Critical behavior of the field-dependent susceptibility in single-crystal $La_{0.73}Ba_{0.27}MnO_3$

Wei Li, H. P. Kunkel, X. Z. Zhou,* and Gwyn Williams
Department of Physics and Astronomy, University of Manitoba, Winnipeg, Manitoba R3T 2N2, Canada

Y. Mukovskii and D. Shulyatev
Moscow State Steel and Alloys Institute, Moscow 119049, Russia

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Detailed field- and temperature-dependent ac susceptibility data on single-crystal $La_{0.73}Ba_{0.27}MnO_3$, while confirming the applicability of Heisenberg model exponents, indicate that the associated scaling law extends over a wide region of the (H,T) plane around T_c, with no evidence of crossover effects. From this it can be inferred that the system is magnetically homogeneous (at least in applied fields exceeding any anisotropy/coercive field), an unanticipated result in view of the existence of various sources of disorder (double- and superexchange coupling, for example), and reports of spontaneous electronic phase separation and Griffiths singularities in doped manganese perovskites.

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Field- and temperature-dependent ac susceptibility measurements, $\chi(H,T)$, provide a powerful technique for estimating critical exponents at a continuous/second-order paramagnetic to ferromagnetic phase transition.[1] While the zero-field ac susceptibility, $\chi(0,T)$, exhibits the usual power-law increase with decreasing temperature immediately above the ordering temperature, T_C, viz.

$$\chi(0,T) \propto t^{-\gamma}; \quad t = \frac{T - T_C}{T_C}. \tag{1}$$

Measurements in nonzero static biasing (applied) fields, H_a, reveal a series of peaks/critical maxima, the temperature (T_M) of which increases while the amplitude, $\chi(H,T_M)$, decreases as H_a is increased. The locus of these maxima delineates the location of a crossover line in the $(H\text{-}T)$ plane, above which the response is thermally dominated, whereas below this line it is field dominated. The appearance of these critical maxima can be understood generally in terms of the fluctuation-dissipation theorem,[2] and their variation with field and temperature are—according to the conventional static scaling law[3]—governed by a series of power laws from which the critical exponents can be estimated.[1,4] Specifically the amplitude, $\chi(h,t_m)$, and (reduced) temperature $t_m = |T_M - T_C|/T_C$ of these critical maxima can be written as follows in terms of the linear scaling fields h $(\sim H_i/T_C$, where the internal field $H_i = H_a - NM$ in the usual notation) and t, as follows:

$$\chi(h,t_m) \propto h^{1/\delta - 1}, \tag{2}$$

$$\chi(h,t_m) \propto t_m^{-\gamma}, \tag{3}$$

$$t_m \propto h^{1/\gamma + \beta}. \tag{4}$$

The first of these, Eq. (2), enables the exponent δ to be estimated *directly* from the field dependence of the amplitude (height) of these critical peaks, *independent of any choice for* T_C. The latter represents a distinct advantage over conventional magnetization-based estimates in which T_C needs to be determined prior to measurements being carried out (and stabilized) along the critical isotherm $(T = T_C)$. By contrast, in the ac susceptibility measurement, H is fixed and then the temperature increased (as summarized in Fig. 1) and from the ensuing peak amplitude, δ can be extracted using Eq. (2). Estimates of γ and β from Eqs. (3) and (4), the latter defining the crossover line, require that an estimate for T_C be made. The manner in which this is done is described subsequently.

The scaling law and the equations derived from it represent the asymptotic behavior as $t \to 0 (T \to T_C)$ and $h \to 0$. Here again, ac susceptibility measurements can provide an advantage. In magnetically soft, low-moment systems, where the technical/regular contributions to the response (arising from domain wall motion, coherent domain rotation) are driven to saturation in low fields, critical peak structure (and hence, critical analysis) can be observed in the most favorable cases[5] in fields as low as 0.1 Oe. By contrast, conventional approaches based on a modified Arrott-Noakes equa-

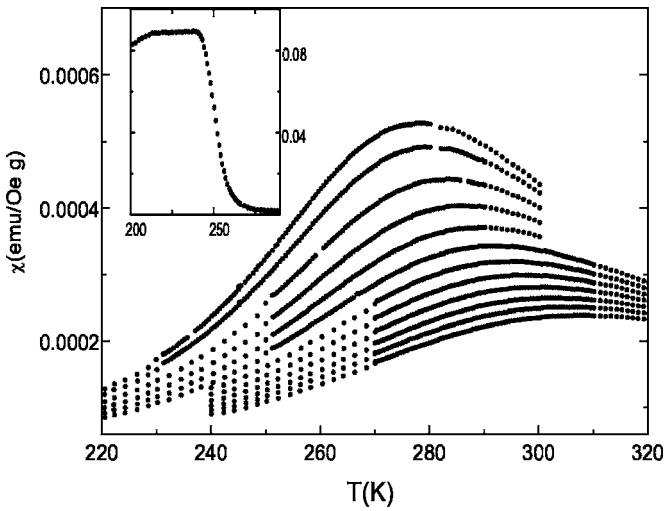

FIG. 1. AC susceptibility isokaps, i.e., measurements in fixed static biasing fields as a function of temperature, in fields of 32 kOe (top), then 35 kOe to 85 kOe (bottom) in 5 kOe steps. Inset: The zero-field ac susceptibility.

tion of state usually involve extrapolations from data acquired in fields above typically 1 kOe in an attempt to eliminate the region of response dominated by regular components, rather than by their critical counterparts. In this regard the manganites and pyrochlores provide interesting counterexamples in that they exhibit a technical hardness far greater than would be inferred from their coercive fields.[1,6]

An interesting example of this dichotomy is provided by a comparison of the measurements of the field and temperature-dependent ac susceptibility on single-crystal $La_{0.73}Ba_{0.27}MnO_3$ presented below, and earlier studies and analysis of its magnetization.[7] The sample was a single crystal of mass 0.163 g (a semicylindrical rod of length 6 mm and average radius 1.6 mm) grown using the floating zone technique at the Moscow State Steel and Alloys Institute;[8] its mosaicity was less than 1° and its coercive field did not exceed 4 Oe at temperatures below T_C. The latter is an important parameter in the current discussion since, in the most favorable cases mentioned above, applied fields comparable to the coercive field near T_C are often sufficient to first resolve critical peak structure in the ac susceptibility.[5] This is clearly *not* the case here, as Fig. 1 confirms. Here applied fields well in excess of 10 kOe [i.e., $10^4 H_C(T \sim T_C)$] are necessary to suppress the technical background so that the critical maxima are revealed. [These ac susceptibility data were acquired in a Quantum Design model 6000 PPMS using an ac driving field of 0.1 Oe rms at 2.4 kHz; the latter and the superimposed static biasing fields were applied along the largest (cylindrical) axis of the sample.]

Figures 2(a)–2(d) summarize the analysis of these ac susceptibility data. Figure 2(a) tests the validity of Eq. (2) for the present data. This double-logarithmic plot of the critical peak susceptibility, $\chi(h,t_m)$ (corrected for background and demagnetizing affects), against the internal field, H_i (with the demagnetizing factor N estimated from the maximum in the zero-field ac susceptibility, the insert in Fig. 1), confirms the power-law dependence, with the slope of this plot yielding

$$\delta = 4.84 \pm 0.09; \quad 20 \text{ kOe} \leqslant 85 \text{ kOe}.$$

This estimate is clearly *independent* of any choice for T_C. Test of the remaining power laws, Eqs. (3) and (4), obviously require such a choice to be made. Figure 2(b) indicates how this is accomplished, by initially plotting the measured peak temperatures, T_M, against $H_i^{0.57}$ [$0.57=(\gamma+\beta)^{-1}$ for Heisenberg model exponents, as the δ estimate above suggests]. The straight line dependence of T_M on H_i using these exponent values indirectly confirms their applicability, while the extrapolation of this line to $H_i=0$ yields an estimate for T_C of

$$T_C = 243.2 \pm 0.4 \text{ K}.$$

This estimate is then used in the double-logarithmic plots of Figs. 2(c) and 2(d), power law tests of the dependence of the (reduced) peak temperatures (t_m), and the peak amplitude [$\chi(h,t_m)$] on the internal field, Eqs. (4) and (3), respectively. These plots confirm such a power-law dependence, and their slopes yield

$$(\gamma+\beta) = 1.74 \pm 0.02; \quad 20 \text{ kOe} \leqslant H_i \leqslant 85 \text{ kOe}$$

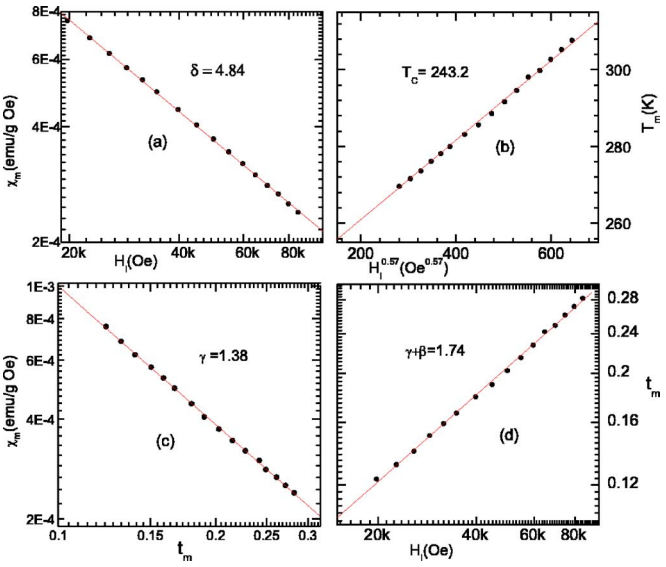

FIG. 2. (Color online) (a) The peak susceptibility, χ_m (in emu/g) taken from Fig. 1, plotted against the internal field, H_i (in kOe), on a double-logarithmic scale. The solid line confirms the power law, Eq. (2), and its slope yields $\delta=4.84\pm0.09$. (b) The peak temperature, T_m (in Kelvin) taken from Fig. 1, plotted against $H_i^{0.57}$ [i.e., Eq. (4) with Heisenberg model exponents]. The zero-field intercept yield a value for T_c of 243.2±0.4 K. (c) The peak susceptibility, χ_m (in emu/g), taken from Fig. 1, plotted against the reduced temperature on a double-logarithmic scale. The straight line drawn confirms the power-law prediction, Eq. (3), and its slope yields $\gamma=1.38\pm0.01$. (d) A double-logarithmic plot of the reduced peak temperature, t_m, using the T_c estimate from Fig. 2(b), against the internal field, H_i (in Oe). The straight line confirms the power-law dependence [Eq. (4)], and its slope yields $\gamma+\beta=1.74\pm0.02$. In all figures, the size of the data points are representative of the associated error.

$$\gamma = 1.38 \pm 0.01; \quad 0.1 < t_m < 0.3$$

from which $\beta=0.36\pm0.005$.

The significance of these results is twofold. First, they are in excellent agreement with exponent estimates deduced from the magnetization data,[7] noting in particular that the δ estimate obtained here does not depend on any choice for T_C, unlike that from magnetization data. Both sets of estimates confirm the applicability of Heisenberg model exponents[4] to this system, a result predicted by model simulations for double-exchange coupled spins, at least when anisotropy can be neglected.[9] Nevertheless these data indicate that manganites continue to display subtleties in their properties, here a technical hardness irreconcilable with the measured coercive field, manifested here via the magnitude of the applied field necessary to first resolve critical peak structure in the field-dependent ac susceptibility.

Second, and more important, as a corollary to the above, the ac susceptibility data indicate that the region of validity of the scaling law equation of state [on which Eqs. (2)–(4) are based] extends over a wide region of the (h,t) plane. Specifically, the scaling law extends to fields of some 85 kOe, with no indication whatsoever of crossover effects

(frequently to mean-field values).[12] The latter provides strong, though indirect, evidence of the homogeneous magnetic character of the present system, a homogeneity unexpected based on the presence of both mixed spin (Mn^{3+},Mn^{4+}) and variable-exchange couplings (double- and superexchange). Mean-field model predictions[4,10] indicate that for an assumed Gaussian distribution[11] of exchange coupling strengths, while a ferromagnetic ground state evolves when the ratio η ($=Jo/J$) of the first (Jo) to second (J) moment of this distribution exceeds unity, the region of validity of the associated critical/scaling behavior collapses in the (h,t) plane as η approaches unity from above. Numerous experiments[10,12] confirm the general validity of such predictions, though the exponents are usually Heisenberg rather than mean-field. A corollary to the above is that the observation of critical behavior over an extended range of field (or temperature) implies that $\eta \gg 1$, i.e., that the exchange coupling distribution is a very narrow one. In the context of the manganites, this would require that ferromagnetic double-exchange dominate (anti)ferromagnetic superexchange significantly, at least for the present Ba doping level. Such a result has not been anticipated, particularly in view of other reports of the effects of disorder—the appearance of a Griffiths phase, for example, in several other doped Mn perovskites.[13] The above result also suggests, again indirectly, that any spatial fluctuations associated with spontaneous electronic phase separation (SEPS) exists on a scale far smaller than the prevailing correlation length in the current experiments. However, given that the data presented above were acquired in the critical regime above T_c in substantial applied fields, the latter is not unexpected; while the onset of SEPS occurs near T_c, the application of magnetic fields enhances the ferromagnetic metallic phase near T_c, allowing this phase to dominate and form percolative paths (with the corresponding suppression of the antiferromagnetic insulating component), an effect that underlies colossal magnetoresistance in some model approaches.[14] Similarly the characteristic features of the Griffith's phase are also suppressed by applied fields.[15] The uniform applied field is the conjugate field for (collinear) ferromagnetism, so applying fields of increasing magnitude will promote the growth of ferromagnetism at the expense of nonuniform/Griffith's phases.

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*Corresponding author. Electronic address: zhou@cc.umanitoba.ca

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