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# The current-induced electroresistance in strain-modulated $Pr_{0.5}Sr_{0.5}MnO_3$ film

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$Pr_{0.5}Sr_{0.5}MnO_3$ films were grown on ferroelectric substrates of $0.67Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.33PbTiO_3$ by pulsed-laser deposition method. The film structure and lattice change with electric field applied on the substrate are examined by X-ray diffraction. The electric field dependence of resistivity was compared with electric field dependence of lattice at room temperature, revealing a relation between resistance and strain. Current-induced electroresistance (CER) was studied by using different measuring current. With increasing electric filed a colossal decrease of CER at low temperature was achieved, indicating great strain effect. The piezoelectric strain effect on the magnetoelectric coupling at multiferroic interface was discussed.
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## I. INTRODUCTION

The influence of electric-current on physical properties of ferromagnetic materials have been widely studied.[1–5] Besides the Joule heating effect,[6] the current-induced reversible resistance switching and magnetization switching are focused due to its potential applications and significant physics of magnetoelectric coupling.[7–9] The current-induced electroresistance (CER) effect, where the resistance is dependent on the electric-current, has been observed in several manganites such as $Pr_{1-x}Ca_xMnO_3$[3] and $Pr_{0.5}Sr_{0.5}MnO_3$ (PSMO).[9] The forcedly moved carriers can revive ferromagnetic interaction and trigger the phase transition without a magnetic field.[10] The filamentary conductivity and thus self-magnetic field effect on the phase separation are proposed.[11] Other possible origins such as local field coming from the spin polarization effect have also been discussed.[12] However, the mechanism of CER effect is still not fully understood.

The strain effect on CER should been investigated both for physics research and device application, though the *in-situ* strain effect on CER is rarely studied. Recently, ferroelectric substrates like $0.67Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.33PbTiO_3$ (PMN-PT) are utilized to deposit ferromagnetic (FM)/ferroelectric (FE) heterostructures for the investigation on strain effect. The lattice of ferroelectric layer can be *in-situ* tuned by the applied electric field due to piezoelectricity, which transfers *in-situ* tunable strain to the film. However, other simultaneous magnetoelectric coupling effects caused by ferroelectric field such as charge simulation at the interface may also make the properties of FM layer change.[13] Fortunately, these two originals could be distinguished because the strain effect is symmetrically modified by electric field. Therefore, utilizing this structure, we may explore the *in-situ* strain effect on CER.

The phase diagram of PSMO are extremely sensitive to strain. For example, a ferromagnetic metal (FMM) to antiferromagnetic insulator (AFI) transition takes place at temperature of ~150 K in PSMO bulk. By contrast, such a transition can not be observed in the (001)-oriented $PSMO/LaAlO_3$

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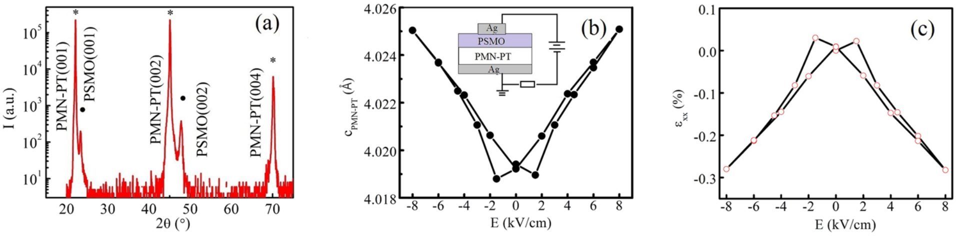

FIG. 1. (a) the $\theta$-2$\theta$ pattern of the initial polarized PSMO/PMN-PT. The diffraction of PSMO and PMN-PT are denoted by the dot (•) and star (*), respectively. (b) the out-of-plane lattice of PMN-PT versus perpendicular electric field. The inset shows the schematic circuit to apply the electric field. (c) in-plane strain of PMN-PT versus electric field.

and PSMO/SrTiO$_3$ thick films but appears in the greatly strained thin films.[14] Ogimoto et al. have reported that, the square template of (001)-oriented cubic substrates constrain the structural distortion required at the FMM-AFI transition.[15–17] On the other hand, both the tensile and compressive strain enhance AFI phase and consequent FMM-AFI transition. Therefore, the FMM-AFI transition is absent in little strained or strain-relaxed films but present in greatly strained cases.[14,18] The previous study on PSMO films also has shown that the CER is dependent on film thickness,[19] indicating strain effect on CER. However, the discrepancy between samples of different thickness makes the research complicated. In this paper, using FM/FE epitaxial heterostructure of PSMO/PMN-PT the influence of *in-situ* strain effect on CER are studied.

## II. EXPERIMENTAL DETAILS

The PSMO films are grown on single crystal substrates of (001)-oriented PMN-PT by pulsed-laser deposition (PLD) method. The film thickness measured by a Dektak stylus profiler is ~40 nm. The x-ray diffraction (XRD) scans of $\theta$-2$\theta$ and $\phi$ are performed to investigate the crystal structures of samples. To study lattice distortion induced by piezoelectric effect, the samples are initially polarized by a field of 12 kV/cm. Then *in-situ* XRD measurements under various electric fields are performed. The inset of Fig. 1(b) shows the schematic diagram of circuit used to apply electric field. In measurements of electrical properties, all films are patterned into microbridges with length of 100 μm and width of 30 μm. Using a Physical Properties Measurement System (PPMS), the resistivity ($\rho$) of the films are measured by the standard four-probe technique. The CER effect is studied by detecting resistivity using different measuring current. To prevent the excitation of metastable states, the currents are controlled to be smaller than 1 mA. In our experiments all the resistivity changes induced by electric field and electric current are reproducible and reversible.

## III. RESULTS AND DISCUSSION

The XRD scans of $\theta$-2$\theta$ and $\phi$ reveal an epitaxial growth of PSMO film with tetragonal symmetry, which is described previously.[19] Fig. 1(a) shows the $\theta$-2$\theta$ pattern of the initial polarized film. The film has an out-of-plane lattice of ($c_{\mathrm{PSMO}}$) ~3.79 Å, which is smaller than that of PSMO bulk (pseudocubic, $a_p$~3.84 Å), indicating an in-plane tensile strain in the film. Fig. 1(b) gives a symmetrical hysteresis loop of the out-of-plane lattice of PMN-PT ($c_{PMN\text{-}PT}$) to the electric field. At $E$=8 kV/cm the $c_{PMN\text{-}PT}$ is extended by 0.14%. According the relation between in-plane strain $\mathcal{E}_{xx}$ and out-of-plane strain $\mathcal{E}_{zz}$ as $\mathcal{E}_{xx}$=-$\mathcal{E}_{zz}$(1-$\upsilon$)/(2$\upsilon$), where $\upsilon$ is the Poisson ratio of PMN-PT $\upsilon$=0.2,[20,21] the in-plane lattice distortion versus electric field can be estimated, which is shown in the Fig. 1(c). A hysteresis loop is found, which is consistent with the result reported by Thiele *et al.*[22] The in-plane strains in the substrate are negative and tend to weaken the tensile strain in the as-grown PSMO.

Fig. 2(a) gives the resistivity percentage change ($\Delta\rho/\rho$) against electric field at room temperature (RT). A symmetrical hysteretic characteristic appears. For the increasing electric field from 0 to ~1.4 kV/cm, the RT resistivity increases by 1.7%. Thereafter, it decreases with an increasing field, and gets lowered by 6.1% at $E$=8 kV/cm. Such a $\Delta\rho/\rho$-$E$ curve is similar to that of $\mathcal{E}_{xx}$-$E$ in Fig 1(c). We gave $\Delta\rho/\rho$ of the film versus $\mathcal{E}_{xx}$ of the substrate in the onset of Fig. 2(a). The resistivity change is

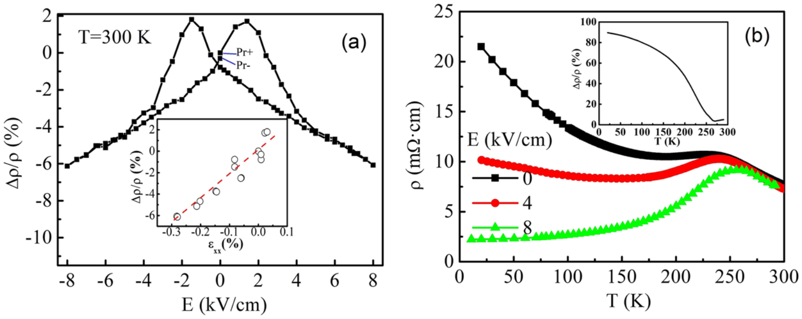

FIG. 2. (a) the percentage resistivity change of $\Delta\rho/\rho$=[$\rho(E)$-$\rho(0)$]/$\rho(0)$ versus electric field at room temperature. The onset of Fig. 2(a) shows the relationship between $\Delta\rho/\rho$ of the film and substrate strain (b) the typical $\rho$-$T$ curves under various fields. The inset shows the temperature dependent $\Delta\rho/\rho$ for $E$=8 kV/cm.

linearly dependent on the in-plane strain. As denoted in Fig. 2(a) the resistivity discrepancy between positive and negative remnant polarizations (Pr+, Pr-) is little.

The opposite Pr+ and Pr- polarizations lead to the opposite charge simulations in the film side near the interface between film and substrate. As seen above, the resistivity response to electric field is quite symmetrical and little dependent on the direction of electric field. This observation demonstrates that here the carrier simulation is not the dominant reason for resistivity change. The little resistivity difference between Pr+ and Pr- polarization state is due to the same strain induced in these states. The modulation of conductivity is mostly determined by the strain.

It has been discussed that phase separation could be electric-field controlled in manganite/ferroelectric heterostructures.[18,23] The symmetrical response of resistivity on electric field indicates in our research the piezostrain effect should be the main reason for the resistivity changes dominated by the modutlation in phase separation. The in-plane strains in the substrate are negative as above and tend to weaken the tensile strain in the as-grown PSMO, which favors the FM phase due to the weakening of Jahn–Teller distortion.

Fig. 2(b) displays the typical $\rho$-$T$ curves under various electric fields. In the initial polarized state ($E$=0 kV/cm), the film undergoes a paramagnetic insulator-ferromagnetic metal transition upon cooling with a resistivity peak at temperature of $T_p$ ~225 K, and then becomes insulating with a resistivity valley at temperature of $T_v$ ~190 K. An electric field of 4 kV/cm leads to a lower $T_v$ ~145 K with a higher $T_p$ ~240 K. Under an electric field of 8 kV/cm, the film becomes metallic at further higher temperature of $T_p$ ~260 K and remains in metallic state at low temperature. Therefore, the electric field leads to an insulator to metal (I-M) transition at low temperature. The inset of Fig. 2(b) shows the temperature dependent resistivity change $\Delta\rho/\rho$ induced by $E$=8 kV/cm. The decrease of resistivity is greater with lower temperature, and reaches as high as ~90 % at 25 K.

As reported by the previous papers,[14–16] the phase diagram of PSMO/PMN-PT films are affected by substrate from two originals. One is the square template of the substrate constrains the lattice transition accompanied with FMM-AFI transition of the film, resulting in absence of the transition in little strained films. The other is the tensile strain enhances FMM-AFI transition in strained films. The strain effect on the phase diagram could be explained by the phase separation and competition in the film. The subtle balance of free energy between the coexistent AFI and FMM phases at low temperature is sensitive to perturbs such as temperature, strain and magnetic field. At the initial polarized state, the large substrate-induced tensile strain leads to a strong AFI state ground phase in PSMO film. When the electric field increases to 8 kV/cm, the in-plane lattice of PMN-PT roughly gets compressed by 0.28%, which reduces the tensile strain in PSMO film. As a result of strain reduction, suppressed AFI phase and enhanced FMM phase lead to higher $T_p$, lower $Tv$ and eventually a metallic ground state.

Fig. 3 shows the current dependence of $\rho$-$T$ when the applied electric field is (a) 0 kV/cm, (b) 4 kV/cm and (c) 8 kV/cm, respectively. The insets give the corresponding resistivity changes between the current of 1 μA and 1 mA as CER=[$\rho$(1 mA)-$\rho$(1 μA)]/$\rho$(1 μA) *100%. In the initial polarization, the negative CER reaches a relative maximum near the FM transition temperature, similar to that

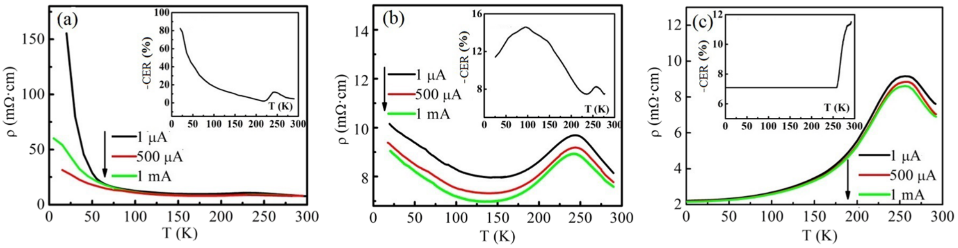

FIG. 3. the typical $\rho$-$T$ curves measured using various current when the applied electric field is (a) 0 kV/cm, (b) 4 kV/cm and (c) 8 kV/cm, respectively. The insets give the responding resistivity change from the current of 1 μA to 1 mA as CER=[$\rho$(1 mA)-$\rho$(1 μA)]/$\rho$(1 μA)*100%, respectively.

of other FM manganites.[21,22] At low temperature, the negative CER sharply increases with lower temperature, and reaches as high as 83% at 25 K. Higher electric field leads to smaller negative CER. Under $E$=8 kV/cm, without FMM to AFI transition at low temperature, the negative CER remains ~7% at 25 K, much lower than that in the initial polarized state. In short, the electric field brings great drop of colossal CER of 76% in low temperature AFI phase. However, in any polarizations, the $T_p$ change littles with increasing current.

CER in manganites might ascribe to two aspects: one is the strong interaction between carrier spins and localized spins in Mn ions, and the other is the percolative mechanism of phase separation. The former means that the interaction between carrier spins and localized spins in Mn ions would force the localized spins to be parallel. A direct consequence is the high-temperature shift of $T_p$. However, in our study no obvious movement of $T_p$ has been observed. When a current with high density is applied on the film, the Joule-heating is strong and the $T_p$ may could be detected exactly. It seems that the CER observation favors the percolative mechanism of phase separation. At the initial polarization ($E$=0 kV/cm), the film is under great in-plane tensile strain. We might consider a state of equilibrium in which FMM clusters are embedded in an AFI matrix. An applied current or its accompanying electric field perturbs the coexistence of different phases and sets up filamentary currents across the insulating region, bringing a great resistivity change at low temperature. Under $E$=8 kV/cm, the weakened tensile strain makes FMM phase is dominantly existing. With a metallic background, though more filamentary conductive paths are induced by the current, the resistivity would not change a lot. Therefore, the CER effect here could be explained by the percolation of the separated phases.

## IV. CONCLUSIONS

In summary, the electric-field control of CER is realized in PSMO/PMN-PT multiferroic heterostructures. A great drop of CER (76%) with an I-M transition at low temperature is induced by an electric field of 8 kV/cm, showing efficient magnetoelectric coupling at the interface. The CER effect changes with electric field could be understood by the percolative mechanism of phase separation which is able to be significantly *in-situ* modulated by the piezostrain.

## ACKNOWLEDGMENTS

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