Observation of three-dimensional Heisenberg-like ferromagnetism in single crystal $La_{0.875}Sr_{0.125}MnO_3$

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We report measurements and analysis of the magnetic critical phenomena in a single crystal of $La_{0.875}Sr_{0.125}MnO_3$. The critical exponents associated with the ferromagnetic transition have been determined from ac susceptibility and dc magnetization data. Techniques like the Kouvel-Fischer plots, the modified Arrots plots, and the critical isotherm analysis were used for this purpose. The values of the exponents γ, β, and δ obtained are found to match very well with those predicted for the three-dimensional Heisenberg model. Our results are consistent with recent numerical calculations and suggest that though the double exchange interaction is driven by the motion of conduction electrons, the effective magnetic interaction near the transition is renormalized to a short range one.

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A universality class (based on the dimensionality of the lattice and the order parameter) is assigned to a system depending on the values of the asymptotic critical exponents that characterizes the phase transition.[1] This practice of assigning such universal classes based on theoretical spin-spin interaction models (like the Heisenberg, mean field or the Ising) has been extremely useful in trying to discern the intricacies of magnetic transitions in real systems.

The nature of the para-ferro magnetic phase transition in mixed valent manganites of the general formula $La_{1-x}A_xMnO_3$ (where A is a divalent atom) is an area where the findings of experiments are yet to converge with theoretical models. The nature of the phase transitions in this class of materials was qualitatively explained by the double exchange (DE) interaction,[2] which originates from the motion of conduction electrons, and its coupling with spins localized at the lattice. Thus these materials provide us with a unique class of strongly correlated electron systems where the lattice, charge, and spin degrees of freedom are strongly coupled. It is interesting to determine how the kinetics of electrons which favor extended states affect the critical fluctuations and renormalizes the DE interaction in the vicinity of the phase transition.

There have been very few reports on the characterization of the critical regime in metallic double exchange driven manganites, and their results have been extremely inconclusive. In systems with a hole doping $>10\%$, which is necessary to observe a DE driven phase transition, the values of the critical exponent associated with magnetization (β) is seen to vary from 0.37 (Ref. 3) to 0.50.[4] This encompasses the values of the exponent as predicted by the three-dimensional (3D) Heisenberg and the 3D Ising and the mean field models, making it difficult to ascertain the universality class to which these systems belong. Even in the same study, different exponents have shown values falling into different universality classes, and the fact that no concensus could emerge was reason enough for many to doubt whether the metallic double exchange driven transition could actually be ascribed to a single universality class. Besides the above mentioned values which were determined using bulk magnetic measurements, the values of the critical exponents determined using other techniques like microwave absorption,[5] microwave surface impedance,[6] and neutron diffraction[7] have also varied, adding to the ambiguity in this issue.

The quality of the samples used in the study of criticality in this class of materials is a probable culprit for the present state of affairs. These samples are notoriously inhomogenous, thus causing a smeared transition (with a distribution of T_C's), which leads to an erroneous determination of the asymptotic critical exponents. Thus, it is imperative that homogenous single crystals be used for experimental investigations of this nature.

In an attempt to resolve the inconsistencies existing in literature, and to determine the nature of ferromagnetism in metallic DE manganites, we have done ac susceptibility and dc magnetization measurements in the critical region of a high purity single crystal of orthorhombic $La_{0.875}Sr_{0.125}MnO_3$. The samples were prepared by the standard solid state route using 99.999% pure La_2O_3, $SrCO_3$ and MnO_2. Single crystals were grown using a four mirror floating zone furnace under 6–8 atm of an Ar/O_2 mixture. The crystallographic phase purity was checked repeatedly by x ray diffraction, and the samples were found to be of single phase. The oxygen stochiometry of this sample was found to be 3.00 ± 0.02. The chemical homogenity of the sample was checked using electron probe microanalysis (EPMA) and variation was found to be within 1%. Further details of sample preparation can be found in Ref. 8.

Dc magnetization and ac susceptibility measurements were carried out using home made setups.[9,10] In the critical region, the temperature was controlled to an accuracy of 0.01 K using commercial Lakeshore controllers DRC93CA and 340. All the measurements were made with the applied field parallel to the *ab* plane of the crystal. The low field linear region of M-H scans were used to determine the value of the

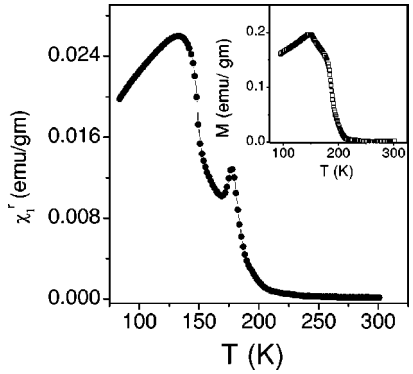

FIG. 1. The real part of the first order susceptibility measured as a function of temperature for the sample $La_{0.875}Sr_{0.125}MnO_3$ at a exciting frequency of 133.33 Hz and a driving ac field of 0.92 Oe. The inset shows the dc magnetization measured at a field of 3.42 Oe.

demagnetization factor, which in our case was found to be 0.22. This value matches very well with the one computed using the dimensions of the sample used for the measurements. All the data used for determining the critical exponents were duly corrected for the demagnetization fields.

The composition $La_{0.875}Sr_{0.125}MnO_3$ ($La_{1-x}A_xMnO_3$, with $x=1/8$) occupies a prominent place in the scheme of manganites, being in the transition region from the superexchange dominated antiferromagnetic (AFM) manganites to the DE driven FM ones.[11] It has been extensively studied in the past,[12] and is reported to show two transitions in reasonably close succession, a paramagnetic insulator-ferromagnetic metal transition, followed by another low temperature transition to an insulating state. This state is now being explained by invoking polaronic[13] and orbital ordering[14] models. However, in this study, we have concentrated on the higher temperature transition which is likely to be driven by the DE interaction. Figure 1 shows the real part

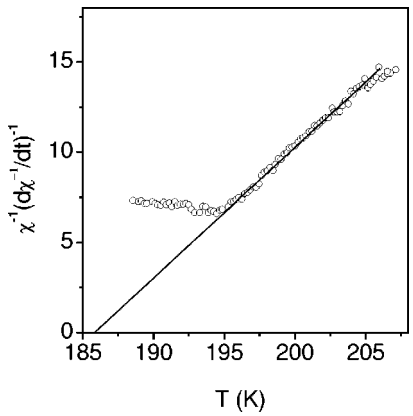

FIG. 2. Kouvel Fischer plots of the real part of the first order susceptibility for the sample $La_{0.875}Sr_{0.125}MnO_3$. The inverse of the slope gives the value of the susceptibility exponent (γ), and the intercept on the temperature axis provides the transition temperature (T_C). We have obtained $\gamma=1.38$ and $T_C=186$ K. Increasing nonlinearity causes deviations from the straight line fit close to the transition.

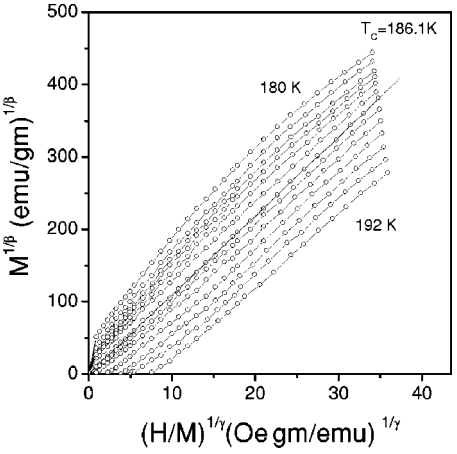

FIG. 3. Modified Arrot plots using the equation of state $(H/M)^{1/\gamma}=a[(T-T_C)/T]+bM^{1/\beta}$. We have used $\gamma=1.38$, and the best fit was obtained with $\beta=0.37$. All the isotherms measured in the critical region have not been shown for the sake of clarity.

of the first order ac susceptibility and the dc magnetization measured in this sample where the two transitions mentioned above are clearly seen.

Conventional methods of analyzing the critical region involves the use of Arrot plots.[15] These M^2 vs H/M isotherms should be straight lines in the critical region, with a zero intercept at $T=T_C$. The intercepts for $T<T_C$ and $T>T_C$ is then used to determine the saturation magnetization (M_S) and the inverse susceptibility ($1/\chi_0$) respectively. It is to be noted that this method implicitly assumes mean field values of the exponents, and a more general form of analysis (called the modified Arrot plots) involves plotting $M^{1/\beta}$ vs $(H/M)^{1/\gamma}$ in the critical region.[16] However, these methods do tend to introduce some systematic errors due to the presence of two (β and γ) free parameters in the fitting procedure.

We have circumvented this problem by using low field ac susceptibility measurements in the critical region to determine γ. Low field ac susceptibility is an extremely useful tool in the study of a para-ferro phase transition, as it enables us to directly determine the true initial susceptibility (χ_0), which otherwise has to be estimated from the extrapolation

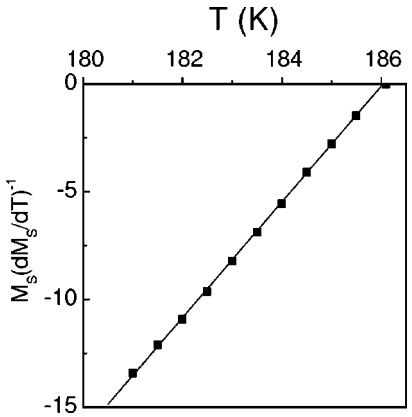

FIG. 4. Kouvel Fisher analysis of the saturation magnetization (M_S) plotted as a function of temperature. The inverse of the slope gives a value $\beta=0.372$.

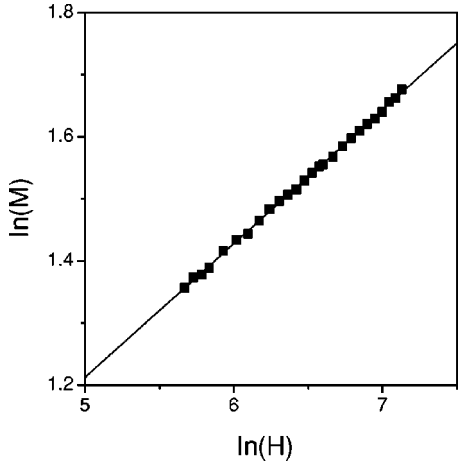

FIG. 5. $\ln M$ plotted as a function of $\ln H$ for the isotherm measured at $T=T_C$. The inverse of the slope of the linear region gives a value of $\delta=4.72$.

of data measured at high applied fields[17] which besides suppressing effects like inhomogeniety in the sample can mask the true critical behavior of the system. This is specially important in systems like the hole doped manganites where short range correlations are known to exist till temperatures as large as $2T_C$.[18] Also, the Kouvel-Fischer (KF) (Ref. 19) analysis can then be used to independently estimate both the transition temperature (T_C) and γ. Ac susceptibility data analyzed using the KF formalism $[1/\chi_0 d/dt(\chi_0^{-1})$ vs $T]$ is shown in Fig. 2. We have obtained $\gamma=1.38$ which matches very well the isotropic 3D Heisenberg value of 1.396. *It is to be noted that to the best of our knowledge this is the first report of a KF analysis of low field ac susceptibility in this class of materials*. Though manganites have been speculated to be magnetically inhomogeneous,[20,21] the fact that a reasonable fit could be obtained to the low field data, and the fact that we could obtain self-consistent values of the exponents using both low and high field data (as is shown below) adds credence to our claim that the sample used is homogenous at least as far as the time scales of our experimental probes are concerned.

Modified Arrot plots[22] based on the equation of state

$$\left(\frac{H}{M}\right)^{1/\gamma} = a\frac{(T-T_C)}{T} + bM^{1/\beta}$$

is shown in Fig. 3. The value of γ determined using the KF analysis of ac susceptibility is used, and β is varied continuously, so as to obtain isotherms almost parallel to each other in the critical region. These isotherms are curved at low fields, as they represent averaging over domains which are magnetized over different directions,[15,22,23] and care has to be taken to discard this region from the fitting procedure. As is clearly seen in Fig. 3, at higher fields, the isotherms are straight lines, and the best fit was obtained with a value $\beta=0.37$. The value of (M_S) determined by the intercepts of the modified Arrot plots in the region $T<T_C$ is then analyzed using the Kouvel Fischer formalism [plotting $M_S(dM_S/dT)^{-1}$ vs T] to reconfirm the value of β. As is shown in Fig. 4, a good fit was obtained with a value $\beta=0.37$.

Figure 5 shows $\ln(M)$ plotted as a function of $\ln(H)$ (using the critical isotherm), the inverse slope of which is used for determining the value of the critical exponent δ. We obtain a value of $\delta=4.72$, which matches very well with the one ($\delta=4.709$) determined by using the Widom scaling equation $\delta=1+\gamma/\beta$. A chart comparing the values of the exponents which we have obtained with those reported in literature for metallic double exchange systems is shown in Table I. As is clearly seen, we have obtained values which match very well with the isotropic 3D Heisenberg values.

It has been shown analytically[24] that if in a 3D ferromagnet, the effective exchange interaction $J(r)$ decays with distance (r) at a rate faster than r^{-5}, then the short range (Heisenberg like) exponents are valid. However, if $J(r)$ decays at a rate slower than $r^{-4.5}$, then the classical (mean-field-like) exponents are valid. According to the theory of double exchange, the effective ferromagnetic interaction is

TABLE I. Values of the exponents β, γ and δ as determined from the Kouvel-Fischer analysis of the first order susceptibility, modified Arrot plots, and the critical isotherm. The values determined by the earlier workers in similar metallic double exchange systems and that expected for various universal models is given for the sake of comparison.

Composition	Ref.	Technique	β	γ	δ
$La_{0.875}Sr_{0.125}MnO_3$[a]	This work	Bulk magnetization	0.37 ± 0.02	1.38 ± 0.03	4.72 ± 0.04
$La_{0.7}Sr_{0.3}MnO_3$[a]	3	Bulk magnetization	0.37 ± 0.04	1.22 ± 0.03	4.25 ± 0.2
$La_{0.7}Sr_{0.3}MnO_3$[a]	5	Microwave absorption	0.45 ± 0.05	–	–
$La_{0.7}Sr_{0.3}MnO_3$[a]	7	Neutron diffraction	0.295 ± 0.004	–	–
$La_{0.8}Sr_{0.2}MnO_3$[b]	4	Bulk magnetization	0.50 ± 0.02	1.08 ± 0.03	3.13 ± 0.20
$La_{0.8}Sr_{0.2}MnO_3$[a]	6	Microwave surface impedance	0.45 ± 0.05	–	–
Mean field model	16	Theory	0.5	1.0	3.0
3D Ising model	29	Theory	0.3265	1.2372	4.789
3D Heisenberg model	29	Theory	0.3689	1.396	4.783

[a]Single crystals
[b]Polycrystals.

driven by the kinetics of the electrons which favor extended states, and hence one would expect a (long range) mean field like universality class. However, recent numerical calculations[25–28] show that the exponents in the DE model are consistent with those of the isotropic short range 3D Heisenberg model.

Our results match well with these calculations and are the first of its kind to show unambigously that the para-ferro phase transition in metallic manganites fall into the isotropic short range 3D Heisenberg universality class. The consistency of the exponent values determined using low field, and high field magnetization data ensures that the values are intrinsic to the system and are not arising either due to sample inhomogeniety or any artifacts in the measurement/fitting processes. The 3D Heisenberg like values of the critical exponents thus imply that the effective interactions in a canonical DE model are relatively short range-near the transition. This is important in the context of phase separation models being used to explain various intruiging features of the hole doped manganites.[11] This intrinsic phase separation (into hole rich and hole poor regions) could possibly be the reason for the short range nature of the magnetic interactions, and hence the 3D Heisenberg like values of the critical exponents. Further studies on various dopants and doping levels would be required before this finding can be generalized to all double exchange driven manganites, since there are reports suggesting a first order transition in the $La_{2/3}Ca_{1/3}MnO_3$ system.[30]

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