S-8

Spin-glass behavior in (*A,B*)-site deficient manganese perovskites

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In the past years, a giant magnetoresistance (GMR) effect found in perovskite-like structured materials has attracted considerable attention among scientists and manufacturers, since, a practical point of view, the capacity of producing magnetic and sensing sensors [1]. In a stream of this interest, further efforts to understand the underlying mechanism that leads to the GMR effect relative to the correlation between transport and magnetic properties, have been extensively devoted [2-4]. In these cases, spin-glass-like behaviors are ascribed to the frustration of random competing exchange interactions, namely the ferromagnetic double-exchange interaction between Co^{3+} (or Mn^{3+}) and Co^{4+} (or Mn^{4+}) and the antiferromagnetic one like spins. Noticeably, the distinction of spin-glass region from cluster-glass one, involved in the remarkable changes in transport and magnetic properties at a critical value of doping concentration, was observed [2,3]. Magnetic anomalies in zero-field-cooled (ZFC) magnetization as well as ac magnetic susceptibility below Curie temperature T_C and the charge/orbital fluctuation were also realized [4]. In this work, we present a study of magnetic properties of a deficient manganese perovskites system of $La_{0.6}Sr_xMnTi_yO_3$, and particularly provide its new magnetic phase diagram.

$La_{0.6}Sr_xMnTi_yO_3$ (x=0.15, 0.2, 0.25, 0.3, 0.35, 0.4, and y=0.4-x, respectively) polycrystalline samples were prepared by a conventional solid-state reaction method. A detailed procedure of sample preparation will be described elsewhere. X-ray diffraction data and scanning electron microscope confirmed the quality of the samples. The magnetic measurements were performed using a Quantum Design MPMS-5 SQUID magnetometer or a PPMS-7 magnetometer.

In a primary material of $LaMnO_3$ with a cubic structure antiferromagnetic and insulator characteristics are present. Substitution of divalent elements such as Ca^{2+}, Sr^{2+}, Ba^{2+}, etc. for La^{3+} in $LaMnO_3$ introduces a paramagnetic to ferromagnetic transition accompanying an insulator to metal one [3,4]. The correlation between metallic conduction and ferromagnetism results from the Zener's double-exchange interaction between Mn^{3+} (electronic configuration $t_{2g}^3 e_g^1$, S = 2) and Mn^{4+} ($t_{2g}^3 e_g^0$, S = 3/2) via oxygen 2p orbital. The nature of magnetic ordering in the compositional range strongly depends on the relative concentrations of Mn^{3+} and Mn^{4+} ions as well as the <Mn-O-Mn> angle and <Mn-O> bond distance. A complex magnetic phase might arise due to the competing ferromagnetic $Mn^{3+}(e_g)$-$O(2p_\sigma)$-$Mn^{4+}(e_g)$ double-exchange and antiferromagnetic $Mn^{3+}(t_{2g})$-$O(2p_\pi)$-$Mn^{4+}(t_{2g})$ super-exchange. To the present work, the deficiency at both A-site (=La,Sr) and B-site (=Mn,Ti) led to a decrease in T_C as the titanium and strontium concentration increases and decreases, respectively (see Fig. 1). The drop in T_C is attributed to the disorder induced by the random substitution of Ti^{4+} in the manganese sublattice, expected to result in

regions of different T_C. Besides, the decrease of Sr-deficient contents here also causes an additional drop in T_C arising from the decrease of A-site average ion radius. In addition, the magnetization measured at T=5K and at a maximum field of 5T decreases as the Ti-deficient content, y, is increased between 0 and 0.25 (and, the Sr-deficient content, x, is decreased between 0.4 and 0.15, respectively). The magnetization is far from saturation at a magnetic field of 5T, but a considerable value of spontaneous magnetization is still observed. The decrease of magnetization here should be related to the dilution of the Mn sublattice, as the Mn^{4+} content does not change much in comparison with y=0.05. This dilution is assumed to result in weakening of the effective exchange interaction between the Mn magnetic moments with a reduced number of magnetic next neighbors. More interestingly, a prominent divergence between zero-field-cooled (ZFC) and field-cooled (FC) magnetization measured at H=50 Oe below T_C for all compositions was observed. An appearance of a sharp peak of ZFC magnetization curves below T_C for $y \geq 0.1$ prefer to a spin-glass-like behavior, rather than the case of the samples with y=0 and 0.5. This cusp is the so-called spin-glass freezing temperature, T_g [2,3]. It is evidently confirmed by ac magnetic susceptibility measurements where a peak of this curve is coincident with that of the ZFC curve. In Fig. 1, a magnetic phase diagram of $La_{0.6}Sr_xMnTi_yO_3$ where SG, FM, and PM denote "spin-glass", "ferromagnetic", and "paramagnetic", respectively, is depicted.

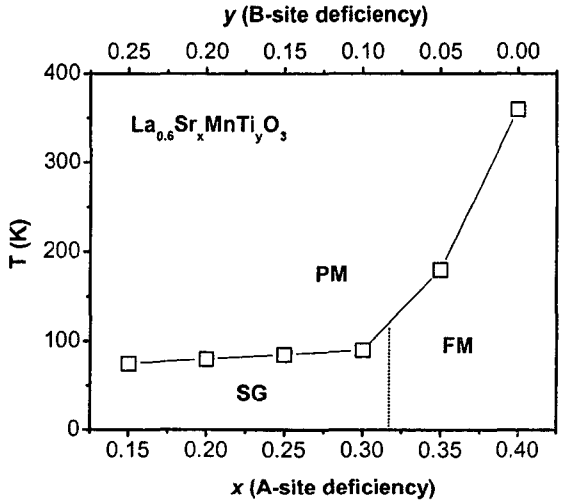

Fig. 1. The magnetic phase diagram of $La_{0.6}Sr_xMnTi_yO_3$ as a function of Sr-deficient content (or Ti-deficient content).

In summary, the magnetic properties of deficient manganese perovskites of $La_{0.6}Sr_xMnTi_yO_3$ have been studied by magnetization measurements in low magnetic fields. It is very instructive that there exists a spin-glass region for $y \geq 0.1$ and $x \leq 0.3$ but disappears at y=0 and 0.05 (or x=0.4 and 0.35, respectively). Nevertheless, the clarification in the correlation between transport and magnetic properties needs further study, and will be published elsewhere.

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