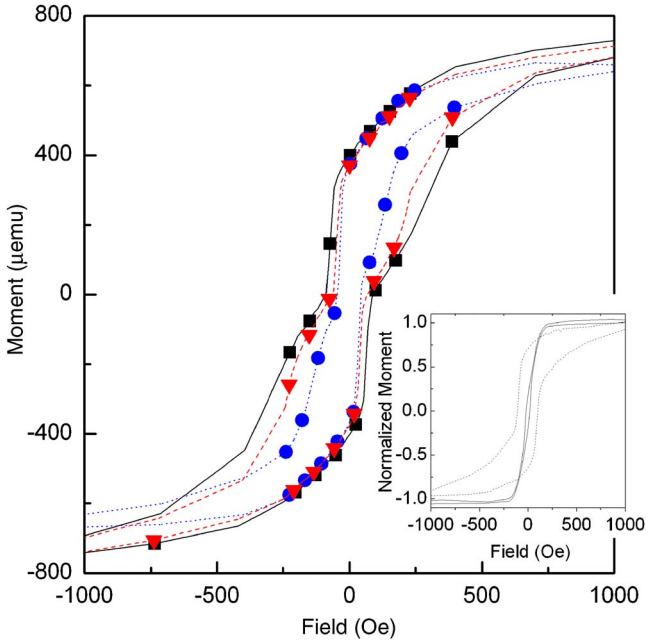

FIG. 2. (Color online) Hysteresis loops of a LNO\LSMO\CoFe trilayer film at 10 (square), 50 (triangles), and 100 K (circles). Inset: hysteresis loops of LNO\LSMO (dotted line) and CoFe (solid line) films on LAO (001) substrates measured at 100 K.

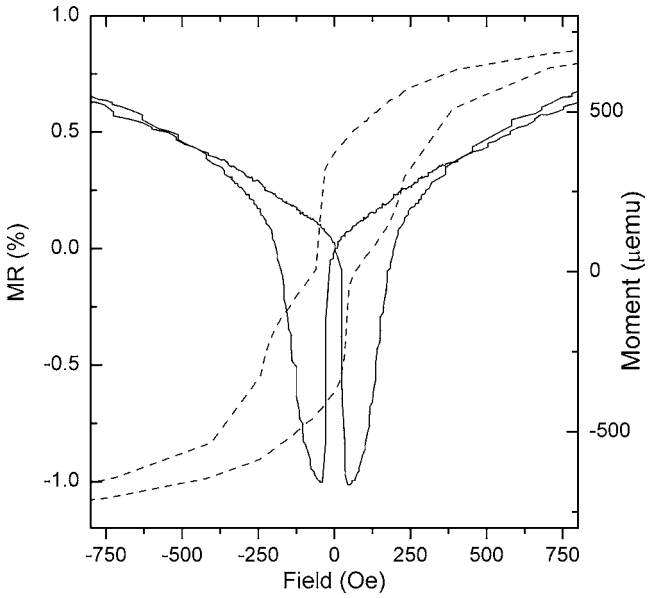

FIG. 3. Magnetoresistance of a LNO\LSMO\CoFe junction (solid line) and the magnetization of a trilayer film sample (dashed line), as a function of external in-plane field, at 50 K.

$-R_0)/R_0]$, where $R_{ap}(R_0)$ is the four-point resistance of the junction when the magnetization vectors of LSMO and CoFe layers are antiparallel (when the external applied field is zero). As shown in Fig. 3, a high resistance state was observed when a saturating magnetic field of 1 kOe was applied to the sample. As the magnetic field gradually decreased to −30 Oe (coercive field of CoFe), switching of CoFe layer magnetization occurred. An antiparallel magnetization configuration was obtained, and the MR curve rapidly switched to a low resistance state. As the magnetic field was further decreased, the magnetization of the LSMO layer was switched at about −220 Oe. The CoFe and LSMO layer magnetization became parallel aligned, and the junction switched back to the high resistance state accordingly. The same behavior was repeated for the branches of hysteresis loop and MR curve with increasing external field.

The results in Fig. 3 strongly suggested that the observed resistance changes were due to the GMR effect. The switching of the relative magnetization directions in the plane film coincided with the resistance changes of the junction. The sharp switching of the CoFe magnetization matched well with a rapid resistance drop of the junction. The resistance changes were not abrupt during the switching of the LSMO layer, which indicates an imperfect parallel/antiparallel alignment of magnetization in the magnetic layers. Nevertheless, this observation matched with gradual magnetization switching behavior of the LSMO layer displayed in the hysteresis loop in Fig. 3.

The use of nonperfect conductors (LNO and Pt) as electrodes could lead to observation of AMR effect, due to a nonzero in-plane current component in the magnetic layers. However, we suggest that the effect was minimal in our results. In case of AMR, the resistive maxima or minima of the $R(H)$ plots coincide with the coercive fields of the magnetic layers, depending on the relative orientations of the (in-plane) current and magnetizations. More importantly, the AMR behavior should be fairly symmetric about the resistance maxima/minima.[17] The resistance changes in our junction, as observed in Fig. 3, were steep at small fields. On the other hand, a more gradual switching behavior was seen at high fields. We therefore conclude that the observed magnetoresistive behavior arose from the GMR effect, with resistance changes due to the magnetization reversal of the CoFe and LSMO layers.

Our transport measurement results have shown a positive magnetoresistive behavior. That is, the resistance of the sample increased upon the application of a saturating magnetic field, which led to a *negative* MR ratio. This was in contrast to the negative MR obtained from LSMO\NiFe system studied by Ruotolo *et al.* Whether the observed GMR is positive or negative is known to be dependent on the relative sign of spin polarization in the neighboring magnetic layers:[18] Materials with opposite signs of spin polarizations have different majority spin bands, which lead to contrasting scattering probabilities for opposite spin populations when their magnetization vectors are aligned. This leads to a high resistive state at high magnetic field and hence a positive MR.[19] However, negative MR behavior has been reported for NiFe–Co (or CoFe) spin valves[20] and tunnel junctions,[21] and the two types of alloys should possess the same sign of spin polarization.[18] It is therefore expected that the same sign of MR would appear in LSMO\NiFe and LSMO\CoFe spin valves. Our results were in direct contrast to the expectation.

A more complete picture on GMR and TMR should also take into account the effect of interfacial spin asymmetry.[18] For example, Du *et al.*[22] have managed to observe both signs of GMR in FeCo\AlO_x\Co MTJs by varying the oxidation time for the aluminum oxide barrier By means of electron microscopy and holography, they speculated that the effect originated from overoxidation of the Al layer during the barrier formation process. Interfacial FeCo oxides, possessing opposite spin polarization compared to CoFe and Co, could have led to the observed effects. These results demonstrated that the magnetoresistive behavior is not simply related to the bulk of the ferromagnetic electrodes, but also the interfacial conditions have to be taken into account when investigating the magnetoresistive behavior of such devices.

Finally, Fig. 4 displays the temperature dependence of magnetoresistance in the junction. At all temperatures, positive magnetoresistive behavior was observed, but with a decreasing magnitude with the rise of temperature. A maximum magnitude of 1.2% was observed at 10 K (inset). With an increasing temperature, the fields at which the junction resistance fell showed little changes. However, the fields corresponding to the branch of increasing junction resistance dropped more quickly with temperature. This observation matched with the observations for the coercive field behavior of CoFe and LSMO layers in the temperature-dependent hysteresis measurements shown in Fig. 2. Again, this pointed to the GMR effect as the origin of the observed magnetoresistive behavior.

As seen in the inset of Fig. 4, no magnetoresistive behavior was observed above 200 K. Resistivity measurements of single-layered LSMO films, however, registered a Curie temperature of ~350 K.[23] The rapid degradation of magne-

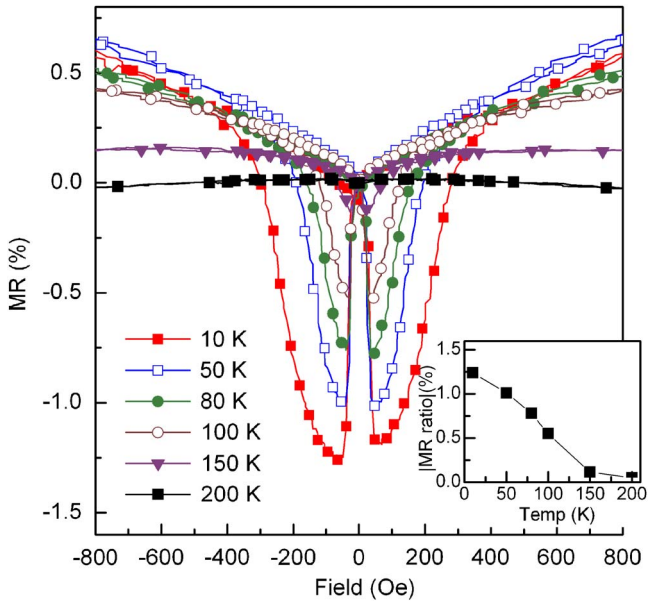

FIG. 4. (Color online) Temperature dependence of magnetoresistance for a LNO\LSMO\CoFe junction. The magnitude of MR ratio against temperature is shown as inset.

toresistance with temperature well below Curie temperature has been persistently observed in LSMO-based MTJs, but no conclusions have been drawn so far on the observation. Suggested origins for the rapid decrease of MR ratio include the inhomogeneous distribution of conductive (ferromagnetic) phases of manganites at the interfaces,[6] and the reduction of polarization at the LSMO surfaces.[24]

The exact nature of the spacer layer is of interest here, as any optimization of the magnetoresistive behavior depends on a critical understanding of the carrier transport properties at the interface. Two of the possible scenario at the interfaces include the presence of the (magnetically) dead LSMO layer[12] and the oxidation of the CoFe surface.[22] It should, however, be noted that their occurrences are not mutually exclusive. Direct visualization of the layers by high-resolution transmission electron microscopy, as well as element-sensitive surface analysis techniques, will provide a clearer picture for the mechanism of the observed magnetoresistive behavior. Further investigations are therefore under progress to examine the nature of the interfacial layer and its relation to the MR properties.

Epitaxial growth of oxide films is usually performed at elevated temperatures. This is highly undesirable for the fabrication of magnetoresistive multilayered structures. As mentioned before, typical spintronic devices impose stringent requirements on the layer thicknesses for optimized magnetoresistive properties, particularly for the spacer layers. Atomic diffusion across boundaries, assisted by the high temperature growth environment, can substantially degrade the transport behavior of the otherwise optimized structures. The present structure, with the growth of a single epitaxial layer under high temperature, provides a simple route for incorporating oxide materials into spintronic devices. By utilizing a natural barrier layer between the metal and manganite, the deposition process eliminates the need of precisely controlled multilayered deposition processes, making it a practical route for fabricating oxide-based spintronic devices with simple thin-film deposition facilities.

IV. CONCLUSION

We have fabricated LNO\LSMO\CoFe spacerless pseudo-spin-valve structure and studied their magnetic and transport properties. Magnetization and transport measurements at various temperatures suggested that GMR effect was observed in such structures. A negative MR ratio of −1.2% was observed at 10 K, and the magnetoresistive behavior persisted up to ~150 K. Given the current interest in oxide-based spintronics, the present study provides a route for incorporating oxide materials into silicon-based device technologies. Besides, the spacerless structure avoids the complications arising from the deposition of ultrathin layers, opening the possibility of studying spintronics with more robust materials preparation and thin-film deposition techniques.

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