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Study of magnetoresistance and conductance of bicrystal grain boundary in $La_{0.67}Ba_{0.33}MnO_3$ thin film

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Abstract. $La_{0.67}Ba_{0.33}MnO_3$ (LBMO) thin film is deposited on a 36.7°C $SrTiO_3$ bicrystal substrate using laser ablation technique. A microbridge is created across bicrystal grain boundary and its characteristics are compared with a microbridge on the LBMO film having no grain boundary. Presence of grain boundary exhibits substantial magnetoresistance ratio (MRR) in the low field and low temperature region. Bicrystal grain boundary contribution in MRR disappears at temperature $T > 175$ K. At low temperature, I–V characteristic of the microbridge across bicrystal grain boundary is nonlinear. Analysis of temperature dependence of dynamic conductance–voltage characteristics of the bicrystal grain boundary indicates that at low temperatures ($T < 175$ K) carrier transport across the grain boundary in LBMO film is dominated by inelastic tunneling via pairs of manganese atoms and tunneling through disordered oxides. At higher temperatures ($T > 175$ K), magnetic scattering process is dominating. Decrease of bicrystal grain boundary contribution in magnetoresistance with the increase in temperature is due to enhanced spin-flip scattering process.

Keywords. Colossal magnetoresistance; bicrystal junction; grain boundary; LBMO; spin-polarized tunneling.

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1. Introduction

Observation of colossal magnetoresistance (CMR) in perovskite manganese oxides has generated a lot of scientific interest [1–3]. Epitaxial thin films of doped $LaMnO_3$ show large magnetoresistance at a large field ($\sim$ few Tesla) and in a narrow temperature range at ferromagnetic transition temperature [4,5]. From a practical point of view, large magnetoresistance at low field and over a wide temperature range is desirable. Unlike the epitaxial thin films, bulk samples and polycrystalline films of doped $LaMnO_3$ show large magnetoresistance effects in the low-field region and even at temperature much lower than the ferromagnetic transition temperature [4–8]. The electrical and magnetic properties of CMR oxides in the ferromagnetic state are explained in the framework of double exchange model [9]. The need to consider the role of strong electron–phonon coupling arising due to John–Teller splitting of outer d orbital of Mn has also been emphasized [10]. In the polycrystalline film and bulk samples electrical transport across the grain boundaries is proposed to be due to spin polarized tunneling [7] or due to spin-dependent scattering of polarized electrons at grain boundaries [3,6]. A mesoscopic magnetoresistance model based on a grain boundary region with strongly suppressed T_c has also been proposed [11]. Recently, artificially created grain boundaries in epitaxial CMR films such as bicrystal junction in LSMO or LCMO films [12,13], edge junction in LCMO film [14] have been found to exhibit substantial low-field magnetoresistance at low temperature. Realization of a single artificial grain boundary in CMR epitaxial film provides a good opportunity for studying the role of grain boundary in CMR materials. This paper reports temperature dependence of magnetoresistance and dynamic conductance of a bicrystal grain boundary in $La_{0.67}Ba_{0.33}MnO_3$ thin film.

2. Experimental

Thin film of $La_{0.67}Ba_{0.33}MnO_3$ (LBMO) was prepared on 36.7°C bicrystal substrate of $SrTiO_3$ from a stoichiometric LBMO target using pulsed laser deposition technique. Deposition of the film was carried out in a partial oxygen atmosphere (400 mTorr) and the substrate temperature was kept at 750°C during deposition. On either side of the bicrystal grain boundary the LBMO films grow epitaxially forming an artificial grain boundary at the fusion line of bicrystal. For studying the effect of the grain boundary on transport characteristics one microbridge was created across the bicrystal grain boundary and the other one away from the bicrystal grain boundary. The width of these microbridges were $\sim$ 700 μm. Resistance–temperature (R–T) and current–voltage (I–V) characteristics of these microbridges were studied using four-probe technique. For magnetoresistance measurements, magnetic field has been applied in the plane of the film, parallel to the grain boundary. Magnetoresistance ratio (MRR) has been calculated using the relation: $\mathrm{MRR} = \{[R(0) - R(H)]/R(0)\} \times 100\%$, where $R(H)$ and $R(0)$ are resistances of the microbridge in the presence and in the absence of magnetic field, respectively. I–V curves were recorded by applying slow varying current. Dynamic conductance vs. voltage plots were obtained numerically from I–V data.

3. Results and discussion

Figure 1 shows R–T curves for both microbridges. These curves show that both the microbridges undergo a transition from a semiconducting like ($\mathrm{d}R/\mathrm{d}T < 0$) to a metallic-like ($\mathrm{d}R/\mathrm{d}T > 0$) behavior as the temperature is decreased from room temperature. The microbridge across the grain boundary showed larger resistance and a smaller value of peak temperature (T_p) in the R–T curve. For the microbridge away from the grain boundary T_p was ~250 K whereas for the microbridge across the grain boundary T_p was ~236 K. The observed smaller value of T_p of the microbridge across the bicrystal grain boundary is most likely due to oxygen deficient layer at the grain boundary. Figure 2 shows temperature dependence of magnetoresistance ratio (MRR) for both the microbridges at 1 kOe magnetic field. At lower temperatures behavior of MRR-T curves are different. For the microbridge which is away from the grain boundary, values of MRR keep on decreasing as temperature

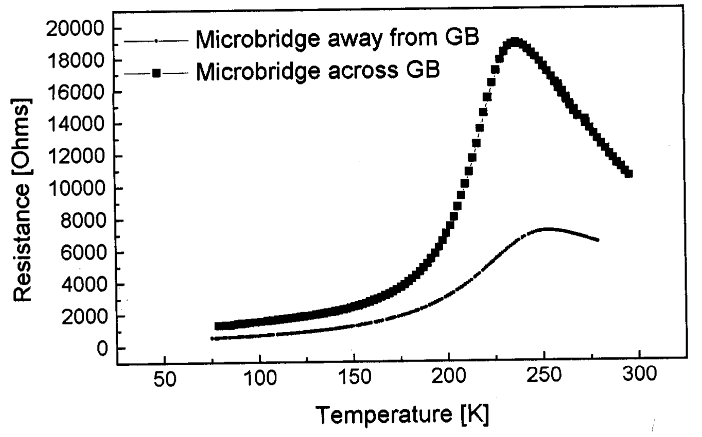

Figure 1. Resistance vs. temperature curves for the LBMO thin film microbridges across bicrystal grain boundary and away from the grain boundary.

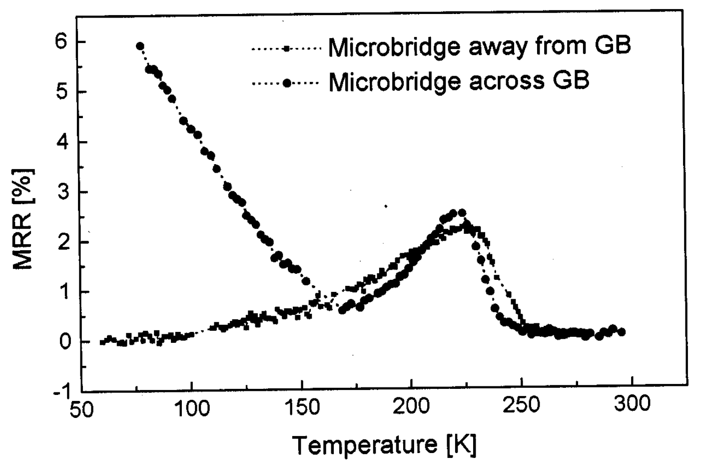

Figure 2. Temperature dependence of magnetoresistance ratio (MRR) of LBMO thin film microbridges across bicrystal grain boundary and away from the grain boundary.

is decreased from 225 K. The values of MRR for the microbridge across the bicrystal grain boundary shows a decrease as the temperature is decreased from 225 K but as the temperature is further lowered from 175 K, the value of MRR shows an increase. At 77 K, the value of MRR for the microbridge away from the grain boundary was 0.02% whereas for the microbridge across the grain boundary, MRR was 6%.

Figure 3 shows I–V curves for both the LBMO film microbridges at 4.2 K. The microbridge at the grain boundary exhibits nonlinear I–V characteristic whereas the microbridge away from the grain boundary shows linear I–V characteristic. In order to explore the temperature dependence of nonlinearity of bicrystal grain boundary, we have studied dynamic

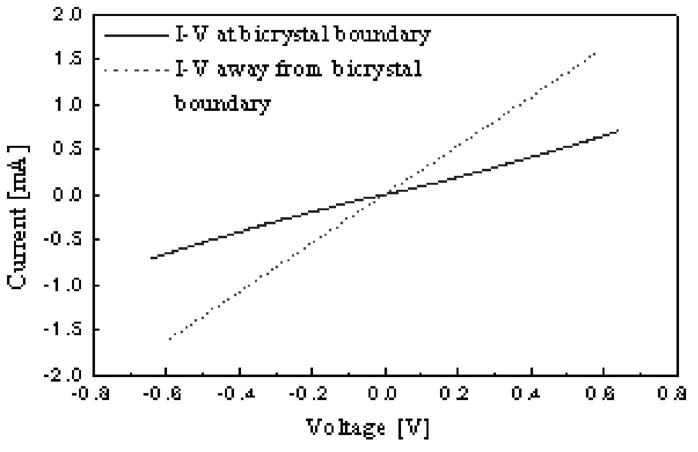

Figure 3. I–V curves for the LBMO thin film microbridge across bicrystal grain boundary and for the microbridge away from the grain boundary.

conductance ($G = \mathrm{d}I/\mathrm{d}V$) of the grain boundary junction as a function of voltage drop across the junction. Figure 4a shows normalized dynamic conductance (G/G_0) vs. voltage curves for the bicrystal junction at different temperatures. It is clear that nonlinearity in I–V curves changes with the increase in temperature.

For a quantitative analysis, the dynamic conductance (G) of the grain boundary junction is analyzed using the expression

$$G/G_0 = 1 + kV^{\alpha}\,, \tag{1}$$

where G_0 is the conductance of the junction at zero voltage, k and α are constants. A theoretical value of $\alpha = 1.3$ is predicted for quasiparticle tunneling via pairs of localized state [15] and a value of $\alpha = 2$ is predicted for direct tunneling [16]. A value of $\alpha = 2$ is also predicted for spin-flip scattering process [17]. Tunneling through disordered oxide gives a value of $\alpha = 0.5$ [18]. We have obtained the value of exponent α by fitting eq. (1) to the experimental G/G_0 vs. V curves. Figure 4b shows the variation of the exponent α with the temperature. The value of α is $\sim$ 1.3 between 100 to 175 K and it increases for $T > 175$ K approaching to a value of 1.8. The value of α decreases to 0.6 for temperatures below 100 K. A comparison of the experimental value of α with theoretical value indicates that for $T < 175$ K in LBMO film bicrystal junction carrier transport is dominated by inelastic tunneling via pairs of manganese and tunneling through disordered oxides. At higher temperature spin flip scattering is dominating. We notice a correlation between the bicrystal junction contribution in MRR and nonlinearity in the I–V characteristics across the bicrystal junction. The magnetoresistance observed in this bicrystal grain boundary is related with tunneling. Decrease of magnetoresistance ratio of bicrystal grain boundary with the increase in temperature is due to enhanced spin-flip scattering at the grain boundary. Spin-flip scattering becomes dominant at $T > 175$ K and contribution of bicrystal junction in MRR also disappears for $T > 175$ K.

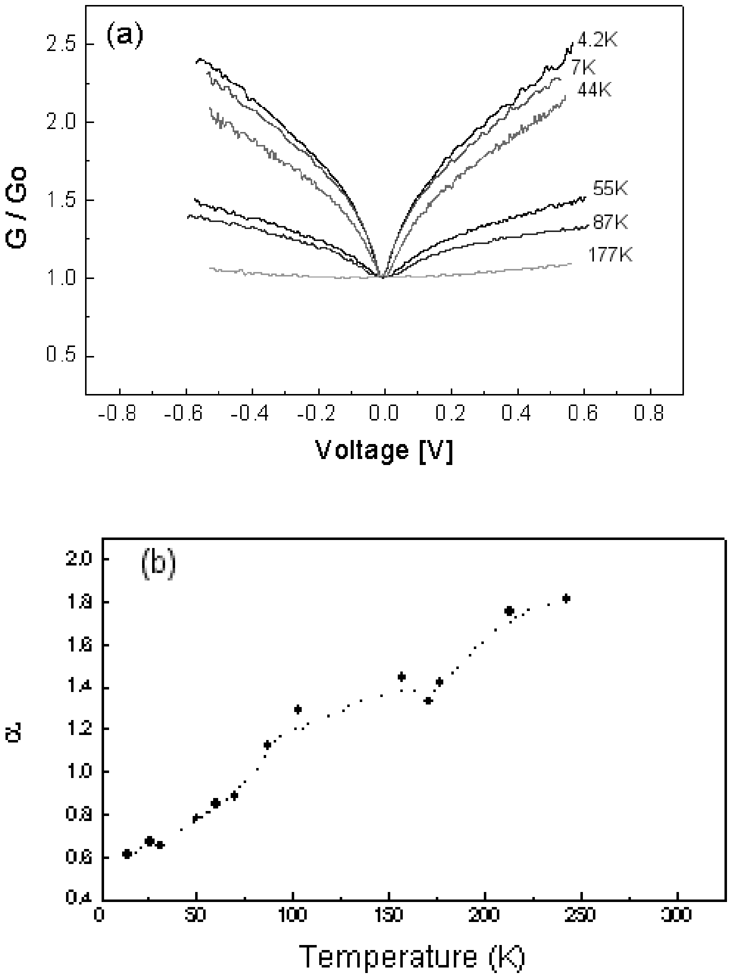

Figure 4. (**a**) Variation of normalized dynamic conductance (G/G_0) with voltage drop across the bicrystal junction in LBMO films at different temperatures. (**b**) Variation of α with temperature. The value of α is obtained from the fitting of eq. (1) to the experimental G/G_0 vs. voltage curves.

4. Conclusion

Presence of a bicrystal grain boundary in LBMO epitaxial film microbridge enhances low-field magnetoresistance and introduces nonlinearity in I–V characteristics. At low temperatures the carrier transport across the grain boundary is due to inelastic tunneling via Mn pairs of atoms and tunneling through disordered oxide. The spin-polarized tunneling at the grain boundary is responsible for the magnetoresistance. At higher temperature, spin-flip scattering becomes dominant which reduces the magnetoresistance ratio.

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