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Critical behavior near the ferromagnetic to paramagnetic phase transition temperature in polycrystalline $La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(0 \leq x \leq 0.1)$

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ABSTRACT

In this paper we report on the critical analysis of the $La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(0 \leqslant x \leqslant 0.1)$ manganites near the ferromagnetic-paramagnatic phase transition temperature. Various techniques such as modified Arrott plot, Kouvel–Fisher method and critical isotherm were used to analyze the magnetic-field dependence of magnetization. The Curie temperature (T_C) could be tuned over a wide temperature range, from 251 K to 310 K, with varying in content. Though the nature of this transition is found to be of second order, the estimated critical exponents β, γ, and δ obtained for different values of x are close to the theoretically predicted values for the three-dimensional (3D)-Ising interaction model ($\beta = 0.324 \pm 0.01$, $\gamma = 1.240 \pm 0.13$ at $T_C = 310$ K for $x = 0.00$); ($\beta = 0.329 \pm 0.04$, $\gamma = 1.241 \pm 0.001$ at $T_C = 294$ K for $x = 0.05$); ($\beta = 0.332 \pm 0.01$, $\gamma = 1.250 \pm 0.04$ at $T_C = 251$ K for $x = 0.10$) and are very far away from any other known universality class. The critical isotherm M (T_C, μ_0H) gives $\delta = 5.02 \pm 0.01$ for $x = 0.00$. Thus, the scaling law $\delta = 1 + \gamma/\beta$ is fulfilled. The critical exponents obey the single scaling equation of $M(\mu_0H,\varepsilon) = \varepsilon^{\beta} f_{\pm}(\mu_0H/\varepsilon^{\beta+\gamma})$; where f_+ for $T > T_C$ and f_- for $T < T_C$.

1. Introduction

Over the last few decades, intensive research has been focused on the colossal magnetoresistance (CMR) of perovskite manganites with a general formula $Ln_{1-x}A_xMnO_3$ associated with simultaneous ferromagnetic to paramagnetic and metal to insulator transitions [1–3]. These properties were traditionally ascribed to the double exchange interaction between ferromagnetically coupled Mn^{3+} and Mn^{4+} ions and the Jahn–Teller effect [4,5]. Furthermore, to better understand the relation between insulator–metal transition and CMR effect, two important questions about PM–FM transition must be answered: the first question concerns the common universality class. The second one is the order of phase transition. Earlier studies have concentrated on the critical behavior of perovskite manganites at the region of the PM–FM transition in order to investigate their nature (first or second order) and to derive the critical exponents of magnetization (β) and magnetic susceptibility (γ). The analysis of the critical exponents in the temperature range near the magnetic phase transition is a powerful tool to examine in details the mechanisms of the magnetic interaction responsible for the transition [6–8]. An important issue when studying the critical behavior is the choice of the appropriate samples. It is well known that single crystals are ideal for that kind of study as polycrystalline samples present a strong smearing in the phase transitions, making it difficult to evaluate the critical parameters. Besides, in order to fully reveal the critical behavior, high quality crystals are needed, with good stoichiometry and as few defects as possible. The real critical behavior might be masked if these conditions are not achieved.

In earlier theoretical studies [9,10], the critical behavior in the double exchange model was first described with long-range mean-field theory. However, the recent theoretical calculations have predicted the critical exponents in manganites in agreement with the short-range exchange interaction model. Sequentially, depending on the computational technology for the CMR of manganites, Furulawa and Motome suggested that the critical behavior should be attributed to short-range Heisenberg model. Moreover, a few relevant experimental investigations on the critical phenomena also supported this viewpoint due to the obtained value of the critical exponents consistently with that in the conventional ferromagnet of 3D Heisenberg model. Ghosh et al. [11] reported that the calculated values of the critical exponent β is equal to 0.37 for $La_{0.7}Sr_{0.3}MnO_3$ (β = 0.365 in Heisenberg model). However,

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a relatively high value of $\beta = 0.5$ obtained in the polycrystalline $La_{0.8}Sr_{0.2}MnO_3$ is in good agreement with that in mean field model [12].

On the other hand, a very low critical exponent of $\beta = 0.14$ identified in the single crystal $La_{0.7}Ca_{0.3}MnO_3$ suggested that the PM-FM transition in this system is of a first rather than a second order type [13]. Therefore, in view of the varied values of the critical exponent β from 0.1 to 0.5, four kinds of different theoretical models were used to explain the critical properties in manganites. These models are mean field values, three dimensional, 3D Ising values, 3D Heisenberg interaction and tricritical mean-field model. Due to the divergence in these reported critical values, it is worthwhile to study the critical behavior in the same perovskite manganites.

In this paper, we present a detailed study of the critical phenomena, based on different techniques including the modified Arrott plot, Kouvel-Fisher method and critical isotherm analysis in $La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(0 \le x \le 0.1)$ manganites.

2. Experiment

$La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(0 \le x \le 0.1)$ compounds were prepared by sol-gel method described in our earlier study [14]. Room temperature X-ray diffraction patterns show that all samples crystallize in the rhombohedral structure with $R\bar{3}c$ space group [14]. Indeed, magnetic measurements were performed using a BS1 and BS2 magnetometer developed in Louis Neel Laboratory, Grenoble. The isotherms were corrected by a demagnetization factor D_a that was determined by a standard procedure from the low-field linear-response regime at a low temperature (H_{appl}-D_aM).

3. Results and discussion

To analyze the exact nature of the magnetic phase transition in $La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(0 \le x \le 0.1)$ compounds, we analyzed the critical behavior near T_C. It is worth-mentioning that the critical exponents, β, γ and δ correspond to the spontaneous magnetization (M_S), the inverse of initial susceptibility (χ_0^{-1}) and the critical magnetization isotherm (magnetization M versus magnetic field μ_0H at T_C), respectively. The mathematical definitions of the exponents from magnetization measurements are given in the following relations:

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

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

$$M = D(\mu_0 H)^{1/\delta}, \quad \varepsilon = 0, \quad T = T_C \tag{3}$$

Here ε is the reduced temperature ($\varepsilon = (T - T_C)/T_C$); h_0 as well as M_0 and D are critical amplitudes.

Fig.1 shows the Arrott plot M^2 vs. μ_0H/M constructed from the raw M–μ_0H isotherms after correcting the external magnetic field for demagnetization effects.

According to the mean field theory ($\beta = 0.5$, $\gamma = 1$ and $\delta = 3$) such curves should give a series of straight lines for different temperatures and the intercept of these straight lines on the μ_0H/M axis is negative/positive below/ above T_C, and the line of M^2 vs. (μ_0H/M) at T_C should pass through the origin. In our work, the curves are nonlinear and show up ward curvature even the high field indicating that the mean field theory cannot be used to describe the critical behavior in the present system. Consequently, the reliability of the critical exponents and the Curie temperature T_C were determined from the modified Arrott plots (MAP) (also called Arrott-Noakes plots). In this technique, the $M = f(\mu_0H)$ data

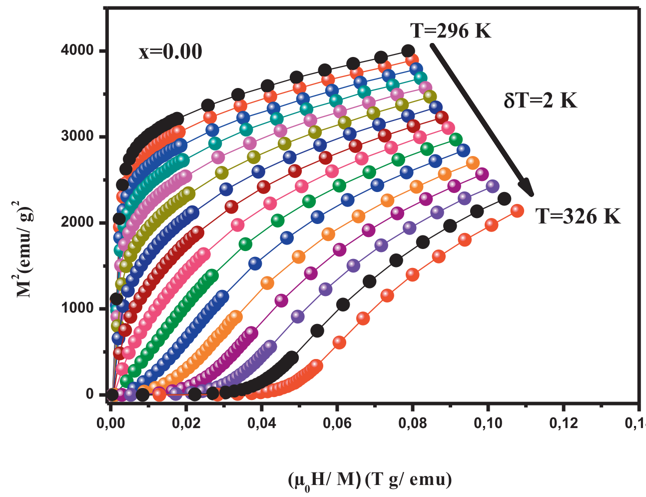

Fig. 1. Standard Arrott plot (isotherms M^2 vs. μ_0H/M).

are converted into series of isotherms ($M^{1/\beta}$ vs $(\mu_0H/M)^{1/\gamma}$) depending on the following relation [15]:

$$(\mu_0 H/M)^{1/\gamma} = (T - T_C)/T_1 + (M/M_1)^{1/\beta} \tag{4}$$

where β and γ are the critical exponents and M is a material constant.

Fig.2 shows the modified Arrott plots (MAP) based on the Arrott–Noakes equation of state Eq. (4), at different temperatures by using four models of critical exponents for $La_{0.5}Sm_{0.1}Sr_{0.4}Mn_{1-x}In_xO_3$ $(x = 0)$ compound: Fig.2(a): 3D Ising model ($\beta = 0.325$, $\gamma = 1.240$), Fig.2(b): 3D Heisenberg model ($\beta = 0.365$, $\gamma = 1.336$) and Fig.2(c): Tricritical mean field ($\beta = 0.25$, $\gamma = 1$). One can see that all models yield quasi straight and nearly parallel lines in the high field region, so it becomes difficult to decide which model is the most appropriate to determine the critical exponents. Thus, to select the model that better describes this system, we calculated the so called relative slope (RS) defined at the critical point as $RS = S(T)/S(T_C)$.

The most satisfactory model should be the one with the closest RS to 1 (unity) [16].

Fig. 2(d) shows the RS vs T curve for $La_{0.5}Sm_{0.1}Sr_{0.4}MnO_3$ sample for the four models, mean field model, 3D Heisenberg, 3D Ising and tricritical mean field model. It's clear from this figure that the 3D-Ising model is the best model which can describe our system and for the determination of the critical exponents for $x = 0.0$ sample. Based on these isotherms, the spontaneous magnetization $M_S(T, 0)$ and the inverse susceptibility $\chi_0^{-1}(T)$ data can be obtained from the linear extrapolation in the high field region to the coordinate axes $M^{1/\beta}$ and $(\mu_0H)^{1/\gamma}$, respectively. In Fig. 3 we have plotted the temperature dependence of $M_S(T, 0)$ and $\chi_0^{-1}(T)$ for the sample with $x = 0.0$. Similarly we can also obtain $M_S(T, 0)$ vs. T and $\chi_0^{-1}(T)$ vs. T for samples with $x = 0.05$ and 0.10 (not shown here). By fitting these plots with Eqs. (1) and (2), new values of β, γ and T_C will be obtained. The best fits give the values ($\beta = 0.324 \pm 0.01$, $T_C = 314.97 \pm 0.36$ K) and ($\gamma = 1.240 \pm 0.13$, $T_C = 311.75 \pm 0.75$ K) for $La_{0.5}Sm_{0.1}Sr_{0.4}MnO_3$ sample. Alternatively, the values of β, γ as well as T_C are also obtained by Kouvel–Fisher (KF) method [17].

$$M_S(T)[dM_S(T)/dT\,]^{-1} = (T - T_C)/\beta \tag{5}$$

$$\chi_0^{-1}(T)[d\chi_0^{-1}(T)/dT\,]^{-1} = (T - T_C)/\gamma \tag{6}$$

Under this method we plotted $\frac{M_S(T)}{dM_S(T)/dT}$ vs. T and $\frac{\chi_0^{-1}(T)}{d\chi_0^{-1}(T)/dT}$ vs. T which should yield straight lines with slopes $1/\beta$ and $1/\gamma$, respectively. When extrapolated to the ordinate equal to zero, these