

Rectifying characteristics and magnetoresistance in $\text{La}_{0.9}\text{Sr}_{0.1}\text{MnO}_3/\text{Nb-doped SrTiO}_3$ heterojunctions

Z. Luo, J. Gao*

Department of Physics, The University of Hong Kong, Pokfulam Road, Hong Kong, China

Abstract

Manganite-based heterojunctions have attracted lots of attention as one of the most promising practical applications of colossal magnetoresistance materials. In this work, heterojunctions were fabricated by depositing $\text{La}_{0.9}\text{Sr}_{0.1}\text{MnO}_3$ (LSMO) films on substrates of 0.7 wt.% Nb-doped SrTiO_3 using pulsed laser deposition technique. X-ray diffraction spectra confirmed that the grown films are of single phase and have an orientation with the *c*-axis perpendicular to the substrate surface. As temperature decreases, the resistivity of LSMO films first increases gradually and then increases abruptly at temperature lower than 150 K. These junctions showed clear rectifying characteristics and strong temperature dependent current–voltage relation. Diffusion voltage decreases as temperature increases. Under forward bias, current is proportion to $\exp(eV/nkT)$. Ideal factor increases quickly and tunneling current plays more and more important role as temperature decreases. At 50 K, tunneling current becomes nearly dominant. Large magnetoresistance was observed. The sign and value of such magnetoresistance depends on the direction and value of current.

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1. Introduction

Manganite-based heterojunctions have attracted lots of attention as one of the most promising practical applications of colossal magnetoresistance materials. Clear rectifying characteristics and photovoltaic effect have been observed in many manganite-based heterojunctions [1,2]. Strong electric field modulation of ferromagnetism in a manganite-based p–n junction, by carrier injection, at room temperature was observed [3]. Recent studies also suggested that rectifying characteristics and photovoltaic effect depend strongly on the electrical and magnetic properties of manganese oxide layers. A synchronous variation of diffusion voltage (V_d), photovoltage and the magnetization of $\text{La}_{0.32}\text{Pr}_{0.35}\text{Ca}_{0.33}\text{MnO}_3$ (LPCMO) has been observed in LPCMO/Nb: SrTiO_3 (STON) heterojunction [2]. This implies that the properties of a manganite-based heterojunction can be modified by changing the magnetic state of the manganite layer.

One of the most interesting properties of manganese oxides is that their resistance will decrease dramatically once a magnetic field is applied. This phenomenon is called colossal magnetoresis-

tance (CMR). Within double-exchange picture, the procedure of electron transferring from Mn^{3+} to the adjacent Mn^{4+} in manganese oxides ($\text{R}_{1-x}\text{A}_x\text{MnO}_3$ (R = rare-earth cation, A = alkali or alkaline earth cation) can be visualized as one electron transfers from Mn^{3+} to O^{2-} and simultaneously one electron transfers from O^{2-} to Mn^{4+} . However, due to strong Hund's coupling, the exchange integral depends strongly on the spin of the adjacent Mn ions. The exchange integral is proportion to $\cos(\theta/2)$, where θ is the angle between the two spin directions [4]. Under a magnetic field, spins of adjacent Mn ions are aligned. It will be easier for electrons to transfer from one Mn ion to another, leading to a decrease of resistance. As magnetic field can influence the electronic structure of manganese oxides, a magnetoresistance (MR) is expected in a manganite-based heterojunction too. MR is really observed in many manganite-based heterojunctions [5–7]. However, the observed phenomenon is very complex. In some cases it is positive [7], while in others it is negative [6]. Sun et al. also reported that, in a LPCMO/STON heterojunction, MR can be either positive or negative, depending on temperature and applied current [5]. In order to make the nature of junction MR clear, more experimental data is necessary.

In this work, transport properties of $\text{La}_{0.9}\text{Sr}_{0.1}\text{MnO}_3$ (LSMO)/STON were investigated. These junctions showed clear rectifying characteristics and strong temperature dependent current–voltage relation. Tunneling current plays more

* Corresponding author.

E-mail address: jugao@hku.hk (J. Gao).

and more important role as temperature decreases and become dominant at 50 K. Complex junction magnetoresistance was observed.

2. Experimental

LSMO/STON heterojunctions were fabricated by depositing LSMO thin films on 0.7 wt.% Nb-doped SrTiO₃ single crystal substrates with (100) orientation using pulsed laser deposition technique. Details about the growth of the films were described elsewhere [8]. During the deposition procedure, the substrate temperature was kept at about 923 K and the oxygen partial pressure was about 10 Pa. The thickness of the LSMO thin film is about 370 nm controlled by the deposition time. LSMO films were also prepared on SrTiO₃ (STO) single crystal substrates for resistance measurement of LSMO films. Then LSMO films were patterned into blocks with conventional photolithography. Silver electrodes were evaporated on the LSMO and STON thermally. X-ray diffraction was made using a Phillips D5000 diffractometer. Resistivity–temperature (R – T) relationships were measured with standard four-probe technique. Current–voltage (I – V) curves were measured at various temperatures using systems equipped with closed cycle refrigerator and superconductivity magnet. I – V curves measurement were performed with two-terminal technique using a tunable current source.

3. Results and discussion

XRD suggested that the grown films are of single phase and excellent texture. Fig. 1 is a typical θ – 2θ scan pattern. Only (100) peaks of LSMO were observed in addition to those of STO substrate. LSMO has an orthorhombic ($Pnma$) crystal structure with $a = 0.552$ nm, $b = 0.556$ nm and $c = 0.777$ nm [9]. STO has a cubic structure with $a = 0.3905$ nm or equivalently a tetragonal structure with lattice parameters: $a = b = 0.5522$ nm and $c = 0.3905$ nm, by rotating 45° in a – b plane [10]. The lattice mismatch between LSMO and STO substrate is very small. If LSMO grows epitaxially with c -axis normal to a – b plane

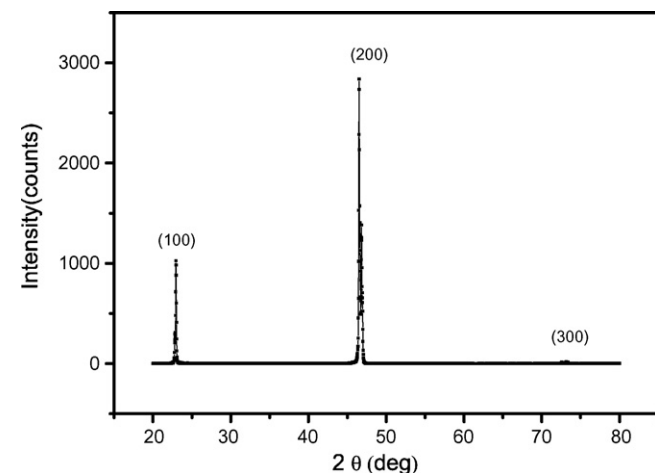


Fig. 1. Typical XRD pattern of La_{0.9}Sr_{0.1}MnO₃ films.

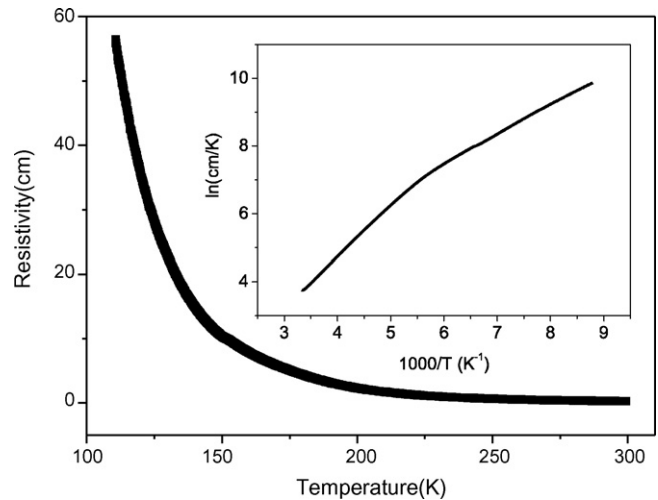


Fig. 2. Typical temperature dependence of resistivity of La_{0.9}Sr_{0.1}MnO₃ films. Inset is plot of $\ln(\rho/T)$ vs. $1000/T$.

of STO, LSMO films will be slightly compressive strained. In this work, XRD pattern was indexed assuming a cubic symmetry of LSMO for simplicity. Calculated from (200) peak, the LSMO film has a lattice parameter of $a_p = 0.388$ nm. This value is slightly smaller than $a/\sqrt{2}$ or $b/\sqrt{2}$, implying that the grown film may be slightly strained.

Fig. 2 shows temperature dependence of LSMO resistivity. As temperature decreases, the resistivity of LSMO first increases slowly and then very quickly at temperature lower than 150 K. For temperature lower than 100 K, the resistance is so large that no stable current can be applied to the sample. This suggested that LSMO films are non-metal in temperature region of our measurement (10–300 K). In high temperature range, $\ln(\rho/T)$ is proportional to $1/T$ (inset of Fig. 2), which is predicted by the adiabatic small-polaron hopping conduction $\rho = AT \exp(E_A/k_B T)$. At about 180 K, the slope of $\ln(\rho/T)$ – $(1/T)$ curve changes, implying a change of active energy. This small-polaron hopping conduction has also been observed in strained LSMO films [11]. LSMO single crystal is paramagnetic insulator at high temperature and ferromagnetic insulator at lower temperature ($T_C = 145$ K) [9]. Previous studies also suggested that oxygen content have great influence on magnetic and transport properties of LSMO. Samples with high oxygen deficient are non-metal. Curie temperature decreases as oxygen deficiency increases, while the ferromagnetism does not disappear [12]. Films fabricated in 10 Pa partial oxygen pressure may be oxygen deficient. The Curie temperature of the grown films would be lower than that of single crystal. However, the strain effect will lead to an increase of Curie temperature [11]. Taking oxygen deficient, strain effect and the strong correlation between resistance and magnetic state in LSMO system into account, the grown films should have a Curie temperature close to 180 K, judging from the change of the slope of the $\ln(\rho/T)$ – $(1/T)$ curve.

I – V curves at various temperatures were shown in Fig. 3. Forward bias was defined as a positive dc voltage on LSMO films. The junction clearly exhibits rectification. Under a small forward bias, the current is small. When the applied bias increases to a

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