Magnetotransport in manganite trilayer junctions grown by 90° off-axis sputtering

J. S. Noh, T. K. Nath, and C. B. Eom[a)]
Department of Materials Science and Engineering, University of Wisconsin—Madison, Madison, Wisconsin 53706

J. Z. Sun
IBM T. J. Watson Research Center, P.O. Box 218, Yorktown Heights, New York 10598

W. Tian and X. Q. Pan
Department of Materials Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109

(Received 1 September 2000; accepted for publication 14 May 2001)

We report magnetotransport studies on $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.67}Sr_{0.33}MnO_3$ trilayer junctions, fabricated using 90° off-axis sputtering. Films were grown on both (001) $(LaAlO_3)_{0.3}$–$(Sr_2AlTaO_6)_{0.7}$ and (110) $NdGaO_3$ substrates. The sputtered trilayers show improved junction resistance uniformity over those made using pulsed laser deposition. Cross-sectional transmission electron microscopy and atomic force microscopy studies confirm smooth interfaces and a uniform barrier. Magnetoresistances up to ~100% are observed for junctions on (001) $(LaAlO_3)_{0.3}$–$(Sr_2AlTaO_6)_{0.7}$ with a 30 Å barrier at 13 K and around 100 Oe. Junction magnetoresistance versus magnetic field behavior is more stable, indicating improved transport and magnetic homogeneity across the junction.
[DOI: 10.1063/1.1383276]

Trilayer tunnel junctions based on the doped manganites[1] exhibit large low-field magnetoresistances (MRs) up to a factor of 10 in change in resistance, and are therefore good model systems for studies of spin-polarized transport across interfaces.[2–5] One factor that made these manganite junctions unsuitable for applications is the premature disappearance of their large magnetoresistance at temperatures well below the Curie temperature of the electrodes.[2,3,5] It is important to refine the materials preparation process for these junctions so as to establish a solid understanding as to whether this limitation is related to interface materials properties or whether it is intrinsic to the doped manganites at the junction interface.

One possible cause for the degraded junction MR at elevated temperatures is the presence of structural disorder at the barrier interface. The interfaces of ferromagnetic layers adjacent to the barrier layer are structurally more disordered, which could induce electronic and magnetic disorders that reduce the degree of spin polarization at the interfaces. Indeed, a reduction of surface spin polarization was reported by Park *et al.*[6] Its origin and relationship to the surface materials property, however, remain unexplored. Other imperfections of the junction interface include lateral inhomogeneity, roughness, and possible interdiffusion of magnetic impurities into the barrier. These in turn will cause inhomogeneous current conduction, parasitic interface-roughness-mediated ferromagnetic coupling (the so-called orange-peel effect),[7] and spin-flip scattering at the junction interface. All such interfacial effects should be minimized.

In this letter, we report on results obtained from junctions fabricated from epitaxial $La_{0.67}Sr_{0.33}MnO_3$(LSMO)/$SrTiO_3$(STO)/$La_{0.67}Sr_{0.33}MnO_3$ trilayers grown by 90° off-axis sputtering. It is known that 90° off-axis sputtering can produce high quality epitaxial films without surface particulates that are usually associated with pulsed laser ablation.[8,9] The magnetotransport properties are investigated using junctions lithographically fabricated from the trilayers. With improved trilayer film quality, we obtained junctions with a more uniform magnetic switching, a better resistance to area scaling, and better high temperature MR.

Both (110) $NdGaO_3$(NGO) and (001) $(LaAlO_3)_{0.3}$–$(Sr_2AlTaO_6)_{0.7}$(LSAT) substrates were used for the growth of LSMO/STO/LSMO trilayer films. The bottom and top LSMO films were grown at a substrate temperature of 750 °C in an atmosphere of 120 mTorr Ar and 80 mTorr O_2, with a thickness of 600 and 400 Å, respectively. The STO barrier was grown in a mixture of 180 mTorr Ar and 20 mTorr O_2 at 720 °C. The barrier thickness was varied from 20 to 40 Å. A silver overlayer of 700 Å was deposited for film protection during photolithography. Optical lithography was used for junction definition. The process involves three mask levels, with pattern-transfer at each step done with ion-milling. Details of the process are described elsewhere.[10] For this experiment, junction sizes vary from 1 to 32 μm. The junctions are rectangular in shape, with aspect ratios of 1:1 or 1:2.

For current-perpendicular-to-plane transport measurements a magnetic field was applied parallel to the film surface. A close-cycle refrigerator together with a heater assembly gives a temperature range from 13 K to ambient. Prior to transport measurements, surface-damage-free junctions were sorted out through a visual inspection under an optical microscope. To obtain statistical information on junction behaviors, for each chip the magnetotransport measurements were

[a)]Electronic mail: eom@engr.wisc.edu

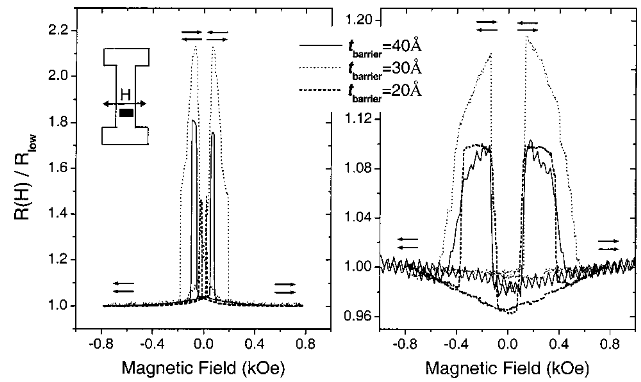

FIG. 1. Resistance vs magnetic field at 13 K. Left: Trilayer junctions on (001) LSAT substrate, right: Trilayer junctions on (110) NGO substrate. The inset shows the schematics of the geometry for the base stripe, the junction, and the direction of applied field.

done on at least 10 junctions that are located at different positions.

The resistance (R) versus magnetic field (H) data normalized to the junction resistance in its resistive-low state (R_{low}) at 13 K are shown in Fig. 1 for six groups of junctions with different combinations of substrate and barrier thickness. All junctions measured exhibit symmetric and well-defined $R(H)$ curves, reflecting a uniform magnetic domain rotation. Furthermore different junctions with the same geometry on each chip showed comparable junction resistance and MR values that represent good spatial uniformity. Here MR is defined as $(R_{\mathrm{high}}-R_{\mathrm{low}})/R_{\mathrm{low}}$, where R_{high} and R_{low} are the junction resistance in its resistive-high and -low states respectively. Junctions with a 30 Å barrier give the largest MR for both types of substrates. The lower switching fields for 40 Å and 30 Å barrier junctions seem to be similar, although the upper switching field of a 30 Å barrier junction is larger than that of a 40 Å barrier junction. On the other hand, both lower and upper switching fields for junctions on the 20 Å barrier chips are appreciably reduced compared to junctions with the other sets of barrier thickness. Here the lower and upper switching fields correspond to a parallel to antiparallel, and an antiparallel to parallel transition of the junction magnetic states, respectively. The largest MR obtained from this work is about 100% for a $2\times4\ \mu\mathrm{m}^2$ sized junction on a (001) LSAT substrate. Spin polarization and the corresponding density of states ratio of up-spin to down-spin in $La_{0.67}Sr_{0.33}MnO_3$ are calculated to be 0.58 and 3.8:1, respectively, for the MR value, when evaluated using the Julliere model.[11] One interesting point is for junctions of the same barrier thickness, those on the LSAT substrate in general, exhibit larger MR and lower switching fields than those on NGO substrate. This suggests the involvement of a substrate in determining the junction's micromagnetic state.

An atomic force microscopy (AFM) study was carried out on separate samples grown under identical conditions to examine layer-by-layer surface structures. LSMO single layer, STO/LSMO bilayer, and LSMO/STO/LSMO trilayer on both LSAT and NGO all showed smooth surfaces with a rms roughness between 5 and 10 Å but no significant difference in surface roughness with the substrate at each layer. There were no surface particulates observed that have been reported to appear on the same LSMO layer grown by laser ablation,[8] reflecting improved micron-scale homogeneity in surface structure. The main difference of the two types of substrates came from the barrier layer surface morphology in STO/LSMO bilayers, which follows that of the bottom LSMO. The barrier surface on LSAT is streaky while its counterpart on NGO is granular, although miscut angles were measured to be very small (0.03°–0.11°) and almost the same for both substrates. But it turned out to be not directly related to the magnetic anisotropy axes, as determined from angle-dependent magnetization measurements.

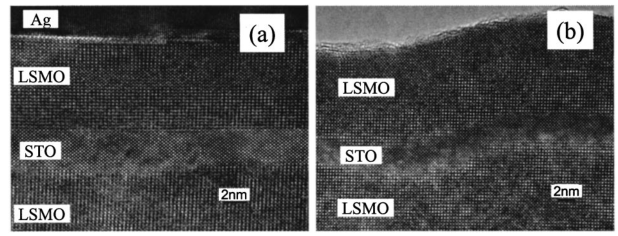

FIG. 2. Cross-sectional TEM images for trilayer films with a nominal barrier thickness of 30 Å: (a) trilayer on (001) LSAT, (b) trilayer on (110) NGO.

A cross-sectional transmission electron microscopy (TEM) study was also conducted to illuminate atomic scale interface structure. Figure 2 shows the high resolution TEM images of trilayers on both LSAT and NGO. High-quality barrier and interfaces of the film on (001) LSAT are seen in Fig. 2(a). No pin-hole, impurity, or sizable interdiffusion layer are observed at both interfaces of the barrier or inside the barrier. On the other hand, the surface of the bottom LSMO layer grown on NGO is rougher than that of its counterpart on LSAT, leading to rougher barrier interfaces. This, together with the reduced device transport performance in Fig. 1, suggests that interface uniformity, plays an important role in the performance of a tunnel junction.

One useful way of checking the transport uniformity in a junction is junction resistance scaling over different junction sizes. Figure 3(a) is the area scaling of our junctions fabricated from sputtered films together with the previous result of laser ablated trilayers. The resistance–area (RA) products for the sputtered junctions stay constant at both ambient and low temperature except for junctions with 40 Å barrier on NGO, which also showed the worst MR. In contrast, the LSMO trilayers made with pulsed laser ablation show a couple of orders of magnitude fluctuation in the RA products. The area scaling can be an indicator of a junction's

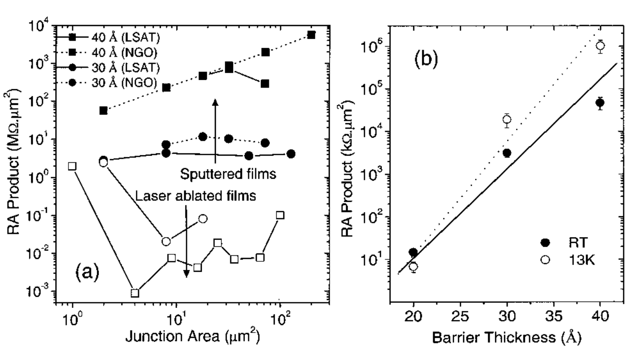

FIG. 3. Junction resistance scaling: (a) RA product vs junction area at low temperatures for junctions defined from both sputtered films (solid symbol) and laser ablated films (Ref. 18) (open symbol), (b) RA product vs barrier thickness both at room temperature and at 13 K.

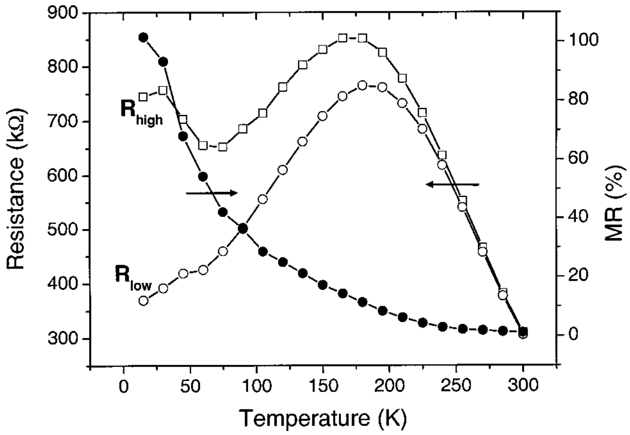

FIG. 4. Temperature dependence of R_{high}, R_{low}, and MR for a $2\times4\ \mu m^2$ sized junction on LSAT with 30 Å barrier. They were extracted from a series of $R(H)$ loops taken at 15 K temperature intervals.

interface uniformity because junction resistance should increase linearly with decreasing junction area, if the junction has a good interface and barrier uniformity.[12] There have been, however, few scaling studies reported and almost no satisfactory results on manganite tunnel junctions, mainly due to the complicated structural, magnetic, and compositional inhomogeneities at interface. From that point, the improved area scaling of our sputtered junctions over the laser ablated ones implies that our trilayer films have more uniform interfaces. This theory was also supported by the particle-free and smooth AFM images. The barrier-thickness scaling in Fig. 3(b) displays an exponential dependence of the junction resistance on barrier thickness, suggesting that a tunneling process is at play across the barrier.[13,14] In addition, it demonstrates that our sputtered trilayers have a statistically reliable uniformity as the RA products were calculated over all junctions tested irrespective of its geometry and location.

An improved temperature dependence of MR is observed for such junctions on LSAT. The temperature-dependent MR for the junction on LSAT that gave the largest MR at low temperatures in our batch is shown in Fig. 4. MR disappears at room temperature, but several percent of MR is still visible above 200 K. The highest temperature, to which we could observe distinguishable MR spikes in $R(H)$ curves, is around 270 K for this junction. This result is better than previously reported for manganite-based tunnel junctions.[2,3,5,15] Obata *et al.* have reported MR to be observed at similar temperatures for $La_{0.8}Sr_{0.2}MnO_3$-based tunnel junctions.[16] However their result seems somewhat controversial because such a high temperature MR appeared only for a junction with a thin barrier (16 Å). Accordingly the junction resistance was measured to be 200–300 Ω at 5 K and in the low resistance range spurious MR effects generated by nonuniform current distribution over the junction area might be involved, as Moodera *et al.* pointed out.[17] Our high temperature MR behavior suggests that the premature disappearance of MR is at least improvable to some degree upon improved interface materials quality.

The underlying causes of the substrate dependence of MR (Fig. 1) remain unresolved. It is difficult to explain the substrate dependence by a lattice-mismatch-induced strain, since the lattice parameters of both substrates are very close [$(a^2+b^2)^{1/2}/2=3.862$ Å for orthorhombic NGO and $a=3.868$ Å for pseudocubic LSAT, hence, an in-plane lattice parameter difference of only 0.16%]. Different surface morphology of the film on each substrate did not appear to directly produce distinct magnetic anisotropy behavior. However the influences of the optimal LSMO growth condition on each substrate, different surface energy and crystal symmetry of the substrates on microstructures and micromagnetic states of the film, are still yet to be investigated for a solid understanding of the interesting founding.

In conclusion, we have grown epitaxial $La_{0.67}Sr_{0.33}MnO_3/SrTiO_3/La_{0.67}Sr_{0.33}MnO_3$ trilayer films on both (110) NGO and (001) LSAT substrates by 90° off-axis sputtering. Junctions fabricated from these films showed distinct MR behaviors for most chips, with a maximum MR of around 100% at 13 K. We found that RA products of the junctions increase exponentially with increasing barrier thickness and remain almost constant over the measured junction area, reflecting good area scaling and hence good junction uniformity. A TEM study confirmed that our sputtered films have a smooth and pin-hole-free barrier. These overall improvements in materials quality indeed result in an improvement in the high temperature junction MR.

This work was partly supported under NSF Grant No. DMR-9802444, NSF Young Investigator Award (C.B.E.), and by IBM. The authors would like to thank Roger Koch, Bill Gallagher, Steve Brown, and John Connolly at IBM Research Yorktown Heights for assistance at various stages of the experiment.

[1] Doped manganites in this case refer to $La_{1-x}(Ca, Sr)_xMnO_3$, the so-called colossal magnetoresistance materials, such as described by S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).
[2] J. Z. Sun, L. Krusin-Elbaum, P. R. Duncombe, A. Gupta, and R. B. Laibowitz, Appl. Phys. Lett. **70**, 1769 (1997).
[3] M. Viret, M. Drouet, J. P. Contour, C. Fermon, and A. Fert, Europhys. Lett. **39**, 545 (1997).
[4] J. O'Donnell, A. E. Andrus, S. Oh, E. V. Colla, and J. N. Eckstein, Appl. Phys. Lett. **76**, 1914 (2000).
[5] M.-H. Jo, N. D. Mathur, N. K. Todd, and M. G. Blamire, Phys. Rev. B **61**, R14905 (2000).
[6] J.-H. Park, E. Vescovo, H.-J. Kim, C. Kwon, R. Ramesh, and T. Venkatesan, Phys. Rev. Lett. **81**, 1953 (1998).
[7] L. Néel, C. R. Acad. Sci. **255**, 1676 (1962).
[8] J. Z. Sun, D. W. Abraham, K. Roche, and S. S. P. Parkin, Appl. Phys. Lett. **73**, 1008 (1998).
[9] A. Gupta and J. Z. Sun, J. Magn. Magn. Mater. **200**, 24 (1999).
[10] J. Z. Sun, W. J. Gallagher, P. R. Duncombe, L. Krusin-Elbaum, R. A. Altman, A. Gupta, Yu Lu, G. Q. Gong, and G. Xiao, Appl. Phys. Lett. **69**, 3266 (1996).
[11] M. Julliere, Phys. Lett. **54A**, 225 (1975).
[12] W. J. Gallagher, S. S. P. Parkin, Yu Lu, X. P. Bian, A. Marley, K. P. Roche, R. A. Altman, S. A. Rishton, C. Jahnes, T. M. Shaw, and G. Xiao, J. Appl. Phys. **81**, 3741 (1997).
[13] W. F. Brinkman, R. C. Dynes, and J. M. Rowell, J. Appl. Phys. **41**, 1915 (1970).
[14] K. Knorr and J. D. Leslic, Solid State Commun. **12**, 615 (1973).
[15] X. W. Li, Yu Lu, G. Q. Gong, G. Xiao, A. Gupta, P. Lecoeur, J. Z. Sun, Y. Y. Wang, and V. P. Dravid, J. Appl. Phys. **81**, 5509 (1997).
[16] T. Obata, T. Manako, Y. Shimakawa, and Y. Kubo, Appl. Phys. Lett. **74**, 290 (1999).
[17] J. S. Moodera, L. R. Kinder, J. Nowak, P. LeClair, and R. Meservey, Appl. Phys. Lett. **69**, 708 (1996).
[18] J. Z. Sun, Philos Trans. R. Soc. London **356**, 1693 (1998).