Coaction of electric field induced strain and polarization effects in $La_{0.7}Ca_{0.3}MnO_3$/PMN-PT structures

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The coaction of the electric field induced strain and polarization effect on magnetic and electrical properties in field-effect structures, composing of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) films and $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) crystal, have been studied. The strain state of the films can continuously be tuned by application of bias electric field. The resistance variation in LCMO film is about −4.34% and +3.48% when a negative (−8 kV/cm) and positive (+8 kV/cm) bias field is applied, respectively, at 50 K. With increasing temperature, the field-dependent shuttlelike loop of relative resistance changes to butterflylike hysteresis loop under the coaction of field induced strain and polarization effect. Moreover, a sharp, stable, and reversal magnetization controlling effect caused by electric field has also been found in the structure. The magnetization decreases as much as −8.64% when a positive field 9.6 kV/cm is applied at 10 K. The data clearly show that the electric field induced strain effect is dominant for the magnetization, while the polarization is dominant for the effect on electrical transport properties when the LCMO is in ferromagnetic metal state. These results reveal that the electric field can tune the lattice distortion and carry distribution artificially as well as magnetic and electrical properties by using manganite/PMN-PT structure.

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I. INTRODUCTION

The perovskite-type $R_{1-x}A_xMnO_3$(R and A are trivalent rare-earth and divalent alkaline-earth ions, respectively) manganites have received a great deal of interest in recent years because they show a wide variety of phases associated with an interplay among spin, charge, orbital, and lattice degrees of freedom.[1] Application of a slight external field such as magnetic field,[1] electric field,[2,3] photon,[4,5] or pressure[6] to a manganite which is located at the phase boundary will get huge and quick response. Among them, the electric field induced polarization effect on manganites has attracted much attention for its possible application in new field-effect devices. Recently, Pallecchi *et al.*[2] observed a shift of the metal-insulator transition temperature as high as 43 K and a resistivity modulation of up to 250% in $La_{0.7}Sr_{0.3}MnO_3$ side-gate channels. Hong *et al.*[7] found that the transport characteristics of LSMO vary sharply using the polarization field from a ferroelectric oxide, Pb(Zr,Ti)O_3. They showed us the clear depletion of holes in manganite films due to the polarization effect. A large electroresistance ~76% at 40 kV/cm was also found in $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) films with Pb(Zr,Ti)O_3 gate and explained by polarization effect on percolative phase separation.[8]

Besides the electric induced polarization effect, lattice deformations caused by some kinds of strain are another important factor to control the electronic states in manganites because of the strong electron-lattice coupling.[9] The commonly applied method for the study of the lattice strain effect is the growth of thin films on substrates with a certain lattice mismatch. The substrate-induced lattice strain, which could change the strength of the double-exchange (DE) interaction and Jahn-Teller (J-T) electron-lattice coupling via modifying Mn-O bond lengths and/or Mn-O-Mn bond angles, is one of the most important factors that strongly affect the properties of manganite thin films—e.g., Curie temperature (T_C), magnetoresistance, and phase separation. For example, the metal-insulator transition temperature T_{MI} of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) can be reduced from 365 K for unstrained films/bulk to 270 K in 16-nm-thick LSMO/LaAlO(001) films strained compressively.[10] Millis *et al.*[11] proved that the ferromagnetic T_C is extremely sensitive to biaxial strain. Gao *et al.*[12] found that the transition temperature is correlated with the strain state in epitaxial LCMO films. Generally, it is difficult to modulate the strain effect in manganite films continuously and reversely by using conventional substrates. To overcome this obstacle, some experiments, in which a ferroelectric crystal with electrostrictive or piezoelectric properties were used as substrate, have been done.[13–15] The strain-induced shifts of the T_C and enhancement of magnetization of $La_{0.7}A_{0.3}MnO_3$ films have been found.[15] Zheng *et al.* also reported that the charge-ordered state and paramagnetic state of $Pr_{0.5}Ca_{0.5}MnO_3$ film can be modulated continuously by applying dc or ac electric fields across ferroelectric crystal.[14] As a result of using ferroelectric crystal, the field-induced polarization effect should also be considered except field-induced strain effect. Hence, thorough knowledge of the coaction of these two factors benefits not only to clarification of the electric-induced effects in manganites but also to the designing of artificial materials. In this paper, the field-effect-transistor (FET) structure composing of a piezoelectric material, $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ [PMN-PT, with 30% of $PbTiO_3$ (PT)],[16] and LCMO films has been fabricated. The coaction of the electric field induced strain and polarization effect on electrical and magnetic properties of the structure have been studied carefully.

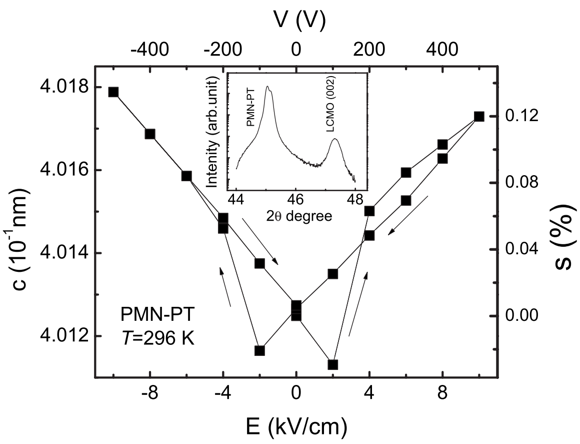

FIG. 1. The variation in the out-of-plane lattice constant c of a PMN-PT(001) crystal and strain s under an electric field E along the c axis. The inset shows the x-ray diffraction pattern (θ-2θ) at angles in the vicinity of the pesudocubic (002) reflection for LCMO/PMN-PT.

II. EXPERIMENTAL

The LCMO films were grown on the (001)-oriented single-crystal substrates of PMN-PT using pulsed laser deposition. The substrate temperature was 700 °C. The deposition took place in a pure oxygen atmosphere of the pressure of 0.5 mbar. The energy of the laser beam is ~400 mJ, the wavelength is 248 nm, and the pulse frequency is 3 Hz. The thickness of the film was about 90 nm, controlled by deposition time. The inset of Fig. 1 shows a typical θ-2θ x-ray diffraction (XRD) pattern of the LCMO/PMN-PT structures. Despite a lattice misfit of +4% between PMN-PT (4.02 Å) compared to the pseudocubic lattice parameter of 3.863 Å of LCMO, the films grow in cube on cube epitaxy, as considered from the (103) phi pattern of LCMO film (not shown here). The electrical transport properties were measured in the temperature range of 20–300 K using a closed-cycle refrigerator. To provide ohmic contact, Ag electrodes are evaporated on film surface by lithography mask. The magnetization of the sample was measured on a Quantum Design superconducting quantum interference device (SQUID) magnetic property measurement system ($1.9 \le T \le 400$ K, $0 \le H \le 5$ T).

III. RESULTS AND DISCUSSION

X-ray diffraction measurements (θ-2θ) performed in LCMO/PMN-PT exhibit the response of the c lattice parameter of PMN-PT(001) crystal under applied electric field. The typical results are shown in Fig. 1. It is found that the c constant depends on a nearly linear way on the bias electric field. It increases by about 0.12% with an electric field of $E=10$ kV/cm ($V_{\mathrm{bias}}=500$ V). The relation between lattice strain s and E indicates a converse piezoelectric nature of the crystal. More precisely, it can be found that the slope of strain decreases slightly with increasing voltage. Cycling the field from positive to negative values, a butterflylike hysteresis loop with coercive fields of 2 kV/cm($V_{\mathrm{bias}}=100$ V) is observed. Assuming approximate volume conservation, 0.12% of out-of-plane expansion would be accompanied by 0.06% of contraction in both in-plane directions of the PMN-PT(001) single crystal.[15,17] If we consider that the in plane of LCMO film is compressed due to the contraction strain coming from PMN-PT substrate, the in-plane distortion ratio $\varepsilon_{xx}=\varepsilon_{yy}$ should be −0.06% and the out-of-plane distortion ratio ε_{zz} is about +0.043% using $\varepsilon_{xx}=-2\nu/(1-\nu)\ \varepsilon_{zz}$ and Poission coefficient $\nu \sim 0.41$.[18,19]

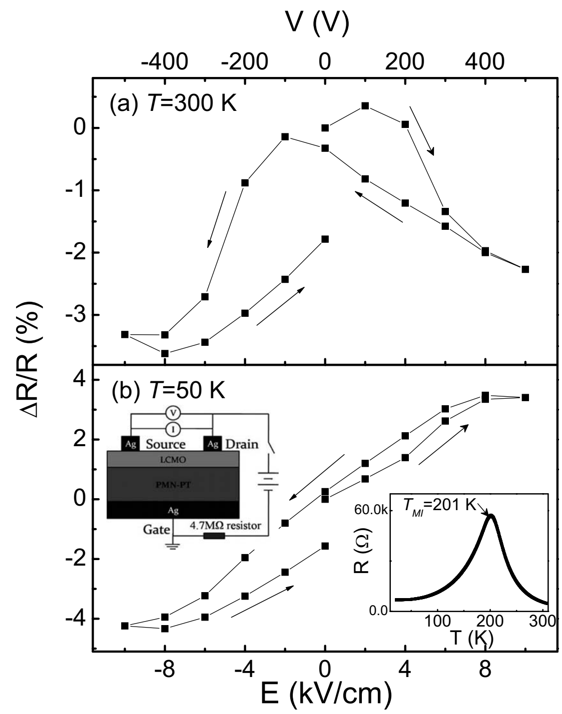

FIG. 2. Resistance hysteresis of the LCMO film vs bias electric field at (a) 300 and (b) 50 K. The left inset of (b) is the schematic for the LCMO-PMNT structure in FET configuration and the circuit for electrical measurements. The right inset of (b) shows the typical temperature dependence of electrical resistance $R(T)$ of LCMO film.

The temperature dependence of the resistance of LCMO film is plotted in the right inset of Fig. 2(b). A metal-insulator transition at $T_{\mathrm{MI}}=201$ K was observed. The electric field control of electrical properties in LCMO films was measured in the FET configuration as shown in the left inset of Fig. 2(b). The resistance of LCMO was measured through source and drain electrodes and the bias electric voltages were applied to the LCMO/PMN-PT structure through the gate and drain. A resistor of 4.7 MΩ was connected in series with the gate in order to protect the electrical meters in case a dielectric breakdown took place in the PMN-PT crystal. Results of transport measurements of LCMO film under bias electric field are shown in Fig. 2. The resistance R at room temperature drops by about −3.61% $\{\Delta R/R=[R(E)-R(0)]/R(0)\}$ at $E=-8$ kV/cm due to the electric-induced in-plane strain in the film. (The positive bias is defined as when the electrode on the PMN-PT gate is positive while the electrode on LCMO is negative.) The change in resistance with variation in V is not linear. It is similar to the strain coefficient s shown in Fig. 1 that a butterflylike hysteresis loop with coercive

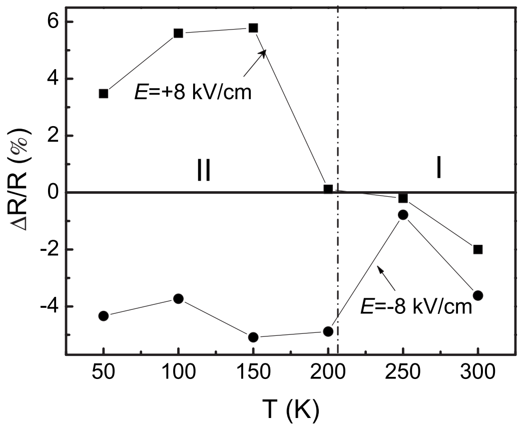

FIG. 3. The relative change in resistance $\Delta R/R$ as a function of temperatures with bias electric field $E=+8$ and -8 kV/cm, respectively.

fields of 2 kV/cm is also observed when we cycle the electric field from the positive to negative values. Basing on the theory of strain effect on manganite films, it is easy to understand this butterflylike behavior as follows. In the simplest scenario, two aspects caused by strain in manganite films can be envisaged: a uniform compression will tend to increase the bare electron-hopping amplitude and thereby reduce the importance of the electron-lattice coupling, at the same time a biaxial distortion will also compete with the J-T distortion and lattice distortion caused by tolerance factor, thus affecting the properties of manganite films especially for magnetic structure (it will be discussed later). Due to the first aspect, the enhancement of electron-hopping amplitude will contribute to the decreasing of resistance as shown in Fig. 2(a).

As temperature decreases, an interesting phenomenon occurs as shown in Fig. 2(b). In the negative branch, the resistance of LCMO film decreases with increasing negative bias voltage. In the positive branch, however, the resistance increases with increasing positive bias voltage, which is quite different from that of high temperatures. As we can see in Fig. 2, the butterflylike hysteresis loop changes to shuttlelike loop with decreasing of temperature. At 50 K, the $\Delta R/R$ is about +3.48% when the electric field E is +8 V/cm, while it reaches −4.34% as $E=-8$ V/cm. The difference reaction to the direction of bias voltage in different temperature can be clearly reflected in Fig. 3. The relative change in resistance $\Delta R/R$ as a function of temperatures, as bias field $E=+8$ and −8 V/cm, respectively, was depicted in this figure. It can be found that the resistance decreases no matter what the direction of bias electric field at high temperature. We named this area as region I for simplicity reason. The change in resistance of the LCMO film at low temperature, however, has a tight relation with the direction of bias field as shown in region II. It is interesting that the transition point from region I to region II is located near $T=200$ K which is close to the metal-insulator transition temperature and Curie temperature ($T_{MI}=201$ K, $T_C=203$ K). These results imply that the transition from field-induced butterflylike hysteresis loop to shuttlelike behavior might has an intimate relation with the electrical and magnetic state of the LCMO films.

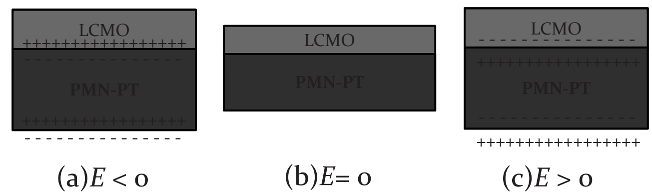

FIG. 4. Schematic picture of coaction of the electric field induced strain and polarization effect on the LCMO/PMN-PT structure.

If only the electric field induced strain effect is considered, the shutterlike loop of the relative change in resistance as a function of electric field cannot be explained.[11,12] It is difficult for the piezoelectric effect to explain the different behavior of LCMO films under different directional bias field. It should be noted that the electric field cannot only induce the piezoelectric effect but also polarize the LCMO film because of using the ferroelectric crystal PMN-PT. Therefore, the bias electric field applied in the FET configuration not only results in the expansion of the lattice along the direction of the electric field but also cause the depletion or accumulation of holes in the LCMO films layer as shown in Fig. 4. It is known that the LCMO is a p-type material, the majority of the charge carriers in the LCMO films are holes. The application of a positive (or negative) field to the gate will polarize the PMN-PT layer such that the electric-dipole moments in the PMN-PT point upward (or downward) into the LCMO film, thereby leading to a depletion (or accumulation) of holes in the interface part of LCMO film as shown in Figs. 4(a) and 4(c). If the polarization effect caused by external bias is taken into account, the depletion of holes will happen in the LCMO film within screen length with positive bias as shown in Fig. 4(c). However, the screen length in LCMO films is very short because of the high carrier density of the LCMO films with doping lever $x=0.3$.[7,8] The applied electric field will be screened within one or two lattice constants of the interface and could not affect bulk transport of the film. In our experiments, therefore, another process may happen and it allows the electric field induced effect to penetrate more deeply into the film. The possible process for the polarization effect observed in our experiments may be relevant to the phase separation of the LCMO films. Recent experimental and theoretical studies implied that the doped manganites are intrinsically inhomogeneous and that phase separation is common in these materials.[20,21] The phase separation and percolative transport properties in LCMO films have also been proven.[8] In an electronically inhomogeneous channel, they are partly metallic and partly insulating. The application of high electric field would change the relative volume fractions of metallic and insulating regions by accumulation of charge between metallic and insulating phases, thereby causing the interface between these phases to move.[7,8] The nature of accumulating charge, holes, or electrons will depend on field polarity and thereby control the direction of interface movement. The field effect on the transport properties, therefore, is also field-polarity dependent. In our case, the polarization effect on phase separation as well as percolative transport caused by positive external bias will make the resistance increase as shown in Fig. 2(b).

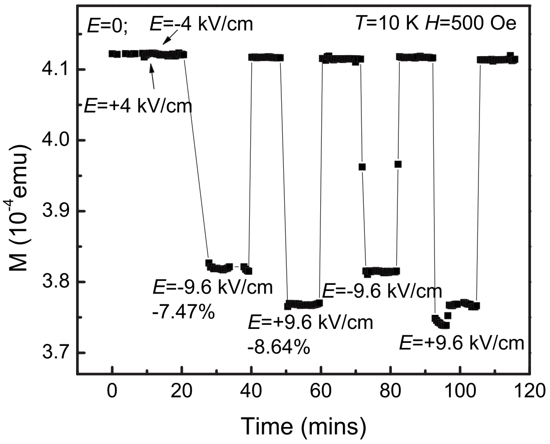

FIG. 5. Magnetization of the LCMO/PMN-PT structure as a function of time measured at 10 K with a magnetic field of 500 Oe.

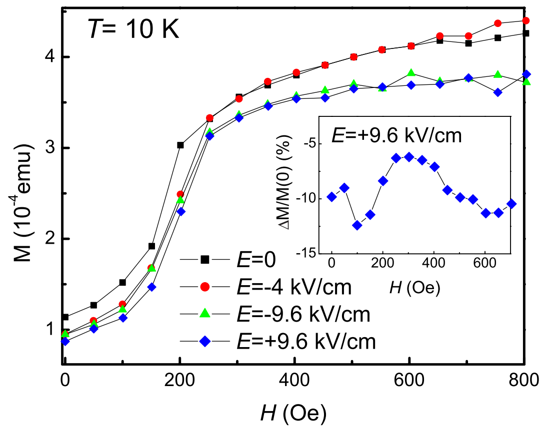

FIG. 6. (Color online) Magnetic field dependence of the magnetization of the LCMO/PMN-PT structure with selected bias electric field $E=-4$, -9.6, and $+9.6$ kV/cm.

A negative voltage does the opposite, leading to a decrease in resistance. Considering the coaction of field-induced strain and polarization effect together, we can understand the behavior that the absolute value of $\Delta R/R$ under negative bias (4.34%) is larger than that of positive bias (3.48%) at $T=50$ K.

In the Fig. 3, it can be found that the relative change in R has a maximum value near the temperature of 150 K. This value is consistent with the temperature at which the resistance of LCMO films has a steeper slope as shown in inset of Fig. 2(b). That is to say the modulation ΔR is clearly larger at temperature where the change in R is steeper. This feature is another evidence for the percolative transport in LCMO films with multiphase. Another point in Fig. 3 should be noted that the polarization effect derived from ferroelectric PMN-PT crystal is dominant at lower temperatures and negligible at higher temperature $T>T_{MI}$. It reveals that the effect on phase separation of LCMO film caused by PMN-PT polarization is more effective at temperatures below the T_{MI}. This point also agrees with the feature that the phase separation is significant at lower temperature in manganites film.[20]

To test whether there is any influence on magnetization under coaction of strain and polarization effect, the measurements of magnetization with and without external-bias electric field have been done. The data were obtained by monitoring the in-plane magnetic response to a voltage applied between the film and substrate bottom. Figure 5 exemplifies the response of magnetization of LCMO/PMN-PT structure to the external-bias field measured at a selected temperature $T=10$ K. When a $E=+9.6$ V/cm bias is applied, a sizeable decrease in the magnetization is observed, and the magnetization shows a quick switch between the original state and lower magnetization state corresponding to the "bias on" and "bias off." The lower magnetization state is stable for a constant bias voltage, unaffected by the repeated bias voltage on and off. The relative variation $\Delta M/M(0)$ [defined as $[M(V)-M(0)]/M(0)$] is about -8.64%. As for the negative bias field $E=-9.6$ V/cm, the relative variation is about -7.47% which is smaller than that of the positive bias. Before the application of larger bias field, the bias field $E=\pm 4$ kV/cm have been added to the LCMO/PMN-PT structure in order. However, there is no obvious response as shown in the left side of Fig. 5 and it implies that the electric field $E\leq\pm 4$ kV/cm is insufficient to modulate the magnetic domains in LCMO films. This behavior is more clearly shown in Fig. 6 in which the magnetic field dependence of magnetization with selected bias field $E=-4$, -9.6, and $+9.6$ kV/cm were shown. We observed that the M-H curves with $E=0$ and -4 kV/cm, respectively, are almost coincident and there is an obvious gap when the bias field reaches ± 9.6 kV/cm as the magnetic field is more than 300 Oe. The inset of Fig. 6 presents the typical results of $\Delta M/M(0)$ as a function of magnetic field with fixed bias field $E=+9.6$ kV/cm. It shows that the magnitude of $\Delta M/M(0)$ varies with magnetic field but has no clear dependence law with a fixed bias field.

It has been known that each orbit of Mn ion in manganite films has a different anisotropy of the wave function and it is coupled to the displacement of the O atoms surrounding the transition-metal ion which has a tight relation with strain. Moreover, the epitaxial strain could induce anisotropic hopping between orbits to cause orbit ordering at the interface. By varying the strain condition, the orbital ordering changes which in turn changes the magnetic ordering at the interface. For example, the energy of the $d_x^2-y^2$ orbit becomes higher than the $d_{3z}^2-r^2$ orbit in Mn-O octahedral when the two apical O atoms move away from the Mn ion.[22] As a result, the C-type antiferromagnetic order would occur in LCMO films if the compressive strain on the ab plane is strong enough. This point has been proven by many experiments.[23,24] As stated above, except the influence on the average Mn-O-Mn bond distance and angles, the compressive strain can also compete with the J-T distortion and lattice distortion caused by tolerance factor and thus magnetic structure of the LCMO film. Therefore, the field-induced strain effect in our experiments could affect the lattice distortion of LCMO films and thus contribute to the decreasing of magnetization. Theoretical and experimental studies have revealed that the coexistence of multiple phases [e.g., ferromagnetic (FM) and antiferromagnetic orbital order (AFM-OO)] and mutual interaction between them are very important in manganites as stated before.[20,21] The electric field induced compressive strain and biaxial strain favor the formation of AFM-OO as well as changing the relative volume fraction of FM and AFM-OO region, and then the magnetization decreases. Although the strain in films is nonuniform because the elastic strain in the film decreases with increasing thickness, we can also use the strain value to denote the average strain state of films. The bulk and biaxial strain are determined by $\varepsilon_B = 1/3(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})$ and $\varepsilon_{bi} = 1/4(2\varepsilon_{zz}-\varepsilon_{xx}-\varepsilon_{yy})$, respectively.[11] In our case, the bulk and biaxial strain is about −0.026% and +0.051%, respectively, at $E=9.6$ kV/cm. That is to say the 0.051% biaxial strain will cause more than 8% decrease in magnetization in LCMO/PMN-PT structures. Besides strain effect, the polarization effect induced by the electric field should also be employed to discuss the field-direction dependence of $\Delta M/M$. It is known that the charge accumulation or depletion has some impact on the magnetization in manganite. In LCMO compounds, the increasing effective hole density near the interface will be the benefit of the enhancement of magnetization as the doping level is smaller than 0.33.[1] Moreover, the accumulation or depletion caused by the polarization effect could also affect the relative volume fractions of FM and AFM-OO phase in LCMO film as discussed before. Hence, the magnetization of LCMO film with negative bias field will be higher and robust than that with positive bias. Our results shown in Fig. 5 are consistent with this point. The sharp, stable, and reversal magnetization controlling effect caused by electric field observed here reveals that the electric field can tune the lattice distortion and effective carrier density artificially as well as magnetic structure by using the manganite/PMN-PT structure. The change in magnetization due to the coaction of the applied electric field induced strain and polarization effect observed here may be of practical interest. The magnetoelectric coupling coefficient $\alpha = \mu_0 \Delta M/\Delta E$ for the LCMO/PMN-PT structure is about 3.4×10^{-8} sm^{-1} which is similar to the results reported by Thiele *et al.*[15] and smaller than obtained in the $La_{0.67}Sr_{0.33}MnO_3/BaTiO_3$ heterostructure.[25] Basing on the discussion above, one can deduce simply that the coupling between the magnetic and lattice degrees of freedom is about −0.3285 emu ($-0.1512\mu_B$/Mn), and the coupling between the magnetic and electric degrees of freedom is about 0.0255 emu ($0.011\mu_B$/Mn) in our LCMO films under the action of electric field, 9.6 kV/cm.

In the results shown above, one point worthy of special attention is that the electric field induced strain effect is dominant for the magnetization at low temperature, while the polarization is dominant for the electrical transport properties as shown in Fig. 2(b). This point implies that the magnetic structure of LCMO films is more sensitive to the local distortion of Mn-O octahedral than to the change in the effective carrier density. Although the magnetization drops about 8%, the LCMO is still in ferromagnetic metal state and the decreasing magnetization has little influence on the double-exchange effect and percolative behavior as well as the electrical transport process. These features are consistent with previous results.[1,26] As mentioned in those works, the resistivity of manganites ρ varies with carrier density n and changing of magnetization ΔM in an empirical relation of $\rho \sim 1/n \exp[-\Delta M/M_s]$, where M_s is the saturation magnetization. This relation reveals that the resistivity varies faster with the change in the effective carrier density n than with the relative change in magnetization ΔM as $\Delta M/M_s < 1$. In this work, we only give a possible explanation for the observed field control of the magnetic and electrical properties of LCMO/PMN-PT structures. To understand the coaction of strain and polarization effect comprehensively, further experiments about the electron-lattice coupling of the manganite films are required.

IV. CONCLUSION

In summary, the electric field control electrical and magnetic properties of LCMO/PMN-PT structures have been studied. It is found that the variation in resistance of the LCMO film is about −4.34% and +3.48% when a −8 and +8 kV/cm bias voltage is applied, respectively, at 50 K. However, the field-dependent shuttlelike loop of the relative resistance at $T < T_{MI}$ changes to butterflylike hysteresis loop when $T > T_{MI}$ under coaction of field induced strain and polarization effect. Moreover, a sharp, stable, and reversal magnetization controlling effect caused by electric field has also been found in the structure. Our results indicate that the field-induced strain and polarization effect both can modulate the electrical and magnetic properties of manganite films. As for field effect on transport properties, the polarization effect is dominant at $T < T_{MI}$ while the strain effect is dominant at $T > T_{MI}$. Moreover, we also observed that the electric field induced strain effect is dominant for the magnetization when the LCMO is in ferromagnetic metal state, while the polarization effect is dominant for the electrical transport properties.

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