CRITICAL PHENOMENA AND EXCHANGE INTERACTIONS OF AN AMORPHOUS FERROMAGNET: GADOLINIUM-GOLD

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ABSTRACT

Magnetization was measured between 4 and 290°K in fields up to 70 kOe on liquid-quenched $\text{Gd}_{80}\text{Au}_{20}$ amorphous alloys. The Curie temperature and critical exponents β , γ and δ are found to be 149.45°K, 0.439, 1.294 and 3.948 respectively. These values are compared with the Heisenberg and molecular field values. The data are fitted to an equation of state previously derived for second order phase transition in fluid systems. The results illustrate clearly a second order phase transition in the amorphous state. A discussion in terms of the Heisenberg model is presented. The effective magnetic moment in the paramagnetic state has a value of 9.37 Bohr magnetons per gadolinium atom. The saturation moment extrapolated to 0°K equals 7.0 Bohr magnetons per gadolinium atom. The low temperature saturation magnetization observes the $T^{3/2}$ law from 0.13 T_c to 0.80 T_c . The effective exchange integrals J_n determined from the Rushbrooke-Wood formula and spin-wave theory are found to be 2.28 and 1.34°K respectively. The differences in J_n , and that between the effective moment and saturation moment are attributed to the nearest-neighbor anti-ferromagnetic couplings below T_c . The possible effects of structural disorder on the magnetic properties of Gd in the amorphous state are discussed.

I. INTRODUCTION

Magnetism in structurally amorphous alloys has been the subject of considerable interest not only as a topic in solid state physics, but also because of their technological potential. Rare earth-transition metal binary alloys in the amorphous state have been prepared in bulk form by d.c. rapid sputtering and by coevaporation as thin films.^{1, 2, 3} Amorphous rare earth-noble metal alloys have also been prepared by these techniques^{3, 4, 5}, and by the method of liquid quenching.⁶ The compositional ranges of the systems investigated with the exception of Gd-Ag alloys⁴ are on the transition metal side of the binary phase diagrams. Previous attention has been focused on compositions corresponding to some crystalline counterparts (e.g. $R-Fe_2$, where $R = Ho, Gd, Tb$ and Y).¹ Neutron scattering measurements⁷ on the bulk samples of these alloys have shown them to be amorphous both structurally and magnetically with a dense random packing of R and Fe atoms. Magnetization and Curie temperature measurements have been carried out to study the magnetic properties (e.g. magnetic moments, local anisotropy, and exchange interactions etc.) of the alloys. A mean-field model of the magnetic properties of $R-Fe_2$ amorphous alloys has been developed^{8, 9} which incorporates a Fe spin whose dependence on parameters is derivable from Mössbauer spectra.

Besides these measurements, there was a basic question of whether or not a second order magnetic phase transition exists in an amorphous ferromagnet. Mizoguchi et al¹⁰ demonstrated experimentally that the answer is positive. Their results indicated that the critical exponents satisfy an equality relation, and the reduced magnetization and field

satisfy an equation of state derived for second order phase transition in fluid systems.¹¹ Previous experiments have been performed to investigate the critical behavior of ferromagnets in the crystalline state.¹²⁻¹⁵ Most of the materials studied have critical exponents which agree well with the theoretical values derived from the Heisenberg model. Such agreement was also observed in amorphous ferromagnets^{10, 16} near the Curie temperature. Up to this point, a question concerning the temperature domain of the applicability of different models still exists.

So far, the rare earth-noble metal and transition metal alloys which can be liquid quenched into the amorphous state are the superconducting La-T (where T = Au, Cu and Ni) alloys.⁶ It is interesting to obtain splat-cooled ferromagnetic rare earth-noble metal alloys which might be structurally different from the coevaporated amorphous alloys. We report here the results of magnetization measurements for bulk amorphous $\text{Gd}_{80}\text{Au}_{20}$ alloys obtained by liquid quenching. Gold is chosen as the glass former both for metallurgical reason and for the fact that it contains no unfilled d- and f-shells. Since the magnetic properties of Gd are mainly determined by the d- and f-electron interactions at the Fermi level¹⁷, the presence of Au is not expected to affect appreciably the magnetic properties of amorphous Gd besides the local structure. The purpose of this paper is two fold. First, a detail study is made on the critical behavior of amorphous $\text{Gd}_{80}\text{Au}_{20}$ alloys around its Curie temperature. We present convenient methods for determining the spontaneous magnetization and initial susceptibility values in the critical region. This allows accurate determinations of the critical exponents and T_c . Magnetization results of single crystal Gd^{18, 19} were found to depend strongly on the crystal axis along which the field was applied. Measure-

ments on amorphous Gd are expected to yield an averaged result of some corresponding crystalline values. Possible asymptotic equations of state are to be investigated following the work of Kouvel and Comly.²⁰ Second, we study the effect of structural randomness on the magnetic properties (effective moment μ_{eff} , saturation moment μ_{Gd} , T_c , and saturation magnetization $M(\infty, T)$) of $\text{Gd}_{80}\text{Au}_{20}$. Comparison of the present results with those of crystalline compounds and solid solutions are made. These results are extrapolated to the case of pure amorphous Gd and compared with theoretical predictions whenever possible.

The format of this paper is as follows. In Section II, experimental procedures and results are presented. Section III consists of three subsections from A to C. Section A describes the processing of raw data to obtain final data for analysis. In Section B (i), critical behavior around T_c is investigated using data from detail magnetization measurements up to ~ 40 kOe for every degree in the temperature range $|T - T_c| \leq 10^\circ\text{K}$. The critical exponents are discussed in terms of models and local interactions. Section B (ii) presents an equation of state graphically. The parameters involved come from the scaling law hypothesis. Asymptotic equations of state very close to T_c are also obtained and discussed. Section C gives the values of μ_{eff} , μ_{Gd} and $M(\infty, T)$. These values are compared and discussed with those of crystalline systems. The values of the nearest-neighbor exchange constants are obtained from the spin-wave theory²¹ and the Rushbrooke-Wood formula.²² Their difference is discussed together with the lowering in μ_{Gd} as compared to the value in crystalline Gd. The possible effects of structural randomness on the magnetic properties are discussed. The last section is the summary and conclusion.

II. EXPERIMENTAL PROCEDURES AND RESULTS

The $\text{Gd}_{80}\text{Au}_{20}$ alloys were prepared by induction melting of the appropriate constituents on a silver boat under an argon atmosphere. Samples were then quenched from the liquid state using the "piston and anvil" technique described in reference 23. The cooling rate is estimated to be of the order 10^6 °C/sec. Samples prepared by this technique were in the form of foils with surface area of $\sim 2 \times 2 \text{ cm}^2$ and thickness of $\sim 10 \mu\text{m}$. The structure of each sample was checked by X-ray scanning with a Norelco diffractometer. Only samples containing a single amorphous phase were retained for detailed experimental studies. The X-ray diffraction patterns of these samples were characterized by a broad maximum centered at 32.8° with a full width at half maximum of 4.6° . According to the Sherrer formula, this corresponds to an effective microcrystal size of $\sim 17\text{\AA}$, which is typical of a glassy metal.²⁴ It should be added that by analyzing our magnetization data we found a small amount of crystallization in the samples ($\sim 2\%$ of a sample) which cannot be detected by the rapid X-ray scanning technique. No significant annealing effect is observed for the amorphous phase at room temperature during periods of several weeks. Spontaneous crystallization is observed at temperatures of about 200 to 250°C .

The ferromagnetic transition temperature (Curie temperature T_c) was observed using a standard ac inductance bridge technique. The T_c estimated from the inflection point of the signal intensity was found to be $\sim 149^\circ\text{K}$. Magnetization measurements as functions of magnetic field and temperature were carried out by using the Faraday method

with an Oxford Instruments Magnetometer described elsewhere.²⁵ Samples used in the $M(H, T)$ measurements were in the form of disks ~ 3 mm in diameter punched from foils. The magnetic force and the field magnitude were determined with two high sensitivity digital voltmeters. The emf of a Au-Fe thermocouple and the field gradient which were supposedly constant were checked frequently for every magnetic isotherm measurement. The thermal output control for the temperature has an accuracy of $\sim 0.1^\circ\text{K}$. Measurements for $M(H)$ were made approximately every 20°K from 4°K to 290°K except near T_c , and for fields up to 70 kOe. Near T_c ($\sim 149^\circ\text{K}$), measurements were made every 1°K in the temperature range of 136°K to 160°K , and for fields up to 40 kOe.

Figure 1 illustrates representative high field isotherms $M(H)$. In figure 2, $M(T)$ for constant magnetic fields were plotted. It is noticed that the small kink or inflection point expected at T_c is not detectable for fields higher than 2 kOe. For small H , the Curie temperature is given by the inflection point because for T just greater than T_c , M is dominated by a large initial susceptibility χ_0 which has a hyperbolic dependence on temperature. For large H , the disappearance of the inflection point at T_c is attributed to the magnetic inhomogeneities.²⁶

III. ANALYSIS AND DISCUSSION

A. Processing of Data.

In order to discuss the results, it is necessary to know the $M(H, T)$ values of the magnetic amorphous phase and the magnetizing field H corrected from the applied field H_0 . Thus corrections for any possible

Fig. 1. Magnetization versus applied field for different temperatures ($4.2^{\circ}\text{K} \leq T < 230^{\circ}\text{K}$). The dashed line indicates the demagnetizing effect correction.

Fig. 2. Magnetization versus temperature for different applied fields ($0 \leq H \leq 70$ kOe).

magnetic precipitates from the matrix and demagnetizing effects must be made. The demagnetizing constant is determined from averaging the initial slopes of the $M(H)$ curves below T_c . The average value of the initial slopes over the temperature range $4^\circ\text{K} < T \leq 140^\circ\text{K}$ is approximately 75 emu/g kOe. This correction method simply assumes that $\chi_o^{-1} = 0$ below T_c . At low fields, the samples tend to deflect slightly from the center to a position where the field direction is not known.²⁵ Thus the geometric interpretation of this demagnetizing constant is not obvious and should not be taken seriously. It is just an averaged result. The value 75 emu/g kOe for the demagnetizing constant can be verified consistently by noticing that the $M(H)$ curves around T_c (determined later) should intercept the demagnetizing line only at the origin as shown in figure 1.

The possibility of crystalline gadolinium precipitates in the matrix can be observed by noticing that for $T \gtrsim 190^\circ\text{K}$ (far from T_c), $M(H)$ is linear for $H > 5$ kOe but exhibits appreciable curvature at lower fields. The latter effect is isolated by plotting as shown in figure 3, $M'(H) = M(H) - \chi_o H$ for temperatures above 190°K , where χ_o is the initial susceptibility determined from the linear portion of $M(H)$ for $H > 5$ kOe. A general feature of these curves is immediately observed. There is a saturation field $H > 5$ kOe for $M'(H)$ almost independent of temperature, and a systematic decrease in M'_s (saturation magnetization) for increasing temperature, characteristic of ferromagnetic precipitates. As found in other amorphous splat-cooled alloys²⁷, this amount of ferromagnetic precipitates varies between the center and the foil edges, but this quantity once subtracted from the total magnetization, the remaining contributions attributed to the amorphous phase was found to be the same for a given foil. The demagnetizing constant determined above is

Fig. 3. Magnetization resulting from precipitates in amorphous matrix versus applied field for different temperatures.

not affected by this small correction with a maximum $M'_s(T)$ of ~ 5 emu/g. To verify that the $M'(H)$ values are in fact due to ferromagnetic contributions from macroscopic gadolinium precipitates in the amorphous matrix, we plot, in figure 4 the Debye-Weiss quantum theory curve for $S = \frac{7}{2}$ using one $M'_s(T)$ value from figure 3 and $T_c \approx 293^\circ\text{K}$.¹⁸ The other $M'_s(T)$ values for $T > 175^\circ\text{K}$ are also plotted and observed to agree well with the theory. The values of crystalline gadolinium for $T/T_c \gtrsim 0.6$ had been reported^{18, 28} to agree well with the Debye-Weiss curve for $S = \frac{7}{2}$. Thus the raw data presented in section II are processed to give corrected values of $M(H, T)$ for use in the next section.

B. Critical Behavior and the Magnetic Equation of State.

(i) Critical Exponents

The second order phase transtion around the Curie point is characterized by a set of critical exponents and a magnetic equation of state. Relations between the exponents are obtained by the scaling hypothesis approach developed by several authors.^{29, 30, 31} The results of the static scaling approach indicate that the exponents should be adequately described by two parameters. Here we will discuss the three exponents β , γ and δ for the present system. The exponents β and γ are those describing the temperature dependence of the spontaneous magnetization ($H = 0$) just below T_c

$$M_s \propto |T - T_c|^\beta \quad (1)$$

and that of the initial susceptibility just above T_c

$$\chi_o \propto |T - T_c|^{-\gamma} \quad (2)$$

Fig. 4. Saturation magnetization M'_s from figure 3 versus reduced temperature T/T_c . The Debye-Weiss theory curve for spin $S = 7/2$ is included for comparison.

The exponent δ describes a relation between M and H at T_c

$$M \propto H^{1/\delta} \quad (3)$$

Scaling law implies that

$$\gamma = \beta (\delta - 1) \quad (4)$$

and a simple form of the magnetic equation of state in the critical region given by

$$h/m = f_{\pm}(m) \quad (5)$$

where the plus and minus signs denote temperatures above and below T_c respectively. The normalized quantities are $m = M/|1 - T/T_c|^{\beta}$ and $h = H/|1 - T/T_c|^{\beta\delta}$.

The exponents of the amorphous $Gd_{80}Au_{20}$ alloys will be determined by the graphical method using equations (1) to (3). A conventional technique developed by Arrott³² and Kouvel³³ was to plot the data at various temperatures as M^2 versus H/M . This convenient method was based on the molecular-field theory. It can be understood by expanding the self-consistent mean field relation $M = M_0 B_s \left(\frac{\mu(H + \lambda M)}{kT} \right)$ for small H around T_c , where B_s is the Brillouin function for spin quantum number S , μ is the magnetic moment per atom, and λ is the exchange coupling constant. It can be shown that $\beta = \frac{1}{2}$, $\gamma = 1$, and $\delta = 3$ using this approach. The intercepts along the M^2 axis (i.e. $\frac{H}{M} = 0$) and $\frac{H}{M}$ axis (i.e. $M^2 = 0$) are expected to give the square of the spontaneous

magnetization M_s^2 and inverse initial susceptibility χ_o^{-1} respectively. If $\delta = 3$, the initial slopes of M^2 versus $\frac{H}{M}$ should be almost equal by considering the equation of state around T_c , and the intercepts can be determined conveniently. Unfortunately, it has been observed that δ is closer to 4 than 3 which gives large gradients $dM^2/d(H/M) \propto |T - T_c|^{\beta(3-\delta)}$ near T_c . The fact that the isotherms $M^2(H/M)$ and the M^2 -axis are almost parallel introduces difficulties in determining M_s^2 accurately. In fact, experimental results in the literature¹² indicate that it is easier to obtain $(H/M)_{M=0}$ than M_s^2 because of the large gradients near T_c .

Following the work of Arrott and Noakes¹³, Mizoguchi etc.¹⁰ obtained M_s and χ_o by plotting $M^{2.5}$ versus $(H/M)^{0.75}$. However, there is no a priori reason to plot $M^{1/\beta}$ versus $(H/M)^{1/\gamma}$ since the exponents are not yet determined. Even if β and γ are known, it is not clear that this plot is physically meaningful or that it does give the correct values of M_s^2 and χ_o^{-1} . In fact, the equation of state proposed in reference 13 is in conflict with the results of Kouvel et al²⁰ at low fields. We are going to proceed in a slightly different fashion. We first determine $\chi_o^{-1}(T)$ using the conventional M^2 versus H/M plot. Equation (2) tells us that a plot of the inverse of $\frac{d \log \chi_o^{-1}(T)}{dT}$ versus T should give a straight line near T_c . The reciprocal gradient of the straight line and its intercept with the T -axis should give the exponent γ and T_c respectively. Knowing T_c , we plot $\log M$ versus $\log H$ from equation (3) to obtain δ . These values of γ and T_c can be checked consistently when we determine β later. Then we plot M^p versus H/M to determine M_s^p , where p is a convenient integer slightly larger than $\delta - 1$. It is hoped that the gradients $dM^p/d(H/M) \propto |T - T_c|^{\beta(p+1-\delta)}$ near T_c are finite and almost equal to the value of $dM^{\delta-1}/d(H/M)$ at T_c . Using equation (1),

a plot of the inverse of $d \log M_x / dT$ versus T should give a straight line with gradient equal to β^{-1} and intercept at T_c along the T axis.

In figure 5, M^2 is plotted against H/M for $T > T_c$. The intercepts at $M = 0$ give the reciprocal initial susceptibility χ_o^{-1} . From a plot of $\log \chi_o^{-1}$ versus T , the derivatives $\frac{d \log \chi_o^{-1}}{dT}$ are obtained at various temperatures. Figure 6 shows a plot of the inverse of these derivatives versus T . The straight line obtained from least square fitting gives values of $\gamma = 1.294$ and $T_c = 149.45^\circ K$. It should be mentioned that these results have been checked by an iterating scheme together with the determinations of β and δ . Although the routine process is not presented here, yet these values should be considered as final results. In figure 7, values of $\log M$ versus $\log H$ are presented at the two temperatures $149^\circ K$ and $150^\circ K$ for fields ranging from 500 Oe to 70 kOe. The reciprocal gradients so obtained represent the values of δ for the two temperatures. They are found equal to 3.976 and 3.936 respectively. Since these two values are almost equal for temperatures $\pm 0.5^\circ K$ from T_c , it might not be meaningful to determine δ by repeating a measurement of $M(H)$ at $149.45^\circ K$. Perhaps it is more relevant to compare these values of δ with the one obtained from equation (4) knowing β and γ . Knowing δ , the aforementioned parameter $p (\approx \delta - 1)$ is taken to be 3. In figure 8, M^3 is plotted versus H/M . It is seen that the lines are almost parallel at small H/M for temperatures ranging from $149^\circ K$ to $136^\circ K$. The finite initial gradients make it easier to determine M_s^3 and thus M_s . The inverse of $\frac{d \log M_s}{dT}$ plotted against T is illustrated in figure 9. The straight line from a least square fit determines $\beta = 0.439$ and $T_c = 149.45^\circ K$ after the iterating process. Substituting $\gamma = 1.294$ and $\beta = 0.439$ in equation (4) implies $\delta = 3.948$ which is

Fig. 5. M^2 versus H/M for every °K from 150°K to 160°K.

Fig. 6. The inverse of $\left(\frac{d \log \chi_0^{-1}}{dT}\right)$ versus temperature gives the critical exponent γ and Curie temperature T_c according to text.

Fig. 7. $\log M$ versus $\log H$ for 149°K and 150°K .

Fig. 8. M^3 versus H/M for every °K from 140°K to 150°K. The curve for 138°K is also included.

Fig. 9. The inverse of $\left(\frac{d \log M_s}{dT}\right)$ versus temperature gives the critical exponent β and Curie temperature according to text.

intermediate of the values determined earlier for $T = 149^{\circ}\text{K}$ and 150°K . It should be noticed that the asymptotic relations of the critical phenomena seem to have different regions of validity for temperatures below and above T_c . For $T < T_c$, it is observed to be $|1 - T/T_c| < 0.06$; while for $T > T_c$, it is $|1 - T/T_c| < 0.04$.

Model calculations of the critical exponents have been made in the past. Besides the well known molecular field results, values of γ , β and δ have been evaluated for the Ising and Heisenberg models for several dimensionalities D and spin quantum numbers S . In these model calculations, two assumptions are important, namely the configurational and short-range nature of the interactions. Experiments have given different critical-point properties for various ferromagnetic systems. A summary of theoretical and representative experimental results on different ferromagnetic materials are presented in Table 1. Two gadolinium monocrystal results denoted by (e) and (f) are included. Results (e) represent measurements on a spherical sample and results (f) are those made along the $\langle 0001 \rangle$ easy axis. The experimental values, with the exception of CrO_2 lie between the Heisenberg (isotropic, nearest-neighbor interactions) values and molecular field (isotropic, long-range interactions) values. It has been suggested³⁴ that the molecular field effect on β might be noticed for experiments done under the condition $(1 - T/T_c) \ll (\mu M_o / kT_c)^{1/\beta(\delta-1)}$. For nickel, this condition corresponds to $(1 - T/T_c) \ll 0.01$. For the Gd-Au alloys, the region of validity for β mentioned earlier falls well inside this mean field region. The small dipole forces ignored in the Hamiltonian do not modify the thermodynamic functions very much at lower temperatures. However, very close to T_c when the delicate and long-range

Table 1. Experimental values of critical exponents γ , β , and δ for amorphous and crystalline materials. Theoretical values obtained from the high temperature series expansion method are also included.

	γ	β	δ
Ising 3D ^(a)	5/4	5/16	5.2
Heisenberg ($S = \infty$)	4/3 ^(b)	0.38 ^(c)	4.2
Molecular field	1	1/2	3
Nickel ^(b)	1.34	0.378	4.58
CrO ₂ ^(d)	1.63	0.34	5.79
Gadolinium (monocrystal) ^(e)	1.196	0.381	3.61
Gadolinium (monocrystal) ^(f)	4/3		4
Amorphous Co ₇₀ P ₂₀ B ₁₀ ^(g)	1.342	0.402	4.39
Amorphous Fe ₈₀ P ₁₃ P ₇ ^(h)	1.30	0.38	4.47
Amorphous Gd ₈₀ Au ₂₀ ⁽ⁱ⁾	1.294	0.438	3.948

(a) Ref. 15 and 20, and references therein.

(b) Ref. 20 and references therein.

(c) R. L. Stephenson and P. J. Wood, J. Phys. (c), Solid State Physics 3, 90 (1970).

(d) Ref. 12

(e) Ref. 15

(f) Ref. 19

(g) Ref. 10

(h) Ref. 16

correlations are very sensitive to small perturbations, it might be expected to produce quite different critical behavior from that produced by the short-range exchange forces alone. Thus the small departures from the Heisenberg values might not be surprising. Besides the dipole forces, one should also consider anisotropic interactions due to crystal-field effects and the possibility of long-range antiferromagnetic couplings which will be discussed later. The anisotropic crystal-field effects arise from the fact that long-range isotropy does not exclude local anisotropy in the amorphous state. However, any antiferromagnetic couplings limited to within nearest-neighbors do not change the critical exponents obtained from the Heisenberg model.³⁴ One should also notice that the value $\beta = 0.38$ was obtained by the method of high-temperature expansions to evaluate the low-temperature critical exponents. The discrepancies between the experimental and model values are not totally understood in a quantitative sense.

It will be shown that the M-H-T data ($136^\circ\text{K} < T < 160^\circ\text{K}$, $0 < H < 40 \text{ kOe}$) for $\text{Gd}_{80}\text{Au}_{20}$ alloys satisfy equations (4) and (5), the scaling law result representing a continuous phase transition. A definite Curie temperature is also observed. The usual scheme of the scaling hypothesis³¹ is to mentally divide an Ising model lattice into square cells of length La_0 , where a_0 is the lattice constant; L is chosen such that it is smaller than a coherence length ξ , but large enough to contain many spin sites. Correspondingly, in an amorphous matrix it might be appropriate to introduce the average nearest spin site distance r_0 as the relevant parameter (c. f. the nearest-neighbor distance from a radial distribution function). For an appreciable number of spin sites within a cell, i.e. $L \gg 1$, the microscopic randomness is averaged out. In the critical region when the magnetic fluctuation becomes long-ranged

(i.e. $\xi \gg Lr_0$), these cells might couple each other the same way as in the crystalline case. It might be fair to say that the very long-range fluctuations "overlook" the structural long-range disorder in the critical region. To interpret the critical exponents of an amorphous ferromagnet, the discussion of the last paragraph holds except that the strength of the antiferromagnetic couplings is now reduced by an exponential factor $e^{-r/\ell}$ (ℓ is the electronic mean free path)³⁵ due to long-range structural disorder.⁶

(ii) Magnetic Equation of State.

The asymptotic form of equation (5) for large m , that is when the field effect dominates over the thermal effect is $f_{\pm}(m) \rightarrow m^{\delta-1}$. It can be said that equation (5) is another version of equation (3) when $m \rightarrow \infty$. However, it is not clear how much can H be increased before equation (3) which is supposed to be valid for small M breaks down. In the present study, it is observed that equation (3) is valid for fields up to 70 kOe. It might be interesting to check equation (3) at even higher fields (e.g. $H \sim 150$ kOe). For small m , the interplay of magnetic field and thermal effects becomes important. The asymptotic forms are then obtained experimentally for materials with different magnetic properties. In CrBr_3 , Ho and Lister³⁶ observed the relation $h \propto m$ for small m . Kouvel and Comly²⁰ obtained $h/m \propto m^2$ for nickel and CrO_2 . Thus it is expected that the general asymptotic form of equation (5) to be given by $h/m = A_{\pm} + B_{\pm} m^2$ where the $\pm$ signs have the usual meaning. Using the definition of h and m , one can show that A_+ is given by the constant in equation (2), and $(\frac{A_-}{B_-})^{\frac{1}{2}}$ is the constant in equation (1) which can be determined using the $M_s(T)$ and $\chi_o^{-1}(T)$ data. Other forms of equation (5) have been obtained^{37, 38} and tested experimentally³⁹ for all m and h .

using the polar parameters r and θ . Here, we do not attempt to deal with this problem in detail. Instead, we just plot m^2 versus h/m using the present data. Figure 10 shows this plot in two curves for temperatures below and above T_c . The central dash line represents the asymptotic form of the curves for large m . Thus equation (5) is obeyed over the entire range of the normalized variables.

In order to check the formerly established asymptotic relations for small m , we make a log-log plot of the small m data to verify the power law relation between m^2 and h/m . In figure 11, $\log m^2$ is plotted against $\log (h/m - A_+)$ where A_+ as determined from the $\chi_o^{-1}(T)$ data using equation (2) is found to be 1.86×10^3 Oe g/emu. Similarly, $\log (m^2 - \frac{A_-}{B_-})$ versus $\log (h/m)$ is plotted in figure 12 with $\frac{A_-}{B_-} = 3.46 \times 10^4$ emu²/g² determined from $M_s(T)$ data using equation (1). Two features can be realized from these plots. First, the asymptotic relations $h/m \rightarrow A_{\pm} + B_{\pm} m^2$ as $m \rightarrow 0$ are established for temperatures below and above T_c for small m . The relations for temperatures higher and lower than T_c are given by $h/m = 1.86 \times 10^3 + 0.19m^2$ and $h/m = 0.164m^2 - 0.566 \times 10^4$ respectively. Second, for a given temperature there is a critical field above which the asymptotic relations ($m \rightarrow 0$) are no longer obeyed. The gradients become smaller, probably tending towards the large m asymptotic gradient given by equation (3). The magnitude of the critical field increases with the temperature difference $|T - T_c|$, meaning higher fields are required to compensate for the thermal effects introduced by the temperature difference $|T - T_c|$ mentioned earlier. It is interesting to note that for $|T - T_c| \sim 12^\circ\text{K}$, the asymptotic relations are valid up to 40 kOe.

Fig. 10. The reduced magnetization m^2 versus reduced inverse susceptibility χ_0^{-1} for temperatures around T_c . The dashed line indicates asymptotic behavior of the two curves for large m above and below T_c .

Fig. 11. $\log m^2$ versus $\log (h/m - A_+)$ determines the asymptotic equation of state for $T = T_c^+$.

Fig. 12. $\log (m^2 - \frac{A_-}{B_-})$ versus $\log h/m$ determines the asymptotic equation of state for $T = T_c^-$.

C. Magnetic Moments and Exchange Interactions.

(i) Determination of magnetic moments.

The saturation magnetization was determined by the semi-empirical expression⁴⁰

$$M(H, T) = M(\infty, T) \left(1 - \frac{a}{H}\right) + \chi_{\infty} H \quad (6)$$

where χ_{∞} is the high field susceptibility, and $M(\infty, T)$ is the saturation magnetization at $T^{\circ}\text{K}$, and $a = \text{constant}$. The value of χ_{∞} varies between different foils of the same nominal composition. The values of $M(\infty, T)$ thus obtained are plotted as a function of $T^{3/2}$ in figure 13. It is observed that the data agree well over the temperature range $0.13 < T/T_c < 0.80$ with an expression of the Bloch law form

$$M(\infty, T) = M(\infty, 0) (1 - bT^{3/2}) \quad (7)$$

where $M(\infty, 0)$ is the absolute saturation magnetization, $b = \text{constant}$. The departure from the straight line below 20°K is small. Several attempts have been made to explain similar departure below 50°K in crystalline gadolinium.^{41, 42}

The $\text{Gd}_{80}\text{Au}_{20}$ sample which was measured in full detail as a function of temperature in section B gives $M(\infty, 0) = 192 \text{ emu/g}$. Low temperature magnetization measurements on three different foils of the same nominal composition exhibited some scattering (~ 5 per cent) between the values of $M(\infty, 0)$. The averaged value of the mean moment per atom is estimated to be $5.6 \mu_B$. The concentration range in which the amorphous phase can

Fig. 13. Inverse susceptibility χ_0^{-1} versus temperature for $T > T_c$.

be retained from splat cooling is rather small (a few per cent around 20 at.% Au). From measurements on $\text{Gd}_{78}\text{Au}_{22}$ samples, it is believed that the mean moment per atom $\bar{\mu}$ varies with c (the Au content) according to the dilution law $\bar{\mu}(c) = \mu_{\text{Gd}}(1 - c)$, where μ_{Gd} is the moment per atom in pure amorphous Gd. Thus, the Au atoms are unlikely to carry a substantial moment, and μ_{Gd} is estimated to lie between 6.9 and 7.0 μ_B .

In figure 14, the ratio of $\frac{M(\infty, T)}{M(\infty, 0)}$ is plotted as a function of the ratio T/T_c . Two curves obtained from the Debye-Weiss Theory of ferromagnetism (molecular field) for $J = \frac{7}{2}$ and ∞ are also plotted. The crystalline Gd data are included for comparison. The $\text{Gd}_{80}\text{Au}_{20}$ data fall between the crystalline gadolinium data and the $J = \infty$ curve. However, the meaning of this result must not be taken too seriously, for as discussed in section B(i), $\text{Gd}_{80}\text{Au}_{20}$ does not characterize a molecular field magnet even near T_c .

The agreement of the temperature dependence of the saturation magnetization for crystalline Gd with the Bloch law expression (7) for $0.17 \leq T/T_c \leq 0.80$ was explained⁴³ by including the higher order terms beyond the quadratic spin-wave dispersion in spin waves calculations. In the case of crystalline gadolinium with a hexagonal structure, it was found that the higher order contributions cancel each other to yield a $T^{3/2}$ behavior. However, it is not clear that the same argument holds for the amorphous $\text{Gd}_{80}\text{Au}_{20}$ alloys. The short-range order as determined from RDF on $\text{La}_{80}\text{Au}_{20}$ ⁴⁴ implies that each Gd atom on the average has only eight nearest-neighboring Gd atoms and three nearest-neighboring Au atoms.

In figure 15, the initial susceptibility $\chi_0^{-1}(T)$ is plotted as a function of temperature. It is seen that $\chi_0^{-1}(T)$ is linear for $T > 175^\circ\text{K}$. The

Fig. 14. Saturation magnetization $M(\infty, T)$ versus $T^{3/2}$.

Fig. 15. Reduced saturation magnetization $M(\infty, T)/M(\infty, 0)$ versus reduced temperature T/T_c . The Debye-Weiss theory curves for $J = 7/2$ and ∞ , and Gd crystalline data are included.

linear portion of the plot is a continuation of the non linear portion of $T = T_c^+$ where the critical phenomena are observed. The paramagnetic Curie temperature θ_p is determined to be $\sim 164^\circ\text{K}$. Assuming that the effective magnetic moment μ_{eff} is mainly due to the Gd atoms since the f shells of the Au atoms are filled, the value of μ_{eff} determined from the slope of $\chi_o^{-1}(T)$ equals 9.37 Bohr magnetons. This gives a total angular quantum number of $gJ = 8.37$ using the relation $\mu_{\text{eff}} = g [J(J + 1)]^{\frac{1}{2}}$, where g is the spectroscopic splitting factor. It is higher than the theoretical saturation moment of 7.0 Bohr magnetons for the ionic $^8\text{S}_{7/2}$ state which does not include conduction electron effects. The values of $M(\infty, 0)$, μ_{eff} , T_c and θ_p for amorphous $\text{Gd}_{80}\text{Au}_{20}$ and crystalline Gd are listed in Table 2.

(ii) Comparison between the values of magnetic moments in the amorphous and crystalline cases.

In crystalline Gd and most of the rare-earth elements, the values obtained for the saturation magnetization and the effective moment are very close, and the experimental values of μ_{eff} are practically the same as the ionic values.⁴⁵ But in quite a number of Gd alloys and intermetallic compounds containing Gd, the saturation moments at 0°K were found to be smaller and the effective moments greater than the corresponding values in pure Gd.^{46, 47, 48} This effect was accounted for by the antiferromagnetic couplings of some Gd moments at low temperatures. Gold is known to favor the antiferromagnetic alignments of the Gd atoms, as in the case of the stoichiometric Gd-Au antiferromagnetic compound.⁴⁹ In amorphous $\text{Gd}_{80}\text{Au}_{20}$, this relatively important effect should come mainly from the nearest-neighbors interactions.

Table 2. Parameters derived from magnetization measurements for amorphous $\text{Gd}_{80}\text{Au}_{20}$ alloys and monocrystalline Gd.

	$T_c(^{\circ}\text{K})$	$\theta_p(^{\circ}\text{K})$	$\mu_{\text{eff}}^{(a)}(\mu_B)$	$M(\infty, 0)(\text{emu/g})$
Amorphous $\text{Gd}_{80}\text{Au}_{20}$	149.45	165	9.37 ^(b)	192
Crystalline Gd ^(c)	293	320	7.94	268.4

(a) $\mu_{\text{eff}} = g[J(J + 1)]^{\frac{1}{2}}$, some authors use p_{eff} .

(b) μ_{eff} in μ_B per gadolinium atom.

(c) Ref. 18, $M(\infty, 0)$ for c axis.

The enhanced moment μ_{eff} observed in Gd alloys and compounds is usually explained by the polarization of the s-d conduction electrons in Gd.⁴⁶ Any possible polarization of the 6s electron in Au should result in a smaller effect. For Gd metals, Harmon et al¹⁷ found by using the augmented-plane-wave method that the conduction-electron spin density is of mostly d character. The nearly complete d-like character of the wave functions found at the Fermi level and the overlap between the 4f states and the d-like wave functions account for the magnetic properties of gadolinium. That is, the d electrons determine the coupling of the 4f local moments below the Curie temperature. In Gd, the enhancement of $0.55 \mu_B$ over the ionic value $7.0 \mu_B$ below T_c is possibly explained by the conduction electron polarization effect. However, this effect is apparently absent above T_c . On alloying with a second element, the diluent impurities potential might add an orbital contribution to the Gd spin angular momentum through an s-d-f interaction, or might change the probabilities of the conduction electron-f state interactions. The detail mechanism of the enhancement in μ_{eff} is not totally understood at present. It might be related to the local ordering in the amorphous phase. This explanation in terms of band structure suggests that the electronic structure of amorphous $\text{Gd}_{80}\text{Au}_{20}$ does not differ significantly from that of some equivalent crystalline compound.

In addition, it might also be possible that the observed results of $\mu_{\text{Gd}} < gJ$ and $\mu_{\text{eff}} > [gJ(J+1)]^{\frac{1}{2}}$ are caused by the amorphousness of the matrix. An explanation of the reduced μ_{Gd} in the amorphous state based on the crystal field effects has been formulated by Darby.⁵⁰ Beck and Flynn⁵¹ proposed that the enhanced μ_{eff} in crystalline nickel was caused by the short-range magnetic ordering above the Curie temperature.

This local ordering persists up to $\sim 2T_c$ in crystalline Ni. Although this effect is apparently absent in crystalline Gd, yet one should not ignore its possible role in the amorphous state when local interactions are particularly important.

(iii) Exchange interactions.

There are two conventional methods of estimating values of the effective nearest-neighbor exchange interaction J_n in ferromagnets based on the Heisenberg model. One is by fitting the spin-wave theory to the experimental saturation magnetization at low temperature using the following expression²¹

$$b = \frac{0.0587}{QS} \left\{ \frac{k_B}{2J_n S} \right\}^{3/2} \quad (8)$$

where b is the constant in equation (7), S is the ionic spin quantum number, Q equals 1, 2, 4 for simple cubic, bcc, fcc structures respectively. However, it must be remembered that (7) is valid over a large temperature range ($0.13 < T/T_c < 0.8$). A second method due to Rushbrooke and Wood²² results from expansion of the susceptibility above the Curie temperature in powers of $J_n/k_B T_c$ which gives

$$\frac{k_B T_c}{J_n} = \left(\frac{5}{96}\right) (z-1) [11 S(S+1) - 1] \quad (9)$$

where z is the number of nearest neighbors. In crystalline gadolinium one obtains $J_n = 2.9^\circ\text{K}$ from (9) knowing T_c . However, a value of 1.8°K was obtained²⁸ on the basis of elementary spin-wave theory which gives (8). The discrepancy between the high-and low-temperature J_n values was

explained by Goodings.⁴³ It is probably caused by the interactions beyond nearest neighbors and it was conjectured that these interactions are of the antiferromagnetic RKKY type.⁵² The magnitude of these long-range exchange interactions increases from the high-temperature molecular field regime (χ_o^{-1} linear in T) to the low-temperature spin-wave regime. In the amorphous state, the long-range interactions are expected to be attenuated significantly due to structural randomness as discussed in section B (i).

In evaluating the values of J_n for $Gd_{80}Au_{20}$ alloys using (8) and (9), several considerations must be made. It has been a custom^{53, 54} to evaluate the average exchange integral J'_n in Gd alloys using the spin quantum number $\frac{7}{2}$ and the crystalline Gd structure factors (i.e. $Q = 4$, $z = 12$). Then the effective integral of the indirect exchange interaction J_n between two neighboring Gd ions is obtained from the formula

$$J_n = \frac{J'_n}{(1 - x)^2} \quad (10)$$

where x is the atomic concentration of the impurity atoms. However, it is not clear physically that this simple dilution model (i.e. averaging the effects of Gd-Gd couplings and Gd-non-magnetic ions couplings) should give the correct value of J_n in the present case. Hence, we would rather use a dilution model which takes into account the short-range ordering in the amorphous phase. That is, instead of using the dilution model from (10), appropriate values of Q and z from RDF results are taken. Based on the results of reference 44 together with the assumption that the major interactions come from Gd-Gd nearest neighbors, $z = 8$ and $Q = 2$ for the bcc values are used. Taking

$T_c = 149.45^\circ\text{K}$ and $gS = 7$, the values of J_n obtained from (8) and (9) are found to be 1.34°K and 2.28°K respectively, which are lower than the crystalline values. If the conduction-electron polarizations are included, then one should use $gJ = 8.37$ in place of $gS = 7$ in (9). A value of $J_n \sim 1.66^\circ\text{K}$ is obtained. It is rather difficult to attribute the difference between the two J_n values to the long-range antiferromagnetic couplings as suggested by Goodings since the nearest-neighbors antiferromagnetic couplings seem to be dominating below T_c in the amorphous state. However, the role of impurity Au atoms and Gd-Gd local ordering on the diminishing of the indirect exchange integral J_n is not known.

Summarizing, by alloying with Au, the Gd moments experience some trend of nearest-neighbors antiferromagnetic couplings in the amorphous case as well as in the crystalline Gd alloys and compounds. However, this must be distinguished from the long-range antiferromagnetic interactions of the RKKY type. The structural disorder in amorphous $\text{Gd}_{80}\text{Au}_{20}$ results in a considerable lowering of the long-range interactions. It is suggested that the difference in J_n values is mainly caused by nearest-neighbors antiferromagnetic couplings between Gd atoms below the Curie point.

(iv) Effects of structural disorder in amorphous $\text{Gd}_{80}\text{Au}_{20}$.

We mentioned earlier the attenuation of the long-range interactions in the amorphous $\text{Gd}_{80}\text{Au}_{20}$ alloys. Recent calculations were made on the effects of local anisotropy (crystal-field) on the saturation moment and Curie temperature of amorphous rare-earths.⁵⁰ Unfortunately, we are not able to give an estimation of this effect on the present system, since the major reduction in $M(\infty, 0)$ is due to the short-range antiferromagnetic

couplings of Gd atoms in the presence of Au impurities. However, if the antiferromagnetic couplings are not taken into account, then the structural disorder should result in a maximum lowering of 7 per cent for μ_{Gd} . On the other hand, by using the Rushbrooke-Wood formula, it is possible to give a rough estimation of the T_c for amorphous Gd. Assuming the same number of Gd-Gd nearest neighbors in the amorphous state as in the crystalline state and $J_n \sim 2.28^\circ\text{K}$ for $T > \theta_p$ (i.e. in the absence of antiferromagnetic couplings and conduction-electron effects), T_c is estimated to be 228°K . That is, a lowering of 22 per cent for T_c in the amorphous state. Thus, structural disorder effect seems to be more detrimental for T_c than for μ_{Gd} , which is in contrary to Darby's calculations. A molecular field model so far does not seem appropriate for amorphous Gd, and Handrich's approach based on the Heisenberg model would be more realistic in this case.⁵⁵ It is found that $\text{Gd}_{78}\text{Au}_{22}$ alloys have essentially the same $T_c (\pm 1^\circ\text{K})$ as $\text{Gd}_{80}\text{Au}_{20}$ alloys. The insensitivity of T_c to small variation in Au content probably results from the inertness of the f-electrons in the presence of Au impurities.

IV. SUMMARY AND CONCLUSION

From magnetization measurements on amorphous samples of $\text{Gd}_{80}\text{Au}_{20}$, Curie temperature T_c and critical exponents β , γ and δ are determined. The data are also fitted to an equation of state previously derived for a second order phase transition in fluid systems. The criteria of a second order phase transition are satisfied. The values of $\beta = 0.439$, $\gamma = 1.294$, and $\delta = 3.95$ so obtained are compared with the theoretical Heisenberg and molecular field values. The

effects of crystal-fields due to short-range anisotropy and the growing importance of dipole forces as T_c is approached on the critical behavior in the amorphous state are emphasized. The long-range interactions are significantly reduced by the very short electronic mean free path as noted in amorphous alloys. The magnetic moments are determined both from the low-temperature saturation magnetization and high-temperature susceptibility data. The effective moment in the paramagnetic state has a value of 9.37 Bohr magnetons per Gd atom which is higher than the $^8S_{7/2}$ ionic value of 7.94. The low temperature saturation magnetization observes the $T^{3/2}$ law up to $0.8 T_c$. The saturation moment extrapolated to 0°K gives $7.0 \mu_B$ per Gd atom which is lower than the value $7.55 \mu_B$ observed in crystalline Gd. The effective exchange integral J_n are determined both from the Rushbrooke-Wood formula for $T > T_c$ and low-temperature spin-wave theory. They are found to be 2.28°K and 1.34 respectively. The difference is attributed to the nearest-neighbor antiferromagnetic couplings in the presence of Au atoms below T_c . The effect of structural disorder on the magnetic properties of Gd in the amorphous state is found to be rather small.

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