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Crossover from first-to-second-order ferromagnetic transition in narrowband manganite $\text{Sm}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x = 0.48$)

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ABSTRACT

We have studied the nature of ferromagnetic (FM) to paramagnetic phase transition in $\text{Sm}_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ single crystal. At ambient condition, the transition is strongly first-order in nature. External perturbations like magnetic field (H) and pressure (P), and internal perturbation, such as Nd doping (y) at the Sm site of $\text{Sm}_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ change the first-order FM transition to second-order or the transition becomes a crossover at the critical points. We have identified the location of three such critical points expressed in terms of H , P , and y and are given by ($H_{\text{cr}} \approx 4$ T, $T_{\text{cr}} \approx 160$ K), ($P_{\text{cr}} \approx 2.5$ GPa, $T_{\text{cr}} \approx 160$ K), and ($y_{\text{cr}} \approx 0.4$, $T_{\text{cr}} \approx 175$ K).

KEYWORDS : Phase transition, Critical point, Manganite

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INTRODUCTION

Unusual magneto electronic properties as well as the rich phase diagram in narrowband manganites $RE_{1-x}AE_xMnO_3$ (RE : rare earth ions, AE : alkaline earth ions) are the manifestations of strong interplay between spin, charge, orbital, and lattice degrees of freedom^{1,2}. Several studies on narrowband manganites have revealed that the physical properties of these systems are strongly affected by the presence of quenched disorder (QD), arising mainly due to the size mismatch between RE and AE cations^{2,3}. Thus, one can introduce a large QD in the system by selecting the rare earth element of smaller ionic radius such as Sm, Eu, Gd, and the alkaline earth element of larger ionic radius, such as Sr and Ba. Among these kinds of systems, $Sm_{1-x}Sr_xMnO_3$ may be considered as a narrowband manganite with large QD.

In this paper, we have extensively studied on $Sm_{0.52}Sr_{0.48}MnO_3$ (SSMO) single crystal to understand the nature of magnetic and electronic phase transition both in the ambient condition as well as in presence of external and internal perturbations. The reason why we have chosen the doping level of $x = 0.48$ is that near the half-doping $x = 0.5$, the phase competition and the resultant phase variation are most clearly seen, but that the exact $x = 0.5$ crystals are known to show the special stability of the charge ordering/ orbital ordering phase unique to the commensuration effect⁴.

MATERIALS AND METHODS

The single crystals of $(Sm_{1-y}Nd_y)_{0.52}Sr_{0.48}MnO_3$ with $y = 0, 0.05, 0.1, 0.2,$ and 0.3 have been prepared by floating zone technique⁵. The quality of the crystals were carefully checked by various techniques such as x-ray diffraction, Laue diffraction, ac susceptibility, etc. The magnetization measurements were done in a superconducting quantum interference device magnetometer in fields up to 7 T and in a vibrating sample magnetometer up to 11.5. Resistivity measurements were performed by a conventional four-probe method. Specific-heat measurements were performed using semiadiabatic techniques in a wide range of temperature (5–300 K) and magnetic field (0–10 T). Thermoelectric power measurements were carried out by a differential technique.

RESULTS AND DISCUSSION

Figure 1 (a) shows the temperature (T) dependence of resistivity (ρ) of SSMO single crystal for different applied magnetic fields (H). For $H = 0$, ρ increases exponentially upon cooling from high temperature and then it suddenly drops as much as 3 orders of magnitude at the metal-insulator transition (MIT) along with thermal hysteresis.

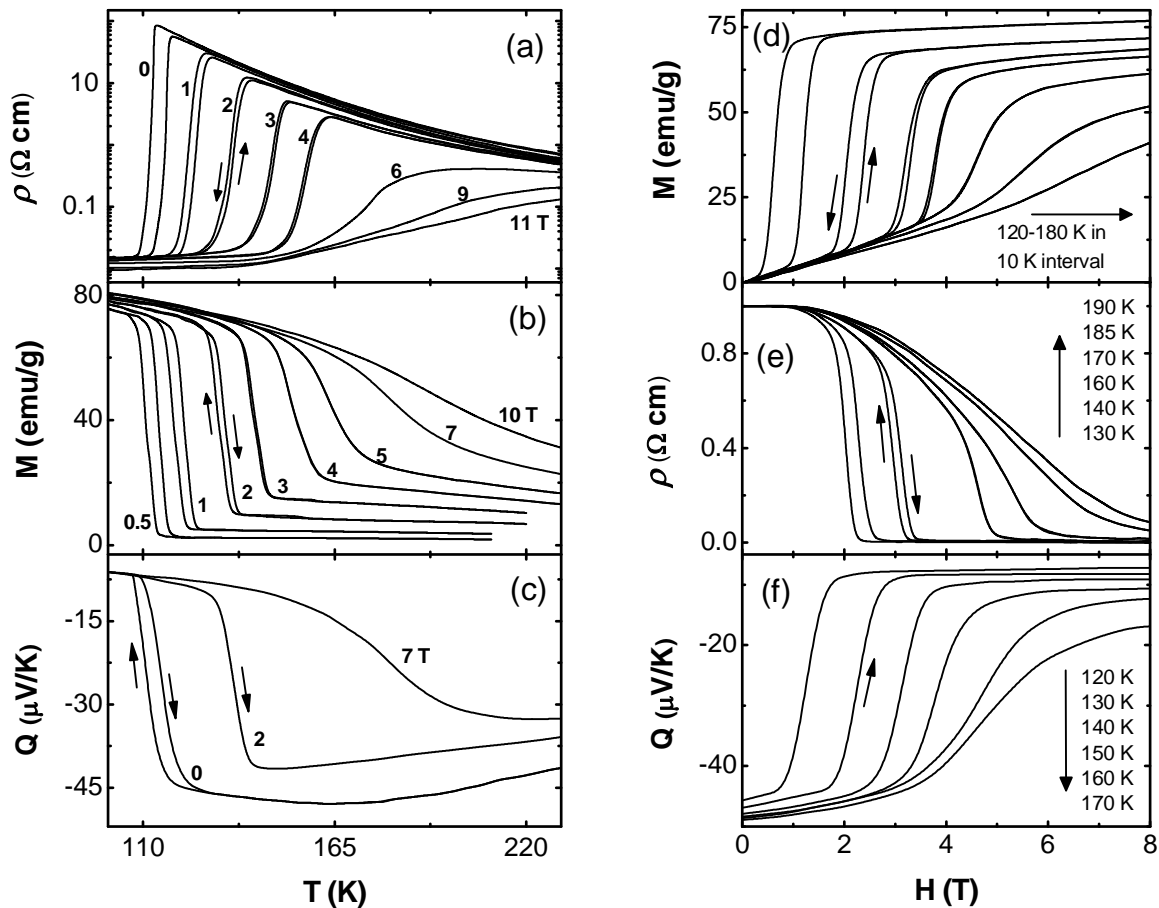


Figure. 1 - Temperature (T) dependence of (a) resistivity (ρ), (b) magnetization (M), and (c) thermoelectric power (Q) of $\text{Sm}_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ (SSMO) single crystal for different magnetic fields (H) measured both in heating and cooling cycles. Magnetic field dependence (both increasing and decreasing) of (d) M , (e) ρ , and (f) Q for different temperatures.

With increasing H , resistivity substantially decreases, MIT shifts toward the higher temperature, and the width of the hysteresis narrows down progressively. As MIT in manganites is accompanied by ferromagnetic (FM) to paramagnetic (PM) phase transition, we have also investigated the temperature dependence of magnetization (M) for different H as shown in Figure 1 (b). Similar to ρ , M also shows strong hysteresis which diminishes with increasing H . Additionally, we have measured the temperature dependence of thermoelectric power (Q) for different H [Figure 1 (c)]. At $H = 0$, Q increases sharply just below the Curie temperature (T_C) and the sharpness decreases at high fields as in the case of magnetization. The sharp change in ρ , M and Q at T_C and the presence of thermal hysteresis indicate that the FM to PM transition in SSMO is first order in nature in the low-field regime, which gets weakened at high fields, as clearly reflected by the suppression of the discontinuity in ρ , M , and Q at the transition and reduction in the hysteresis width. Above a critical magnetic field $H_{cr} \approx 4$ T, the hysteresis completely vanishes and the first-order transition becomes a crossover. Figure 1 (d) shows a series of isotherms of magnetization for some selected temperatures.

Initially, M increases linearly with H up to a transition field above which it suffers a step-like jump. Such a step-like jump in M and the hysteresis between increasing and decreasing field are the manifestation of first-order PM-FM phase. With increasing temperature, the sharpness of the jump as well as the width of the hysteresis in $M(H)$ decrease. In Figure 1 (e), the magnetoresistance, $\rho(H)/\rho(0)$ for SSMO crystal is also shown. It is clear from the figure that at low fields, the change in magnetoresistance is steep along with hysteresis. Similar to ρ and M , Q also shows a field-induced step-like jump which disappears at high temperatures [as shown in Figure 1 (f)]. So it is clear that the first-order character of the field-driven transition is weakened with increasing temperature and the hysteresis vanishes above a critical temperature, $T_{cr} \approx 160$ K. We define (H_{cr}, T_{cr}) as a critical point above which the first-order FM transition becomes a crossover, because the transition exists only in the zero-field limit and becomes a crossover in the presence of an external magnetic field.

Due to the coexistence of several phases in manganites, it is difficult to analyze the nature of phase transition from magnetic measurement only. In order to confirm the nature of the magnetic phase transition, we have measured the temperature dependence of specific heat (C_p) of SSMO crystal for different H [inset of Figure 2 (a)]. C_p exhibits strong anomaly near T_C both in the presence and absence of magnetic field. To estimate the magnetic contribution in C_p , C_{mag} , a polynomial fit from 40 to 300 K [excluding the region $(T_C - 30) < T < (T_C + 30)$] was subtracted.

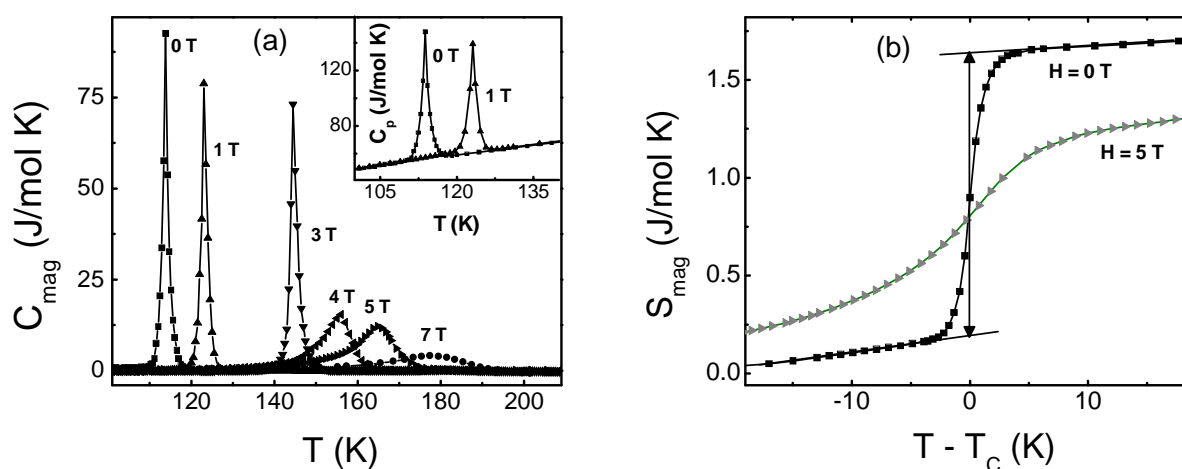


Figure. 2 - (a) Molar specific heat after subtracting the background, C_{mag} as a function of temperature. Inset shows T dependence of specific heat (C_p) of SSMO single crystal for different H (heating cycle). (b) Magnetic entropy S_{mag} calculated by integrating C_{mag}/T around T_C for $H = 0$ and $5T$.

Figure 2 (a) shows the thermal distribution of C_{mag} for different magnetic fields. For $H = 0$, C_{mag} shows a large, narrow, and symmetric peak at T_C . With increasing H , the sharpness of the peak remains almost unaltered for $H < 4$ T, but above that the sharpness decreases and the peak becomes λ -like. The shape and field dependence of C_{mag} indicate that $H = 4$ T is the boundary between the region where the FM transition is first order ($H < 4$ T) and the region where the transition becomes a

crossover ($H > 4$ T). The order of the magnetic phase transition can also be understood from the behavior of the magnetic entropy (S_{mag}) at the transition point. For a first-order transition one expects a discontinuous jump in entropy. For this purpose, we have calculated S_{mag} by integrating C_{mag}/T over temperature. In Figure 2 (b), S_{mag} is plotted as a function of T within a temperature interval of 20 K around T_C . The discontinuous and smooth changes of S_{mag} around T_C for $H = 0$ and 5 T, respectively, indicate that the first-order FM becomes a crossover with increasing magnetic field.

The magnetic phase diagram of SSMO in terms of the evolution of T_C as a function of external H is shown in Figure 3. With the decrease of H , the phase transition curve splits into two lines for $H < 4$ T: one of them corresponds to the FM to PM phase transition, which occurs on heating, while the other corresponds to the inverse transition from PM to FM phase that occurs on cooling. In between the region, the magnetic state of the system is determined by the way through which the sample arrives at this region, i.e., by the increase or decrease of temperature. This confirms that the system has a critical end-point ($H_{cr} \approx 4$ T, $T_{cr} \approx 160$ K) at which first-order FM transition becomes a crossover.

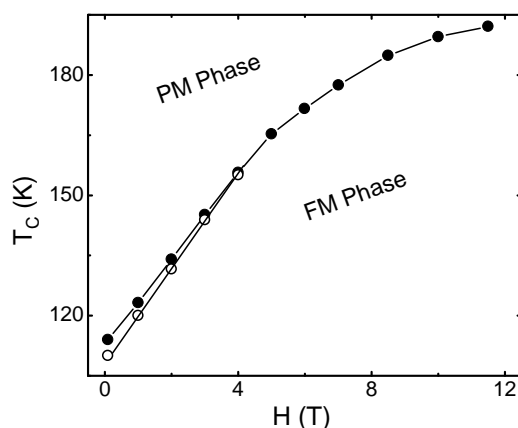


Figure. 3 - $H - T$ phase diagram of SSMO single crystal. Closed and open symbols correspond to the transition temperatures in the heating and cooling cycles, respectively.

Similar to magnetic field, we have also studied the effect of external hydrostatic pressure (P) on the nature of FM to PM phase transition in SSMO.

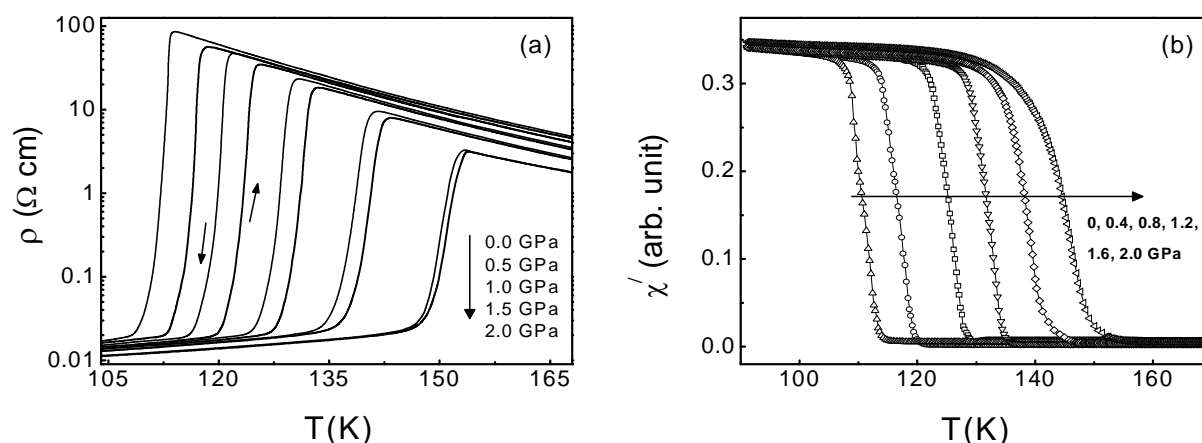


Figure. 4 - Temperature dependence of (a) resistivity (both heating and cooling cycles) and (b) real part of ac susceptibility (χ') (cooling cycle) of SSMO crystal for different hydrostatic pressures.

Figure 4 (a) shows the temperature dependence of the resistivity of SSMO crystal at different applied pressures up to 2 GPa. The MIT is quite sharp for $P = 0$ and the sharpness does not decrease appreciably up to the highest pressure 2 GPa. Similar to resistivity, the real part of ac susceptibility (χ') also shows a sharp drop at T_C as shown in Figure 4 (b). With increasing P , resistivity decreases, T_C shifts toward the higher temperature at the rate of 19 K/GPa (as shown in Figure 5), and the width of the thermal hysteresis (ΔT) gradually decreases (1.7 K/GPa). If both T_C and ΔT continue to change with P in the same fashion then the hysteresis in $\rho(T)$ will disappear above a critical pressure $P_{cr} \approx 2.5$ GPa and the corresponding value of T_{cr} would be ~ 160 K (see Figure 5). This implies that the nature of FM transition in SSMO changes from first- to second-order at a critical point ($P_{cr} \approx 2.5$ GPa, $T_{cr} \approx 160$ K).

Beside external perturbations, we have also investigated the effect of internal perturbation, originated due to the substitution of Nd at SSMO [$(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$] on the nature of FM to PM phase transition. Figure 6 (a) shows the T dependence of ρ of $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ single crystals for different y . All $\rho(T)$ curves exhibit MIT at different temperatures similar to that observed in case of T dependence of ρ at different H and P . With increasing y , resistivity decreases and T_C (taken as the temperature at which ρ just start to increase) increases almost linearly [see Figure 6 (b)].

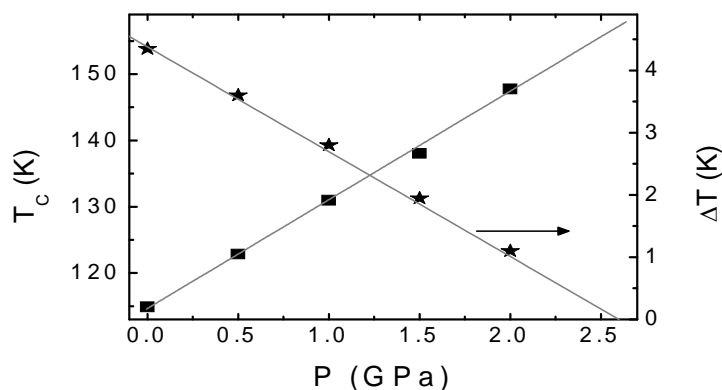


Figure. 5 - Hydrostatic pressure (P) dependence of T_C and thermal hysteresis width (ΔT) of SSMO single crystal.

One can also observe that the thermal hysteresis width, ΔT , becomes narrower and the discontinuity in $\rho(T)$ at T_C diminishes with increasing y . If both T_C and ΔT continue to change in the same fashion then the hysteresis would vanish just above a critical concentration, $y_{cr} = 0.4$ and the corresponding T_{cr} would be ~ 175 K as shown in Figure 6 (b). Thus the first-order FM transition in SSMO switches over to second-order at a critical point ($y_{cr} \approx 0.4$, $T_{cr} \approx 175$ K).

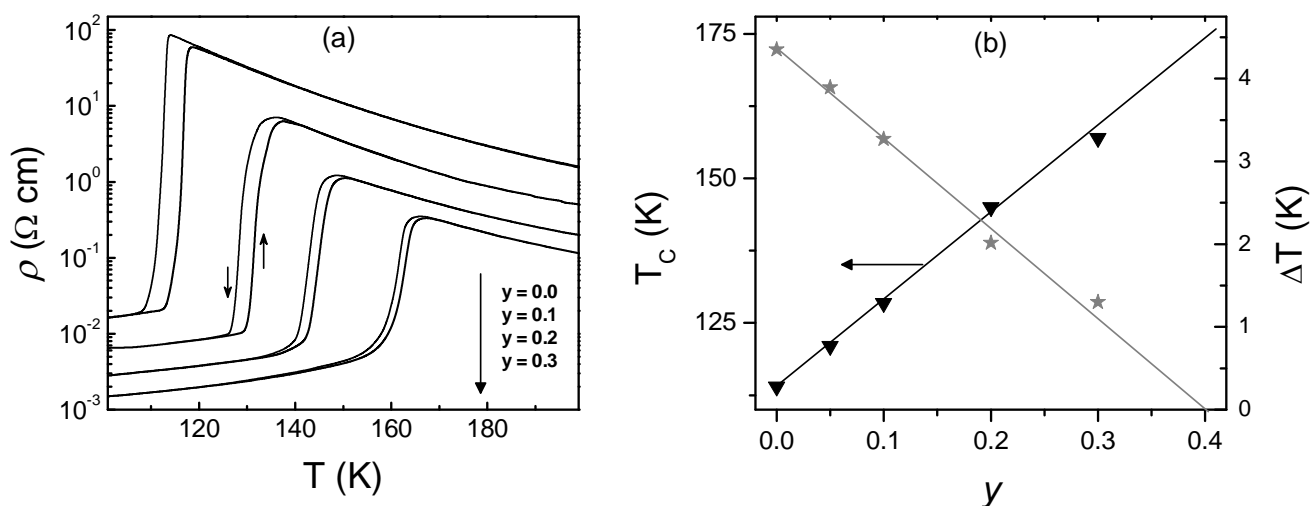


Figure 6. (a) Temperature dependence of resistivity of $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ single crystals with $y = 0, 0.1, 0.2,$ and 0.3 . (b) y dependence of T_C (heating cycle) and ΔT .

So it is clear that the system possesses several critical end points at which the first-order FM transition truncates. The effect of P on the nature of FM transition have also been examined in $(\text{Sm}_{1-y}\text{Nd}_y)_{0.52}\text{Sr}_{0.48}\text{MnO}_3$ with $y = 0.2$ and 0.3 and found that in both cases, the effects are qualitatively similar to that of SSMO, but the value of P_{cr} reduces with increasing y and becomes 10 kbar or 1 GPa for $y = 0.3$. 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. We have analyzed the critical behavior for $y = 0.3$ crystal in presence of $P = 12.1$ kbar, which is larger than P_{cr} . The detailed investigation of critical exponents and critical amplitudes reveal that the FM–PM transition under $P = 12.1$ kbar becomes purely second-order belonging to Heisenberg universality class.

CONCLUSIONS

In summary, we have studied the nature of FM to PM phase transition in SSMO single crystal and observed that the transition is strongly first-order in the ambient condition. The application of external H increases T_C , diminishes the first-order character of the transition and above a critical point ($H_{cr} \approx 4$ T, $T_{cr} \approx 160$ K), the transition becomes a crossover. The effect of pressure on the nature of FM transition is quite similar to that of H and the corresponding critical point, where the character of FM transition changes from first- to second-order is ($P_{cr} \approx 2.5$ GPa, $T_{cr} \approx 160$ K). We have also determined the location of critical point expressed in terms of Nd concentration and found that the transition changes to second-order at ($y_{cr} \approx 0.4$, $T_{cr} \approx 175$ K).

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