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Tunneling anisotropic magnetoresistance in $La_{2/3}Sr_{1/3}MnO_3$/$LaAlO_3$/Pt tunnel junctions

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The magnetotransport properties of $La_{2/3}Sr_{1/3}MnO_3$(LSMO)/ $LaAlO_3$(LAO)/Pt tunneling junctions have been analyzed as a function of temperature and magnetic field. The junctions exhibit magnetoresistance (MR) values of about 37%, at H=90 kOe at low temperature. However, the temperature dependence of MR indicates a clear distinct origin than that of conventional colossal MR. In addition, tunneling anisotropic MR (TAMR) values around 4% are found at low temperature and its angular dependence reflects the expected uniaxial anisotropy. The use of TAMR response could be an alternative of much easier technological implementation than conventional MTJs since only one magnetic electrode is required, thus opening the door to the implementation of more versatile devices. However, further studies are required in order to improve the strong temperature dependence at the present stage. [http://dx.doi.org/10.1063/1.4946851]

INTRODUCTION

Magnetic tunneling junctions (MTJ) have been a subject of intense research during the last decade due to their potential interest for the implementation of high-density magnetic random access memories (MRAM).[1] Their response relies on the strong dependence of the magnetoresistance on the relative orientation of the magnetization directions in each magnetic electrode and their spin polarizations. However, the requirement of two independently controlled ferromagnetic electrodes and spin-coherent tunneling imposes some technical challenges, such as the uniformity of the magnetic properties of the electrodes, the insulating barrier uniformity or the thermal stability, for the implementation of MTJ-based devices that, in spite of the work already done, are not fully resolved yet.[2] Different attempts have been made to overcome these problems such as, for instance, increasing the magnitude of the tunneling magnetoresistance (TMR) response by using half-metallic materials as electrodes.[3,4] Even though TMR values of several hundred percent have been achieved in some cases[5–8] by using those materials, they become vanishing small well below room temperature,[5,6,9,10] therefore severely hampering technological applications. To overcome these challenges requires exploring new possibilities. An alternative to conventional TMR could be tunneling anisotropic magnetoresistance (TAMR) phenomena, i.e. the dependence of the magnetoresistance on the orientation of the magnetization of the electrodes with respect to the crystallographic axes or the current flow direction.[11,12] This angular-dependent contribution is due to the coupling of orbital and electronic spin degrees of freedom and its origins may be diverse including: (a) spin-orbit induced changes of the density of states of the ferromagnetic electrode, (b) the interference between Bychkov-Rashba and Dresselhaus spin-orbit couplings at junctions interfaces and in the tunneling region, (c) resonant states whose coupling to the scattering channels depends on magnetization direction. It is worth mentioning that TAMR is present even in the case of only one ferromagnetic electrode when conventional TMR is absent.[13,14] Since TAMR-based devices can operate with only one magnetic electrode they could be an alternative of much easier technological application than conventional MTJs, thus opening the door to the implementation of more versatile devices, avoiding the need for two independently controlled ferromagnetic electrodes and spin-coherent tunneling. Even thought TAMR was initially found in magnetic semiconductors, it has also been found in TJs with ferromagnetic metal electrodes.[14,15] Studies in ferromagnetic metal systems were initially conducted in most of the cases in spin valve-like systems with two magnetic electrodes and a complex stacking sequence.[16] However, TAMR reports in system with only one magnetic electrode are much more recent and scarce.[17,18] In these cases values of TAMR around 5-10 % at low temperature and with a strong dependence of the bias voltage are reported. In this context, the introduction of manganite electrodes could be of interest for combining different functionalities and hopefully enhancing the TAMR response while reducing the complex stacking sequence typically used in metallic MTJs. Among manganites $La_{2/3}Sr_{1/3}MnO_3$ (LSMO), with the highest Curie temperature ($T_C \sim 370$ K) of this family of materials, is the most interesting one for the implementation of devices.

In this work we report on the tunneling magnetotransport properties of the $La_{2/3}Sr_{1/3}MnO_3$ (LSMO)/ $LaAlO_3$(LAO) bilayer system as a function of temperature and applied magnetic field with the aim to analyze their TAMR response and its potential for technological applications. A Pt metallic electrode has been used because of its good adhesion properties. The conduction across the LSMO/LAO/Pt heterostructure exhibits typical features expected for a tunneling conduction process. However, the temperature dependence of the junction resistance exhibits a peak at about 170 K whose origin is not well understood yet. A clear TAMR response that amounts about 4% at low temperature is observed when a magnetic field higher than the anisotropy field, ($H_a \cong 10$ kOe) is applied parallel or perpendicular to the current flow direction.

RESULTS AND DISCUSSION

LSMO/LAO bilayers used in this work have been prepared by RF-magnetron sputtering on top of (001)-oriented $SrTiO_3$ (STO) substrates treated before deposition to select a unique atomic termination, likely to be TiO_2.[19] Bilayers are highly crystalline with a STO(001)/LSMO(001) /LAO(001) epitaxial relationship and sharp clean interfaces (see Fig. 1). LAO was chosen as insulating barrier because it has demonstrated to be less harmful for the magnetic and electronic properties of LSMO than other oxides such as $SrTiO_3$ or MgO.[20–22] LSMO layer thickness was about 40 nm while the thickness of the LAO layer was about 1.5 nm, as corroborated from TEM images (see Fig. 1), and in good agreement with the estimation from growth rate.[23]

For the fabrication of metallic contacts samples were covered with a layer of polymethyl methacrylate (PMMA) resist by spin coating, then electron beam lithography was used to define square holes in the PMMA resist. After developing, Pt was deposited by evaporation through a shadow mask defining the upper metallic electrode precisely on top of the openings of PMMA resist engraved by lithography. Further details about sample preparation can be found elsewhere.[23]

Electronic transport properties were measured in a commercial physical properties measurement system (PPMS) by Quantum Design using a 3-point configuration between a large area macro-contact and top contacts, this configuration was adopted to avoid contributions from the bottom electrode. The current was applied perpendicular to the film plane while the magnetic field was rotated from out of plane, i.e., parallel to the current direction, [001] direction, to in plane parallel to [100] direction. The dependencies on temperature, magnetic field and angle orientation between magnetization and current directions were proved.

The conduction across the LSMO/LAO/Pt heterostructure exhibits typical features expected for a tunneling conduction process (see inset in Fig. 2). However, due to the slimness of the LAO barrier and the procedure to prepare it, the existence of microstructural defects and randomly distributed pinholes through the barrier cannot be fully excluded. The temperature dependence of the junction resistance, R(T), of a 12 μm^2 LSMO/LAO/Pt junction is shown in Fig. 2. The R(T) curve exhibits a smooth increase of the resistance on lowering temperature compatible with direct

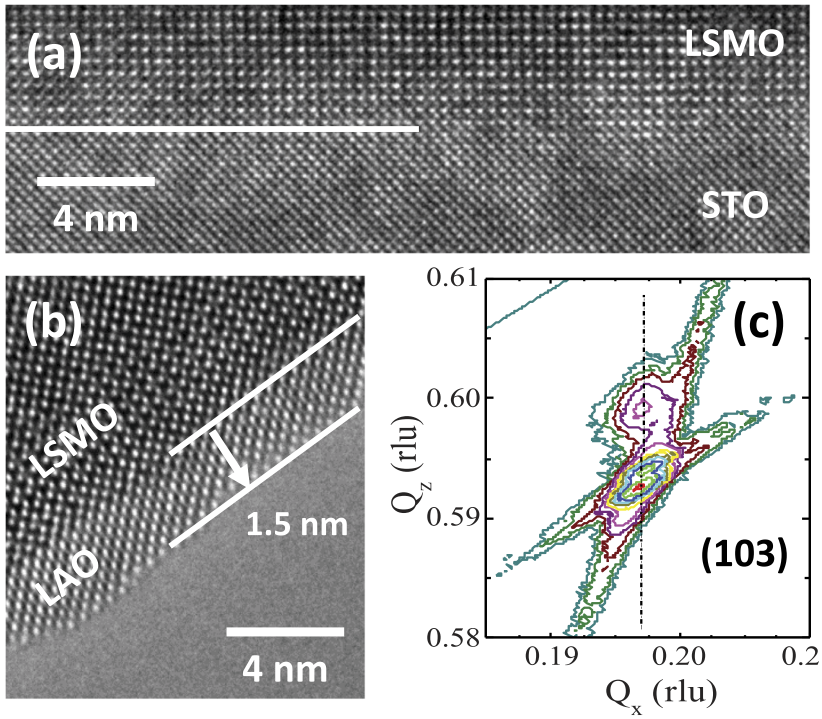

FIG. 1. HRTEM picture of the STO/LSMO (a) and LSMO/LAO (b) interfaces. (c) Reciprocal space map around the STO (103) reflection for a STO/LSMO/LAO heterostructure.

tunneling transport.[24] However, R(T) exhibits a peak with a maximum at about 170 K that seems to be a common feature in MTJs containing manganite electrodes and whose origin is not well established yet.[7,25,26] As shown by the figure the application of a magnetic field (regardless of its orientation) results in an important reduction of the junction's resistance. Interestingly, in contrast

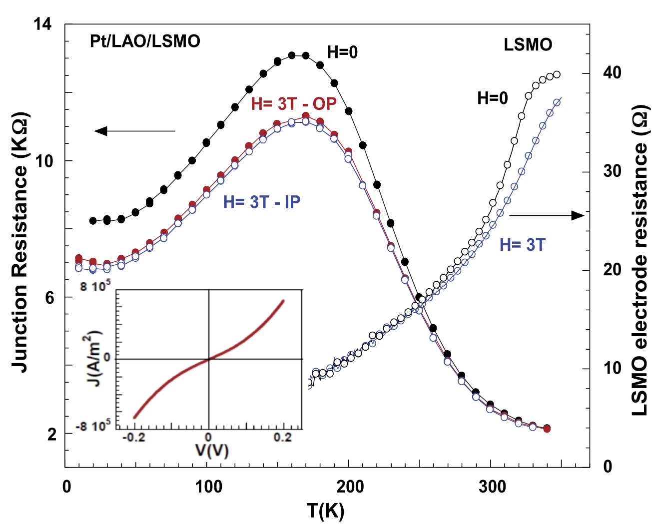

FIG. 2. Temperature dependence of the Pt/LAO/LSMO junction resistance measured in 3- terminal configuration in a 12 μm^2 junction for applied bias voltage of 100 mV. Two different values of the magnetic field are shown (H=0 (black dots) and H=3 T. The latter has been applied parallel to the junctions' plane (IP, blue open dots) and perpendicular to it (OP, red dots). The temperature dependence of the R(T) of the LSMO bottom electrode is also shown for comparison purposes (the field (H=3 T) was applied perpendicular to the film plane). Measurements were done after a zero field cooling process. The inset shows a typical I-V characteristic curve of the junction at T= 10 K.

with the typical behavior of CMR materials, no shift in the temperature of the peak of the junction resistance is observed. This key observation indicates that the measured MR has nothing to do with the typical behavior of double-exchange mediated CMR materials thus, strongly suggesting that the origin of the peak in R(T) is not related to an oxygen-deficient layer at the LSMO/LAO interface as reported previously.[26,27] The temperature dependence of the resistance of the LSMO electrode is also shown in Fig. 2. It exhibits the typical behavior of a good quality LSMO film with T_C slightly above 350 K, large CMR values close to T_C, small residual resistance at low temperature and saturation magnetization slightly below bulk saturation values. Thus, excluding a major role of effects related to phase segregation and magnetic frustration that could be important for very thin LSMO films. Worth to mention that the resistance of the LSMO electrode is almost three orders of magnitude smaller than the resistance of the TJ, thus the contribution of the LSMO electrode to the resistance of the junction is almost negligible.

The MR of the junction is better appreciated in the R(H) curves depicted in Fig. 3. A negative MR of about 37% at 90 kOe is measured at low temperature, and then it smoothly decreases when increasing temperature below 5% at room temperature (see Fig. 4) as the ferromagnetic transition temperature of the LSMO electrode ($T_C \sim 350$ K) is approached. This MR response is observed independently of the orientation of the magnetic field. The fact that this high-field MR is larger at low temperatures (10 K) than at the peak temperature of the junction R(T) (~170 K, Fig. 2 left) or the LSMO transition temperature ($T_C \sim 350$ K) further corroborates that this MR contribution cannot be ascribed to conventional CMR. However, the curve exhibits a clear signature of the R(T) peak at 170 K. The origin of this high-field MR contribution is not clear and it may well be related to magnetic disorder at the interface, which would give a MR contribution similar to that observed in LSMO granular systems.

Additionally, a MR response is also observed when the orientation of the magnetic field is changed from parallel to perpendicular to the sample surface, i.e., parallel to the current (see Figures 2 and 3). This MR response, unexpected, in principle, because the junction has only one magnetic electrode, (LSMO), corresponds to the so-called TAMR and it amounts about 4% at low temperatures. As temperature is increased, TAMR slowly decreases and vanishes close to room temperature (see Fig. 4). As mentioned in the introduction, MR in magnetic tunneling junctions may depend on the orientation of the magnetization with respect to the crystallographic axis or the direction of the current flow.[13,14] The geometry of the present LSMO/LAO/Pt stack is shown in the

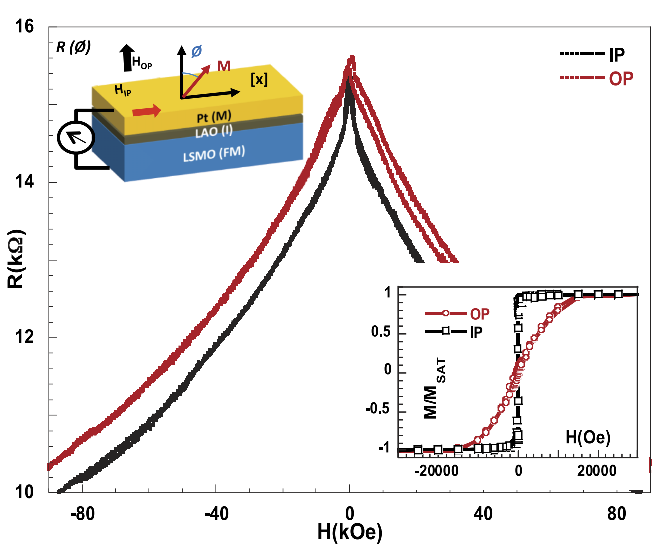

FIG. 3. R(H) curves measured at 10 K, and voltage bias of 10 mV, with the applied magnetic field in-plane (IP) and out-of-plane (OP) configurations for a 12 μm^2 Pt/LAO/LSMO junction. The corresponding hysteresis loops are shown in the lower inset. Upper inset: Sketch of the measurement configuration for the corresponding in-plane (IP) and out-of-plane (OP) applied magnetic field.

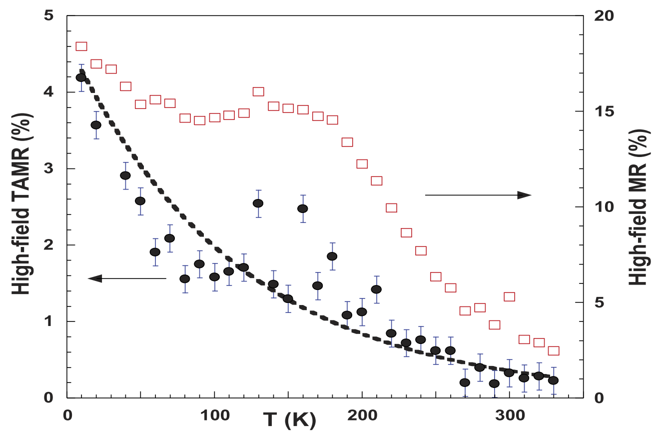

FIG. 4. Temperature dependence of the different high-field MR contributions for a 12 μm^2 Pt/LAO/LSMO junction measured at 100 mV. Red squares, referred to the right axis, depict the difference between the resistance at H=0 and 30 kOe applied in-plane (IP). Black dots, referred to the left axis, represent the difference in resistance for a fixed magnetic field of 30 kOe applied in-plane (IP) and out-of-plane (OP).

upper inset of Fig. 3 and corresponds to the so-called out-of-plane TAMR with a single magnetic electrode.[28] In these circumstances TAMR reflects the change in the tunneling magnetoresistance when the magnetization is rotated from parallel to the plane of the junction ($\phi = 90°$) to perpendicular to it ($\phi = 0°$) and is given by: TAMR(ϕ) = (R(ϕ)-R(0))/R(0), being ϕ the angle between the normal to the junction plane and the direction of the magnetization. Since the LSMO/LAO/Pt stack exhibits in-plane easy magnetization direction, a field H applied perpendicular to the stacking plane progressively drags the magnetization M out from the film plane until it is saturated in the field direction, (i.e. parallel to the current direction) once the anisotropy field is surpassed (see lower inset of Fig. 3). Thus, above the anisotropy field TAMR should be constant as evidenced in Fig. 3 where in-plane and out-of-plane branches of the R(H) curves are shown to be parallel in the high field regime. The angular dependence of TAMR is better appreciated in Fig. 5 where the

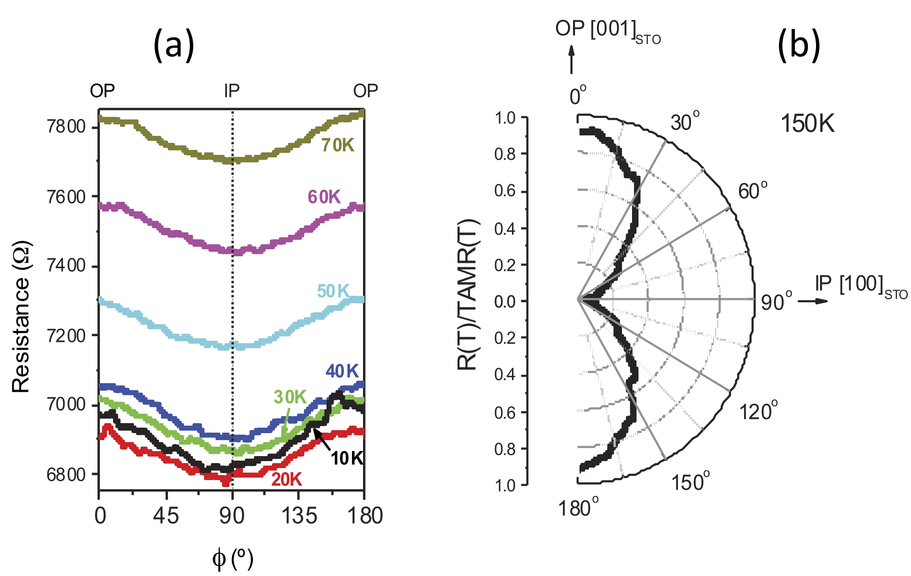

FIG. 5. TAMR at high magnetic fields of a 12 μm^2 Pt/LAO/LSMO junction. (a) Resistance as a function of the angle between the current (normal to the sample surface) and the applied magnetic field, ϕ, for several temperatures. Different colors represent measurements taken at different temperatures, from 10 K to 70 K; (b) Polar plot of the normalized R(ϕ)/TAMR at 150 K. IP and OP correspond to the configurations where magnetization is aligned in-plane and out-of-plane, respectively.

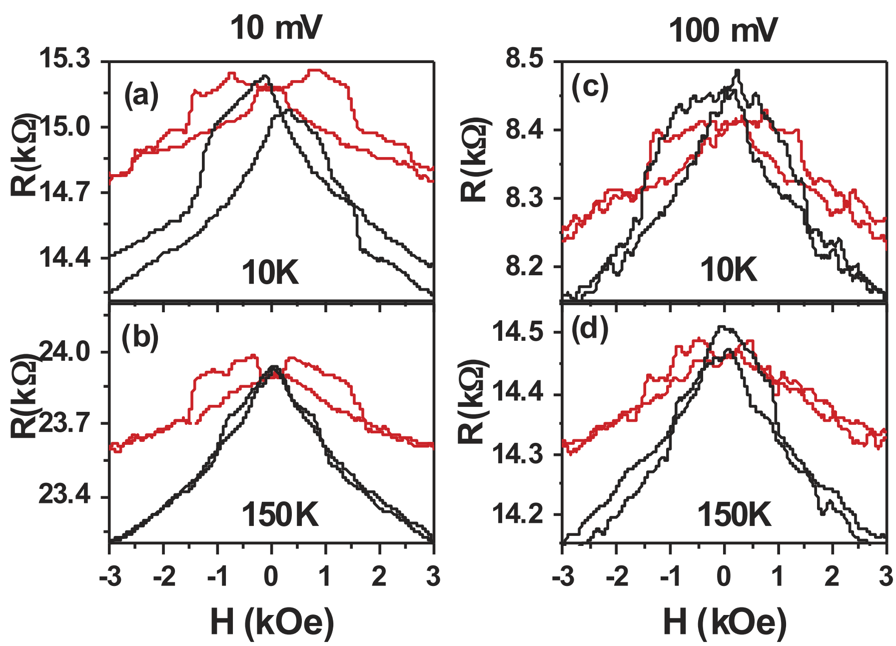

FIG. 6. TAMR at low magnetic fields of a 12 μm^2 Pt/LAO/LSMO junction: R(H) curves taken for the in-plane (IP) (black lines) and out-of-plane (OP) (red lines) configurations, at different bias voltage values (10 mV (a and b) and 100 mV (c and d)), for different temperatures (T=10 K (a and c) and T=150 K (b and d).

junction resistance is depicted as a function of the angle between the magnetization and current directions, R(ϕ), for several temperatures when the magnetic field is rotated from the [001] to the [100]. A polar plot of the normalized R(ϕ)/TAMR at 150 K is also shown in the figure making evident the expected uniaxial anisotropy character. Similar values of TAMR are obtained when the magnetic field is rotated from the [001] direction to the [010]. On the other hand, TAMR values are substantially reduced when increasing bias voltages as already observed in metallic MTJs.[16] As mentioned in the introduction, this angular-dependent contribution is due to the coupling of orbital and electronic spin degrees of freedom however; our macroscopic measurements do not allow determining the microscopic origin.

In the low field regime, i.e. below the anisotropy field, M and H may not be fully aligned and R(H) curves exhibit switching between distinct resistance states in a spin-valve-like fashion (see Fig. 6). As our system has a single ferromagnetic electrode, TAMR must be at the origin of such behavior through a magnetization reversal that takes place in a multi-step process, so that the jumps in resistance correspond to the rotation of different magnetic domains.[14,29] As shown in Fig. 6 the effect is stronger for smaller bias voltages and lower temperatures; TAMR is hardly distinguishable from the noise for the measurement taken at 150 K and 100 mV. This could be attributed to the fact that providing more energy to the system (via thermal energy or via electric field) leads to a faster magnetization reversal, with no intermediate magnetization steps.

In summary, the magnetotransport properties of LSMO/LAO/Pt tunneling junctions have been analyzed as a function of temperature and magnetic field demonstrating the existence of TAMR response. The temperature dependence of the junction resistance, R(T), exhibits a peak at about 170 K , as in the case of other tunneling junctions with manganite electrodes, whose origin is not well understood yet. Our system exhibits MR values of about 37%, at H=90 kOe and low temperature, and its temperature dependence, which cannot be explained through conventional CMR mechanisms, strongly suggests that the previously reported scenario of an interfacial layer of under-doped manganite with reduced ordering temperature is very unlikely. This MR response may well be related to magnetic disorder at the interface. In addition, the system exhibits TAMR values of around 4% at low temperature, despite the weak spin-orbit coupling in this system, and its angular dependence reflects the expected uniaxial anisotropy. These values are very similar to that reported for ferromagnetic metal MTJ but with a much more simple stacking sequence which would be of major interest for the implementation of TAMR-based devices. The use of TAMR response could be an alternative of much easier technological implementation than conventional MTJs opening the door to the implementation of more versatile devices avoiding the need for two independently controlled ferromagnetic electrodes and spin-coherent tunneling. However, further studies are required in order to improve the strong temperature dependence that severely restricts potential technological applications in the present stage.

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