Magnetically dilute metallic glasses. II. 4*f* moments

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Magnetic-susceptibility and high-field magnetization measurements are presented for amorphous $Zr_{40}Cu_{60-x}M_x$ (M denoting Gd and Tb), with x ranging from zero to ten. Effective moments of magnetic solutes were determined by fitting susceptibility data to the Curie-Weiss expression. The moments of Gd and Tb are very close to those expected for trivalent ions and the paramagnetic Weiss temperatures are positive. Saturation did not occur in any of the samples, even at 80 kOe and 1.3 K; however, the alloys containing Gd approached normalized magnetization values of unity. High-field hysteresis loops were used to obtain the temperature dependence of the coercive fields H_c and to determine ordering temperatures, defined as those temperatures for which H_c vanishes. The zero-field susceptibility for both the Gd and Tb alloys shows sharp peaks at low temperatures. The magnetically ordered state of the Gd alloys is characterized as spin-glass-like; that is, the spins are frozen in random directions and there are both ferromagnetic and antiferromagnetic exchange interactions, with the former dominant. The Tb alloys also have low-temperature magnetic states characterized as spin-glass-like in this sense. However, in the Tb alloys, the presence of local random anisotropy affects the high-field magnetization considerably. Attempts to fit the high-field magnetization of the Tb alloys to the local-random-anisotropy theory of Harris *et al.* are described.

I. INTRODUCTION

This paper presents results on the magnetic properties and magnetic ordering of the metallic glasses $Zr_{40}Cu_{60-x}M_x$, with M = Gd and Tb and $x = 0, 3, 6, 10$. The general motivation for studying the magnetic properties of magnetically dilute glasses was discussed in the Introduction to the preceding paper (hereafter denoted as Paper I). The primary motivation for studying metallic glasses with rare-earth solutes was to investigate the effect of local random anisotropy. Because Gd dissolves as an *S*-state ion and Tb as an *F*-state ion, one would expect anisotropy effects to be considerably more pronounced in the Tb alloys than in the Gd alloys. Since these alloys have the same host, one would expect that only small differences in the fluctuations in the rare-earth–rare-earth exchange coupling $\mathcal{J}$ for alloys

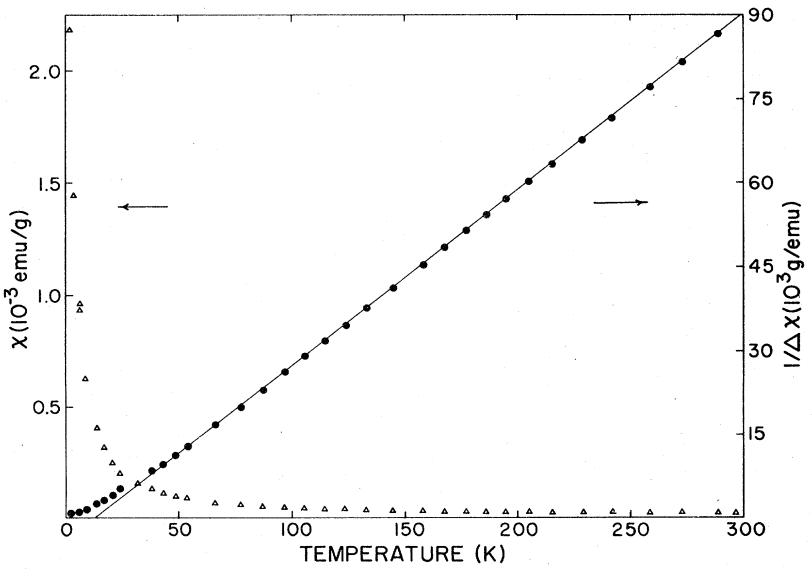

FIG. 1. $\chi(T)$ of $Zr_{40}Cu_{57}Gd_3$ (open triangles) at 0.23 kOe. Also plotted are $1/\Delta\chi$ and Curie-Weiss fit to data above 60 K.

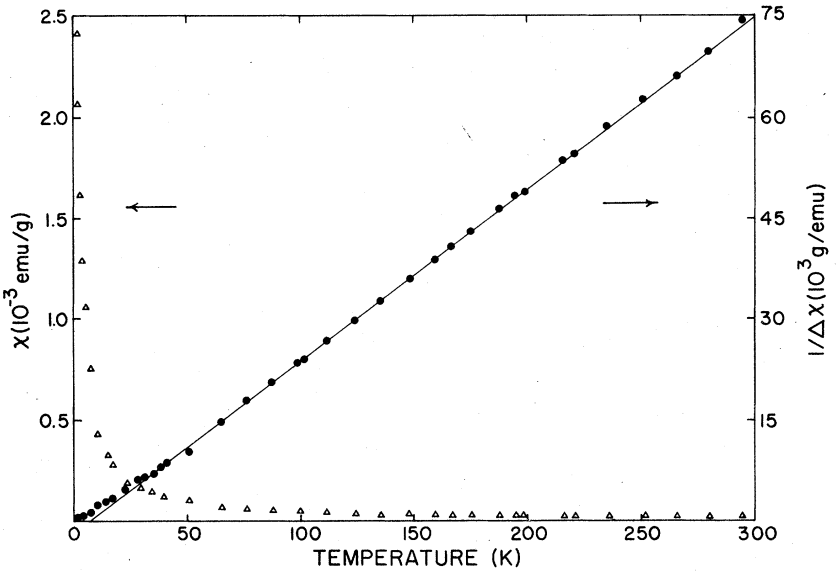

FIG. 2. $\chi(T)$ of $Zr_{40}Cu_{57}Tb_3$ (open triangles) at 0.23 kOe. Also plotted are $1/\Delta\chi$ and Curie-Weiss fit to data above 100 K.

of equal concentration. Therefore, any differences in the magnetic behavior of these two systems, that cannot be accounted for by a different de Gennes factor (which for rare-earth ions is determined by the Hund's-rule ground state) and/or different interaction between the conduction electrons and localized moments, may be due to differences in the strength of the local random anisotropy. Furthermore, in magnetically dilute alloys of this type, it is possible to decrease the concentration to arbitrarily small values so that, in alloys for which random anisotropy is present, the effect of random anisotropy can be made to dominate that of interimpurity exchange. This allows for a good test of the local-random-anisotropy (LRA) model of amorphous magnetism.[1,2]

The sample preparation and experimental methods employed are discussed in Paper I. In this paper the experimental results are presented in Sec. II and compared with various theoretical models in Sec. III. A brief summary and conclusion is given in Sec. IV.

II. EXPERIMENTAL RESULTS

The low-field room-temperature magnetization was measured for $Zr_{40}Cu_{60-x}M_x$, where M denotes Gd and Tb and $x=3$ and 6 is the solute concentration. For $Zr_{40}Cu_{57}Gd_3$, there is evidence of a very small amount of clustering; for the other samples, no curvature was exhibited. The low-field magnetic suscep-

TABLE I. Curie-Weiss parameters and T_c (see text). p_{eff} was obtained from C using the expression $p_{\text{eff}}=(3k_BC/N_i)^{1/2}\mu_B^{-1}$, where N_i is the number of impurity atoms per unit mass of alloy. Values of the angular momenta J were obtained using $p_{\text{eff}}=g[J(J+1)]^{1/2}$. Here g, the Landé g factor, was calculated assuming trivalent ions. $\chi(T)$ for the host alloy shows essentially no temperature dependence and equals about 0.89 μemu/g for $Zr_{40}Cu_{60}$. ND denotes not determined.

Sample	χ_0 (μemu/g)	Θ (K)	p_{eff}	J	T_c (K)
$Zr_{40}Cu_{57}Gd_3$	6.9	13	8.0	3.5	<5
$Zr_{40}Cu_{54}Gd_6$	3.3	30	8.0	3.5	~15
$Zr_{40}Cu_{57}Tb_3$	3.6	8	8.9	5.5	~4.8
$Zr_{40}Cu_{54}Tb_6$	1.6	6	9.4	5.8	~6.8
$Zr_{40}Cu_{50}Gd_{10}$	ND	ND	ND	ND	~45
$Zr_{40}Cu_{50}Tb_{10}$	ND	ND	ND	ND	~16

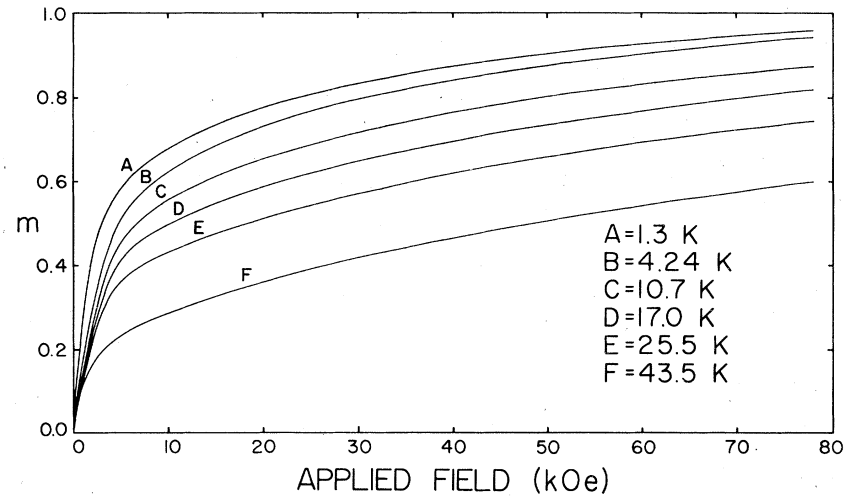

FIG. 3. $m(H)$ of $Zr_{40}Cu_{50}Gd_{10}$ at various temperatures.

tibility of the $x=3$ and 6 samples was measured at 0.23 kOe. Examples of the temperature dependence of the magnetic susceptibility for these samples are shown in Figs. 1 and 2. Curie-Weiss fits and $1/\Delta\chi$ are also shown in these figures. The susceptibility data of these four alloys follow a Curie-Weiss law quite well except at the lowest temperatures; the resulting parameters are in Table I. The values of p_{eff} are very close to those expected for trivalent ions subject to Hund's rule (7.94 for Gd^{3+} and 9.72 for Tb^{3+}). The positive Θ values indicate a predominance of ferromagnetic interimpurity coupling.

High-field magnetization measurements were made on all the samples of various temperatures between 1.3 and 48 K. Examples of these data are shown in Figs. 3 and 4. In these figures, $m=M/g\mu_B J$ is the reduced magnetization. Here M is the magnetization per rare-earth ion and gJ is calculated from Hund's rule assuming trivalent ions ($gJ=7$ for Gd^{3+} and $gJ=9$ for Tb^{3+}). With this definition, the reduced magnetization is normalized to unity.

Figures 3 – 5 clearly show that the approach to saturation of the Gd alloys is different from that of the Tb alloys. This is evident in that the values of $m(H)$ for a given temperature are much smaller for the Tb alloys than for the Gd alloys. A closer look also shows that the shape of the $m(H)$ curves for the Tb alloys differs from that for the Gd alloys. For the Gd alloys, $m(H)$ lies above a simple Brillouin function at high temperatures, which substantiates the idea that there exists a predominance of ferromagnetic interimpurity coupling in this alloy. However, at lower

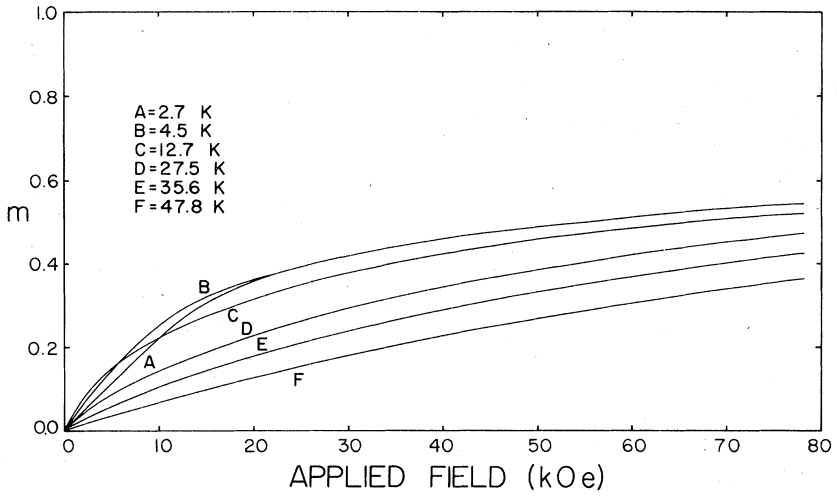

FIG. 4. $m(H)$ of $Zr_{40}Cu_{50}Tb_{10}$ at various temperatures.

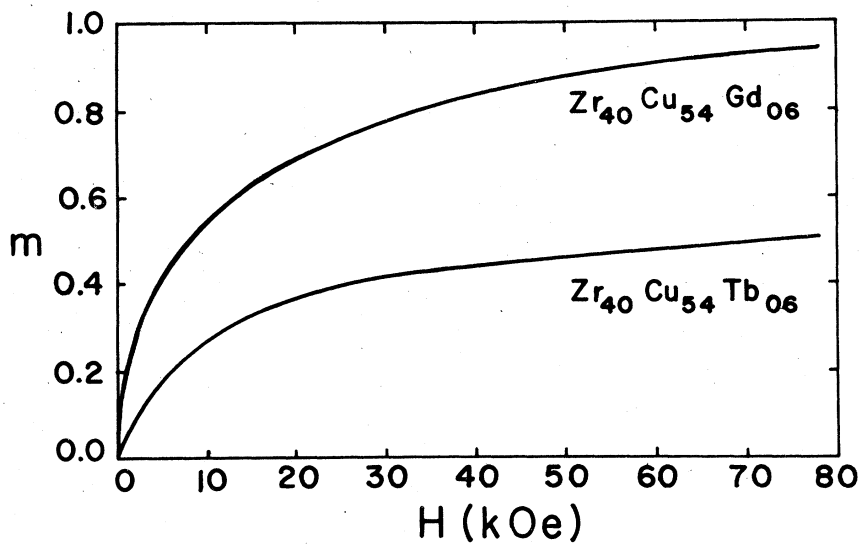

FIG. 5. $m(H)$ at 1.3 K showing the difference in approach to saturation for $Zr_{40}Cu_{54}Gd_6$ and $Zr_{40}Cu_{54}Tb_6$.

temperatures, $m(H)$ approaches saturation more slowly than does a simple Brillouin function. This is consistent with the idea that at low temperatures the local moments are frozen in directions with a large degree of randomness, i.e., in a spin-glass-like state. More will be said regarding this behavior later. For the Tb alloys, $m(H)$ approaches saturation much more slowly than does a simple Brillouin function over the entire temperature range.

High-field hysteresis loops were taken on all of the samples. In all cases, the field was taken to 80 kOe in both directions and no "squaring" off of the hysteresis loop was observed.

The temperature dependence of the coercive field (H_c) was determined from the high-field hysteresis data. $H_c(T)$ for the 10-at.% Gd and Tb alloys is shown in Fig. 3 of Ref. 3. The values of H_c for the Tb alloys are an order of magnitude larger than for the Gd alloy. As was done for the Fe and Mn alloys (Paper I), the hysteresis data were used to determine T_c, that temperature for which H_c vanishes. Values of T_c for the 3- and 6-at.% samples are given in Table I and values for all the samples are displayed in Fig. 6.

The values obtained for T_c were compared with random ferromagnetic ordering temperatures suggested by plotting curves of m^2 vs H/m. This was done for three samples and two examples of such plots are shown in Figs. 7 and 8. These plots were constructed using $m(H)$ data obtained by increasing H. An example of one case (Fig. 8, dashed line) is given for which the plot was constructed by starting in the magnetized state and decreasing H. The $m(H)$ curves were not virgin curves in the usual sense because the samples were not heated to above the ordering temperature between $m(H)$ sweeps.

The "extrapolation" behavior of $m^2(H/m)$ in Fig. 7 suggests a random ferromagnetic ordering temperature of $Zr_{40}Cu_{50}Gd_6$ between 10 and 22.4 K. This temperature is similar to T_c for this alloy; however, the maximum in the zero-field susceptibility data lies at a considerably lower temperature. For $Zr_{40}Cu_{54}Tb_6$ the peak in $x(T)$ occurs at a temperature near T_c. In Fig. 8, m^2 vs H/m and zero-field susceptibility for $Zr_{40}Cu_{50}Tb_{10}$ are displayed. The ordering temperature estimated from the straight line intercept and from the peak in the zero-field susceptibility both are in the vicinity of T_c.

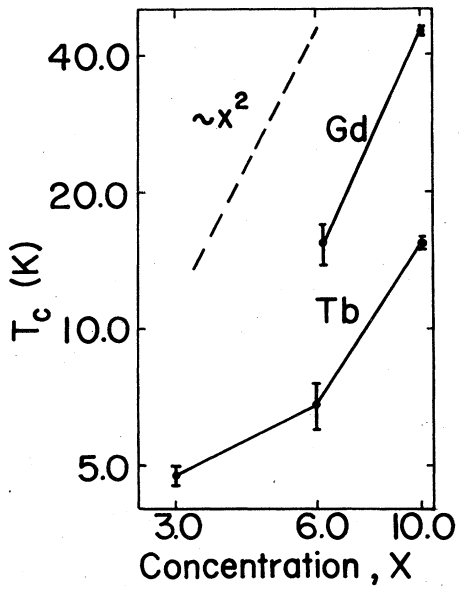

FIG. 6. $T_c(x)$ for $Zr_{40}Cu_{60-x}M_x$, M = Gd and Tb. For $Zr_{40}Cu_{57}Gd_3$, $T_c < 5$ K. The dashed line goes as x^2.

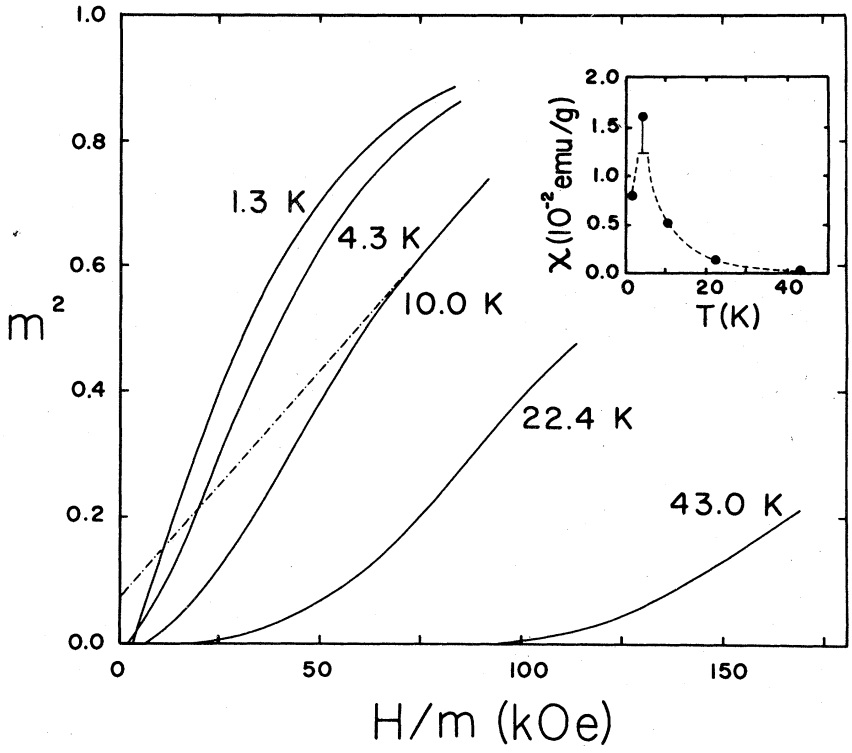

FIG. 7. Curves of m^2 vs H/m and $\chi(T)$, obtained from H/m intercepts, for $Zr_{40}Cu_{54}Gd_6$. The initial slopes of four separate 4.3-K $m(H)$ curves lie within error bar below the $\chi(T=4.3\ \text{K})$ datum. For this alloy, 1 gOe/μemu $\simeq$ 29,215 kOe per unit of m.

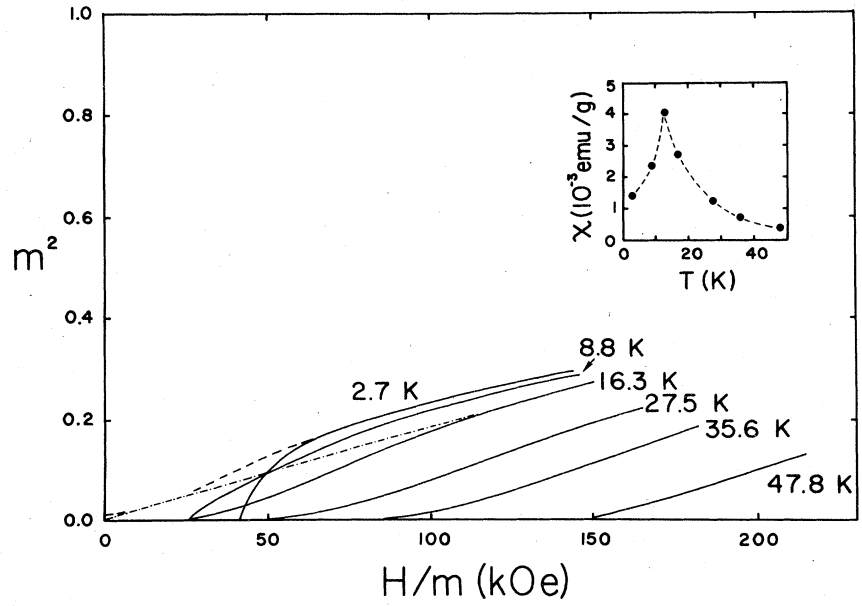

FIG. 8. Curves of m^2 vs H/m and $\chi(T)$, obtained from H/m intercepts, for $Zr_{40}Cu_{50}Tb_{10}$. For one case (dashed line), a plot was constructed from $m(H)$ data obtained by starting in the magnetized state and decreasing H. The $\chi(T=12.7\ \text{K})$ datum was obtained from the initial slope of curve C in Fig. 4. For this alloy, 1 gOe/μemu $\simeq$ 59,948 kOe per unit of m.

III. DISCUSSION

A. Gd alloys

It seems likely that the magnetic behavior of the Gd alloys is explicable in terms of Gubanov's[4] idea regarding a Heisenberg model including fluctuations in the interimpurity exchange coupling $\mathcal{J}$. However, the available theories involving fluctuations in $\mathcal{J}$ either are very simplified or are primarily for crystalline metallic spin glasses and assume long-range interimpurity interactions of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type.[5] From the values of the resistivity one can estimate the electron mean free path in metallic glasses to be of the order of the interatomic distances, whence long-range oscillatory interactions are probably inappropriate. Nevertheless, $m(H)$ data for the Gd alloys were compared with two theories.

R. Harris and M. J. Zuckermann[6] have compared the present $m(H)$ data for $Zr_{40}Cu_{54}Gd_6$ at 15.4 K to an expression due to Handrich.[7] Writing this expression in a form similar to that used for Eqs. (3) and (4) of Paper I,[8] one obtains

$$m = B_J(x_+) + B_J(x_-) \quad , \tag{1}$$

where $x_\pm$ is given by

$$x_\pm = g\mu_B J[H + \lambda(1 \pm \Delta)M]/k_B T \quad , \tag{2}$$

λ is a molecular-field constant related to the paramagnetic Curie temperature as in Eq. (7) of Paper I, and Δ is a measure of the fluctuation in $\mathcal{J}$. For $\Delta > 1$ and $H = 0$ Handrich's expression for m approaches zero as $T \to 0$. Since this behavior was not observed experimentally, a comparison between data and Eq. (1) was not made for $\Delta > 1$. For $\Delta \leq 1$, the data approached saturation more slowly than did Handrich's expression for m. That the experimental values of m lie below those values of m given by Eq. (1) supports the above-mentioned idea that the Gd alloys develop a spin-glass-like state at lower temperatures.

A comparison between experiment and a theory due to Larkin and Khmel'nitskii (LK)[9] was also made. LK assume a long-range RKKY interimpurity interaction (their theory is applicable primarily to crystalline spin glasses). At large distances r between local moments, the spatial dependence of the RKKY interaction is given by

$$V(r) = V_0 \cos(2k_F r)/r^3 \quad , \tag{3}$$

where k_F is the Fermi wave number. For $g\mu_B H >> k_B T$ and $g\mu_B >> nV_0$, n being the solute concentration, LK obtained the following expression for the reduced magnetization:

$$m = 1 - \frac{2}{3}\frac{V_0 n}{g\mu_B H}(2S+1) \quad . \tag{4}$$

(Note that for Gd^{3+} ions, $J = S$, the spin angular momentum.) Equation (4) is applicable only when the second term on the right-hand side is a small correction. Thus, the high-field ($H > 50$ kOe) $m(H)$ data for $Zr_{40}Cu_{50}Gd_{10}$ at 1.3 K were used to test this expression. m vs $1/H$ was plotted and, although there was a slight concave-upward curvature in the resulting curve, a straight line with slope -7100 Oe could be drawn reasonably through the data. Extrapolation of this line to $1/H = 0$ gave $m \simeq 1.05$, which is a reasonable result. Using Eq. (4) gave $V_0 \simeq 5 \times 10^{-39}$ erg cm^3, where the solute concentration was taken to be $n \simeq 5 \times 10^{21}$/cm^3. Information that readily can be estimated from V_0 includes the effective value of the exchange interaction Γ between the local moments and conduction electrons, and the spin-glass ordering temperature T_{sg}. Γ can be obtained from the following expression[10]:

$$\Gamma^2 = \frac{V_0 32(2)^{1/2} E_F k_F^3}{9\pi^2 S(S+1)} \quad , \tag{5}$$

where E_F is the Fermi energy. Using free-electron parameters $E_F = 9.56$ eV and $k_F = 1.59 \times 10^8$ cm^{-1}, the value $\Gamma = 0.056$ eV was determined. Sherrington and Southern[11] give the following expression for T_{sg}:

$$T_{sg} = \frac{4\pi n V_0}{9k_B}\left[[S(S+1)]^2 + \frac{S(S+1)}{2}\right]^{1/2} \quad . \tag{6}$$

In obtaining Eq. (6), $h^2\tilde{J}$ of Ref. 11 was replaced by the average value of V_0/r_{12}^3, (Ref. 12) where $\frac{1}{3}(4\pi r_{12}^3) = n^{-1}$ is the average volume per magnetic ion. Equation (6) gives $T_{sg} = 4.9$ K, a temperature an order of magnitude lower than T_c. It therefore appears that either Eq. (4) or Eq. (6), or possibly both, is not applicable to the Gd alloys.

It certainly is reasonable that the Gd ions interact by means of indirect exchange via the conduction electrons. Also, even though an RKKY interaction is unlikely in amorphous metals, a similar interaction is expected, probably one falling off more rapidly and oscillating differently from that given in Eq. (3). Therefore, because the high-field low-temperature $m(H)$ data have an H dependence similar to that given by Eq. (4) and the $1/H = 0$ intercept is a reasonable one, it is possible that the resulting value for V_0 is a rough measure of the interimpurity interaction. Similarly, the value obtained for Γ may be a rough estimate of the exchange interaction between the local moments and the conduction electrons. In this connection it may be noted that Felsch[13] determined that $\Gamma = 0.08$ eV in $LaGd_6$(Gd) alloys, a value which is similar to that obtained above using Eq. (5).

If the above reasoning is valid, it appears that Eq. (6) probably is inappropriate, i.e., the Gd alloys probably are not spin-glasses in the sense that the exchange interactions are distributed randomly about a zero mean. The question which arises is, what kind

of magnetic order develops in the Gd alloys? Since the magnetic behavior is probably affected mainly by fluctuations in $\mathcal{J}$, it may be appropriate to discuss the different types of magnetism arising from a Heisenberg model with fluctuating $\mathcal{J}$. For simplicity, only nearest-neighbor interactions are considered.

If the average fluctuations in $\mathcal{J}$ and the average value of $\mathcal{J}$ are given by $\Delta\mathcal{J}(>0)$ and $\bar{\mathcal{J}}$, respectively, then three distinct phases are possible. The first phase is characterized by $\bar{\mathcal{J}}>0$ and $\Delta\mathcal{J}<\bar{\mathcal{J}}$. The precise meaning of this depends upon how $\Delta\mathcal{J}$ is defined; however, the idea is that there exists a clear predominance of ferromagnetic nearest-neighbor exchange interactions and the effect of random antiferromagnetic interactions is negligible. A system characterized in this way belongs to that group of systems called random ferromagnetic. The second phase is characterized by $\bar{\mathcal{J}}<0$ and $\Delta\bar{\mathcal{J}}<|\bar{\mathcal{J}}|$. Such a system may be called a random antiferromagnet or, perhaps, a spin-glass. However, for this discussion, the term spin-glass will be reserved for the third possible phase, which is characterized by $\Delta\mathcal{J}>|\bar{\mathcal{J}}|$. In a system of this kind, there exists a nearly equal mixture of positive and negative exchange. That the three phases mentioned above give rise to random ferromagnetism, random antiferromagnetism, and spin-glassiness, probably would not be disputed. Yet, an attempt to demarcate where one phase ends and another begins probably would not be readily accepted. Defining $P(\ )$ to be the probability distribution function of exchange interactions in a magnetic glass, one might define an *ideal* spin-glass as one in which $P(\mathcal{J})$ is a symmetrical function with its maximum centered on $\mathcal{J}=0$, i.e., $\bar{\mathcal{J}}=0$. It is suggested that glasses for which $P(\mathcal{J})$ is not centered on $\mathcal{J}=0$ (so that $\bar{\mathcal{J}}\neq 0$) be denoted as *spin-glass-like*. Thus, we characterize the phase of magnetic order in the Gd alloys as spin-glass-like with positive $\bar{\mathcal{J}}$.

B. Tb alloys

Section III A confronted the question of what kind of order develops in the Gd alloys. A Heisenberg Hamiltonian with fluctuating $\mathcal{J}$ was used to define the character of the ordered phase in terms of $\bar{\mathcal{J}}$ and $\Delta\mathcal{J}$ More generally, however, the character of the ordered phase can be determined by the average fluctuations in the molecular fields which may be due to fluctuations in $\mathcal{J}$ or some other mechanism.

For example, an effect of local random anisotropy is to introduce fluctuations in the local molecular fields. Patterson *et al.*,[14] using a local-mean-field approximation, have shown that the LRA model allows for metastable spin-glass-like states for large enough values of $D/\mathcal{J}=D/\bar{\mathcal{J}}$. If these metastable states are ignored, however, the theoretical results show that (for $\Delta\mathcal{J}=0$) the presence of local random anisotropy does not affect the *character* of the (equilibrium) ordered states.[15] That is, if $\mathcal{J}=\bar{\mathcal{J}}>0$ (or $\bar{\mathcal{J}}<0$), then the character of the ordered equilibrium state is random ferromagnetic (or random antiferromagnetic), regardless of the size of D; the fluctuations in the molecular field effected by the presence of random anisotropy do not change the character of a state for which $\Delta\mathcal{J}=0$. However, the *type* of magnetism is significantly influenced by the size of D, e.g., the amount of spread in the fan structure increases with D.

For $\Delta\mathcal{J}\neq 0$, the existence of local random anisotropy may still have only a small effect on the character of the (equilibrium) ordered states. Of course, since the presence of local anisotropy increases the fluctuations in the local molecular field, it must have some effect in this regard. For example, it is likely that a system with exchange couplings such that $\bar{\mathcal{J}}\neq 0$ (denoted as spin-glass-like in Sec. III A) would be driven somewhat closer to the spin-glass phase as D is increased from zero; it is clear that an effect of D would not be to drive the state farther from the spin-glass phase. In the following this reasoning will be used to discuss the character of the ordered phase of the Tb alloys.

It is reasonable that there are only small differences in $\Delta\mathcal{J}/\bar{\mathcal{J}}$ for alloys of Gd and Tb of equal concentrations. The behavior of the high-temperature susceptibility indicates a predominance of ferromagnetic exchange interactions and the zero-field susceptibility exhibits a maximum at low temperatures for both the Gd and Tb alloys. By a direct comparison between experiment and theory, it was concluded that the Gd alloys are spin-glass-like with positive $\bar{\mathcal{J}}$. Because present theories do not consider both fluctuations in $\mathcal{J}$ and random anisotropy, such a comparison between experiment and theories including fluctuating $\mathcal{J}$ could not be made for the Tb alloys. However, in light of what has been said about the effect of random anisotropy on the character of the ordered phase, and because of the existing similarities between the Tb and Gd alloys regarding $\bar{\mathcal{J}}$ and maxima in the susceptibilities, it is likely that the ordered phase of the Tb alloys is also spin-glass-like with positive $\bar{\mathcal{J}}$. In this connection we note that the remanence $m(0)$ for $Zr_{40}Cu_{50}Tb_{10}$ is only about 0.1 (see Fig. 2 of Ref. 3). If the character of the magnetic order were describable as random ferromagnetic, due to ferromagnetic coupling only, then one would expect $m(0)>0.5$ for $T\ll T_c$. This result, therefore, is an indication that the state of magnetic order does contain some spin-glass character.

It has been stated that the temperature dependence and magnitude of H_c and the behavior of $m(H)$ are significantly influenced by local random anisotropy in the Tb alloys. This follows for several reasons. First of all, the rapid increase in H_c with decreasing temperature for $Zr_{40}Cu_{50}Tb_{10}$ is reminiscent of the results

of Patterson *et al.*[14] using the random anisotropy model. Second, the large values of H_c for the Tb alloys as compared to those for the Gd alloys can be understood in terms of local moments getting "blocked" in the Tb alloys by the energy barriers produced by crystal-field anisotropy. Third, the extremely slow approach to saturation for the Tb alloys *vis-à-vis* Gd alloys is explicable in terms of the random anisotropy model. A further discussion of this last point is given below.

A rough estimate of D can be determined by considering the random anisotropy model classically for $T=0$, and in a mean-field approximation, as do Callen *et al.*[16] They show that if D is large enough, $m(H)$ increases linearly with H for a range of fields such that H is large enough to ignore domain effects, yet small enough so that

$$D >> \tfrac{2}{3}\mu_{\text{eff}}H + k_B\Theta \ . \tag{7}$$

Here D has units of energy, μ_{eff} is the classical effective magnetic moment, and Θ is the paramagnetic Curie temperature. For this range of H, Callen *et al.* show that the random anisotropy strength is given by

$$D = \frac{g\mu_B}{3J}\left(\frac{dm}{dH}\right)^{-1} \ . \tag{8}$$

Values of dm/dH were obtained for all the Tb alloys from the nearly-linear high-field $m(H)$ values. The values of D (in temperature units) determined from Eq. (8) were between 4 and 5 K; however, these values clearly are not large enough to satisfy Eq. (7). Nevertheless, these values may be considered as approximate estimates of D.

In principle, a better way to obtain the anisotropy strength D is by fitting $m(H)$ over the whole field range to the quantum-mechanical LRA theory of Harris, Plischke, and Zuckermann (HPZ).[2] We have pursued this problem in collaboration with Harris, Zuckermann, and Ferrer and briefly describe the procedure, following Ferrer *et al.*[3] The Hamiltonian of the HPZ model is

$$H = -g\mu_B H\sum_i J_{iz} - \sum_{i,j}\mathcal{J}_{ij}\vec{J}_i\cdot\vec{J}_j - D\sum_i J_{iz_i'}^2 \ . \tag{9}$$

In the molecular-field approximation (MFA) this becomes

$$H^{\text{MFA}} = -g\mu_B\sum_i (H^{\text{mol}}+H)J_{iz} - D\sum_i J_{iz_i'}^2 \ , \tag{10}$$

where

$$H^{\text{mol}} = 2\nu\mathcal{J}\langle J_z\rangle/g\mu_B \ , \tag{11}$$

$\mathcal{J}$ is the nearest-neighbor exchange constant, ν the number of rare-earth nearest neighbors, z is the direction of the bulk magnetization, and z' is the local easy axis. The Hamiltonian of Eq. (10) can be diagonalized for any value of J and a self-consistent equation for the magnetization determined. Also, in the MFA the ordering temperature T_c is given by

$$T_c = 2\nu\mathcal{J}J(J+1)/3k_B \ . \tag{12}$$

Ferrer *et al.* have shown that the above procedure leads to reasonable agreement with the $m(H)$ and T_c data for amorphous HoAg, TbAg, and DyAg, at least above 100 kOe. Below 100 kOe, where the theory and data diverge significantly, Ferrer *et al.* suggest that metastable spin-glass-like states exist which drive the magnetization far below the calculated values.

Harris *et al.*[17] have fitted our Tb alloy data to the LRA theory. The fit of the $m(H)$ data for $Zr_{40}Cu_{50}Tb_{10}$ at 47.8 K (far above $T_c=16$ K) is shown in Fig. 9. The fit gives a value of $D=11.6$ K and $\nu\mathcal{J}=0.42$ K. Using these parameter values, $m(H)$ was computed for 27.5 and 16.3 K. Figure 9 shows that the agreement between the computed curves and experimental data is not very good, the experimental $m(H)$ values falling increasingly below the calculated ones as $T\rightarrow T_c$. In addition, the same value of D was used to compute $m(H)$ curves for $Zr_{40}Cu_{57}Tb_3$, as shown in Fig. 10. It is seen that these fits are only semiquantitatively correct, even with $\nu\mathcal{J}=0$; good fits could be achieved only with a much larger value of D. Two comments regarding these fits are appropriate. First, the LRA model ignores fluctuations in the strength of both the exchange coupling and the local anisotropy; yet, it has been argued that ΔJ is large in these alloys, and it is almost certain that fluctuations in the local anisotropy strength are present. In connection with this, it is noted that $[m(H)]_{\text{theor}}$ increases with respect to $[m(H)]_{\text{exp}}$ as T approaches T_c; this is entirely consistent with the conclusion that a substantial amount of antiferromagnetic coupling is present in these al-

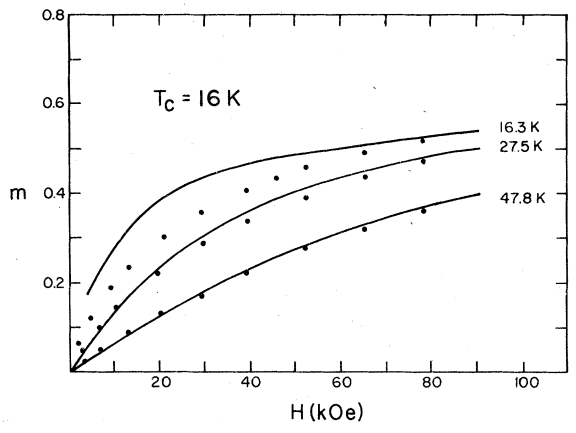

FIG. 9. Computed curves (full lines) using the LRA model and experimental $m(H)$ points, for $Zr_{40}Cu_{50}Tb_{10}$. The parameters $D=11.6$ K and $\nu\mathcal{J}=0.42$ K were determined by fitting the data obtained at 47.8 K.

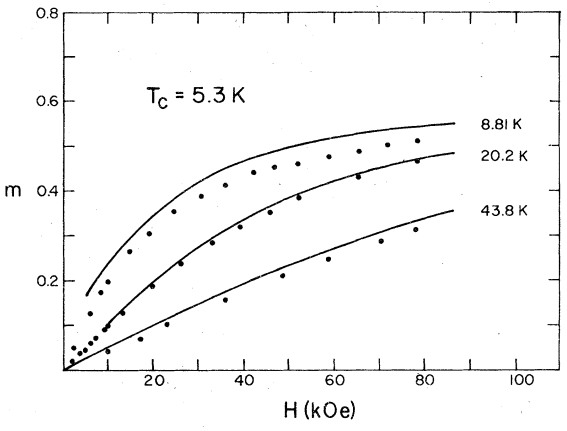

FIG. 10. Computed curves (full lines) using the LRA model and experimental $m(H)$ points for $Zr_{40}Cu_{57}Tb_3$. The parameters $D = 11.6$ K and $\nu\mathcal{J} = 0$ were used.

loys. Second, the fits of Figs. 9 and 10 are for one value of D. To get good fits for different Tb concentrations different D values had to be used. Because the local environments of the Tb atoms will vary significantly, in terms of rare-earth versus nonrare-earth neighbors, as one goes from 3 at.% Tb to 10 at.% Tb, the strength of the local random anisotropy (as well as the size of the exchange fluctuations) will also vary. It is perhaps not too surprising that such a simple theory is incapable of fitting the data with only one D value for arbitrary Tb concentration. To our knowledge, no other attempts have been made to characterize the LRA strength in alloys of varying rare-earth concentration. This is an interesting question for future research.

IV. CONCLUSIONS

The magnetic behavior of the Gd alloys is understandable in terms of a Heisenberg Hamiltonian including fluctuations in $\mathcal{J}$. The behavior of the high-temperature susceptibility and magnetization both indicate that the predominant exchange interaction is ferromagnetic, i.e., $\overline{\mathcal{J}} > 0$. However, the slow approach to saturation at low temperatures indicates the presence of some random antiferromagnetic interactions along with the ferromagnetic interactions. The sharp maximum in the zero-field susceptibility is consistent with this interpretation. It is concluded that the presence of the antiferromagnetic interactions gives rise to a spin-glass-like phase with positive $\overline{\mathcal{J}}$.

For the Tb alloys, it has been shown that there exists a predominance of ferromagnetic exchange interactions. Also, it has been inferred that a significant amount of antiferromagnetic coupling exists, and that the presence of this antiferromagnetic coupling causes the Tb alloys to order in a spin-glass-like phase with positive $\overline{\mathcal{J}}$. It has also been shown that the magnetic behavior of the Tb alloys is significantly influenced by local random anisotropy and that the $m(H)$ data can be fitted approximately to the random anisotropy model of amorphous magnetism with D values of order 10 K.

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