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SQUID measurements of susceptibility χ versus field H are fitted by $\chi = \chi_0 - \chi_1 H^{a-1}$ at a given temperature T, for two classic spin-glasses: 4.6% Mn in Cu (polycrystalline) and 37% Gd in Al (amorphous). $\chi_1(T)$ shows a much stronger anomaly than $\chi_0(T)$, while $a(T)$ has a minimum at the freezing temperature T_f. The data can also be scaled according to $\chi_0 - \chi = H^{2/\delta} f((T - T_f)/H^{2/\varphi})$ with $\delta = 5 \pm 1$ and $\varphi = 4.5 \pm 0.5$.

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In this paper we study the critical behavior of the nonlinear magnetic susceptibility near the freezing temperature T_f of two well-known spin-glasses, polycrystalline 4.6% Mn in Cu[1-6] and amorphous 37% Gd in Al.[5-7]

A great deal of attention has of course been focused on the cusp at T_f in the linear susceptibility χ_0, that is, the zero-field limit of the susceptibility $\chi(H)$. However, it has been recognized that in many theoretical models,[8-16] the strongest critical behavior occurs not in χ_0, but in the nonlinear susceptibility $\chi_0 - \chi$. This has been experimentally demonstrated first by Miyako *et al.*[17] using an ac technique on PtMn and $(Ti_{1-x}V_x)_2O_3$, and more recently by Monod and Bouchiat[18] using dc measurements on AgMn. In this paper we also show the effect, but in contrast to these authors who *assumed* that $\chi - \chi_0$ followed an H^2 dependence, we consider a more general form H^{a-1} with the exponent $a(T)$ varying as a function of temperature. This is an important difference since Toulouse and Gabay[15] have shown that $a(T)$ should vary through the freezing in a mean-field model, and, in any case, our data do not permit an H^2 fit near T_f.

Furthermore, we use a scaling form proposed earlier[11] for the nonlinear susceptibility and show that the data obey it reasonably well. This is the first experimental demonstration of scaling in both H and T for the spin-glass susceptibility (earlier scaling by Salamon, Rao, and Yeshurun[19] for the paramagnetic-to-spin-glass transition considered only temperature and composition, not field).[20]

The magnetization measurements have been performed with use of a SHE VTS-50 SQUID magnetometer, in which fields from 1 to 50 000 Oe can be applied. Further details on the samples and apparatus are given elsewhere.[5,6] An important point is that the measurements were dc with a long waiting period (6 min) between data points, so that magnetization appears to be in equilibrium.[5,6] By contrast, the ac data of Miyako *et al.* are, as they themselves have shown,[16] frequency dependent. Thus, they are not in thermodynamic equilibrium, and this can have a drastic effect on comparison with thermodynamic theories particularly below T_f.

In a first step to analyze the data, which have been described earlier,[6] we use the field expansion of the magnetization:

$$M(T)/H = \chi_0(T) - \chi_1(T) H^{a(T)-1}, \quad (1)$$

where the exponent $a(T)$ has to be determined at different temperatures T. We have determined $\chi_0(T)$ for both samples by a roughly linear extrapolation down to zero field of the measured M/H vs H curves between 50 and 2 Oe. Then a log-log plot of $\chi_0 - M/H$ vs H allows us to determine at each temperature $\chi_1(T)$ and $a(T)$. Results for GdAl are shown in Fig. 1; MnCu gives similar results. The variation of $\chi_1(T)$ is very rapid, especially if compared to $\chi_0(T)$, and its maximum determines the phase transition temperature T_f. By contrast, $\chi_0(T)$ does not appear to be singular [note offset of scale in Fig. 1(a)], as suggested by Toulouse and Gabay.[15] $a(T)$ equals 3 for both samples at high temperatures, as normally expected. Below 20 K in GdAl and 30 K in MnCu, $a(T)$ decreases down to, respectively, 1.5 ± 0.1 and 1.8 ± 0.1 at the temperature T_f. Below T_f it increases again. This similar behavior in the two samples supports the mean-field results of Toulouse and Gabay,[15] who predicted a minimum of $a(T)$ at T_f. Our experimental values are not far from their prediction of $a = 3$ above T_f, $a = 2$ at T_f, and $a = \frac{7}{5}$ below T_f. While the value of 3 at high temperature is a consequence of analyticity and symmetry and must be true, the about 25% discrepancy in $a(T_f)$ may be associated with deviations from the mean-field approximation used in the theory. However, there is also concern that even though we used very low fields in our extrapolation for χ_0, higher terms in the field

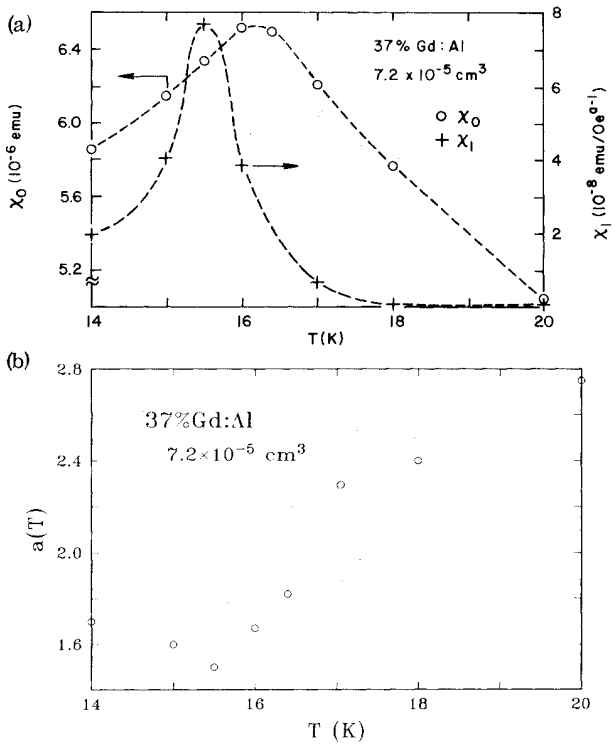

FIG. 1. Results of fits by Eq. (1) for GdAl sample: (a) linear susceptibility $\chi_0(T)$ (note scale offset!) and nonlinear susceptibility coefficient $\chi_1(T)$, and (b) nonlinear susceptibility exponent $a(T)$ (error bars ± 0.1).

expanison might contribute, leading to an underevaluation of $a(T)$. To avoid these problems, we next consider a more general scaling approach to treat the data.

From a theoretical point of view, the first observation of the role of the nonlinear susceptibility $\chi_s = \chi_0 - \chi$ in a spin-glass-like phase can be made in the context of the Mattis model.[8] There, it can be shown[9,10] that the second derivative of the regular susceptibility χ with respect to the magnetic field diverges like $t^{-\gamma}$, where γ is the (spin-glass) order-parameter susceptibility exponent and $t = (T - T_f)/T_f$, with T_f the spin-glass transition temperature. This relation has also been argued to be valid for more general Edwards-Anderson spin-glass models,[11,12] and is also valid in the mean-field approximation.

On the other hand, Chalupa[11] has shown that for the Edwards-Anderson spin-glass at T_f, the singular part χ_s should go as $H^{2/\delta}$, where δ is again derived from the relation between the spin-glass order-parameter q and its conjugate field h_q, $q \sim h_q^{1/\delta}$ at T_f. The above limiting forms suggest the more general scaling relation for $\chi_s \equiv \chi_0(T) - M/H$:

$$\chi_s(H,t) = H^{2/\delta} f(t/H^{2/\varphi}), \tag{2}$$

where

$$f(x) = \text{const}, \quad x \to 0;$$
$$f(x) = x^{-\gamma}, \quad x \to \infty; \tag{3}$$
$$\varphi = \gamma\delta/(\delta - 1) \equiv \beta\delta.$$

The scaling form Eq. (2) also follows from the more general one proposed by Suzuki earlier.[12] This kind of scaling near the transition would follow in a renormalization-group calculation if near the fixed point H would scale with some power of the length. Thus, measuring the behavior as a function of H is the only handle on exponents that are nonthermal.

At $t = 0$ ($T = T_f$), Eq. (2) is the same as Eq. (1), and as will be seen below, the $2/\delta$ exponents indeed come out close to $(a-1)_{t=0}$ (the values range from 0.2 to 0.5). But away from T_f, Eq. (2) predicts a different H dependence which approaches $H^{2/\delta}$ only at high fields and yields the crossover between the different values of $a(t)$. In fact, the data are not described by a simple power law, if a large field range is included, and therefore Eq. (2) is a better representation of a larger range of data than Eq. (1), as will be seen below.

We have fitted the nonlinear susceptibility, i. e., the difference $\chi_s = \chi_0 - \chi$, with Eq. (2) by plotting $\log[\chi_s(t)/H^{2/\delta}]$ vs $\log(t/H^{2/\varphi})$ for a very large number of different values of the pair (φ, δ). T_f was taken at the maximum of $\chi_1(T)$. The result was generally a very broad distribution of points lying in one plane without any obvious correlation. However, for the case $T > T_f$, these points converged towards a single curve as shown in Fig. 2 for the two samples, for a relatively narrow range of values of δ and φ:

$$5.9 \leq \delta \leq 6.3, \quad 3.5 \leq \varphi \leq 4.5 \text{ (37\% GdAl)};$$
$$4.0 \leq \delta \leq 4.3, \quad 4.5 \leq \varphi \leq 5.5 \text{ (4.6\% MnCu)}.$$

The observed clustering is quite remarkable especially if one considers the wide ranges of field (2–10 000 Oe) and temperature ($T_f - 2T_f$), which are consistent with the assumption of a large critical region. Furthermore, the curves and exponents are quite similar for two very different spin-glasses. Scaling can also be achieved below T_f, but these results as well as an investigation of the size of the critical region will be discussed elsewhere.

Using Eq. (3), we get the third exponent $\gamma = 3.8 \pm 0.5$ for GdAl and 3.4 ± 0.4 for MnCu. These high

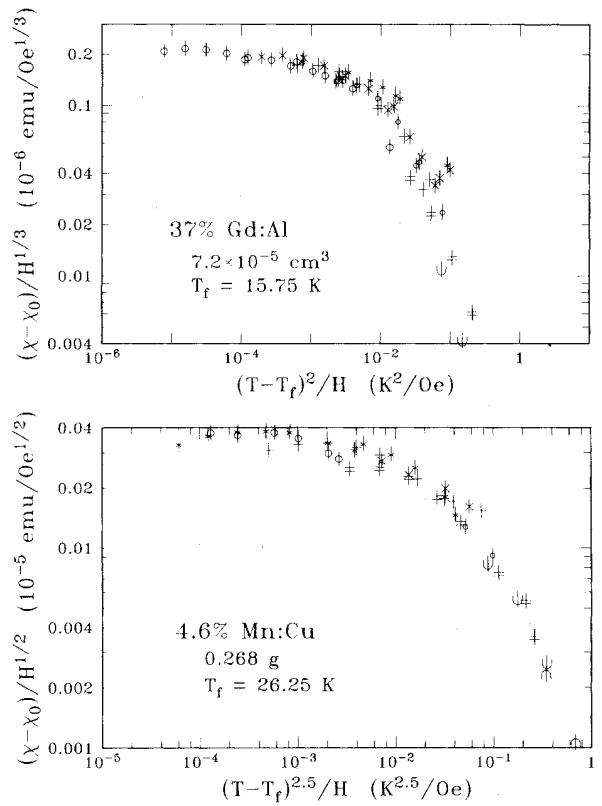

FIG. 2. Scaling fits according to Eq. (2) for the nonlinear susceptibility as a function of field and temperature above T_f. For GdAl and MnCu samples, respectively, large circles represent data at 16 and 26.5 K; small ×'s, 16.4 and 27; large ×'s, 17 and 27.5; crosses, 18 and 28; small circles, 20 and 29; equals signs, 30 and 30; ∪'s, 40 and 40; and ∩'s, 50 and 50.

exponent values imply a very rapid thermal variation of $\chi_s(t) \propto H^2 t^{-\gamma}$, even near T_f if H is small enough. These large γ values can be compared with the value 1.5 ± 0.5 obtained by Monod and Bouchiat.[18]

The fitting of the data with the scaling relation of Eq. (2) implies that the nonlinear susceptibility in these two spin-glasses appears to diverge like $t^{-\gamma}$ as T_f is approached from above with sufficiently low field, and that it goes as $H^{2/\delta}$ at T_f. The values of the exponents appear to be $\gamma = 3.6 \pm 0.6$, $\delta = 5 \pm 1$ for both systems. The mean-field values[15] for these exponents are $\gamma = 1$, $\delta = 2$ while the first-order results using an ϵ expansion[21] about dimensionality $d = 6$ yield $\gamma = 2.8$, $\delta = 3.2$ for three dimensions. Of course, the ϵ expansion is not reliable for three dimensions, i.e., $\epsilon = 3$, but the trends and the substantial changes from the mean-field theory appear to be reasonable.

These results support the possibility of a second-order phase transition at T_f. We still cannot rule out a small intrinsic width of this transition. Such a width is possible if T_f is related to a fixed point in the renormalization-group formulation which is not stable but rather weakly unstable so that at least an apparent transition follows. Still, the observed width seems to be totally accounted for by sample inhomogeneities.[5]

The scaling analysis presented here also has further thermodynamic consequences. A well-known thermodynamic relation,

$$\delta^2\chi/\delta T^2 = T^{-1}\delta^2 C_v/\delta H^2, \tag{4}$$

where C_v is the specific heat at constant volume, means that the curvature of $\chi(T)$ should be given by $1/T$ times the (temperature-dependent) coefficient of H^2 in the expansion of $C(H, T)$ in powers of H. Fogle *et al.*[22] have recently found that this relation is not satisfied when the measured slowly field-cooled *linear* $\chi_0(T)$ is used. This was interpreted as a nonergodicity of the system. However, from our scaling relation Eq. (2) we find that for small H at fixed T, the nonlinear part of χ gives

$$\delta^2\chi_s/\delta T^2 \propto t^{-\gamma-2}H^2. \tag{5}$$

This implies an H^4 term in $C_v(T)$ which increases very fast as $t \to 0$ (eventually when $t \to 0$ this crosses over to the relation for constant H and $t \to 0$). Thus, to effect a meaningful check of Eq. (4), one should keep the $O(H^4)$ terms and the nonlinear χ_s.

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Thermodiffusion of High-Density Electron-Hole Plasmas in Semiconductors

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The spatial distributions of temperature and density in electron-hole plasmas in surface-excited semiconductors are investigated with use of linear irreversible thermodynamics and a microscopic plasma theory. Above a certain threshold the density distribution is dominated by a characteristic density, which increases with temperature. Experimental results for Ge, unstressed Si, and Si under high uniaxial stress are in agreement with the theory.

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Contrary to the well understood phenomenon of electron-hole liquid (EHL) condensation,[1] the properties of the charge-carrier system above the critical point for liquid formation are discussed rather controversely to date: In Ge the luminescence band has been assigned to the overlapping emissions of a low-density electron-hole plasma (EHP), free excitons (FE), and of various higher-order bound states.[2] In GaAs the emission band was ascribed to a high-density EHP and was observed up to temperatures of 70 K.[3] A surprisingly weak dependence of the plasma density n_{EHP} on the excitation intensity and an increase of n_{EHP} with temperature were reported.[3] Recent experiments on Si reported similar results as obtained for GaAs and displayed no indication of noticeable densities of higher-order complexes.[4] Furthermore it was demonstrated that the band structure influenced the plasma via the density of states masses,[4] which ruled out earlier speculations on a correlation of the plasma density and the density of the metal-insulator transition.[3]

In spite of the contradicting conclusions drawn from the individual experiments, a direct comparison of the high excitation spectra obtained from the very different semiconductors Ge,[2] Si,[4] GaP,[5] CdS,[6] and GaAs[3] displays striking resemblances: At temperatures above the critical point for liquid formation a broad "liquidlike" emission is observed in the energy range just below the FE. Equilibrium thermodynamics, however, gives no indication for the occurrence of distinguished states above the critical temperature. Nonequilibrium phenomena, on the other hand, have been discussed so far mainly in the context of EHL nucleation and under isothermal conditions.[6,7]

In this Letter we present the basic features of a new approach to describe the properties of high-

