Modification of the spin state in $Sm_{0.52}Sr_{0.48}MnO_3$ by external magnetic field

P. Sarkar[1] and P. Mandal[2,a)]
[1]*Department of Physics, Serampore College, Serampore 712 201, India*
[2]*Saha Institute of Nuclear Physics, 1/AF Bidhannagar, Calcutta 700 064, India*

(Received 5 October 2007; accepted 15 January 2008; published online 4 February 2008)

The effect of applied magnetic field (H) on the magnetic properties of $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal in the paramagnetic (PM) state has been investigated. We observe a field induced steplike jump in magnetization (M) above T_C (110 K). The temperature and magnetic field dependence of susceptibility reveal that the PM phase of this system is quite complicated due to the coexistence of ferromagnetic (FM) and antiferromagnetic (AFM) interactions. The nature of magnetic interaction changes abruptly from AFM to FM at around 3.5 T, above which M ceases the steplike jump. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2840184]

Unusual magnetoelectronic properties as well as the rich phase diagram in narrowband manganites $R_{1-x}A_xMnO_3$ (R: rare earth ions, A: alkaline earth ions) are the manifestations of strong interplay between spin, charge, and orbital degrees of freedom. Several studies on narrowband manganites have revealed that the physical properties of these systems are strongly affected by the presence of local disorder arising mainly due to the size mismatch between R and A ions.[1–4] The magnitude of disorder can be identified as the variance of the ionic radii, $\sigma^2=[(1-x)r_R^2+xr_A^2-r_0^2]$, where r_R and r_A are the ionic radii of R and A ions, respectively, and the mean ionic radius is $r_0=(1-x)r_R+xr_A$.[5] Thus, one can introduce a large local disorder 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, $Sm_{1-x}Sr_xMnO_3$ has been studied extensively over a wide range of doping concentration (x); in particular, close to $x=0.5$, where a ferromagnetic (FM) metal (M) to an antiferromagnetic (AFM) or charge ordering insulator (I) transition occurs.[3,4,6–9] Because, when the system is close to MI transition, even a weak perturbation, such as magnetic field, pressure, irradiation, isotope substitution, quenching, annealing, etc. may change the physical properties dramatically or induce a phase transition.[6,10–12] Among these external parameters, magnetic field is most suitable to tune the magnetic states because it interacts mainly with the spin degrees of freedom. Moreover, magnetic field is continuously tunable, nondestructive, and reversible.

In this letter, we focus on the nature of the paramagnetic (PM) state of $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal in the presence of external magnetic field (H). Unlike conventional ferromagnet, magnetic susceptibility of this system shows a strong field dependence in the PM state. Our analysis also reveals the coexistence of FM and AFM interactions in the PM state and the nature as well as the strength of the interaction can be tuned with the application of magnetic field.

The single crystals of $Sm_{0.52}Sr_{0.48}MnO_3$ have been prepared by the floating zone technique using an infrared image furnace.[13,14] The quality of the crystal was carefully checked by various techniques such as electron dispersive x-ray analysis, x-ray diffraction, ac susceptibility, etc. The dc magnetization (M) and ac magnetic susceptibility were measured in a superconducting quantum interference device magnetometer (MPMS, Quantum Design) in the temperature range from 5 to 300 K.

Figure 1 shows a series of isotherms of magnetization of $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal in the vicinity of the Curie temperature ($T_C=110$ K). Just above T_C, M shows a two-step behavior and a large hysteresis between increasing and decreasing field is observed (inset of Fig. 1). Initially, M increases sublinearly with field up to ~0.3 T and then increases sharply before it reaches the saturation. One can see that the behavior of M is extremely sensitive to temperature and field. Above 116 K, 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 (H_c) followed by a steplike jump. Such a discontinuous change in M along with hysteresis persists for $H_c<4$ T. This steplike increase of M is possibly due to the formation of inhomogeneous metastable state in presence of local disorder.[4]

To perceive the signature of inhomogeneity, we have also studied ac magnetic susceptibility. The temperature de-

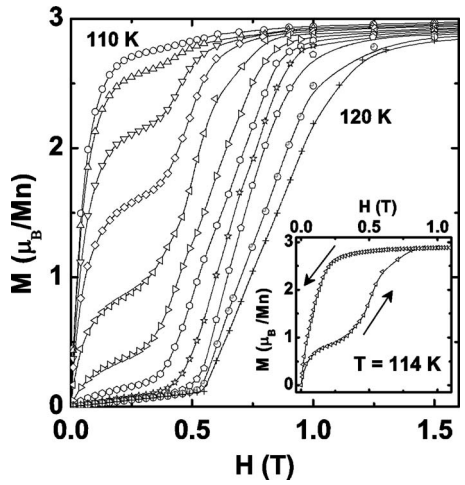

FIG. 1. Isothermal magnetization (M vs H) of $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal between 110 K (top) and 120 K (bottom) in 1 K interval (data with increasing field are shown). Inset: hysteresis in $M(H)$ at $T=114$ K.

a)Electronic mail: prabhat.mandal@saha.ac.in.

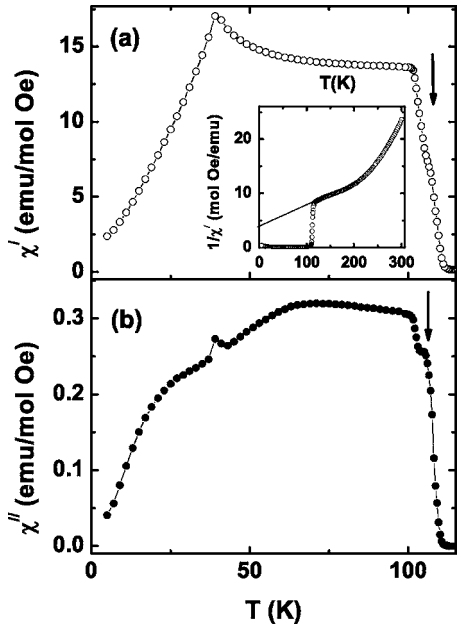

FIG. 2. Temperature dependence of ac susceptibility at 10 Hz for $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal: (a) real part (χ') and (b) imaginary part (χ''). The appearance of the anomaly near T_C (110 K) is indicated by the arrow. Inset: temperature dependence of $1/\chi'$ in the PM state.

pendence of the real (χ') and imaginary (χ'') parts of the ac susceptibility for $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal are presented in Fig. 2. Though the overall behavior of χ' (T) is qualitatively similar to earlier work, our data exhibit some additional important features.[6] With decreasing T, both χ' and χ'' show an abrupt increase at FM transition and then increase slowly with further decrease of T over a span of 40–50 K. Just below T_C, χ' shows a weak anomaly that appears quite prominently in χ'', is probably related to the structural phase transition.[8,15] We would like to mention that this anomaly was not reported earlier in case of polycrystalline samples.[6] At 40 K, χ' exhibits a sharp cusp while χ'' shows a small peak possibly due to the AFM ordering of Sm moment. The decrease of χ' below the cusp may be related to the clusterlike inhomogeneity.[6] Interestingly, we observe that χ' decreases precisely as $T^{3/2}$ for $T \leqslant 40$ K. Apart from the low-temperature decrease of susceptibility, the large value of χ'' in the FM state is possibly due to the presence of microscopic inhomogeneity. The temperature dependence of $1/\chi'$ is shown in the inset of Fig. 2. $1/\chi'$ above T_C shows a tendency for AFM interaction that competes with FM interaction at relatively high temperatures, i.e., both FM and AFM interaction coexist. The coexistence of both FM and AFM interaction in PM state is also reported from inelastic neutron scattering.[7] Another important observation is that χ' does not exhibit critical behavior above T_C as in the case of second order FM to PM phase transition.

For a better understanding of the nature of PM state, we have investigated the temperature dependence of M for different H. Figure 3 shows T dependence of inverse dc susceptibility χ_{dc}^{-1} $(=H/M)$ for some selected magnetic fields. The behavior of low-field χ_{dc} is similar to χ'. At relatively high temperatures, above 230 K, $\chi_{dc}^{-1}(T)$ for different magnetic fields merge to each other. With lowering of temperature, however, $\chi_{dc}^{-1}(T)$ tends to flatten and shows a strong dispersion with H. With increasing H, $\chi_{dc}^{-1}(T)$ does not decrease significantly up to a certain field but for $H \geqslant 4$ T, it decreases

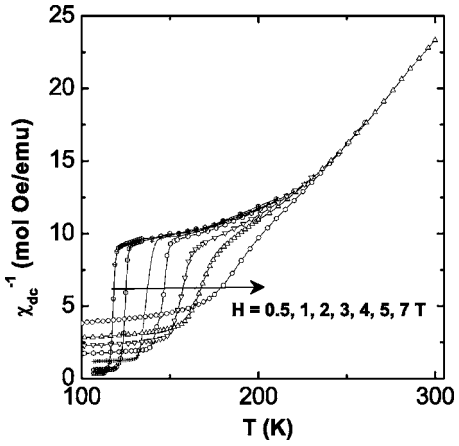

FIG. 3. Temperature dependence of inverse dc susceptibility (χ_{dc}^{-1}) for $Sm_{0.52}Sr_{0.48}MnO_3$ single crystal for different magnetic fields. The arrow indicates the direction of increasing field.

monotonically and the flattening nature becomes less and less prominent and disappears at high field. The decrease of H/M with increasing H implies the superlinear dependence of M on H. Such a strong field dependence of magnetic susceptibility in the PM state has not been reported earlier in this class of materials. From the nature of T and H dependence of M, it appears that the PM state of this system is quite different from that of conventional ferromagnet. To analyze the nature of PM state more precisely, we have calculated the effective moment (p_{eff}) and molecular field constant (λ) for different magnetic fields both below and above 230 K. The Curie constant (C) is determined from the approximate linear region of χ_{dc}^{-1} $(=T/C-\theta/C)$. The Weiss constant θ is related to the molecular field constant via $\theta=\lambda C$, which measures the strength as well as the nature of magnetic interaction. A positive value of λ indicates that the molecular field is adding to the applied field and, therefore, tending to make the elementary magnetic moments parallel to each other and to the applied field, i.e., FM interaction. If λ is negative, the molecular field opposes the applied field and tends to decrease the susceptibility, i.e., AFM interaction.

The magnetic field dependence of p_{eff} [calculated from $C=(\mu_B p_{eff})^2 n/3k$] and λ in two different temperature regions are shown in Fig. 4. In both the regions, the values of p_{eff} at

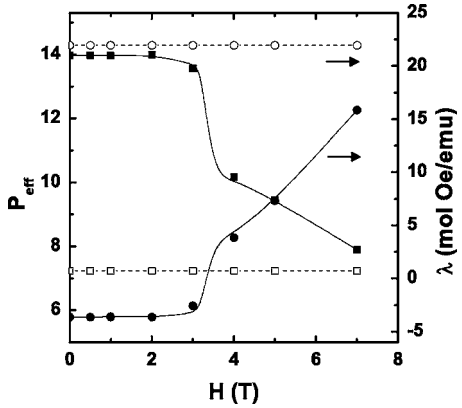

FIG. 4. Magnetic field dependence of p_{eff} (square) and λ (circle) for $Sm_{0.52}Sr_{0.48}MnO_3$ in two different temperature regions. The solid and the dashed lines are the guides to the eye, connecting data points below and above 230 K, respectively.

various magnetic fields are higher than that expected from the Curie-Weiss law for free Mn moments (p_{eff}=4.44). The high value of p_{eff} suggests the formation of magnetic clusters of a few Mn^{3+} and Mn^{4+} ions together.[16] The field dependence of p_{eff} and λ below 230 K indicate that the intracluster coupling is FM (large value of p_{eff}) while the intercluster coupling is AFM (negative λ) for the small value of applied field. As field increases, there is almost no change in p_{eff} and λ up to a certain field but at ~3.5 T, λ increases abruptly and becomes positive while p_{eff} drops sharply. So the application of the magnetic field leads to a change in the phase diagram: the weakening of AFM interaction and stabilization of FM interaction. In this context, it may be mentioned that the observed field-induced steplike jump in M (Fig. 1) persists as long as the magnetic interaction in the PM state is AFM. Although the heterogeneous nature of the PM state in narrowband manganites is well known, in the present work we have explicitly shown that, in ferromagnetic $Sm_{0.52}Sr_{0.48}MnO_3$, the effective interaction in the PM phase is AFM below 3.5 T and the interaction changes abruptly to FM at around 3.5 T. We believe that this kind of behavior has not been reported earlier in ferromagnetic manganites. In the present system, the change of the magnetic interaction from AFM to FM induced by temperature and magnetic field may be compared with the spin-state transition in metallic double exchange systems $MnAs_{1-x}P_x$ and $Cr_xMn_{1-x}As$.[17] These systems exhibit a low-spin to high-spin state transition in the PM phase with increasing temperature. The effective magnetic interaction is AFM in the low-spin state and FM in the high-spin state. It may also be mentioned that the present results contrast strikingly with the phase separation phenomena observed in other manganites, where the competing FM and AFM interactions for different ground states exist in the ferromagnetic state well below T_C.[18] In a previous work, we have shown from magnetization, specific heat, and resistivity data that this system exhibits a first order FM-PM phase transition when the applied magnetic field is small but the order of transition changes to second order at around H=4 T.[14] Comparing the present result with the earlier one, it is clear that the order of the magnetic transition in $Sm_{0.52}Sr_{0.48}MnO_3$ is related to the nature of the magnetic interaction in the PM phase.

In summary, the present study on the magnetic properties of $Sm_{0.52}Sr_{0.48}MnO_3$ single crystals provides several interesting results. We observe a steplike metamagnetic transition above T_C. Magnetic susceptibility reveals that in the PM ground state the effective interaction is FM above 230 K and AFM below this temperature. The most noteworthy is that susceptibility below 230 K exhibits strong field dependence and the analysis shows that the effective interaction is AFM for small H but changes abruptly to FM at around H =3.5 T. Furthermore, M exhibits the steplike metamagnetic transition as long as the interaction in the PM state is AFM. We believe, the present investigation will help to understand the nature of the magnetic state of narrowband manganites, where disorder plays an important role.

The authors would like to thank P. Choudhury for enlightening discussions and A. Pal for technical assistance.

[1] E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. **344**, 1 (2001).
[2] C. Martin, A. Maignan, M. Hervieu, and B. Raveau, Phys. Rev. B **60**, 12191 (1999).
[3] Y. Tomioka and Y. Tokura, Phys. Rev. B **70**, 014432 (2004).
[4] L. M. Fisher, A. V. Kalinov, I. F. Voloshin, N. A. Babushkina, D. I. Khomshkii, Y. Zhang, and T. T. M. Palstra, Phys. Rev. B **70**, 212411 (2004).
[5] L. M. Rodriguez-Martinez and J. P. Attfield, Phys. Rev. B **54**, R15622 (1998); **63**, 024424 (2000).
[6] N. A. Babushkina, E. A. Chistotina, O. Yu. Gorbenko, A. R. Kaul, D. I. Khomski, and K. I. Kugel, Phys. Rev. B **67**, 100410(R) (2003).
[7] Y. Endoh, H. Hiraka, Y. Tomioka, Y. Tokura, N. Nagaosa, and T. Fujiwara, Phys. Rev. Lett. **94**, 017206 (2005).
[8] A. N. Styka, Y. Ren, O. Yu. Gorbenko, N. A. Babushkina, and D. E. Brown, J. Appl. Phys. **100**, 103520 (2006).
[9] Y. Tomioka, H. Kuwahara, A. Asamitsu, M. Kasai, and Y. Tokura, Appl. Phys. Lett. **70**, 3609 (1997).
[10] H. Y. Hwang, T. T. M. Palstra, S. W. Cheong, and B. Batlogg, Phys. Rev. B **52**, 15046 (1995).
[11] V. Kiryukhin, D. Casa, J. P. Hill, B. Keimer, A. Vigliante, Y. Tomioka, and Y. Tokura, Nature (London) **386**, 813 (1997).
[12] M. Egilmez, K. H. Chow, and J. Junga, Appl. Phys. Lett. **89**, 062511 (2006).
[13] P. Mandal, B. Bandyopadhyay, and B. Ghosh, Phys. Rev. B **64**, 180405R (2001); P. Mandal and B. Ghosh, Phys. Rev. B **68**, 014422 (2003).
[14] P. Sarkar, P. Mandal, S. K. Bera, and S. M. Yusuf (unpublished).
[15] A. I. Kurbakov, A. V. Lazuta, V. A. Ryzhov, V. A. Trounov, I. I. Larionov, C. Martin, A. Maignan, and M. Hervieu, Phys. Rev. B **72**, 184432 (2005).
[16] R. P. Borges, F. Ott, R. M. Thomas, V. Skumryev, J. M. D. Coey, J. I. Arnaudas, and L. Ranno, Phys. Rev. B **60**, 12847 (1999).
[17] R. Wöhl and K. Bärner, J. Magn. Magn. Mater. **21**, 80 (1980); K. Bärner, in *Double Exchange in Heusler Alloys and Related Materials*, edited by K. Baerner (Research Signpost, Kerala, 2007), p. 1.
[18] M. Fäth, S. Freisem, A. A. Menovsky, Y. Tomioka, J. Aarts, and J. A. Mydosh, Science **285**, 1540 (1999); A. Yakubovskii, K. Kumagai, Y. Furukawa, N. Babushkina, A. Taldenkov, A. Kaul, and O. Gorbenko, Phys. Rev. B **62**, 5337 (2000).