Magnetic phase diagram $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6

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Abstract

We have established a magnetic phase diagram of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 by means of magnetization measurements. The magnetization curve exhibits a rectangular hysteresis loop below T^*, implying the ferromagnetic behavior. A metamagnetic transition was observed also above T^* without a hysteresis. Combined with our previous data of the elastic constants, it was found that there were mainly three magnetic phases in the Field-Temperature ($H-T$) phase diagram. The magnetic properties of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 at low temperatures is discussed in terms of spin- and orbital-glass state.

Key words: spin-glass; phase diagram; $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$; magnetization

In recent intensive experimental studies on double-layered Manganites: $La_{2-2x}Sr_{1+2x}Mn_2O_7$, it has been found to exhibit various exotic physical phenomena as represented by colossal negative magnetoresistance (CMR)[1-3]. It is strongly depending on La concentration, which provides a opportunity to change the carrier concentration and a local lattice deformation: Jahn-Teller distortion (JT)[4-5]. Furthermore, the substitution of Pr at the La site can realize to keep the carrier concentration constant but change a local lattice deformation[6-7]. Double-layered Manganites exhibits a long-range magnetic ordering. $La_{1.2}Sr_{1.8}Mn_2O_7$ shows the ferromagnetic ordering at T_c=125 K, accompanied Metal-Insulator (MI) transition[1-2]. In this undoped system (z=0), the planar $d(x^2\text{-}y^2)$ orbital occupancy is dominant, leading to the planar ferromagnetic (FM) ordering. FM transition temperature T_c decreases and CMR-effect is also enhanced with increasing z. Ultimately, the tremendously large CMR effect appeared in $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6[6-7]. The resistivity shows a semiconductor-like behavior with a kink at about 40 K (T^*) as decreasing temperatures in zero field. On the other hand, the resistivity shows metallic behavior in applying magnetic fields, in which it decreases very fast at low temperature and the transition becomes more distinct and larger with increase of magnetic field. The magnetoresistance shows a large decrease by a factor of about 2000 at 25 K with a large hysteresis[6-7].

Besides, the JT of Mn-O_6 octahedra, *i.e.*, the ratio of the averaged apical Mn-O bond length to the equatorial Mn-O bond length increases with increasing the Pr concentration z[6, 8]. This indicates a change in the orbital occupancy of $d\gamma$-state in Mn ion: increase of the $d(3z^2\text{-}r^2)$ orbital polarization.

Although such interesting experimental results were reported, the understanding of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ has not been clear, yet. It is suggested that a increase of the $d(3z^2\text{-}r^2)$ orbital polarization make the FM interaction weak. Consequently this leads to the disappearance of the spontaneous magnetic phase transition around z=0.5[7]. Especially the low-temperature property for z=0.6 has been not well-established so far. In this paper, in order to clarify the low-temperature

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property of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6, the detailed magnetization measurement was performed.

Single crystals of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 were grown from a sinted rods of the same nominal composition by the floating-zone technique by using a minor furnace. The dimension of the sample were 5 × 4mm in the *ab*-plane and 1 mm along the *c*-axis. The magnetization (M) measurements were performed by the vibrating sample method (VSM) up to 14 T and at temperatures down to 3.7 K.

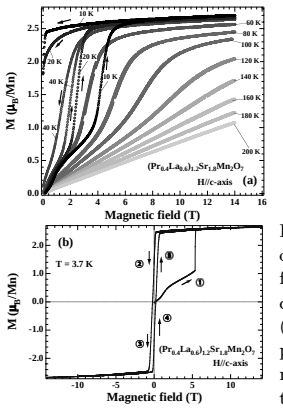

Fig. 1. (a) M-H curves of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 under fields along c-axis above T^*= 40 K and (b) below T^* at selected temperature. The number denotes order of the magnetization process.

Figure1(a) shows the magnetization curves under the field along *c*-axis at selected temperature above T^*= 40 K. A metamagnetic transition behavior gradually becomes broader with increasing a temperature. The magnetization curves at selected temperature below T^*= 40 K is also shown in the Fig. 1(b). This result is almost same as that of the previous measurement[6]. A clear rectangular hysteresis loop was observed below T^*, implying ferromagnetic behavior. The number denotes order of the magnetization process. In contrast, the transition does not accompany a hysteresis above T^*.

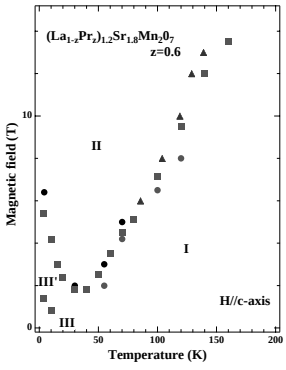

Fig. 2. The magnetic phase diagram of $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6 for $H//c$-axis. The squares, triangles, circles denote the transition points determined by the M-H curves, M-T curves (Ref. 9) and the previous elastic constant measurement (Ref. 9).

From these results combined with our previous data of ultrasonic measurement and the temperature dependence of magnetization[9], one can draw the (H-T) phase diagram. There are three main phases as labeled by Greek number in Fig. 2. The phase I is paramagnetic, and the phase is probably induced ferromagnetic phase superimposed on the orbital ordering. It seems that the phase III' is ascribed to the domain rotation. Here, let us consider the phase III and III'. It is noted that the boundary going down to about 40 K is obscure in the absence of any fields. Actually, no distinct anomaly has been observed in the magnetic susceptibility and resistivity. However, the anomaly is enhanced by applying magnetic fields, and a magnetoresistive memory effect was observed in the phase III. Time-dependent magnetization measurements performed by Gordon *et al.*, indicates the spin glass-like behavior in the phase III[7]. Furthermore, our previous ultrasonic measurement indicates that the longitudinal elastic constant C_{11} and C_{33} exhibits a pronounced softening with a large hysteresis in the pahse III[9]. It seems that this observed hysteresis is ascribed to the strain susceptibility. Actually, the striction measurement also shows a pronounced anomaly on the boundary between II and III, accompanied large hysteresis[6, 8]. At present we propose the following interpretation of the phase III. Since the field dependence of elastic constant accompanies a large hysteresis below T^*, the orbital of 3d state such as $d(xy)$, $d(yz)$ and $d(zx)$ has glass state. Following this orbital restriction, the corresponding spin has the resultant spin-glass state, *i.e.*, the magnetoresistive memory effect. In summary, we performed the magnetization measurement on $(La_{1-z}Pr_z)_{1.2}Sr_{1.8}Mn_2O_7$ for z=0.6. From these present results and our previous ones of the ultrasonic measurement, magnetic phase diagram was established. There are three main phases in which the ground state is suggested to be orbital- *i.e.*, resultant spin-glass state. The comparison of the same study with different concentration for z=0.2 and 0.4 will provide us significant information, and it is now in progress.

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