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Scaling study of magnetic phase transition and critical behavior in $Nd_{0.55}Sr_{0.45}Mn_{0.98}Ga_{0.02}O_3$ manganite

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ABSTRACT

Microscopic magnetic interactions and magnetism can be probed by studying the critical paramagnetic–ferromagnetic phase transition of magnetic materials. In this work, the critical behavior of the lightly Ga-doped manganites $Nd_{0.55}Sr_{0.45}Mn_{0.98}Ga_{0.02}O_3$ (NSMGO) has been investigated. Scaling analysis reveals the critical exponents $\beta = 0.308$, $\gamma = 1.197$, and $\delta = 4.916$, where their self-consistency and reliability are verified by the Widom scaling law and the scaling equation. These exponents indicate that the magnetic phase transition in NSMGO belongs to the anisotropic 3D-Ising universality class. Nevertheless, the asymptotic critical exponents β_{eff} and γ_{eff} are found to approach to the values of 3D-Heisenberg model, implying the existence of isotropic short-range magnetic interactions. We suggest that, although the Nd-based manganites possess a strong magnetocrystalline anisotropy, this effect can be partly averaged out in view of the present polycrystalline NSMGO. Therefore, in the asymptotic critical region, the phase transition reveals the crossover from the scaling of 3D-Ising toward 3D-Heisenberg universality class.

1. Introduction

Perovskite oxides have attracted intense research interests during past decades in both materials science research and condensed matter physics, which comprise of a broad spectrum of interesting functionalities, such as high temperature superconducting (high T_C copper oxides) [1], colossal magnetoresistance (manganite) [2], and more recently multiferroicity ($BiFeO_3$/$TbMnO_3$) [3,4]. The wide range of material properties and rich physics involved have led to extensive studies to understand the fundamental nature of existing systems, so as to better control/design novel materials for applications. Among them, perovskite manganites with the general formula $A_{1-x}B_xMnO_3$ (A = rare earth element, B = divalent alkaline earth element) have been paid great attention due to the strong coupling between the structure, electron and magnetism which causes an incredibly rich panoply of phenomena including paramagnetic–ferromagnetic (PM–FM) transition, insulator-metal transition (IMT), charge/orbital ordering, and colossal magnetoresistance effect [5,6].

Extensive efforts to understand these mechanism have shown that the doping concentration x controls the band filling and the e_g electron transfer in $A_{1-x}B_xMnO_3$ [7]. By partial substitution with divalent elements B for A, a corresponding amount of Mn^{3+} was converted into Mn^{4+}, leading to the appearance of mixed-valence state Mn^{3+}/Mn^{4+}. Many studies have indicated that the double-exchange (DE) interaction between Mn^{3+} and Mn^{4+} can cause a strong PM–FM transition together with IMT around the Curie temperature [8–12]. Therefore, the DE interaction together with electron-lattice coupling theories were proposed to understand the correlation between magnetic properties and electronic transport [13]. On the other hand, The perovskite structure generally shows lattice distortion from the cubic structure to orthorhombic or rhombohedral ones. This distortion is mainly due to the Jahn–Teller (JT) effect, inducing the deformation of the MnO_6 octahedron. It has been noted that cooperative JT distortions are present in the orthorhombic phase, but not allowed in the rhombohedral one due to a higher symmetry of MnO_6 octahedra [14,15]. Accordingly, the DE interaction between Mn^{3+} and Mn^{4+} in these manganites is strongly related to JT effect and distortion of MnO_6 octahedron [16]. Therefore, the current theory can only partly explain the magnetic properties and electronic transport in perovskite manganites because many distinct deviations from the above DE and JT theory have been reported in experiments [17,18]. To better understand these exotic properties in the doping manganites, it is essential to perform comprehensive studies on the uppermost nature of PM–FM phase transition [19,20]. One of the effective methods is to investigate the critical behavior in close vicinity

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of this phase transition [21–24]. In fact, scaling analysis of the critical behavior in the vicinity of magnetic phase transition is a powerful tool to obtain important microscopic information about the underlying magnetic interactions. Moreover, the study of critical exponents allows the determination of the correlation of these exponents with those of the appropriate model, existing in the literature, and the exchange integral in the ferromagnetic state. Not only that, scaling method have been recently applied to study chiral helimagnetic FeGe and $Cr_{1/3}NbS_2$ [25–28]. These materials generally show a special spin textures, such as the conical magnetic ordering, the magnetic skyrmion, and the chiral magnetic bubble. The analysis of critical behavior can further give us a penetrating insight into the potential mechanism of magnetic coupling and clarify the whole process of development and evolution of their spin textures.

Perovskite manganites $Nd_{0.55}Sr_{0.45}MnO_3$ is ferromagnetic metallic and has high Curie temperature T_C = 273 K. Scaling study of the second-order PM–FM phase transition shows that it belongs to the framework of mean field model [29]. Generally, for the most isotropic doping manganites, the ionic substitution for Mn^{3+}/Mn^{4+} of B-site sublattice prevents the DE effect from forming long-range exchange coupling but yields short-range interaction [30,31]. Thus, the magnetic exchange interactions are usually close to 3D-Heisenberg model. However, in contrast to the general belief, in this paper, we find the lightly Ga-doped $Nd_{0.55}Sr_{0.45}Mn_{0.98}Ga_{0.02}O_3$(NSMGO) shows 3D-Ising-Like phase transition based on the estimated critical exponents derived from scaling study. Although both models all describe short-range interaction, the 3D-Heisenberg interaction represents isotropic interaction while the 3D-Ising model is anisotropic interaction. Therefore, the unusual 3D-Ising model observed in NSMGO is associated with the strong anisotropic magnetism of pristine Nd-based compounds and the disorder effect of Ga^{3+}-substitution.

2. Experiment

Polycrystalline NSMGO sample was prepared by traditional solid state reaction method. Stoichiometric quantities of high-purity oxides of Nd_2O_3, $SrCO_3$, Ga_2O_3, and MnO_2 were thoroughly mixed and ground, then preheated at 1173 K for 24 h. With intermediate grinding, they reacted at 1473 K for 24 h. After pressed into pellets, a final sintering was carried out at 1673 K for 30 h. The structure and phase purity of the sample were checked by powder X-ray diffraction (XRD) using Cu Kα radiation at room temperature. The XRD patterns prove that the sample is pure and a single-phase with orthorhombic structure. The magnetization versus magnetic field were measured by using a magnetic property measurement system (Quantum Design MPMS 7T-XL) with a superconducting quantum interference device (SQUID) magnetometer.

3. Results and discussion

According to whether there is the phase transition latent heat, the PM–FM phase transition can be classified into first-order and second-order. Generally, the first-order has large latent heat while the second-order phase transition has no latent heat. The phenomena associated with the second-order phase transitions are called critical phenomena. On the basis of the scaling hypothesis [32], for a magnetic material with second-order magnetic phase transition, the critical behavior near the Curie point can be 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), which is defined from magnetization as:

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

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

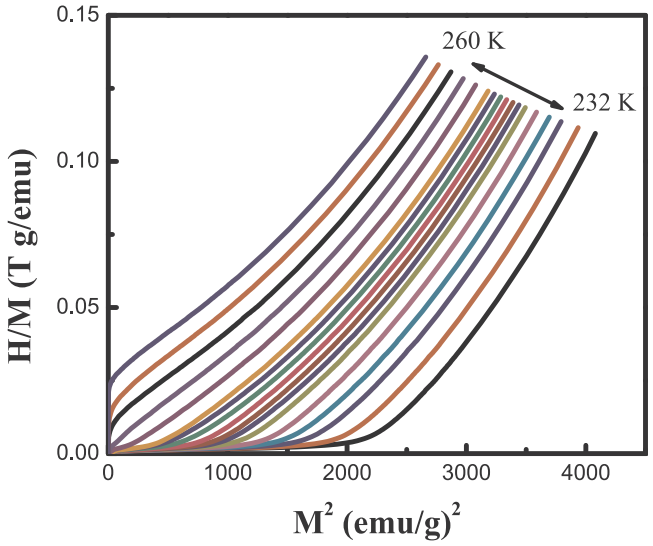

Fig. 1. Arrott plot: isotherms of H/M vs M^2 of $Nd_{0.55}Sr_{0.45}Mn_{0.98}Ga_{0.02}O_3$ at different temperatures close to the Curie temperature (T_C = 247 K).

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

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

According to thermodynamics, 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: $G(T, M) = G_0 + aM^2 + bM^4 - MH$, 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: $H/M = 2a + 4bM^2$. Thus, the relationship of M^2 vs H/M should be shown as a linear behavior around T_C. Banerjee proposed that the order of magnetic transition could be determined from the slope of straight line [33]. The positive slope corresponds to the second-order transition while the negative slope corresponds to the first-order transition. Fig. 1 shows the Arrott plot of M^2 vs H/M for NSMGO based on the isothermal magnetization measured around Curie point T_C = 247 K. 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 concave, indicating that the magnetization of NSMGO cannot be described by mean-field theory, i.e., with critical exponents β = 0.5 and γ = 1.0. [32] Moreover, the Arrott plot of Fig. 1 cannot be used to obtain spontaneous magnetization $M_s(T)$ and inverse susceptibility $\chi_0^{-1}(T)$ from the intercepts of the ordinate and abscissa, respectively.

In the asymptotic critical region ($|\varepsilon| < 0.1$), the critical exponents β and γ follow the Arrott–Noakes equation of state [34]:

$$(H/M)^{1/\gamma} = (T - T_C)/T_C + (M/M_1)^{1/\beta} \tag{4}$$

Thus, for the scaling analysis the isothermal magnetization data can be rescaled and presented as $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ in the modified Arrott plots. If the critical exponents β and γ are correctly determined, the modified Arrott plot will show a set of parallel straight lines. Here, we first choose three kinds of most possible critical exponents to construct the modified Arrott plots. They are 3D-Heisenberg model (β = 0.365 and γ = 1.336), tricritical mean-field model (β = 0.25 and γ = 1.0), and 3D-Ising model (β = 0.325 and γ = 1.24), respectively [35]. As shown in Fig. 2(a–c), three Arrott plots all exhibit quasi-straight lines in high field region and all lines of Fig. 2(a–c) are almost parallel to each other. In order to estimate which kind of critical exponents yields the best effect of parallel, we adopt the method of relative slope (RS) which is defined as $RS = S(T)/S(T_C)$, where $S(T) = dM^{1/\beta}/d(H/M)^{1/\gamma}$ is the slope of one parallel line at the region of high magnetic field [23]. As