Converse piezoelectric control of the lattice strain and resistance in $Pr_{0.5}Ca_{0.5}MnO_3$/PMN-PT structures

R. K. Zheng,[1,*] C. Chao,[1] H. L. W. Chan,[1] C. L. Choy,[1] and H. S. Luo[2]
[1]*Department of Applied Physics and Materials Research Center, The Hong Kong Polytechnic University, Hong Kong, China*
[2]*State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 201800, China*
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Thin films of the charge-ordered $Pr_{0.5}Ca_{0.5}MnO_3$ (PCMO) compound have been deposited on (001)-oriented ferroelectric $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$ $(x\sim 0.3)$ (PMN-PT) single-crystal substrates. The resistance of the PCMO films in the charge-ordered state and paramagnetic state can be modulated by applying dc or ac electric fields across the PMN-PT substrates. Piezoelectric displacement and x-ray diffraction measurements indicate that the electric field induces lattice strains in the PMN-PT substrates via the converse piezoelectric effect, which subsequently changes the strain state and resistance of the PCMO films. Quantitative relationships between the resistance of the PCMO films and the electric-field induced lattice strains in the PMN-PT substrates have been established, which would be important for designing converse piezoelectric effect based functional devices.

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I. INTRODUCTION

Recently, perovskite-type manganites have received great attention due to a variety of interesting properties such as colossal magnetoresistance, charge ordering, and electronic phase separation.[1] Among all the properties of manganites, the charge ordering, an ordering of charge carriers in two different Mn sublattices below the charge ordering transition temperature T_{CO}, is one of the most fascinating phenomena that have been studied extensively in bulk and thin films.[2–9] The half-doped $Pr_{0.5}Ca_{0.5}MnO_3$ (PCMO) compound is a prototype manganite exhibiting charge-ordered (CO) state characterized by the charge and orbital ordering below T_{CO} ($\sim$240 K).[2] Recent studies have shown that the resistance and stability of the CO state in the PCMO films are very sensitive to substrate-induced lattice strain (either compressive or tensile). Using electrical transport and electron microscopy, Prellier *et al.*,[3–5] Ogimoto *et al.*,[6] and Yang *et al.*[7,8] have found that the substrate-induced tensile strain has a strong stabilizing effect on the CO state in the PCMO films. In contrast, Nelson *et al.*[9] reported that the CO state in the $Pr_{0.6}Ca_{0.4}MnO_3$ films becomes more robust when the films are under compressive strain. Moreover, they found that the tensile strain disfavors the formation of CO state in the $Pr_{0.6}Ca_{0.4}MnO_3$ films grown on $NdGaO_3$ substrates. On the other hand, Biswas *et al.*[10] found that the substrate-induced compressive strain can induce CO phase in the $La_{0.67}Ca_{0.33}MnO_3$ thin films at low temperatures. These results suggest that the compressive strain stabilizes the CO state. Thus, the effects of substrate-induced lattice strain on the resistance and stability of CO state in charge-ordered compounds, e.g., $Pr_{1-x}Ca_xMnO_3$, are quite complex and far from fully understood.

Strained thin films are usually obtained by growing thin films with different thicknesses on substrates with certain lattice parameters. However, it should be noted that, besides the substrate-induced lattice strain, some other extrinsic factors, e.g., oxygen nonstoichiometry, crystallinity, strain relaxation, grain sizes and boundaries, growth related disorder, and defects, also have significant influences on the properties of thin films.[11–15] Thus, it is quite difficult to isolate property changes due to substrate-induced lattice strain from those resulting from above mentioned extrinsic factors. An intrinsic study of the substrate-induced lattice strain effect requires that the lattice strain in a substrate could be *in situ* changed systematically while aforementioned extrinsic factors are kept constant.

The PMN-PT single crystals are perovskite ferroelectric materials showing excellent piezoelectric activity.[16–18] If the PMN-PT single crystals are used as substrates to grow perovskite thin films, the lattice strain in the PMN-PT substrates and the subsequent strain state of thin films can be changed by the converse piezoelectric effect of the PMN-PT. Using this method, the above mentioned extrinsic factors can be kept constant while the lattice strains in the PMN-PT substrates and thin films can be changed systematically by applying electric fields across the PMN-PT substrates.

In this paper, we have studied the electrical properties of the PCMO thin films grown on the PMN-PT single-crystal substrates. We manipulated the strain state and resistance of the PCMO films by *in situ* inducing lattice strains in the PMN-PT substrates via the converse piezoelectric effect, and quantified the relationships between the resistance of the PCMO films and the electric-field induced in-plane and out-of-plane strains in the PMN-PT substrates.

II. EXPERIMENTAL

PMN-PT single crystals were grown by a modified Bridgman technique at the Shanghai Institute of Ceramics.[18] The single crystals were cut into plates of dimensions $10\times 3\times 0.55\ mm^3$ and with the plate normal in the $\langle 001\rangle$ crystal direction and polished until the average surface roughness was less than 1 nm. The PCMO films were deposited on the (001)-oriented PMN-PT substrates by dc magnetron sputtering in an argon-oxygen flow $(Ar:O_2=1:1)$ at a pressure of

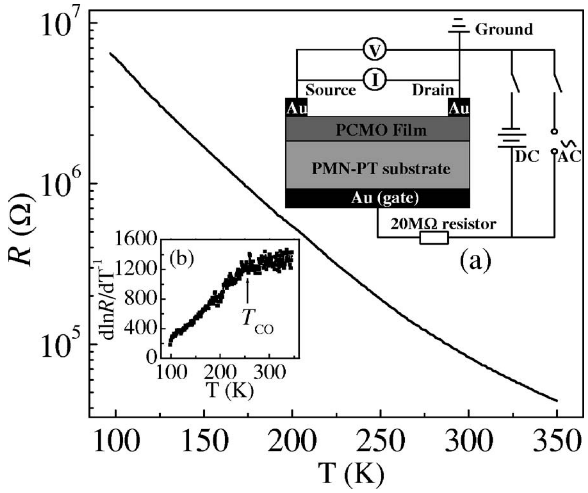

FIG. 1. Temperature dependence of the resistance for the PCMO film. Inset (a) is a schematic diagram showing the PCMO/PMN-PT structure and the circuit for electrical measurements. Inset (b) shows the $d \ln R/dT^{-1} - T$ curve for the PCMO film.

4.35 Pa and at a substrate temperature of 700 °C. After deposition, the PCMO films were slowly cooled to room temperature *in situ* and postannealed in air at 700 °C for 30 min.

X-ray diffraction (XRD) measurements using a Bruker D8 Discover x-ray diffractometer equipped with Cu $K\alpha$ radiation show that the PCMO films are single phase, c-axis preferentially oriented, and of good crystallinity. Low frequency (1 Hz) ferroelectric measurements using a standard Sawyer-Tower cicuit indicate that the polarization-electric field hysteresis loops of the PMN-PT substrates are square shape with a remnant ferroelectric polarization of $\sim 25\ \mu C/cm^2$. The longitudinal piezoelectric coefficients (d_{33}) of the PMN-PT substrates were measured using a Berlincourt-type quasistatic d_{33} meter while the transverse piezoelectric coefficients (d_{31}) of the PMN-PT substrates were measured by a resonance technique using a HP4294A impedance analyzer. Piezoelectric vibration of the PMN-PT substrates was generated by applying dc/ac electric voltages across the PMN-PT substrates. The longitudinal lattice displacements of the PMN-PT substrates due to the converse piezoelectric effect were measured using a modified double-beam laser interferometer.

The resistance of the PCMO films was examined in the ferroelectric field effect transistor configuration schematically shown in the inset (a) of Fig. 1. External electric voltages were applied to the PMN-PT substrates through the gate and the drain. A 20 MΩ resistor was connected in series with the gate in order to protect the current source and voltage meters in case a dielectric breakdown took place in the PMN-PT substrates. The gate leakage current was measured by applying electric voltages to the PMN-PT substrates through the gate and the drain using a Keithley 6517A Electrometer. The temperature dependence of the resistance for the PCMO film is shown in Fig. 1. The resistance increases with decreasing temperature from 350 K to lower temperatures, consistent with previous reports.[4–6] The $d \ln R/dT^{-1} - T$ curve shows a change in slope near 255 K, implying that the charge ordering phase transition occurs near that temperature.

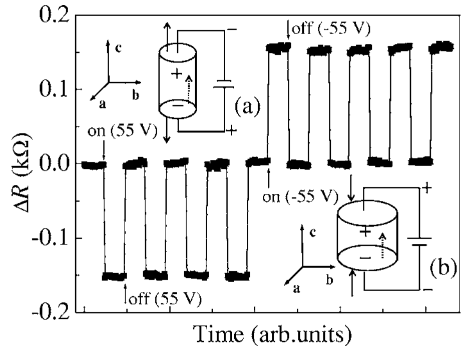

FIG. 2. Resistance response of the PCMO film at 296 K as a function of time when ±0.1 kV/mm dc electric fields (i.e., ±55 V electric voltages) are switched on and off. Inset (a) and (b) are schematic diagrams showing the electric-field induced expansion and contraction of the lattice of the PMN-PT substrates via the converse piezoelectric effect, respectively. The dotted arrows represent the direction of polarization.

III. RESULTS AND DISCUSSION

It is known that only polarized ferroelectric materials possess the piezoelectric effect and the converse piezoelectric effect. Therefore, before the measurements of the resistance of the PCMO film, we applied a dc electric voltage of +550 V (corresponding to an electric field of $E = +1$ kV/mm, much larger than the coercive field of the PMN-PT) across the PCMO/PMN-PT structure at 140 °C in a silicone oil bath so that the PMN-PT was positively polarized (i.e., electric dipole moments in the PMN-PT substrate point toward the PCMO film). After poling for 15 min at that temperature the PCMO/PMN-PT was slowly cooled to room temperature in the presence of the electric field. Such polarized PMN-PT has a longitudinal piezoelectric coefficient $d_{33} \sim 1500$ pC/N (or pm/V) at room temperature, indicating that the PMN-PT substrate has been well polarized.

Figure 2 shows that the resistance (ΔR) of the PCMO film at 296 K was repeatedly modulated by applying a positive (negative) 0.1 kV/mm dc electric field to the PMN-PT substrate. Note that the resistance has been normalized to that for $E = 0$ kV/mm, i.e., $\Delta R = R(E) - R(0)$ where $R(E)$ and $R(0)$ are the resistance of the PCMO film under electric field and zero electric field, respectively. The resistance decreases sharply when a +0.1 kV/mm electric field has been switched on and returns to its initial value upon the removal of the electric field. In contrast, the resistance increases sharply when a negative (−0.1 kV/mm) electric field has been switched on. The resistance also returns to its initial value when the electric field has been turned off. To further examine the resistance change due to the application of the electric field to the PMN-PT substrate, we measured the resistance response of the PCMO film as a function of time by applying sinusoidal and triangular ac electric fields to the PMN-PT substrate, respectively, and the results are shown in Fig. 3. Once again, the resistance has been normalized to that for

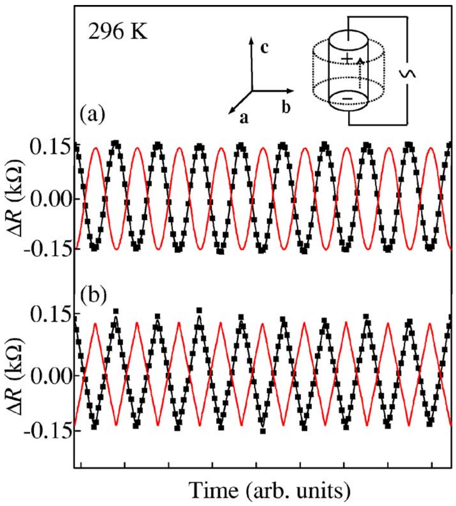

FIG. 3. (Color online) Resistance response of the PCMO film at 296 K as a function of time when peak-to-peak 0.2 kV/mm sinusoidal and triangular ac electric fields are applied to the positively polarized PMN-PT substrate, respectively. The red (lighter) curves represent the sinusoidal and triangular ac electric fields. Inset is a schematic diagram showing the ac electric-field induced vibrations of the lattice of the PMN-PT substrate via the converse piezoelectric effect. The dotted arrow represents the direction of polarization.

$E=0$ kV/mm. It can be seen that the resistance of the PCMO film was modulated with the same frequency as that of the sinusoidal/triangular ac electric field. Nevertheless, the phase difference between ΔR and the ac electric field is π, which means that the positive electric field induces a decrease in the resistance while a negative electric field induces an increase in the resistance. The relationship between ΔR and the electric field (E) can be described by $\Delta R=-aE$ where a is a positive constant and the units for ΔR and E are kΩ and kV/mm, respectively. This indicates that the change in the resistance of the PCMO films is proportional to the electric fields applied to the PMN-PT substrates.

It is noted that the gate leakage current may influence the resistance of the PCMO film. We have measured the gate leakage current and found that it is less than 0.5 nA under a 0.1 kV/mm electric field. Such a small leakage current is negligible as compared with the source/drain current (5 μA). Thus, the electric-field induced modulation of the resistance of the PCMO film cannot be explained by the leakage current. On the other hand, the PMN-PT substrate had been well polarized before the measurements of the resistance of the PCMO film when the PMN-PT substrate was subjected to the 0.1 kV/mm dc or ac electric field. This field is far lower than the coercive field ($E_C \sim 0.32$ kV/mm) of the PMN-PT substrate. Moreover, the measurements of the resistance were made at 296 K which is about 120 K lower than the Curie temperature of the PMN-PT substrate. Thus, the application of 0.1 kV/mm electric field at 296 K cannot change the polarization state of the PMN-PT substrate. For this reason, it is concluded that the modulation of the resistance is not due to the charge injection (or depletion) caused by the ferroelectric field effect.

Ferroelectric materials exhibit the piezoelectric effect and the converse piezoelectric effect after they have been polarized, that is, electric charges appear on the surface of polarized ferroelectric materials when they are subjected to mechanical deformations and conversely, when electric fields are applied to polarized ferroelectric materials, strains is induced in the materials.[19] If the converse piezoelectric effect is considered, the electric-field induced modulation of the resistance can be explained as follows. When a positive 0.1 kV/mm electric field is applied to the positively polarized PMN-PT substrate, the lattice of the PMN-PT substrate will expand along the c axis and contract perpendicular to that direction, as schematically shown in the inset (a) of Fig. 2. Such changes in the lattice of the PMN-PT will subsequently impose in-plane compressive strains on the PCMO film with respective to the strain state of the PCMO film for $E=0$ kV/mm, causing a decrease of the Jahn-Teller electron-lattice coupling and an enhancement of the double-exchange interaction in the PCMO film.[20–22] Consequently, the resistance of the PCMO film decreases. When the electric field is switched off, the lattice of the PMN-PT substrate restores to its initial positively polarized state. Accordingly, the strain state and resistance of the PCMO film return to their initial values. In contrast, when a negative electric field (−0.1 kV/mm) is applied to the positively polarized PMN-PT substrate, the lattice of the PMN-PT will contract along the c axis and expand perpendicular to that direction, as schematically shown in the inset (b) of Fig. 2. Such changes will impose in-plane tensile strains on the PCMO film, leading to an increase in the resistance. When a sinusoidal/triangular ac electric field is applied to the PMN-PT substrate, the lattice of the PMN-PT will periodically expand and contract along the c axis with the same frequency as that of the driving ac electric field (inset of Fig. 3). As a result, the lattice strain and resistance of the PCMO film are modulated with the same frequency as that of the driving ac electric fields. We noted that the modulation of resistance due to converse piezoelectric effect induced lattice strain has been observed in the $La_{0.7}Sr_{0.3}MnO_3/Pb(Zr,Ti)O_3$, $La_{0.7}Sr_{0.3}MnO_3/Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$, and $La_{0.75}Ca_{0.25}MnO_3$/PMN-PT systems.[23–26]

To check the converse piezoelectric effect in the PMN-PT substrate, we measured the electric-field induced longitudinal lattice displacements (Δl) of the PMN-PT substrate when dc and ac electric voltages were applied to it, respectively. As seen in Fig. 4(a), applying a positive 20 V dc electric voltage to the positively polarized PMN-PT causes the lattice of the PMN-PT substrate expands along the c-axis by $\sim$31 nm while applying a negative 20 V dc electric voltage to the positively polarized PMN-PT substrate causes the lattice of the PMN-PT contracts along the c axis by the same magnitude. When 40 V peak-to-peak sinusoidal or triangular ac electric voltages were applied to the PMN-PT substrate, the lattice of the PMN-PT periodically expands and contracts along the c-axis with the same frequency and phase as that of the driving ac electric fields, as shown in Figs. 4(b) and 4(c), respectively. It is known that, when an electric voltage (V) is applied to a polarized ferroelectric material, the electric-field induced relative lattice displacements (Δl) of the material due to the converse piezoelectric effect is given by $\Delta l=d_{33}V$.[19] Using $d_{33}=1500$ pm/V, one obtains $\Delta l=30$ nm

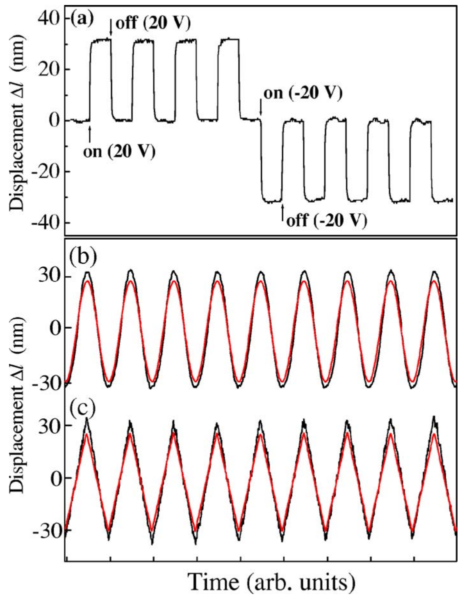

FIG. 4. (Color online) Electric-field induced longitudinal lattice displacements of the PMN-PT substrate via the converse piezoelectric effect at 296 K when ±20 V dc electric voltages and peak-to-peak 40 V sinusoidal and triangular ac electric voltages are applied to the positively polarized PMN-PT substrate, respectively. The red curves represent the sinusoidal and triangular ac electric voltages.

when a 20 V electric voltage is applied to the PMN-PT. This value agrees well with the experimental results. Thus, the piezoelectric displacement measurements confirm that the electric field induces lattice displacements (or strains) in the PMN-PT substrate via the converse piezoelectric effect.

Figure 5 shows the resistance response of the PCMO film as a function of time by applying a 0.1 kV/mm sinusoidal ac electric field to the PMN-PT substrate when the PCMO film is in the charge-ordered state (77 K). Being similar to that observed at 296 K, the resistance of the PCMO film was modulated by the sinusoidal ac electric field, implying that the converse piezoelectric effect of the PMN-PT substrate plays a role in controlling the lattice strain and resistance of the PCMO film in the CO state. The inset of Fig. 5 shows the resistance response of the PCMO film at 77 K as a function of time when ±0.1 kV/mm dc electric fields were switched on and off. Also being similar to that observed at 296 K, applying a positive electric field to the PMN-PT substrate causes a decrease in the resistance of the PCMO film. When the electric field switches from +0.1 kV/mm to −0.1 kV/mm (i.e., corresponding to exert an in-plane compressive strain on the PCMO film), the resistance decreases by ∼25.5 kΩ. This feature indicates that the compressive strain reduces the resistance and thus destabilizes the CO state while the tensile strain enhances the resistance and stabilizes the CO state in the PCMO film, consistent with those reported in Refs. 3–8.

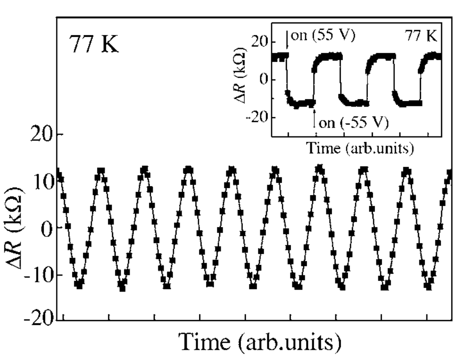

FIG. 5. Resistance (ΔR) response of the PCMO film at 77 K as a function of time (T) when a peak-to-peak 0.2 kV/mm sinusoidal ac electric field is applied to the positively polarized PMN-PT substrate. Inset shows the ΔR versus T curve at 77 K when ±0.1 kV/mm dc electric fields (i.e., ±55 V electric voltages) are switched on and off.

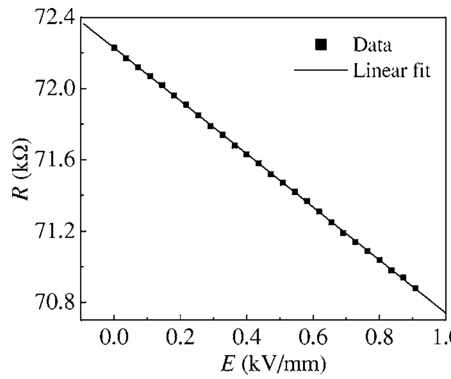

FIG. 6. Resistance of the PCMO film at 296 K as a function of the electric field E applied to the positively polarized PMN-PT substrate. The solid line is the fitting using Eq. (1).

Figure 6 shows the resistance (R) of the PCMO film at 296 K as a function of the electric field E applied to the PMN-PT substrate. The resistance decreases linearly with increasing E. The relationship between R and E can be described by

$$R(E) = R(0) - bE, \tag{1}$$

where b is a positive constant. To quantitatively investigate the correlation between the resistance of the PCMO film and the electric-field induced lattice strain in the PMN-PT substrate; we have measured the lattice parameter c of the PMN-PT substrate when electric fields of 0, 0.25, 0.5, 0.75, and 1 kV/mm were applied to the PMN-PT substrate, respectively. Inset (a) of Fig. 7 shows the PMN-PT(002) reflection under electric fields. Note that the 2θ angle has been normalized to that for $E=0$ kV/mm, i.e., $\Delta 2\theta = 2\theta(E) - 2\theta(0)$. The PMN-PT(002) reflection clearly shifts to lower 2θ angles with increasing electric field. This implies that the electric field has induced an increase in the lattice parameter c of the PMN-PT substrate. Thus, the electric-field induced lattice strains along the direction of the electric field in the PMN-PT substrate (i.e., out-of-plane strain ε_{zz}) can be estimated by $\varepsilon_{zz} = \frac{c(E)-c(0)}{c(0)}$ where $c(E)$ and $c(0)$ are the lattice parameter c of the PMN-PT substrate under electric field and zero electric field, respectively. As shown in Fig. 7, ε_{zz} increases linearly with increasing electric field E. The relationship between ε_{zz} and E can be described by

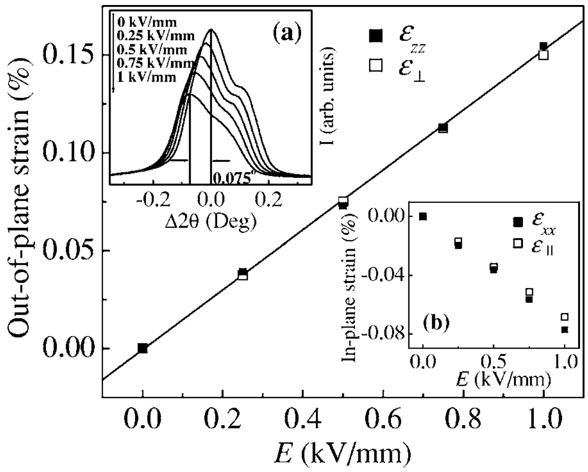

FIG. 7. Out-of-plane strains in the PMN-PT substrate as a function of the electric field applied to the PMN-PT substrate at 296 K. The filled squares represent the out-of-plane strain (ε_{zz}) in the PMN-PT substrate obtained by the XRD measurements while the open squares represent the out-of-plane strain ($\varepsilon_{\perp}$) in the PMN-PT substrate calculated using Eq. (3). Inset (a) shows the XRD patterns of the PMN-PT(002) reflection, as measured under electric fields. Inset (b) shows the in-plane strains ε_{xx} and $\varepsilon_{\parallel}$ in the PMN-PT substrate as a function of E.

$$\varepsilon_{zz} = cE, \tag{2}$$

where c is a positive constant. According to the converse piezoelectric effect, the electric-field induced lattice strains along the direction of electric field in the PMN-PT substrate (i.e., out-of-plane strain $\varepsilon_{\perp}$) due to the converse piezoelectric effect can be calculated by[19]

$$\varepsilon_{\perp} = d_{33}E. \tag{3}$$

Using d_{33}=1500 pC/N, we calculated $\varepsilon_{\perp}$ when electric fields of 0, 0.25, 0.5, 0.75, and 1 kV/mm were applied to the PMN-PT and plotted $\varepsilon_{\perp}$ as a function of E in Fig. 7. It can be seen that $\varepsilon_{\perp}$ agrees well with ε_{zz} obtained by XRD measurements. This gives further evidence that the electric-field induced lattice strain in the PMN-PT substrates has its origin in the converse piezoelectric effect. Combining Eq. (1) with Eq. (2), the relationship between the resistance of the PCMO film and the electric-field induced out-of-plane strain in the PMN-PT substrate can be expressed as

$$R(E) = R(0) - b\varepsilon_{zz}/c. \tag{4}$$

Equation (4) shows that the resistance of the PCMO film is proportional to the induced out-of-plane strain in the PMN-PT substrate. Since the converse piezoelectric effect induces an in-plane biaxial compressive strain in the PMN-PT substrate when a positive electric field is applied to the positively polarized PMN-PT, and with in-plane strain $\varepsilon_{xx}=\varepsilon_{yy}<0$ (compressive strain) and ν=Poisson's ratio, the relationship between ε_{zz} and ε_{xx} in the PMN-PT can be expressed as[27–29]

$$\varepsilon_{zz} = -\frac{2\nu}{1-\nu}\varepsilon_{xx}. \tag{5}$$

In a first-order approximation one can assume volume conservation, i.e., $\varepsilon_{zz}=-2\varepsilon_{xx}$ corresponding to ν=0.5.[26–29] Thus, the electric-field induced in-plane compressive strain in the PMN-PT substrate can be calculated using equation $\varepsilon_{zz}=-2\varepsilon_{xx}$ and ε_{zz} data shown in Fig. 7. ε_{xx} was plotted as a function of electric field in the inset (b) of Fig. 7. Clearly, ε_{xx} is also linearly dependent on the electric field applied to the PMN-PT, implying piezoelectric nature of the strain.[24–26] Once again, according to the converse piezoelectric effect, the electric-field induced lattice strain perpendicular to the direction of the electric field in the PMN-PT substrate (i.e., in-plane compressive strain $\varepsilon_{\parallel}$) can be calculated by[19]

$$\varepsilon_{\parallel} = -d_{31}E. \tag{6}$$

where d_{31} is the transverse piezoelectric coefficient of the PMN-PT substrate. For the PMN-PT substrate, d_{31} is about 686 pC/N. Using Eq. (6) and $d_{31}\sim$686 pC/N, we calculated $\varepsilon_{\parallel}$ for E=0, 0.25, 0.5, 0.75, and 1 kV/mm, respectively, and plotted $\varepsilon_{\parallel}$ as a function of E in the inset (b) of Fig. 7. $\varepsilon_{\parallel}$ also decreases linearly with increasing E. The values of $\varepsilon_{\parallel}$ roughly agrees with ε_{xx}. The linear decrease in ε_{xx} ($\varepsilon_{\parallel}$) indicates that the in-plane lattice of the PMN-PT substrate is compressed with the increase of the electric field. Combing Eq. (1) with Eq. (6), one can obtain the relationship between the resistance of the PCMO film and the electric-field induced in-plane compressive strain in the PMN-PT substrate and can be written as

$$R(E) = R(0) + b\varepsilon_{\parallel}/d_{31}, \tag{7}$$

where $\varepsilon_{\parallel}$ is a negative value. Equation (7) indicates that the resistance of the PCMO film is linearly dependent on the induced in-plane strain in the PMN-PT substrate, which implies that the relative change in the resistance ($\Delta R/R$) of the PCMO films due to substrate-induced lattice strain is proportional to the relative change in the in-plane lattice strain in the PMN-PT substrates.

IV. CONCLUSIONS

We have found that the resistance of the PCMO films grown on ferroelectric PMN-PT substrates can be modulated by applying electric fields across the PMN-PT substrates. Piezoelectric displacement and x-ray diffraction measurements revealed that the modulation of the resistance is caused by the electric-field induced lattice strain in the PMN-PT substrates via the converse piezoelectric effect. The resistance of the PCMO films was found to be linearly dependent on the induced in-plane and out-of-plane strains in the PMN-PT substrates, similar to those observed in $La_{0.7}Sr_{0.3}MnO_3$/PMN-PT and $La_{0.75}Ca_{0.25}MnO_3$/PMN-PT systems where the resistance of the $La_{0.7}Sr_{0.3}MnO_3$ and $La_{0.75}Ca_{0.25}MnO_3$ depends linearly on the induced lattice strain in the PMN-PT substrates.[25,26] Moreover, the relationships between the resistance of the PCMO films and the induced in-plane and out-of-plane strains in the PMN-PT substrates have been quantified, which may be important for understanding the physics of substrate-induced lattice strain effect and device fabrication. The results also show that the in-plane compressive strain destabilizes the CO state while the in-plane tensile strain stabilizes the CO state in the PCMO films, which supports the results reported in Refs. 3–8. These tuning of resistance by *in situ* inducing lattice strain in the PMN-PT substrates provides a good method that can be extended to study intrinsic effects of substrate-induced lattice strain on other properties of thin films.

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*Electronic mail: zrk@ustc.edu

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