Thickness dependent magnetotransport in ultra-thin manganite films

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Abstract

To understand the near-interface magnetism in manganites, uniform, ultra-thin films of La$_{0.67}$Sr$_{0.33}$MnO$_3$ were grown epitaxially on single crystal (001) LaAlO$_3$ and (110) NdGaO$_3$ substrates. The temperature and magnetic field dependent film resistance is used to probe the film’s structural and magnetic properties. A surface and/or interface related dead-layer is inferred from the thickness dependent resistance and magnetoresistance. The total thickness of the dead layer is estimated to be $\sim 30$ Å for films on NdGaO$_3$ and $\sim 50$ Å for films on LaAlO$_3$.

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One issue in manganite trilayer junction is the premature disappearance of magnetoresistance (MR) upon temperature increase. The MR in La$_{0.67}$Sr$_{0.33}$MnO$_3$ based trilayer junctions disappears above 150 K, well below the electrode’s Curie temperature of 360 K [1–7]. Recently, spin-resolved photoemission measured the spin-polarization of surface electronic density-of-state of epitaxial thin films of La$_{0.67}$Sr$_{0.33}$MnO$_3$ [8,9]. It revealed, as a function of increasing temperature, a more rapid decrease of the surface spin polarization than the film’s overall magnetization. Both transport and photoemission experiments suggest the possible existence of a surface dead-layer with depressed magnetic order at elevated temperatures. Questions remain as to the depth of this dead-layer, and its physical origin. In this paper, we experimentally establish some estimates about the dead-layer thickness. The approach is to study the thickness dependent electrical transport, including resistance and magnetoresistance, in ultra-thin epitaxial films of La$_{0.67}$Sr$_{0.33}$MnO$_3$ (LSMO). To estimate the effect of lattice strain, films on two types of substrates are compared — those grown on (001) LaAlO$_3$ (LAO) and on (110) NdGaO$_3$ (NGO). The film thickness range covered by this study is between 15 Å and 240 Å.

Films were grown epitaxially using laser ablation from a stoichiometric La$_{0.67}$Sr$_{0.33}$MnO$_3$ target. A Nd-YAG laser was used for ablation, operating in frequency-tripled mode at 355 nm, with a pulse energy of 140 mJ/pulse at 10 Hz repetition. The deposition rate was about 0.6 Å/sec. A series of films were made in the thickness range between 15 Å and 240 Å. Two substrates, one LAO and one NGO, each 1 cm × 2 mm × 0.5 mm in size, were loaded side-by-side for simultaneous deposition at each thickness. The substrate holder’s temperature during growth was 750 C, and the growth ambient consisted of 300 mτ of oxygen at a flow rate of 50 sccm.

The films were subsequently coated with four in-line contact pads, 1 mm diameter and 2.2 mm apart from each other, for transport measurement. These contact pads, usually silver or gold, about 1000 Å thick, were sputter deposited through a stencil mask. The four-point resistance thus measured was assumed to be the resistance-square $R_{\square}$. The film thickness was deduced from deposition time normalized to calibration runs.
A summary of the temperature dependent film resistivity, $\rho(T)$, is shown in Fig.1(a). An increase of resistivity is seen as the film thickness is decreased, for films both on NGO and on LAO. Films on LAO show faster increase in resistivity upon thickness reduction. At 15 Å, the $\rho(T)$ for film on NGO just begins to show a negative temperature slope at 14 K, with its $\rho(T)$ peak temperature moved down to around 240 K from the bulk value of 360K. The $\rho(T)$ for the 15 Å film on LAO, on the other hand, already shows a pronounced resistivity rise for $T < 150$ K. The increase of resistivity with decreasing film thickness indicates the presence of a dead-layer with reduced conductivity. This can be seen more clearly in the thickness dependence of the film’s total conductance $G$ (defined as $G = 1/R_{\Box}$, where $R_{\Box}$ is the film’s 4-probe-measured resistance).

Fig.1(b) shows the thickness dependence of $G$ at $T = 14$ K, the lowest temperature the film resistance was measured down to. A linear thickness dependence of $G$ is expected. If the films were uniform, the line should intercept the horizontal axis at zero thickness. Data in Fig.1(b) shows a linear thickness dependence for films thicker than 60 Å, but with a finite intercept. This intercept represents the total thickness of the dead-layer(s), if these dead-layers are much less conducting than the center part of the film. The total dead-layer thickness according to this estimate is around 30 Å for films grown on NGO, and around 50 Å for films on LAO. This thickness estimate combines the contribution of the surface dead-layer and the film-substrate interface dead-layer. It is not yet clear whether the thicknesses of these two types of dead-layers are the same for a given film.

Next we rule out two other possibilities that could contribute to a thickness-dependent film resistivity, namely the film’s strain variation, and the incomplete film coverage over the substrate during initial growth.

Fig.4 shows results of X-ray diffraction measurements on lattice parameters of the films. Both out-of-plane and in-plane lattice parameters were measured, using normal and grazing incident X-ray diffraction [10]. The film’s unit cell dimensions deviate from their bulk values for the entire thickness range, presumably due to the combined effect of epitaxial strain and oxygen defects. The deviation is greater for films on LAO than for films on
NGO. A thickness-dependent change of lattice parameter is seen. This change, < 0.76% for out-of-plane, < 1.2% for in-plane lattice in the case of film on LAO, is small compared to the difference in corresponding lattice parameters between films grown on LAO and NGO.

The main contribution to resistivity’s thickness dependence is not from film strain variation. This can be seen by comparing the strain-field difference and resistivity difference for films grown on different substrates. For the 240 Å film on LAO and NGO, their out-of-plane lattice constants differ from bulk value by 2.12% and 0.83%, respectively. This is a difference in strain-field of over a factor of 2.5 in the out-of-plane direction. Yet their averaged resistivity at 14 K differ only by a factor of 1.3, with dead-layer effects included. Thus, a relatively small strain variation of 0.2% ~ 0.5% over the thickness range is not expected to cause much variation to the resistivity. The observed thickness-dependent resistivity varies over an order of magnitude, and can not be explained away by strain variation.

The continuous coverage of such thin films over the substrate surface was confirmed using atomic force microscopy (AFM). Fig.3 shows a morphology image of two 30 Å films, one on LAO, another on NGO. From these images the film’s surface roughness is estimated to be about 5 Å peak-to-peak in both cases, indicating continuous film coverage without large voids.

With strain effect and initial coverage issues resolved, we thus conclude that the thickness-dependent resistivity is most likely caused by the presence of a surface and/or interface dead-layer.

The MR of the films also show thickness and substrate dependence. Fig.4 shows MR vs. temperature for films on NGO and LAO at different thicknesses. Two types of behaviors are distinguishable. In the first type the MR rises monotonically as a function of temperature. This is what normally expected, since the Curie temperature of bulk LSMO is around 360 K which is above the maximum measurement temperature of 300 K. This normal behavior is associated with thicker films. The second type of MR vs. temperature shows a peak between 100 ~ 200 K. This only appears in very thin films (15 Å film on NGO, 30 and 60 Å film on LAO). It is the signature MR behavior of the dead-layer. The peak temperature in MR
could be used as an empirical estimate \cite{11} to the Curie temperature, \( T_c \) of the dead-layer, which thus comes to be around 100 \( \sim \) 200 \( K \).

In conclusion, we have obtained an experimental estimate of the total thickness of the dead-layers associated with the surface and film-substrate interface in epitaxial films of LSMO on NGO and LAO substrates. The total dead-layer thickness in these two cases are estimated to be about 30 \( \text{Å} \) and 50 \( \text{Å} \), respectively. The dead-layer has a magnetoresistance peak temperature of around 100 to 200 \( K \), and is likely to have a magnetic Curie temperature in the same range.

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FIGURES

FIG. 1. (a) Temperature-dependent resistivity for films of different thicknesses. Resistivity increases for thinner films. (b) Thickness dependence of the total conductance of films at 14K. For films that are metallic, \( R(T) \) is flat enough in this temperature region to consider this value representative of the residual resistance. The thickness dependence of conductance is linear, but with a finite intercept on the horizontal axis, suggesting the existence of an electrically less conducting dead-layer. From the intercept the total thickness of the dead layers can be estimated. For films on NGO, it is about 30 Å; on LAO, about 50 Å.

FIG. 2. Lattice parameters’ thickness dependence as measured using a 4-circle X-ray diffractometer. Error bars reflect the diffraction peak-width. A small variation of film lattice constant is seen to occur when the film reaches a thickness of around 60 to 120 Å. The small increase of out-of-plane lattice constant vs. thickness for films on NGO is not well understood. The presence of large substrate peaks nearby may account for some variation in diffraction peak position (and therefore lattice constant) for the very thin films of 15 Å ∼ 30 Å.

FIG. 3. AFM images of two 30 Å films, one grown on NGO (a), the other on LAO (b). Both films have similar peak-to-peak surface roughness, estimated to be less than 5 Å, excluding large particles believed to be laser-ablation-related particulates. This suggests continuous film coverage.

FIG. 4. Thickness dependent evolution of magnetoresistance for films on NGO (a) and on LAO (b). Magnetoresistance are evaluated with a field sweeping of ±3 kOe. A low-temperature peak in magnetoresistance is seen for the very thin films. The threshold for this behavior is < 60 Å for films on LAO and ∼ 15 Å for films on NGO, consistent with estimates of the dead layer thicknesses of data from Fig.1(b).
Figure 1: (a) Temperature dependence of resistivity $\rho$ for different thicknesses of NGO and LAO bilayers. (b) Electrical conductivity $G$ as a function of thickness for NGO and LAO bilayers.

J. Z. Sun et al., Fig.1
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(a) NGO

(b) LAO

\[ \frac{[R(0) - R(H)]}{R(H)} \]

\[ T \text{ (K)} \]

J. Z. Sun et al., Fig. 4
This figure "Figfile3.jpg" is available in "jpg" format from:

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