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## New approach to critical dynamic scaling in random magnets

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A new approach to critical dynamic scaling of  $\chi''$  is presented wherein  $\beta/zv$ , zv, and  $T_c$  may all be *separately* determined *independent* of each other. Application is made to Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te where by use of this new procedure it is shown that the data in the critical region above  $T_c$  are inconsistent with activated dynamics. This new procedure applied to other systems, reveals frequent underestimates of zv and overestimates of  $T_c$ . Apparent systematic deviations from scaling which are found near and below  $T_c$  are briefly discussed.

Dynamic scaling of the ac susceptibility  $\chi'(\omega, T)$ + $i\chi''(\omega, T)$  has been used extensively as supporting evidence for critical behavior associated with phase transitions in random magnetic systems. In conventional dynamic scaling the relaxation time  $\tau$  is related to the correlation length  $\xi \sim |t|^{-\nu}$  via  $\tau \sim \xi^z \sim |t|^{-z\nu}$  where  $t = (T - T_c)/T_c$  is the reduced temperature. A different form of critical dynamics has been suggested <sup>1,2</sup> in which  $\tau$ is activated, i.e.,  $\tau \sim \tau_0 e^{B/T}$  where  $\tau_0$  is a microscopic time and the barrier *B* diverges as  $B \sim \xi^{\psi}$ , with  $\psi$  positive. This activated dynamics is expected to apply to random-field systems<sup>1,2</sup> and systems where  $T_c = 0$ , and it also has been suggested <sup>3</sup> for spin glasses with  $T_c > 0$ .

In a frequent application of conventional dynamic scaling, one uses the relation  $\phi = \chi''/\chi' = \omega \tau_{av} \approx \omega t^{-z_{av}v}$  valid for *small*  $\phi$  and from a best straight-line plot of  $\log_{10}t$  vs  $\log_{10}\omega$  at fixed  $\phi$  finds  $T_c$  and  $z_{av}v$ .<sup>4</sup> However, in many instances the criterion of *small*  $\phi$ , i.e., that  $\phi/\omega$  be independent of  $\omega$  for constant T, may be shown to have been violated, leading to an underestimate of  $z_{av}v$ . Moreover, by using small  $\chi''$  one is restricted to a region of the data with the largest experimental errors. An improvement on this has been to use the complete scaling form<sup>5</sup>

$$\chi''/\chi_{eq} \text{ or } \chi''T \approx \omega^{\beta/z\nu} f(\omega/t^{z\nu}),$$
 (1a)

$$(\chi_{eq} - \chi')/\chi_{eq} \approx \omega^{\beta/z\nu} g(\omega/t^{z\nu}),$$
 (1b)

where  $\chi_{eq} = \chi(\omega = 0)$ , f and g are scaling functions, and  $zv = z_{av}v + \beta$ .  $T_c$ , zv, and  $\beta$  are then selected to give the best collapse of all the data (as determined by visual examination) onto a single scaling plot. For  $\chi''$  this is  $\chi''T/\omega^{\beta/zv}$  vs  $\omega/t^{zv}$  and is usually done on a log-log plot since the abscissa extends over many decades as  $t \rightarrow 0$ . Such a log-log plot may conceal departures from good scaling that are considerably larger than the experimental error.

Our new scaling procedure starts with a simple but important recasting of the argument of the scaling function to  $t/\omega^{1/z\nu}$  instead of  $\omega/t^{z\nu}$ , i.e.,

$$\chi'' T/\omega^{\beta/z\nu} \approx \tilde{f}(t/\omega^{1/z\nu}).$$
<sup>(2)</sup>

The modification has profound implications for the scaling plot. Since 1/2v and  $\beta/2v \ll 1$ , the frequency dependence

is rather weak and now the argument of  $\tilde{f}$  is linear in t so that a linear scaling plot of  $\chi''T/\omega^{\beta/zv}$  vs  $t/\omega^{1/zv}$  resembles  $\chi''$  vs T itself. Thus any departures from perfect scaling may be more readily judged relative to the experimental error. Moreover, since the argument of the scaling function is no longer singular for  $t \rightarrow 0$ , the scaling plot may be extended to  $t \leq 0$  as well. More important, however, is that this modification immediately suggests a method of *separately* and *independently* determining  $\beta/zv$ , zv, and  $T_c$  from  $\chi''(\omega, T)$ , as described below.

This new method is illustrated with  $Cd_{0.6}Mn_{0.4}Te$ . Both first- and second-neighbor Mn interactions are antiferromagnetic in this naturally frustrated fcc lattice. The combination of frustration and dilution is expected to result in spin-glass behavior. See Ref. 7 for citations to extensive previous work on this system. The experimental work reported here is on  $\chi''(\omega, T)$  and supplements previous work on  $\chi'(\omega,T)$ , but is also subjected to the more exacting analyses of our new method. The Faraday rotation technique, described elsewhere,<sup>7</sup> was used to measure  $\chi' + i\chi''$ . The ac magnetic field  $h_{ac}$  was in all cases < 3 G so as to avoid any nonlinear effects greater than experimental error. The results are plotted in Figs. 1(a) and 1(b). In order to allow comparison of our particular sample and temperature scale with other works on this compound we specify the peak in  $\chi'$  (97.5 Hz) which is at T = 13.19 K.

We now show how, assuming conventional dynamic scaling (2),  $\beta/zv$ , 1/zv, and  $T_c$  can be separately and independently determined. This process depends on  $\chi''$  having a readily identifiable and accurately measured feature, namely its peak for a given  $\omega$ . This maximum occurs at  $T_p(\omega)$ , where  $\chi'' \equiv \chi_p''(\omega)$ . Since the peaks in  $\chi''T$  must coalesce to the same point on the scaling plot, according to Eq. (2), one has  $\chi_p''(\omega)T_p(\omega) \sim \omega^{\beta/zv}$ . Thus the slope of the straight line in the log-log plot in Fig. 2(a) yields  $\beta/zv = 0.053 \pm 0.002$ . (The temperatures at which  $\chi''T$  and  $\chi''$  peak are indistinguishable within experimental error.)

Generally, it would be more appropriate to scale  $\chi''/\chi_{eq}$ with  $\chi_{eq} \sim 1/T$  only for a symmetric (in sign) distribution of exchange interactions.  $\chi_{eq}(T)$  is determined directly at high enough T, and for lower T, is extrapolated from a power series fit<sup>7</sup> at higher T. We estimate that any error introduced in  $\chi''/\chi_{eq}$  by this extrapolation is less than



FIG. 1. (a) Relative  $\chi''(\omega, T)$  in Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te. Frequencies in multiples of 0.975 Hz. (b) Relative  $\chi'(\omega, T)$ . Vertical scale same as in (a).



FIG. 2. (a) Determination of  $\beta/zv$  from peaks  $\chi_p^{\nu}$  in  $\chi''(\omega)$ , i.e.,  $\chi_p^{\nu}/\chi_{eq}(T_p)$  or  $\chi_p^{\nu}(\omega)T_p(\omega) \sim \omega^{\beta/zv}$ . The curvature in the dashed line shows the departure from an attempted fit to activated dynamics (see text). (b) Determinations of 1/zv from  $(T_B - T_A) \sim \omega^{1/zv}$  where the symbols  $\bigcirc$ ,  $\textcircled{\bullet}$ , and  $\bigtriangleup$  correspond to  $\chi_n^{\nu}$  pairs in Fig. 3 (0.8*R*-0.8*L*), (0.6*R*-0.92*L*), and (0.3*R*-0.8*R*), respectively, and *R* and *L* refer to points to the right- and left-hand sides of the peak in  $\chi_n^{\nu}$  (see text). Note the systematic increase in the effective zv as lower temperature data is used.

0.3%, which is far smaller than the experimental error in  $\chi''$ . In Fig. 2,  $\log_{10}[\chi_p''(\omega)/\chi_{eq}(T_p)]$  is plotted versus  $\log_{10}\omega$  giving  $\beta/zv = 0.051 \pm 0.02$  compared to  $\beta/zv = 0.053$  when using  $\chi_p''T_p$ . While only very slight differences of this sort enter any of our analyses, we prefer  $\chi''/\chi_{eq}$ .

Activated dynamic scaling (3) can be recast in the form

$$(\chi''/\chi_{eq})(|\log_{10}\omega\tau_0|)^{p/Q} = G(t|\log_{10}\omega\tau_0|^{1/Q}), \quad (3)$$

where  $Q = \psi v$ . Similar scaling of  $\chi_p''(\omega)$  to determine p/Q is given by plotting  $\log_{10}[\chi_p''(\omega)/\chi_{eq}]$  vs  $\log_{10} \times |\log_{10}\omega\tau_0|$ , and looking for a straight-line fit, but one now has an extra constant  $\tau_0$  which must be selected. This is shown for the same six data points  $\chi_p''T_p$  which are connected in Fig. 2(a) by the dashed line; for these the absicissa is now  $\log_{10}|\log_{10}\omega\tau_0|$  (with  $\tau_0 = 10^{-12}$  s) instead of  $\log_{10}\omega$  and the scale has been adjusted linearly to match the two end points. The systematic curvature in this plot can only be eliminated by selecting unphysical values of  $\tau_0 < 10^{-15}$ , which rules against activated dynamics.

To find zv we now normalize  $\chi''(\omega, T)/\chi_{eq}$  at its peak to unity by dividing by  $\chi''_{p}(\omega)/\chi_{eq}(T_{p})$  as shown in Fig. 3, and call this quantity  $\chi''_{N}$ . Conventional dynamic scaling says  $\chi''_{N} \approx X(t/\omega^{1/zv})$ , while activated scaling has  $\chi''_{N}$  $\approx \tilde{X}(t|\log_{10}\omega\tau_{0}|^{1/2})$ , with X,  $\tilde{X}$  scaling functions. Therefore if we chose any two values of  $\chi''_{N}$ , e.g.,  $(A)\chi''_{N} = 0.92$  to the left-hand side of the peak (0.92L) and (B) 0.6 to the right-hand side of the peak (0.6R) as shown in Fig. 3, the temperature difference should scale as  $(T_{B} - T_{A}) \sim \omega^{1/zv}$  or  $(T_{B} - T_{A}) \sim |\log_{10}\omega\tau_{0}|^{1/Q}$ , respectively. Thus for conventional scaling, without any reference to  $T_{c}$  or  $\beta$  we can find zv by the slope of  $\log_{10}(T_{B} - T_{A})$  vs  $\log_{10}\omega$  as shown in Fig. 2(b) for several different selections of the pair of values of  $\chi''_{N}$ . While for



FIG. 3. Scaled  $\chi''/\chi_{eq}$  (or  $\chi''T$ ) vs T using data of Fig. 1 (see text). Conventional dynamic scaling has the temperature difference  $(T_B - T_A)$  between any corresponding pair of points such as (B,A), (B',A'), etc., varying as  $(T_B - T_A) \sim \omega^{1/zv}$ . Thus zv may be determined without any reference to  $T_c$  as shown in Fig. 2(b). Failure of curves to intersect at  $T_c$  is discussed in text.

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these different pairs of reference points the average value of zv is about 11.3, there is a systematic trend towards *larger* zv as the selected pair moves to lower T, close to  $T_c$ . Finally, since  $\chi_N^{"}$  should be independent of  $\omega$  at  $T_c$ , the convergence of the plots in Fig. 3 suggests  $T_c \sim 12.0$ K. If the data fully agreed with either scaling form, Eqs. (2) or (3), all the curves for  $\chi_N^{"}$  would cross at  $T_c$ . The imperfect crossing plus the systematic variation of zv will be discussed below.

From Fig. 3 one also determines that the data on the steepest parts of  $\chi_N''$  fit conventional but not activated scaling. Consider  $\Delta \chi_N'' \equiv \chi_N''(\omega, T) - \chi_N''(\omega/10, T)$ . For conventional scaling the maximum value of  $\Delta \chi_N''$  should be  $\omega$  independent, while for activated scaling it should vanish as  $1/|\ln\omega\tau_0|$  for low  $\omega$ . We find that it varies between 0.5 and 0.6, with no particular trend with  $\omega$ , thus again ruling against activated dynamics.<sup>8</sup> A previous suggestion<sup>7</sup> of activated dynamics above  $T_c$  in Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te was based on large zv obtained in a log-log plot scaling fit of  $\Delta \chi'$  to conventional dynamics and the presence of significant short-range type-III order (See Ref. 7 for appropriate references.) Our new more exacting method of analyses of  $\chi''$  rules against activated dynamics over most



FIG. 4. (a) Scaling of  $\chi''/\chi_{eq}$ . Similar results are found for  $\chi''T$ . Best parameters vary according to which region of temperature is emphasized. Region around peak of  $\chi''$  is emphasized here (see text). The symbols a, v, +, x, o, and e denote frequencies from 0.975 Hz to 97.5 kHz in decade steps. (b) Scaling of  $\chi_{eq} - \chi'/\chi_{eq}$  with the same parameters as (a).

of the critical temperature range except perhaps at lower T around  $T_c$ .

Guided by values of  $\beta/zv$ , zv, and  $T_c$  individually determined above, the full scaling plots for  $\chi''$  and  $\Delta \chi'$  are presented in Fig. 4. The indicated parameters give the best scaling in the range 0.9L to 0.6R for  $\chi_N''$  where the average departure from good scaling is < 3%, comparable to experimental error. However, note the departure from scaling at lower T where it is found that progressively larger values of zv and lower  $T_c$  are needed for a better fit, corresponding to the absence of a sharp crossing in Fig. 3. We find almost identical results when a similar analysis<sup>9</sup> is performed on the published data of Gunnarson et al.<sup>10</sup> for Fe0.5Mn0.5TiO3, a supposed Ising spin glass and Svedlindh et al.<sup>11</sup> for (Fe<sub>0.15</sub>Ni<sub>0.85</sub>)<sub>75</sub>B<sub>16</sub>P<sub>6</sub>Al<sub>3</sub>, an amorphous metallic spin glass. In both cases we find a lower  $T_c$ and higher zv where the best parameters are respectively  $\beta/zv = 0.05$  and 0.06; zv = 11.5 and 10.3;  $T_c = 20.5$  and 21.9.

Often-made claims of complementarity of static and dynamic scaling data in yielding the same values of  $T_c$ and other critical parameters are suspect due to the correlation between the parameters, the wide choice of fits possible with the log-log plots, and the tendency to overestimate  $T_c$  in any experiment because of lack of equilibrium due to the inordinately long relaxation times near  $T_c$ . For example, in a reanalysis<sup>9</sup> of the published nonlinear susceptibility,  $\chi_{\rm NL}$  data for Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te (Ref. 12) using a *linear* scaling plot of  $\chi_{\rm NL}/H^{2\beta/(\gamma+\beta)}$  vs  $t/H^{2/(\gamma+\beta)}$ , we find at least as good a fit with  $\gamma$  = 4.4,  $\beta$  = 0.6, and  $T_c$  = 12.14 compared to  $\gamma$  = 3.3,  $\beta$  = 0.9, and  $T_c$  = 12.37 in Ref. 12. Levy<sup>13</sup> finds  $\gamma$  = 4.8 in Cd<sub>0.75</sub>Mn<sub>0.25</sub>Te.

One explanation for the deviations from scaling near and below  $T_c$  seen here may be a lack of complete equilibrium. Excess values of  $\chi''(\omega)$  as a function of waiting time  $t_w$  have been observed<sup>14</sup> well below  $T_c$  for low  $\omega$  but the situation is ambiguous at  $T_c$ . The departures from scaling seen here for 1 Hz at low T are  $\sim 15\%$  and correspond to a phase angle of  $\sim 0.04^{\circ}$ . A  $t_w$  of many hours and a phase stability of  $\lesssim 0.02^{\circ}$  would be needed during this long time to check for a decrease of  $\chi''$  with  $t_w$ . While we could maintain phase stability of  $\lesssim 0.01^{\circ}$  for  $\sim 15$ min, the low S/N in Cd<sub>0.6</sub>Mn<sub>0.4</sub>Te, due to the small  $\chi$  and  $h_{\rm ac}$ , prevented 0.02° stability for the needed hours to check for nonequilibrium. However a  $t_w$  of 15 min is  $\sim 10^3 \tau_p$  where  $\tau_p$  is the characteristic time at  $\chi_p''$  for 1 Hz  $(\tau_p \sim 1/\omega)$  so that equilibrium is presumably attained at  $\chi_p''$  even for 1 Hz. Thus our determination of  $\beta/zv$  or zvnear and above  $T_{\rho}(\omega)$ , and thus well above  $T_{c}$ , is in no way affected, as may also be seen by the fact that the slopes in Fig. 2 would be essentially the same even if the lowest  $\omega$  were neglected.

Another possible reason for the observed deviations from scaling may be strong corrections to scaling due to being near the lower critical dimension. This explanation has been advocated for qualitatively similar systematic deviations from finite-size scaling seen in simulations of the  $\pm J$  Ising model.<sup>15</sup> Yet another possibility is that at higher T the systems appear to be headed for a conventional transition and then cross over to some other behavior at lower T. While these deviations from scaling are interesting, the main focus here is the new method of separately determining  $\beta/zv$ , zv, and  $T_c$  from  $\chi''$ . This procedure has also allowed us to distinguish between conventional versus activated dynamics and has revealed a frequent underestimate of zv and overestimate of  $T_c$ . The linear, as opposed

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to logarithmic, scaling plots suggested by Eqs. (2) and (3) should clearly be used in any careful analysis of critical behavior in random magnets.

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