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Heisenberg-like ferromagnetism and percolative conductivity in the half-doped manganite $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$

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ABSTRACT

The magnetic behavior and electronic transport in the half-doped manganite $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$ have been investigated. The critical exponents are studied by using isothermal magnetization methods. The results show that the paramagnetic–ferromagnetic transition is second order and the magnetic interaction is satisfied with the prediction of three-dimensional Heisenberg model. The electronic transport belongs to the percolation mechanism. These findings demonstrate that the critical behavior of the magnetic transition and conductivity for manganites are related to Mn-site ordering degree.

1. Introduction

Perovskite manganites $R_{1-x}A_xMnO_3$ (R=trivalent rare earth, A=divalent alkaline earth) show a great variety of fascinating properties such as colossal magnetoresistance (CMR) effect and electronic phase separation [1–4]. It has been extensively accepted that the double exchange (DE) effect in which e_g electrons hop between Mn^{3+} and Mn^{4+} visa oxygen ions with a strong on-site Hunds-rule coupling is essential for understanding the magnetic and transport properties in manganites [5]. However, if the DE effect is solely considered, the calculated resistivity obviously deviated from the experimental observations [6,7]. Then, a electron–phonon coupling was included due to Jahn–Teller lattice distortion [8,9].

Besides CMR effect, another intriguing phenomenon of charge ordering (CO) phase has been frequently observed in manganite. As we know, for the half-doped manganites $R_{0.5}A_{0.5}MnO_3$, the CO phase reveals a periodic arrangement of Mn^{3+} and Mn^{4+} ions. Usually, the CO state can be kept stably in manganites, except for some extreme exoteric perturbations. For example, Tokunaga et al. [10] found that the CO state still remained stable even under magnetic fields up to 27 T in $Pr(Nd)_{0.5}Ca_{0.5}MnO_3$. In addition to the external field, quenched disorder arising from the local lattice distortion and/or doped impurities can significantly modify CO state. A few percent substitutions of Cr easily destroy the long-range CO phase and induce the ferromagnetic phase locally, which results in the phase separated ground state with both CO and FM clusters randomly distributed [11–13]. However, our recent research shows that the paramagnetism–ferromagnetism (PM–FM) transition and insulator–metal (IM) transition are observed in the half-doped manganite $Nd_{0.5}Sr_{0.25}Ca_{0.25}MnO_3$ [14]. Due to the large mismatch of ionic size between Ca^{2+} and Sr^{2+} (r_{Ca}=1.18 Å, r_{Sr}=1.31 Å), a considerable disorder on A-site causes inhomogeneous strain field so that it is impossible to form a long-rang CO phase. However, for the reason of PM–FM phase transition and IM transition, it is not very clear in the present system. In this paper, we measured the static magnetization around Curie temperature (T_C) and investigated the critical properties. The obtained critical exponents are close to that predicted by a three-dimensional (3D) Heisenberg model. It suggests that the observed ferromagnetism is short-range magnetic interaction.

2. Experiment

Polycrystalline sample $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$ was synthesized by the conventional solid-state reaction method with high pure Nd_2O_3, $SrCO_3$, $CaCO_3$, MnO_2. The detailed experimental process has been reported in Ref. [14]. The Powder X-ray diffraction was employed on Japan Rigaku D/max-$\gamma\alpha$ rotating powder diffractometer using Cu Kα radiation to check the structure and phase purity. The samples were proved to be single-phase orthorhombic

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structure. The resistivity (ρ) was measured by standard four-probe method. The magnetization (M) measurement was performed using the Quantum Design Superconducting Quantum Interference Device under 0.01 T magnetic field in the range of 5–300 K. The $M(H)$ curve was measured with sweep field from 0 to 3.0 T. Moreover, in order to ensure a perfect demagnetization of the samples, each measurement was performed only after the sample was heated firstly above T_C.

3. Results and discussion

Fig. 1(a) shows the temperature dependence of magnetization and resistance for $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$ sample. The conductivity of $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$ displays an IM transition at $T_{Im}=175$ K. Correspondingly, a PM–FM transition occurs at the same temperature. Generally, as for the half-doped manganites, the charge ordering state and antiferromagnetic phase are ubiquitous. However, in the present materials, these characters have not been observed. In our previous investigation [14], we have suggested that the A-site cation disorder induced by the size mismatch between Sr^{2+} ion and Ca^{2+} ion was mainly responsible for this phenomenon. However, for further understanding the above behavior, two important questions about PM–FM transition should be firstly considered: one is the order of phase transition, the other is the common universality class. To make these issues clear, we investigated the critical exponents around the region of the PM–FM transition based on the measurement of isothermal magnetization as shown in Fig. 1(b).

As we known, the thermodynamic function around the critical point can be expressed by a power law form. According to the scaling hypothesis [15,16], a second-order magnetic phase transition near Curie point is characterized by a set of critical exponents, β (associated with the spontaneous magnetization M_s), γ (associated with the initial magnetic susceptibility χ_0), δ (associated with the critical magnetization isotherm at T_C). The mathematical definitions of the exponents from magnetization measurements are given below:

$$M_S(T)=M_0(-\varepsilon)^{\beta},\quad \varepsilon<0,\ T<T_C \tag{1}$$

$$\chi_0^{-1}(T)=(h_0/M_0)\varepsilon^{\gamma},\quad \varepsilon>0,\ T>T_C \tag{2}$$

$$M=DH^{1/\delta},\quad \varepsilon=0,\ T=T_C \tag{3}$$

where ε is the reduced temperature $(T-T_C)/T_C$, and M_0, h_0/M_0, and D are the critical amplitudes.

In addition, near the critical point of a second-order transition, the free energy G can be expressed in terms of the order parameter M in the following form:

$$G(T,M)=G_0+aM^2+bM^4-MH \tag{4}$$

where the coefficients of a and b are temperature-dependent parameters. For the condition of equilibrium, i.e., energy minimization, $\partial G/\partial M=0$, the magnetic equation of state is obtained as

$$H/M=2a+4bM^2 \tag{5}$$

Thus, the relationship of M^2 vs. H/M should be shown as a linear behavior around T_C. According to the criterion proposed by Banerjee [17], the order of magnetic transition can be determined from the slope of straight line. The positive slope corresponds to the second-order transition while the negative slope corresponds to the first-order transition. Fig. 2(a) is an Arrott plot of M^2 vs. H/M. Clearly, in the present case the positive slope of M^2 vs. H/M curves indicates the phase transition is a second-order PM–FM phase transition. However, all the curves in the Arrott plot are nonlinear and shows upward curvature even at high field indicating the critical exponent of $\beta=0.5$ and $\gamma=1.0$ is not satisfied according to Arrott–Noakes equation of state $(H/M)^{1/\gamma}=(T-T_C)/T_C+(M/M_1)^{1/\beta}$ [18]. Namely, the mean-field theory [16,19], which exponent is of $\beta=0.5$ and $\gamma=1.0$, cannot be used to describe the critical behavior in present $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$ system.

In order to determine the critical exponents accurately, a modified Arrott plot with Arrott–Noakes equation is used. As the critical exponents β and γ are correctly chosen, the modified Arrott plot will produce a set of parallel straight lines. Using a polynomial fit with Eq. (5) and extrapolating the data in Fig. 2(a), the intercepts of the isotherms on the H/M and M^2 axes give the values of $\chi_0^{-1}(T)$ for $T>T_C$ and $M_S(0,T)$ for $T<T_C$, respectively. Thus, one can plot the $M_S(0,T)$ vs. T and $\chi_0^{-1}(T)$ vs. T curves, as shown in set of Fig. 2(b). Then, using Eqs. (1) and (2), the fitting results produce two new critical exponents β and γ. These two new values of β and γ are used to make the Arrott plot. The procedure is performed repeatedly until the values of β and γ do not change. Finally, the modified Arrott plot is shown in Fig. 2(b). With the final value of $\beta=0.386(2)$ and $\gamma=1.174(4)$, one can obtain a set of parallel lines around critical region. The obtained critical exponents are close to theoretically predicted values of 3D-Heisenberg magnets ($\beta=0.365$ and $\gamma=1.336$) [20,21]. Alternatively, the critical exponents can be obtained from the

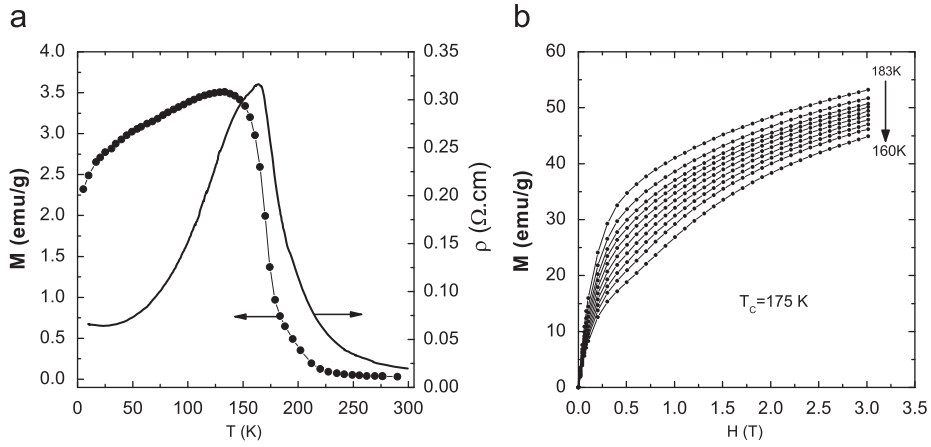

Fig. 1. (a) Magnetization vs. temperature and resistivity vs. temperature of $Nd_{0.5}Ca_{0.25}Sr_{0.25}MnO_3$; (b) magnetization vs. magnetic field at different temperatures around the Curie temperature ($T_C=175$ K).