

# Thermal transport of Cr-doped double-layered $\text{LaSr}_2\text{Mn}_2\text{O}_7$

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## Abstract

We have carried out thermoelectric power ( $S$ ), magnetization and magnetoresistance measurements on  $\text{LaSr}_2\text{Mn}_{2-y}\text{Cr}_y\text{O}_7$  compounds ( $y=0.1, 0.2$  and  $0.4$ ). In the bilayer manganite  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (hole doping level  $x=1/2$ ), it is well known that the coexistence of metallic A-type antiferromagnetic (AF) phase and CE-type charge/orbital order (CO/OO) phase is essential to understand physical properties. The Cr-doping strongly suppressed the AF transition temperature and its associated peak in  $S$  down to lower temperatures. The Cr impurities on the Mn site in the bilayer manganite system did not cause such a dramatic influence as the insulator to metal transition in cubic manganites. This finding is discussed on the basis of phase separation model concerning the A-type majority and CE-type minority phases.

*Key words:* bilayer manganite ; thermoelectric power; Cr-doping;

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The CMR bilayer compounds  $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$ , in which  $\text{MnO}_2$  bilayers and  $(\text{La},\text{Sr})_2\text{O}_2$  blocking layers are alternatively stacked, have a rich variety of physical properties depending on hole doping ( $x$ ). In  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  (hole doping level  $x=1/2$ ), neutron diffraction studies have revealed the coexistence of A-type antiferromagnetic (AF) phase and the CE-type charge/orbital ordered (CO/OO) phase [1]. It has been reported that the Cr-doping on the Mn site in the cubic manganites with the CO/OO phase cause the phase separation into the metallic and insulating states associated with occurrence of the CMR [2].

In order to investigate the Cr-doping effect on physical properties in the bilayer manganites,  $\text{LaSr}_2\text{Mn}_2\text{O}_7$ , we have performed thermopower ( $S$ ), magnetization and magnetoresistance measurements on  $\text{LaSr}_2\text{Mn}_{2-y}\text{Cr}_y\text{O}_7$  ( $y=0.1, 0.2$  and  $0.4$ ) compounds. Thermopower measurements provide direct information about the energy derivative of density of states and polaronic states. The  $\text{Cr}^{3+}$  has the electronic con-

figuration of  $t_{2g}^3e_{2g}^0$  (spin quantum number  $S=3/2$ ) which is the same as the  $\text{Mn}^{4+}$ , and acts on the Mn site as the  $e_g$ -orbital deficiency. Cr-doped polycrystalline samples were prepared with the solid-state reaction method. A  $y=0$  single crystal with high quality was grown with the floating zone method. The thermopower was measured with a steady-state heat-flow method using GM-type refrigerator. Thermoelectric voltage and thermal gradient generated in the longitudinal direction of samples were detected with Cu wires and differential thermocouples (AuFe-Chromel), respectively. Magnetoresistance was measured with a four-probe technique at IMR of Tohoku Univ. Magnetic measurement was carried out using a SQUID magnetometer.

First, we showed in Fig.1 the temperature dependence of magnetization in the Cr-doped  $\text{LaSr}_2\text{Mn}_2\text{O}_7$  bilayer manganites. The Cr-doping strongly suppressed the AF transition temperature from 210K ( $y=0$ ) down to 125K ( $y=0.2$ ). On the other hand, the low-T second peak was more enhanced with an increase of Cr-impurities. The resistivity data of Cr-

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doped samples showed a temperature variation of thermal activation type and were in the magnitude more enhanced with an increase of Cr-content , in contrast with those in cubic manganites (not shown here). A pronounced peak of the pure crystal near the AF and CO transition temperature disappeared due to Cr-doping.

Next, in Fig.2 , we displayed the thermopower of our samples as a function of temperatures. For all samples, the  $S$  data showed almost negative sign over the measured temperature range and the observed peak in  $S$  reached a very small-value around  $T_N$ . The  $S$  behavior at  $y=0.1$  is almost similar to the  $T$ -dependence of  $S_{ab}$  of the  $y=0$  single crystal. It should be noted that a strong decrease in  $S_{ab}$  of the  $y=0$  crystal at higher  $T$  is associated with the formation of Zener-polarons in [3], where Zener pairs are two-manganese clusters of  $Mn^{3+}$ -O- $Mn^{4+}$  and are confined to  $MnO_2$  double layers. Thus, it is expected that the formation of Zener pairs is disturbed due to Cr-impurities as orbital-deficiencies so that the value of  $S$  of Cr-doped samples showed a weak dependence at higher  $T$ .

Finally, Magnetoresistance data at selected temperatures are shown in Fig.3, where the current supplied is parallel to the direction of applied field. Magnetoresistance at  $y=0.1$  showed a relatively large decrease of 40 percent (in a field of 8T at 80K) comparable with reported data at  $y=0$ , which seems to originate from the low temperture orbital-disordered state [4].

In half hole-doped bilayer manganites, the volume fraction of the CE type phase is estimated to be about 18 percent at 115 K by Kubota et al. in other words, this system has been separated into the A-type majority and CE-type minority regions. If we assume this situation is also realized in our samples, it is expected that most of Cr-impurities are doped for the majority phase of the A-type AF state in comparison with the minority phase of the CE-type CO state. This finding is consistent with our experimental results on a strong degradation of  $T_N$  due to Cr-doping. The Cr-impurities act on the Mn site as orbital-deficiencies and through the spin-orbital interaction suppress a AF long-range spin correlation in the major phase.

In summary, we have carried out thermopower , magnetization and magnetoresistance measurements on Cr-doped  $LaSr_2Mn_2O_7$  compounds. Our data strongly support the phase separation model in which in the half-doped bilayer manganite system coexist both the A-type AF metallic and CE-type CO/OO insulating phases.

## References

- [1] M.Kubota et al.,J.Phys.Soc.Jpn. 68(1999)2202.
- [2] T.Kimura et al.,Phys.Rev.B62(2000)15021.

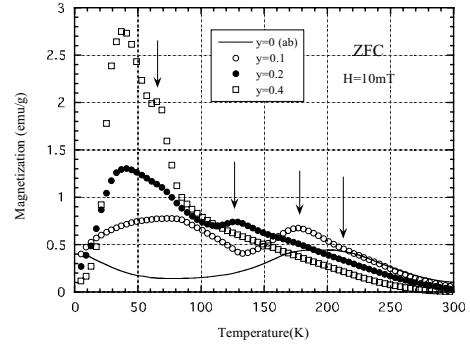


Fig. 1. Temperature dependence of magnetization in the Cr-doped  $LaSr_2Mn_2O_7$  bilayer manganites. The  $y=0$  single-crystal data are enlarged by a factor of ten. The arrows denote the AF transition temperature.

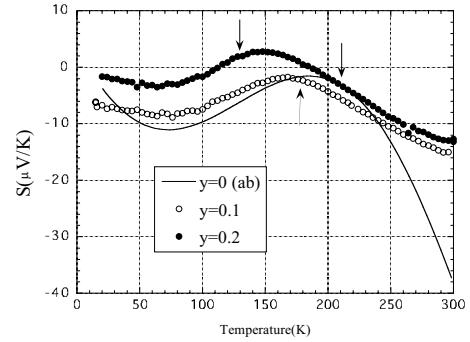


Fig. 2. Temperature dependence of thermoelectric power in the Cr-doped  $LaSr_2Mn_2O_7$  bilayer manganites. The arrows denote the AF transition temperature determined from magnetic measurements.

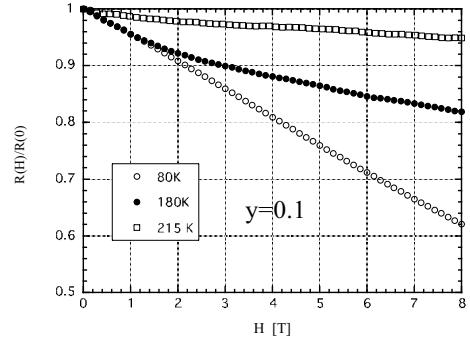


Fig. 3. Magnetoresistance at selected temperatures in the Cr-doped  $LaSr_2Mn_2O_7$  ( $y=0.1$ ).

[3] L.S.Zhou et al. Phys.Rev.B58 (1998)R579.

[4] T.Kimura et al., Phys.Rev.B58(1998)11081.