Rapid Communication

Effect of film thickness on interfacial barrier of manganite-based heterojunctions*

Xie Yan-Wu (谢燕武)[a], Guo De-Feng (郭得峰)[a],
Sun Ji-Rong(孙继荣)[b], and Shen Bao-Gen(沈保根)[b]

[a] *State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao 066004, China*
[b] *Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*

(Received 15 June 2010; revised manuscript received 20 July 2010)

Interfacial barrier is a key factor that determines the performances of heterojunctions. In this work, we study the effect of manganite film thickness on the effective interfacial barrier for $La_{0.67}Sr_{0.33}MnO_3$/Nb:$SrTiO_3$ junctions. The barrier is extracted from the forward current–voltage characteristics. Our results demonstrate that the barrier decreases gradually from ∼0.85 eV to ∼0.60 eV when the film thickness decreases from 150 nm to 2 nm. The overall value of the barrier is only about 50% of the corresponding one determined from the photovoltaic effect.

Keywords: manganite, heterojunction, thickness
PACC: 7340, 7530V, 7360T

1. Introduction

Manganite-based bilayer heterojunctions have aroused considerable interest in recent years because of their interesting physical properties and potential applications in high-performance oxide based electronic devices.[1−5] In addition to rectifying behaviour, these junctions also show a lot of extraordinary properties such as strongly bias-dependent magnetoresistance[1,3,4] and temperature/magnetic field-dependent photovoltaic effect.[6] The height of the barrier that forms at the interface of these junctions is an essential parameter that determines their performances. Generally, three methods have been used to indentify the height of interfacial barrier (V_B) of manganite junction. The first two methods are based on the analyses of the current–voltage (I–V) or capacitance–voltage (C–V) characteristics, respectively, through adopting the conventional semiconductor model.[7,8] The third one is based on the internal photoemission measurements,[9] and the height of V_B is also extracted from the photovoltaic effect of junctions.[10] Although the relevant studies have extensively been carried out, the understanding of the interfacial state in manganite junctions is still very limited due to the extreme complexity involved in manganite, i.e., the strong coupling between spin/charge/orbital degrees of freedom and lattice strains.[11]

It is well known that physical properties of manganite film significantly vary with film thickness.[12] Therefore, a systematic study of the effect of film thickness on manganite junction is highly desired. A systematic thickness-dependent study by photovoltaic technique has been performed.[10] Photovoltaic method, as well as the photoemission spectroscopy method, is useful in the sense that it can accurately measure the intrinsic V_B, without any difficulty encountered in analysing I–V and C–V characteristics, especially junctions where the transports severely deviate from the thermal diffusion or thermal emission models.[9,10] However, from another viewpoint, this merit could also be a shortcoming. As shown in Fig. 1, in a manganite junction the charge carriers may tunnel through, instead of surmount over, the interfacial barrier. As a result, the effective barrier (V_{EB}) that actually restricts the transport will be lower than V_B. For practical application, it is still of great importance to study how the V_{EB}, instead of V_B, develops with the thickness of manganite film.

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10804094, 50832007, 50721001 and 50821001), and the Natural Science Foundation of Hebei Province, China (Grant No. A2009000339).
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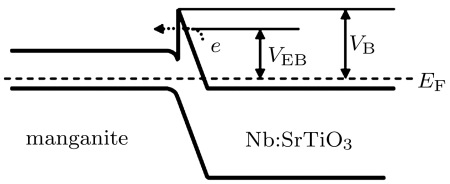


Fig. 1. Schematic band diagram of a typical manganite heterojunction.

Here, we report on a systematic study of the thickness-dependent V_{EB} on a series of junctions based on $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) films. The V_{EB} is derived from the I–V relations. Our results demonstrate that V_{EB} decreases gradually from ∼0.85 eV to ∼0.60 eV when the film thickness decreases from 150 nm to 2 nm. The value of V_{EB} is only about 50% of the corresponding V_B determined from the photovoltaic effect.

2. Experiment

The junctions were fabricated by growing LSMO films on 0.05 % Nb-doped (001) $SrTiO_3$ (NSTO) substrates through using a pulsed laser deposition technique. During deposition the substrate temperature was kept at 750 °C and the oxygen pressure was maintained at 50 Pa. The film thickness was controlled by the deposition time. Similar LSMO films were also deposited on the undoped $SrTiO_3$ substrates, and they were used to measure the in-plane resistivity of the LSMO films.

The x-ray diffraction analysis reveals epitaxial growth of the (001)-oriented LSMO film (not shown). Atomic force microscopy analysis indicates a rather flat film surface. The root-mean-square roughness is ∼0.24 nm for the 2 nm film, showing that even in the thinnest case the substrate was fully covered by the LSMO film.[13] The lateral size of the junctions is 1 mm×1 mm, patterned by the conventional photolithography and chemical etching technique. Two Ag pads were deposited, respectively, on LSMO and NSTO as electrodes and the contacts are quasi-ohmic.

3. Results and discussion

Figure 2 shows the temperature dependences of in-plane resistivity of LSMO films of different thicknesses, fabricated on the undoped $SrTiO_3$ substrates under the same conductions as those for the corresponding junctions. The thinnest LSMO film (2 nm) exhibits a totally insulating conduction with a resistivity at least two order of magnitude larger than that of thicker film. As the film thickness increases to 5 nm, a metallic behaviour is observed in the whole temperature range and a transition in resistivity is found to appear at around 320 K. Compared with the resistivity of 5 nm thick film, the resistivity of 10 nm thick film decreases about 4 times in the whole temperature range and the transition temperature rises up to 350 K and more. Further increase in film thickness to 150 nm only causes small variation in resistivity. These results confirm that the film thickness has a significant influence on the properties of LSMO films and the influence is much stronger when the film is thin.

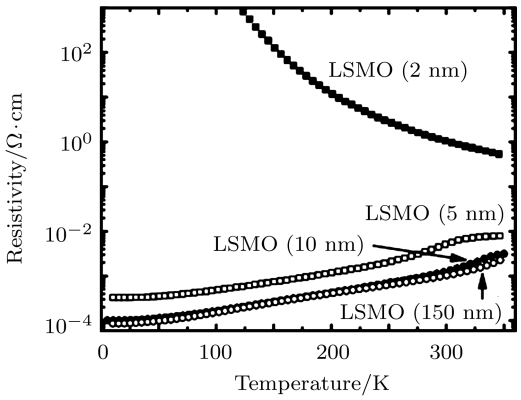


Fig. 2. Temperature-dependent in-plane resistivities of LSMO films of different thicknesses.

To have a general impression on the transport behaviours, in Fig. 3 we show the I–V characteristics of the 2 nm (the thinnest) and the 150 nm (the thickest) junctions. The two junctions have rather similar transport behaviours, in spite of the huge differences in the film thickness and conduction. Each of them shows an excellent rectifying behaviour (see Figs. 3(a) and 3(b)). In general, the current increases exponentially with positive bias voltage, while a reverse bias, even it is as large as −5 V, only yields negligible current, suggesting a depression of the tunnelling conduction. We emphasize that the junctions of all thicknesses studied in this work each show a good I–V relation that follows the conventional semiconductor model, which is different from the case in the previous study.[10] We ascribe this improvement to the low doping density of NSTO and the high quality of the junctions, both of which suppress the leakage conduction.

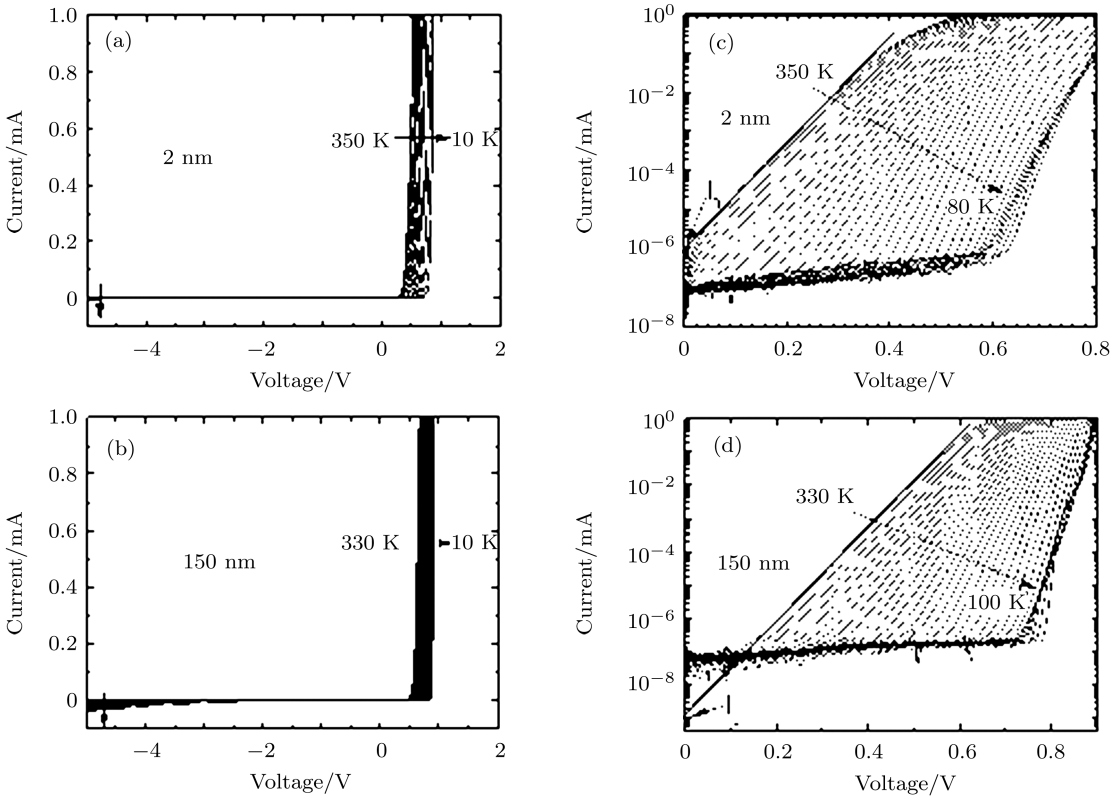


Fig. 3. Current–voltage characteristics of (a) 2 nm and (b) 150 nm junctions. The forward characteristics are also re-plotted in semi-logarithm coordinates (c) for 2 nm and (d) for 150 nm junctions. The thick lines are guides for eyes. The deviation from linear relation at high currents is caused by the circuit resistance. The shutdown at low currents is limited by the measuring ability.

In previous studies two models, p–n junction (thermal diffusion) and Schottky junction (thermal emission), have been used for extracting $V_{\rm EB}$.[7,8] Although the their mechanisms might be quite different, the two models yield similar I–V relations and also give roughly the same values of $V_{\rm EB}$.[7] Obviously it is unreasonable to treat the LSMO (2 nm)/ NSTO junction as a Schottky junction, owing to the insulating conduction of LSMO (2 nm). So, for simplicity, here we analyse the junctions of all thicknesses using the p–n junction model with a band diagram as shown in Fig. 1 (An analysis from the Schottky model gives a similar result). In a p–n junction the forward bias current can be expressed approximately as[7,14]

$$I_{\rm F} = I_0 \exp(qV/nk_{\rm B}T), \quad (1)$$

$$I_0 \propto T \exp(-qV_{\rm EB}/k_{\rm B}T), \quad (2)$$

where q is the unit charge, n is the ideal factor, and $k_{\rm B}$ is the Boltzmann constant. Equation (1) implies a linear dependence of log $I_{\rm F}$ on bias V when $qV > k_{\rm B}T$. This relation is exactly mirrored in Figs. 3(c) ($T > 80$ K) and 3(d) ($T > 100$ K). At lower temperatures the log I–V curves show deviations from the systematic trend. This is caused by the onset of quantum paraelectricity in the depletion layer of Nb:STO, where the dielectric constant strongly depends on the bias voltage.[15] And also, the dielectric constant increases sharply at low temperatures, which may cause further deviation.[16]

Next, we evaluate $V_{\rm EB}$ from the temperature dependence of I_0. From Eq. (2) one can see that the temperature dependence of I_0 satisfies a relation log $(I_0/T) \propto qV_{\rm EB}/k_{\rm B}T$. Therefore, $V_{\rm EB}$ can be calculated from the slope of log(I_0/T)–$1/T$. As shown in Eq. (1), I_0 can be extrapolated from the intersection of the log $I_{\rm F}$–V line and the zero-bias axis (indicated in Figs. 3(c) and 3(d)). Figure 4 shows the log(I_0/T)–$1000/T$ characteristics for the 2 nm and 150 nm junctions. To avoid the influence of low-temperature effect, we only use the data extracted from the I–V curves measured at a temperature larger than 200 K. As expected, log(I_0/T)–$1000/T$ is in a good linear relation, and the derived values of $V_{\rm EB}$ are 0.64 V and 0.85 V for the 2 nm and 150 nm junction, re-

spectively. These values are much smaller than the interfacial barrier values evaluated from the photoemission spectroscopy[9] and the photovoltaic effect,[10] but consistent well with those evaluated from I–V and C–V characteristics,[7] confirming that the barrier actually restricting the transport might be much smaller than the intrinsic one.

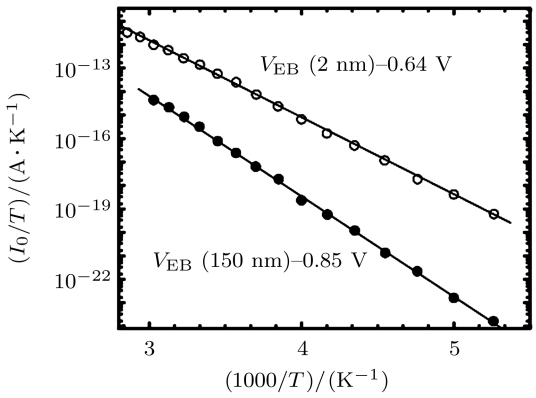


Fig. 4. Linear relations of $\log(I_0/T)$ versus $1000/T$ for the 2 nm and 150 nm junctions. V_{EB} is derived from their slopes.

Figure 5 shows the V_{EB} values of junctions with thickness varying from 2 nm to 150 nm. The data are taken from three sets of junctions fabricated under the same conditions. There is no sharp change in V_{EB} when the film thickness decreases from 5 nm to 2 nm, although a dramatic change of in-plane resistivity can be observed (see Fig. 2). This indicates that the manganite layer near the interface, which is assumed to be insulting, plays a dominant role in the transporting of the junction. Given a film thickness, a large variation in V_{EB} is observed. This might be caused by unavoidable interface defects which induce different interfacial states.[14] How to well control the interfacial states is still a big challenge in this field. Despite the large variation in V_{EB}, we can still see an overall trend that V_{EB} decreases with film thickness decreasing. This trend is consistent with the result achieved by photovoltaic study, in which the variation in barrier was related to the thickness-dependent lattice constant.[10] However, there is also a large difference between the results from the two methods. The V_{EB} value deduced here is only half of the V_B value obtained from the photovoltaic effect and shows no saturation,[10] demonstrating that besides the intrinsic height, the width/shape of interfacial barrier plays a great role in determining V_{EB}.

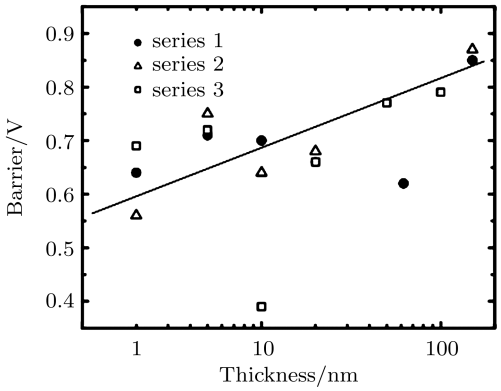


Fig. 5. Variation of V_{EB} with LSMO thickness. The data are taken from three sets of junctions fabricated under the same conditions. The solid line is guide for eyes.

4. Summary

We have studied the thickness-dependent interfacial barrier of LSMO/NSTO junctions by analysing the forward I–V characteristics. In spite of the existence of large variation that might be caused by the unavoidable interfacial detects, the barrier shows an overall decreasing trend with film thickness decreasing, which is consistent with the previous study by using photovoltaic effect. However, the barrier value deduced here is only half of that deduced from photovoltaic effect and shows no saturation. Our result confirms that the film thickness of manganite has a nontrivial effect on the interfacial barrier of the junctions.

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