

Resistive switching behavior of BaTiO₃/La_{0.8}Ca_{0.2}MnO₃ heterostructuresS.Y. Wang^{a,*}, Meng Li^a, W.F. Liu^{c,*}, J. Gao^b^a College of Physics and Material Science, Tianjin Normal University, Tianjin 300074, China^b Department of Physics, the University of Hong Kong, Pokfulam Road, Hong Kong, China^c Department of Applied Physics, Faculty of Science, Tianjin University, Tianjin 300072, China

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ABSTRACT

The electric transport properties of BaTiO₃/La_{0.8}Ca_{0.2}MnO₃ heterostructures were investigated in the temperature range 100–320 K. It is found that the leakage current versus voltage (*I*–*V*) curves show strong temperature dependence, and at lower temperature (<250 K) the *I*–*V* curves are diode-like asymmetric characteristics; while they exhibit resistive switching behavior at higher temperature. Moreover, the resistive switching coefficient increases with the elevated temperature. Such switching conduction has been ascribed to the modulation of interfacial barrier height in BaTiO₃/La_{0.8}Ca_{0.2}MnO₃ heterostructures upon the ferroelectric polarization flipping. Such type of resistive switching behavior based on a correlation between ferroelectric polarization and conductivity might become of particular interest for nonvolatile memory applications.

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1. Introduction

In recent years, with the on-going miniaturization process of the silicon-based devices and demand for the increasing storage capacity per unit area for future electronic circuitries, great deal of research activities has been focused in the exploration of novel materials for utilizing as memory and switching-based devices [1–3]. Wherein, nonvolatile random access memory concepts based on a ferroelectric resistance random access memory (RRAM) have been considered as the most promising candidate for future universal memory, since they not only unify fast read/write speeds, lower power consumption, nonvolatile characteristics, but also are in principle capable of being highly scalable below 10 nm [4–7].

Ferroelectric materials possess a spontaneous electric polarization that can be switched by an external electric field. One type of resistive switching (RS) devices called ferroelectric tunnel junction (FTJ) can be constructed employing ferroelectric films with thickness of several dozens of nanometer or less, in which dominating electric transport mechanism is quantum mechanical tunneling. Upon switching the ferroelectric polarization, the resistive states of the device could switch between high resistance state and low resistance state [3–7]. Another type of structure exhibiting RS effect are so-called ferroelectric diodes, such as Au/PbTiO₃/LaSrCoO₃ [8], Pt/PbTiO₃/SrRuO₃ [9], and Pt/PbTiO₃/LaSrCoO₃ [10] heterostructures, in which the thickness of ferroelectric film is several hun-

dred of nanometers and large enough to avoid the electron tunneling, and the underlying mechanism for RS phenomena has been ascribed to the polarization reversal-induced variable barrier height, or to the asymmetric distribution of electron trapping centers [11]. However, the resistive switching coefficient in ferroelectric diodes is usually less than 200. And, most of these resistive switching effects are based on a certain type of defect (ionic or electronic) mediated phenomenon, suggesting the inherent difficulty in precise control of the switching behavior. Recently, Jiang et al. [12] proposed a new type resistive memory in which the bistable diode currents can be obtained through changing the ferroelectric polarization direction in Pt/BiFeO₃/SrRuO₃ thin-film capacitors. Wherein the BiFeO₃ films were thought as semiconducting layer in nature, hence the ferroelectricity and conductivity could coexist and interact in a single phase of BiFeO₃ layer. In Pt/BiFeO₃/SrRuO₃ capacitors the RS effect is ascribed to the modulation of conductivity of BiFeO₃ layer by ferroelectricity in itself, rather than to the interfacial effects due to high conductivity for both Pt and SrRuO₃ layers.

As proposed by R. Myer and R. Waser [13] that the metal–ferroelectric–semiconductor (MFS) structure employing semiconductor layer as electrode is more promising for better RS coefficient. As compared to a metal electrode, the screening effect of the bound charges is less efficient, due to lower concentration of carriers in a semiconductor. As a result, a bending of the energy band could be induced at the region near interface, which could dramatically modify the potential profile, promising to gain a large resistive switching coefficient in MFS structure. Moreover, the quality of semiconducting manganites is believed to be substantially supe-

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rior because of their chemical stability with the ferroelectrics and their rich electronic phases [14]. It is known that RS phenomena have been observed for manganites sandwiched by two metallic electrodes in a capacitor-like structure. The physical origin of RS effect in manganites is attributed to local changes of oxygen vacancy concentration at the interface of metal/manganites induced by electrical perturbation [15,16]. Recently, Bryant et al. provide direct image of oxygen defects and their motion at a manganite surface by using scanning tunneling microscopy [17]. In manganites the delicate balance between electronic phases is sensitive to external perturbation including the applications of electric/magnetic fields or pressure. Thus, a dramatic response in conduction is expected to external stimuli in MFS structure with semiconducting manganite layer [18].

In this paper we present an experimental result about the electric transport properties in a composite structure composed of $\text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$ (LCMO) and ferroelectric BaTiO_3 (BTO) thin films. The electric current versus bias voltage (I–V) curves of the heterostructure exhibits different characteristics in various temperatures (100–320 K). At around room temperature the I–V curve shows resistive switching behavior between a low-current state and a high-current state. The RS coefficient shows strong dependence on the ambient temperature. The fundamental basics for RS effect in heterostructures were discussed.

2. Experiment

In this work, LCMO thin films as the bottom electrode with thickness of 150 nm were deposited on (001)-oriented SrTiO_3 single-crystal substrates. BTO layers of 350 nm are employed as the ferroelectric component to construct heterojunctions. Both LCMO and BTO layers were deposited by pulsed-laser ablation, which taken place at substrate temperature of 720°C and in a pure oxygen atmosphere of the pressure of 6×10^{-1} mba and 2×10^{-1} mba, respectively. The energy of 300 mJ, wavelength of 308 nm, and pulse frequency of 2 Hz were employed for the laser beam. A post annealing at 700°C was made for 30 min in a pure oxygen atmosphere of the pressure 1 atm. The structural quality of the films was examined by X-ray diffraction (XRD) using $\text{CuK}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$). For electric measurements, silver pad electrodes were deposited by thermal evaporation onto the BTO and LCMO films. A schematic drawing for the Ag/BTO/LCMO heterostructure is shown in Fig. 1(a). Electric properties were examined with Keithly 2400 in a closed-cycle cryostat. In voltage swept process, a voltage ramp rate of 0.1 V/step and a delay time of 2 seconds were used. The polarity of the bias voltage is defined as positive when the bias is applied to top electrode Ag with area of $3 \times 10^{-4} \text{ mm}^2$. P–E loops were measured with an Aixacct model TF 2000 ferroelectric analyzer.

3. Results and discussions

Fig. 1(b) presents the typical XRD patterns of θ – 2θ scans for BTO/LCMO on SrTiO_3 substrate. In the scanned range of 2θ from 10° to 80° , there are only well defined and high density (001) reflections corresponding to BTO, LCMO layers and STO substrates, indicating that both LCMO and BTO layers are in single phase without any impurity, and that both layers are preferentially oriented along the c-axis perpendicular to the surface of substrate. The full width at half maximum (FWHM) of the rocking curves, recorded around the fundamental (002) diffraction peak are less than 0.5° , confirming a good crystallinity for both BTO and LCMO layers.

In order to confirm the polarization (P) switching property for BTO layer in Ag/BTO/LCMO capacitors, we measured the dependence of switching current and ferroelectric polarization as a function of biased voltage with a frequency of 5 kHz at room temper-

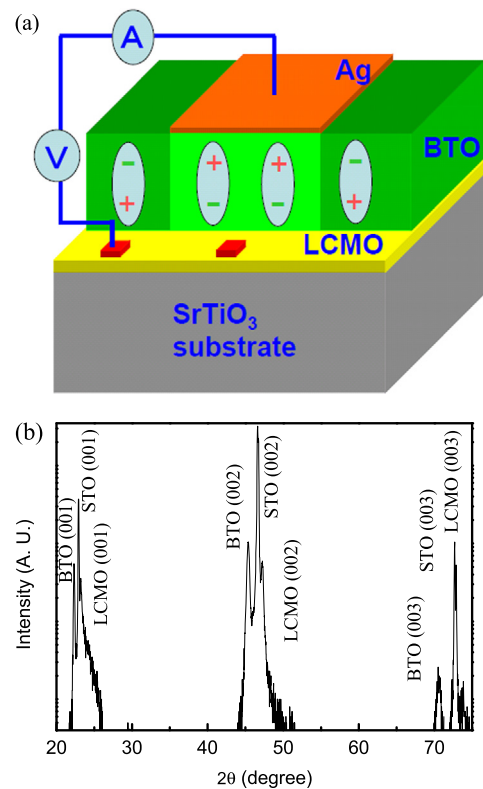


Fig. 1. (a) Schematic structure of Ag/BTO/LCMO heterostructure on SrTiO_3 substrate. (b) The typical XRD patterns of θ – 2θ scans in 20 – 80° for the heterostructure.

ature. As the biased voltage is ramped to 5 V, the displacement current exhibits a summit as shown in Fig. 2(a), demonstrating the occurrence of polarization switching in the BTO layer. It is known that the bias voltage can lead to a change in the surface charge of the sample. One contribution is from the ferroelectric domain switching that can be represented by the sharp increase of displacement current. As the polarization dynamics cease after a certain time, switching current subsides, and further change in the surface charge can be attributed to another contribution, i.e., the leakage currents through the BTO layer. If the BTO is not highly resistive and the leakage currents are large enough to overlay the ferroelectric domain switching current, the sharp increases in the displacement current cannot be observed. The forward and backward coercive voltage (around 3.7 V and -4.1 V) can be determined from the peak position of switching current. The coercive voltages are consistent with those values corresponding to voltage at which $\frac{dP}{dV}$ shows a maximum in the P–V loops (Fig. 2(b)). Both ferroelectric loops and clear peaks in switching current confirm a reversal of ferroelectric polarization in the BTO layer. Fig. 2(d) shows a representative R–T curve for an LCMO film with 30 nm. The R–T curve shows an insulator–metal transition at temperature ~ 233 K, and it exhibits typical semiconductor characteristics with a resistance $\sim 8 \text{ k}\Omega$ at room temperature. As a consequence, the charge carrier concentration in the LCMO can be calculated to be $\sim 10^{19} \text{ cm}^{-3}$ at room temperature.

Fig. 3 shows the typical I–V characteristics recorded with current by sweeping the biased voltage in temperature range from 100 K to 320 K. In all measuring loops the applied voltage follows $-5 \rightarrow 0 \rightarrow 5 \rightarrow 0 \rightarrow -5$ V cycles. As shown in Fig. 3(a), at low temperature (< 250 K) all I–V curves clearly show a reverse diode like rectifying characteristics, and leakage current values grow larger and larger as temperature increases. Such as at bias voltage of -4 V, the current value is $2 \times 10^{-8} \text{ A}$ at 100 K that is smaller than that of $1.3 \times 10^{-7} \text{ A}$ at 160 K, and $1.8 \times 10^{-6} \text{ A}$ at

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