KMS-1-R2

Magnetic, magnetotransport and magnetocaloric properties of $La_{0.7}Ca_{0.3-x}Sr_xMnO_3$ manganites under the orthorhombic-rhombohedral phase transition

Alexander N. Ulyanov,[1] Jin-Sun Kim[1], Young-Min Kang,[1] Seong-Gi Min,[2] Kyu-Won Lee,[3]
Seong-Cho Yu,[2] and Sang-Im Yoo[1]*
[1]School of Materials Science and Engineering, Seoul National University, Seoul 151-744, Korea
[2]Department of Physics, Chungbuk National University, Cheongju 361-763, Korea
[3]Korea Research Institute of Standards and Science, Yusong, Taejon 305-600, Korea

Perovskite-like lanthanum manganites attracted a great attention last decades because of the colossal magnetoresistivity (CMR) effect at the metal-insulator transition, which is very close to Curie temperature, where the giant magnetocaloric (MCE) effect was observed. The magnetoresistivity effect was initially explained by the "double exchange" (DE) model [1]. According to Zener, electrons move between two partially filled 3*d* shells of Mn^{3+} and Mn^{4+} ions with strong on-site Hund's coupling. The DE model, revealing the connections between the electrical and magnetic subsystems of lanthanum manganites, was complemented by the electron-phonon interaction caused by the Jahn-Teller splitting of external *d*-level of manganese [2]. However, the nature of the CMR has not been fully clarified yet. The difficulties consist in a close connection between structure, electronic and magnetic properties of perovskite-type manganites. We present here a study of $La_{0.7}Ca_{0.3-x}Sr_xMnO_3$ compositions ($0.105 \le x \le 0.18$) with a small change in *x* to know the properties of manganites at the cross sections of both the orthorhombic-rhombohedral (*O-R*) structural phase transition and ferromagnetic-paramagnetic transition, where the spin-lattice interaction is known very strong

At room temperature, samples of x = 0.105, 0.12 and 0.135 were orthorhombic ***Pbnm*** phase, and those of x=0.15, 0.165 and 0.18 were rhombohedral $R\bar{3}c$ phase. The temperature dependence of magnetization, $M(T)$, for the samples (except the sample of x=0.135) showed a

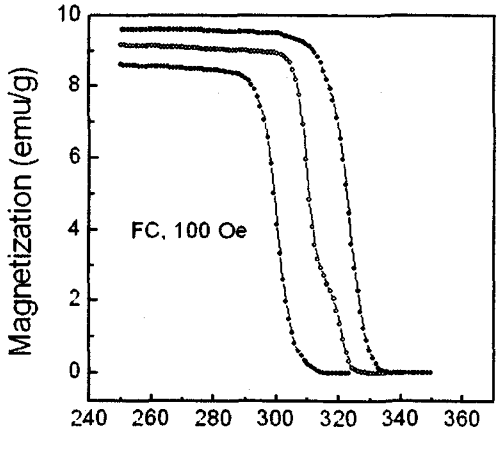

Fig.1. Field-cooled magnetization of $La_{0.7}Ca_{0.3-x}Sr_xMnO_3$ compositions (x=0.12, 0.135, and 0.15, from the left to the right).

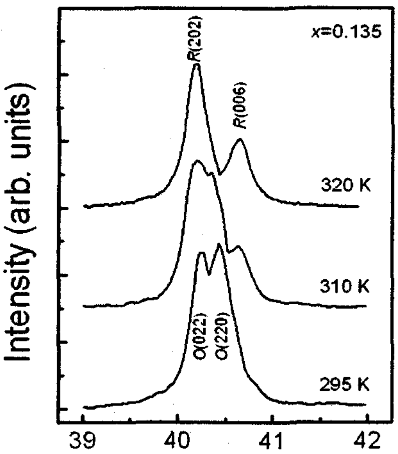

Fig.2. Some diffraction peaks for the x=0.135 composition at selected temperatures.

normal behavior (without any "kink") (see Fig. 1). However, The *M*(*T*) curve for x=0.135 sample revealed a unusual two-step dependence with the inflexion points at 309 and 320 K. For this kink behavior, we argue that the above inflexion points correspond to the Curie points of ***Pbnm*** and $R\bar{3}c$ phases, respectively since the structural orthorhombic- rhombohedral (*O*-*R*) phase transition occurs within the above temperature interval (see Fig.2).

The magnetic entropy changes $\Delta S_m(T,H) = S_m(T,H) - S_m(T,0) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH$ were calculated using measured isothermal *M*(*H*) curves, and presented in Fig. 3. The peak values of magnetic entropy change $\left|\Delta S_m^{\max}\right|$ are 1.87 J/kg K, 1.72 J/kg K, and 1.7 J/kg K for the samples of x=0.12, 0.135, and 0.15, respectively. The temperature dependence of ΔS_m for the x=0.135 sample shows two peaks, corresponding to Curie points of orthorhombic and rhombohedral phases.

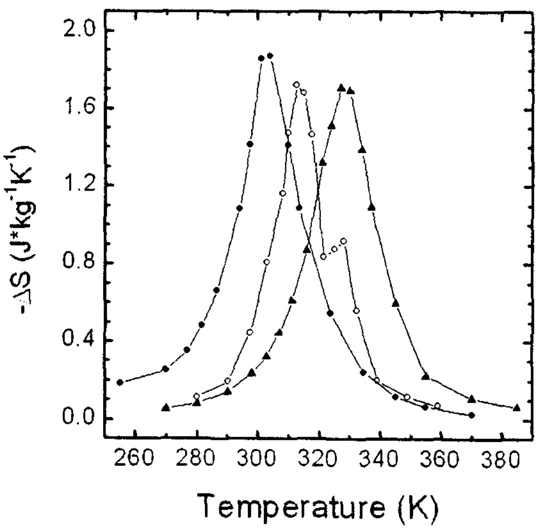

Fig. 3. Magnetic entropy change vs temperature for applied magnetic field changes of 0 to 2 T (x=0.12, 0.135, and 0.15, from the left to the right).

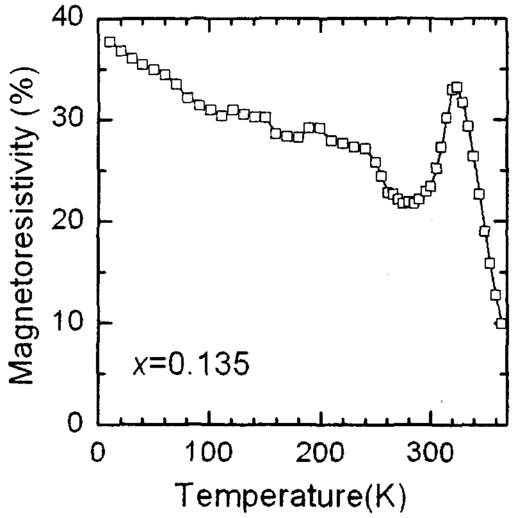

Fig. 4. Magnetoresistivity ratios of x=0.135 composition in field of 5 T.

Magnetoresistivity ratio (MR) in 5 T applied field, defined as MR = $-\{[\rho(T, H)-\rho(T, 0)]/\rho(T, 0)\}*100\%$, is shown in Fig. 4. No anomaly is observed near the structural phase transition point.

Our data, for the first time, show a direct evidence for higher Curie temperature in the rhombohedral phase than that in the orthorhombic phase, indicating that double exchange interaction is stronger in the *R*-phase than in the *O*-phase. This also confirms a close connection between the magnetic properties and crystal structure. Interestingly, it is found that the magnetocaloric property is strongly related to the change of crystal symmetry but the magnetoresistivity is insensitive to it.

[1] N. Zener, Phys. Rev. **82** (1951) 403.
[2] A.J. Millis, P.B. Littlewood, and B.I. Shraiman, Phys. Rev. Lett. 74 (1995) 5144.