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Dramatic change of magnetic property in the A-site ordered/disordered manganites $PrBaMn_2O_6/Pr_{0.5}Ba_{0.5}MnO_3$

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Abstract

The magnetic properties of the A-site ordered/disordered manganese perovskite $PrBaMn_2O_6/Pr_{0.5}Ba_{0.5}MnO_3$ have been investigated in terms of the extent of the A-site randomness. The A-site disorder suppresses both ferromagnetic and A-type antiferromagnetic transitions in the A-site ordered $PrBaMn_2O_6$, and leads to a magnetic glassy state in the strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$. Ultrasharp and stepwise changes of magnetization and resistivity have been observed in the strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$.

Keywords: New metal-ordered/disordered manganites; $RBaMn_2O_6$

Due to the rich physical properties including the colossal magnetoresistance (CMR), manganese perovskites have drawn much attention in the fields of physics and engineering [1]. Many experimental and theoretical studies have revealed that the interesting properties such as CMR and electronic phase separation come from a critical competition between a ferromagnetic metal and an antiferromagnetic charge–orbital order interaction, which could be significantly influenced by the A-site randomness or a fluctuation of composition [2,3]. Recently, we synthesized the A-site ordered manganese perovskites $RBaMn_2O_6$ (R = Y and rare earth elements) and reported the electronic phase diagram of $RBaMn_2O_6$ [4]. The novel properties observed in $RBaMn_2O_6$ are not only due to the absence of the A-site randomness but also due to the layer-type structure with the $BaO–MnO_2–RO–MnO_2$ stacking sequence along the *c*-axis [4–6]. We also successfully synthesized the A-site disordered $R_{0.5}Ba_{0.5}MnO_3$ and have studied the A-site randomness effect in Ba-based manganites. In this paper, we report the magnetic properties of the ordered/disordered $PrBaMn_2O_6/Pr_{0.5}Ba_{0.5}MnO_3$, which strongly depend on the extent of randomness with Pr and Ba in the A-site.

Powder samples of the ordered $PrBaMn_2O_6$ were synthesized by a solid-state reaction of Pr_6O_{11}, $BaCO_3$ and MnO_2 as described in Ref. [4]. The extent of the A-site disorder strongly depended on the preparation process. The weak disordered samples were obtained by annealing the ordered samples at 900−1300° C in O_2 gas. The strong disordered samples were prepared by a solid- state reaction of the same starting materials in at 1350° C 1% O_2/Ar gas, followed by annealing at 900° C in O_2 gas. The obtained products were checked to be single phase by X-ray diffraction using CuK_α radiation. The extent of order/disorder with Pr and Ba was estimated by the peak intensity of $(0\,0\,\frac{1}{2})_p$ reflection indexed with the primitive cell.

The ordered $PrBaMn_2O_6$ with the strong $(0\,0\,\frac{1}{2})_p$ reflection has a tetragonal structure ($a = 3.9088(1)$ Å, $c = 7.7649(4)$ Å), while the strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$ without $(0\,0\,\frac{1}{2})_p$ reflection has a cubic structure ($a = 3.8951(1)$ Å). The weak disordered $Pr_{0.5}Ba_{0.5}MnO_3$, which shows the weak $(00\frac{1}{2})_p$ reflection has a tetragonal structure ($a = 3.9038(2)$ Å, $c = 7.7906(7)$ Å).

The magnetic susceptibilities (M/H) measured under 1000 Oe are shown in Fig. 1. The ordered $PrBaMn_2O_6$ shows the ferromagnetic transition at $T_C = 310$ K, followed by the A-type antiferromagnetic transition at $T_N = 270$ K (Fig. 1(a)). The weak disordered

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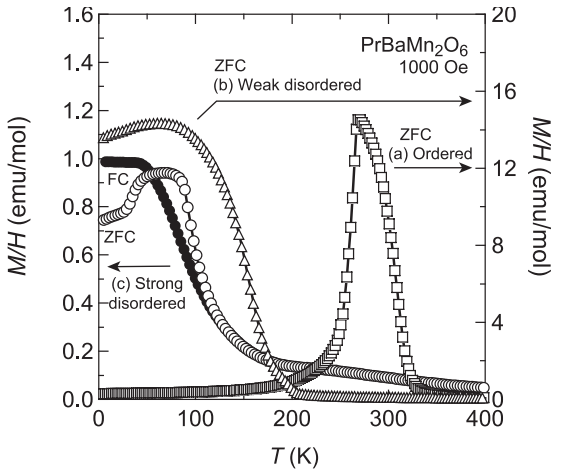

Fig. 1. The magnetic susceptibilities (M/H) of (a) ordered (□:ZFC), (b) weak disordered (△:ZFC) and (c) strong disordered (○:ZFC, ●:FC) $PrBaMn_2O_6$ as a function of temperature.

$Pr_{0.5}Ba_{0.5}MnO_3$ exhibits the ferromagnetic transition at $T_C = 160\,K$ (Fig. 1(b)) but does no transition below 160 K. The strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$ has a magnetic transition at 160 K, showing a very small spontaneous magnetic moment about $0.2\,\mu_B$ and a significant difference of M/H–T curves on zero-field cooled (ZFC) and field cooled (FC) processes. The A-site randomness clearly suppresses not only the ferromagnetic transition but also the A-type antiferromagnetic transition to much extent. It is natural that the A-site disorder destabilizes the A-type antiferromagnetic state associated with the $d_{x^2-y^2}$ orbital order (layer type). On the other hand, the magnetic glassy behavior observed in the strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$ could be due to a disorder effect that hinders the long-range magnetic ordering, namely that could occur as a result of the competition between randomly distributed ferromagnetic and antiferromagnetic interactions. Since the ionic radius of Ba^{2+} is much larger than Sr^{2+} ($= 1.44$ Å) [7], the strong disordered $R_{0.5}Ba_{0.5}MnO_3$ may include the special heterogeneity in nanometer size, which leads to the magnetic nonhomogeneous state. Very interestingly, ultrasharp and stepwise changes of magnetization and resistivity have been observed in the strong disordered $Pr_{0.5}Ba_{0.5}MnO_3$ [8]. The microscopic measurements such as neutron diffraction and electron microscopy are now in progress in order to fully understand these behaviors.

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