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Spin wave and percolation studies in epitaxial $La_{2/3}Sr_{1/3}MnO_3$ thin films grown by pulsed laser deposition

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ABSTRACT

We investigate the magnetic and transport properties of high quality $La_{2/3}Sr_{1/3}MnO_3$ thin films grown by pulsed laser deposition. X-ray diffraction shows that the deposited films are epitaxial with the expected pseudo-cubic structure. Using the spin wave theory, the temperature dependence of magnetization was satisfactory modeled at low temperature, in which several fundamental magnetic parameters were obtained (spin wave stiffness, exchange constants, Fermi wave-vector, Mn–Mn interatomic distance). The transport properties were studied via the temperature dependence of electrical resistivity [$\rho(T)$], which shows a peak at Curie temperature due to metal to insulator transition. The percolation theory was used to simulate $\rho(T)$ in both the ferromagnetic and paramagnetic phases. Reasonable agreement with the experimental data is reported.

1. Introduction

$La_{2/3}Sr_{1/3}MnO_3$ (LSMO) has been extensively studied in recent years due to its particular physical properties, with high potential applications in spintronic devices [1], such as magnetic memory-storage cells or magnetic-field sensors. This compound exhibits some interesting properties, such as a half-metallic character, colossal magnetoresistance, and a high Curie temperature ($T_{C,bulk}$ = 370 K) [2–5]. In such systems, the magnetic interaction is described by the double-exchange mechanism between the Mn^{3+} and Mn^{4+} ions [6], where the coupling is induced by the electron hoping between two partially filled d shells. The existence of $Mn^{3+}$and Mn^{4+} mixed valence is essential to interpret both the ferromagnetism and metallic conductivity states. However, it was demonstrated that the double-exchange interaction alone cannot fully explain the magnetic interaction in LSMO. It was suggested that other mechanisms may influence the exchange interaction, such as the charge ordering, the size and the disorder at the cation site [7,8], the oxygen vacancies [9] or the polarons effect induced by strong electron–phonon interaction originating from the Jahn–Teller distortion [10]. On the other hand, a relationship between electrical and magnetic properties has been demonstrated. In fact, LSMO exhibits a metal-insulator transition, which is accompanied by a ferromagnetic to paramagnetic transition evidencing the strong correlation between the magnetic and transport properties. However, the simulation of the change of resistivity as a function of temperature is limited by the metal-to-insulator transition. To be able to model $\rho(T)$ in the entire temperature window, Li et al. [11] have developed a percolation model based on phase segregation, which we use in this present study. In order to better understand the properties and exploit LSMO for applications a detailed fundamental understanding of the magnetic and electrical properties is needed [12,13]. Here, we report on the magnetic and transport properties of high quality LSMO thin films using spin wave and percolation theories, respectively [14–16].

2. Experimental methods

LSMO thin films were deposited using pulsed laser deposition technique on $SrTiO_3$(001) (STO) substrates [17]. The background pressure was about 10^{-8} mbar. Before deposition, the substrate was heated at 900 °C for 30 min to clean the surface. During growth, the substrate temperature was kept at 750 °C and the oxygen pressure at 0.5 mbar. A KrF excimer laser (λ=248 nm), with a frequency of 10 Hz and an energy density of 1.5 J/cm^2 was used for ablation [18,19]. The film thicknesses were about 38 nm. The LSMO target was prepared via conventional solid state reaction technique using stoechiomteric mixture of La_2O_3, $SrCO_3$, and $MnCO_3$ powders used as starting materials. The mixture was

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annealed at 1200 °C during 16 h. Then, the obtained powder was pressed into a disk by applying a pressure of 600 bar, and sintered at 1450 °C during 16 hours in air. The crystalline structure was investigated by X-ray diffraction (XRD) using a Rigaku Smartlab (9 kW) diffractometer equipped with a monochromatic Cu $K\alpha_1$ source (λ=0.154056 nm) and a Ge(220) 2-bounce front monochromator. Magnetic measurements were carried out on a superconducting quantum interference device (SQUID) magnetometer. Electrical resistivity as a function of temperature was carried out using a four-probe method.

3. Results and discussion

Fig. 1a shows the XRD pattern of a LSMO thin film grown on $SrTiO_3$(001) substrate. Only the (0 *k* 2*l*) reflections of the rhombohedral LSMO phase[i] are observed suggesting that the film has a well defined texture. No spurious phases could be identified in the detection limit of the XRD technique. The average grain size along the growth axis was calculated using Debye–Scherrer formula: $G=\frac{0.9\cdot\lambda}{\gamma\cdot\cos\theta}$ where, G is the grain size, λ is the wavelength of the x-ray source, γ is the full width at half maximum (FWHM) of the diffraction peaks. In order to determine the FWHM, the LSMO (036) peak was fitted using a Gaussian function. The calculated grain size is found to be 25 ± 2 nm. In order to check if there is an in-plane structural relation between the substrate and the layer, ϕ scans were performed for both the substrate and the layer. For this purpose the {2,0,2} and {1,1,3} plane families of STO and LSMO were analyzed, respectively. The choice of the {1,1,3} LSMO plane family is motivated by the fact that no STO peak can be found nearby. As expected the scan showed four peaks corresponding to the cubic (fourfold symmetry) structure of STO (Fig. 1b). Although the symmetry of the LSMO structure is lower, four peaks are observed as well, suggesting the existence of at least two variants. It appears that there is an in-plane relation between the substrate and the layer, and therefore, LSMO is perfectly epitaxied on the STO substrate. This epitaxial relation can be written (for two LSMO variants) as $[1\pm10]STO(001)\|[2-10]LSMO(012)$. This relation is compatible with the very small lattice mismatch of about 0.5% existing at the STO(001)/LSMO(012) interface and with the almost square lattice inside the LSMO(012) plane (angles of 90.4° and 89.6° instead of 90°).

The change of magnetization as a function of temperature of a LSMO thin film is shown in Fig. 2. The measurements were made upon warming the film after a zero field cooling (ZFC) starting from 5 K in a field of 500 Oe parallel to the [100]$SrTiO_3$(001) direction. The T_C is found to be around 340 K. This value is lower than that reported for LSMO bulk samples (~370 K). Song, et al. [20] have shown that T_C depends sensitively on the deposition parameters such as the oxygen pressure during growth or the laser beam defocus on the LSMO target. Thus, the lower T_C can be attributed to these parameters.

For ferromagnetic materials, the temperature dependence of magnetization follows Block's law ($T^{3/2}$) which is associated with the thermal excitation of spin waves. Using spin wave theory, the change of magnetization as a function of temperature for ferromagnets can be written as [21]:

$$\frac{M(5\text{ K})-M(T)}{M(5\text{ K})}=BT^{3/2} \quad (1)$$

[i] Although the structure of LSMO is rhombohedral (R-3c), the crystalline structure of LSMO is often described as pseudocubic. In this case the (024)LSMO peak in the rhombohedral notation corresponds to the (002)LSMO peak in the pseudocubic notation.

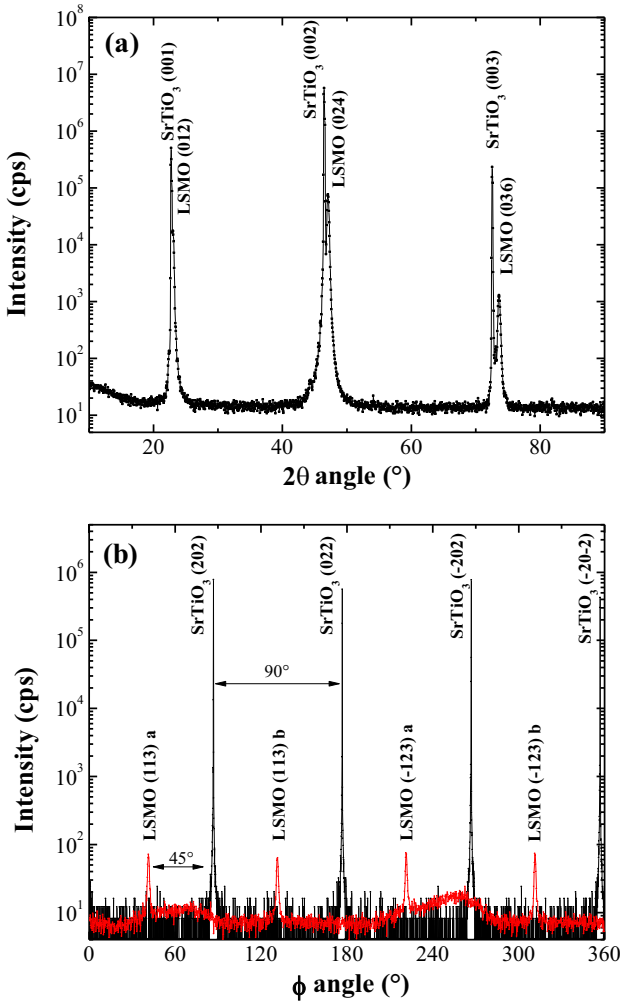

Fig. 1. (a) X-ray diffraction pattern of a 25-nm-thick LSMO film grown on $SrTiO_3$(001) substrate. (b) ϕ Scans of the STO {2,0,2} and LSMO {1,1,3} planes showing 90° periodicities for the monovariant STO and multivariant (a and b) LSMO.
Note: Although at first glance the (113) and (−123) LSMO planes do not seem to be part of the same plane family, this is indeed the case. In the four indices notation (hexagonal structure) these planes are (11–23) and (−12–13) which are indeed part of the same plane family.

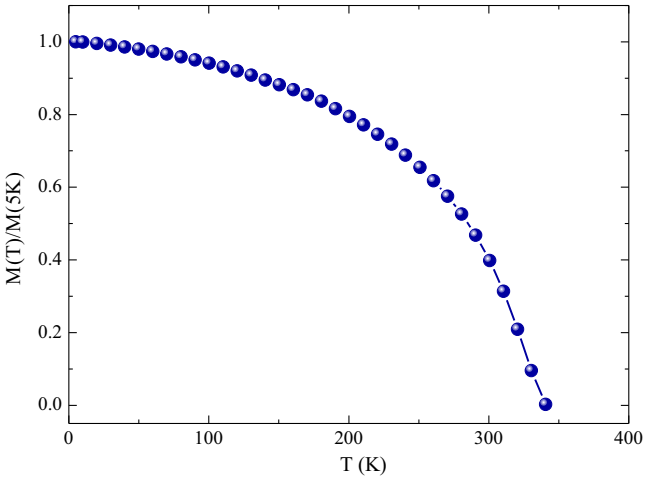

Fig. 2. Temperature dependence of normalized magnetization for LSMO thin films obtained at an applied magnetic field of 500 Oe. The magnetic field was parallel to the [100]$SrTiO_3$(001) direction.