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Effect of Al doping on the magnetic and electrical properties of layered perovskite $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$

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Abstract

The effect of Al substitution for Mn site in layered manganese oxides $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$ on the magnetic and electrical properties has been investigated. It is interesting that all the samples undergo a similar and complex transition with lowing temperature; they transform from the two-dimensional short-range ferromagnetic order at T^*, then enter the three-dimensional long-range ferromagnetic state at T_C, at last they display the canted antiferromagnetic state below T_N. T^*, T_C and T_N are all reduced with Al content. Resistivity increases sharply with increasing Al concentration, and the metal–insulator transition disappears when x reaches 10%. Additionally, magnetoresistance (MR) effect is weakened. Al substitution dilutes the magnetic active Mn–O–Mn network and weakens the double exchange interaction, and further suppresses FM ordering and metallic conduction. Owing to the anisotropic interaction in the layered perovskite, the magnetic and electrical properties are more sensitive to Al doping level than those in ABO_3-type perovskite.

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Keywords: Layered perovskite; Magnetic property; Electrical property; Magnetoresistance

1. Introduction

The discovery of the colossal magnetoresistance (CMR) effect in doped rare-earth perovskite manganite system has motivated much interest in recent years [1,2]. It is well known that perovskite structure manganite oxides belong to Ruddlesden–Popper (RP) series with the general formula of $Ln_{n+1}Mn_nO_{3n+1}$, where Ln is lanthanide or alkaline-earth metal (such as La or Sr etc.), n is the dimension. Doped perovskite manganites $Ln_{1-x}A_xMnO_3$ can be regarded as the $n=\infty$ member of RP family, which has a three-dimensional (3D) crystal structure. In recent years, many studies [3–5] have been performed for the $n=2$ member of RP series, $Ln_{2-2x}A_{1+2x}Mn_2O_7$, which are a stack of ferromagnetic (FM) metal sheets composed of MnO_2 bilayers separated by a $(Ln, A)_2O_2$ rocksalt layer and thus form a natural array of FM–insulator–FM junction. The special two or quasi-two-dimensional (2D) structure makes its physical properties strongly anisotropic. Recent magnetic neutron scattering [6] measurement on $La_{1.2}Sr_{1.8}Mn_2O_7$ has provided evidence for the coexistence of FM fluctuations between Mn^{3+} and Mn^{4+} ions within a single layer and more persistent antiferromagnetic (AFM) cluster above the Curie temperature. Therefore, we think the complex magnetic order must influence the electrical properties of layered perovskite manganese oxides. For $Ln_{2-2x}A_{1+2x}Mn_2O_7$, there are so many reports [7–10] on the substitution by other 3d transitional metals such as Fe and Co for Mn site. Generally speaking, substation of other elements at Mn site changes the ratio of Mn^{3+}/Mn^{4+}, lattice parameters, band filling, the electronic band structure and magnetic properties due to the nonmagnetic or magnetic nature of the doped ions simultaneously. In this paper, we have chosen Al to make the substitution for Mn because its atomic radius matches very well with that of the Mn^{4+}. Thus, one would expect minimal lattice distortion along with a preferential

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replacement of Mn^{4+}. Most importantly, Al with an empty d shell is nonmagnetic and hence a simple dilution of the magnetic lattice would be expected.

2. Experiment

Samples of $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$ ($x=0$, 0.05, 0.1) were prepared by the conventional ceramic technique. Stoichiometric amounts of La_2O_3 (99.95%, preheated to 800 °C for 6 h before weighing), $SrCO_3$ (99.9%), MnO_2 (99.9%) and Al_2O_3 (99.9%) were mixed, ground and calcined at 1000 °C for 24 h and 1300 °C for 40 h in air. The product was ground and calcined at 1400 °C for 72 h. The obtained powder was ground and pressed into pellets with the dimension of 12 mm diameter and 2 mm thickness and then sintered at 1450 °C in the flowing oxygen for 40 h cooling to room temperature. The structure of the bulk was analyzed using Cu Kα radiation ($\lambda=0.15406$ nm) and Rigaku D/Max-2400 X-ray diffractometer. The magnetic measurements were detected by a SQUID magnetometer in the temperature range of 5–300 K. The zero field-cooled (ZFC) and field-cooled (FC) curves were checked under an applied field of 100 Oe. The electrical resistivities were measured using the standard direct current four-probe method from 25 to 300 K within the accuracy of 0.1 K. The magnetoresistance (MR) is defined as the following relation:

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

where $\rho(H)$ and $\rho(0)$ are the resistivities at zero field and applied field, respectively [11]. The external field was applied vertically to the direction of the current.

3. Results and discussion

The X-ray diffraction (XRD) patterns for samples of $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$ ($x=0$, 0.05, 0.1) are shown in Fig. 1. All the diffraction peaks are well indexed with the $Sr_3Ti_2O_7$-type tetragonal structure (I4/mmm) without an impurity phase like $La_{1-x}Sr_xMnO_3$ or $La_{1-x}Sr_{1+x}MnO_4$ [12], which indicate that all samples are of a single phase. The cell parameters and the volume of unit cell calculated from XRD data are listed in Table 1. It is obviously seen that the cell parameters and cell volume decrease with increasing the Al doping level; this is due to the fact that radius of the Al^{3+} (0.0535 nm) [1] is smaller than that of Mn^{3+} (0.0645 nm) and matches well with that of Mn^{4+} (0.053 nm).

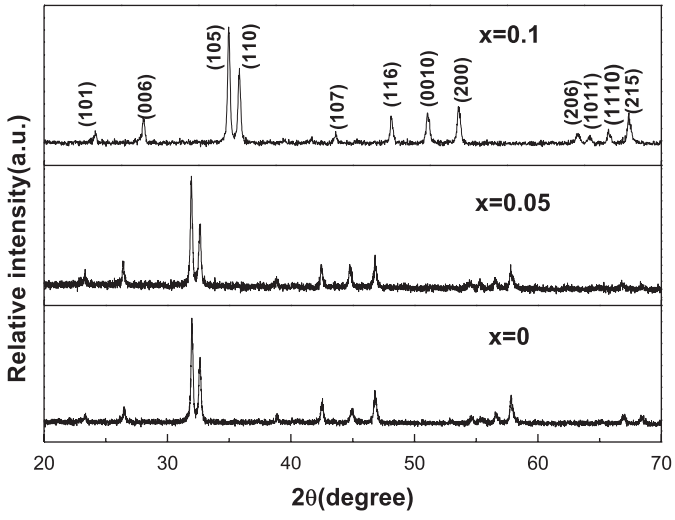

Fig. 1. XRD patterns of $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$ ($x=0$, 0.05, 0.1).

Fig. 2 shows the temperature dependence of ZFC and FC magnetization measured in a field of 0.01 T for $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$. It clearly exhibits a similar trend for the three samples. For the undoped sample, from the room temperature downward, the magnetization increases slowly and then reaches a plateau labeled as T^* (defined as

Table 1
Cell parameters and volume of unit cell of $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$

X	a (nm)	c (nm)	V (nm^3)
0	0.3863	1.9786	296.6
0.05	0.3858	1.9847	295.4
0.1	0.3846	1.9901	294.3

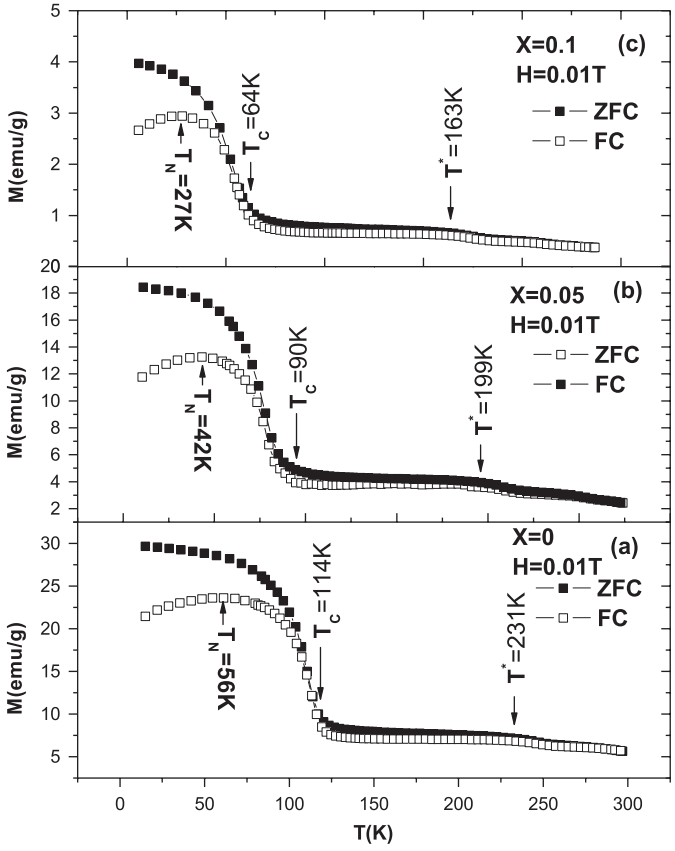

Fig. 2. Temperature dependence of ZFC and FC magnetization measured in a field of 0.01 T for $La_{1.3}Sr_{1.7}Mn_{2-x}Al_xO_7$. (a) $x=0$, (b) $x=0.05$, (c) $x=0.1$.