Critical behavior in $La_{0.5}Sr_{0.5}CoO_3$

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We have studied the critical behavior in $La_{0.5}Sr_{0.5}CoO_3$ near the paramagnetic to ferromagnetic transition temperature. We have analyzed our dc magnetization data near the transition temperature with the help of the modified Arrot plot, Kouvel-Fisher method. We have determined the critical temperature T_c and the critical exponents β, γ, and δ. With the values of T_c, β, and γ, we plot $M/(1-T/T_c)^{\beta}$ vs $H/(1-T/T_c)^{\gamma}$. All the data collapse on one of the two curves. This suggests that the data below and above T_c obeys scaling, following a single equation of state. The exponents are close to Heisenberg values.

I. INTRODUCTION

$La_{1-x}Sr_xCoO_3$ shows the onset of a ferromagnetic (FM) transition for $x>0.2$.[1–3] Sr doping in the parent compound $LaCoO_3$ generates hole-rich, metallic ferromagnetic regions. In $La_{1-x}Sr_xCoO_3$, for $x<0.2$, the hole-rich regions are isolated from each other and show superparamagnetic behavior below $T_c \sim 240$ K.[1,4] Metallic ferromagnetism has been suggested for the range $0.30 \leqslant x \leqslant 0.50$.[1] However, the hole-poor matrix interpenetrating the ferromagnetic regions persists to $x=0.5$.[1,4,5] A previous study of critical exponents has been performed for $La_{0.5}Sr_{0.5}CoO_3$.[6] The study does not suggest a single universality class. The γ value is close to the three-dimensional Ising value whereas δ is close to the mean-field one. Their study does not incorporate the value of β. Recently[7] critical exponents of the paramagnetic-ferromagnetic (PM-FM) transition of $La_{1-x}Sr_xCoO_3$ ($0.2 \leqslant x \leqslant 0.3$) compounds have been calculated from the magnetization data. According to Mira *et al.*,[7] the value of γ was shown to correspond to a Heisenberg model but β is mean-field like. They have suggested that the system behaves like a Heisenberg model as the dilution of the magnetic lattice by the hole-poor regions prevent the occurrence of long-range order.[7] Although metallic ferromagnetism has been suggested for the range $0.3 \leqslant x \leqslant 0.5$, the hole-rich regions increase with increasing x even in this region. Hence we think, in order to understand the nature of the ferromagnetic transition in this system it is useful to study the critical exponents in detail associated with the transition in the extreme ferromagnetic limit $La_{0.5}Sr_{0.5}CoO_3$.

II. EXPERIMENTAL

The sample, $La_{0.5}Sr_{0.5}CoO_3$, was prepared by a solid state reaction method starting with preheated La_2O_3, CoO, and $SrCO_3$. The appropriate mixture was ground and calcined at 1000 °C for 1 day. The mixture was then ground again and heated at 1100 °C in air for 2 days with intermediate grindings. It was then pelletized and fired in air at 1300 °C for 1 day. The phase purity was checked with x rays and the sample was found to be of single phase and the diffraction pattern compared well with the reported data.

The magnetization measurements were performed using superconducting quantum interference device magnetometer (Quantum Design). The data were collected at 2 K intervals over the temperature range from 202 to 270 K, in fields from 100 to 55 kOe. The maximum deviation in the temperature was ± 0.02 K at each measuring temperature.

III. RESULTS AND DISCUSSION

The second-order magnetic phase transition near the Curie point is characterized by a set of critical exponents, β (associated with the spontaneous magnetization), γ (associated with the initial susceptibility), and δ (related to the critical magnetization isotherm). They are defined as

$$M_s(T)=M_0(-\epsilon)^{\beta}, \quad \epsilon<0, \tag{1}$$

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

$$M=A_0(H)^{1/\delta}, \quad \epsilon=0 \tag{3}$$

where $\epsilon=(T-T_C)/T_C$, T_C is the Curie temperature and M_0, h_0/M_0, and A_0 are the critical amplitudes. Our aim is to determine the critical exponents and the critical temperature from the magnetization data as a function of the field at different temperatures.

Figure 1(a) shows the M^2 vs H/M plot or the Arrot plot. In agreement with Mira *et al.*,[7] we also observe a positive slope of the M^2 vs H/M plot and we analyze our data assuming the PM-FM transition to be of second order in this compound. According to the mean-field theory near T_c, M^2 vs H/M at various temperatures should show a series of parallel lines. The line at $T=T_c$ should pass through the origin. In our case the curves in the Arrot plot are not linear. This suggests that the mean-field theory is not valid. We then tried to analyze our data according to the modified Arrot plot method, based on the Arrot-Noakes equation of state.[8] Figure 1(b) shows the modified Arrot plot, $M^{1/\beta}$ versus $(H/M)^{1/\gamma}$. The isotherms are almost parallel straight lines for $\beta=0.365$ and $\gamma=1.336$. The corresponding value of δ can be obtained from Widom scaling relation, i.e., $\delta=1+\gamma/\beta=4.66$. The high field straight line portions of the isotherms can be linearly extrapolated to obtain the spontaneous magnetization $M_s(T)$ and the inverse susceptibility $\chi_0^{-1}(T)$. The temperature variation of $M_s(T)$ and $\chi_0^{-1}(T)$, obtained from Fig. 1(b) are shown in Fig. 1(c). The continuous curves in

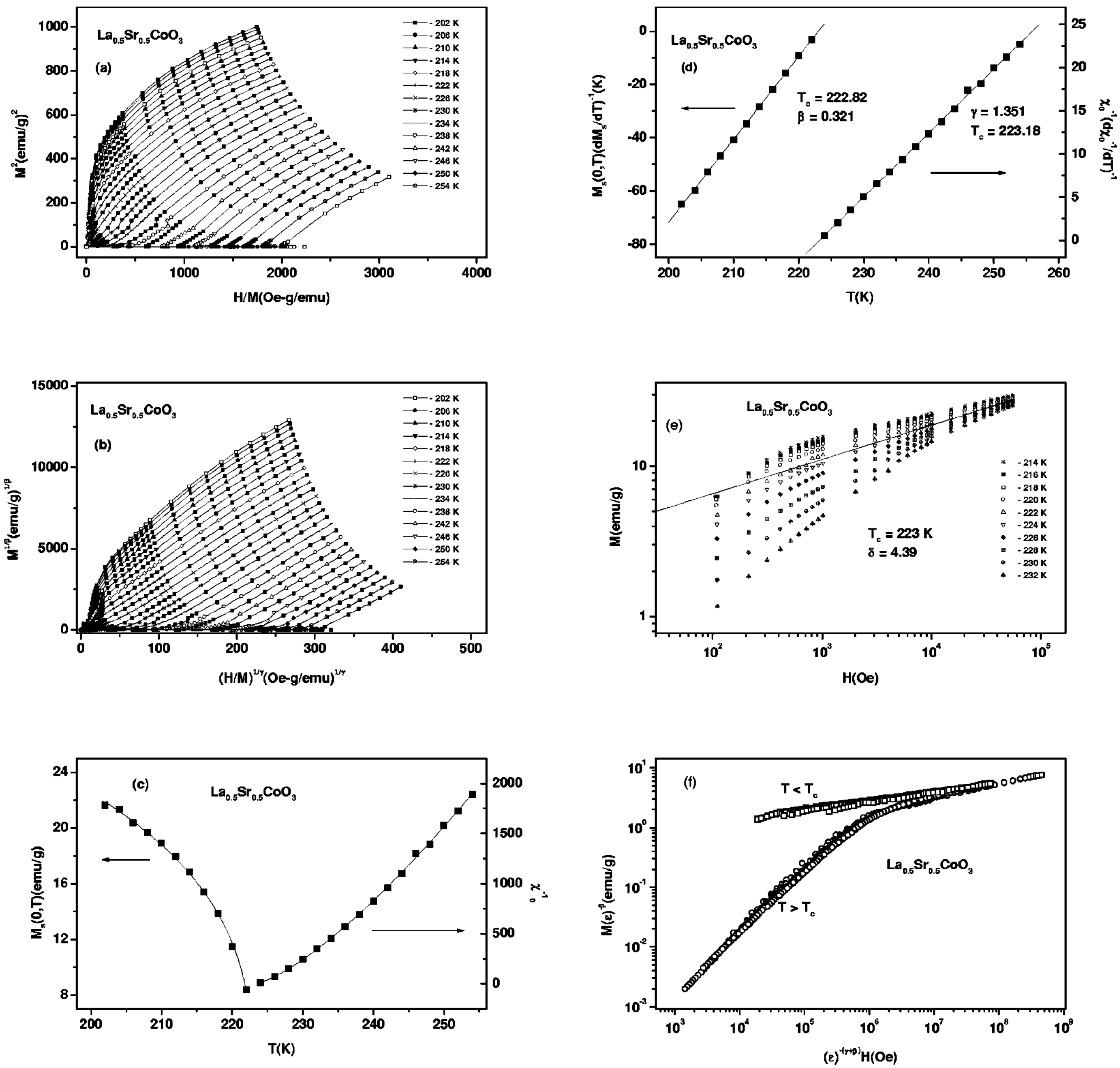

FIG. 1. (a) Isotherms of M^2 vs H/M. (b) Modified Arrot plot isotherms. (c) The temperature variation of the spontaneous magnetization along with the fit obtained with the help of the power law; the temperature variation of the inverse initial susceptibility along with the fit obtained with the help of the power law. (d) Kouvel-Fisher plot for the spontaneous magnetization; Kouvel-Fisher plot for the inverse initial susceptibility. (e) M vs H on a log scale at several temperatures close to T_c. (f) The scaling plot on a log scale.

Fig. 1(c) show the power-law fits obtained from Eqs. (1) and (2), respectively. The Kouvel-Fisher (KF) method[9] suggests that the quantities $M_s(T)[dM_s(T)/dT]^{-1}$ and $\chi_0^{-1}(T)\times[d\chi_0^{-1}(T)/dT]^{-1}$ plotted against temperature, give straight lines with slopes $(1/\beta)$ and $(1/\gamma)$, respectively, with the intercepts on the T axes that are equal to T_c/β and T_c/γ, respectively. The linear fit to the plots following the KF method [Fig. 1(d)] give β as 0.321 ± 0.002 with T_c as 222.82 K and γ as 1.351 ± 0.009 with T_c as 223.18 K. Figure 1(e) shows the M vs H plot on a log scale at few temperatures close to T_C. The straight line shows the fit for the interpolated data at $T_c=223$ K. This gives the value of δ as 4.39 ±0.02.

Next we compare our data with the prediction of the scaling theory[10]

$$M/|\epsilon|^{\beta}=f_{\pm}(H/|\epsilon|^{(\beta+\gamma)}), \tag{4}$$

where $(+)$ and $(-)$ signs are for above and below T_c, respectively. This relation further predicts that $M/|\epsilon|^{\beta}$ plotted as a function of $H/|\epsilon|^{\beta+\gamma}$ give two different curves, one for temperatures below T_c and the other for temperatures above T_c. Taking the values of β, γ obtained from the Kouvel-Fisher method with T_c equal to 223 K, the scaled data are plotted in Fig. 1(f). All the points fall on two curves, one for $T<T_c$ and the other for $T>T_c$. This suggests that the value of the exponents and T_c are reasonably accurate.

The values of the critical exponents depend on the range of the exchange interaction $J(r)$. Fisher *et al.*[11] have performed a renormalization group analysis of systems with an exchange interaction of the form $J(r)=1/r^{d+\sigma}$ (d is the dimension of the system, σ is the range of the interaction). If σ is greater than 2, then the Heisenberg exponents ($\beta=0.365$, $\gamma=1.386$, and $\delta=4.8$) are valid. The mean-field exponents ($\beta=0.5$, $\gamma=1.0$, and $\delta=3.0$) are valid for σ less than 1/2. For $1/2<\sigma<2$, the exponents belong to different universality classes which depend upon σ. A useful discussion on the critical exponents of the manganites has been given in Ref. 7. Menyuk *et al.*[6] studied the critical behavior in $La_{0.5}Sr_{0.5}CoO_3$. In $La_{0.5}Sr_{0.5}CoO_3$ the cobalt ions occupy a single cubic structure. For this case the high-temperature expansion calculation predicts $\gamma=1.4$. The calculation was carried out for a Heisenberg exchange with nearest-neighbor interactions. The experimental study gives $\gamma=1.27\pm0.02$ and $\delta=3.05\pm0.06$. Recent studies of critical exponents have been done in $La_{1-x}Sr_xCoO_3$ system in the concentration range $0.2\leqslant x\leqslant0.3$. They have analyzed their data using modified Arrot plot method. The results are $0.43\leqslant\beta\leqslant0.46$, $1.39\leqslant\gamma\leqslant1.43$ and $4.02\leqslant\delta\leqslant4.38$.[7] The results suggest that γ value is Heisenberg like and β value is mean-field like. They suggest, the difference is because the β is calculated from fittings below T_c whereas γ is from above T_c. However, they concluded that the system is better described by a Heisenberg model than by a mean-field one. This is different from the manganites. This is because of the absence of long-range order due to hole-poor regions. The reason for the high β value is predicted to be the spin transition of the Co^{+3} ions at T_c.

In our measurement, we observe $0.321\leqslant\beta\leqslant0.365$, $1.336\leqslant\gamma\leqslant1.351$, $4.39\leqslant\delta\leqslant4.66$ and $T_c\approx223$ K. We have analyzed our data using modified Arrot plot and Kouvel-Fisher method. Finally the scaling confirms that the exponents and T_c appear reasonable. These values are closer to Heisenberg values than mean-field values. Our calculated value of T_c is in good agreement with Mira *et al.*[7] for $x=0.3$. The calculated γ and δ values are close to them although β values differ. Our γ value is also consistent with the theoretical prediction of Stanley[12] based on Heisenberg exchange with nearest-neighbor interactions. The Heisenberg model does not apply to metallic conductors. However, as Mira *et al.*[7] have mentioned, $La_{0.5}Sr_{0.5}CoO_3$ is not a simple metallic ferromagnetic system. The system consists of hole-rich metallic ferromagnetic regions and a hole-poor matrix similar to $LaCoO_3$. The ferromagnetic clusters reach a percolation threshold at $x=0.2$. However, the hole-poor matrix still persists even in $x=0.5$. In this hole-poor matrix the Co^{3+} ions exist in the diamagnetic low-spin state (t_{2g}^6) as well as the high-spin state ($t_{2g}^4e_g^2$).[1] These diamagnetic ions can prevent the onset of long-range order. This may be the reason that the values of the exponents are closer to Heisenberg values rather than mean-field values. However, in contrast to Ref. 7, in this study, our value of β is also closer to the Heisenberg value. Hence we believe that our detailed analysis with the help of the modified Arrot plot, the Kouvel-Fisher method, and scaling confirms that the system is a Heisenberg one.

IV. CONCLUSION

We have studied the critical behavior of $La_{0.5}Sr_{0.5}CoO_3$ polycrystalline sample from dc magnetization measurement near T_c. We have determined the values of T_c, β, γ, δ. The values of the exponents are close to the Heisenberg values rather than mean field values. The Co ions in the low spin diamagnetic state may be the cause for preventing the long-range order.

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