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Low-field magnetoresistance anisotropy in ultrathin $Pr_{0.67}Sr_{0.33}MnO_3$ films grown on different substrates

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We have conducted a comparative study of the strain effect on the anisotropic low-field magnetoresistance (LFMR) in ultrathin $Pr_{0.67}Sr_{0.33}MnO_3$ films epitaxially grown on $LaAlO_3$(LAO), $NdGaO_3$(NGO), and $SrTiO_3$(STO) substrates. Distinctive LFMR effects have been observed on films with compressive (on LAO), tensile (on STO), and nearly zero (on NGO) strains. The films with compressive strain show very large negative LFMR and MR hysteresis when a magnetic field is applied perpendicular to the film plane, while those with tensile strain show positive LFMR for the same field orientation. All samples show negative MR in a parallel magnetic field. These results can be qualitatively interpreted based on the strain-induced magnetic anisotropy. © *1999 American Institute of Physics.* [S0003-6951(99)01815-X]

The doped manganese oxides exhibit a variety of interesting properties, such as the colossal magnetoresistance (CMR) effect, which are potentially useful in device applications. Most of the proposed applications involve thin films in which the physical properties can be quite different from their bulk counterparts due to strain. It has been shown that in bulk materials, the Curie temperature T_c and the CMR in high magnetic fields are extremely sensitive to stoichiometry and hydrostatic pressure.[1,2] Recently, several groups have reported that lattice strain affects the peak resistance temperature T_p and the CMR properties of $La_{0.7}Sr_{0.3}MnO_3$,[3] $La_{0.7}Ca_{0.3}MnO_3$,[4] $La_{0.8}Ca_{0.2}MnO_3$[5] thin films, and the strain-induced magnetoelastic interaction plays a dominant role in the magnetic anisotropy.[3,6,7] We have previously reported that a large low-field magnetoresistance (LFMR) has been obtained in compressively strained ultrathin $Pr_{0.67}Sr_{0.33}MnO_3$ (PSMO) films.[8] These results demonstrate that lattice distortions in manganite films can dramatically alter their physical properties.

In this letter, we report a systematic study of strain effect on the anisotropic LFMR properties of ultrathin $Pr_{0.67}Sr_{0.33}MnO_3$ films grown on $LaAlO_3$ (001) (LAO), $SrTiO_3$ (001) (STO), and $NdGaO_3$ (110) (NGO) substrates, which impose compressive, tensile, and nearly zero strains respectively in the films. We have focused our attention to ultrathin films (5–15 nm) in which the strain distribution is relatively uniform. We have found very different effects of compressive and tensile strain on the magnetic and the LFMR properties. The compressive strains cause the easy magnetization axis to be perpendicular to the film plane, whereas tensile strains result in an easy axis parallel to the plane. When a magnetic field is applied perpendicular to the film plane, the compressive-strain films show a very large negative LFMR, while the tensile-strain films show a positive LFMR. The nearly strain-free films show a very small negative LFMR. All samples show negative MRs when the field is applied parallel to the plane. The results demonstrate that strain-induced anisotropic magnetization plays a major role in determining the LFMR properties of the films.

Epitaxial PSMO films were grown on LAO, NGO, and STO substrates by pulsed-laser deposition. Details of the film preparation and MR measurement have been described previously.[8] LAO is nearly cubic with a lattice constant of $a=3.79$ Å, STO is cubic with $a=3.905$ Å, and NGO is orthorhombic with the lattice parameters of $(a^2+b^2)^{1/2}/2=3.862$ Å and $c/2=3.854$ Å. Bulk PSMO is orthorhombic with $a/\sqrt{2}=3.879$ Å, $b/\sqrt{2}=3.866$ Å, and $c/2=3.856$ Å.[9] Therefore, among the three types of films studied, the PSMO/NGO films have the smallest film-substrate lattice mismatch ($\sim-0.3\%$), and hence the smallest lattice distortion. Both PSMO/LAO ($\sim-2\%$) and PSMO/STO ($\sim1.0\%$) films have large lattice mismatches with their in-plane lattice parameters compressed and expanded respectively. X-ray diffraction (XRD) experiments reveal that the films are *c*-axis oriented. The *c*-axis lattice parameter of the PSMO/NGO film is essentially thickness independent and very close to the bulk value, while those of the ultrathin PSMO/LAO and PSMO/STO films are enlarged to 3.95 Å and reduced to 3.82 Å, respectively. The resistivities of all the samples show a crossover at T_p from a high temperature semiconducting state to a low temperature metallic state, typical for manganites.

The anisotropic LFMR properties of the three different types of samples are shown in Figs. 1–3. Figure 1 shows the MR results of a 7.5-nm-thick PSMO/LAO film measured with the field parallel and perpendicular to the film plane at 50 K. In the perpendicular-field geometry, a very large MR with pronounced hysteresis is observed. The peak resistance (R_p) occurs at a field of about 800 Oe. The temperature dependence of the LFMR, defined as $[R(5\,\mathrm{kOe})-R_p]/R(5\,\mathrm{kOe})$, is shown in the inset. In contrast, in the parallel-field geometry, the MR hysteresis is much weaker and the LFMR ratio is less temperature dependent. Figure 2 shows the MR hysteresis loops of a 7.5-nm-thick PSMO/NGO film in different field and current directions. The MR characteristics are very different from those of the PSMO/

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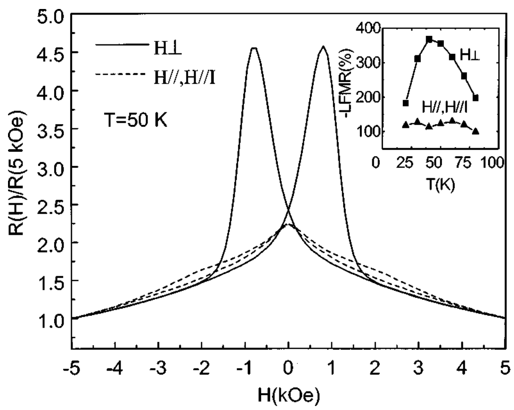

FIG. 1. The magnetoresistance $R(H)$, normalized to its value at 5 kOe, of a 7.5-nm-thick PSMO/LAO film measured with the magnetic field parallel ($H_{\|}$) and perpendicular ($H_{\perp}$) to the film plane. Inset is the temperature dependence of the LFMR as defined in the text.

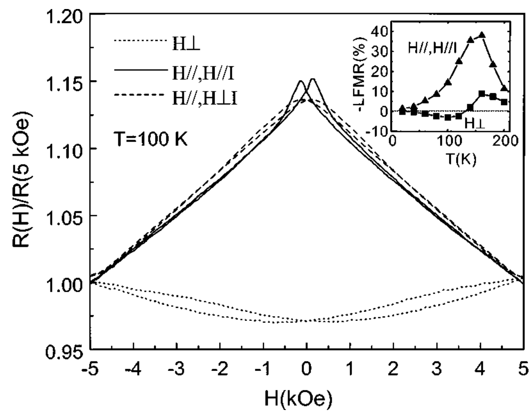

FIG. 3. Normalized magnetoresistance of a 15-nm-thick PSMO/STO film measured with the magnetic field normal to the film plane ($H_{\perp}$), in-plane and parallel to current ($H_{\|}, H_{\|}I$) and in-plane but perpendicular to current ($H_{\|}, H_{\perp}I$). Inset is the temperature dependence of the LFMR for the cases of ($H_{\perp}$) and ($H_{\|}, H_{\|}I$).

LAO, in particular, the MR ratio is orders of magnitude smaller. Note that the MR hysteresis was observed in all three cases, although the magnitudes and the peak positions are different.

Shown in Fig. 3 are the MR results of a 15-nm-thick PSMO/STO film. Strikingly, the MR has different signs for different field directions. It is *positive* for the perpendicular field and negative for the parallel field. Also, the MR ratios are different for the two field directions and both are orders of magnitude smaller than those of the PSMO/LAO samples. The positive MR effect in the perpendicular-field geometry is only present in small magnetic fields. When the applied field is larger than 1–2 T, the MR becomes negative. The positive MR also has a strong temperature dependence. It is readily observable when the temperature is about 20–30 K below T_p. Approaching T_p, the positive MR crosses over to a negative MR as shown in the inset (here the definition of the LFMR is the same as in Fig. 1, except R_p now represents for the minimum resistance).

It should be noted that the MR curves presented in Figs. 1–3 are not at the same temperature, but at the temperature where the largest LFMR is obtained for each sample. At low temperatures the qualitative features of the MR results are similar to those in Figs. 1–3.

With the exception of the PSMO/NGO samples, the measured MR properties depend strongly on the film thickness. In Fig. 4, we show $(R_p - R_0)/R_0$ as a function of the film thickness for a group of PSMO/LAO films, where the MR ratio decreases rapidly with increasing film thickness. For a 20-nm-thick sample, the LFMR is reduced to less than 10%. In thicker PSMO/STO films, the positive MR behavior also disappears. These effects are probably due to the gradual release of strain and size effect as the thickness of the sample increases.

In bulk manganites, there is essentially no MR anisotropy.[10] The striking difference of the MR hysteresis behaviors of the films on different substrates are closely related to their strain state and magnetization properties. Figure 5(a) and 5(b) show the magnetization curves of a 6-nm-thick PSMO/LAO film and a 10-nm-thick PSMO/STO film measured in two different field directions. The nearly square shaped magnetization hysteresis loop for the perpendicular field ($H_{\perp}$) in Fig. 5(a) is a clear indication of the out-of-plane easy axis of the PSMO/LAO film. In contrast, the curves in Fig. 5(b) demonstrate an in-plane easy axis of the PSMO/STO film. We were unable to determine the easy axis of the PSMO/NGO samples due to the very large paramagnetic signal from the NGO substrate. However, since the demagnetization field is in the film plane and the strain an-

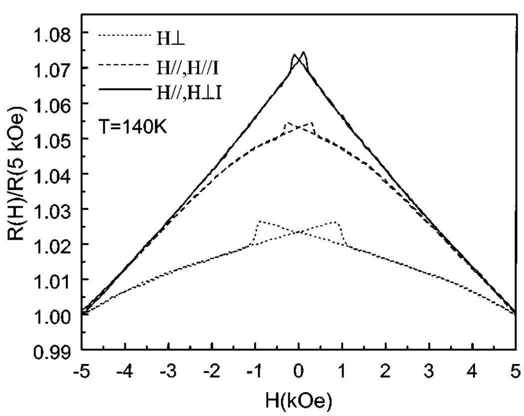

FIG. 2. Normalized magnetoresistance of a 7.5-nm-thick PSMO/NGO film measured with the magnetic field normal to the film plane ($H_{\perp}$), in-plane and parallel to current ($H_{\|}, H_{\|}I$) and in-plane but perpendicular to current ($H_{\|}, H_{\perp}I$).

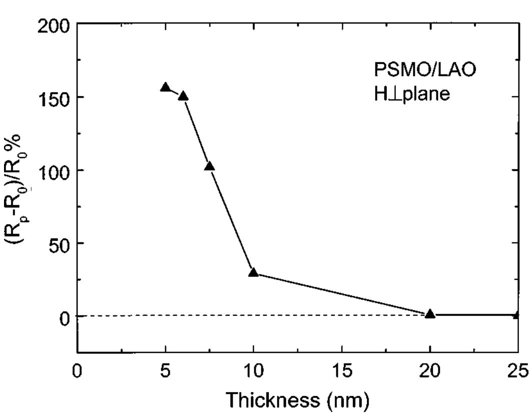

FIG. 4. Thickness dependence of $(R_p R_0)/R_0$ of PSMO/LAO films, obtained from the $R-H$ loops with perpendicular field at temperatures where the MR hysteresis is the largest for each sample. The solid line is guide to the eyes.

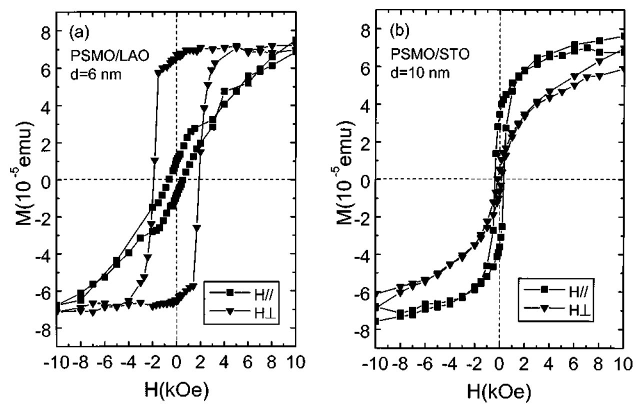

FIG. 5. Magnetization curves of (a) PSMO/LAO and (b) PSMO/STO films measured with magnetic field perpendicular ($H_{\perp}$) and parallel ($H_{\parallel}$) to the film plane.

isotropy field is rather small, it is reasonable to expect that the easy axis lies in the plane.

The anomalous anisotropic LFMR properties of the differently strained samples can be explained in part by the magnetic anisotropy and the domain movement. In the PSMO/LAO films, the strain-induced anisotropy field, which favors an out-of-plane magnetization, is larger than the demagnetization field, resulting in an out-of-plane easy axis.[7] Under a large perpendicular field, all magnetic domains are aligned along the field direction, and hence the resistance is low. When the field is near the coercive field, oppositely aligned domains are present. The resulting high resistance is most likely due to spin dependent scattering from the oppositely aligned domains. On the other hand, when the field is applied in the plane, a much higher field is required to align all the domains along the magnetic hard axis and hence the MR ratio at low field is relatively small. Since a large MR due to spin-dependent scattering relies on a high density of unaligned ferromagnetic entities, it suggests the existence of numerous perpendicularly magnetized domains with very thin walls in ultrathin PSMO/LAO films. The strong thickness dependence of the LFMR may be due to the changes of the domain wall width as a function of the film thickness.

In the PSMO/STO samples, the tensile strain induces a magnetic anisotropy, which favors an in-plane magnetization. When a perpendicular field is applied, the magnetization will rotate out of the plane and be perpendicular to the film plane, when H is larger than a threshold field of $H_t = H_K + H_D$, where H_K is the strain-induced anisotropy field, and $H_D = 4\pi M$ is the demagnetization field. As discussed in detail by Eckstein *et al.*,[11] the resistance increases with field due to the increasing angle between the measuring current and the magnetization, resulting in a positive MR. The positive MR crosses over to a negative MR due to the intrinsic negative CMR when $H > H_t$, as the magnetization increases with field. In the temperature ranges of 60–100 K, the measured H_t for a 10-nm-thick PSMO/STO film is about 1 T, comparable to the sum of the demagnetization field and the anisotropy field measured.[3,6,12] In a parallel field, the LFMR hysteresis is due to the domain rotation and movement within the film plane and the negative MR is due to the increased magnetization in the field direction. It should be noted that unlike the perpendicular domains in the PSMO/LAO samples, the domains in this case do not cause a large MR as the anisotropic MR in usual ferromagnetic materials.

For the PSMO/NGO samples, though we expect the spontaneous magnetization to lie in the plane, we did not observe a positive MR in a perpendicular field in most of the samples (except one at very low temperatures). This is probably because of the small H_t due to the small anisotropic field H_K (caused by the small compressive strain) which partially cancels out H_D.

Based on these considerations, the strain-induced magnetic anisotropy plays a crucial role in determining the LFMR properties in the ultrathin PSMO films. However, this mechanism alone may not be sufficient to account for all aspects of the experimental results such as the thickness and temperature dependencies of the LFMR. Other factors such as structural defects and spin disorder must also be considered in order to analyze quantitatively the LFMR properties of films on different substrates. Due to the large lattice mismatch, structural disorder may be introduced during film processing. Our XRD experiments have in fact shown that the diffraction peaks of the ultrathin PSMO/LAO and PSMO/STO films are broader than those of the unstrained PSMO/NGO films. However, the distinctive features we have observed in different samples are mainly due to the strain-induced magnetic anisotropy.

In summary, we have studied the strain effects on the magnetoresistance of the ultrathin PSMO films. Dramatic differences in the low-field MR properties have been observed in films with different types of strains. The anomalous anisotropic MR effects are mainly attributed to the strain-induced magnetic anisotropy caused by lattice distortions.

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