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Domain structure and magnetotransport in epitaxial colossal magnetoresistance thin films

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Our studies of compressively strained $La_{0.7}Sr_{0.3}MnO_7$ (LSMO) thin films reveal the importance of domain structure and strain effects in the magnetization reversal and magnetotransport. Normal and grazing incidence x-ray diffraction indicate that the compressive strain on these LSMO thin films on (100) $LaAlO_3$ is not completely relaxed up to thicknesses on the order of 1000 Å. The effect of the compressive strain is evident in the shape of the magnetization loops and the magnetotransport measurements at various temperatures. Although the domain wall contribution to the magnetoresistance is significantly larger than that predicted from a simple double exchange picture, the contribution is a small fraction of the measured magnetoresistance. © *2000 American Institute of Physics.* [S0021-8979(00)55608-4]

The role of domain structure in transport properties has been the focus of recent research in epitaxial colossal magnetoresistance (CMR) films. In elemental magnetic thin films, the effect of domain walls (DWs) on resistivity has been explained in different ways; the effect of internal fields and surface scattering on electron trajectories, a Hall effect mechanism, a two channel conduction model in ferromagnets, and spin dependent scattering.[1–3] In these systems, the contribution of domain walls to resistivity has been found to be small compared to conventional anisotropic transport effects, and can be either positive (increase resistivity) or negative. In colossal MR films where mean free paths are small, the effect of internal fields on the diffuse scattering of electrons at the film interfaces is negligible and the overall contribution of the DW is to enhance the resistivity.

In CMR thin films, Mathur *et al.*[4] report that the measured resistivity of a magnetic domain wall is four orders of magnitude larger than that predicted by a simple double exchange picture. Wang *et al.*[5] have suggested that DW scattering is the mechanism explaining a low field MR observed in compressively strained epitaxial (Pr,Sr)MnO_3 thin films. We present, in this paper, results that indicate that although the DW contribution to the resistivity is significantly larger than that predicted from a simple double exchange picture, the contribution is a small fraction of the measured MR.

We have studied the effect of DW transport in compressively strained epitaxial LSMO thin films on $LaAlO_3$ substrates. These films are grown using conventional pulsed laser deposition (PLD) where three-dimensional island growth gives rise to partially relaxed films. Details of the growth conditions are described elsewhere.[6] Rutherford backscattering spectroscopy indicates that the film has, to within the accuracy of the measurement, the composition of the target. Normal incidence x-ray diffraction indicates that the films are oriented in and out of the plane of the film and thus can be called single crystalline. Normal incidence diffraction, in combination with grazing incidence diffraction (GID), provides us with in-plane and out-of-plane lattice parameters. Since GID only penetrates 100 Å beneath the surface of the film, it provides insight into the relaxation of the lattice that occurs for thicker films. Figure 1 below shows a plot of LSMO/LAO films from 280 to 4500 Å thick. For comparison, the LSMO bulk lattice parameters are $a_{\mathrm{bulk}}=b_{\mathrm{bulk}}=c_{\mathrm{bulk}}=3.88$ Å and the substrate lattice parameters are $a=b=c=3.79$ Å. Figure 1 indicates that even in films on the order of 1000 Å, the strain is not completely relaxed. The strain in these films can be written down in terms of a bulk strain ($\epsilon_B=\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz}$), associated with a hydrostatic volume distortion, and a volume preserving Jahn-Teller strain ($\epsilon_{\mathrm{JT}}=(1/\sqrt{6})(2\epsilon_{zz}-\epsilon_{xx}-\epsilon_{yy})$), associated with the lattice mismatch between film and substrate.[7] Since we have direct measurements of the lattice parameters both in and out of the plane, we can easily separate these two contributions. In agreement with the results of Rao *et al.*[8] and Millis *et al.*,[9] we observe an increasing Jahn-Teller strain with decreasing

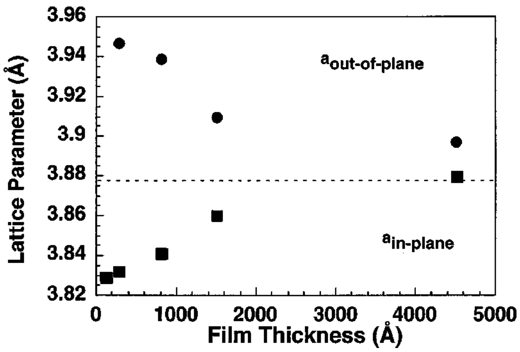

FIG. 1. Relaxation of the lattice parameters in and out of the plane of the film is observed in CMR films of thickness 100–4500 Å.

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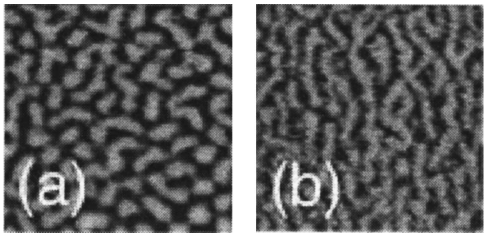

FIG. 2. Magnetic domain structure of a 1400 Å LSMO/LAO film after saturation in a direction (a) perpendicular to the plane of the film and (b) parallel to the film plane at room temperature.

thickness while the bulk strain contribution becomes more negative with decreasing thickness. Therefore volume preserving Jahn-Teller strain is a dominant factor in the epitaxial thin LSMO films of all thicknesses. These GID measurements are consistent with a biaxial compressive strain due to the lattice mismatch between the film and the substrate. This biaxial strain is predicted to give rise to a perpendicular anisotropy.[10]

Magnetic force microscopy (MFM) provides us with images of the domain structure of our films in their zero field state after saturation. In Fig. 2 are images of a 1400 Å thick LSMO/LAO thin film in its zero field state after saturation in a direction normal to the film plane (a) and in the plane of the film (b). In (b), the domains are predominantly elongated along the direction of the saturating magnetic field. Therefore, if scattering at DW's contributes significantly to the resistivity, the DW effect should manifest itself in the zero field resistance of the same sample poled in different directions.

We measured the MR of films of varying thicknesses and found that the main MR effects can be understood in terms of bulk colossal MR and anisotropic MR. Figure 3 shows MR curves for a 800 Å thick LSMO/LAO film from room temperature down to 4 K with the magnetic field applied in the plane of the film and perpendicular to the current. The MR curves exhibit a low field hysteretic behavior and a sublinear deviation from the linear high field behavior. In the limit of extremely high magnetic field, we would expect the MR to approach a constant value. However our MR measurements up to 6 T, as well as those of other groups,[11] indicate that we continue to suppress spin fluctuations even at these high magnetic field values. Below magnetic fields corresponding to saturation in the MH loop (Fig. 3), the MR deviates from linearity. At these fields, moments rotating out from the applied field direction and out of the film plane direction, as observed, would lead to a superlinear deviation of the MR. However, the resistivity anisotropy in our films is such that the resistivity is smallest when the magnetization is perpendicular to the film plane and for this reason the MR deviation is sublinear.[12]

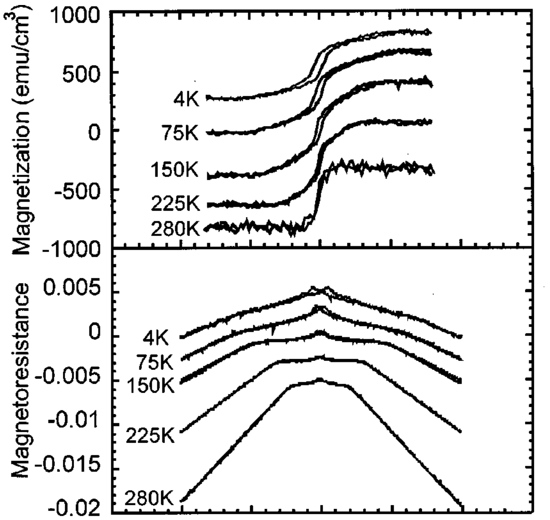

FIG. 3. Magnetization and magnetotransport curves of a 800 Å thick LSMO film with the magnetic field applied in the plane of the film and perpendicular to the current.

As the temperature is reduced below room temperature, we observe an increase in the coercive field that directly correlates with the hysteretic behavior in the MR measurements, thus confirming that the MR hysteresis is associated with domain wall motion. With decreasing temperature, the remnant magnetization increases significantly. This remnant magnetization indicates that the moments have a non-negligible projection of magnetization in the plane of the film. Therefore moments in the demagnetized state do not exhibit true perpendicular anisotropy but are most likely canted out of the film normal direction.

Now let us turn to MR measurements where the applied magnetic field is perpendicular to the current and the plane of the film. We observe a negative MR with a high field linear dependence on the applied field. We attribute this negative MR to the suppression of spin fluctuations at high fields (CMR). At lower fields, there is a sublinear deviation of the MR from the linear high field behavior, with the resistivity approximately field independent. The field at which the deviation occurs corresponds to the magnetic saturation field (Fig. 4). As the field is lowered below the saturation field, reversed magnetic domains form and, while the applied field is varying, the internal magnetic field B is approximately constant and the CMR is suppressed.

As a function of temperature, we observe very little change in the MR and MH loops (Fig. 4). With decreasing temperature, the perpendicular anisotropy associated with the biaxial compressive strain increases faster than the magnetization, thus leading to higher remanence. At lower temperatures, we also observe an increase in the coercive field which is consistent with an increasing in-plane component of magnetization.

The difference between the in-plane transverse and perpendicular MR at $H=0$ reflects the effect of domain configurations on film resistivity. However we need to take into account the fact that in the ''maze'' configuration [Fig. 2(a)] a portion of the current is shunted by the domains and need not cross domain walls, thus producing a smaller resistivity. With this consideration in mind, we have estimated the DW MR from the MR data to be 3×10^{-3} at room temperature.[13] A simple double exchange picture predicts a DW MR of 1.5×10^{-4}.[13] As the temperature is lowered, the DW contribution to the MR remains the same order of magnitude. An

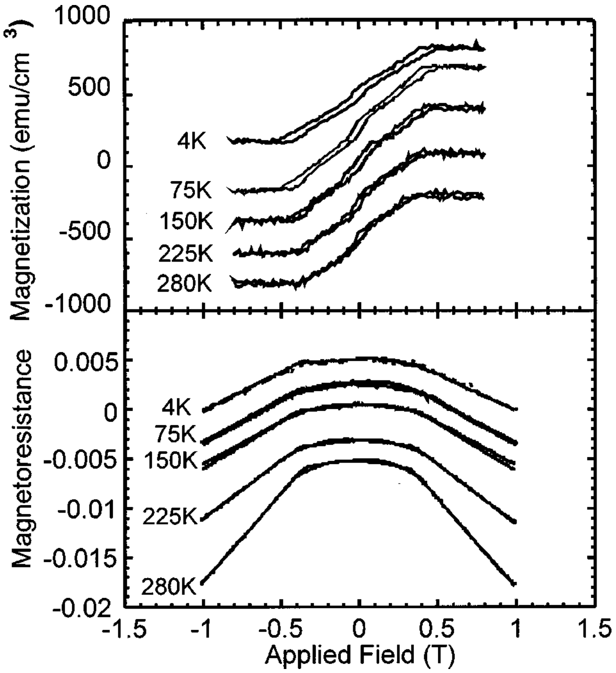

FIG. 4. Magnetization and magnetotransport curves of an 800 Å thick LSMO film with the magnetic field applied normal to the plane of the film.

increasing in-plane magnetization component renders the DW resistivity estimate a limiting case, thus emphasizing even more the discrepancy between experiment and a simple double exchange model. A more detailed model of the domain wall MR, in double exchange ferromagnets, predicts a 1%–2% effect which is consistent with our measured MR.[14]

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[12] In magnetic crystals, the spin–orbit interaction, which is at the microscopic origin of this resistivity anisotropy or anisotropic MR (AMR) effect, leads to a resistivity which depends not only on the orientation of magnetization and current and but also on the relation of these vectors to the crystal structure. For example, resistivity anisotropy of this type has recently been reported for transition metal epitaxial thin films.

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