Correlation between A-site randomness and magnetic phase transition in half-doped manganite $Pr_{0.5}Ba_{0.5}MnO_3$

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Correlation between A-site randomness and magnetic phase transition in half-doped manganite $Pr_{0.5}Ba_{0.5}MnO_3$

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Abstract. The interplay between A-site randomness and magnetic phase transition in half-doped manganite $Pr_{0.5}Ba_{0.5}MnO_3$ (PBMO) was extensively investigated. The *H/M* versus M^2 isotherms show that the A-site ordered PBMO undergoes a second-order magnetic phase transition from paramagnetism to ferromagnetism, whereas the A-site disordered PBMO exhibits a fluctuation-driven first-order transition arising from a competing order phase *possibly* existing in the paramagnetic state. A step-like transition in magnetization and resistivity with a sharp width of $\Delta H/H \sim 10^{-3}$ was only observed in the A-site partially ordered PBMO at 2 K, indicating that the metamagnetic transition is associated with a competition between randomly distributed short-range ferromagnetic and antiferromagnetic phases. These findings provide evidence that the A-site randomness not only suppresses A-type antiferromagnetism also moderately weakens long-range ferromagnetism in the A-site ordered PBMO.

1. Introduction

The half-doped manganites $R_{0.5}A_{0.5}MnO_3$ (R=rare-earth and A=alkaline-earth cations) have attracted considerable attention due to a wide variety of emergent properties such as colossal magnetoresistance (CMR), ferromagnetic (FM) metal-antiferromagnetic (AFM) insulator transition, and field-induced transition from the CE-type charge ordered (CO) AFM state to FM metallic phase [1-6]. It is widely believed that these striking phenomena are caused by the strong correlations/competitions of spin, charge, orbital, and lattice degrees of freedom which could be substantially tuned by the tolerance factor, the A-site cation size mismatch, and the Mn-site doping [7]. Among them, the effect of random potential due to quenched disorder arising from the mismatch effect of the A-site cations has a significant influence on electronic phase separation. Therefore, to quantify the dramatic effect of the quenched disorder on the CMR physics, a detailed study on manganites that are free of the quenched disorder is preferable. Owing to the large difference in the ionic radii of Pr^{3+} and Ba^{2+} as well as the complete solid solution of Pr^{3+} and Ba^{2+} ions, the half-doped manganite $Pr_{0.5}Ba_{0.5}MnO_3$ is a good candidate for studying the effect of the A-site quenched disorder on the physics of phase competition and related phenomena. Recently, Ueda *et al.* have studied structural and magnetic properties of the A-site ordered/disordered $Pr_{0.5}Ba_{0.5}MnO_3$ [8-10]. In this work, we have successfully synthesized three polycrystalline samples of $Pr_{0.5}Ba_{0.5}MnO_3$ with various amount of the A-site disorder by the standard solid-state reaction method. An attempt was made to systematically investigate the interplay between A-site quenched disorder (A-site randomness) and magnetic phase transition.

2. Experimental

The A-site ordered sample was synthesized by calcining a well-ground stoichiometric mixture of Pr_6O_{11}, $BaCO_3$ and Mn_2O_3 powders at 1000 ^oC in flowing Ar gas for 24 h, followed by sintering at 1300 ^oC in high-pressure Ar atmosphere for 48 h and then annealing at 500 ^oC in O_2 for 60 h. The A-site partially ordered sample was obtained by further annealing the as-prepared A-site ordered sample at 1000 ^oC in O_2 for 120 h. In addition, the A-site disordered sample was made by calcining a stoichiometric mixture of starting powder at 1100 ^oC in air for 2 h, followed by sintering at 1500 ^oC in air for 2 h and then annealing at 900 ^oC in air for 96 h. The structure analysis was characterized by powder x-ray diffraction (XRD) method using a Rigaku RINT-2100 diffractometer with Cu K_α radiation. Magnetoresistance and magnetic measurements were performed in a 9-T QUANTUM DESIGN physical property measurement system (PPMS).

3. Results and Discussion

The crystal structure of the A-site ordered PBMO with alternate stack of the Pr-O and Ba-O rock-salt layers separated by the MnO_2 plane and that of the A-site disordered (solid solution) one are illustrated in Fig. 1(a) for comparison. Figure 1(b) shows the observed and refined XRD patterns for samples studied. All Bragg peaks can be indexed with a tetragonal lattice of space group *P4/mmm* for the A-site ordered PBMO and with a cubic lattice of space group $Pm\bar{3}m$ for the A-site disordered PBMO. The refined occupation number of Pr and Ba with decent weighted profile *R*-factor R_{wp} indicates that the "ordered" sample contains 98 % of the A-site ordered PBMO, whereas the "disordered" sample is an A-site disordered PBMO. In addition, the "mixed" sample is an A-site partially ordered PBMO which includes 32 % of the A-site ordered PBMO and 68 % of the A-site disordered PBMO. It is further confirmed by a progressive decrease in the intensity of the (001) peak, corresponding to an increase of the A-site randomness in half-doped manganites [10,11], from the ordered sample to the disordered sample as revealed in the inset of Fig. 1(b).

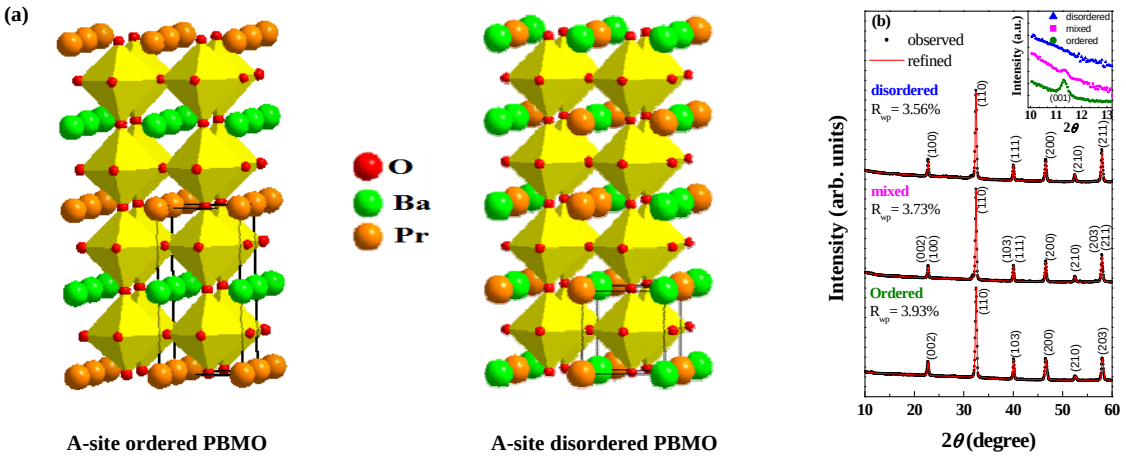

Figure 1. (a) Crystal structure of A-site ordered/disordered PBMO. (b) The observed and refined XRD patterns for the ordered, mixed, and disordered samples, respectively. The inset shows the intensity of the (001) peak for samples studied.

The temperature dependence of the zero-field-cooled (ZFC) magnetic susceptibility in a field of 100 G is displayed in Fig. 2(a). It clearly demonstrates that the samples with various amount of A-site randomness exhibit different magnetic states at low temperatures. The ordered sample undergoes a ferromagnetic (FM) transition at T_C = 311 K and then evolves into an A-type antiferromagnetic (AFM) state below 265 K, confirmed by neutron diffraction studies [8]. It is likely associated with the

$d_{x^2-y^2}$ orbital stabilization arising from the local lattice distortion in MnO_6 octahedra [12], whereas the disordered sample with the largest A-site randomness among samples investigated goes through a magnetic transition around 125 K. Comparing with the disordered sample, the mixed sample reveals a much wider transition width and a smaller magnetization at low temperatures, suggesting that a larger amount of short-range AFM phase exists in the mixed sample.

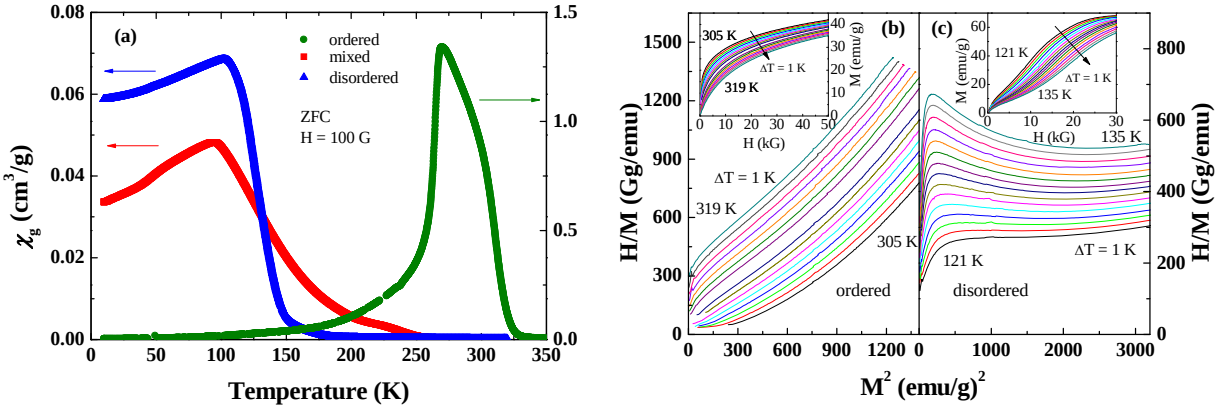

Figure 2. (a) Zero-field cooled magnetic susceptibility as a function of temperature for ordered, mixed, and disordered samples. (b) and (c) The Arrott plot for ordered and disordered samples, respectively in the vicinity of transition temperature. The insets display corresponding $M(H)$ isotherms.

To analyze the nature of the magnetic phase transition in PBMO with different amount of A-site randomness, the slope of the Arrott plot (H/M vs M^2) is examined. Based on the Banerjee criterion [13], the slope of H/M vs M^2 is negative for a first-order transition and positive for a second-order transition, respectively. As shown in Fig. 2(b), the slope of H/M vs M^2 isotherms for the ordered sample near 311 K is positive throughout, indicative of a second-order transition with a Brillouin-like M-H isotherms displayed in the inset of Fig. 2(b). In contrast, the corresponding plots for the disordered sample near 125 K evidently exhibit a negative slope shown in Fig. 2(c), indicative of a fluctuation-driven first-order transition arising from a competing order phase *possibly* existing in the paramagnetic state.

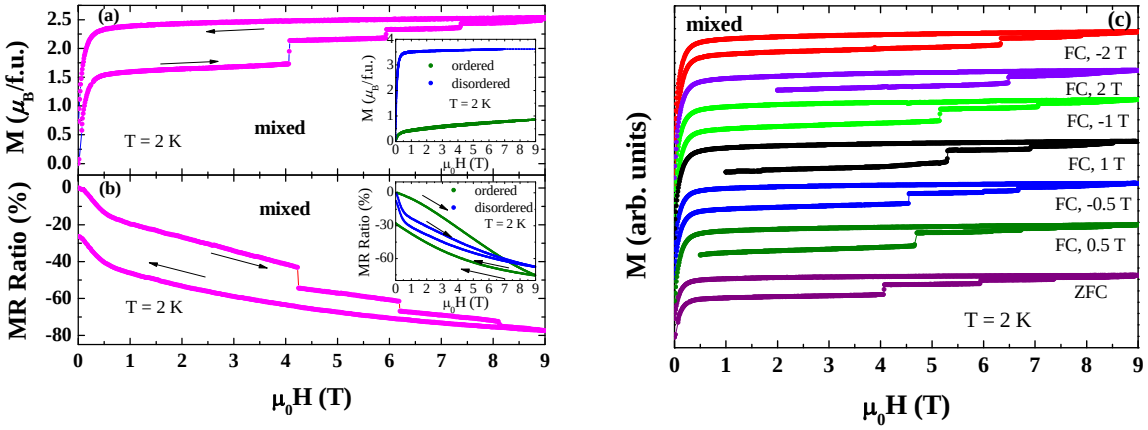

Figure 3. (a) M-H curve for the mixed sample at 2 K. The inset displays M-H curve for the ordered and disordered samples at 2 K. (b) MR ratio versus H for the mixed sample at 2 K. The corresponding plot for the ordered and disordered samples is displayed in the inset for comparison. (c) M-H curve for

the mixed sample at 2 K under various field-cooled processes. Each curve is placed separately for clarity.

As shown in Figs. 3(a) and 3(b), magnetization exhibits three large jumps at 4.08 T, 5.93 T and 7.37 T, respectively, whereas resistivity accordingly decreases stepwise around similar fields at 2 K for the mixed sample. It should be noted that the multistep transition in magnetization and resistivity is not reversible in the field-ramping-down process and only observed in the mixed sample as revealed in the inset of Figs. 3(a) and 3(b). The successive step-like transition with extraordinarily sharp width of $\Delta H/H \sim 10^{-3}$ is likely associated with a competition between randomly distributed short-range FM and AFM phases. The magnetic moment at 9 T is smaller than the theoretical value of 4.38 μ_B, suggesting that the magnetic-field-induced metamagnetic transition is associated with an *abrupt precipitation* of the FM regions at the expense of the AFM clusters. To have a better understanding of the competing magnetic ground states in the mixed sample (A-site partially ordered PBMO), $M(H)$ at 2 K under ZFC and positive/negative field-cooled (FC) processes is shown in Fig. 3 (c). Note that the step transition remarkably shifts to higher fields after cooling the mixed sample in a higher applied field regardless of the polarity. It is quite striking because the field-cooled process normally weakens the AFM exchange interaction and is expected to reduce the threshold fields for the step transition. Further investigations are needed to resolve the origin of the intriguing metamagnetic transition.
In summary, we have demonstrated that the A-site randomness has a significant influence on magnetic properties of the half-doped PBMO. The presence of the A-site randomness leads to a metamagnetic transition at low temperatures in the A-site partially ordered PBMO and induces a first-order phase transition in the A-site disordered PBMO.

Acknowledgements
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References
[1] Tomouka Y, Asamitsu A, Moritomo Y, Kuwahara H and Tokura Y 1995 *Phys. Rev. Lett.* **74** 5108
[2] Radealli P G, Cox D E, Marezio M, Cheong S W, Schiffer P and Remirez A P 1995 *Phys. Rev. Lett.* **75** 4488
[3] Maignan A, Hebert S, Hardy V, Martin C, Hervieu M and Raveau B 2002 *J. Phys.: Condens. Matter* **14** 11809
[4] Mahendiran R, Maignan A, Hebert S, Martin C, Hervieu M, Raveau B, Mitchell J F and Schiffer P 2002 *Phys. Rev. Lett.* **89** 286602
[5] Aliaga H, Magnoux D, Moreo A, Poiblang D, Yunoki S and Dagatto E 2003 *Phys. Rev. B* **68** 104405
[6] Murakami S and Nagaosa N 2003 *Phys. Rev. Lett.* **90** 197201
[7] Rao C N R and Raveau B 1998 *Colossal Magnetoresistance, Charge Ordering and Related Properties of Manganese Oxides* (World Scientific)
[8] Nakajima T, Kageyama H, Yoshizawa H, Ohoyama K and Ueda Y 2003 *J. Phys. Soc. Jpn.* **72** 3237
[9] Ueda Y and Nakajima T 2004 *J. Phys.: Condens. Matter* **16** S573
[10] Nakajima T, Yoshizawa H and Ueda Y 2003 *J. Phys. Soc. Jpn*. **73** 2283
[11] Trukhanov S V, Lobanovski L S, Bushinsky M V, Fedotova V V, Troyanchuk I O, Trukhanov A V, Ryzhov V A, Szymczak H, Szymczak R and Baran M 2005 *J. Phys. : Condens. Mater.* **17** 6495
[12] Takeshita N, Terakura C, Akahoshi D, Tokura Y and Takagi H 2004 *Phys. Rev. B* **69** 180405(R)
[13] Banerjee S K 1964 *Phys. Lett.* **12** 16