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Colossal magnetoresistance of $Pr_{0.7}Sr_{0.3}MnO_3$ layer grown on charge-ordered $La_{0.5}Ca_{0.5}MnO_3$ manganite layer

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ABSTRACT

We present a report of the interfacial reaction between ferromagnetic (FM) $Pr_{0.7}Sr_{0.3}MnO_3$ (PSMO-3) in proximity to charge ordered (CO) $La_{0.5}Ca_{0.5}MnO_3$ (LCMO-5) in a bilayer structure. PSMO-3/LCMO-5 bilayer film with high crystalline quality was epitaxially grown on $SrTiO_3$ (001) substrate by pulsed laser deposition. The bilayer loses 20% of its resistance for only 0.5 T, and loses 50% of its resistance for 2 T. This large low-field magnetoresistance (LFMR) is comparable to some FM/CO multilayers and other mixed compounds including strongly competing phases. LFMR of ~−73% has been achieved at H = 1 T and T = 185 K. The maximum MR reaches a value of ~−641% at H = 9 T and T = 205 K and the corresponding MR at room temperature is ~−30%. The formation of CO anti-ferromagnetic state in LCMO-5 and its collapse (melting) under the applied magnetic field may be responsible for the enhanced MR mentioned above. In other words, the enhancement of MR can be explained by a FM phase percolation. Moreover, exchange bias (EB) cannot be observed in the bilayer at low temperature of 3 K, further confirming the occurrence of FM phase percolation at the PSMO-3/LCMO-5 interface. The disappearance of EB can be explained by a combination of the preferable distribution of FM clusters and FM clusters percolation at the PSMO-3/LCMO-5 interface. Large MR can be realized by using a CO material as a sublayer, which can provide potential application in the used devices.

1. Introduction

Due to the discovery of colossal magnetoresistance (CMR) [1], perovskite manganese oxides of general formula $R_{1-x}A_xMnO_3$ (R is rare earth, A is alkaline earth) have attracted widespread attention for their potential application in the field of spintronic devices [2,3]. CMR is usually characterized by a drop of resistance and it is often observed in a doping range of $0.2 < x < 0.45$. A large majority of manganites exhibit ferromagnetic (FM) metallic behavior below the Curie temperature T_C while exhibiting semiconductor-like characteristic at high temperature [4,5]. By applying a magnetic field, one can get a large magnetoresistance (MR) near the metal-insulator (MI) transition temperature T_{MI}. However, in order to get a large MR, it usually requires a high magnetic field for practical application.

Half-doped manganite $La_{0.5}Ca_{0.5}MnO_3$ (LCMO-5) exhibits phase separation and charge ordered (CO) state [6,7]. The CO state is insulating and is accompanied with a sharp increase in resistance at the transition temperature T_{CO}. At close temperature, concomitant phase transitions of structure and magnetism often happen. For example, the unit cell can shift from orthorhombic (*Pnma*) structure to monoclinic ($P2_{1/m}$) structure at T_{CO}, and the so-called CE-type anti-ferromagnetism can appear at Néel temperature T_N [8–10]. The CO state can be melted by exerting a magnetic field larger than H_C and recover a conducting FM phase [11–14]. For example, in half-doped $Nd_{0.5}Ca_{0.5}MnO_3$, H_C can reach a value of 20 T [9,10]. While in LCMO-5 single thin film, this field H_C is suppressed down to 5 T due to lattice strains in substrate [15].

Because of recent improvement in thin-film deposition technique, many studies of thin films give some successful enhancement of the MR [16]. Moreover, combining two materials with different properties to form a multilayer structure can produce very interesting phenomena [17,18]. The interfacial coupling of two competing ground states (the metallic FM state and the CO insulating state) is significant complex [19]. Therefore, a bilayer film of

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FM metallic layer and CO insulating LCMO-5 layer is appropriate for investigations.

In this paper, we study the transport properties of $Pr_{0.7}Sr_{0.3}MnO_3/La_{0.5}Ca_{0.5}MnO_3$ bilayer and a different route is presented to enhance the MR. We use the proximity of a FM metallic $Pr_{0.7}Sr_{0.3}MnO_3$ (PSMO-3) layer to a CO LCMO-5 layer to produce large MR at different magnetic fields of 1–9 T. Low-field MR (LFMR) as large as ~−73% is achieved at an applied field of only 1 T.

2. Experimental method

Solid-state reaction method was used to synthesize the targets of PSMO-3 and LCMO-5 with details elsewhere [15]. The bilayer film of PSMO-3 and LCMO-5 was grown by pulsed laser deposition (PLD) on single-crystal $SrTiO_3$ (001) substrate at a temperature of 720 °C under the oxygen pressure of 5×10^{-1} mbar. At 7 cm from the substrate, targets were ablated by a KrF laser ($\lambda = 248$ nm) at a repetition rate of 2 Hz and an energy density of 3 J/cm^2 over a spot size of 0.1 cm^2. The deposition rate is ~0. 06 nm/s for both materials, which has been evaluated from the thickness measurement of reference specimen. After deposition, the bilayer was annealed in 1 atm pure oxygen for half an hour. The growth was followed by an *in situ* fast cooling (35 °C/min) from 720 to 500 °C under the same oxygen pressure, followed by a 5 °C/min cooling ramp from 500 to 300 °C. The thickness of top PSMO-3 is 36 nm and the thickness of bottom insulating LCMO-5 is 24 nm. During the preparation of bilayer, sufficient oxygen pressure can avoid the possible oxygen deficiency. Wavelength dispersive X-ray spectroscopy suggests that the composition of the PSMO-3 and LCMO-5 layer is the same as that of corresponding target within experimental error. In addition, two 50-nm-thick single layer films of PSMO-3 and LCMO-5 have been grown under the same condition for comparison.

Surface morphology was measured by atomic force microscopy and the crystallographic structure was examined by high resolution X-ray diffraction (HRXRD). X-ray diffraction experiment was performed in beam line 4W1C of Beijing Synchrotron Radiation Facility (BSRF). The X-ray wavelength is 0.15405 nm, and the energy resolution is 4.4×10^{-4}. Resistance as a function of magnetic field and temperature was studied in a physical properties measurement system (PPMS) using a conventional four-probe technique. The contact resistance is negligible compared to the high resistance of samples. Magnetic properties were measured using a PPMS with a vibrating sample magnetometer (VSM) option with the magnetic field in the sample plane.

3. Results and discussion

Typical room-temperature HRXRD curve in the θ-2θ mode is shown in Fig. 1(a) for the region between 40° and 52°. Besides the (002) peak of $SrTiO_3$, a PSMO-3 peak indexed as (002) and a LCMO-5 peak indexed as (002) can be observed. Only (00*l*) reflections are observed, indicating that the orientation relationship between the bilayer in relation to the substrate. Crystalline quality was studied by the full width at half maximum (FWHM) of the on axis rocking curves. The FWHM of $SrTiO_3$ substrate, PSMO-3 (002) peak and LCMO-5 (002) peak is 0.12°, 0.18° and 0.25°, respectively. The FWHM of PSMO-3 (002) peak and LCMO-5 (002) peak is very small, suggesting the high crystalline quality of the PSMO-3/LCMO-5 bilayer. Fig. 1(b) shows the asymmetric X-ray diffraction curve in 2θ-ω mode around (103) reflection plane of PSMO-3/LCMO-5 bilayer on (001) $SrTiO_3$ substrate. In order to get the in-plane epitaxy of the PSMO-3/LCMO-5 bilayer, we carried out the φ scans experiments around asymmetric (103) planes of the bilayer. The scans of (103) reflection planes of PSMO-3 layer, LCMO-5 layer and $SrTiO_3$ substrate were performed with the same starting φ azimuth. The reflection intensity of the asymmetric (103) planes of PSMO-3 layer, LCMO-5 layer and $SrTiO_3$ substrate is shown in Fig. 1(c). The presence of four symmetric peaks with 90° interval confirms the fourfold symmetry of pseudocubic perovskites. The reflection peaks of (103) PSMO-3 layer, (103) LCMO-5 layer and (103) $SrTiO_3$ substrate appear at the same azimuth φ angle, representing an in-plane orientation relationship of $SrTiO_3$ [100]// PSMO-3 [100]//LCMO-5 [100]. Fig. 1(a)–1(c) show the following orientation relationship: $SrTiO_3$ (001) [100]//PSMO-3 (001) [100]// LCMO-5 (001) [100]. The well epitaxial growth is related with small lattice mismatch between the bilayer and the substrate, since the value of lattice parameter in bulk PSMO-3 (pseudo-cubic), bulk LCMO-5 (pseudo-cubic) and $SrTiO_3$ substrate (cubic) is $a_{PSMO} = 0.3874$ nm, $a_{LCMO} = 0.3822$ nm and $a_{STO} = 0.3905$ nm, respectively [15]. The out-of-plane ($a_\perp$) and in-plane ($a_{//}$) lattice parameters for two different sublayers (PSMO-3 and LCMO-5) in the bilayer and associated lattice strains are calculated from the X-ray diffraction θ-2θ scan (see Fig. 1(a)) and the asymmetric X-ray 2θ-ω scans (see Fig. 1(b)). The out-of-plane strain$\varepsilon_\perp$ and the in-plane strain$\varepsilon_{//}$ can be calculated using the formula $\varepsilon = (a-a_0)/a_0$, where a is $a_\perp$ and/or $a_{//}$ and a_0 is the bulk unstressed lattice parameter as measured from power XRD pattern [15]. For PSMO-3 layer, the $a_{//}$(0.3895 nm) was slightly larger than $a_\perp$(0.3829 nm), indicating an in-plane stretching of the PSMO-3 lattice. The corresponding strain values ($\varepsilon_\perp = -1.16\%$, $\varepsilon_{//} = 0.54\%$) show that PSMO layer has slight tensile strain. In the same way, for LCMO-5 layer, $a_{//}$ (0.3874 nm) was larger than $a_\perp$ (0.3731 nm), suggesting an in-plane stretching of the LCMO-5 lattice. The corresponding strain values ($\varepsilon_\perp = -2.39\%$, $\varepsilon_{//} = 1.36\%$) suggest that LCMO layer has tensile strain. The calculated strain values indicate an out-of-plane compressive strain but an in-plane tensile strain in PSMO-3/LCMO-5 bilayer film in order to match the slightly larger lattice parameter of $SrTiO_3$ ($a = 0.3905$ nm) substrate. Strain states of the bilayer film are sketched in Fig. 1(d).

Atomic force microscopy measurement was performed on the surface of the bilayer to characterize the surface morphology and calculate its root-mean-square (*rms*) surface roughness. Fig. 2 shows the atomic force microscopy topographic image of the PSMO/LCMO bilayer on $SrTiO_3$ substrate. The scan is revealed by using an area of 2 μm × 2 μm. The surface is formed with several islands and their average width is ~105 nm. The *rms* surface roughness is ~0.92 nm and the average amplitude between peaks and valleys is ~9 nm.

Fig. 3 shows the resistance as a function of temperature for two reference single layer film of PSMO-3 and LCMO-5 ($d_{PSMO-3} = d_{LCMO-5} = 50$ nm) without and with a magnetic field of 5 T. For the LCMO-5 film, at zero magnetic field, temperature-dependent resistivity (ρ ~ T) shows thermally activated behavior down to about 160 K (T_{CO}) and then an upturn can be observed, indicating the existence of CO state accompanied with an FM to AFM transition [20]. The metallic FM and insulating AFM clusters coexist in LCMO-5 film below T_{CO}. In the ρ ~ T curve, there is a characteristic temperature T* indicated with arrow at about ~100 K. At T*, the tendency of resistivity with temperature changes abruptly. Under 5 T, the resistivity of LCMO-5 film decreases and T_{CO} in LCMO-5 disappears, clearly indicating that CO is weakened under the applied magnetic field. Further decreasing temperature, FM clusters start to percolate, and then a metallic-like transport behavior is observed below T*. In addition, magnetism measurements [see Fig. 7], from another angle, confirm the existence of FM cluster percolation (this will be discussed below). For PSMO-3 film, it shows a transition from metal to insulator at classical MI transition temperature T_{MI} ~ 256 K, which is consistent with the transition temperature of bulk PSMO-3 [21]. Note that the resistivity of LCMO-5 film is more than one hundred times larger than that of PSMO-3