Electrical Transport and Magnetic Properties of $La_{2/3}Pb_{1/3}MnO_3$ Thin Films

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The magnetic and transport properties of epitaxial $La_{2/3}Pb_{1/3}CuO_3$ thin films deposited on $SrLaGaO_4$ substrate using dc magnetron sputtering technique are reported. The giant magnetoresistance effect (of about 50% at magnetic field of 1 T) was observed near the Curie temperature. Several mechanisms responsible for temperature dependence of resistivity are discussed. The effect of annealing was studied. It shifted the Curie temperature to the lower value, probably, because of the loss of oxygen.

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1. Introduction

The $R_{2/3}A_{1/3}MnO_3$ (R = rare earth, A = alkaline earth or lead) compounds often exhibit a large magnetoresistance associated with a metal–insulator transition, which can take place at temperatures from about 30 K to above than 300 K [1–5]. Interest in the perovskite manganites has considerably increased since their fabrication as epitaxial thin films [6]. Until recently, much of the experimental work on the manganites has been devoted to the compounds $La_{2/3}A_{1/3}MnO_3$ with A being alkaline earth, while the properties of epitaxial $La_{2/3}Pb_{1/3}MnO_3$ films have not yet been reported. At the same time it has to be mentioned that the giant magnetoresistance was observed a long time ago [7] in $La_{1-x}Pb_xMnO_3$ single

crystals. This material is unique among the manganites since for $0.26 < x < 0.4$ the Curie temperature is above 320 K. Recently, it has been shown [8–10] using magnetic resonance technique that the magnetic mixed-phase state takes place in $La_{0.7}Pb_{0.3}MnO_3$ single crystals in the vicinity of T_c.

2. Experimental

We present in this paper the measurements of electric transport and magnetic properties of epitaxial $La_{2/3}Pb_{1/3}MnO_3$ thin (≈ 2000 Å) films. The films were grown in 15 mTorr argon–oxygen mixture (1:1) by dc magnetron sputtering at deposition rate of ≈ 0.1 nm/s, plasma current of 100 mA, and substrate-to-target distance of 7 cm. The target of nominal stoichiometry was fabricated by reaction of high purity component oxides at 900°C (36 h) followed by grinding, pressing into disk form (40×1 mm^2) and sintering at 1150°C (10 h). X-ray diffraction measurements indicate a single-phase orthorombically distorted perovskite structure of the films. The films were grown on (001)-oriented $SrLaGaO_4$ substrates at a temperature $T_{sub} = 675$°C. After a deposition the films were cooled to room temperature in oxygen (250 Torr) at the rate of 5 K/min. Next, one of the studied films was annealed at 800°C for 3 h in oxygen flow. The crystal orientation of the studied films was determined by X-ray diffraction measurements. The films were found to be (100)-oriented and to exhibit epitaxial in-plane alignment.

The electrical resistance of the samples was measured as a function of temperature and magnetic field using a standard four-probe dc technique. The measurements were performed with the external field applied parallel to the current direction. Magnetization was measured in a magnetic field up to 5 T over a temperature range of 5–300 K using a SQUID magnetometer (MPMS-5, Quantum Design).

3. Results and discussion

The temperature dependences of magnetization $M(T)$ in an applied field of 0.1 T, for the as deposited and annealed samples, are shown in Fig. 1. The ferromagnetic Curie temperature T_c was determined by linear extrapolation of a maximal slope of the $M(T)$ data to zero. Using this procedure we obtained $T_c = 211$ K for the as deposited film and $T_c = 176$ K for the annealed film. The obtained values of T_c are considerably smaller than $T_c = 355$ K reported for $La_{0.7}Pb_{0.3}MnO_3$ single crystal [7]. The shift of T_c to lower temperatures after annealing is also quite unexpected. It is in contrast to what is observed for $La_{2/3}Ca_{1/3}MnO_3$ films deposited on $LaAlO_3$ [11]. A possible explanation of both effects should, probably, be related to the oxygen deficiency. Similar effect was observed in $La_{0.6}Pb_{0.4}MnO_{3-\delta}$ thin films [12].

The temperature dependences of the resistivity $\rho(T)$ both for the as deposited and annealed films are shown in Fig. 2. The maximum of resistivity occurs
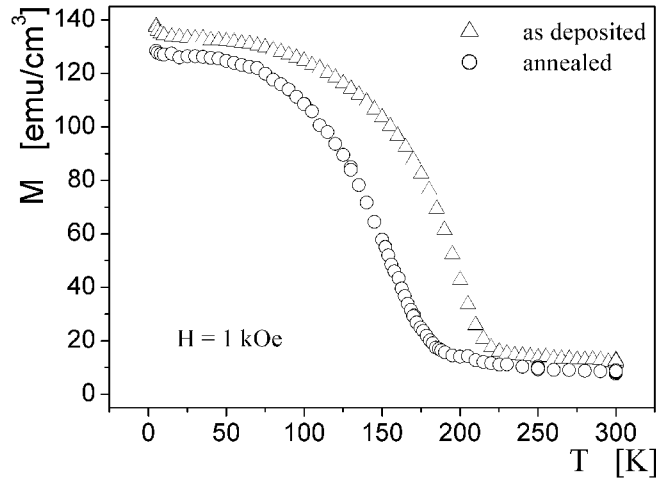
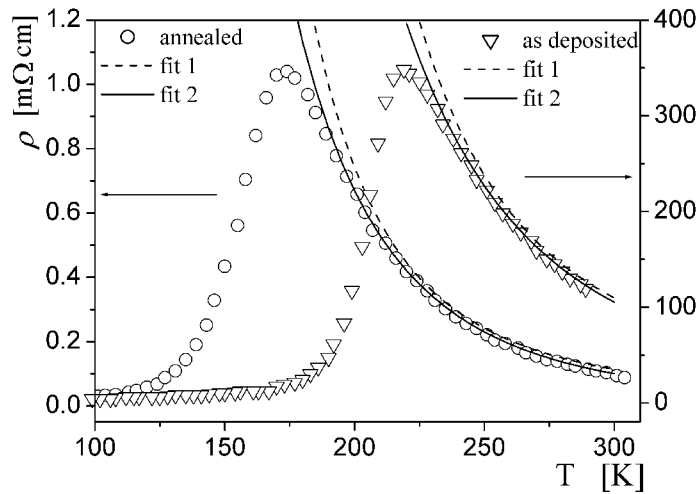Fig. 1.   Magnetization vs. temperature for as deposited ($\triangle$) and annealed (o) films.

Fig. 2.   Resistivity vs. temperature for as deposited (∇) and annealed (o) films. The lines present least squares fits: continuous — small polaron hopping model, dashed — semiconductor model.

very close to T_c determined from the magnetization measurements. One can see that the drop in ρ below T_c is quite sharp, while above T_c it is much less pronounced. This is caused by different mechanisms being responsible for the scattering processes in the temperature ranges below and above T_c. We have considered here two transport mechanisms for $T > T_c$: (i) semiconductor-like mechanism [13] (see fit 1 in Fig. 2) and (ii) hopping of small polarons in adiabatic regime (see fit 2 in Fig. 2). The resistivity in the frame of hopping of small polaron model in

adiabatic regime is described by

$$\rho(T) = T \exp\left(\frac{E_{\mathrm{a}}}{kT}\right), \tag{1}$$

where E_{a} is the activation energy. The activation energy determined as parameter of the fitting procedure was found to be 113 meV and 100 meV for as deposited and annealed film, respectively.
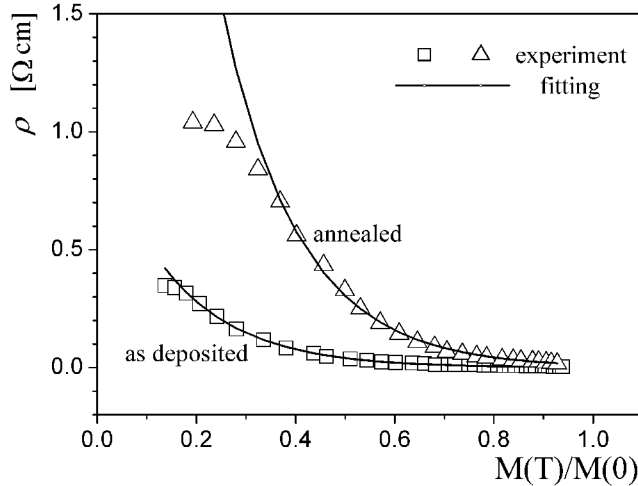

Fig. 3. Correlation between resistivity and magnetization for as deposited ($\square$) and annealed ($\triangle$) films. The lines present fits of Eq. (3) to experimental data. Fitting parameters $\rho_{\mathrm{m}} = 1013$ and 7756, $K = 6.43$ and 6.48 for the as deposited and annealed films, respectively.

For the ferromagnetic region ($T < T_{\mathrm{c}}$) we have confirmed the existence of an interesting correlation between magnetization $M(T)$ and $\rho(T)$, which was observed in $\mathrm{La}_{0.7}\mathrm{Ca}_{0.3}\mathrm{MnO}_3$ thin films [14], and can be expressed as

$$\rho(T) = \rho_{\mathrm{m}} \exp[-M(T)/W], \tag{2}$$

where W and ρ_{m} are fitting parameters. It is suggested in Ref. [14] that the transport below T_{c} described by Eq. (2) involves quasiparticle hopping. By transforming Eq. (2) to

$$\rho(T) = \rho_{\mathrm{m}} \exp\left[-K\frac{M(T)}{M(0)}\right], \tag{3}$$

it can be shown (see Fig. 3) that Eq. (3) describes correctly experimental results both for the as deposited and annealed films with practically the same values of parameter K (6.43 and 6.48, respectively).

The temperature dependences of the magnetoresistance effect $(\rho_0 - \rho_{1T})/\rho_0$, in the magnetic field of 1 T, are shown in Fig. 4, both for the as deposited and annealed films.
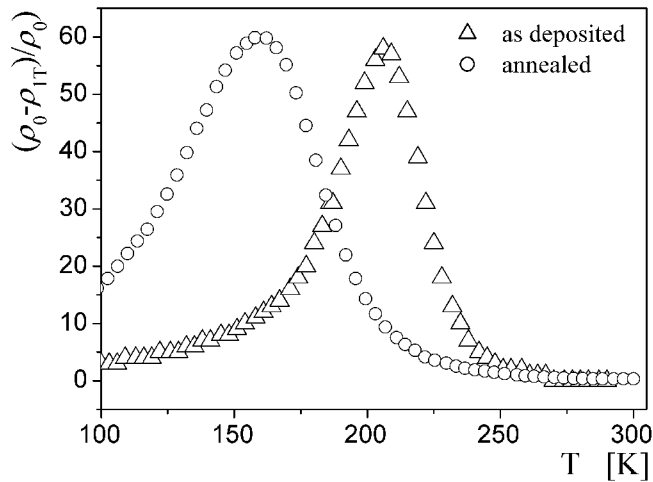
Fig. 4. Magnetoresistance effect vs. temperature for both studied films.

The results presented in this paper confirm the important role of quasiparticles in the determination of magnetic and transport properties of the investigated films.

To summarize, we have found giant magnetoresistance effect in $La_{2/3}Pb_{1/3}MnO_3$ films deposited on $SrLaGaO_4$ substrate. We suggest that the mechanism responsible for this effect is related to the different roles played by quasiparticles below and above the Curie temperature.

Acknowledgment

References

[1] R. von Helmolt, J. Wecker, B. Holzapfer, L. Schultz, K. Samwer, *Phys. Rev. Lett.* **71**, 2331 (1993).

[2] M. McCormack, S. Jin, T.H. Tiefel, R.M. Fleming, J.M. Phillip, R. Ramesh, *Appl. Phys. Lett.* **644**, 3045 (1994).

[3] S. Jin, T.H. Tiefel, M. McCormack, R.A. Fastnacht, R. Ramesh, L. Chen, *Science* **264**, 113 (1994).

[4] S. Jin, M. McCormack, T.H. Tiefel, R. Ramesh, *J. Appl. Phys.* **76**, 6929 (1994).

[5] J.Z. Sun, L. Krusin-Elbaum, S.S.P. Parkin, G. Xiao, *Appl. Phys. Lett.* **67,** 2726 (1995).

[6] H.L. Ju, C. Kwon, Q. Li, L. Greene, T. Venkatesan, *Appl. Phys. Lett.* **65,** 2108 (1994).

[7] C.W. Seale, S.T. Wang, *Can. J. Phys.* **48,** 2024 (1970).

[8] N.V. Volkov, G.A. Petrakovski, V.N. Vasilev, K.A. Sablina, *Fiz. Tverd. Tela* **44,** 1290 (2002).

[9] N.V. Volkov, G.A. Petrakovski, K. Sablina, K. Patrin, *Acta Phys. Pol. A* **105,** (2004).

[10] N.V. Volkov, G.A. Petrakovski, V.N. Vasilev, K.A. Sablina, K.G. Patrin, *J. Magn. Magn. Mater.* **258-259,** 302 (2003).

[11] D.C. Worledge, G.J. Snyder, M.R. Beasley, T.H. Geballe, *J. Appl. Phys.* **80,** 5158 (1996).

[12] K.M. Satyalaksmi, S. Sundar Manoharan, M.S. Hedge, V. Prasad, S.V. Subramanyam, *J. Appl. Phys.* **78,** 6861 (1995).

[13] G.J. Snyder, R. Hiskes, S. DiCarolis, M.R. Beasley, T.H. Geballe, *Phys. Rev. B* **53,** 14434 (1996).

[14] M.F. Hundley, M. Hawley, R.H. Heffner, Q.X. Jia, J.J. Neumeier, J. Tesmer, J.D. Thompson, X.D. Wu, *Appl. Phys. Lett.* **67,** 860 (1995).