

## The investigation of reversible strain and polarization effect in (011)-La<sub>0.9</sub>Ba<sub>0.1</sub>MnO<sub>3</sub> film using field effect configuration

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We have investigated the influence of the electric bias field on the magnetic and transport properties of (011)-oriented La<sub>0.9</sub>Ba<sub>0.1</sub>MnO<sub>3</sub> (LBMO) thin film epitaxially grown on (011)-0.7Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.3PbTiO<sub>3</sub> (PMN-PT) single crystal substrate. It was found that strain and polarization effects induced by electric bias coexist in the whole temperature range and both of them can modulate the transport properties of (011)-LBMO on PMN-PT. The relative change of resistance  $\Delta R/R$  exhibits peak values of  $-22\%$  and  $-32\%$  for bias fields of  $+12$  and  $-12$  kV/cm, respectively, around metal-insulator transition temperature,  $T_{MI}$ . However, the sign of  $\Delta R/R$  shows polarity dependence at temperature far below  $T_{MI}$ . Further careful analysis demonstrates that these two opposite behaviors can be ascribed to the different influence of strain and polarization effects on transport properties. © 2011 American Institute of Physics. [doi:10.1063/1.3545805]

### I. INTRODUCTION

The perovskite-type R<sub>1-x</sub>A<sub>x</sub>MnO<sub>3</sub> (R and A are trivalent rare-earth and divalent alkaline-earth ions, respectively) manganites have attracted a great deal of interest in recent years because of their various intriguing phenomena due to the interplay among spin, charge, orbital, and lattice degrees of freedom.<sup>1</sup> The manganite thin films are of special interest due to their advantages in practical applications. The biaxial strain due to lattice mismatch between film and substrate plays a very important role in controlling the properties of manganite thin films because of the strong electron-lattice coupling, especially for Curie temperature  $T_C$ , magnetic anisotropy, and transport properties.<sup>2-4</sup>

However, a thorough understanding of the strain effect is still unavailable and challenging, partially because the substrate-induced lattice strain is entangled with additional parameters (defects, microstructure, compositions). Recently, the electrically controlled strain from a piezoelectric substrate has been suggested as a versatile route to do the direct and quantitative measurements of strain dependent ferroic properties.<sup>5,6</sup> Taking advantage of the well inverse piezoelectric characteristic of the ferroelectric substrate, Thiele *et al.* have found the strain-induced shifts of Curie temperature  $T_C$  and enhancement of magnetization of La<sub>0.7</sub>A<sub>0.3</sub>MnO<sub>3</sub> films (A = Sr, Ca).<sup>7</sup> By applying dc electric fields across ferroelectric crystal and dynamically tuning the strain state, Zheng *et al.* also reported that the ferroelectric-poling-induced strain reduction gives rise to a decrease in the resistivity and an increase in Curie temperature, ferromagnetism, and magnetoresistance of (001)-oriented La<sub>0.875</sub>Ba<sub>0.125</sub>MnO<sub>3</sub> film.<sup>8</sup> On the other hand, the polarization effect should be taken into account since a ferroelectric substrate is used in such investigations. Wu *et al.* have reported a

large electroresistance,  $\sim 76\%$  at 40 kV/cm, in La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub> films due to the polarization of Pb(Zr, Ti)O<sub>3</sub> gate.<sup>9</sup> Thiele *et al.* found, in La<sub>0.7</sub>Sr<sub>0.3</sub>MnO<sub>3</sub>/Pb(Zr, Ti)O<sub>3</sub>, a thickness dependent competition between polarization effect and field-induced strain effect.<sup>10</sup> Furthermore, Sheng *et al.* found coactions of strain and polarization effects in La<sub>0.7</sub>Ca<sub>0.3</sub>MnO<sub>3</sub>/0.7Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.3PbTiO<sub>3</sub> structures.<sup>11</sup> These results indicate the field-induced polarization effect should also be considered in the studies of the field-induced strain effect and deserves to be investigated carefully. In this paper, we present the results of the competition between the electric-field-induced strain and the polarization effect in a field-effect-transistor (FET) structure containing ferroelectric (011)-0.7Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-0.3PbTiO<sub>3</sub> (PMN-PT) and lightly doped La<sub>0.9</sub>Ba<sub>0.1</sub>MnO<sub>3</sub> (LBMO) thin film. The inset of Fig. 1 shows the schematic FET structure.

### II. EXPERIMENTAL DETAILS

Although most investigations of field-induced strain effects are performed in (001)-oriented manganite films, previous reports have demonstrated that the piezoelectric coefficients with poling direction along [011] is nearly twice as much as that along [001] direction.<sup>12</sup> It is reasonable to expect a diverse behavior due to the field-induced strain and the polarization effect in (011)-films. Thus the (011)-oriented PMN-PT single crystal (coercivity field  $E_c \sim 3.6$  kV/cm) was chosen as the substrate. The LBMO film with thickness of 50 nm was grown on it using pulsed laser deposition. The stoichiometric target was ablated by KrF excimer laser (248 nm) with a pulsed energy of 200 mJ and a frequency of 2 Hz. The deposition took place at an oxygen pressure of 1 mbar and a substrate temperature of 750 °C. After deposition, in a pure oxygen atmosphere of 1 atm, the film was cooled to room temperature and then postannealed at 800 °C for 1 h. Au electrodes

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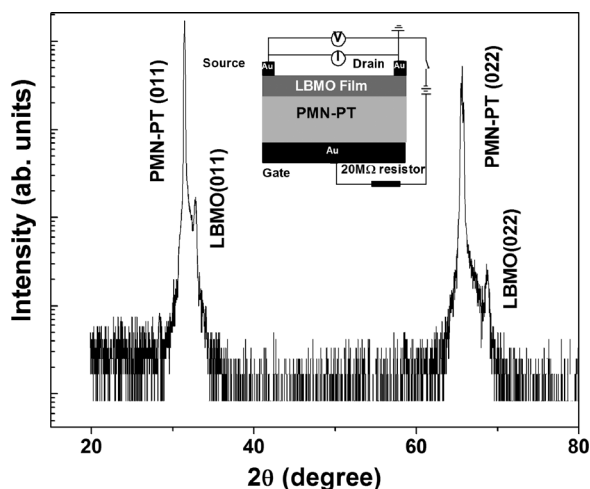


FIG. 1. X-ray diffraction patterns of (011)-oriented LBMO/PMN-PT film with thickness of 50 nm. (Inset) A schematic diagram of the field-effect transistor (FET) structure and the circuit for electrical measurements.

were vapor deposited on LBMO film and the back of PMN-PT substrates as the leads of source, drain, and gate of the FET structure, and a 20 MΩ resistor is presented in the circuit for protection (see the inset of Fig. 1)

Although the lattice constants ( $a \sim b \sim c \sim 4.017 \text{ \AA}$ ) of the PMN-PT substrate are larger than those ( $a \sim b \sim c \sim 3.902 \text{ \AA}$ ) of the LBMO bulk material,<sup>13</sup> previous reports<sup>14,15</sup> and our recent work<sup>16</sup> demonstrated that the epitaxial growth of manganite thin films on PMN-PT substrates is possible. The experiments of x-ray diffraction further verified such conclusion (see Fig. 1). An out-of-plane compressive strain of  $\sim -0.42\%$  (compared with the bulk one), due to the large lattice mismatch ( $\sim 3.31\%$ ), was found in the (011)-oriented LBMO film.

Magnetic and transport measurements were performed by a superconducting quantum interference device magnetometer with the magnetic field applied parallel to the [0-11] direction of the PMN-PT substrate. A static bias field up to  $\pm 12 \text{ kV/cm}$  was applied across the PMN-PT substrate (see the inset of Fig. 1) to change the poling state of PMN-PT and the corresponding strains in the LBMO film. The resistance of PMN-PT ( $\sim 10 \text{ G}\Omega$ ), measured using a Keithley 6517A electrometer, is much higher than that of the LBMO film ( $< 30 \text{ k}\Omega$ ), resulting in a negligible small leakage current (less than 15 nA under a 12 kV/cm electric field).

### III. RESULTS AND DISCUSSION

Figure 2 displays the temperature dependent resistivities of the LBMO film under different magnetic field of 0 and 5 T when the (011)-PMN-PT substrate is in the unpoled, negatively poled, and positively poled state, respectively. The poling electric field was set to  $\pm 12 \text{ kV/cm}$ . One could find that the resistivity of the film shows a clear metal-insulator transition near  $T_{MI} \sim 237 \text{ K}$  at the unpoled state. With the application of the electric bias on the structure of LBMO/PMN-PT, the resistivity decreases noticeably around the transition temperature, meanwhile  $T_{MI}$  slightly shifts to a higher temperature ( $T_{MI} \sim 241 \text{ K}$  for +12 kV/cm, while 243 K for -12 kV/cm). It has been reported that the field-induced strain of the piezoelec-

tric substrate is nearly symmetric with respect to the sign of the electric field,<sup>7,17,18</sup> while the polarization loop is antisymmetric.<sup>18</sup> This fundamentally different characteristic can be used to distinguish the modulation of transport properties from strain and polarization effects. The butterflylike resistance loop with respect to the field, i.e., the resistance decreases for both positive and negative fields even at the polarization saturation field of the piezoelectric substrate, is related to the field-induced strain effect.<sup>7,14</sup> In contrast, a polarity dependence of the resistance variation on the electric field, i.e., the resistance increases (decreases) at the positive (negative) electric field, reflects the influence of polarization.<sup>18</sup> In our case, the similar decrease of resistivity and  $T_{MI}$  under the bias field with opposite direction indicates that the bias-field-induced strain effect should be responsible for the transport properties around  $T_{MI}$ . The observed butterfly-like resistance loop at 250 K [see Fig. 2(a), inset] further confirms the strain origin.<sup>18</sup> Previous studies have demonstrated that the bias-field-induced compressive strain and biaxial strain in a piezoelectric substrate could transfer directly to the oxide films grown on it,<sup>17</sup> resulting in a reduction in the tensile strain in films. In the present case, such a reduction in the tensile strained LBMO film leads to a decrease of the JT distortion to some extent and delocalizes charge carriers somewhat, thus enhancing the electron hopping and double exchange (DE) interaction, resulting in a reduction in resistivity and an increase in  $T_{MI}$ .

On the other hand, one could also distinguish a clear enhancement for the resistivity, under a positive bias field, at temperatures far below  $T_{MI}$  [see Fig. 2(b), inset]. Such behaviors diverge from the feature of the modulation of resistance by the field-induced strain effect, indicating the dominant role of polarization within this temperature range.<sup>18</sup> To clarify the relation between the strain effect and the polarization effect in the whole temperature range, we present the relative change in resistance  $\Delta R/R (= [R(E) - R(0)]/R(0))$  as a function of

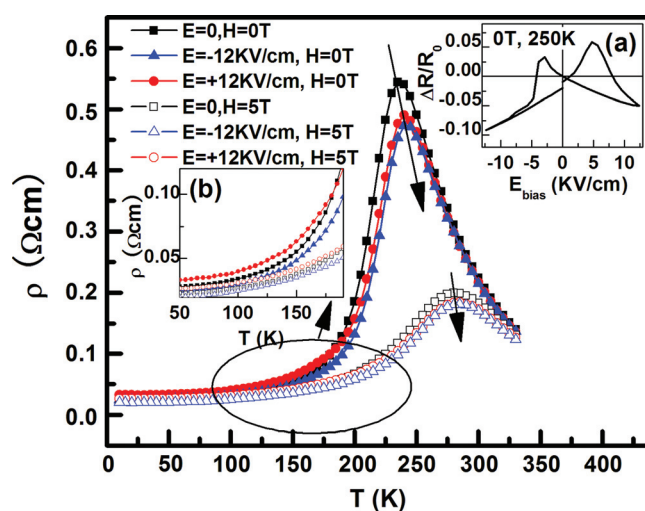


FIG. 2. (Color online) Temperature dependent resistivity of LBMO film under a magnetic field of 0 and 5 T when the (011)-PMN-PT substrate is in the unpoled, negatively poled, and positively poled state, respectively. The poling electric field was set to  $\pm 12 \text{ kV/cm}$ . (a, inset) The relative change in the resistance of the (011)-LBMO film as a function of the electric field applied to the PMN-PT substrate. (b, inset) The expanded view of the resistivity of LBMO/PMN-PT in the region of low temperature.

temperature, with bias field  $E = +12$  kV/cm,  $-12$  kV/cm, and magnetic field  $H = 0, 5$  T in Fig. 3. First, one could find that  $\Delta R/R$  peaks at a temperature a bit below  $T_{MI}$  for both polarities. The peak value is  $-22\%$  for the positive polarity and  $-32\%$  for the opposite, respectively. With the increase of temperature, the absolute value of  $\Delta R/R$  decreases above  $T_{MI}$ , independent of the sign of polarity. However, the sign of  $\Delta R/R$  is found to be polarity dependent with a small difference in the absolute value at low temperatures. These results indicate that the polarization effect derived from the ferroelectric PMN-PT crystal is dominant at low temperatures while the electric-field-induced strain effect plays a dominant role at high temperatures. Previous studies have indicated that a multiphase composed of partly insulating and partly metallic coexistence is common in doped manganites. The electric field would change the relative volume fractions of metallic and insulating regions by accumulating charges between metallic and insulating phases and move the interface.<sup>9,19</sup> In the present film of (011)-LBMO/PMN-PT, the polarization effect caused by a positive bias field will push the interface into metallic phase, leading to an increase of insulating fraction and thus an increase of resistance, while a negative field does the opposite, leading to a decrease of resistance. Careful analysis of the curves in Fig. 2 shows slight differences in the increase of  $T_{MI}$  ( $\sim 4$  K for positive polarity and  $\sim 6$  K for negative one) and also in the reduction of resistivity under different polarity of bias field. These small but distinct discrepancies indicate the coexistence of strain and polarization effect, although the strain effect dominates the modulation of transport properties around  $T_{MI}$ . Such different reactions to the direction of the bias field in different temperature range may be ascribed to the coactions and competition of the electric-field-induced strain effect and the polarization effect in the whole temperature range.<sup>11,18</sup>

The influence of the applied magnetic field on these two effects was also investigated and displayed in Figs. 2 and 3. It was found that a magnetic field of 5 T almost vanishes the difference in the transition temperature,  $T_{MI}$ , for positive and negative electric field ( $< 1$  K), which indicates the magnetic

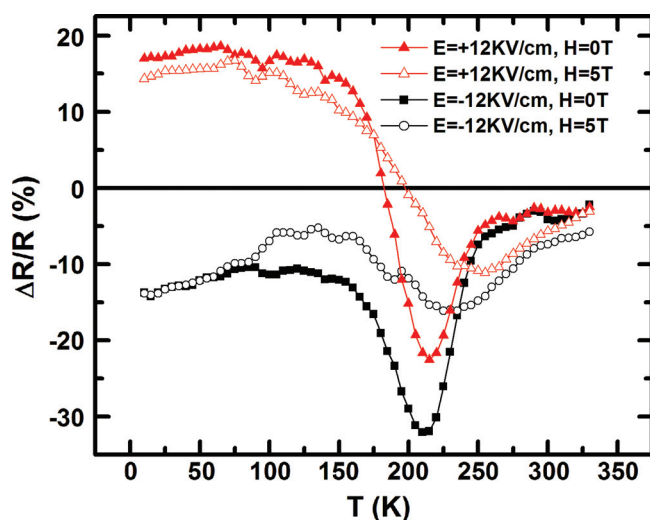


FIG. 3. (Color online) Relative change in resistance  $\Delta R/R$  ( $= [R(E) - R(0)]/R(0)$ ) as a function of temperature, with bias fields  $E = +12, -12$  kV/cm and magnetic fields  $H = 0, 5$  T.

field might suppress the polarization effect. Further, when a magnetic field of 5 T is applied, the absolute values of  $\Delta R/R$  due to the electric bias field increase in the temperature range above  $T_{MI}$  and decrease below  $T_{MI}$  (as seen from Fig. 3). These complete opposite responses of  $\Delta R/R$  to magnetic field indicate a different impact of the magnetic field on the polarization effect and the electric-field-induced strain effect. However, the underlying mechanism is still unclear at the moment and further investigations are under way.

#### IV. CONCLUSION

In summary, the influence of the electric field on magnetic and transport properties of (011)-LBMO/PMN-PT film has been studied. It was found that both electric-bias-induced strain effect and polarization effect can modulate the transport properties of LBMO/PMN-PT. The relative change in resistance  $\Delta R/R$  exhibits peak values of  $-22\%$  and  $-32\%$  for the positive and the negative bias field, respectively, around  $T_{MI}$ . However, the sign of  $\Delta R/R$  shows polarity dependence at temperatures far below  $T_{MI}$ . Further analysis indicates that the electric-field-induced strain effect is dominant around  $T_{MI}$ , while the polarization effect plays a dominant role at the temperature far below  $T_{MI}$ .

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