Journal of the Korean Vacuum Society
Vol.12, No.S1, 2003, pp.100~103

Magnetoresistance behavior of $La_{1-x}Sr_xCoO_{3-\delta}$ films around the metal-insulator transition

J. S. Park, H. G. Park, C. O. Kim, Y. P. Lee†, and V. G. Prokhorov*

q-Psi and Dept. of Physics, Hanyang Univ., Korea
**Institute of Metal Physics, NASU, Ukraine*

Abstract

The magnetoresistance (MR) of $La_{1-x}Sr_xCoO_{3-\delta}$ films prepared by pulsed-laser deposition were investigated in order to clarify the magnetotransport properties around the metal-insulator transition. For the films in the metallic state ($x > 0.25$), the MR(T) manifests a small peak at the Curie temperature due to the spin-disorder scattering. The transition of the film into the insulating state ($x \leq 0.25$) is accompanied by an essential growth of the MR and results in a significant increase in the MR(T) with decreasing temperature, due to a phase separation into the ferromagnetic-metal clusters and the insulating matrix.

Keywords : Magnetic oxide film, Spin disorder scattering, Phase separation

1. Introduction

The discovery of colossal magnetoresistance (CMR) in manganites with perovskite structure has stimulated researches on compounds exhibiting a large magneto-resistance (MR) because of the scientific interests and the potential technological applications to new devices such as magnetic-read heads, field sensors and memories [1,2]. Recently, a large negative MR observed in $La_{1-x}Sr_xCoO_3$ (LSCO) thin films with $0.15 < x \leq 0.4$ has triggered a reinvestigation on this perovskite system [3].

The parent compound $LaCoO_3$ is a unique diamagnetic semiconductor with a spin gap of 30 meV, a charge gap of 0.1 eV and a rhombohedrally-distorted perovskite structure at low temperatures [4]. It exhibits remarkable transport and magnetic properties as a result of thermally induced transitions from a low-spin to high-spin or intermediate-spin state at the trivalent Co atoms [5]. Sr-substituted LSCO leads to a mixed valence of the Co ions, which gives rise to ferromagnetism for $x > 0.15$ and transition to the metallic state for $x > 0.2$ ($\equiv x_c$) [6]. The ferromagnetic interaction between Co^{3+} and Co^{4+} is supposed to arise from a double-exchange mechanism as in $La_{1-x}Sr_xMnO_3$ [7]. Since the magnetic and the transport properties of LSCO depend on the Co spin configuration, the MR is strongly influenced by the spin state of Co ions. Although various properties of LSCO have been widely studied, the MR behavior has been reported only a few [8,9] In order to clarify the mechanism of the MR around the metal-insulator transition, we investigated the magnetotransport properties of LSCO films.

2. Experiment

LSCO films were prepared by pulsed-laser deposition. Nd-YAG laser was employed with a wavelength of 1064 nm, a pulse duration of 7.8-10.5 ns, and an energy of 0.3-0.4 J/pulse. The film deposition was carried out at a pulse-repetition rate of 12 Hz. For the preparation

† E-mail : yplee@hanyang.ac.kr

of LSCO target, first of all, La_2O_3, SrO_3 and CoO powders were mixed in proportion to the stoichiometric composition and then annealed at 900℃ in air for 2 days. The cooled-down powders were then hot-pressed and heated at 1100℃ for 2 h to manufacture the targets. The substrate was $SrTiO_3$ (100) single crystal. The substrate temperature during deposition was ~ 850℃. The oxygen pressure in chamber was 450 mTorr during deposition and 750 Torr during cooling. At these conditions LSCO films were produced with a thickness of 200 nm. A specular θ-2θ x-ray diffraction (XRD) was performed using a Rigaku diffractometer with Cu-K_α radiation. The resistance measurements were carried out by using the four-point-probe method in a temperature range of 4.2-300 K and a magnetic field up to 5 T.

3. Results and Discussion

Figure 1 presents the θ-2θ XRD scans for films with four different compositions: x=0.15, 0.25, 0.35 and 0.5 (from the bottom in Fig. 1). A high intensity of the (00l) peak manifests that the deposition results in a highly c-oriented film. Therefore, in spite of the presence of (011), (022) and (112) peaks with much smaller intensities, all the samples can be treated as chemically-homogeneous epitaxial films.

Figure 2 shows the temperature dependence of resistivity, ρ(T), without (solid circles) and with (open

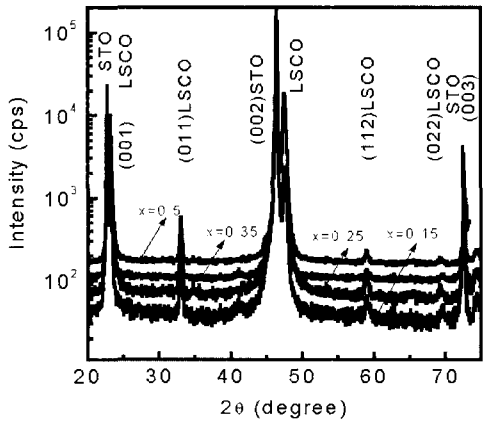

Fig. 1. θ-2θ XRD patterns of $La_{1-x}Sr_xCoO_{3-\delta}$ films.

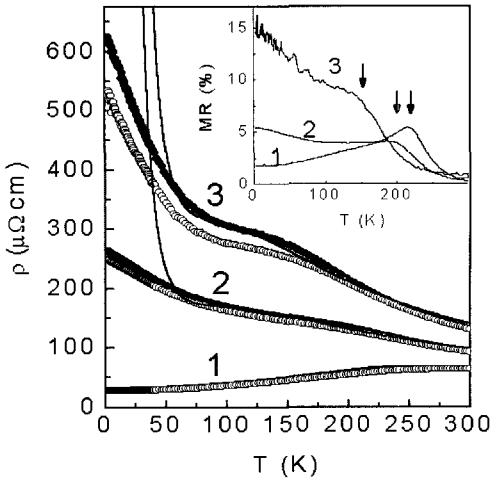

Fig. 2. Temperature-dependent resistivity of LSCO films with x=0.35 (1), 0.25 (2), and 0.15 (3), measured in a magnetic of 0 (solid circles) and 5 T (open circles).Solid lines are the theoretical curves. Inset displays the MR ratio for these films in a perpendicular magnetic field of 5 T.

circles) an applied magnetic field of 5 T for the as-deposited LSCO films. The film prepared with x=0.25 already manifests a semiconducting behavior of the conductivity. The reason for the difference in x_c between the film and the bulk can be ascribed to a lattice strain leading to a slight distortion of the crystal lattice and a reduction in the oxygen content during the deposition. Inset in Fig. 2 displays the temperature dependence of negative MR for the LSCO films. The MR value was estimated by using MR(%)=100%×[R(0)(R(H)]/R(0), where R(0) and R(H) are the film resistance without and with a magnetic field of 5 T, respectively. For the metallic film (x=0.35), the MR (T) has a peak (indicated by arrow) at 230 K, which is coincident with the published value for the Curie temperature [10]. This MR behavior of the metallic film is a normal result from the ferromagnetic metal, attributed to the spin-disorder scattering. The transition of films into the insulating state (x=0.15 and 0.25) is accompanied by a significant increase in the MR with decreasing temperature.

Figure 3 reveals a plot of $\ln\rho$-vs.-$T^{-1/4}$ for the insulating

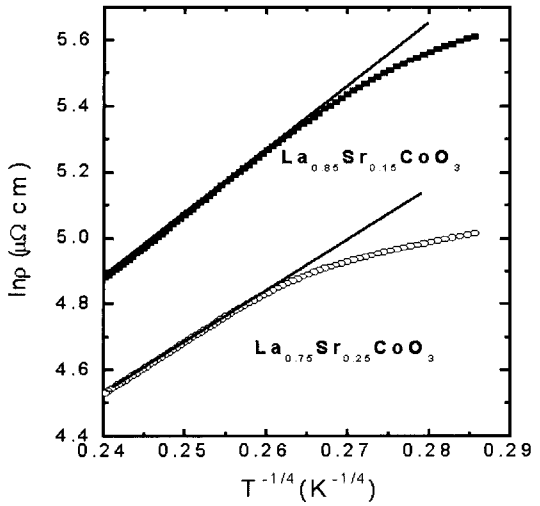

Fig. 3. $\ln\rho$-vs.-$T^{-1/4}$ plot for the insulating samples in a high temperature regime (150 K ≤ T ≤ 300 K).

films in a high temperature regime (150 K ≤ T ≤ 300 K). These curves are nearly linear for 200 K ≤ T ≤ 300 K. Therefore, we conclude that the transport of carriers at high temperatures in LSCO (x=0.15 and 0.25) is governed by the Mott-type variable-range hopping. There is a noticeable change in the slope near Tc. This can be interpreted as the onset of formation of the ferromagnetic metal (FM) clusters in the insulating matrix (IM). Consequently, these phase-separated compounds can be treated as a random resistor network with percolating FM clusters. Therefore, the analysis of ρ(T) was performed by using the two-parallel-resistor model, where one resistor corresponds to the FM clusters and the other to the IM. It is shown that the ρ(T) of insulating films is excellently fitted to the two-parallel-resistor model;

$$\frac{1}{\rho(T)} = \frac{n'f}{(\rho_0 + \alpha T^2)} + \frac{(1-f)}{\rho_\infty \exp[(T_0/T)^{1/4}]}$$

where $n' = \nu/[\nu + \exp(\Delta/k_B T)]$ is the normalized number of excited Co^{3+} ions from the low-spin to the high-spin state, ν=15 which is the multiplicity of the high-spin states, and Δ is the spin gap, and f is the fraction of the FM phase, which is equal to the Sr doping in our consideration. The first term describes the metallic behavior of resistivity through the FM-cluster channel, and the second describes the insulating behavior of resistivity, based on the Mott-type VRH in the IM. It is suggested that the MR of insulating state is increased by a phase separation into the FM clusters and the IM.

4. Conclusions

We have found two mechanisms of MR in the investigated films. In the metallic state (x > 0.25), the MR is caused by a change in the electron scattering with spin disorder, and exhibits a small peak at the Curie temperature. In the insulating state (x ≤ 0.25), the phase separation into the FM clusters and the IM occurs, and an applied magnetic field leads to a growth of the FM phase.

Acknowledgments

This work was supported by the KOSEF through Quantum Photonic Science Research Center, by a Korea Research Foundation Grant (2001-015-DS0015), and by MOST, Korea.

References

[1] S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **264**, 413 (1994).

[2] H. Y. Hwang, S.-W. Cheong, N. P. Ong, and B. Batlogg, Phys. Rev. Lett. **77**, 2041 (1996).

[3] T. Saitoh, T. Mizokawa, A. Fujimori, M. Abbate, Y. Takeda, and M. Takano, Phys. Rev. B **56**, 1290 (1997).

[4] S. Yamaguchi, Y. Okimoto, H. Taniguchi, and Y. Tokura, Phys. Rev. B **53**, R2926 (1996).

[5] R. R. Heikes, R. C. Miller, and R. Mazelsky, Physica **30**, 1600 (1964).

[6] V. G. Bhide, D. S. Rajoria, C. N. R. Rao, and R. G. Rama, Phys. Rev. B **12**, 2832 (1975).

[7] N. Furukawa, J. Phys. Soc. Jpn. **64**, 3164 (1995).
[8] R. Mahendiran, A. K. Raychaudhuri, A. Chainani, and D. D. Sarma, J. Phys.: Condens. Matter. **7**, L561 (1995).
[9] S. Yamaguchi, H. Taniguchi, H. Takagi, T. Arima, and Y. Tokura, J. Phys. Soc. Jpn. **54**, 1885 (1995).
[10] M. Itoh, I. Natori, S. Kubota, and K. Motoya, J. Phys. Soc. Jpn. **63**, 1486 (1994).