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

S. Mukherjee, P. Raychaudhuri, A. K. Nigam

Tata Institute of Fundamental Research,

Homi Bhabha Road, Colaba, Mumbai - 400 005, India.
Abstract

We have studied the critical behaviour in $La_{0.5}Sr_{0.5}CoO_3$ near the paramagnetic-ferromagnetic transition temperature. We have analysed our dc magnetisation data near the transition temperature with the help of modified Arrott plots, Kouvel-Fisher method. We have determined the critical temperature $T_c$ and the critical exponents, $\beta$ and $\gamma$. With these values of $T_c$, $\beta$ and $\gamma$, 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.
I. INTRODUCTION

The parent compound $LaCoO_3$ has been studied extensively.\textsuperscript{1-4} This is a semiconductor with a conductivity gap of almost 0.6 eV and shows a decrease in susceptibility below 100 K with a broad maximum and a gradual reduction obeying Curie-Weiss law at higher temperatures. The magnetic properties of the system are explained by a progressive conversion of low-spin $Co^{III}$ into high-spin $Co^{3+}$. In the temperature range $100K < T < 350K$, the ratio of the high-spin to low-spin Co reaches 50:50 with short range ordering of low-spin and high-spin Co ions. Above 600 K it behaves metallic. Substitution of $Sr^{2+}$ for $La^{3+}$ causes a remarkable change in the system. Due to Sr doping the material segregates into hole-rich, metallic ferromagnetic regions and a hole poor matrix like $LaCoO_3$. The Co ions in the ferromagnetic phase are in intermediate-spin configurations.\textsuperscript{5} In $La_{1-x}Sr_xCoO_3$, for $x < 0.2$, the hole rich regions are isolated from each other and show superparamagnetic behaviour below $T_c \sim 240K$.\textsuperscript{5,6} These clusters freeze at a lower temperature. For $x > 0.2$, the onset of ferromagnetic transition is observed.\textsuperscript{5,7,8} Metallic ferromagnetism has been suggested for the range $0.30 \leq x \leq 0.50$.\textsuperscript{5} However,
the hole-poor matrix interpenetrating the ferromagnetic regions persists to 
\[ x = 0.5 \].

In order to fully understand the nature of the ferromagnetic transition 
we carried out the study of the critical exponents in detail associated with 
the transition. For our study we have chosen the extreme ferromagnetic 
limit \( \text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3 \). Experimental studies of critical phenomena have been 
previously made on manganese oxides \(^{10-12}\). Recently similar study has been 
done in \( \text{La}_{1-x}\text{Sr}_x\text{MnO}_3 \) system \(^{13,14}\).

II. EXPERIMENTAL

The sample, \( \text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3 \) was prepared by a solid state reaction method 
starting with preheated \( \text{La}_2\text{O}_3 \), CoO and \( \text{SrCO}_3 \). The appropriate mixture 
was ground and calcined at 1000\(^\circ\)C for 1 day. The mixture was then ground 
again and heated at 1100\(^\circ\)C in air for 2 days with intermediate grindings. It 
was then pelletized and fired in air at 1300\(^\circ\)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 magnetisation measurements were performed using SQUID magnetometer (Quantum Design). The data were collected at 2 K intervals over the temperature range from 202 K to 270 K, in fields from 100 Oe 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 characterised by a set of critical exponents, \( \beta \) (associated with the spontaneous magnetisation), \( \gamma \) (associated with the initial susceptibility) and \( \delta \) (related to the critical magnetisation isotherm). They are defined as

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

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

\[
M = A_0(H)^{1/\delta}, \epsilon = 0. \tag{3}
\]

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 magnetisation data as a function of
the field at different temperatures.

Figure 1 shows the zero field cooled (ZFC) and field cooled (FC) mag-
netisation data as a function of temperature in a field of 100 Oe. A sharp
rise in the ZFC magnetisation below 240 K is considered as the signature
of ferromagnetic ordering. Below the Curie temperature, FC magnetisation
value is different from the ZFC value. Figure 2 shows hysteresis loop at 5
K. The loop is similar to that of a ferromagnet with small coercive force and
remanence.

Figure 3 and figure 4 show the magnetisation data as a function of the
magnetic field at different temperatures. Figure 5 shows the $M^2$ vs $H/M$ plot
or the Arrot plot. 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. Then we
tried to analyse our data according to the modified Arrot plot method. This
method is based on the Arrot-Noakes equation of state.\textsuperscript{15} According to this
method, the plot of $M^{1/\beta}$ and $(H/M)^{1/\gamma}$ at several temperatures close to $T_c$
should give a series of parallel straight lines. We adopted this method and ob-
tained almost parallel straight lines with the values of $\beta$ and $\gamma$ equal to 0.365 and 1.336 respectively. Figure 6 shows the modified Arrot plot in our case. This plot shows that the isotherms are almost parallel straight lines in the high-field region. The high field straight line portions of the isotherms can be linearly extrapolated to obtain the intercepts on the $M^{1/\beta}$ and $(H/M)^{1/\gamma}$ axes. From these intercepts the spontaneous magnetisation $M_s(T)$ and the inverse susceptibility $\chi_0^{-1}(T)$ can be computed. The temperature variation of $M_s(T)$ and the same for $\chi_0^{-1}(T)$, obtained from figure 6 are shown in figure 7 and figure 8. The continuous curves in figures 7 and 8 show the power-law fit obtained from equation (1) and (3) respectively. Then we apply Kouvel-Fisher method\textsuperscript{16} to obtain $T_c$, $\beta$ and $\gamma$. The Kouvel-Fisher method suggests that the quantities $M_s(T)(dM_s(T)/dT)^{-1}$ and $\chi_0^{-1}(T)(d\chi_0^{-1}(T)/dT)^{-1}$ plotted against temperature, give straight lines with slopes $(1/\beta)$ and $(1/\gamma)$ respectively, and the intercepts on the T-axes are equal to $T_c/\beta$ and $T_c/\gamma$ respectively. From figures 7 and 8 we have computed $M_s(T)(dM_s(T)/dT)^{-1}$ and $\chi_0^{-1}(T)(d\chi_0^{-1}(T)/dT)^{-1}$. Figure 9 and figure 10 show respectively the plots of $M_s(T)(dM_s(T)/dT)^{-1}$ and $\chi_0^{-1}(T)(d\chi_0^{-1}(T)/dT)^{-1}$ vs temperature. The linear fit to the plot shown in figure 9 gives the value of $\beta$ as $0.321 \pm 0.002$ and $T_c$ as 222.82 K. The linear fit to the plot shown in figure 10 gives
the value of $\gamma$ as $1.351 \pm 0.009$ and $T_c$ as $223.18$ K. Figure 11 shows $M$ 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 $\delta$ as $4.39 \pm 0.02$.

Next we compare our data with the prediction of the scaling theory

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

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 $\beta$, $\gamma$ and $T_c$ obtained from our analysis, the scaled data are plotted in figure 12. 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 accurate enough.

We started analysing our data with the help of modified Arrott plot. Finally we got the exponent values as $\beta = 0.321 \pm 0.002$, $\gamma = 1.351 \pm 0.009$, $\delta = 4.39 \pm 0.02$. Our experimental value of $\beta$ is very close to 3D Ising value. However, the value of $\gamma$ and $\delta$ do not match very accurately. The value of the critical exponents depend on the range of the exchange interaction $J(r)$. 

According to Fisher et al.\textsuperscript{18} $J(r)$ varies as $1/r^{d+\sigma}$, where $d$ gives the dimension of the system and $\sigma$ gives a measure of the range of the interaction. If $\sigma$ is greater than 2, then the Heisenberg exponents ($\beta = 0.365$, $\gamma = 1.386$ and $\delta = 4.8$) are valid. The mean field exponents ($\beta = 0.5$, $\gamma = 1$ and $\delta = 3.0$) are valid for $\sigma$ less than half. For $1/2 < \sigma < 2$, the exponents belong to different universality class which depends upon $\sigma$. $La_{1-x}Sr_xCoO_3$ is not a very simple ferromagnetic system. It shows a large difference between the ZFC and FC magnetisation below $T_c$. The hole-poor matrix is also present in $La_{0.5}Sr_{0.5}CoO_3$.\textsuperscript{5} Hence one cannot expect the exponents to belong to any universality class.

**IV. CONCLUSION**

We have studied the the critical behaviour of $La_{0.5}Sr_{0.5}CoO_3$ polycrystalline sample from dc magnetisation measurement near $T_c$. We have determined the values of $T_c$, $\beta$, $\gamma$, $\delta$. The value of $\beta$ is very close to 3D Ising value. However, the values of $\gamma$ and $\delta$ do not completely agree with any universality class.
V. ACKNOWLEDGMENT

We thank K.V. Gopalakrishnan for his sincere help in the experiment.
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FIGURE CAPTIONS

**figure 1:** Magnetisation as a function of temperature under zero field cooled (ZFC) and field cooled (FC) conditions.

**figure 2:** Hysteresis loop drawn at 5 K.

**figure 3:** Magnetisation as a function of magnetic field at several temperatures in the range 202 K - 232 K.

**figure 4:** Magnetisation as a function of the magnetic field at several temperatures in the range 234 K - 270 K.

**figure 5:** Isotherms of $M^2$ vs H/M.

**figure 6:** Modified Arrot plot isotherms.

**figure 7:** The temperature variation of the spontaneous magnetisation along with the fit obtained with the help of the power law.

**figure 8:** The temperature variation of the inverse initial susceptibility along with the fit obtained with the help of the power law.

**figure 9:** Kouvel-Fisher plot for the spontaneous magnetisation
Figure 10:- Kouvel-Fisher plot for the inverse initial susceptibility.

Figure 11:- M vs H on a log scale at several temperatures close to $T_c$.

Figure 12:- The scaling plot on a log scale.
The plot shows the magnetic moment $M$ (emu/g) as a function of temperature $T$ (K) for La$_{0.5}$Sr$_{0.5}$CoO$_3$ under a magnetic field $H = 100$ Oe. The data is represented by two cooling curves: FC (full cooling) and ZFC (zero-field cooling).
La$_{0.5}$Sr$_{0.5}$CoO$_3$

$\gamma = 1.351$

$T_c = 223.18$
La$_{0.5}$Sr$_{0.5}$CoO$_3$

$T_c = 223$ K

$\delta = 4.39$
$M(\varepsilon)^{\beta} (\text{emu/g})$

$T < T_c$

$T > T_c$

$\text{La}_{0.5} \text{Sr}_{0.5} \text{CoO}_3$
$\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$
The graph shows the variation of $M_s(0, T)$ (emu/g) with temperature $T$ (K) for $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$. The data points form a downward trend, indicating a decrease in magnetization with increasing temperature.
$\chi^{-1}$

$\La_{0.5}\Sr_{0.5}\CoO_3$

$T(K)$
$\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$

$T_c = 222.82$

$\beta = 0.321$