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In amorphous GdAl spin glass, as in a number of other spin glasses, the susceptibility cusp first increases, then decreases in temperature with increasing field. An explanation for this behavior is found in terms of a competition between the noncritical linear susceptibility and the critical nonlinear susceptibility.

Early lore about spin glasses held that the temperature of the susceptibility cusp shifts *down* with increasing magnetic field. Nevertheless, even the early results of Cannella and Mydosh[1] on AuFe spin glasses showed the opposite tendency. More recently, our own studies[2] of the dc susceptibility of CuMn and amorphous GdAl spin glasses also showed the peak temperature shifting up with field. Yeshurun[3] found a similar effect in an amorphous FeMnPBAl spin glass. Results of a more complete study[4] on our amorphous GdAl sample[5] are shown as circles in Fig. 1(a). The peak temperature T_P first increases with field H, but later decreases as the peak broadens. In terms of a reduced temperature $t = (T - T_G)/T_G$, with the zero-field transition temperature T_G at 15.5 K for our sample, the initial shift with field follows $t = 0.0061H^{0.50}$.

Ours is dc equilibrium data, while most other studies have been done with ac techniques, which introduce nonequilibrium effects. Aside from our GdAl and CuMn results,[2] and those of Yeshurun,[3] the only other detailed dc equilibrium study of the field dependence is that of Chamberlin *et al.*[6] on a AgMn spin glass, where T_P *decreases* with H. We interpret the present results as indicating that both behaviors are possible, depending on the material, and a satisfactory theory should explain why.

In this paper we show how a non-mean-field scaling theory can explain the remarkable behavior of Fig. 1. As described in more detail elsewhere,[4,7] such a theory predicts a singular contribution $\chi_{\rm sing}$ to the equilibrium susceptibility within a critical region near and above the phase transition temperature T_G. In terms of the reduced temperature $t = (T - T_G) T_G$, scaling exponents β, ϕ, δ, and γ, and a scaling function g, the singular susceptibility has the form

$$\chi_{\rm sing} = t^{\beta} g(H^2/t^{\phi}), \tag{1}$$

$$g(x) \to g_0 - g_1 x, \text{ for } x \to 0,$$

$$g(x) \to -g_2 x^{1/\delta}, \text{ for } x \to \infty,$$

$$\delta = \phi/\beta,$$

$$\gamma = \phi - \beta = \beta(\delta - 1),$$

where g_0, g_1, and g_2 are constants. Therefore, for H fixed, $\chi_{\rm sing}$ has the limits

$$\chi_{\rm sing} \to g_0 t^{\beta} - g_1 H^2 t^{-\gamma}, \text{ for } t \to \infty, \tag{2}$$

$$\to -g_2 H^{2/\delta}, \text{ for } t \to 0.$$

Plots of $\chi_{\rm sing}/H^{2/\delta}$ vs $t/H^{2/\phi}$ [which is an equivalent way to test Eq. (1)] have indeed shown scaling.[7] A specific form for $\chi_{\rm sing}$ which is consistent with Eqs. (1) and (2) and which smoothly interpolates between the limits is[8]

$$\chi_{\rm sing} = -g_1 H^{2/\delta}/[(t/H^{2/\phi}) + (g_1/g_2)^{1/\gamma}]^{\gamma}. \tag{3}$$

In the low-field limit, this form is regular in H^2 as indicated by Eq. (2), and in the high-field or low-temperature limit it is regular in t. Both these properties are important in achieving an accurate fit to the data, to be described below. It should also be mentioned that the g_0 term of Eq. (2) is ignored in Eq. (3), because it can be shown to be negligibly small in fits to the data.[9]

Next we ask, can the behavior of the susceptibility peak be understood from Eqs. (1) or (3)? Differentiating Eq. (1), we find the condition for an extremum:

$$\beta g(x) = \phi x g'(x), \quad x = h^2 t^{-\phi}. \tag{4}$$

Any real solution of this equation corresponds to an extremum line following $t \propto h^{2/\phi}$. That is, such a line is governed by the same exponent ϕ which describes the crossover between the low or high temperature limits of Eq. (1). However, the existence of such an extremum line depends on the specific form of g. We have recently argued elsewhere[4] that on physical grounds, g_0, g_1, and g_2 should all be positive for $t > 0$. In this case, it is easy to see from Eqs. (3) and (4) that there is no solution at all for $t > 0$. This conclusion is in contradiction to the observed tendency of the peak to shift to higher t in our sample.

The resolution of this difficulty, we believe, lies in recognizing the importance of regular (nonsingular) contributions to the susceptibility. From a physical point of view, such contributions represent spins or clusters of spins which do not participate in the critical fluctuations. From the point of view of analyzing experimental susceptibility data, they play a more important role in spin glasses than in ferromagnets for two reasons. (1) The singular behavior for a ferromagnet occurs in the linear susceptibility, while for a spin glass it occurs in the nonlinear susceptibility. Determination of the nonlinear susceptibility is more subtle, requiring proper subtraction of the linear terms and greater experimental
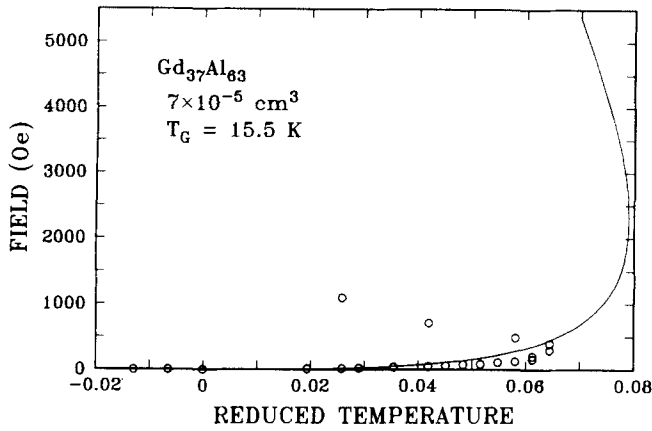
(a)
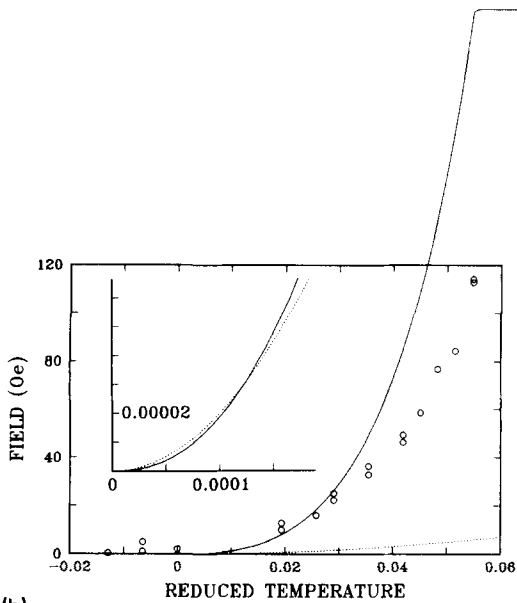
(b)

FIG. 1. Susceptibility peak position (points) for a $Gd_{37}Al_{63}$ amorphous spin glass. Parts (a) and (b) and the insert show different scales. The reduced temperature is $(T - 15.5\ K)/15.5\ K$. The solid line is the predicted line of maxima, from Eq. (3) and fit 1 of the table. The dotted line is the crossover line $t = \{g_1/g_2\}^{1/\gamma}H^{2/\phi}$.
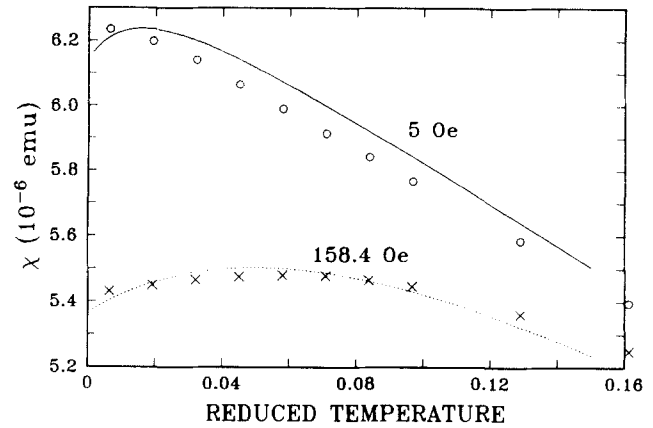

FIG. 2. Susceptibility vs reduced temperature (points) for the GdAl sample at two fields. Points are experimental results and lines are predictions of Eq. (3) for the parameters of fit 1 of the table. Dividing χ by the sample volume of 7×10^{-5} cm^3 gives cgs units of emu/cm^3.

accuracy. (2) So far susceptibility peaks of spin glasses have been found smeared $(\Delta t \geqslant 1\%)$ in temperature, presumably because of inhomogeneity problems characteristic of these systems. On the other hand, there are reasons to expect their critical region to be broader than in ferromagnets.[7] Therefore, the data on critical behavior of spin glasses comes from a wider temperature region relative to the transition temperature than in ferromagnets. In this wider temperature region, variation of the regular susceptibility is more substantial.

We expect the regular susceptibility to have a Curie-like behavior plus possible H^2 corrections. With such terms, χ_{reg} can be expanded in a series

$$\chi_{reg} = \chi_p - a_1 t - a_2 H^2 - a_3 H^2 t - a_4 t^2 \ldots . \quad (5)$$

Since the coefficients a_i are not known a priori, one should in principle include all of them in fitting the data. Given the many parameters already needed to characterize the singular susceptibility, this is impractical. We have used χ_p and a_1 in conjunction with Eq. (3) to obtain the fit 1 of Table I. These parameters imply $\beta = 0.7$ and $\phi = 4.0$. To obtain

TABLE I. Least-squares nonlinear fits of $\chi_{reg} + \chi_{sing}$ [Eqs. (3) and (5) of text] to dc susceptibility of 7×10^{-5} cm^3 of $Gd_{37}Al_{63}$ spin glass[7] in range $15.5 \leqslant T$ (K) $\leqslant 18$, $0.5 \leqslant H$ (Oe) $\leqslant 2000$. All nonexponent units in 10^{-6} emu (i.e., emu/cm^3 after dividing by the volume). A dash indicates this term not used in the fit. MSQD is mean square deviation, that is, the square root of chi-squared divided 240, the number of points in the fit.

Fit No.:	1	2	3	4
χ_{reg}:				
χ_p	6.47 ± 0.01	6.47 ± 0.01	6.47 ± 0.01	6.53 ± 0.01
a_1	6.43 ± 0.07	6.38 ± 0.08	6.40 ± 0.08	7.9 ± 0.2
a_2	...	$-(6 \pm 3) \times 10^{-8}$	...	...
a_3	...	...	$-(2 \pm 2) \times 10^{-7}$	...
a_4	...	...	...	-8 ± 1
χ_{sing}:				
γ	3.3 ± 0.1	3.3 ± 0.1	3.3 ± 0.1	3.2 ± 0.1
δ	5.6 ± 0.1	5.5 ± 0.1	5.6 ± 0.1	6.1 ± 0.1
g_1	6.9×10^{-7}	7.2×10^{-7}	7.1×10^{-7}	5.6×10^{-7}
g_2	0.19 ± 0.01	0.17 ± 0.01	0.18 ± 0.01	0.22 ± 0.01
MSQD	0.0323	0.0321	0.0323	0.0294

some measure of the importance of the other terms in Eq. (5), we have done fits with each of the remaining parameters in turn, giving the fits 2–4 in the table. We find that the mean square deviation, the critical exponents and coefficients are not significantly affected. The $-a_1 t$ term in the regular susceptibility of Eq. (5) is the most important regular term, and in what follows we ignore the higher order terms. Since an expansion of the Curie law near T_G gives $\chi_p \, T_g/(T_g - \theta)$ for a_1, the close similarity of χ_p and a_1 in the table implies $\theta = 0$. Although the paramagnetic Curie temperature for our sample has previously been reported[10] to be of order 30 K, our result is not inconsistent because of well-known deviations from the Curie law near a spin glass transition.[11]

The presence of the regular $-a_1 t$ term significantly modifies our earlier conclusions about the conditions for extrema. In the limit of low field [or large t in Eq. (2)] we now find a maximum at

$$t_{\max 0} = (\beta g_0/a_1)^{1/(1-\beta)}. \tag{6}$$

Thus, surprisingly, in the $H \to 0$ limit and for nonzero g_0, the susceptibility peak is not precisely at the transition but slightly above it. In practice, however, g_0 is negligibly small and the shift is experimentally unobservable considering our temperature smearing of order $\Delta t \sim 2 \times 10^{-2}$.

At higher field, but providing $t > (g_1/g_2)^{1/\gamma} H^{2/\phi}$ (the crossover line), the peak position rises with a vertical tangent in the $H \cdot t$ plane and then rolls over to follow the line

$$t = (\gamma g_1/a_1)^{1/(\gamma + 1)} H^{2/(\gamma + 1)}, \tag{7}$$

obtained by differentiating the g_1 and a_1 terms of Eqs. (2) and (5). Such a formula was found earlier by Bouchiat,[12] but with a sign reversal which put the peak below T_G (i.e., $t < 0$). Plugging in the parameters of fit 1, we find $t = 0.032 H^{0.47}$, in approximate agreement with our experimental observation. However, as long as $2/(\gamma + 1)$ is less than $2/\phi$, i.e., as long as $\beta < 1$, Eq. (7) will cross the dotted crossover line as shown in the inset to Fig. 1(b). This happens at very small t and H for the parameters of fit 1, and at higher H Eq. (7) breaks down. Direct calculation from Eq. (3) and (5) using the parameters of fit 1 gives the full contour of susceptibility maxima shown in Fig. 1(a). The temperature shift of the susceptibility maxima reaches a maximum at $t \sim 0.08$ and $H \sim 2000$ Oe. While this is a factor of 5 off in field, the overall shape of the behavior is in qualitative agreement with the experiment. The quantitative discrepancy is most likely due to approaching

the high-field boundary of the critical region. Figure 2 shows a comparison of experimental and calculated $\chi(t)$ curves, with approximate agreement for the maxima even when the susceptibility peak becomes very broad. In other materials where the upward shift of the susceptibility peak has not been observed,[6] we suppose that the parameters g_1, g_2, etc., are such as to cause the upward shift to be small and to occur at low fields.

In summary, we find that a non-mean-field scaling theory explains in a semiquantitative way the observed tendency of the equilibrium susceptibility maxima to shift first up, and then down with increasing field. Although nonequilibrium Monte Carlo calculations have shown such an effect,[13] so far ours is the only *equilibrium* theory of spin glass behavior to do so.

[1]V. Cannella and J. A. Mydosh, Phys. Rev. B **6**, 4220 (1976).

[2]B. Barbara, A. P. Malozemoff, and Y. Imry, Physica B **108**, 1289 (1981).

[3]Y. Yeshurun (private communication).

[4]B. Barbara and A. P. Malozemoff, J. Less Common Metals **94**, 45 (1983).

[5]For details on the sample, see A. P. Malozemoff, and Y. Imry, Phys. B **24**, 489 (1981).

[6]R. V. Chamberlin, M. Hardiman, L. A. Turkevich, and R. Orbach, Phys. Rev. B **25**, 6720 (1982).

[7]B. Barbara, A. P. Malozemoff, and Y. Imry, Phys. Rev. Lett. **47**, 1852 (1981); A. P. Malozemoff, B. Barbara, and Y. Imry, J. Appl. Phys. **53**, 2205 (1981).

[8]In Refs. 4 and 7 we used a slightly different form which, in the $t \to 0$ limit, gave a correction irregular in t. The mean square deviations of those earlier fits were about double those in the table of this article. The form of Eq. (3), used in this paper, avoids the irregularity in t and gives a slightly better fit to the data with parameters not significantly different from those obtained earlier (see Table I).

[9]See Ref. 4, where g_0 was shown to be small. In Ref. 7, g_0 appeared anomalously large because the temperature dependence of the regular susceptibility was mistakenly neglected.

[10]T. Mizoguchi, T. R. McGuire, S. Kirkpatrick, and R. J. Gambino, Phys. Rev. Lett. **38**, 89 (1977).

[11]A. F. J. Morgownik, J. A. Mydosh, and C. van Dijk, J. Magn. Magn. Mater. **31-34**, 1334 (1983).

[12]H. Bouchiat, J. Phys. C **16**, 445 (1983).

[13]K. Binder and W. Kinzel, Proceedings of the Heidelberg Colloquium on Spin Glasses, May 30-June 2, 1983, Springer Lecture Notes (to be published).