Phase-Transition Behavior in a Random-Anisotropy System

D. J. Sellmyer and S. Nafis
Behlen Laboratory of Physics, University of Nebraska, Lincoln, Nebraska 68588
(Received 18 February 1986)

The character of the magnetic transition is studied for a magnetic glass with strong random-anisotropy and ferromagnetic interactions. Field-dependent ac susceptibility measurements are presented near the ordering temperature of $Tb_{64}Fe_{20}Ga_{16}$. The data are analyzed with a scaling theory for the singular susceptibility. An excellent collapse of the H,T data is obtained for reduced temperatures in the range 0.002–0.13. These results, which are compared with recent theoretical work, provide strong evidence for a true phase transition in a random-anisotropy system.

PACS numbers: 75.50.Kj, 75.30.Gw, 75.40.−s

Among the most challenging and significant problems in condensed-matter and statistical physics is the development of an understanding of phase transitions in random and disordered systems. Particularly intriguing are those in which it is difficult, both experimentally and theoretically, to determine whether the putative transitions are true *phase* transitions based on equilibrium statistical mechanics, or *glass* "transitions" in which the relaxation times smoothly increase as the temperature is lowered, and finally become equal to the measuring time at T_g. These considerations apply to a rather wide variety of materials including spin-glasses (SG),[1] systems with random magnetic fields[2,3] or random magnetic anisotropies (RMA),[4] localized electron glasses,[5] ferroelectric glasses,[6] and metastable charge-density–wave states.[7] Such systems tend to be characterized by the existence of metastable states, a high degree of ground-state degeneracy, and a difficulty in determining a satisfactory order parameter. In addition, it is often hard to ascertain a diverging susceptibility or relaxation time which would clearly signal a phase-transition temperature.

The character of the transition exhibited by spin-glasses has been debated at length. At present there seems to be considerable evidence from both theory[8] and experiment[1] that the phase-transition picture is relevant to spin-glasses, even though magnetic relaxations over energy barriers must be accounted for at temperatures below and even somewhat above T_c.[1] In comparison with spin-glasses, the transitions exhibited by random-field and, especially, random-anisotropy systems have been much less studied.[4,9] In this paper we present compelling evidence for the validity of a phase-transition description of the spin freezing in an amorphous material with strong RMA: a-$Tb_{64}Fe_{20}Ga_{16}$. Critical exponents are determined by a scaling analysis and the behavior in an applied field also is discussed.

a-TbFeGa develops a random, spin-glass-like order below a temperature $T_g \cong 141$ K. The origin of the randomness is RMA associated with electric field gradients interacting with the Tb^{3+} ions. Were it not for the strong RMA, the magnetic order would be ferrimagnetic, as is the case for the analogous glass without RMA: a-$Gd_{64}Fe_{20}Ga_{16}$. In the Gd glass, Cornelison and Sellmyer[10] have shown that the susceptibility diverges at a critical temperature, T_c, below which a ferrimagnetic structure obtains. Thus, despite the rare-earth (RE)-Fe exchange interactions which result in antiparallel RE and Fe moments, there are strong *ferromagnetic* correlations (RE/RE and Fe/Fe) in both the Tb and Gd glasses.[10]

The glasses were prepared by rapid quenching from the melt. High-resolution x-ray diffraction measurements confirmed the amorphous structure of the samples. In previous work[10] we have studied the magnetization of a-TbFeGa in fields up to 80 kOe and temperatures down to 1.5 K. However, the present studies are confined to low-field ac susceptibility measurements at and above T_g. The frequency was varied from 10 to 10^4 Hz, but mostly was held at 280 Hz. The amplitude of the ac field was about 0.1 Oe and a dc field, H, parallel to H_{ac}, was applied.

In previous work on strong RMA glasses we and others have noted several similarities in the transitions exhibited by RMA and SG systems.[11] The dc susceptibility in the field-cooled and zero-field–cooled states shows common features. Both types of material exhibit low-temperature magnetic viscosity effects which are qualitatively similar. In RMA magnetic glasses with strong ferromagnetic correlations (e.g., a-TbFeGa and a-$TbFe_2$) the correlation length[9] and susceptibility[4,10] can get large, but the freezing of the spins at T_g prevents the total susceptibility χ from diverging. The nondiverging susceptibility is another similarity of RMA and SG transitions. However, the sharpness of the transitions can be much more pronounced in RMA glasses. For example, in a-$Nd_{64}Co_{20}Ga_{16}$ we have observed a very narrow susceptibility peak at T_g for which $\delta T/T_g \cong 0.07$, where δT is the FWHM.[4] Theoretically, it was shown in the limit $D/J_0 \rightarrow \infty$, where D and J_0 are measures of the average RMA and exchange strengths, respectively, that the RMA energy density has the same form as the random-bond SG model with Ising symmetry.[12] For these and other reasons it is desirable to consider the *singular susceptibility*, $\chi_s \equiv \chi_0 - \chi$, where χ_0 and χ are the linear and total susceptibilities, respectively. For temperatures $T > T_g$, and in the small-field limit, the *nonlinear* susceptibility is defined by $\chi_{nl} = \chi_s / H^2$. These definitions follow from the theoretical results for the SG problem, first obtained by Suzuki.[13]

Figure 1 shows the ac susceptibility of a-TbFeGa for several applid dc fields ranging from 0 to 288 Oe. The lower part of the figure gives the temperature dependence of χ_s for several fields. In order to determine whether a phase-transition picture is relevant to this transition it is necessary to determine an appropriate order parameter, and then to have a scaling theory in terms of which critical exponents can be determined. For the RMA problem the order parameter can be chosen as $q = N^{-1} \sum \langle \mathbf{J}_i \rangle \cdot \langle \mathbf{J}_i \rangle$, which measures the correlation of a moment with its own direction.[14] As in the SG problem, it is the nonlinear susceptibility, χ_{nl}, which should exhibit critical behavior if there exists a phase transition. The static scaling hypothesis then implies for this case[1]

$$\chi_s = t^{\beta} f(H^2 / t^{\gamma+\beta}), \tag{1}$$

where $f(x)$ is the scaling function and $t \equiv (T - T_c)/T_c$ is the reduced temperature. The theory also gives

$$\chi_s \propto t^{-\gamma}, \quad t > 0, \tag{2}$$

$$\chi_s \propto H^{2/\delta} = H^{2\beta/(\gamma+\beta)}, \quad t \to 0^+, \tag{3}$$

with

$$\beta\delta = \gamma + \beta. \tag{4}$$

Equations (2) and (3) hold in the limit of low enough t and H.

Figure 2 shows the results of the scalng analysis. In this figure both the susceptibility and field have been corrected for demagnetization effects. That is, $\chi \to \chi_{ac}[1 - N_d \chi_{ac}]^{-1}$ and $H \to H_a - N_d M$, where χ_{ac}, N_d, and H_a denote the measured ac susceptibility, demagnetization factor, and applied field, respectively. N_d was determined from a ferromagnetic sample of the same shape as the a-TbFeGa sample and had the value 2.2 g Oe/emu. The demagnetization corrections were at most 10% of χ_{ac} and H_a. The range of reduced temperature is $0.002 \leq t \leq 0.13$. The critical exponents obtained are $\beta = 1.7 \pm 0.1$, $\gamma = 3.7 \pm 0.1$, and $T_c = 140.5 \pm 0.6$ K. The errors on β, γ, and T_c were estimated by our noting the sensitivity of computer-generated scaling fits to variations in these parameters. In addition, the uncertainty in T_c contains a contribu-

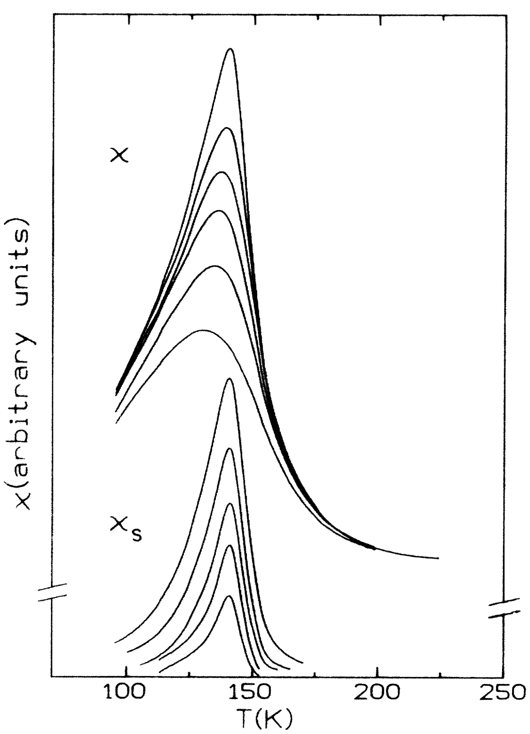

FIG. 1. ac susceptibility results at 280 Hz for a-$Tb_{64}Fe_{20}Ga_{16}$. The top set of curves gives the total susceptibility χ in dc fields of $H = 0$, 48, 78, 120, 180, and 288 Oe (top to bottom). The bottom set of curves gives the singular susceptibility χ_s for fields of 288, 180, 120, 78, and 48 Oe (top to bottom). The peak of χ corresponds to $0.10 N_d^{-1}$, where N_d is the demagnetization coefficient.

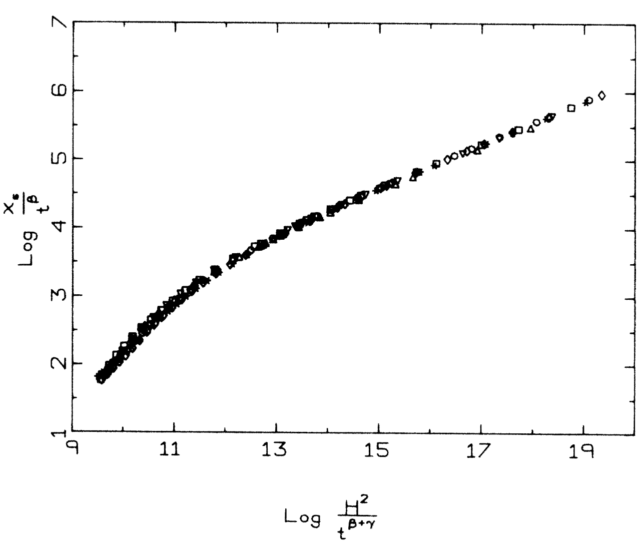

FIG. 2. Scaling of χ_s / t^{β} as a function of $H^2 / t^{\beta+\gamma}$. The fields are as follows: (inverted triangles) 40 Oe; (triangles) 78 Oe; (squares) 120 Oe; (circles) 180 Oe; (lozenges) 240 Oe; (stars) 288 Oe. χ_s and H in this plot are corrected for demagnetization effects as explained in the text.

tion (~ 0.3 K) due to possible systematic error in the thermometer calibration. From the scaling relation [Eq. (4)] it follows that $\delta = 3.2$. It should be emphasized that the quality of the scaling fit is excellent over about ten decades in the argument of the scaling function. Note also that the high-temperature slope of the curve (left-hand side) tends toward 1, and the high-field slope tends toward $\delta^{-1} = \beta/(\gamma+\beta)$, as expected from Eqs. (1) and (3). This scaling analysis is much more satisfying than earlier attempts[4,15] to understand transitions in similar materials because the previous methods to determine γ at a single field value broke down for $t \to 0$, precisely where one would expect critical behavior to be observed.

As seen in Fig. 1, the experimentally measured χ_s does not diverge as $t \to 0$. This is to be expected since it is the nonlinear susceptibility $\chi_{nl} = \chi_s/H^2$ which should diverge as $T \to T_c^+$. The intercept of a plot of $\chi_s H^{-2}$ vs H^2, as $H \to 0$, does, in fact, tend to diverge as $t \to 0$ and this is equivalent to, and confirmed by, the excellent scaling fit of the data shown in Fig. 2, over the reduced temperature range $\sim 10^{-3}$ to 10^{-1}. If the spin freezing were regarded as a glass "transition" one would expect that the frequency dependence of T_g would fit a Fulcher law or perhaps a power law indicating slowing down of the magnetic relaxations as $T \to T_g^+$.[1] For this reason the frequency dependence of χ was measured between 10 and 10^4 Hz. However, within our relative error of about 0.3 K ($2\times 10^{-3} T_g$), no frequency dependence of T_g could be detected. These results are thus consistent with the scaling theory for a phase transition in which χ_{nl} diverges at T_c.

Figure 1 shows clearly a decrease in the peak temperature at the maximum of χ, as the field increases. Earlier dc magnetization studies show the presence of irreversibility below the peak temperature.[10] If it is assumed that $T_g(H)$ represents a field-dependent transition line, then a phase diagram can be constructed as shown in Fig. 3. This indicates that the data fit an expression of the form

$$H = H_0[1 - T_g(H)/T_g(0)]^{\zeta}, \tag{5}$$

with $H_0 = 7003$ Oe, $T_g(0) = 140.5$ K, $\zeta = 1.23 \pm 0.05$. The expected behavior for $H(T_g)$ in the SG case has been the subject of much experimental and theoretical work. In the low-field, Ising-type limit of the theory[16,17] it is found that $\zeta = \frac{3}{2}$. In addition, Fischer and Zippelius[18] have shown that the high-temperature phase becomes unstable along a critical line similar to Eq. (5) with $\zeta = \frac{3}{2}$. These authors employed an infinite-range (mean-field) RMA model, with additional cubic anisotropy, in the limit of infinite anisotropy. At this stage, it is not clear whether any of these these theoretical results[16-18] are relevant to an RMA system such as the one under discussion here. More-

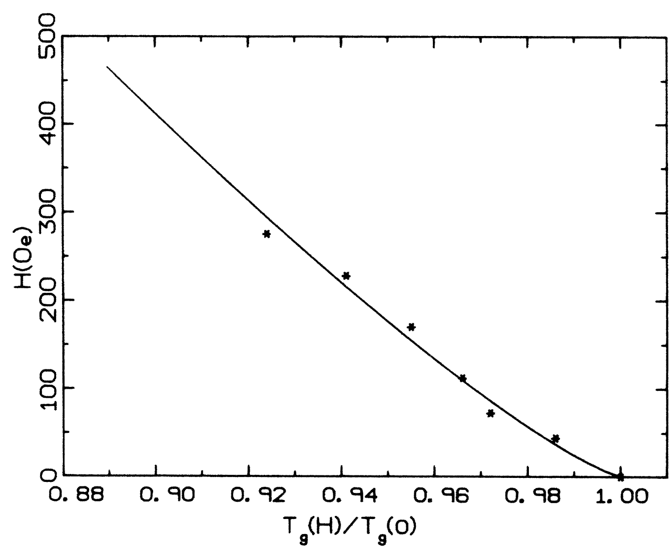

FIG. 3. $H(T_g)$ diagram showing field dependence of T_g at 280 Hz. The curve is that given by Eq. (5). H in this plot is corrected for demagnetization effects.

over, it should be noted that Eq. (1) predicts a field dependence like Eq. (5) for the peak in χ_s, with $\zeta = (\beta+\gamma)/2$. But Fig. 1 shows that there is essentially no shift of the peak of χ_s in a field. Thus, the results of Fig. 3 may reflect a field dependence of the linear susceptibility in a way that is not readily correlated with theory.

It is natural to ask whether there are theoretical results pertinent to the assumed phase transition and scaling results of Fig. 2. Aharony[19] was the first to study the critical behavior of the RMA Hamiltonian and he found, for space dimensionality $d \leq 4$, that no stable fixed point was approached. Pelcovits, Pytte, and Rudnick[20] showed that for $m \geq 2$, there is no long-range *ferromagnetic* order for $d \leq 4$, where m is the number of spin components. Thus the lower critical dimensionality for a ferromagnetic transition is 4. Aharony and Pytte[21] and Goldschmidt[22] showed that the susceptibility was limited for $T < T_c$ to $\chi_{max} \propto (J_0/D)^4$. This is in agreement with experiment which shows a nondiverging susceptibility for all strong RMA glasses.[4] Questions concerning the existence of spin-glass-like transitions or quasiferromagnetic[23] transitions (for small D) are much more subtle and controversial. In the $m \to \infty$ limit it was shown that there exists a spin-glass-like phase for $2 < d < 4$, for any nonzero D.[22,24] However, this result has been questioned by Fisher[3] who argues that the theoretical proof of a quasiferromagnetic of spin-glass-like state in the presence of RMA is an open question. Bray and Moore[25] concluded, on the basis of a numerical study, that in the $D \to \infty$ limit the RMA model displays an Ising SG transition at $T = 0$, for $d = 2$; this is consistent with the picture of a spin-glass-like transition in models with RMA. Recent numerical studies[8] on the Ising SG problem suggest a genuine phase transition at a nonzero temperature, for $d=3$. On the basis of this work Morgenestern[26] has conjectured that the RMA model with strong positive (ferromagnetic) exchange also should possess a phase transition. We are aware of no theoretical attempts to calculate the critical behavior of the nonlinear susceptibility of the RMA model, although Fähnle[27] has discussed the temperature dependence of an effective nonlinear suscepbitility exponent, on the basis of a correlated molecular-field theory.

In summary, we have presented clear evidence for the existence of a phase transition in a strong RMA glass. In order to test the generality of our analysis we have performed experiments on an independent system based on *a*-DyFeB. The results, to be published elsewhere are similar qualitatively and quantitatively to those presented here. We hope that these studies will stimulate further theoretical work on the phase-transition aspects of random-anisotropy and related systems.

We thank D. S. Fisher and Y. Goldschmidt for critical comments and advice on theoretical matters. R. D. Kirby, M. J. O'Shea, J. J. Rhyne, W. Saslow, and J. Souletie have provided us assistance and informative discussions. We are most grateful to the National Science Foundation for financial support under Grants No. DMR-8110520 and INT-8419546.

Note added.—After this paper was submitted, M. A. Feigel'man informed us of recent theoretical work[28] involving a model with long- (but finite-) range ferromagnetic interactions and totally uncorrelated random axes. The predicted behavior, which confirms the conclusions of the present paper, is that a crossover takes place between the preasymptotic "ferromagnetic" behavior and genuinely asymptotic critical behavior which is the same as for the Ising spin-glass.

[1]See J. Souletie, Ann. Phys. (Paris) **10**, 69 (1983); J. Souletie and J. L. Tholence, Phys. Rev. B **32**, 516 (1985), and references therein.

[2]See A. Houghton, A. Khurana, and F. J. Seco, Phys. Rev. Lett. **55**, (1985), and references therein.

[3]D. Fisher, Phys. Rev. B **31**, 7233 (1985).

[4]D. J. Sellmyer and S. Nafis, J. Appl. Phys. **57**, 3584 (1985), and references therein.

[5]J. H. Davies, P. A. Lee, and T. M. Rice, Phys. Rev. B **29**, 4260 (1984).

[6]G. A. Samara, Phys. Rev. Lett. **53**, 298 (1984).

[7]G. Mihaly and L. Mihaly, Phys. Rev. Lett. **52**, 149 (1984).

[8]A. Ogielski and I. Morgenstern, Phys. Rev. Lett. **54**, 928 (1985), and J. Appl. Phys. **57**, 3382 (1985); R. W. Bhatt and A. P. Young, Phys. Rev. Lett. **54**, 924 (1985); W. L. McMillan, Phys. Rev. B **31**, 340 (1985).

[9]J. J. Rhyne, IEEE Trans. Magn. **21**, 1990 (1985).

[10]S. G. Cornelison and D. J. Sellmyer, Phys. Rev. B **30**, 2845 (1984).

[11]For a review and references up to 1983, see K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, New York, 1984).

[12]J. H. Chen and T. C. Lubensky, Phys. Rev. B **16**, 2106 (1977).

[13]M. Suzuki, Prog. Theor. Phys. **58**, 1151 (1977).

[14]J. D. Patterson, G. R. Gruzalski, and D. J. Sellmyer, Phys. Rev. B **18**, 1377 (1978).

[15]D. J. Sellmyer and S. Nafis, J. Magn. Magn. Mater. **54–57**, 113 (1986).

[16]G. Kotliar and H. Sompolinsky, Phys. Rev. Lett. **53**, 1751 (1984).

[17]D. S. Fisher and H. Sompolinsky, Phys. Rev. Lett. **54**, 1063 (1985).

[18]K. H. Fischer and A. Zippelius, J. Phys. C **18**, L1139 (1985).

[19]A. Aharony, Phys. Rev. B **12**, 1038 (1975).

[20]R. A. Pelcovits, E. Pytte, and J. Rudnick, Phys. Rev. Lett. **40**, 476 (1978), and **48**, 1297 (1982).

[21]A. Aharony and E. Pytte, Phys. Rev. B **27**, 5872 (1983).

[22]Y. Y. Goldschmidt, Nucl. Phys. B **225 [FS9]**, 123 (1983).

[23]E. M. Chudnovsky and R. A. Serota, J. Phys. C **16**, 4181 (1983); E. M. Chudnovsky, W. Saslow, and R. A. Serota, Phys. Rev. B **33**, 251 (1986).

[24]Y. Y. Goldschmidt, Phys. Rev. B **30**, 1632 (1985).

[25]A. J. Bray and M. A. Moore, J. Phys. C **18**, L139 (1985).

[26]I. Morgenstern, private communication.

[27]M. Fähnle, Solid State Commun. **55**, 743 (1985).

[28]M. V. Tsodex and M. A. Feigel'man, to be published.