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Effects of electric-field-induced piezoelectric strain on the electronic transport properties of $La_{0.9}Ce_{0.1}MnO_3$ thin films

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ABSTRACT

The authors constructed multiferroic structures by growing $La_{0.9}Ce_{0.1}MnO_3$ (LCEMO) thin films on piezoelectric $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMN-PT) single-crystal substrates. Due to the efficient elastic coupling at the interface, the electric-field-induced piezoelectric strain in PMN-PT substrates is effectively transferred to LCEMO films and thus, leads to a decrease in the resistance and an increase in the magnetoresistance of the films. Particularly, it was found that the resistance-strain coefficient $[(\Delta R/R)_{film}/(\Delta \varepsilon_{zz})_{film}]$ of the LCEMO film was considerably enhanced by the application of magnetic fields, demonstrating strong coupling between the lattice and the spin degrees of freedom. $(\Delta R/R)_{film}/(\Delta \varepsilon_{zz})_{film}$ at 122 K was enhanced by ~28.8% by a magnetic field of 1.2 T. An analysis of the overall results demonstrates that the phase separation is crucial to understand strain-mediated modulation of electronic transport properties of manganite film/PMN-PT multiferroic structures.

1. Introduction

Multiferroic structures composed of perovskite manganite thin films $La_{1-x}A_xMnO_3$(A = Ca,Sr,Ba) and ferroelectric single crystals have attracted great interest in recent years because they exhibit magnetoelectric effects with potential applications in multifunctional devices [1,2]. Theoretical calculations and experimental results have shown that the magnetoelectric effect in such complex oxide multiferroic structures, e.g., $Pb(Zr_{0.3}Ti_{0.7})O_3$ film/$La_{1.2}Sr_{1.8}Mn_2O_7$ crystal [3], $La_{1-x}A_xMnO_3$(A = Ca,Sr,Ba) film/$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ crystal [4–9], $La_{1-x}Sr_xCoO_3$ film/$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ crystal [10,11] etc., arises from the interaction of the elastic components of the ferromagnetic and ferroelectric layers. Namely, an electric field induces piezoelectric strain in the ferroelectric crystal, which is passed on to the neighboring ferromagnetic film, thereby modifying the magnetic properties of the magnetic layer. Conversely, a magnetic field induces strain in the magnetic crystal via magnetostriction, which is transferred to the neighboring ferroelectric film in which the electrical properties are modified by the induced strain. It is generally accepted that the interface strain coupling plays an essential role in determining the electronic and magnetic properties of manganite thin films in manganite film/ferroelectric crystal heterostructures [3–11]. So far, the interface strain coupling and its impact on the electronic and magnetic properties of manganite thin films with optimal hole doping level of $0.2 \leq x \leq 0.3$ for manganite film/ferroelectric crystal heterostructures have been studied intensively by several groups [4–9], little research has been done in lightly doped ($x \leq 1/8$) manganite films whose electronic and magnetic properties are different from those with optimal hole doping level. Therefore, a detailed investigation of the interface strain coupling effect and its impact on the electronic transport properties of lightly doped manganite films would help researchers better understand the strain-property coupling effects in manganite thin films.

Perovskite (1-x)$Pb(Mg_{1/3}Nb_{2/3})O_3$-$x$$PbTiO_3$ single crystals with compositions near the morphotropic phase boundary display excellent piezoelectric activities (the piezoelectric coefficient d_{33} ~2400 pC/N [12]). These piezoelectrically active crystals have been used as substrates to grow perovskite ferromagnetic thin films [4–11]. In this paper, we epitaxially grow lightly doped perovskite $La_{0.9}Ce_{0.1}MnO_3$ (LCEMO) thin films on ferroelectric $0.68Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.32PbTiO_3$ (PMN-PT) single crystal substrates and studied the intrinsic effects of the electric-field-induced piezoelectric strain on the electronic transport properties of the LCEMO film. Without the constraint from substrates, the piezoelectric strain was effectively transferred to the LCEMO film, which causes a decrease in the resistance and an enhancement in the magnetoresistance (MR). Particularly, it was observed that the effects of the piezoelectric strain on the electronic transport properties were significantly enhanced by the application of a magnetic field. An analysis of

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the overall results shows that the electric-field-induced piezoelectric-strain effects can be reasonably understood within the framework of the phase separation scenario.

2. Experiment

PMN-PT single crystals with a dimension of Φ50 mm × 80 mm were grown by a modified Bridgman technique as described previously [13]. The as-grown crystals were cut into rectangular plates with a dimension of 10 mm × 2.5 mm × 0.5 mm and with the plate normal in the <001> crystal direction and polished to an average surface roughness of less than 1 nm. LCEMO films were deposited on such polished PMN-PT substrates by pulsed laser deposition using a 248 nm KrF excimer laser with a repetition rate of 4 Hz. The laser energy density irradiated on the rotating LCEMO targets was ~3 J/cm^2. The deposition was carried out at 700 °C with the oxygen pressure fixed at 27 Pa. After deposition, the films were in situ cooled to room temperature.

X-ray diffraction (XRD) patterns of the LCEMO/PMN-PT structure were recorded using a high resolution Bruker D8 Discover X-ray diffractometer equipped with a four-bounce Ge(220) monochromator and Cu *K*α radiation (λ = 1.5406 Å). The inset (1) of Fig. 1(a) shows a schematic diagram for in situ measurements of the electric-field-induced strain in the LCEMO film. The XRD θ-2θ scans were made in the 2θ ranging from 44 to 48° at 296 K during the application of an electric field to the PMN-PT substrate. The inset (2) of Fig. 1(a) shows the in-plane resistance measurement circuit for the LCEMO/PMN-PT structure. A Keithley model 2400 source meter and a Keithley model 2000 voltage meter were employed to measure the resistance of the LCEMO film between the two top–top gold electrodes in the temperature region from 122 to 300 K. During the resistance measurements, a constant current of 0.1 μA was applied between the two top–top gold electrodes and an additional external dc electric field of $E = 0$ or 10 kV/cm was applied to the PMN-PT substrate through the LCEMO film (i.e., top electrode) and the gold (i.e., bottom electrode) using a Keithley 6517A electrometer. The top and bottom electrodes were held at low and high potentials, respectively, so that the direction of the electric field points to the LCEMO film. The resistance of the PMN-PT at room temperature is ~3 × 10^9 Ω while that of the LCEMO at $T = 300$, 200, and 120 K are ~1 × 10^5, ~1.2 × 10^6, and ~5.7 × 10^7 Ω, respectively. These values are 0.0033%, 0.04% and 1.9% of that of the PMN-PT substrate. Therefore, the electric field drop across the LCEMO film is minor and could be neglected. A resistor of 20 MΩ was connected in series with the bottom gold electrode in order to protect the source meter and voltage meters in case a dielectric breakdown took place in the PMN-PT substrate. Magnetic properties of the LCEMO film were measured using a superconducting quantum interference device (SQUID 5T, Quantum Design) magnetometer with magnetic fields applied parallel to the film plane.

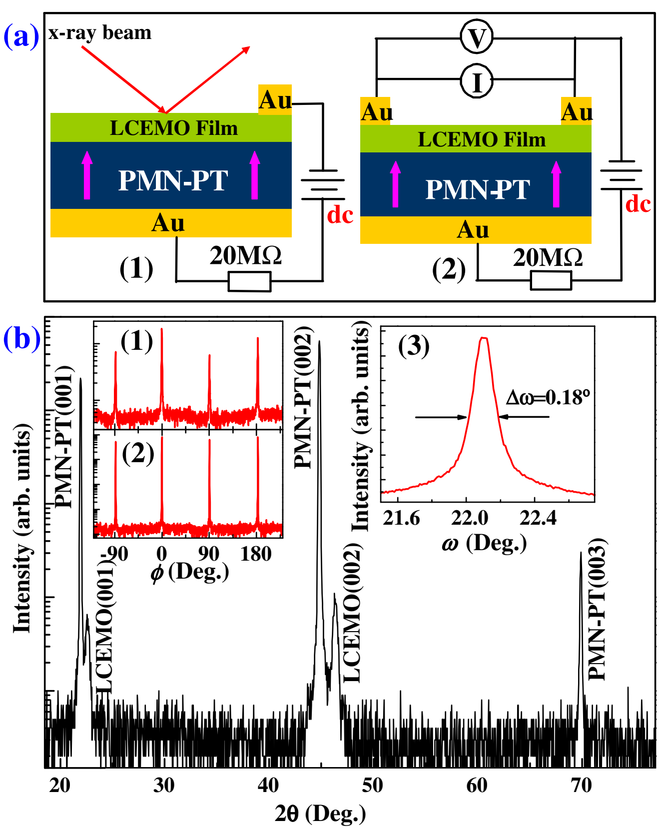

Fig. 1. (a) Schematic diagrams of the LCEMO/PMN-PT structure and the electric field configuration for in situ measurements of the strain (left panel) and the resistance (right panel). The arrow in the PMN-PT represents the poling direction. (b) XRD θ-2θ scan of the LCEMO/PMN-PT structure. The insets (1) and (2) show the XRD phi scans of the LCEMO(101) and PMN-PT(101) planes, respectively. The inset (3) shows the XRD rocking curve taken around the LCEMO(002) reflection.

3. Results and discussion

Fig. 1(b) shows the XRD θ-2θ scan of the LCEMO/PMN-PT structure. Only (00*l*) (*l* = 1, 2) reflections from the LCEMO film appear, indicating that the film is highly *c*-axis preferentially oriented. No reflections were detected that would be indicative of second phases. The XRD rocking curve taken around the LCEMO(002) reflection has a full width at half maximum of ~0.18° [the inset (3) of Fig. 1(b)], implying quite good crystalline quality of the LCEMO film. We examined the in-plane epitaxial relationship between the LCEMO film and the PMN-PT substrate through XRD phi scans of the LCEMO(101) and PMN-PT(101) planes and observed the sharp fourfold symmetrical reflections originating from the LCEMO film and the PMN-PT substrate [the insets (1) and (2) of Fig. 1(b)], respectively, indicating a cube-on-cube epitaxial growth of the LCEMO film on the PMN-PT substrate.

Fig. 2 shows the temperature dependence of the resistance for the LCEMO film when electric fields of $E = 0$ and 10 kV/cm were applied to the PMN-PT substrate. Whether $E = 0$ or 10 kV/cm, the resistance increases with decreasing temperature from 300 to 122 K. A small kink in the *R*–*T* curve for $E = 0$ kV/cm is appreciable near $T = 155$ K. For $T < 122$ K, the film is highly insulating. However, when an electric

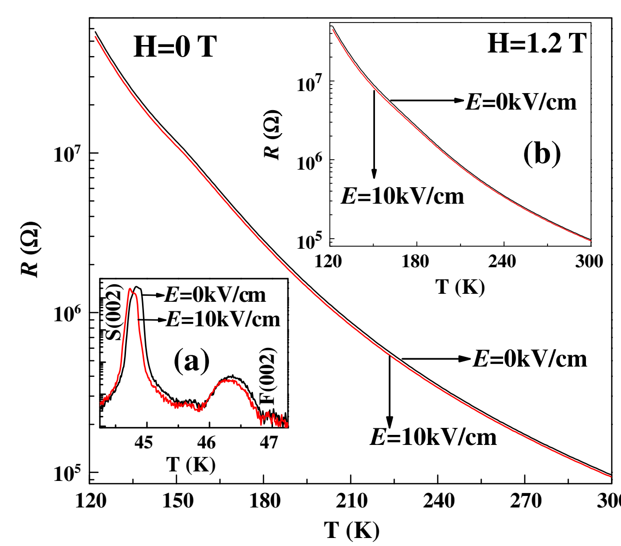

Fig. 2. Temperature dependence of the resistance for the LCEMO film under $E = 0$ and 10 kV/cm for $H = 0$ T. The inset (a) shows the XRD patterns in the vicinity of the PMN-PT(002) and LCEMO(002) reflections under $E = 0$ and 10 kV/cm, respectively. The inset (b) shows the temperature dependence of the resistance for the LCEMO film under $E = 0$ and 10 kV/cm for $H = 1.2$ T.