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# Effect of Cr doping in the bilayer manganite La<sub>1.4</sub>Sr<sub>1.6</sub>Mn<sub>2</sub>O<sub>7</sub>

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# ABSTRACT

The effect of Cr doping on magnetic and electrical properties in the bilayer manganites  $La_{1.4}Sr_{1.6}(Mn_{1-y}Cr_y)_2O_7$  (y=0-0.1) has been investigated. When  $y \le 0.025$ , Cr doping enhances the three-dimensional magnetic transition temperature  $T_c$  and the insulator-metal transition temperature  $T_{IM}$  as well as decreases the peak resistivity at  $T_{IM}$ , and the saturated magnetization decreases slightly. When  $y \ge 0.035$ ,  $T_{IM}$  decreases gradually accompanied by the increase of peak resistivity, but  $T_c$  remains nearly constant, and the saturated magnetization decreases heavily. In the whole doping region, the two-dimensional magnetic transition temperature  $T^*$  monotonously decreases with an increasing of Cr doping level. These results can be explained by considering different magnetic (including ferromagnetic and antiferromagnetic) interactions between Mn ions and Cr ions.

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## 1. Introduction

In the past few years, the bilayer manganites  $La_{2-2x}Sr_{1+2x}Mn_2O_7$ of Ruddlesden–Popper phases (n=2) attracted considerable attention due to their novel physical properties such as the colossal magnetoresistance effect, the tunneling magnetoresistance and fascinating magnetic properties [1-3]. In this system, two ferromagnetic metallic  $MnO_2$  layers are separated by a rock-salt-type block layer  $(La,Sr)_2O_2$ , keeping the quasi-two-dimensional networks of the MnO<sub>6</sub> octahedra. The system presents a very rich phase diagram and displays a rich variety of properties depending strongly on the doping level x [4]. For x=0.3, namely La<sub>1.4</sub>Sr<sub>1.6</sub>Mn<sub>2</sub>O<sub>7</sub>, the magnetic coupling is ferromagnetic (FM) within the constituent MnO<sub>2</sub> bilayer and weakly antiferromagnetic (AFM) between the adjacent bilayers at low temperature, the three-dimensional (3D) magnetic transition temperature being  $T_{\rm C} \sim 100$  K. For x = 0.32 - 0.4, the magnetic couplings mentioned above are FM; the maximum  $T_{\rm C} \sim 131$  K for  $x \sim 0.36$ , where  $Mn^{3+}/Mn^{4+}$  ratio is an optimal ratio. For x=0.5, namely LaSr<sub>2</sub>Mn<sub>2</sub>O<sub>7</sub>, with charge-ordered state the magnetic coupling is FM within the constituent single MnO<sub>2</sub> layer but shows A-type AFM order between the respective MnO<sub>2</sub> layers with a bilayer unit, the AFM ordering temperature being  $T_N \sim 220$  K.

It is well known that the correlation between the magnetic and electrical properties in the mixed-valence manganites is generally understood in terms of the double-exchange (DE) interaction mechanism [5]. Besides the DE mechanism, the Jahn–Teller effect, phase separation (PS) [6] and AFM superexchange and chargeorbital ordering interactions also play an important role. The bilayer manganites,  $La_{2-2x}Sr_{1+2x}Mn_2O_7$ , offer another unique opportunity to investigate the generic features of the mixedvalence manganites.

Mn-site doping in the bilayer manganites can dramatically change the magnetic and electrical properties. Some such studies have been undertaken during the past few years [7–13]. Cr is a very interesting substitution ion for Mn site, because  $Cr^{3+}$  has the same electronic configuration  $(t_{2g}^3 e_g^0)$  as that of  $Mn^{4+}$ , and closely identical ionic radius as that of  $Mn^{3+}$ . There may be FM DE interaction between  $Cr^{3+}$  and  $Mn^{3+}$  just as between  $Mn^{4+}$  and  $Mn^{3+}$ . Some reports on the effect of Cr doping in the cubic perovskite manganites have appeared, and several reports on such studies have also appeared in the case of bilayer manganites [14–18]. But there are a few reports on the effect of Cr doping in the bilayer manganite La<sub>1.4</sub>Sr<sub>1.6</sub>Mn<sub>2</sub>O<sub>7</sub>.

In this paper, we report the effect of Cr doping on magnetic and electrical properties, especially on magnetic properties in the bilayer manganites La<sub>1.4</sub>Sr<sub>1.6</sub>(Mn<sub>1-y</sub>Cr<sub>y</sub>)<sub>2</sub>O<sub>7</sub> (y=0–0.1). With Cr doping, Cr<sup>3+</sup> will replace Mn<sup>3+</sup>, which directly decreases the content of Mn<sup>3+</sup> ions. On the other hand, Cr<sup>3+</sup> can act as Mn<sup>4+</sup> due to the same electronic configuration if there exists FM DE interaction between Cr<sup>3+</sup> and Mn<sup>3+</sup>. Thus, the Mn<sup>3+</sup>/Mn<sup>4+</sup> (including Cr<sup>3+</sup>) ratio will be closer to optimal ratio, and an interesting effect on magnetic and electric properties will be observed in the present Cr-doped system. The present measurements show that the magnetic coupling between Cr ions and Mn ions is related to the Cr doping concentration. When y < 0.035 the coupling is FM and when y > 0.035 it is AFM. The total effect is observed and discussed.

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#### 2. Experiment

The polycrystalline bulk samples of La<sub>1.4</sub>Sr<sub>1.6</sub>(Mn<sub>1-y</sub>Cr<sub>y</sub>)<sub>2</sub>O<sub>7</sub> (y=0-0.1) were synthesized by a standard ceramic process. Well ground stoichiometric mixture of high purity La<sub>2</sub>O<sub>3</sub>, SrCO<sub>3</sub>, MnO<sub>2</sub> and Cr<sub>2</sub>O<sub>3</sub> were calcined at 1200 °C for 24 h in air with intermediate grinding. The powders were well ground again, then pelletized and sintered at 1450 °C for 45 h with an intermediate grinding. The structural characterization was done through X-ray diffraction (XRD) with CuK $\alpha$  radiation at room temperature. The temperature dependence of resistivity was measured by a standard four probe method at zero field. Magnetization was measured using a commercial physical property measurement system (Quantum Design PPMS).

#### 3. Results and discussion

XRD patterns for all the samples of  $La_{1.4}Sr_{1.6}(Mn_{1-y}Cr_{y})_2O_7$ (y=0-0.1) is presented in Fig. 1. All diffraction peaks are indexed using  $Sr_3Ti_2O_7$ -type tetragonal structure (I4/mmm), indicating a single phase of bilayer structure. The lattice parameters obtained from XRD data are listed in Table 1.

Fig. 2 shows the temperature (77–300 K) dependence of resistivity for all samples. For undoped sample (y=0), the insulator-metal transition (IMT) temperature  $T_{IM}$  is 110 K. At



**Fig. 1.** XRD patterns of all samples  $La_{1,4}Sr_{1,6}(Mn_{1-y}Cr_y)_2O_7$  ( $0 \le y \le 0.1$ ).

Table 1	
Lattice parameters for samples La1	$_{4}\mathrm{Sr}_{1.6}(\mathrm{Mn}_{1-y}\mathrm{Cr}_{y})_{2}\mathrm{O}_{7} \ (0 \le y \le 0.1).$

у	a (Å)	c (Å)	$V(Å^3)$
y=0y=0.005y=0.01y=0.015y=0.025y=0.035y=0.05	3.8751 3.8746 3.8729 3.8732 3.8711 3.8704 3.8688	20.171 20.174 20.175 20.181 20.183 20.183 20.187 20.194	302.90 302.86 302.61 302.75 302.45 302.40 302.26
y = 0.1	3.8007	20.202	302.05



Fig. 2. Temperature dependence of resistivity for samples  $La_{1.4}Sr_{1.6}(Mn_{1-y}Cr_y)_2O_7$   $(0\leq y\leq 0.1)$  at zero field.



**Fig. 3.** Temperature dependence of magnetization *M* at 0.01 T for samples  $La_{1.4}Sr_{1.6}(Mn_{1-y}Cr_y)_2O_7$  (y=0, 0.025, 0.035, 0.05, 0.1).

lower Cr doping level ( $y \le 0.025$ ), the IMTs shift gradually to higher temperature with increasing Cr doping level.  $T_{IM}$  reaches the maximum value of 121 K for y=0.025 sample, decreases gradually with increasing Cr doping level and when y=0.1, IMT disappears. Associated with such  $T_{IM}$  change, the resistivity near  $T_{IM}$  decreases gradually for y < 0.025 samples, and increases for  $y \ge 0.025$  samples. Such results for Cr doping are not observed in other manganites (including the bilayer and 3D cubic manganites), which are very interesting. In addition, with increasing Cr doping, IMTs become broader and broader; similar results have also been observed for Cr doping in La<sub>1.2</sub>Sr<sub>1.8</sub>Mn<sub>2</sub>O<sub>7</sub> [16].

The temperature dependence of magnetization *M* measured at a field of 0.01 T for samples  $La_{1.4}Sr_{1.6}(Mn_{1-y}Cr_{y})_2O_7$  (y=0, 0.025, 0.035, 0.05, 0.1) is shown in Fig. 3. All *M*–*T* curves show two magnetic transitions lying at lower temperature near 110 K and higher temperature near 200–240 K. For all samples, a visible plateau in the *M*–*T* curves extends to higher temperature, which has been ascribed to a short range two-dimensional (2D) FM ordering due to the quasi-two-dimensional feature [19–21]. For the undoped sample (y=0), Fig. 3 shows a 3D magnetic transition at  $T_C$  (defined as the temperature at which d*M*/d*T* reaches the maximum) is 106 K; another upper transition, namely 2D magnetic transition at  $T^*$  (defined as another maximum in d*M*/d*T*–*T*  Download English Version:

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