Magnetic and transport properties of bilayered manganites $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ ($x = 0, 0.08$)*

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The magnetic and electrical properties of nonmagnetic Ga^{+3} ion substitution for Mn site are investigated in the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. When the Mn is substituted by Ga, the ferromagnetic property obviously weakens, the magnetic transition temperature decreases and a spin-glass behaviour occurs at low temperature. Meanwhile, doping causes the resistivity to dramatically increase, the metal–insulator transition temperature to disappear, and a greater magneto-resistance effect to occur at low temperature. These effects result from the fact that Ga substitution dilutes the magnetic active Mn–O–Mn network and weakens the double exchange interaction, and further suppresses ferromagnetic ordering and metallic conduction.

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

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1. Introduction

In the 1990s, Moritomo *et al.*[1] discovered rich physical properties such as the colossal magneto-resistance (CMR) effect in the bilayer compound $La_{1.2}Sr_{1.8}Mn_2O_7$. These discoveries were followed by intensive study of bilayered manganites $A_{2-2x}B_{1+2x}Mn_2O_7$ (A = La, Nd, Sm; B = Sr, Ca).[2–7] The bilayered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ is constructed by alternately stacking a rock-salt-type block layer $(La, Sr)_2O_2$ and two ferromagnetic metallic MnO_2 layer along the c axis. The layered perovskites exhibit unique anisotropies of the electronic and magnetic properties due to the intrinsic quasi-two-dimensional layered structure. In the past years, many investigations have focused on substitution of transition metal element at Mn site. The ferromagnetic (FM) transition temperature T_C and metal–insulator (MI) transition temperature T_{MI} both decrease with doping level increasing. Insulating behaviour is observed at high doping level. Doping destroys the long-range FM ordering and suppresses the double-exchange interaction.[8–10] We report the effect of non-magnetic Ga ions substitution for Mn site on the magnetic and the transport properties in the bilayer manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ in the present paper.

2. Experiment

The samples of $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ (x = 0, 0.08) were prepared by the conventional solid-state reaction method. La_2O_3, $SrCO_3$, $MnCO_3$, and Ga_2O_3 with a purity higher than 99.9% were stoichiometrically mixed ground, and calcined at 1000 °C for 24 h in air. The product was ground and pressed into pellets and then sintered until single-phased samples were obtained. The samples were characterized by X-ray diffraction. The magnetic measurements were performed by a superconducting quantum interference device (Quantum Design, MPMS-7). The resistivities in zero field and an applied field (H = 5 T) were mea-

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sured by the standard four-point technique separately.

3. Results and discussion

The X-ray diffraction patterns for the samples $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ ($x = 0$, 0.08) are shown in Fig. 1. The crystal structure was further refined by the Rietveld method. After doping with a little Ga^{3+} ions, we could not find any other impurity diffraction peaks and their position changes obviously, indicating the formation of good-quality single-phase with the substitution of Ga^{3+} ions for Mn site. The sample can be well indexed to an $Sr_3Ti_2O_7$-type tetragonal structure with the space group $I4/mmm$. The radius of Ga^{3+} ion (0.062 nm) is bigger than that of Mn^{4+} ion (0.053 nm) and matches well with that of Mn^{3+} ion (0.0645 nm). So there is a minimal lattice distortion along with a preferential replacement of Mn^{3+}.

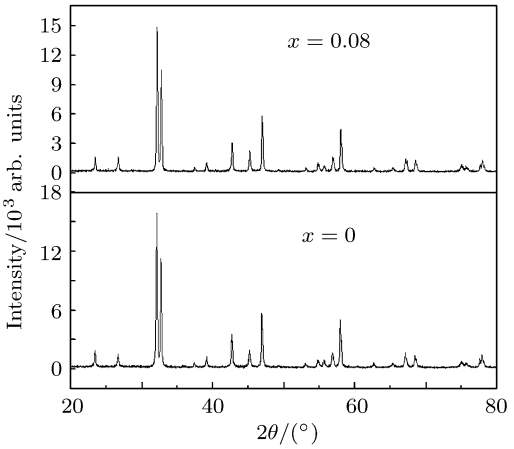


Fig. 1. The X-ray diffraction patterns of $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ ($x = 0$, 0.08).

Figure 2 shows the temperature dependences of magnetization under applied magnetic field of 0.1 T for $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ with $x = 0$ (a) and $x = 0.08$ (b). The undoped sample $La_{1.2}Sr_{1.8}Mn_2O_7$ is ferromagnetic at low temperature, which is consistent with the previously reported.[5] There are two transition temperatures: the lower transition temperature is observed around the temperature $T_C \sim 100$ K, the temperature at which dM/dT reaches a minimum (see the inset in Fig. 2(a)). The three-dimensional long-range FM ordering forms around T_C. With temperature increasing, the higher transition is observed around the temperature $T^* \sim$236 K, the temperature at which the $dM/dT - T$ curve reaches another minimum (see the inset in Fig. 2(a)), which is due to the two-dimensional short-range FM ordering. The two transition temperatures are related to the anisotropic exchange interaction. The lower transition T_C temperature results from the out-plane exchange interaction, and the higher transition temperature T^* originates from the in-plane exchange interaction.[11,12] In Fig. 2(b), Ga^{3+} ion doping causes the magnetic transition temperatures T_C and T^* to reduce to 46 K and 176 K (values of dM/dT are shown in the inset of Fig. 2(b)), respectively. Then, it can be discerned from Fig. 2(b) that the sample shows obviously a characteristic splitting below 31 K, indicating the presence of a low temperature spin-glass state as in ABO3-type perovskites. The spin-glass behaviour is ascribed to the competition between FM and AFM interactions, which leads to the disorder in the system.

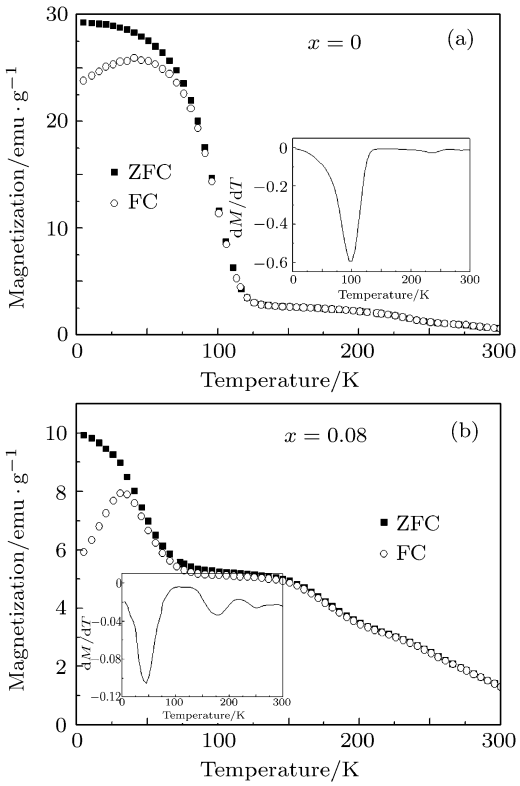


Fig. 2. Temperature dependences of the magnetization in a 0.1-T field for $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ with (a) $x = 0$ and (b) $x = 0.08$. ZFC and FC stand for zero field cooling and field cooling respectively. Values of dM/dT are shown in the insets.

Figure 3 shows the temperature dependences of resistivity and magneto-resistance (MR) for $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ at the fields of 0 T and 5 T. In Fig. 3(a), we find that the parent compound $La_{1.2}Sr_{1.8}Mn_2O_7$ presents the MI transition about $T_{MI} = 70$ K. In Fig. 3(b), we can see that 4% doping on Mn site causes the resistivity to increase, and the MI transition temperature to disappear. In addition, an applied 5-T field induces the resistivity to

decrease for $x = 0$ and $x = 0.08$ samples. The applied field can suppress the magnetic frustration and the spin scattering of electrons, so the resistivity reduces.

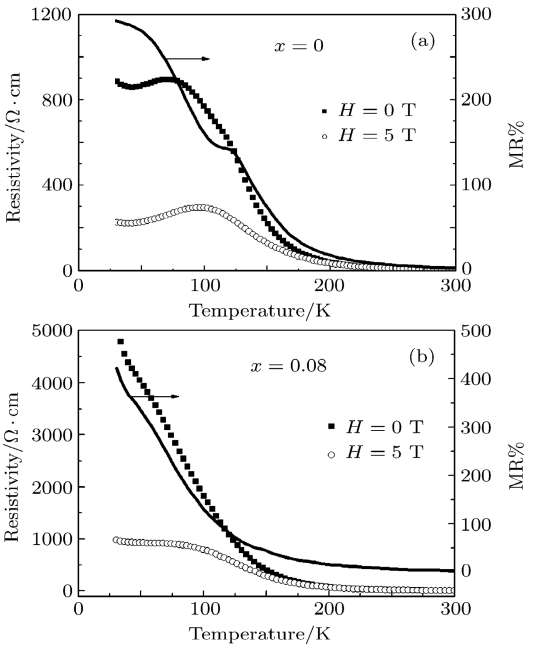


Fig. 3. Temperature dependences of resistivity and magneto-resistance (MR) for $La_{1.2}Sr_{1.8}Mn_{2-x}Ga_xO_7$ samples at the fields of 0 T and 5 T with (a) $x = 0$ and (b) $x = 0.08$.

The MR is defined as $\mathrm{MR}(\%) = [(\rho(0) - \rho(H))/\rho(H)] \times 100$, where $\rho(0)$ is the resistivity in the zero field and $\rho(H)$ is the resistivity in the magnetic field $H = 5$ T. For the undoped sample, the MR peak value reaches up to 292% at about 30 K, and this phenomenon is related to spin-polarized tunneling or grain boundary effect due to a ferromagnetic layer-insulating layer-ferromagnetic layer tunneling structure of its own.[13,14] After doping with Ga^{3+} ions, MR curve exhibits a simple monotonic increase with temperature decreasing, and a greater magnetoresistance effect is obtained at low temperature. This phenomenon probably arises from intergrain tunneling MR effect.

The results described above indicate that the Ga^{3+} doping dilutes the magnetically active Mn–O–Mn network for the Mn^{3+} ions replaced by the nonmagnetic Ga^{3+} ions. In addition, the replacement of Mn^{3+} by Ga^{3+} reduces the Mn^{3+}/Mn^{4+} ratio, the numbers of carriers, and the number of available hopping sites, so the double-exchange interaction weakens, then long-range FM ordering is broken and metallic conduction is suppressed. The antiferromagnetic surper-exchange interaction increases with double-exchange interaction weakening, thereby resulting in the reduction of transition temperature and the presence of the spin-glass state at low temperature. The number of carriers and the number of available hopping sites decrease, so the electrical transport channel is broken, then the resistivity increases. The variations for Ga doping sample are similar to those for Fe and Co doping samples.[8,15]

4. Conclusion

We investigated the effects of nonmagnetic Ga^{3+} ion substitution for Mn site on the magnetic and the transition properties in the bilayer Manganite $La_{1.2}Sr_{1.8}Mn_2O_7$. With Ga^{3+} doping, the transition temperature decreases, and the spin-glass state occurs at low temperature due to the competition between FM and AFM interactions. In addition, the MI transition disappears with Mn ion replaced by Ga ions. These effects are attributed to the weakening of double-exchange interaction, which results from the fact that Ga^{3+} doping dilutes the magnetically active Mn–O–Mn network and reduces the Mn^{3+}/Mn^{4+} ratio.

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