Effects of step-edge conditions on the magnetoresistance of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ tunneling junctions

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Abstract

$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) tunneling magnetoresistance (TMR) junctions have been successfully fabricated on step-edge (001) SrTiO_3 substrates with various step-edge conditions. We report the fabrication techniques of LSMO step-edge TMR junctions in which the standard photolithography and ion-beam etching techniques are used to control the step angle and step height. We study the influence of step-edge conditions on the resistance and MR behavior of LSMO TMR junctions. It is found that the magnitude of junction resistance increases with an increase of step angle or with a decrease of d/H ratio, where d is the film thickness and H the step height. Based on our results, we conclude that a suitable d/H ratio of ~ 0.5 – 0.9 with a high step angle of $\phi \geq 70^\circ$ can be regarded as the optimum conditions for the fabrication of step-edge TMR junctions.

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1. Introduction

At present, various types of tunneling magnetoresistance (TMR) devices have been developed, of which ramp-edge junctions [1], bi-crystal grain boundaries [2], step-edge junctions [3], and trilayer sandwich junctions [4,5] are the most widely used. Of these, bi-crystal junctions are easy to fabricate, but high cost of the substrate and restricted topological freedom limit their applications for multiple-junction devices. However, ramp-edge junctions and trilayer sandwich junctions have topological freedom; however, a fabrication technology for reliable high quality junctions has yet to be developed. Step-edge junctions are low cost, easily fabricated, and reproducible. In addition, they also have topological freedom and are believed to be a good candidate for the TMR junctions. Within the development of high- T_c superconducting Josephson junctions, the effects of step-edge conditions on the transport

properties have been widely studied [6–10]. Detailed microstructural investigations [9,10] of such $\text{YBa}_2\text{Cu}_3\text{O}_y$ films grown on steep steps in cubic single-crystal substrates have shown that nearly 90° tilt grain boundaries, acting as weak links, occur in $\text{YBa}_2\text{Cu}_3\text{O}_y$. Therefore, the magnetotransport properties of step-edge junctions are expected to behave similar as those of other grain boundary junctions. Recently, Ziese et al. [3] have presented the large low-field magnetoresistance (MR) of step-edge structures in 25 nm thick $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ films. However, it has been shown that the electrical and magnetic properties should be strongly dependent on the strain effects, especially for the films with thickness $d \leq 40$ nm [11–13]. Until now, the influence of step-edge conditions and the factors of impact on the junction properties have not been investigated in detail.

In this article, we report the fabrication of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) step-edge TMR junctions using standard photolithography and ion-beam etching techniques. We study the influence of step-edge conditions on the resistance and MR behavior of LSMO TMR junctions. It is found that a large MR ratio should be

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Fig. 1. (a) Schematic diagram of the Ar^+ ion-milling. (b) Schematic diagram of our step-edge arrays. The inset shows its corresponding three-dimensional schematic diagram.

observed on the LSMO step-edge junctions with a higher step angle, $\phi \geq 70^\circ$, as well as a suitable ratio of film thickness to step-edge height, d/H .

2. Experimental procedure

In this work, SrTiO_3 (001) substrates ($0.5 \times 5 \times 5 \text{ mm}^3$) polished on one side were used. A $1.5 \mu\text{m}$ thick photoresist (AZ1500) was used as the etching mask to define the step edge and baked at 70°C before it was patterned. The exposure conditions were tuned to achieve sharp and straight edges for the photoresist mask. The step edges were fabricated using an ion mill with a 500 V, 20 mA argon ion beam. Fig. 1a shows the schematic diagram of the Ar^+ ion-milling. The angle α is defined by the normal of sample stage plane, the z axis, with respect to the direction of the Ar^+ beam. θ is defined by the edge line of photoresist mask with respect to the horizontal x axis in the sample-stage plane. The substrates were fabricated with different orientations. We can obtain sharp and straight edges with a high step angle of $\phi \geq 70^\circ$ for $\alpha \approx 65^\circ$ and $\theta = 0^\circ$. Furthermore, the step angle can be controlled by using different θ angles. We have found that as the θ angle is increased up to 45° , a decreased step angle of

$\phi \approx 10^\circ$ will be obtained. In addition, step height can be controlled by etching time.

Using off-axis r.f.-sputtering deposition, the LSMO films were prepared onto SrTiO_3 substrates in which step edges with various step angles were fabricated. Step-edge arrays with 7 steps in series, heights of 170 or 290 nm and a $15 \mu\text{m}$ wide meander track bridge geometry were produced. An additional gold layer was deposited on the LSMO bridges, but with the $10 \mu\text{m}$ 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. Fig. 1b shows a schematic diagram of our step-edge arrays. The electrical properties were measured by using a d.c. four-terminal method with a magnetic field applied in the film plane and perpendicular to the step-edge line. The inset of Fig. 1b shows its corresponding three-dimensional schematic diagram. The surface morphology and the shape of the step edges were explored by the atomic force microscopy (AFM).

3. Results and discussion

Fig. 2a, b and c show the AFM micrographs of the step-edge ramp surface onto SrTiO_3 substrates for step

Table 1

Step-edge conditions of samples studied in this work and the junction resistance R_j , the resistance area product $R_j A$ and the barrier height V_{bi} for samples at 5 K

samples	ϕ ($^\circ$)	d (nm)	H (nm)	d/H	R_j (5 K) (Ω)	$R_j A$ (5 K) $\mu\Omega \text{ cm}^2$	V_{bi} (5 K) (eV)
S1	70	150	290	0.52	175 192	3942	0.78
S2	50	150	290	0.52	510 06	1148	0.73
S3	26	150	290	0.52	53.79	1.21	0.44
S4	10	150	290	0.52	13.38	0.301	0.39
S5	70	50	170	0.29	245 526 2	18 417	0.85
S6	70	100	170	0.59	369 845	5548	0.80
S7	70	150	170	0.88	809 09	1820	0.75
S8	70	310	170	1.82	28.8	1.34	0.45

Fig. 2. AFM micrographs of the step-edge ramp surface onto SrTiO_3 substrates for step angle $\phi =$ (a) 26° , (b) 50° and (c) 70° . (d) The AFM micrograph of LSMO film on the step-edge ramp surface.

angle $\phi \approx 26^\circ$, 50° and 70° , respectively. All the AFM micrographs show a straight-line shape and a smooth ramp-edge surface with a step-edge height of ~ 290 nm. Fig. 2d shows the AFM micrograph of LSMO film on the step-edge ramp surface. A sharp step-edge angle of 70° as shown in Fig. 2d forms the tunneling barrier in this junction. Somewhat irregular ramp surface is caused by the deposition of LSMO on the miss-orientated step edge. Table 1 illustrates the step-edge conditions of

samples S1–S8 studied in this work. As illustrated in Table 1, the step angles are 70° , 50° , 26° and 10° for samples S1, S2, S3, and S4, respectively, in which the film thickness of $d = 150$ nm and the step-edge height of $H = 290$ nm are fixed. However, with a constant step angle of $\phi = 70^\circ$ and step-edge height of $H = 170$ nm, the film thickness is changed by $d = 50$, 100, 150 and 310 nm for samples S5, S6, S7 and S8, respectively. Some of the transport properties are also shown in Table 1 for discussion below. Fig. 3a shows the temperature dependence of junction resistance R_j in magnetic fields of 0 and 7 T for S1, S2, S3 and S4 samples in comparison to an epitaxial film. We can see that the magnitude of junction resistance increases with an increase of step angle. Moreover, the positive temperature coefficient of the resistance at low temperatures observed in S3 and S4 implies that the possible mechanism for the low-step-angle junctions is due to the grain-boundary scattering, while the non-positive temperature coefficient observed in S1 and S2 implies that the possible mechanism for the high-step-angle junctions is due to tunneling [14,15]. The positive temperature coefficient of the resistivity observed in the polycrystalline $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ films has been suggested to result from a non-tunneling mechanism [14]. In Fig. 3b, we show the temperature dependence of junction resistance for S5, S6, S7 and S8 samples. Clearly the magnitude of junction resistance increases drastically with a decrease of film thickness, and non-positive temperature coefficients at low temperatures can be observed in S5, S6 and S7 samples. In addition, a significant magnitude of high-field MR at low temperatures can be observed in samples with a higher step angle or a thinner film thickness, as seen in Fig. 3a,b. In Table 1 we also illustrate the junction resistance R_j , the resistance area product $R_j A$ and the barrier height V_{bi} for samples at 5 K, where A is the junction area taken by the product of film thickness $d \times$ bridge width of $15 \mu\text{m}$. It can be seen that R_j and $R_j A$ increase intensively with an increase of step angle or a decrease of film thickness. Furthermore the barrier height V_{bi} can be achieved from the data of $R_j A$ by according to the calculations of Klein et al. [16]. Using the Wentzel-Kramer-Brillouin (WKB) 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 $t = (2\varepsilon_0\varepsilon_r V_{bi}/e^2 N)^{1/2}$ with $N \sim 10^{21}/\text{cm}^3$, $\varepsilon_r \sim 10$, and the effective mass $m = 4m_e$ for LSMO. Taking the $R_j A$ values of 10^{-1} – $10^4 \mu\Omega \text{ cm}^2$ at 5 K into Eq. (1), we obtain the barrier heights of 0.39–0.85 eV as shown in Table 1. The estimated barrier heights for our step-edge TMR junc-

Fig. 3. (a) Temperature dependence of junction resistance R_j in magnetic fields of 0 and 7 T for S1, S2, S3, and S4 samples in comparison to an epitaxial film. (b) Temperature dependence of junction resistance for S5, S6, S7 and S8 samples.

tions are a little smaller than that of ~ 1 eV for the bicrystal grain junctions [16].

Fig. 4a, b, c and d show the curves of MR ratio vs. external magnetic field H for S1, S2, S3 and S4 samples at 100 K, respectively. Here the MR ratio is calculated as $MR = [R(H) - R(0)]/R(0)$. Typical butterfly MR curves are clearly observed for S1, S2 and S3 samples, and diminish for the S4 sample. The maximum MR ratio $MR_{\max}$, corresponding to the junction resistance

when the two ferromagnetic electrodes have antiparallel magnetizations, decreases as the step angle is decreased. The decrease of $MR_{\max}$ for samples with low step angle is surmised to be due to the continuous crystal structure on the grain boundary, which suppresses the tunneling and reduces the barrier height. Moreover the continuity of crystal structure on the grain boundary is controllable by varying the d/H ratio. Fig. 5a, b, c and d, respectively, show the curves of MR ratio vs. external magnetic

Fig. 4. The curves of MR ratio vs. external magnetic field H for (a) S1, (b) S2, (c) S3 and (d) S4 samples at 100 K.

Fig. 5. The curves of MR ratio vs. external magnetic field H for (a) S5, (b) S6, (c) S7 and (d) S8 samples at 100 K.

field H at 100 K for S5, S6, S7 and S8 samples. Among these samples the step angle is fixed at 70° and the d/H ratio is varied from 0.29 to 1.82. As seen in Fig. 5b,c, butterfly MR curves are clearly observed for S6 and S7 samples. However, distinguishable MR spikes in the MR(H) curve are not observed for the S8 samples, as seen in Fig. 5d. The diminishing of the MR spike observed in S8 is due to the continuity of the step-edge crystal structure, resulting in the suppression of tunneling. Otherwise, it is significant that distinguishable MR spike also diminishes in S5, which has a low d/H ratio. This can be attributed to the discontinuity and disorder on the grain boundary, which raises the grain-boundary scattering and reduces the tunneling, when the film thickness is much smaller than the step-edge height. Based on these results, we conclude that a suitable d/H ratio of ~ 0.5 – 0.9 and a high step angle of $\phi \geq 70^\circ$ can be regarded as the optimum conditions for the fabrication of step-edge TMR junctions.

4. Summary

In summary, we report the fabrication techniques of LSMO step-edge TMR junctions. Using standard photolithography and ion-beam etching techniques, the step angle and step height can be controlled by the θ angle and etching time, respectively. We study the influence of step-edge conditions on the resistance and MR behav-

ior of LSMO TMR junctions. It is found that the magnitude of junction resistance increases with either an increase of step angle or with a decrease of d/H ratio. Finally, we obtain the optimum step-edge conditions containing a suitable d/H ratio of ~ 0.5 – 0.9 and a high step angle of $\phi \geq 70^\circ$ for the fabrication of step-edge TMR junctions.

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