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Strain effects on transport and magnetic properties of $Pr_{0.65}La_{0.05}Ca_{0.3}MnO_3$ thin films

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ABSTRACT

$Pr_{0.65}La_{0.05}Ca_{0.3}MnO_3$ (PLCMO) thin films in various thickness with either compressive or tensile strains were epitaxially deposited on (0 0 1) $SrTiO_3$ (STO) and (0 0 1) $LaAlO_3$ (LAO) substrates. Epitaxial strain in films relaxes with increasing film thickness, as evidenced from X-ray diffraction studies. Charge ordered insulating phase is stabilized in strained PLCMO films, no matter it is compressive or tensile. The charge ordering temperature decreases with the decrease of strain in PLCMO films. The curie temperature and the saturated magnetic moment in films increases as the strain relaxes with increasing film thickness. The transport and magnetic properties are well explained in terms of substrate induced strain.

1. Introduction

Since the discovery of colossal magnetoresistance and charge-orbital ordering in perovskite rare earth manganites, $R_{1-x}A_xMnO_3$ (R=trivalent rare earth elements, A=divalent alkaline earth elements) materials have attracted considerable attention due to their unique electrical and magnetic characteristics [1,2]. The recent interest in rare earth manganites focuses on the phase separation, which is believed to be a key ingredient for their physical properties [3]. Within this frame, $Pr_{1-x}Ca_xMnO_3$ ($x\sim0.3$–0.5, PCMO) has been recognized as a representative system in which a phase separation leads to microscopic inhomogeneity due to the coexistence of an antiferromagnetic charge ordered insulating phase resulting from coulomb interaction of Mn^{3+} and Mn^{4+} ions, and a ferromagnetic metallic phase originating from Mn^{3+}–O–Mn^{4+} double exchange interactions at low temperatures [4,5]. The competition between the charge ordered phase and the metallic phase is very sensitive to various external stimuli. The charge ordered phase may melt and convert into the metallic state via a percolative way due to X-ray irradiation [6], electric current [7], pressure [8] or magnetic fields [9], giving rise to a large reduction of the resistivity.

Previous works on PCMO system have revealed that the charge ordered insulating phase occurs below a certain temperature, the charge ordering temperature (T_{co}). The charge ordered phase may be transformed into a metallic state upon the application of a critical magnetic field, which is at the origin of the appearance of colossal magnetoresistance [9–11]. A large critical magnetic fields up to 25 T is required to melt the charge ordered phase in bulk PCMO [10]. However, in the case of thin films, the destruction of the charge ordered phase and the induced insulator–metal transition appears at a much lower magnetic field. For instance, the critical melting filed has been reduced to 7 T in 110 nm thick epitaxial PCMO thin films with tensile strain, indicating a strong effect of residual epitaxial strain, due to the lattice mismatch between the films and the substrates, on the charge ordering behavior in PCMO thin films [9]. Epitaxial strains have been widely used to modulate the physical properties in manganite thin films due to their significant effects on the competition among charge, spin, and orbital orders [11–13]. One of the possible mechanisms is that the strain may mediate orbital-ordering of the e_g states, which couples with the Jahn–Teller distortion of MnO_6 octahedra [12,13]. As an example, Millis et al. [11] pointed out that 1% strain would lead to 10% shift in the Curie temperature (T_c) in perovskite manganites such as $La_{0.83}Sr_{0.17}MnO_3$ due to the Jahn–Teller distortion. Although tremendous efforts have been made to study the strain effects on the physic properties of PCMO, the melting behaviors of the charge ordered phase as a function of epitaxial strain is still unclear [9,14–16]. In fully strained PCMO thin films, 15 nm in thickness, on $SrTiO_3$ (STO) or $LaAlO_3$ (LAO), the charge ordered state could not be melted with the application of magnetic field up to 9 T [14]. As the film thickness increases, the critical field of PCMO on STO decreases remarkably. Prellier et al. [9] and Yang et al. [15] reported that the melting filed could be reduced to 7 T and 4 T, respectively, for films of 110 nm and 150 nm on STO. However, PCMO films, 110 nm in thickness, on

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LAO were still an insulator under 7 T [9]. In addition, the evolutions of magnetic properties of PCMO in terms of the residual strain are also uncertain [5,17,18].

On the other hand, the average A-site cation radius also plays important roles in determining the transport and magnetic characteristics in PCMO. The incorporation of elements such as La^{3+}, Ba^{2+} or Sr^{2+}, with larger cation radius, in the A-site could increase the width of e_g band, disturb charge ordered state and induce the development of ferromagnetic regions in the antiferromagnetic matrix in PCMO [19,20]. For example, a critical field which urges the transformation from an antiferromagnetic insulating sate to a ferromagnetic metallic one could be lowered by La^{3+} substitution [19,20].

The effect of epitaxial strain on the melting behavior and the magnetic properties in La^{3+} doped PCMO thin film is rare in the literature. In this paper, we reports systematic studies on the structure, magnetization and transport characteristics of epitaxial $Pr_{0.65}La_{0.05}Ca_{0.3}MnO_3$ (PLCMO) thin film as a function of film thickness from 30 to 130 nm, grown on (0 0 1) STO and LAO substrates, emphasizing the relations between the residual strain and the physic properties.

2. Experimental details

The epitaxial PLCMO films with thickness of 30, 50 and 130 nm were grown on (0 0 1)-oriented STO and LAO substrates by pulsed laser deposition at 800 °C in an oxygen pressure of 0.01 mbar, using a 248 nm KrF excimer laser (CompexPro 205F, Coherent) with a repetition rate of 3 Hz. The crystal structure, lattice constants and strain state were investigated by means of high resolution X-ray diffraction (XRD) using the BL14B1 beamline of Shanghai Synchrotron Radiation Facility, China with an X-ray wavelength of 1.2398 Å. Transport measurements were carried out by a physical property measurement system (PPMS-16, Quantum Design) up to 12 T. Magnetization of the films were characterized as functions of temperature and magnetic field in a superconducting quantum inference device magnetometer (MPMS XL-7, Quantum Design).

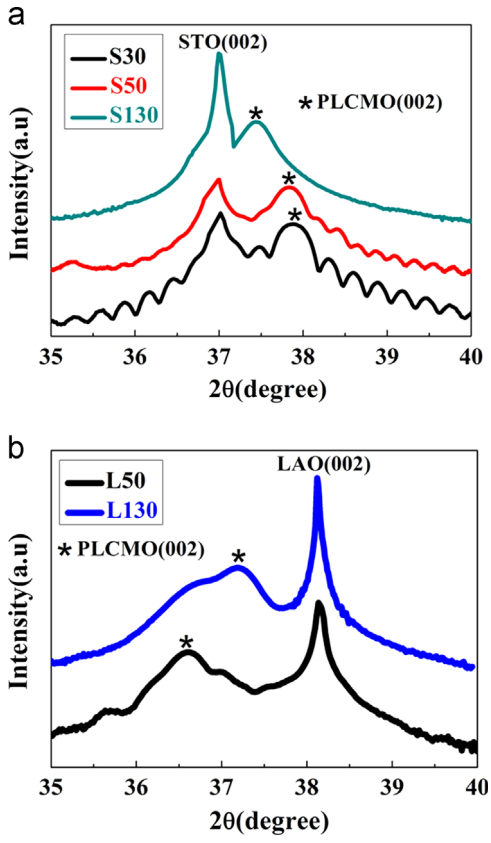

Fig. 1. X-ray diffraction θ–2θ scans recorded around (0 0 2) diffraction peaks of various PLCMO films on (a) STO and (b) LAO substrates.

3. Results and discussions

Fig. 1(a) and (b) show XRD patterns around the (0 0 2) reflections of various PLCMO thin films deposited on (0 0 1) STO and (0 0 1) LAO substrates, respectively. The XRD patterns are indicative of an epitaxial growth on the substrates. The samples are hereafter labeled as Sx for films on STO or Lx for films on LAO, where x is film thickness in nanometers. For example, S30 stands for a 30 nm-thick PLCMO on STO while L130 represents for a 130 nm-thick PLCMO in LAO. The appearance of clear fringes around PLCMO (0 0 2) diffraction peak in the S30 sample indicates the smooth surface and the sharp interface. Bulk PLCMO

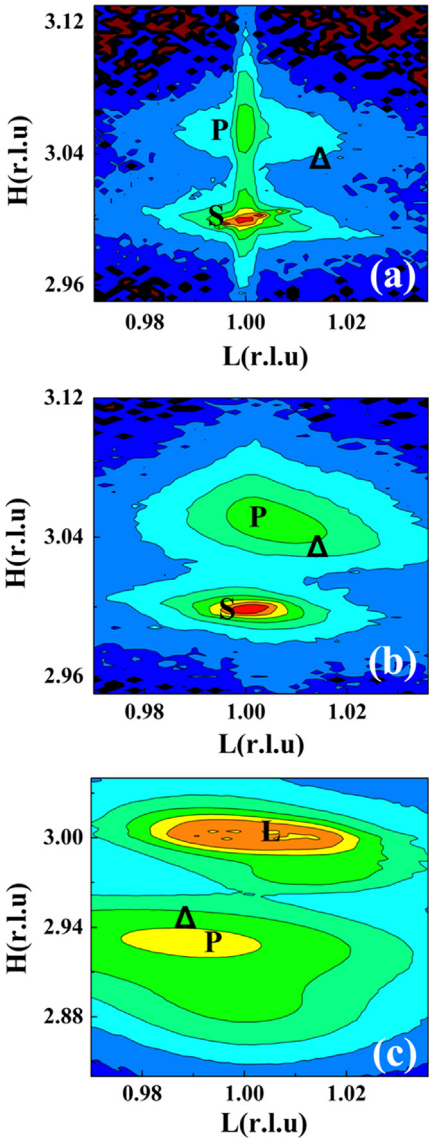

Fig. 2. Two-dimensional diffraction indensity mapping on (1 0 3) reflections from (a) S30, (b) S130 and (c) L130 samples. "P", "S" and "L" indicate the (1 0 3) diffraction peaks from the PLCMO film, the STO and the LAO, respectively. The Δ symbol indicates the differcation peak of PLCMO if the films are completely relaxed.