Pressure Induced Critical Behavior of Ferromagnetic Phase Transition in Sm-Nd-Sr Manganites

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We report on the hydrostatic pressure dependence of the order of ferromagnetic (FM) to paramagnetic (PM) phase transition in a $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ single crystal. At ambient pressure, the system undergoes a first-order FM-PM phase transition at 146 K. The application of pressure increases the T_C, suppresses the hysteresis width, and thus makes the transition second order. We have analyzed the critical behavior associated with the second-order FM-PM transition in the presence of an external pressure (12.1 kbar) and obtained the critical exponents $\beta = 0.358$, $\gamma = 1.297$, and $\delta = 4.536$, which are close to those predicted for the three-dimensional Heisenberg system. Using these values of β, γ, and T_C (~176 K), one can scale the magnetization data below and above T_C following a single equation of state.

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The prototypical $Sm_{1-x}Sr_xMnO_3$ has been extensively studied in order to understand the role of disorder on the nature of the ferromagnetic (FM) to paramagnetic (PM) phase transition in narrow band manganites [1–11]. As the ionic radius of Sm and Sr differs significantly, the value of quenched disorder or local structural disorder is quite large in this system [12]. For such a system with large structural disorder, the formation energy for lattice polarons is considerably lowered, and when these polarons form in the FM state the ferromagnetism is truncated, rendering the transition first order with a reduced Curie temperature (T_C) [6–13]. Several studies have shown that both the magnetic and structural properties of $Sm_{1-x}Sr_xMnO_3$ are extremely sensitive to the applied pressure (P) and magnetic field (H) [2,5–11]. T_C increases while the discontinuity and the width of the thermal hysteresis in magnetization decrease rapidly with increasing P. Above a critical value of P, the hysteresis disappears completely and the transition becomes second order [7,10]. Similarly, with the application of H, the nature of T dependence of lattice strain changes dramatically and the discontinuity in lattice parameters at T_C which is the signature of a first-order phase transition disappears above a critical magnetic field [2,5]. It has also been observed that $^{16}O \rightarrow {}^{18}O$ isotope substitution reduces the T_C and changes the FM metallic phase to the antiferromagnetic insulating state [3]. Such a strong dependence of magnetic properties on P or isotope substitution and structural properties on H clearly indicate that magnetic and structural order parameters are strongly coupled in this system and there exists a critical end point of the first order FM transition.

To confirm that the transition becomes conventional second order above the critical point, one has to show that the system obeys the critical behavior belonging to some universality class, the most important feature of a second-order phase transition. For a true second-order phase transition the critical exponents are independent of the microscopic details of a system due to the divergence of correlation length and correlation time close to the transition point and hence their values are almost same for a transition that may occur in different physical systems [14]. Though there are several studies on the critical behavior in manganites, they are restricted to those systems which show conventional second-order FM transition at the ambient condition [15]. In this Letter, we present the first detailed critical behavior of a system, $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$, in which FM to PM phase transition is first order and the transition becomes second order only under the influence of external pressure. The critical exponents β, γ, and δ, associated with the second-order transition have been determined. Using these exponents it has also been verified that the scaling hypothesis is perfectly obeyed. To the best of our knowledge, this is the first experimental scaling analysis in manganites in presence of an external perturbation.

The single crystals of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ have been prepared by floating zone technique under oxygen atmosphere [11]. The magnetization measurements were performed by using a SQUID magnetometer (MPMS, Quantum Design) in fields up to 5 T and hydrostatic pressure up to 12.1 kbar. The data were collected at 2 to 4 K interval after stabilizing the temperature for about 30 min. External magnetic field was applied along the longest sample direction and data were corrected for the demagnetization effect. We have selected the Nd-doped compound because the FM transition in this system

changes from first order to second order below the maximum available pressure in our magnetic measurement setup in SQUID magnetometer.

Figure 1(a) shows a series of isotherms of magnetization $M(H)$ of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ single crystal in the vicinity of T_C ($\approx$146 K). For $T < T_C$, the data are typical of a ferromagnet. Initially, M increases rapidly with field and then reaches almost to saturation and its magnitude gradually decreases with increasing temperature. But above T_C, there is a significant change in the nature of $M(H)$ curve, particularly in the low-field region, where H dependence of M changes from sublinear to superlinear. M increases superlinearly up to a critical field followed by a rapid increase, until it reaches saturation. This kind of S-shaped $M(H)$ curve along with the hysteresis [inset of Fig. 1(a)] between increasing and decreasing field is the signature of a first-order metamagnetic transition. The S-shaped behavior in $M(H)$ is much stronger in $Sm_{0.52}Sr_{0.48}MnO_3$ and has been attributed to the formation of inhomogeneous metastable state in the presence of quenched disorder [4,6,8]. The partial substitution of Nd at Sm site reduces the magnitude of quenched disorder and hence the steplike behavior in $M(H)$ gets weakened. The occurrence of metamagnetic transition in this system may be understood by taking into account the field dependence of the differential susceptibility (dM/dH). The peak in dM/dH vs H curve above T_C is the clear signature of field-induced magnetic transition from PM to FM state [upper inset of Fig. 1(b)]. The resulting phase diagram, H-T curve, is shown in the lower-inset of Fig. 1(b). The critical magnetic field of PM-FM transition increases with increasing temperature at the rate of 0.065 T/K, a characteristic of a first-order FM transition. The first-order phase transition line, along which two thermodynamic phases coexist, is described by the Clausius-Clapeyron equation. Using this equation, the magnetic entropy change (ΔS_{mag}) associated with FM transition can be estimated as

$$\Delta S_{mag} = \Delta M/(dT_C/dH). \quad (1)$$

To calculate ΔS_{mag}, we take $dT_C/dH = 15.4$ K/T from H-T phase diagram and ΔM as the spontaneous magnetization at $T = 143$ K and obtain $\Delta S_{mag} = 0.13$ J/mol K [6]. The order of magnetic phase transition can also be determined from the slope of Arrott plots (M^2 vs H/M) [16]. According to the Banerjee criterion, the slope of M^2 vs H/M curve is negative for a first-order magnetic transition [17] and the slope is positive for the conventional second-order transition. The finite amount of magnetic entropy change at T_C and the negative slope in M^2 vs H/M plot [Fig. 1(b)] in the low-field regime imply that FM to PM transition is first order.

Figure 2 shows $M(T)$ for different P. At $P = 0$, M shows a drop at T_C along with thermal hysteresis. The thermal hysteresis has also been observed in resistivity and thermopower [11]. The application of P increases M and shifts the T_C towards the higher temperature at the rate of $\sim$2.5 K/kbar [inset of Fig. 2]. The width of the thermal hysteresis (ΔT) at $P = 0$ is about 1.5 K, which narrows progressively with increasing P and completely disappears at a critical pressure ($\leq$10 kbar) [inset of Fig. 2]. These observations suggest that the first-order nature of the FM

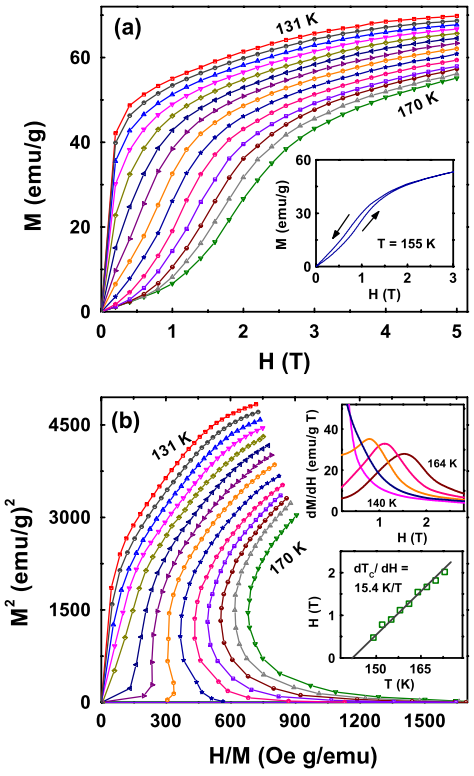

FIG. 1 (color online). (a) Magnetization (M) of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ single crystal as a function of magnetic field (H) between 131 K (top) and 170 K (bottom) in 3 K interval (data with increasing field are shown). Inset shows the hysteresis in $M(H)$ at $T = 155$ K. (b) M^2 vs H/M plot of the above isotherms. Insets show the H dependence of differential susceptibility (dM/dH) for 140 K $\leq T \leq$ 164 K in 6 K interval and H-T phase diagram.

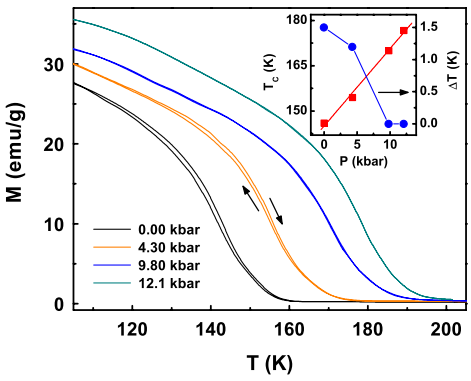

FIG. 2 (color online). Temperature dependence of magnetization (M) of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ single crystal for different pressures (P). Inset shows the P dependence of T_C and the thermal hysteresis width (ΔT).

transition in $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ ends at a critical pressure.

To confirm whether the transition becomes conventional second order above the critical pressure, we have measured a set of $M(H)$ isotherms at $P = 12.1$ kbar, as shown in Fig. 3(a). Unlike in Fig. 1(a), the $M(H)$ curves at 12.1 kbar show a gradual transition between ferromagnetism and paramagnetism with $T_C \approx 176$ K and no hysteresis between the increasing and decreasing field has been observed. Thus, the metamagnetic transition is absent and the magnetization isotherms become conventional Brillouin-like, with a slope that decreases continuously with increasing field as reflected from the H dependence of dM/dH for various T around T_C [inset of Fig. 3(a)]. The magnetization isotherms at $P = 12.1$ kbar are shown as Arrott plots in Fig. 3(b) [16]. It is clear from the figure that the slopes of M^2 vs H/M are positive over the entire range, confirming the second-order nature of the transition [17]. The second-order magnetic phase transition is characterized by a set of critical exponents $-\beta$, γ, and δ [14]. They are defined as

$$M_S(0, T) = M_0(-\varepsilon)^{\beta}, \qquad \varepsilon < 0, \tag{2}$$

$$\chi_0^{-1}(0, T) = (h_0/M_0)(\varepsilon)^{\gamma}, \qquad \varepsilon > 0, \tag{3}$$

$$M(H, T_C) = D(H)^{1/\delta}, \qquad \varepsilon = 0, \tag{4}$$

where M_S, χ_0 and $\varepsilon = (T - T_C)/T_C$ are the spontaneous magnetization, initial susceptibility and reduced temperature, respectively, and M_0, h_0/M_0, and D are the critical amplitudes. The values of T_C, M_S and χ_0^{-1} can be determined from the Arrott plot, which shows a set of parallel curves for different T. The curve for $T = T_C$ should pass through the origin. Moreover, the intersections of these curves for $T < T_C$ on M^2 axis and for $T > T_C$ on H/M axis give the values of M_S and χ_0^{-1}, respectively. To determine M_S and χ_0^{-1}, we have done a polynomial fitting to the M^2 vs H/M data and then extrapolate to $H = 0$. The temperature dependence of M_S and χ_0^{-1} are shown in Fig. 4(a). As T approaches towards T_C, both M_S and χ_0^{-1} go towards zero according to Eqs. (2) and (3), respectively. In order to obtain the precise values of critical exponents (β and γ) and T_C, we have used the Kouvel-Fisher method [18]. In this method, $M_S(dM_S/dT)^{-1}$ and $\chi_0^{-1}(d\chi_0^{-1}/dT)^{-1}$ plotted against T should be straight lines with slopes $1/\beta$ and $1/\gamma$, respectively. These straight lines intercept the T axis at T_C. The linear fit to the plots [Fig. 4(b)] yield β as 0.358 with $T_C = 176.16$ K and γ as 1.297 with $T_C = 176.56$ K. Another critical exponent δ is determined from log$M$ vs log$H$ at T_C. According to Equ. (4), this plot should yield a straight line with a slope $1/\delta$. For this purpose, we have used the M-H curve at $T = 176$ K, the nearest one to the critical isotherm and the linear fit gives $\delta = 4.536$ [inset of Fig. 4(c)]. Comparing these critical exponents with the standard models, it is clear

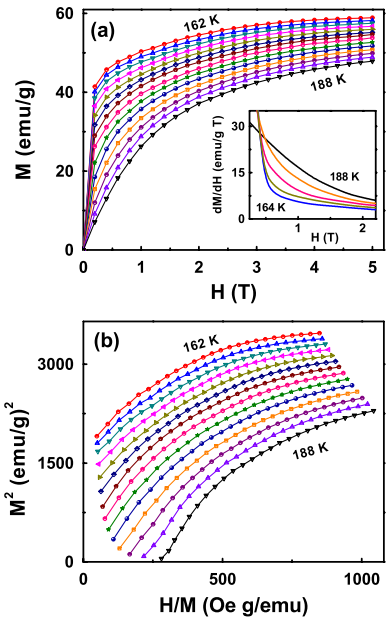

FIG. 3 (color online). (a) M vs H of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ single crystal at $P = 12.1$ kbar between 162 K (top) and 188 K (bottom) in 2 K interval. Inset shows the H dependence of (dM/dH) for 164 K $\leq T \leq$ 188 K in 6 K interval. (b) M^2 vs H/M plot of the above isotherms.

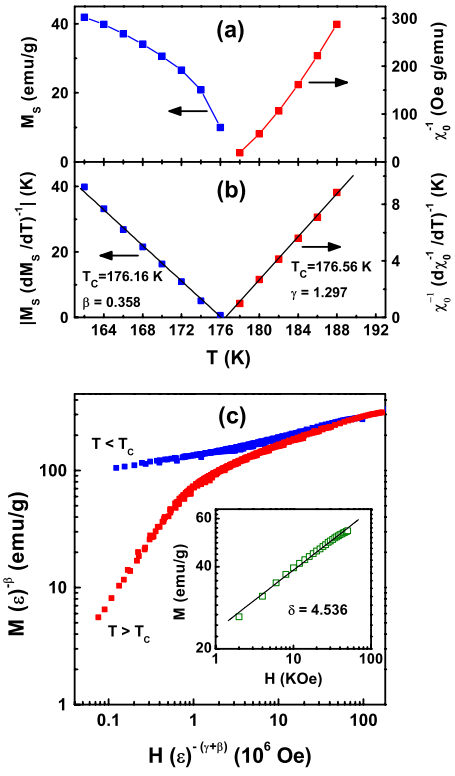

FIG. 4 (color online). (a) Temperature dependence of spontaneous magnetization (M_S) and inverse initial susceptibility (χ_0^{-1}) of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ at $P = 12.1$ kbar. (b) Kouvel-Fisher plots of M_S and χ_0^{-1}. (c) Scaling plots on a log-log scale, indicating two universal curves below and above T_C. Inset shows log-log plot of M vs H at $T = T_C$.

that the values of exponents match well with the 3D Heisenberg system [14]. This implies that the system becomes conventional isotropic ferromagnet belonging to the Heisenberg universality class under high pressure. As a further test, the scaling hypothesis predicts that $M(H, \varepsilon)$ is a universal function of H and ε,

$$M(H, \varepsilon) = (\varepsilon)^{\beta} f_{\pm}[H/\varepsilon^{(\gamma+\beta)}], \quad (5)$$

where f_+ is for $T > T_C$ and f_- is for $T < T_C$ [14]. By plotting $M(\varepsilon)^{-\beta}$ vs $H(\varepsilon)^{-(\gamma+\beta)}$, all the data points for $T > T_C$ are expected to fall on f_+, whereas the data points for $T < T_C$ will be on f_-. The main panel of Fig. 4(c) displays such a scaling plot on a log-log scale. It shows that the data points over the entire range of the variable fall on two branches of the curve depending on the sign of ε. This suggests that the values of critical exponents and T_C are reasonably accurate. Finally, one can see that the Widom scaling relation, $\gamma - \beta(\delta - 1) = 0$ (theory), 0.031 (experiment) is also satisfied [14].

It is thus clear that $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$ exhibits a first-order FM to PM phase transition at ambient condition. The application of hydrostatic pressure increases T_C and makes the transition second order with critical exponents belonging to the Heisenberg universality class. The change in the nature of FM transition with external pressure may be explained on the basis of strong coupling between magnetic order parameter and lattice strain. The free energy in presence of coupling between magnetization and strain can be written as [19]

$$f = f_0 + AM^2 + \sum A_i e_i M^2 + BM^4 + \sum B_i e_i M^4 + CM^6 + 1/2 \sum C_{ij} e_i e_j + \sum e_i P_i, \quad (6)$$

where the e_i are usual elastic strains, the C_{ij} are elastic constants and A_i, B_i are the coupling strength. Using equilibrium conditions ($\partial f/\partial M = 0$ and $\partial f/\partial e_i = 0$) and the stability criterion ($\partial^2 f/\partial M^2 > 0$), one can show that the width of thermal hysteresis decreases with increasing P and disappears above a critical pressure where the first-order FM transition becomes second order [19]. This picture is also consistent with the polaronic model. When the electrical conduction is solely governed by the polaronic hopping, the resistivity (ρ) just below T_C is related to M through a phenomenological relation $\rho(H, T) = \rho_m \exp[-M(H, T)/M_0]$, where ρ_m and M_0 are constants [20]. In a previous study, we have shown that this $\rho - M$ correlation exist in this system [11]. So the polaronic hopping becomes the prevalent conduction mechanism. To understand the role of P on the formation of polaron, we have determined the activation energy (E_g) from the temperature dependence of resistivity [$\rho \sim \exp(E_g/\mathrm{kT})$] above T_C. At ambient pressure, the value of E_g is 87 meV and it decreases monotonically with increasing P [11]. This behavior indicates that pressure suppresses the polaronic state, increases the bandwidth of the system, and as a consequence first-order FM-PM transition becomes second order.

In conclusion, we have made a comprehensive study of critical phenomena in presence of external pressure in a single crystal of $(Sm_{0.7}Nd_{0.3})_{0.52}Sr_{0.48}MnO_3$. At ambient condition, FM to PM phase transition is first order due to the strong coupling between magnetic and structural order parameter. With increasing pressure T_C of the system increases and the first-order character of the transition is suppressed and eventually vanishes at a critical pressure. Above the critical point, the transition becomes conventional second order with critical point exponents $\beta = 0.358$, $\gamma = 1.297$, and $\delta = 4.536$, belonging to the Heisenberg universality class.

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