Magnetic and transport properties of Dy substituted layered perovskite $La_{1.3}Sr_{1.7}Mn_2O_7$*

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The effect of Dy substitution for La site in layered manganese oxides $La_{1.3-x}Dy_xSr_{1.7}Mn_2O_7$ on the magnetic and electrical properties has been investigated. With the La^{3+} substituting by Dy^{3+}, the long range three-dimensional ferromagnetism transition and the insulator–metal transition disappear. These effects are attributed to the lattice distortion due to the substitution of the smaller Dy^{3+}. Addtionally, the small Dy^{3+} is inclined to occupy the R site which is in the rock-salt layer, then the distribution of La, Sr, Dy ions in Dy-doped sample should be more orderly than that in $La_{1.3}Sr_{1.7}Mn_2O_7$, so there is only one insulator–metal transition in the ρ–T curve of the sample with $x = 0.05$ and $x = 0.1$.

Keywords: magnetic property, electrical property, magnetoresistance, metal–insulator transition

PACC: 7530K, 7570P, 7130

1. Introduction

In the past few years, layered manganites of the type $La_{2-2x}Sr_{1+2x}Mn_2O_7$ [LSMO(327)] have been the subject of intensive investigations due to their colossal magnetoresistance (CMR) and a variety of the fascinating physical properties.[1–3] The structure of LSMO(327) is considered as the $n = 2$ member of the Ruddlesden–Popper series $A_{n+1}B_nO_{3n+1}$ in which two perovskite blocks composed by two-dimensional layers of BO_6 corner-sharing octahedral are separated by rocksalt AO layers.

As we know, the properties of the layered perovkites $Ln_{2-2x}M_{1+2x}Mn_2O_7$ are very sensitive to the lanthanide ions. The La-based perovskites $La_{2-2x}Sr_{1+2x}Mn_2O_7$ ($x = 0.3$–0.4) are ferromagnetic (FM),[4] but the Tb-based or Dy-based layered perovkites are paramagnetic.[5] The Gd-doped layered manganite $(La_{0.8}Gd_{0.2})_{1.4}Sr_{1.6}Mn_2O_7$ shows the nonmagnetic spin-glass-like state below 30 K.[6] Partly doping La with Nd and Eu results in the weakening and disappearing of long-range FM ordering.[7] So the different ionic sizes of the Ln site and the Ln site disorder greatly affect the magnetic properties.

In this report we present the effect of Dy substitution for La on the magnetic and transport properties in the $La_{1.3}Sr_{1.7}Mn_2O_7$.

2. Experiment

The samples of $La_{1.3x}Dy_xSr_{1.7}Mn_2O_7$ ($x = 0$, 0.05, 0.1 and 0.2) were prepared by the conventional solid-state reaction method. The oxides La_2O_3, $SrCO_3$, $MnCO_3$ and Dy_2O_3 (with purities higher than 99.9%) were weighed in stoichiometric amount, then mixed and prefired at 1000 °C for 24 h. Then the powder was ground and sintered in air repeatedly until single-phased samples were obtained. The phase purity and the crystal structure for these samples were examined by a Rigaku x-ray diffractometer. The magnetic measurements were performed by a superconducting quantum interface device magnetometer (Quantum Design, MPMS-7). The resistivity measurements were carried out using the standard four-point technique. The magnetoresistance (MR) is defined as the following relation:

$$\mathrm{MR}(\%) = \frac{\rho(0) - \rho(H)}{\rho(0)} \times 100$$

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where $\rho(0)$ and $\rho(H)$ are the resistivities at zero field and applied field, respectively

3. Results and discussion

Figure 1 shows the x-ray diffraction (XRD) patterns for the $La_{1.3}Sr_{1.7}Mn_2O_7$ sample as a representative pattern. All samples with x in the range of 0–0.2 can be indexed to an $Sr_3Ti_2O_7$-type tetragonal perovskite structure with a space group $I4/mmm$. The crystal structure was refined by the Reitveld program,

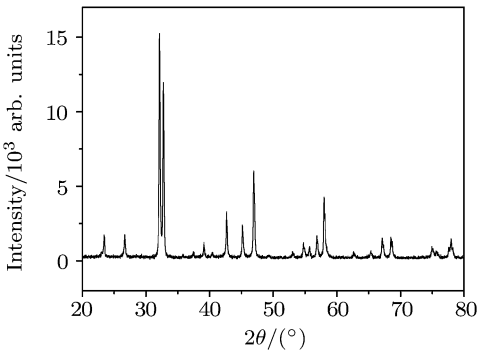


Fig. 1. The x-ray diffraction patterns of $La_{1.3}Sr_{1.7}Mn_2O_7$.

and the fitting results show that the cell parameters of $La_{1.3-x}Dy_xSr_{1.7}Mn_2O_7$ decrease gradually with La^{3+} being replaced by smaller Dy^{3+} which are presented in Table 1.

Table 1. Lattice parameters and c/a ratio for $La_{1.3-x}Dy_xSr_{1.7}Mn_2O_7$, (1 Å=0.1 nm).

x	a/Å	c/Å	c/a
0.0	3.8742	20.0819	5.1835
0.05	3.8731	20.0821	5.1850
0.1	3.8693	20.0758	5.1885
0.2	3.8653	20.0708	5.1926

Figure 2 shows the temperature dependence of magnetization under applied magnetic field of 0.1 T for $La_{1.3-x}Dy_xSr_{1.7}Mn_2O_7$. For the undoped sample, a clear increase in magnetization is observed in the temperature range of 200 K $\leq T \leq$ 250 K, which indicates that the two-dimensional (2D) short-range FM ordering is formed around the temperature $T^* \sim$ 225 K. With further decreasing temperature in the temperature range 100 K $\leq T \leq$ 125 K, the magnetization increases rapidly, forming a three-dimensional (3D) long-range FM ordering at $T_C \sim$ 120 K. Similar phenomenon is also observed in Refs. [2], [7] and [8].

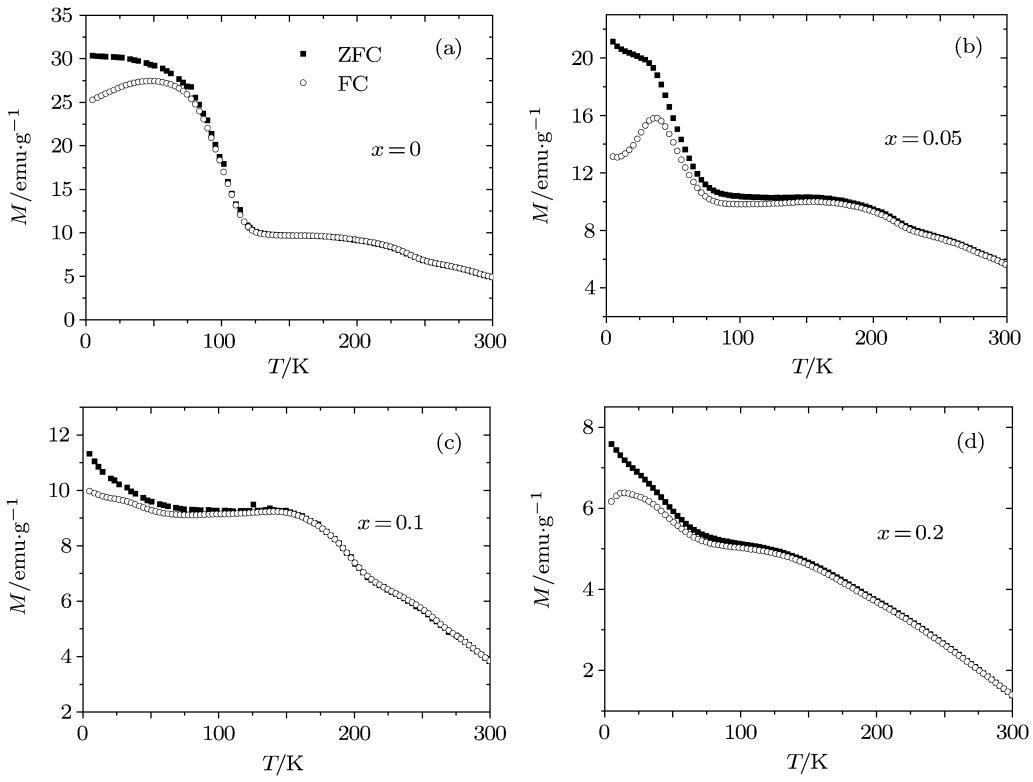


Fig. 2. Temperature dependence of the magnetization M in a 0.1-T field for the samples. ZFC and FC present zero field cooling and field cooling. (a) $x = 0$, (b) 0.05, (c) 0.1, and (d) 0.2.

The two transition temperatures are related to the anisotropic exchange interaction. The higher 2D transition temperature T^* results from the in-plane exchange interaction, and the lower 3D transition T_C temperature results from the out-plane exchange interaction.[2,9] When $x = 0.05$, the magnetic transition temperatures T^* and T_C decrease to 200 K and 75 K, respectively. Then, the ZFC and FC curves exhibit a characteristic splitting below 35 K, indicating the presence of a low temperature spin-glass state as in ABO_3-type perovskites. When $x = 0.1$, 0.2, there is no long-range magnetic ordering at low temperatures, the transition temperature T^* decreases to 170 K for $x = 0.1$, 100 K for $x = 0.2$.

Temperature dependence of resistivities and MR for $La_{1.3-x}Dy_xSr_{1.7}Mn_2O_7$ at 0 T and 5 T are shown in Fig. 3. We find that the resistivities of the samples show a peak with $x = 0$ at 126 K and $x = 0.05$ at 87 K. It indicates that there is an insulator–metal (MI) transition in the samples. For $x \geq 0.1$, the MI transition disappears and the resistivities dramatically increase. At the same time, the applied field greatly suppresses the resistivities for all samples. An application of 5-T field drives an MI transition for $x = 0.1$, but the sample of $x = 0.2$ is still insulating at 5 T. This is because an applied field can suppress the magnetic frustration and the spin scattering of electrons, so the resistivity reduces and drives an MI transition. Moreover, the ρ–T curve in 5 T shows two MI transition temperatures (signed in Fig. 3 with peak A and peak B) for $x = 0$. Similar curve was reported by Liu *et al.*[10] This is because the La or Sr ion has two kinds of occupation sites, a 12-coordinate site (P-site) in the MnO_2 bilayer and a 9-coordinate site (R-site) in the rock-salt layer. The small-sized ion preferentially occupies the R site, but the radii of La^{3+} and Sr^{2+} are close to each other, so the distribution of La^{3+} and Sr^{2+} at P-site and R-site are disorder, the compound can exist in two phases. Then the two peaks of ρ–T curve corresponding two phases. With La^{3+} replacing by Dy^{3+}, the small Dy^{3+} is inclined to occupy the R site, then the distribution of La, Sr, Dy ions in Dy-doped sample should be more orderly than that in $La_{1.3}Sr_{1.7}Mn_2O_7$, so there is only one peak in the ρ–T curve of the sample with $x = 0.05$ and $x = 0.1$.

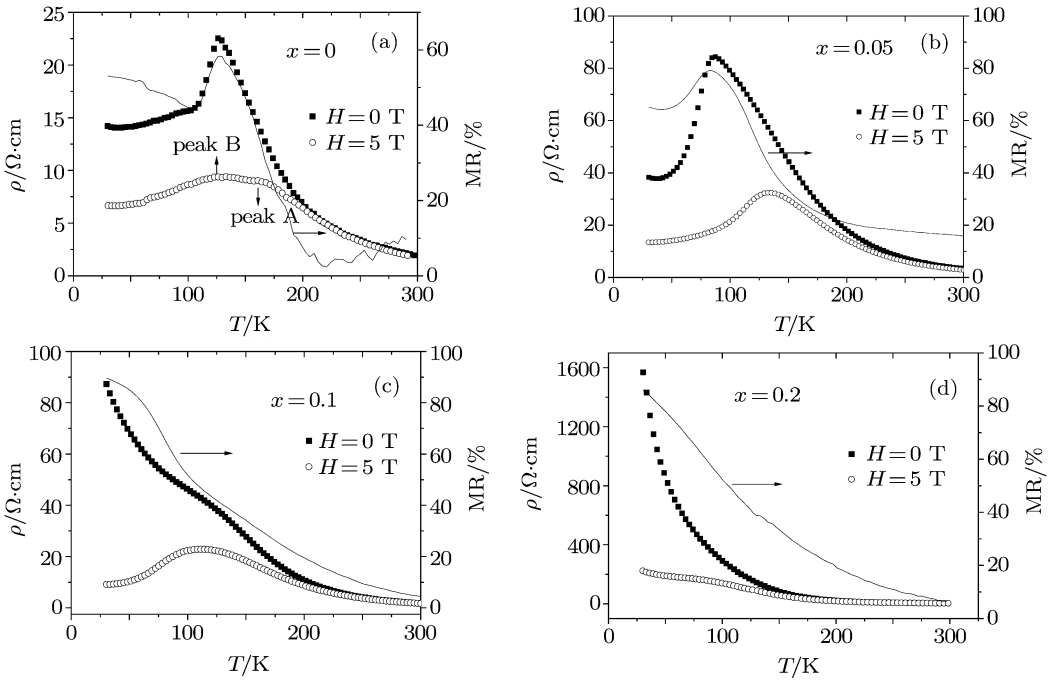


Fig. 3. Temperature dependence of resistivities and magnetoresistance for the samples at the field of the 0 T and 5 T with (a) $x = 0$, (b) 0.05, (c) 0.1 and (d) 0.2.

For undoped sample, the MR maximum is observed at the peak temperature $T_{MR} = 126$ K, which agrees well with the MI transition temperature. The temperature of the MR maximum decreases to 81 K for $x = 0.05$, and the MR maximum reaches 79% which is bigger than the undoped sample. But the MR maximum disappeared over the whole temperature range for $x = 0.1$, 0.2. The results described above can interpret as substituting Dy for La decreases the average ionic radius, then results in the lattice distortion, and consequently, electrons

become more localized, the mobility of electrons is reduced, so the maximum temperatures of MI and MR gradually disappear.

4. Conclusion

The effect on magnetic and transport properties of $La_{1.3}Sr_{1.7}Mn_2O_7$ by substituting La site with Dy has been investigated. It is found that the Dy substitution for La greatly affects the magnetic and transport properties of the parent compound $La_{1.3}Sr_{1.7}Mn_2O_7$. With increasing Dy doping level, the long range 3D FM is weakened and gradually disappearing. For the lightly Dy^{3+}-doped samples, the MI transition occurs accompanying the 3D FM transition and they display the CMR which is the common phenomenon in the mixed-valent manganese oxides. For the heavily Dy^{3+}-doped samples, the MI transition disappears. These effects are attributed to the special lattice distortion due to the substitution of the smaller Dy^{3+}.

References

[1] Moritomo Y, Asamitsu A, Kuwahara H and Tokura Y 1996 *Nature* **380** 141

[2] Kimura T, Tomioka Y, Kuwahara H, Asamitsu A, Tamura M and Tokura Y 1996 *Science* **274** 1698

[3] Argyriou D N, Mitchell J F, Radaelli P G, Bordallo H N, Cox D E, Medarde M and Jorgensen J D 1999 *Phys. Rev.* B **59** 8695

[4] Kubota M, Fujioka H, Ohoyama K, Hirota K, Moritomo Y, Yoshizawaand H and Endoh Y 1999 *J. Phys. Chem. Solids* **60** 1161

[5] Battle P D, Green M A, Laskey N S, Kasmir N, Millburn J E, Spring L E, Sullivan S P, Rosseinsky M J and Vente J F 1997 *J. Mater. Chem.* **7** 977

[6] Joonghoe Dho, Kim W S and Hur N H 2001 *Phys. Rev.* B **65** 024404

[7] Zhang J, Wang F W, Zhang P L and Yan Q W 2000 *Mater. Sci. Eng.* B **76** 6

[8] Wang A H, Liu Y, Zhang Z Y, Long Y and Cao G H 2004 *Solid State Commun.* **130** 293

[9] Asano H, Hayakawa J and Matsui M 1997 *Phys. Rev.* B **56** 5395

[10] Liu L, Xia Z C and Yuan S L 2006 *Mater. Sci. Eng.* B **127** 55