Room-temperature tunneling magnetoresistance in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ step-edge junctions

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$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ tunneling magnetoresistance (TMR) junctions have been fabricated on step-edge (001) SrTiO_3 substrates with a high step-edge angle. The step-edge junctions show nonvanishing TMR spikes in $R(H)$ curves above room temperature (RT). The resistance, dynamic conductance, and TMR ratio are characterized to explore the possible conduction mechanism for the step-edge junctions. The temperature dependence of surface magnetization $M_s(T)$ deduced from the spin polarization $P(T)$, following a $(T_c - T)^{0.92 \pm 0.02}$ dependence, agrees with the theoretical models for $M_s(T)$. From these results, we conclude that tunneling is the dominating mechanism and that the charge carriers at the surface boundary govern the tunneling conductivity. © 2004 American Institute of Physics. [DOI: 10.1063/1.1687994]

I. INTRODUCTION

The manganite-based spin-dependent tunneling devices have attracted much attention in recent years due to their potential applications in magnetic sensors¹ and digital storage technologies.² The manganites of the type $\text{La}_{0.7}\text{A}_{0.3}\text{MnO}_3$ ($\text{A} = \text{Ca}, \text{Sr}, \text{Ba}, \text{and Pb}$) are half-metallic and are therefore ideal candidates for the design of spin-electronics devices. To date, several groups have made tunneling magnetoresistance (TMR) devices with various structures, such as ramp-edge junctions,³ bicrystal grain boundaries,⁴ polycrystalline films,⁵ step-edge junctions,⁶ and trilayer sandwich junctions.⁷ Enhanced low-field MR effects at low temperatures have been reported in these TMR devices. For bicrystal grain boundary junctions, it has been shown that the TMR value can be up to 300% at 4.2 K.⁸ More recently, a TMR ratio of more than 1800% at 4 K for $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3/\text{SrTiO}_3/\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ trilayer junctions was obtained,⁹ leading to an electrode spin polarization of at least 95% inferred. However the reported low-field MR for the manganite-based devices decreases rapidly with an increase of temperature, and even vanishes at temperatures well below the Curie temperature of bulk manganites. In the last few years, a considerable number of studies^{10–12} have been made on the origin of the rapid decrease of the TMR effect with increasing temperature. For the manganite tunneling junctions, the elastic spin-flip scattering of the spin-polarized carriers by the local magnetic impurities within the insulating barrier,¹⁰ the inelastic spin-flip scattering at the interfaces between the insulating barrier and the magnetic electrodes,¹¹ and the stronger temperature dependence of spin polarization at interface,¹² have been regarded as the

basic ingredients proposed to describe this behavior. It is generally believed that tunneling is a mechanism occurring at the surface boundary, and the TMR is dominated by charge carriers at the surface boundary.^{13,14} Yin *et al.*¹⁵ have showed a near RT TMR in their trilayer device with a good magnetic–nonmagnetic interface and a few defects in it. Recently, a similar result, that the TMR extends up to temperatures of more than 270 K for $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3/\text{SrTiO}_3/\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ trilayer junctions, has been obtained by Bowen *et al.*⁹ However, for the manganite-based devices, surprisingly few studies¹⁶ have so far reported an above RT low-field TMR, even though $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) bulk samples exhibit a ferromagnetic Curie temperature of 364 K.¹⁷

In this article, we introduce our step-edge devices, which show a nonvanishing TMR above RT. We measure the MR ratio and the dynamic conductance at various temperatures to study the conduction mechanism of polarized electrons across grain boundaries.

II. EXPERIMENTS

Epitaxial LSMO thin films were prepared by an off-axis rf sputtering configuration. We use a standard photolithographic process with ion-beam etching to fabricate LSMO step-edge junctions on SrTiO_3 substrate. Step-edge arrays with seven steps in series, heights of 170–290 nm, and a 15- μm -wide meander track bridge geometry were produced. An additional gold layer was deposited on the LSMO bridges, but the 10- μm -long sections patterned across the artificial step edge. The gold layer was used to eliminate the signal contribution from the epitaxial LSMO film and reduce the contact resistance. Figure 1(a) shows a photo of our step-edge arrays with a step-edge line along SrTiO_3 [100] crystal direction. The MR ratios were measured by using a dc four-

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FIG. 1. (a) A photo of our step-edge arrays. (b) The AFM micrograph of LSMO film on the step-edge ramp surface.

terminal method with a magnetic field applied to the film plane and perpendicular to the step-edge line. Figure 1(b) shows the atomic force microscopy (AFM) micrograph of LSMO film on the step-edge ramp surface. A sharp step-edge angle of 70° , as shown in Fig. 1(b), forms the tunneling barrier in our junctions. It is notable that the transport properties of these junctions are strongly influenced by the step-edge angle and the ratio of film thickness d to step-edge height H . The details will be described elsewhere. We present herein the TMR data on the samples of S1, S2, and S3, in which the ratios of film thickness to step-edge height (d/H) are 150/290, 150/170, and 100 nm/170 nm, respectively.

III. RESULTS AND DISCUSSION

Figure 2 shows the temperature dependence of junction resistance R_j for the three samples in comparison to an epitaxial film. Since the resistance of the epitaxial film is much smaller than the junction resistance, R_j is believed to be dominated by the step-edge barrier. It is also found that the $R_j(T)$ curves are similar and comparable to those reported on the ramp-edge junctions³ or the trilayer sandwich junctions,^{7,18} respectively. The inset of Fig. 2 shows the resistance area product $R_j A$ for S1, S2, and S3 samples, in which the ratios d/H are 0.52, 0.88, and 0.59, respectively. It can be seen that the $R_j A$ values are in the range of 10^{-1} – $1 \mu\Omega \text{ m}^2$ at temperatures of 5 and 300 K, and decrease with an increase of d/H . The $R_j A$ values for our

FIG. 2. The temperature dependence of junction resistance R_j for samples. The inset shows the resistance area product $R_j A$. The dashed line is for the purpose of guiding the eye.

samples are smaller than those for the bicrystal grain junctions,¹⁹ and are on the same order of magnitude as the sputtered trilayer junctions reported by Noh *et al.*¹⁸ Typical $R_j A$ values of about 10^{-3} – $10^{-1} \mu\Omega \text{ m}^2$ for tunnel junctions have been proposed.²⁰ Additionally, the barrier height V_{bi} can be achieved from the data of $R_j A$ by according to the calculations of Klein *et al.*¹⁹ Using the Wentzel–Kramers–Brillouin approximation for the resistance of a rectangular barrier of height V_{bi} and thickness t , Klein *et al.* estimated the resistance area product $R_j A$ by

$$R_j A = [8\pi^2 \hbar^2 t^2 \exp(2\kappa t) / e^2] (1 + 2\kappa t), \quad (1)$$

where $\kappa = (2mV_{bi}/\hbar^2)^{1/2}$ and $t = (2\epsilon_0\epsilon_r V_{bi}/e^2 N)^{1/2}$ with $N \sim 10^{21}/\text{cm}^3$, $\epsilon_r \sim 10$, and the effective mass $m = 4m_e$ for LSMO. Taking the $R_j A$ values of 0.2–0.5 $\mu\Omega \text{ m}^2$ at 5 K into Eq. (1), we obtain the barrier heights of 0.75–0.80 eV and $t \approx 0.9$ nm for our samples. The estimated values are very close to those deduced from our I – V measurements (see subsequent discussion). The obtained barrier heights for our step-edge TMR junctions are a little smaller than that of ~ 1 eV for the bicrystal grain junctions.¹⁹

Figure 3 shows the curves of MR ratio versus external magnetic field H for S1 sample at 5, 300, 310, and 330 K. Here, the MR ratio is calculated as $\text{MR} = [R(H) - R_p]/R_p$, where R_p is the resistance when both moments in the electrodes are parallel to each other. In many articles,^{3,7,15,16} R_p has been taken by the resistance for a magnetic field in the range of $H = 1$ to 1.5 kOe. We chose $R_p = R(H = 1.4 \text{ kOe})$ to make the MR ratios of our samples comparable to that previously reported. It is also believed that the tunnel resistance will reach saturation, and that MR is originated mainly from the contribution of epitaxial LSMO film for $H \geq 1.4$ kOe. In Figs. 3(b) and 3(c), the $R(H)$ curves of an epitaxial LSMO film are also shown for comparison. The highest temperature at which we could observe distinguishable MR spikes in

FIG. 3. MR ratio versus external field H for the S2 sample at 5, 300, 310, and 330 K. The $R(H)$ curves of epitaxial LSMO film are also shown for comparison.

$R(H)$ curves, is around 330 K for this sample, as seen in Fig. 3(d). This result is better than previously reported for manganite-based tunnel junctions.

Figure 4 shows the $R(H)$ curves for S2 and S3 samples at various temperatures. Typical butterfly MR curves are clearly observed for samples at temperatures below 200 K. As shown in Figs. 4(b) and 4(d), MR spikes in $R(H)$ curves of 300 K are distinguishable for S2 and S3 samples, respectively.

To further clarify the mechanism of MR, it is helpful to measure the I - V characteristics. Figure 5 shows the dynamic conductance $G = dI/dV$ normalized by the conductance G_0 at zero voltage as a function of the voltage drop

FIG. 4. $R(H)$ curves for S2 and S3 samples at various temperatures.

FIG. 5. Dynamic conductance $G = dI/dV$ normalized by the conductance G_0 at zero voltage as a function of the voltage drop across a series of step edges for the S1 sample. Insets (a) and (b) show the dynamic conductance exponent x and the nonlinear tunneling conductance g as determined by a fit of Eq. (2) to the measured curves, respectively. Inset (c) shows the calculated curve of $\Delta I = [I(V, 25 \text{ K}) - I(V, 5 \text{ K})] / I(V, 5 \text{ K})$ and two cusps at around $\pm 0.7 \text{ V}$ are indicated by the arrows.

across a series of step edges for the S1 sample. The nonlinear behavior of I - V characteristics gives support for the tunneling mechanism of conduction process at the boundaries. We analyzed the dynamic conductance with the general expression

$$G/G_0 = 1 + gV^x, \quad (2)$$

where G_0 denotes the low-voltage dynamic conductance, g is the nonlinear tunneling conductance, and the gV^x term corresponds to the inelastic tunneling process.²¹ Equation (2) yields satisfactory fits to the data, as shown by the solid lines in Fig. 5. The conductance exponent x is plotted in inset (a) of Fig. 5. It displays an increase with increasing temperatures, approaching the direct-tunneling value²² of $x=2$. At low temperatures, the value $x \approx 1.3$ is close to the theoretical value of $x=4/3$ for inelastic tunneling via pairs of localized states.²¹

Inset (b) of Fig. 5 shows the nonlinear conductance g as a function of temperature for the S1 sample. As can be seen, the value of g decreases gradually with an increase of temperature, implying that the inelastic tunneling in the tunneling process will diminish at higher temperatures for a low-bias voltage. We find that the temperature dependence of $g(T)$ can be approximately described by $g(T) = 1.1 - 0.003T$, as shown in inset (b) of Fig. 5. Recently, Rotländer et al.²³ have proposed a simple method to determine the barrier height deduced from the temperature dependence of I - V curves. This method is to take two I - V characteristics at different temperatures, one ideally at the absolute zero

$I(V,0)$ and the other at some temperature $I(V,T)$. The normalized temperature dependence $\Delta I \equiv [I(V,T) - I(V,0)]/I(V,0)$ should then have cusps at around the barrier height. Shown in inset (c) of Fig. 5 is the calculated curve of $\Delta I = [I(V,25\text{ K}) - I(V,5\text{ K})]/I(V,5\text{ K})$ for the S1 sample. In this curve, two cusps can be seen at voltages around $\pm 0.7\text{ V}$, indicating that the barrier height V_{bi} is around 0.7 eV for our S1 sample. This value is near that estimated by using the R_p/A data, as previously discussed.

According to the simple model proposed by Jullière,²⁴ the TMR ratio for our junctions just described should be given by

$$\text{TMR} = [R_{\max} - R_p]/R_p = 2P^2/(1 - P^2), \quad (3)$$

where P is the spin-polarization parameter for LSMO and $R_{\max}$ is the maximum resistance in the measurement of field sweep at a fixed temperature, corresponding to the junction resistance when the two ferromagnetic electrodes have antiparallel magnetizations. In our step-edge junctions, if we take $(\text{TMR})_{\max} \approx 30\%$, Eq. (3) leads to $P \approx 0.32$ for LSMO. Recently Höfener *et al.*²⁵ have extended the Jullière model by adding an inelastic, spin-dependent tunneling contribution to describe the voltage and temperature dependences of the TMR for bicrystal grain-boundary junctions. They have derived a modified Jullière formula as follows:

$$\text{TMR} = \frac{I_{\text{ap}}^e}{I_{\text{ap}}^e + I^i}(V, T) \times \text{TMR}_{\text{Jullière}}(T), \quad (4)$$

where I_{ap}^e is the elastic tunneling current for antiparallel magnetization arrangement, and I^i is the inelastic tunneling current. The I_{ap}^e and I^i terms can be obtained from the I - V characteristics of $I = I_{\text{ap}}^e + I^i = G_0 V + G_0 g V^{1+x}$, as described by Eq. (2). Taking $G_0 = G_0(0\text{ K})(1 - P^2)$ for Jullière model and $\text{TMR}_{\text{Jullière}} = 2P^2/(1 - P^2)$, as shown in Eq. (3), into Eq. (4), we have the modified TMR formula as follows:

$$\begin{aligned} \text{TMR} &= \frac{I_{\text{ap}}^e}{I_{\text{ap}}^e + I^i}(V, T) \times \text{TMR}_{\text{Jullière}}(T) \\ &= \frac{(1 - P^2)}{(1 - P^2) + gV^x} \frac{2P^2}{1 - P^2} \\ &= \frac{2P^2}{1 - P^2 + gV^x}. \end{aligned} \quad (5)$$

According to Eq. (5), we can see that the TMR will be dependent on the bias voltage. Figure 6 shows the voltage dependence of the TMR ratio for the S1 sample at 5 and 100 K, which is evolved from the I - V characteristics measured in magnetic field 1400 Oe and in the fields in which $R_{\max}$ occurs (~ 100 and 60 Oe for 5 and 100 K, respectively). Inset (a) of Fig. 6 shows the voltage dependence of normalized ratio $\text{TMR}/\text{TMR}(V=0)$ for $T=5$ and 100 K. We can see the TMR ratio is weakly dependent on the bias voltage for $T=100\text{ K}$. The weak voltage dependence of TMR for higher temperatures is due to the decrease of the g value as temperature is increased. Furthermore, we calculate the temperature dependence of I^i/I_{ap}^e by taking $I^i/I_{\text{ap}}^e = gV^x$, as shown in inset (b) of Fig. 6. It can be seen, for a low-bias voltage, the I^i/I_{ap}^e ratio decreases rapidly with an increase of temperature,

FIG. 6. Voltage dependence of the TMR ratio for the S1 sample at temperatures 5 and 100 K. Inset (a) the voltage dependence of normalized ratio $\text{TMR}/\text{TMR}(V=0)$, and inset (b) the temperature dependence of I^i/I_{ap}^e ratio for voltages of 0.1, 0.5, and 1 V.

indicating that the inelastic tunneling current in the tunneling process can be neglected at higher temperatures. Our result is somewhat different from that observed in the bicrystal grain boundary junctions,²⁵ in which a strong voltage dependence of high-field ($H = 1.5\text{ T}$) MR ratio was presented.

Figure 7 shows the TMR ratio, given by $\text{TMR} = [R_{\max} - R_p]/R_p$, as a function of temperature for the S1, S2, and S3 samples. Taking the measured $\text{TMR} = 30\%$, $g = 1.1$ for $T = 5\text{ K}$, and the bias voltage of around 1 V for the S1 sample

FIG. 7. The TMR ratio as a function of temperature for the S1, S2, and S3 samples. The inset shows the spin polarization P versus temperature for the S1, S2, and S3 samples, and the modified $P^*(T)$ for S1. The solid lines are the fitting curves of $P(T) \propto (T_c - T)^{0.92 \pm 0.02}$. The inset also shows the magnetization $M/M(5\text{ K})$ of LSMO film measured with a SQUID.

in our measurement, we recalculate the modified spin polarization P^* using Eq. (5). The obtained $P^*(5\text{ K})$ is around 0.41, which is slightly larger than that directly obtained by Jullière formula. The modified P^* value is still smaller than that of 0.54–0.95 reported for the trilayer sandwich junctions.^{7,9,18} However, the $(\text{TMR})_{\text{max}}$ value for our S1 sample is a little larger than that of the ramp-edge junctions³ or the $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ step-edge junctions.⁶ Thus, the decrease in spin polarization of our step-edge junctions is not due only to the nonlinear tunneling, but also to other processes, such as the trapping of depolarized electrons induced by the defects in the barrier.²⁶ For grain-boundary junctions in particular, it has been pointed out that the strain and distortions at the grain-boundary interface,²⁷ or the band bending effects²⁸ will dominate the transport mechanism and decrease the TMR value.

In addition, the reductions of TMR and spin polarization due to temperature are explored. By using Eq. (3), the temperature dependences of P estimated from the data of TMR (T) for the S2 and S3 samples are also shown in the inset of Fig. 7. The temperature dependence of P^* for the S1 sample can be estimated by taking account of the contribution of the inelastic tunneling current. As shown in the inset of Fig. 7, we find that the modified $P^*(T)$ for the S1 sample is close to that deduced from the Jullière model for higher temperatures, implying that the nonlinear tunneling is negligible.

Some deviation between P and P^* is caused by the larger contribution of inelastic tunneling at low temperatures. Since there is small differentiation between P and P^* , we analyze the temperature dependence of spin polarization, only taking account of P derived from the Jullière model. The spin polarization P versus temperature shown in the inset of Fig. 7 can be fitted by the curves of $P(T) \propto (T_C - T)^{0.92 \pm 0.02}$ with $T_C = 361, 357$, and 322 K for S1, S2, and S3 samples, respectively. Since tunneling dominates the conduction process in our junctions, and should reflect the local electronic structure near the interface, it is believed that $P(T)$ will reflect the average magnetization near the grain-boundary surface. Therefore, we can assume that the spin polarization $P(T)$ is proportional to the surface magnetization $M_S(T)$.²⁹ As a result, we obtain $M_S(T) \propto P(T) \propto (T_C - T)^{0.92 \pm 0.02}$.

Earlier theoretical studies using the mean field approximation³⁰ and numerical Monte Carlo simulations³¹ according to the semi-infinite Heisenberg model predicted that $M_S(T)$ could be characterized by $\beta = 1$ and $\beta \approx 0.8$ in $(T_C - T)^\beta$ near T_C , while $\beta = 1/2$ and $\beta \approx 1/3$ were predicted for the bulk magnetization, respectively. Also shown in the inset of Fig. 7 is the temperature dependence of magnetization $M/M(5\text{ K})$ in a field of 1000 Oe for LSMO film measured with a superconducting quantum interference device (SQUID). We find that as $T \rightarrow T_C$, from below, the magnetization becomes proportional to $(1 - T/T_C)^{0.35}$ for our LSMO film. This result agrees well with the prediction of numerical Monte Carlo simulations for bulk magnetization. Moreover, the temperature dependence of surface magnetization $M_S(T)$ deduced from $P(T)$, following a $(T_C - T)^{0.92 \pm 0.02}$ dependence, also agrees with the theoretical models for $M_S(T)$. Our data prove clearly that the charge carriers at the surface

(grain/interface) boundary govern the tunneling conductivity for our step-edge junctions, and give the suggestion that the rapid decrease of TMR with increasing temperatures is due to the stronger temperature dependence of surface magnetization at interfaces than that of the bulk. These results are coincident with many arguments presented by other workers.^{12,14,32}

IV. SUMMARY

In summary, we have fabricated LSMO step-edge junctions that show nonvanishing MR spikes in $R(H)$ curves above RT. According to the I – V characteristics, the possible mechanism for our step-edge junctions is tunneling. Additionally, the surface magnetization $M_S(T)$ deduced from $P(T)$, following a $(T_C - T)^{0.92 \pm 0.02}$ dependence, exhibits a great difference from that for the bulk-like LSMO film, and agrees with the theoretical models for $M_S(T)$. These results lead to the conclusion that the charge carriers at the grain-boundary interface govern the tunneling conductivity for our step-edge junctions, and suggest that the decrease of TMR with increasing temperatures is due to the stronger temperature dependence of surface magnetization at interfaces.

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