Transport properties in $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$ system

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Abstract

We investigated the crystallographic, transport and magnetic properties of a strongly electron correlated $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$ series with x = 0-1.0. All our samples are in a rhombohedral $R\bar{3}c$ structure of lattice parameters $a \sim 0.550$ nm and $c \sim 1.338$ nm. We found that the addition of the larger Pb ion in $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$ system does not apparently affect on the metal-insulating and magnetic ordering temperatures as well as the magnetoresistance (MR) ratio. However, it reduces the sintered temperature and improves the electric conductivity. The MR ratios are monotonically linear decreasing with temperature from $\sim$ 25% at 5 K down to $\sim$ 6% at 300 K for all samples. The chemical disorder effect of the Pb^{2+} substitution does not change the magnetic properties at low temperatures. It can be contributed to exhibiting the constant hole concentration due to the same ratios of $[Ba^{2+}]$+$[Pb^{2+}]$/$[La^{3+}]$ and $[Mn^{4+}]$/$[Mn^{3+}]$ in these Pb doping manganese oxides. The cluster glass behavior was apparently observed, even for the parent sample of $x = 0$.

Key words: $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$; Magnetoresistance; Magnetic Properties

The discovery of colossal MR in $La_{1-x}Ca_xMnO_3$ [1] initiated intensive investigations of the $R_{1-x}A_xTO_3$ compounds (R = a trivalent rare earth metal; A = Ca, Sr, Ba, Pb; and T = a transition metal). In general, the perovskite-like $(R_{1-x}A_x)TO_3$ oxides with mixed-valence transition metal exhibit very rich phase diagrams. This is due to subtle competition among interactions involving spin, lattice, and charge degrees of freedom. All the energy scales of those interactions appear to be comparable of 1 eV.

In this work we studied the structural and physical properties of the compounds with nominal composition $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$ with $0 \leq x \leq 1$. All polycrystalline samples were prepared by a usual solid-state reaction method [3-5]. The samples were checked for phase purity by the X-ray diffraction (XRD). In addition, crystallographic parameters were obtained by the Rietveld structural analysis. The powder XRD patterns showed that the samples were almost single-phase in a rhombohedral $R\bar{3}c$ structure of lattice parameters $a \sim 0.550$ nm and $c \sim 1.338$ nm.

Dc magnetization and *ac* susceptibility were measured by an Oxford MagLab^{AC} 2000 susceptometer in the temperature range of 2-305 K under magnetic fields of -5 - +5 T. The results of magnetic measurements show that the addition of the larger Pb ion in $La_{0.7}(Ba_{1-x}Pb_x)_{0.3}MnO_{3+\delta}$ series does not apparently affect on both the metal-insulating and magnetic ordering temperatures as well as the magnetoresistance ratios. However, it can reduce the sintered temperature and improve the electric conductivity.

Fig. 1 shows the zero-field cooled (ZFC) and field-cooled (FC) *dc* magnetizations of all samples as a function of temperature in $H_{dc} = 100$ Oe. The $M(T)$ curves indicate that at least three magnetic transitions occur at various temperature ranges. We believe that the first transition is due to ordinary ferromagnetic ordering transition occurred at an onset temperature $T_C \sim$ 360 K, which is slightly dependent on the applied field. The second one shows strongly field-dependent over a wide temperature range ($T < T_C$). All the magnetic

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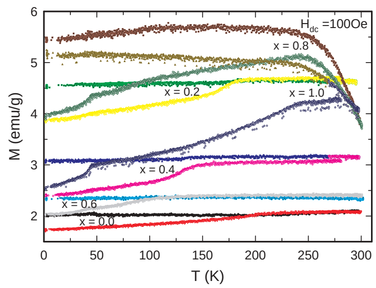

Fig. 1. ZFC and FC *dc* magnetizations as a function of temperature in $H_{dc} = 100$ Oe for all samples.

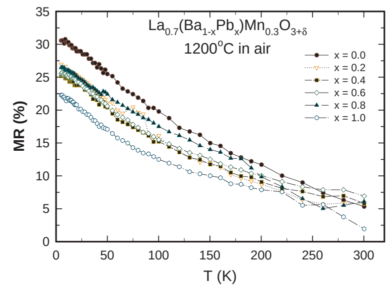

Fig. 3. The negative MR ratios versus temperature for all samples.

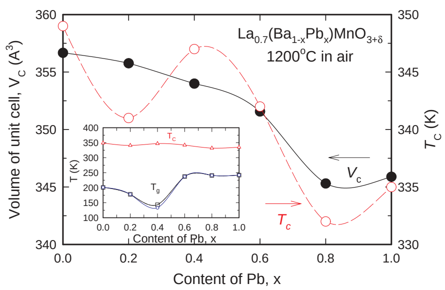

Fig. 2. V_c, T_C and T_g as a function of Pb content, x

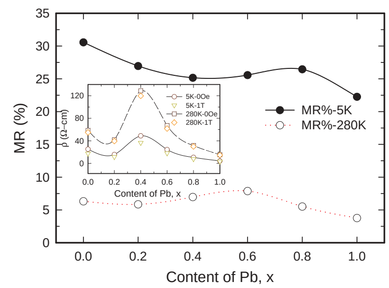

Fig. 4. The variation of negative MR ratio and the electrical resistivity (inset plot) with Pb content at $T = 5$ and 280 K.

features including irreversibility, remnant magnetization and scaling behavior indicate a canonical spin-glass transition in all samples. The cluster glass behavior was clearly observed in a wide temperature range, even for the parent sample of $x = 0$. In Fig. 2 we determined the unit cell volume V_c, Curie temperature T_C, irreversibility T_{irr} and glass transition temperatures T_g (in $H_{dc} = 100$ and 200 Oe), from their ZFC and FC *dc* magnetization measurements, as a function of Pb content. We also observed a field-independent AFM transition occurring at $T_N \sim 45$ K, which have been found in our samples $(La_{0.5}A_{0.5})MnO_3$ with $A =$ Sr and Pb [2,6]. The magnetic data suggest that, at low T, the samples are intimate mixtures of ferromagnetic and antiferromagnetic microregions.

We measured electrical resistivities of all samples as a function of temperature in $H = 0$ and 1 T using a four-points method. The negative MR ratios of all samples are monotonically linear decreasing with temperature down from $\sim 25\%$ at 5 K to $\sim 6\%$ at 300 K as shown in Fig. 3. The variations of the negative MR ratios and the electrical resistivity (inset plot) with Pb content, x, at $T = 5$ and 280 K are displayed in Fig. 4.

The chemical disorder effect of the Pb^{2+} substitution does not significantly change in magnetic properties and the magnetoresistance ratios. We belief that these properties may cause from the fixed hole concentration in all our samples because the ratios of $[Ba^{2+}]+[Pb^{2+}]/[La^{3+}]$ and $[Mn^{4+}]/[Mn^{3+}]$ are the same in these oxides.

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