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Critical behavior in polycrystalline $La_{0.7}Sr_{0.3}CoO_3$ from bulk magnetization study

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The critical behavior of hole-doped perovskite cobaltite $La_{0.7}Sr_{0.3}CoO_3$ has been investigated around the second-order paramagnetic-ferromagnetic (PM-FM) phase transition based on the static magnetization. Reliable critical exponents ($\beta = 0.272 \pm 0.001$, $\gamma = 1.291 \pm 0.004$, and $\delta = 5.54 \pm 0.02$ with critical point $T_C = 227.2 \pm 0.2$ K) have been determined by using different techniques, including the Modified Arrott plot, the Kouvel–Fisher method, and critical isotherm analysis. The obtained critical exponents not only obey the Widom relation $\delta = 1+\gamma/\beta$, but also can collapse the magnetization data $M(H,T)$ into two curves below and above T_C following a single scaling equation $M(H,\varepsilon) = \varepsilon^{\beta} f_{\pm}(H/\varepsilon^{\beta+\gamma})$, which implies the reliability and accuracy of the exponents. Temperature variation of the effective exponents resemble with those for disordered ferromagnets. The exponents analysis related to the magnetocaloric effect is studied. The field dependence of the relative cooling power (RCP) is found to obey $RCP \propto H^{1+1/\delta}$ with the exponent δ. The asymptotic critical exponents (β_{eff} and γ_{eff}) are close to those predicted by 3D-Ising model with the exchange interaction $J(r)$ decaying as $r^{-4.97}$ in the system, which reflects the existence of short-range FM coupling.

1. Introduction

Perovskite cobaltites $La_{1-x}Sr_xCoO_3$ have attracted great attention because they have exhibited diverse physical properties and complex magnetic phase diagram due to the presence of spin state transition of the Co ions induced by the temperature, compositional doping, magnetic field, or pressure [1–9]. $LaCoO_3$ itself is a nonmagnetic insulator with only low-spin Co^{3+} (t_{2g}^6, $S = 0$) at ground state [8–11]. Substituting La^{3+} with Sr^{2+} will generate microscopic hole-rich ferromagnetic (FM) metallic clusters in the hole-poor insulating antiferromagnetic (AFM) matrix, *i.e.*, the magnetoelectric phase separation, which leads to the microscopic inhomogeneity in $La_{1-x}Sr_xCoO_3$ ferromagnet [5–7]. This microscopic inhomogeneity affects the FM interaction between Co^{3+} and Co^{4+} ions, and the FM clusters grow in size and number with increasing Sr content, which leads to enhanced paramagnetic (PM) to FM phase transition in mixed-valence $La_{1-x}Sr_xCoO_3$ ($0 < x \leq 0.5$) series [2,8,12].

Because the Co^{3+} and Co^{4+} ions have complex spin states which affect the magnetic interaction and behavior, to well understand the intrinsic nature of the interactions in hole-doped cobaltites, it is important to clarify the order and nature of the PM-FM phase transition. The analysis of the critical exponents associated with the PM-FM transition is proved to be an effective way to find out the intrinsic nature of cobaltites ferromagnets [12–17]. Recently, N. Khan et al. have studied the critical behavior of $La_{1-x}Sr_xCoO_3$ single crystals with $x = 0.21$, 0.25 and 0.33, where $x = 0.25$ and 0.33 compounds behave as 3D-Heisenberg FM with nearest neighbor interaction, but $x = 0.21$ compound deviates from short range Heisenberg interaction towards long range mean-field like one [13,14]. A previous critical exponents study [15] in polycrystalline $La_{0.5}Sr_{0.5}CoO_3$ has shown that the exponent γ value is close to 3D Ising value whereas the exponent δ approaches to mean-field one. Nevertheless, S. Mukherjee et al. [16] have given a totally different physical scenario for the same composition, where all the values of critical exponents confirm that the system is a Heisenberg one. J. Mira et al. [12] have calculated the critical exponents of polycrystalline $La_{1-x}Sr_xCoO_3$ ($0.2 \leq x \leq 0.3$) by bulk magnetization under field only up to 1.1T. Their study shows that the critical exponent γ agrees with Heisenberg model while β approaches to mean-field

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model.

Since metallic ferromagnetism has been suggested for the $La_{1-x}Sr_xCoO_3$ with the range $0.30 \leq x \leq 0.50$ [16], it is important to clarify the critical behavior in the limit $x = 0.3$. Here, we yield the critical exponents for the polycrystalline $La_{0.7}Sr_{0.3}CoO_3$ ferromagnet by using various techniques through bulk magnetization under magnetic field up to 5T. The obtained exponents are found to obey scaling law and are close to 3D-Ising values.

2. Experiment

Polycrystalline $La_{0.7}Sr_{0.3}CoO_3$ sample was synthesized using the standard solid-state reaction method as described elsewhere [18]. As shown in Fig. 1(a), the phase purity of the as-prepared sample has been checked by the powder X-ray diffraction (XRD) patterns at room temperature. The Rietveld refinement analysis was performed on the XRD data. There is no obviously detectable impurity phase, and all diffraction peaks can be indexed to a rhombohedral perovskite lattice with $R\overline{3}c$ space group, which indicates the high quality of the as-prepared sample. The isothermal magnetizations were performed at 5 K interval over the temperature range from 155 to 280 K with field sweeping up to 50 kOe on a commercial Physical Property Measuring System (PPMS). The external applied magnetic field (H_a) has been corrected for the demagnetization effect to get the internal field (H_i) following the method in Ref. 13. The H_i dependent magnetization is shown in Fig. 1(b). The deduced H_i has been used for the scaling analysis below.

3. Results and discussions

Fig. 1(c) shows the temperature dependence of magnetization $M(T)$ for $La_{0.7}Sr_{0.3}CoO_3$ under a magnetic field of 500 Oe in zero-field-cooled (ZFC) and field-cooled (FC) sequences. With cooling, a sharp PM-FM phase transition arises at $T_C \approx 230$ K, which is determined by the flection in $dM(T)/dT$ curve (as shown in the right axis of Fig. 1(c)). With further cooling below T_C, the $M(T)$ curve shows a obvious divergence between ZFC and FC, which is often observed in perovskite cobaltites and can be attributed to the magnetic inhomogeneity [1–3,8,13,14].

The critical behavior of magnetization in vicinity of the second order magnetic phase transition can be characterized using a set of critical exponents (β, γ and δ), which are defined from magnetization as [19]:

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

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

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

where $\varepsilon=(T-T_C)/T_C$ denotes the reduced temperature, h_0/M_0 and D are the critical amplitudes.

Fig. 1(d) shows the Arrott plots (M^2 vs. H/M, i.e. $\beta = 0.5$ and $\gamma = 1$) near phase transition for the $La_{0.7}Sr_{0.3}CoO_3$ compound. Based on the mean-field theory, the regular Arrott plots near the phase transition should be a group of parallel lines in the high field regions, and the line at T_C should pass through the origin [20]. However, here all curves are nonlinear and show curvature character, which indicate that the mean-field model with critical exponents $\beta = 0.5$ and $\gamma = 1$ is not suitable to the system. By using Eqs. (1) and (2), the temperature variation of $M_S(T,0)$ and $\chi_0^{-1}(T,0)$ are fitted and shown in Fig. 2(a). The fitting result yields two sets of critical exponents of $\beta = 0.270 \pm 0.007$ with $T_C = 226.9 \pm 0.3$ K and $\gamma = 1.282 \pm 0.012$ with $T_C = 228.4 \pm 0.2$ K.

To further support the correctness of the obtained exponents and T_C, Kouvel–Fisher (KF) method [21] is applied to deduce the critical exponents β and γ along with T_C:

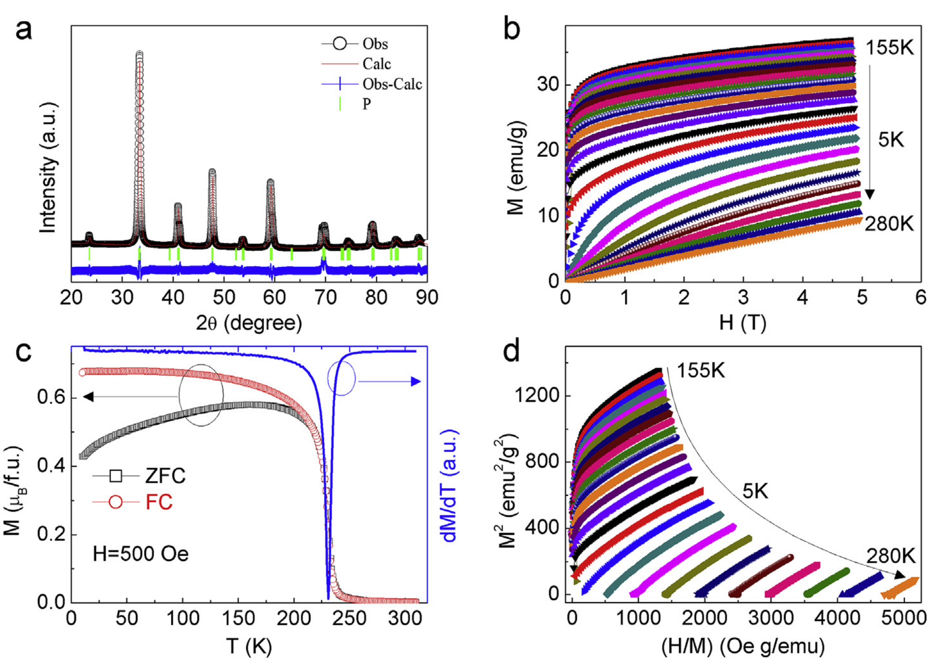

Fig. 1. (a) X-ray-diffraction patterns at room temperature for the sample, the symbols present the experimental data and the red solid curve is the fit from the Rietveld refinement, the vertical marks indicate the position of Bragg peaks and the bottom curve shows the difference between the observed and calculated intensities, (b) The isothermal magnetizations $M(H)$ around T_C, the field H is the internal field corrected for the demagnetization effect, (c) Temperature dependence of magnetization under ZFC and FC sequences (left axis) and dM/dT (right axis) under a 500 Oe field, (d) the corresponding Arrott plots M^2 vs. H/M (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).