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MAGNETIC CLUSTER MEAN-FIELD DESCRIPTION OF SPIN GLASSES IN AMORPHOUS La-Gd-Au ALLOYS

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by

S. J. Poon⁺ and J. Durand⁺⁺

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ABSTRACT

The bulk magnetic properties of splat-cooled amorphous alloys of composition $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ ($0 \leq x \leq 80$) have been studied. Zero-field susceptibility, high-field magnetization (up to 75 kOe) and saturated remanence have been measured for temperatures ranging between 1.8 and 290°K. Detailed analysis of the data based on a cluster mean-field approximation for the spin-glass and micromagnetic alloys ($x \leq 56$) is presented. It is shown that mean-field theories can account for the experimental freezing-temperatures of dilute spin-glasses in which the Ruderman-Kittel-Kasuya-Yosida interaction is dominant. For our dilute alloys, the role of amorphousness on the magnetic interactions is discussed. By extending the mean-field approximation, our concentrated spin-glasses are represented by rigid ferromagnetic clusters as individual spin-entities interacting via random forces. Scaling laws similar to those of Blandin, Souletie, Tholence and Tournier for the magnetization M and saturation remanent magnetization M_{rs} are obtained and presented graphically for the $x \leq 32$ alloys in which $M/x = g(H/x^*, T/x)$, $M_{rs}(T)/x = \frac{M_{rs}(0)}{x} \exp(-\alpha^* T/x^p)$ where x^* is the concentration of clusters, α^* is a constant, and p is the freezing-temperature exponent given by $T_M \propto x^p$. It is found that $p = 1$ and 1.3 for the regions $4 \leq x \leq 40$ respectively. An attempt is also made to account for the freezing temperatures of concentrated spin glasses. The strength of the interaction among clusters is determined from high-field magnetization measurements using the Larkin-Smith method modified for clusters. It is shown that for the $x < 24$ alloys, the size of the clusters can be correlated to the structural short-range order in the amorphous state. More concentrated alloys are marked by the emergence of cluster percolation.

I. INTRODUCTION

Existence and characteristics of spin-glass regimes in amorphous systems have been a subject of interest only in a recent past.¹ In contrast, dilute spin-glasses in crystalline alloys containing 3d magnetic solutes have been investigated quite extensively for a long time. Numerous reviews on both the experimental² and theoretical³ aspects of these studies are available. Experimentally, various thermodynamical parameters (such as magnetization $M(H,T)$ and specific heat) and the concentration dependence of the freezing temperature T_M (defined by a sharp cusp in the zero-field susceptibility) are describable in a rather quantitative fashion. It is believed that the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction⁴ plays an important role on the observed magnetic properties in crystalline dilute spin-glasses. The same conclusion was obtained for dilute amorphous La-Gd-Au alloys from analysis of magnetic and superconducting properties.⁵ For more concentrated alloys, where short-range effects occur together with long-range RKKY interaction, it is generally difficult to compare the experimental results with existing spin-glass theories. However, in concentrated amorphous La-Gd-Au alloys, the magnetoresistivity results can be attributed to conduction electrons scattered off magnetic clusters coupled by the RKKY interaction.⁶ On the other hand, preliminary data⁷ on saturated remanent magnetization on the same alloys were interpreted in a phenomenological model of uncompensated magnetic clouds.⁸ These observations motivated us to investigate in detail the temperature-concentration dependence of the magnetization and spin-glass phenomena (freezing temperature and remanent magnetization) in the amorphous La-Gd-Au system. Our experimental data are analyzed in a cluster mean-field approximation (CMFA)

within which spin-glasses are represented by magnetic clusters as individual spin-entities interacting with random forces. This study is favored by using an amorphous system containing Gd as magnetic solutes. The possession of a large and localized moment ($7\mu_B$) by Gd 4f-ions immersed in a normal matrix (in this case $\text{La}_{80}\text{Au}_{20}$) and the absence of crystal-field effects allow one to focus on the magnetic interactions (e.g. RKKY, dipolar,...) between localized spins. In addition, the complete substitution of Gd for La in the $\text{La}_{80}\text{Au}_{20}$ matrix preserves a 'single phase' in the amorphous state. This eliminates the metallurgical complexities usually encountered in crystalline binary alloys. On the other hand, amorphous alloys obtained by splat-cooling are thought to be macroscopically more homogeneous than amorphous materials produced by other methods (electrodeposition, co-evaporation, sputtering,...) Thus, this splat-cooled amorphous system seems to be a good candidate to undertake a systematic comparison of experimental magnetic properties with theoretical results on spin-glasses.

In a previous report, we have presented an overview of the magnetic phases in the amorphous $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ alloys.⁹ The magnetic phase diagram was schematically divided into four regions. The 'dilute' alloys ($0.24 < x < 1$ at.%) were found⁵ to behave like canonical spin-glass systems, in that the magnetization and initial susceptibility data follow the universal curves of the scaling predictions.¹⁰ For the $1 < x \leq 32$ alloys, the classical scaling laws for $M(H,T)$ are no longer obeyed. However, the reduced saturated remanence $M_{rx}(T)/x$ scales with T/x .⁷ On the other hand, T_M is roughly proportional to x . According to Ododo and Coles¹¹, these alloys could be termed 'cluster glasses'. For more concentrated alloys, none of the aforementioned regularities is observed concerning T_M , $M(H,T)$. On the other hand, from initial susceptibility

and magnetization data, a long range ferromagnetic order does not seem to exist in alloys containing less than about 56 at.% Gd. This intermediate region ($32 \leq x \leq 56$) could be called 'mictomagnetic'.¹² Finally, the $56 \leq x \leq 80$ alloys are 'good' ferromagnets, in which a Curie temperature T_c is well defined from Arrott plots with critical exponents and equation of state characterizing a second order phase transition.¹³ The present paper will focus on the two first magnetic phases. Our magnetic data will be described within the same CMFA formalism, so that all the alloys up to $x \leq 32$ will be called spin-glasses.

A preliminary problem we have to deal with concerns the effect of structural disorder on spin-glass properties. Analysis of the magnetic-superconducting properties in dilute alloys allows us to investigate the role of amorphousness on the RKKY interaction. The possible attenuation of this interaction in an amorphous matrix where the electronic mean-free-path reaches a few interatomic spacings will be discussed in the present paper. Since our analysis of spin glasses is based on the CMFA, it would be appropriate to check the applicability of the mean-field spin-glass theories of Edwards and Anderson¹⁴ (also extended by other authors¹⁵) in dilute spin-glasses. Such can be done by comparing the mean-field freezing temperatures to the experimental values. The only fitting parameter is the strength of the RKKY interaction. Based on our understanding of the dilute regime, we can investigate the spin-glass mechanism in the more concentrated alloys which are represented by rigid ferro-clusters (i.e. we ignore the internal dynamics of the clusters) as individual spin-entities interacting via random forces. We attempt to extract simple laws¹⁰ using the CMFA by suitable scaling transformations of variables (concentration and size of spin-clusters,

interacting forces,...). The magnitude of the variables are determined from magnetization measurements. Various spin-glass phenomena can be accounted for this way. It should be mentioned that recent theoretical treatments of spin-glasses using the cluster interaction picture have been made. In the Monte Carlo results of Binder et al¹⁶, a sharp cusp in the susceptibility and a rounded maximum at higher temperature than T_M in the specific heat are reproduced. Using a mean-field model of dynamical clusters, Soukoulis and Levin¹⁷ also produced a rounded maximum in the specific heat.

The format of this paper is as follows. In section II, experimental procedures are presented. Section III describes the experimental results based on which the phenomenology in the concentrated regime is discussed. Section IV consists of three subsections from A to C. Section IV A discusses the RKKY interaction in an amorphous matrix based on previously published results. In section IV B, the spin-glass freezing temperatures in dilute alloys are discussed in the light of the mean-field theory of Edwards and Anderson. Section IV C presents a cluster mean-field description of spin-glasses in the concentrated regime. The magnetization and other spin-glass phenomena are accounted for within the framework of this model. The last section is the summary and conclusion.

II. EXPERIMENTAL PROCEDURES

The purity of La and Gd used in this study is 99.9 + %. Alloys of composition $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ with $x = 0, 0.24, 0.5, 1.0, 4.0, 6.4, 9.6, 16, 24, 32, 40, 48, 56, 64$ and 80 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 Ref. 18. 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 about 40 μm . 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 pattern (Cu K_α) of the samples are characterized by a broad maximum the center of which ranges from 30.7° in $\text{La}_{80}\text{Au}_{20}$ to 32.8° in $\text{Gd}_{80}\text{Au}_{20}$ with a full width at half maximum of $\sim 4.6^\circ$. According to the Sherrer formula, this corresponds to an effective microcrystal size of $\sim 17 \text{ \AA}$, which is typical of a glassy metal. No significant annealing effect is observed for the amorphous phases at room temperature during periods of several weeks. Spontaneous crystallization is observed at temperatures of about 150 to 200°C .

Magnetization measurements as functions of magnetic field (up to 75 kOe) and temperature (1.8°K to 290°K) were carried out by using the Faraday method with an Oxford Instruments Magnetometer. Samples used in the $M(H,T)$ measurements were in the form of disks (3mm in diameter) punched from foils. The thermal output controls have an accuracy of $\sim 0.05^\circ\text{K}$. Magnetic ordering temperatures were observed using a standard ac inductance bridge technique.

III. EXPERIMENTAL RESULTS

In this section, we present a qualitative description of our experimental results. Alloys in the region $x \leq 56$ are characterized by susceptibility maxima in low-field measurements and thermomagnetic history

effects (isothermal and thermal remanent magnetization) already discussed in Ref. 7. The dependence of T_M on Gd concentration is illustrated on a log-log scale in fig. 1. It can be seen that T_M varies linearly with x for alloys containing less than 12 at.% Gd. At higher concentrations, T_M increases more rapidly with x . For the dilute alloys ($x \leq 1$), the values of T_M are too low to be measured with our equipment. The $T_M(x)$ dependence will be discussed in a more quantitative fashion later. In fig. 2 are shown the $\chi_0^{-1}(T)$ data taken over a wide temperature range for three concentrated alloys. The paramagnetic regions are clearly established at sufficiently high temperatures giving a well-defined paramagnetic Curie temperature θ_p . This value of θ_p ($\approx 3-5 T_M$) is found to increase with x indicating a stronger trend towards ferromagnetic coupling. The large values of $\theta_p - T_M$ also indicate the presence of ferromagnetic clusters around T_M . These clusters which freeze in their local field below T_M disappear gradually when temperature increases above T_M . Using the classical molecular field approach and assuming that the moment per Au atom and per La atom is zero, the effective number of Bohr magnetons per Gd atom is found to remain approximately constant and close to the value 8 which corresponds to the ionic value 7.94. From the linear portion of $\chi_0^{-1}(T)$ we can define a temperature $\theta_s (> \theta_p)$ corresponding to the complete dissociation of magnetic clusters into paramagnetic spins. The values of T_M , θ_p , θ_s and μ_{eff} as a function of x are listed in Table 1.

In fig. 3, we show the low-temperature magnetization data for the $x = 16$ sample measured in fields up to 70 kOe. The difficulty of saturating the Gd moment at low temperature is observed. Using the magnetization data, the classical Arrott plots (M^2 vs. H/M) are obtained. Such

Fig. 1. 'Ordering' temperatures versus Gd concentration (log-log scale) in amorphous $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ alloys. For $x < 56$ at.% T_M is the temperature of the cusp in zero field susceptibility. For $x > 56$ at.%, the Curie temperature is defined consistently from Arrott plots and a.c. measurements.

Fig. 2. Inverse susceptibility χ_s^{-1} versus temperature for amorphous $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ alloys containing 16, 32 and 48 at.% Gd.

Table I. Magnetic properties of some amorphous $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ alloys.

$x(\text{at.}\%)$	T_M (°K)	$\theta_p^{(b)}$ (°K)	θ_s (°K)	$M_\infty(0)$ ($\mu_B/\text{Gd at.}$)	$\mu_{\text{eff}}^{(b)}$ ($\mu_B/\text{Gd at.}$)	x^* (at.%)	S^*/S (Gd spins/cluster)
0.24	—	~ 0	~ 0	8.09	8.34	0.22	1.09
0.50	—	~ 0	~ 0	8.04	8.58	0.46	1.09
1.00	—	~ 0	~ 0	8.04	8.60	0.92	1.09
4.0	1.9	13	40	8.14	8.12	2.28	1.75
6.4	3.2	15	40	8.40	8.22	3.18	2.01
9.6	4.8	20	55	8.14	8.13	3.70	2.59
16	8.5	36	100	7.72	8.25	5.50	2.91
24	15	50	100	7.45	8.15	6.40	3.75
32	21.5	72	120	7.20	8.30	4.95	6.46
40	28	76	130	6.93	8.58		
48	40	92	130	6.93	8.50		
56	54	114	150	6.91	8.22		
64	78 (a)	155	170	7.28	8.20		
80	150 (a)	165	175	7.00	8.90		

(a) Refer to Curie temperature as determined consistently from Arrott plots and a.c. measurement.

(b) θ_p and μ_{eff} are obtained from Curie-Weiss law at high temperature (up to 290°K).

$M_\infty(0)$ is determined by a 1/H extrapolation.

Fig. 3. Magnetization versus applied field for amorphous $\text{La}_{64}\text{Gd}_{16}\text{Au}_{20}$ at different temperatures.

plots for two samples ($x = 32, 48$) are shown in fig. 4. Strong departures from linearity at small and high fields at temperatures below θ_p are seen, so that any spontaneous magnetization and Curie temperature cannot be defined. However, the $M^2(H/M)$ isotherms are observed to approach closer to the M^2 -axis for higher Gd concentrations indicating a gradual onset of spontaneous magnetization for $x > 56$. The absence of spontaneous magnetization from the Arrott plots for all $T < \theta_p$ also points towards inhomogeneous magnetic interactions.

IV. ANALYSIS AND DISCUSSION

A. RKKY interaction in an amorphous matrix.

Since we are dealing with spin-glass phenomena in amorphous alloys, it is appropriate to investigate the effect of amorphousness on the RKKY interaction. Several authors have treated this subject in the case of a disordered matrix. De Gennes¹⁹ pointed out that incoherent interference of the incident and scattered electronic waves in a disordered system would attenuate the RKKY interaction. He conjectured that the interaction in its asymptotic form is modified by an exponential factor $e^{-r/\ell}$, where ℓ is the electronic mean-free path. Other forms²⁰ of the attenuated RKKY interaction have also been proposed. There have been a few experimental attempts to test these theories. Earlier study was made by Heeger et al²¹ who observed a decrease in NMR linewidth when nonmagnetic Al impurities (which decrease the mean-free-path) were introduced into CuMn system (containing only 500 ppm Mn). Likewise, Souletie²² studied the specific heat of AuFe alloys doped with Ti. He checked the strength of the RKKY interaction based on the intuitive exponential law $e^{-r/\ell_{mfp}}$.

Fig. 4. M^2 versus H/M at different temperatures for $x = 32$ and $x = 48$ alloys.

He found that good agreement was obtained when ℓ_{mfp} is greater than $2R_c$, the average distance separating the near-neighbour impurities. However, for $\ell_{\text{mfp}} < 2R_c$, the interaction became less and less attenuated. For example, when $\ell_{\text{mfp}} = R_c$, the interaction was 50% stronger than the theoretical value. More recent investigations were made on the depression of superconductivity in amorphous superconductors doped with magnetic impurities (Mn). Comparison was then made with that in the crystalline counterpart. In the InMn system, the study was limited to 0.1 at.% Mn, since $T_c \sim 1.3^\circ\text{K}$ at 800 ppm Mn. Comparison with theories of Kondo superconductor seemed to indicate an attenuation in the magnetic interaction. In contrast, such an attenuation was not observed in dilute ($0.24 < x < 1$) $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ amorphous alloys, where a value for the strength of the RKKY interaction was determined consistently from the approach to saturation and from the initial gradient of the superconducting transition temperature as a function of concentration.⁵ On the other hand the magnetization and the initial susceptibility were found⁵ to follow the classical scaling laws deduced from the invariance of xr^3 in the asymptotic form of the RKKY function.¹⁰ Such regularities in an amorphous matrix were unexpected in the light of previous experimental and theoretical investigations on the effect of mean-free-path on RKKY interaction.

In the original treatment of De Gennes¹⁹, the exponential factor was intended to describe the spatial decay of the electronic wave functions in a disordered matrix. Therefore it is not clear that the transport mean-free-path ℓ_{mfp} is the relevant length to be included in the exponential decay, since the former quantity results from a time-dependent-perturbation treatment (conductivity is a time-dependent process). Besides, the characteristic length ℓ_c which measures the range of the

RKKY interaction should be related to the spin-correlation length of the eigenstates in a disordered system. The relationship between these two fundamental quantities is not known up to this point. If $\ell_c = \ell_{\text{mfp}} = 10\text{\AA}$, the RKKY interaction would be significantly modified in the dilute alloys. It would follow that the geometrical arguments leading to the scaling laws would be no longer valid. In addition, it would also result in a more rapid depression of T_c in the Gd doped amorphous superconductors. For the dilute alloys we studied previously⁵ ($0.24 < x < 1$) the average distance $2R_c$ separating two near-neighbor spins varies from 20 to $30\text{\AA}$ ($> \ell_{\text{mfp}}$), so that one would expect a significant modification of the magnetic-superconducting properties. However, our analysis indicated that such is not the case. In fact, the experimental observations resembled those observed in the crystalline alloys. One might tentatively conclude that $\ell_c > 20\text{\AA} > \ell_{\text{mfp}}$. This is also consistent with the fact that the strength of the RKKY interaction was found to conserve the same order of magnitude from dilute LaAuGd alloys to the ferromagnet $\text{Gd}_{80}\text{Au}_{20}$.¹³ This suggests that in order to observe the effect due to ℓ_c , one has to investigate the 10^3 ppm range, as in the CuAlMn and InMn cases. The above conjecture might also explain the 'saturation effect' when $\ell_{\text{mfp}} > 2R_c$ observed by Souletie.²² The exact meaning of ℓ_c awaits further theoretical investigations.

B. The freezing temperature in dilute spin-glasses.

To illustrate the adequacy of the mean-field approximation (MFA) in the analysis of magnetization data, we attempt to account for the freezing temperature in dilute alloys within the frame of the MFA. In this section, we shall deal only with the interactions among single spins.

Experimentally, the freezing temperature T_M is defined as the temperature of the cusp in the susceptibility.

Let us define the interactions between spins located at sites i and j by a Heisenberg Hamiltonian:

$$H = \sum_{i < j} J_{ij} S_i S_j$$

where the interactions J_{ij} are distributed in a random fashion with a mean taken to be zero so as to exclude the possibility of conventional order. In typical spin-glass theories, one obtains an expression for the order parameter m defined by $m^2 = \overline{\langle S_i \rangle^2}$ (where $\langle \rangle$ denotes thermal average and the bar refers to a configuration average). The zero of m^2 then determines T_M . A simple deviation for the order parameter is given by Sherrington.²⁴ Despite the simplicity of the mathematics involved, the expression for T_M thus obtained is in fairly good agreement with those obtained by more rigorous treatments.^{14,15,25} He obtains

$$T_M = [n^2 S(S+1)/3 \sqrt{3} k_B] \overline{(\sum_j J_{ij}^2)}^{1/2} \quad (1)$$

where the average $\overline{(\sum_j J_{ij}^2)}^{1/2}$ is related to the half-width of the molecular field distribution Δ by $\Delta = S(\sum_j J_{ij}^2)^{1/2}$. Using this interpretation of the 'average',^{22,26} one obtains $T_M \propto x$ through $\Delta \propto x$. Such concentration dependence of T_M is shown to be true only for low concentrations of magnetic solutes.^{26,28} In canonical spin-glasses, it is observed that $T_M \propto x$ for $x \sim 0.01$ to 0.1 at.%,^{2,10} while $T_M \propto x^p$ ($0.55 < p < 0.75$) for higher concentrations.² Attempts to explain the deviation of $T_M(x)$ from linearity based on the damping of the RKKY interaction^{26,28} and the 'frustration'

theory²⁸ have been made. For the present purpose of comparing with the MFA, it is sufficient to estimate the gradient dT_M/dx for low concentrations. The starting point is equation (1).

We are dealing with metallic spin-glass system, where the dipolar coupling is much weaker than the RKKY interaction. We assume that the mean-free-path is infinite. Thus we assimilate the J_{ij} 's with the RKKY function in its asymptotic form $V_{ij} = V_o \cos(2k_F r_{ij} + \Phi)/r_{ij}^3$. Following the molecular field approach of Souletie²² inspired from Klein and Brout²⁹ we define R_c as the radius of the average sphere containing one magnetic spin

$$\left(\frac{16\pi}{3}\right) Q \times R_c^3 = d^3 \quad (2)$$

where d is the lattice constant of a cubic cell, and Q equals $\frac{1}{4}$, $\frac{1}{2}$ and 1 for the simple cubic, body-centered cubic, and face-centered-cubic structure respectively. The half-width of the molecular-field distribution of the field created by all the spins outside this sphere is given by

$$\Delta^2 = \frac{2Q \times}{d^3} \int_{R_c}^{\infty} \left[S V_o / r^3 \right]^2 4\pi r^2 dr \quad (3)$$

where the square of the cosine in the RKKY term is averaged by $\frac{1}{2}$. Performing the integration in (3) with R_c given in (2) we obtain:

$$\Delta = 11.85 Q \times S V_o d^{-3} \quad (4)$$

Substituting (4) in (1) yields an explicit expression for the freezing temperature

$$T_M = 2.28 Q \times k_B^{-1} S(S+1) V_O d^{-3} \quad (5)$$

Thus knowing S and V_O , one can determine T_M . The value of V_O used here are determined from high field magnetization measurements as described in Ref. 30,31. From structural study on amorphous $\text{La}_{80}\text{Au}_{20}^{32}$, the La-La distance is $3.74\text{\AA}$ and the La-La coordination number is 8. Thus we regarded the amorphous La-Gd-Au alloys as a bcc lattice ($Q = \frac{1}{2}$; $d = 3.74 \times 2/\sqrt{3}$) in order to calculate T_M/x from equation (5). On the other hand, we used for V_O a value of $0.30 \times 10^{-37} \text{ erg cm}^3$. This value differs from that given in reference 5 ($0.20 \times 10^{-37} \text{ erg cm}^3$). Our new evaluation of V_O comes from a more detailed analysis of the data and from a correction in Larkin's formula (see reference 30). In Table II, we list the values of V_O , S , the theoretical Heisenberg values of T_M/x , and the experimental values of T_M/x for a series of dilute alloys. It can be seen that the experimental values agree well with the theoretical Heisenberg values. It should be noted that there is a definite relationship between the strength of the RKKY interaction and the magnitude of T_M . The weakness of the indirect exchange interaction in rare-earth alloys originates from the localization of 4f electrons in Gd. The latter is thus responsible for the small T_M/x values observed in amorphous La-Gd-Au alloys and in crystalline $\text{La}_3\text{In}(\text{Gd})$ compounds as well (Table II). Within a different context, Tholence and Tournier⁸ already pointed out the proportionality between the 'blocking' temperature T_b and the amplitude of the RKKY coupling. According to these authors there is a distribution of T_b corresponding to the distribution of cloud size within their model, the experimental value for T_M being interpreted as the 'blocking' temperature for the largest clouds.

Table II. Comparison of theoretical and experimental values of T_M in dilute spin-glass alloys.

ALLOY	Q	V_0 (10^{-37} erg cm ³) from approach to saturation	S	T_M/x ($^{\circ}$ K/at.%)	
				Calculated	Experimental
Cu-Mn	1	7.5 (Ref.31)	2.0	15.8	9.5 (a)
Ag-Mn	1	3.5 (Ref.31)	2.0	5.1	4.5 (b)
Au-Mn	1	2.4 (Ref.31)	2.2	4.1	5.0 (a)
Zr-Mn	$\frac{1}{2}$	2.7	2.0	2.8	4.0 (c)
Au-Fe	1	11.0	1.5	10.0	9.3 (d)
Mo-Fe	$\frac{1}{2}$	3.8 - 2.4 (e)	0.9 - 1.2 (e)	1.1 - 1.6	0.6 (f)
am. La ₄ Au(Gd)	$\frac{1}{2}$	0.3	3.8	0.55	0.5 (this work)
cryst. La ₃ In(Gd)			3.5		0.6 (g)
cryst. LaAl ₂ (Gd)	2	0.22	3.5	0.21	0.26(h)

- (a) V. Cannella in Amorphous Magnetism, edited by H. O. Hooper and A. M. De Graaf (Plenum, New York, 1973) p. 195
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Our model neglects the influence of dipolar forces and crystal fields on the freezing phenomena. In most metallic spin-glass systems, the dipolar coupling is two orders of magnitude smaller than the RKKY interaction and thus is too weak to affect T_M . Obviously, this would be different for insulators where spin-glass phenomena were observed.³³ On the other hand, crystal-field anisotropy can be as large as the RKKY force and thus it can affect T_M significantly. Recent experiments^{33,34} and calculations³⁵ clearly illustrate this point. The single-ion anisotropy is absent in alloys containing Gd. Thus we have demonstrated that in an ideal case (i.e., dilute alloys, normal metal, and absence of crystal field effect), spin-glass freezing can be accounted for within the MFA by the isotropic RKKY force alone. In the next section, we shall attempt to explain the deviation of $T_M(x)$ from linearity based on the cluster mean-field approximation.

C. Cluster mean-field description of concentrated alloys.

(i) The model

This approach to study the magnetic properties of amorphous concentrated LaAu Gd alloys is motivated by the following observations:

- (a) the magnetoresistivity results can be analysed in terms of conduction electrons scattered off magnetic clusters coupled by the RKKY interaction.⁶
- (b) As mentioned beforehand, in more concentrated canonical spin-glasses the freezing temperature T_M no longer scales with x . The direct exchange interactions must be important and one has to consider clustering effects. In general, one can treat the clusters (whether they are antiferro- or ferromagnetic, chemical, etc...) as individual spin-entities interacting

with each other. One can use a model in which the distribution in cluster size is given by a probability function (e.g. a Gaussian). Or one can use a mathematically simpler approach in which all the clusters are of the same size.¹⁷ In what follows, we will take the latter approach to study the magnetic properties (magnetization, remanence, and freezing temperature) in our concentrated alloys.

We start with the assumption that our spin-glasses can be described by a picture of ferromagnetic clusters (of equal and constant size around T_M) interacting via random forces. A trivial justification for the ferromagnetic coupling is based on the fact that $\theta_s \gg T_M$ for intermediate concentrations (see Table I). We also ignore the internal dynamics (e.g. spin waves) of these clusters. Thus the conservation of spins yields $x^* S^* = xS$ where x^* and S^* are the concentration and size of the clusters respectively. By ignoring the effect of amorphousness according to section IV, the RKKY local field h_v acting on a cluster v by the other clusters $\{\mu\}$ is then given within the Ising model by

$$h_v = \sum_{\mu} h_{v\mu}$$

with

$$h_{v\mu} = \frac{v_o S^2}{S^*} \sum_{ij} \frac{\cos(2k_F r_{ij} + \Phi)}{r_{ij}^3} \quad (6)$$

where i and j denote spins in clusters v and μ respectively. We have left out the $\pm$ signs in (6). The sum of the cosine terms determines the sign of $h_{v\mu}$ since the spins inside a ferromagnetic cluster all align

along the same axis. The number of terms in the sum equals $(S^*/S)^2$. Thus we can abbreviate the sum term by $(S^*/S)^2$ times an 'average'. The latter is expected to be varying rapidly in sign as a function of the intercluster distance $R_{\nu\mu}$ since the cosine term does (c.f. $k_F d \approx \pi$). To first order approximation, we can combine V_o and the 'average' into a product term $(V_o^*/R_{\nu\mu}^3)f_{\nu\mu}$, where f varies rapidly in sign as a function of $R_{\nu\mu}$ and V_o^* is chosen such that the mean square value of f is half. If r_c measures the radial extent of the spins within a cluster, it is admitted that the above argument is most transparent when the ratio R_c^*/r_c (where R_c^* is defined from the spherical volume containing a cluster) is large so that $R_{\nu\mu} \approx r_{ij}$ even for $R_{\nu\mu} \approx R_c^*$. Knowing x^* from magnetization measurements⁶, R_c^* can be calculated from equation (3) modified for clusters. The trivial case would be the dilute limit, where $R_c^*/r_c = \infty$ by assuming a 'point charge' $4f$ spin. This ratio decreases at increasing x which signifies a trend towards overlapping clusters. Let us assume for the time being that the above argument is adequate for not too concentrated alloys, then (6) can be rewritten as

$$h_\nu = V_o^* S^* \sum_{\mu} \frac{f_{\nu\mu}}{R_{\nu\mu}^3} \quad (7)$$

with

$$\langle f^2 \rangle = \frac{1}{2}$$

Equation (7) is similar to the RKKY local field under the transformation $V_o \rightarrow V_o^*$, $S \rightarrow S_o^*$, $r_{ij} \rightarrow R$, and cosine $\rightarrow f$. It can be viewed as an effective RKKY interaction between clusters. Next, we derive an expression for the effective strength V_o^* from a statistical argument

Table III. Values of R_c^* , r_c , and R_c^*/r_c at four concentrations. These values are obtained from magnetoresistivity results of reference 5. (See text for notation).

$x(\text{at.}\%)$	$R_c^* (\text{\AA})$	$r_c (\text{\AA})$	R_c^*/r_c
1	11.8	0	∞
6.4	7.6	~ 1.80	~ 4.2
9.6	7.0	~ 1.80	~ 3.9
$32^\dagger$	6.7	~ 6.7	~ 1.0

$\dagger R_c^* \sim r_c$ in this case because the number of spins inside a cluster doubles the number allowed from the Gd-Gd nearest neighbors consideration.

similar to Ref. 22. We deduce the second moment of the field distribution from two approaches. First, considering the effective interaction V_o^* between the clusters we obtain $\Delta^2 \propto x^{*2} S^{*2} V_o^{*2}$ similar to the derivation of (4). Then we consider the interaction $V(r)$ between individual spins in the matrix. Since the spins within a cluster are strongly correlated, they can only flip in a rigid way. The effective concentration of 'free' spins is reduced to x^* and we then obtain $\Delta^2 \propto x^{*2} S^2 V_o^2$. Comparing the two results we deduce that

$$S^* V_o^* = S V_o \quad (8)$$

Substituting (8) in (7) reduces the cluster dependent variables in (7) to those contained in the sum $\sum_{\mu}$. Returning to the local field expression in (7) and noticing that the new invariance is $x^* R_{\nu\mu}^3$, we can easily extend the Blandin-Souletie-Tournier argument to magnetic clusters.^{10,22} Since both the effective concentration x^* and total field $(H + h)$ appear in the Brillouin function as $x^* S^* (H + h)/Tx^* = (S(H + h)/x^*) / (T/x)$, it is almost routine to show that

$$\frac{M}{x} = g \left(\frac{H}{x^*}, \frac{T}{x} \right) \quad (9)$$

as a scaling law for magnetization. To proceed, we first illustrate (9) based on our magnetization data and determine x^* (and S^*) simultaneously. The present scheme of determining S^* should be distinguished from the more phenomenological approach of fitting the magnetization data to a modified Brillouin function employed in Ref. 6. It is also interesting to determine the critical concentration at which (9) no longer holds. However, it should be noted that the present scaling law relies on the validity

of (8). The latter should be checked by an independent method. This is achieved by determining V_O^* using the Larking-Smith method^{30,31} modified for clusters. The value is then compared to that predicted by (8) knowing S^* .

(ii) Magnetization

1.- Scaling behavior for clusters and determination of the cluster size.

In the presence of irreversible effects in spin glasses, the measured magnetization is composed of both reversible and irreversible contributions. Care must be taken when one desires to extract from the measured values the component which obeys the scaling relation in (9). For our alloys, we demonstrated previously that the remanence is dipolar in origin.⁷ However we have to modify the picture of individual spins interacting with each other to clusters interacting via the dipolar force. Since the dipolar interaction also has a $1/r^3$ dependence as the RKKY interaction, we can carry on the same argument as beforehand and obtain a scaling law similar to (9) for the total magnetization. In addition, it should be mentioned that the saturated remanent magnetization is about five percent of the saturation magnetization for $x < 32$ at.% in our alloys.⁷ Thus taking M as the measured magnetization is rather unlikely to introduce any serious uncertainty. In figure 5 are shown normalized isothermal magnetization curves versus applied field for different concentrations. At first sight, one can hardly observe any correspondence between these curves. The simple case of the scaling law is in the dilute alloys where $x^* = x$. This is shown in figure 6. In this case, the scaling law is expected to hold even for $T \gg T_M$ since

Fig. 5. Reduced magnetization M/x versus applied field in some amorphous $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$ alloys at different temperatures.

Fig. 6. Reduced magnetization M/x as a function of reduced magnetic field H/x at three different temperatures T/x (from ref. 5).

the interactions are among individual spins. As one progresses to higher concentration, the main task is to determine x^* so that the scaling relation can be observed. Of course a limitation is that the temperature must be low enough so that no significant dissociation of clusters occurs. For instance when one tries to correspond the isotherms described by ($x = 1.0$, $T = 1.8^\circ\text{K}$, $T/x = 1.8$) and ($x = 6.4$, $T = 12^\circ\text{K}$, $T/x = 1.87$) to each other, one selects a given value of M/x and determines the ratio of the fields H such that the data points fall on each other. For a given H/x^* , such a procedure yields the ratio of the effective concentrations x^* . Using this ratio of x^* to correspond other points on the two curves, it is observed that a nice scaling relation is obtained. This is shown in Fig. 7. For two given concentrations, one can use the ratio of x^* obtained by scaling two isotherms to generate more scaling relations at other values of T/x from other isotherms. One can also determine the values of x^* at progressing concentration. By performing this procedure, nice scaling relations are obtained in figures 7, 8 and 9. The values of x^* and S^* so determined are listed in Table I. Before discussing the physical significance of S^* , we should mention two points we noticed through the above curve fitting process. First, for $x > 4$ at.% dissociation of clusters is noted for temperatures higher than $\theta_s/3$. This is manifested through the observed deviation from the universal curves. Second, for $x > 32$ no correspondence of isotherms to the T/x curves at lower concentrations can be made. This indicates the failure to describe the magnetization in terms of our simple model as the clusters start to overlap. Such failures for $x = 40$ and 56 are shown in figure 9.

We plotted the values of S^*/S as a function of concentration in figure 10. In the same figure, we included a schematic variation of the

Fig. 7. Reduced magnetization M/x as a function of reduced magnetic field for clusters H/x^* at three different reduced temperatures T/x for $0.5 < x < 6.4$ alloys.

Fig. 8. Modified scaling laws for magnetization in $6.4 < x < 24$ alloys.

Fig. 9. Modified scaling laws for magnetization in $6.4 < x < 32$ alloys. Are shown also departures from scaling behavior for alloys containing 40 and 56 at.% Gd.

Fig. 10. Number of spins per cluster S^*/S and effective coupling between clusters V_O^*/V versus Gd content. The variation of S^*/S (crosses) is compared with that of Z (see text) (straight line). The experimental values for V_O^*/V (crosses) are compared with experimental variation for S/S^* (dotted line).

first nearest neighbors coordination shell (defined by a Gd reference atom and its Gd-Gd nearest neighbors) obtained by the linear extrapolation of RDF (radial distribution function) results³² on amorphous $\text{La}_{80}\text{Au}_{20}$. By comparing the value of S^*/S and Z , one can say that for $x < 24$ at.%, the size of Gd coordination shell determines the size of magnetic clusters. For higher Gd content $S^*/S > Z$ indicating that the clusters involve spins located beyond the first coordination shell. That is, there is a tendency for the Gd atoms to percolate. Different model calculations³⁶ predict percolation limits between 20 and 24 at.% Gd for $Z = 8$. However, the presence of antiferromagnetic interaction delays the onset of ferromagnetism to concentrations far above the percolation limits in our alloys. The small positive deviation of S^*/S from Z at low concentration might be caused by the polarization of conduction electrons. It should be noted that S^*/S is not necessarily an integer as a result of using an 'average' cluster mean-field approximation. This certainly would have an effect on the theoretical freezing temperature to be discussed later. It can be seen that the numbers of spins within a cluster are comparable to those obtained from magnetoresistivity results.⁶ The latter approach is more phenomenological in nature. The maximum number of spins within a cluster for which our simple model is valid equals ~ 6 . The confinement of spins within a cluster constituted by only nearest neighboring atoms for $x < 24$ is consistent with the assumptions of our model.

2.- Determination of the strength of effective interactions between clusters.

As mentioned beforehand, the consistency of this model can be further checked by investigating equation (8). What one has to do is to

determine the strength of the effective interaction V_O^* by an independent method. We adopt the Larkin-Smith^{30,31} approach modified for clusters. By taking the transformation $x \rightarrow x^*$, $S \rightarrow S^*$, $V_O \rightarrow V_O^*$ and $A \rightarrow A^*$ their equation reads

$$M(H,T) = x^* g\mu_B S^* \left[1 - \frac{2(2S^* + 1) x^* V_O^*}{3 g\mu_B H} - \frac{A^* k_B T}{H} \right] \quad (10)$$

for

$$g\mu_B H > k_B T$$

Using similar plots as in references 6 and 23, one obtains the value $(2 S^* + 1)x^* V_O^* \approx 2 S^* x^* V_O^* = 2 S x V_O^*$ from which one determines V_O^* . A typical plot of M versus $1/H$ is shown in figure 11 for the $x = 4$ alloys. The values of V_O^* so determined are normalized by V_O determined for the dilute alloys ($x < 1$). They are shown in figure 10. It should be mentioned that the physical meaning of the A^* term is not known up to this point. The values of V_O^* obtained here are comparable to those obtained previously in magnetoresistivity studies.⁶ To test the validity of (8) we plotted (fig. 10) the curve S/S^* versus x determined earlier. It can be seen that relation (8) is reasonably respected. We did not try to determine an effective V_O^* for $x > 32$ alloys in which a spontaneous magnetization can be defined from Arrott plots. The $1/H$ term in approach to saturation is then more complex in nature. In particular, important contributions may arise from magnetoelastic coupling between internal stresses and the spontaneous magnetization as studied by Kronmüller in some amorphous ferromagnets.³⁷

The effective intercluster interaction, the strength of which we

Fig. 11. Magnetization M as a function of reciprocal magnetic field $1/H$ for amorphous $\text{La}_{76}\text{Gd}_4\text{Au}_{20}$ at different temperatures.

determined from approach to saturation, has a $1/r^3$ dependence, since the modified scaling laws of equation (9) are obeyed. Both dipolar and RKKY interactions have such a $1/r^3$ behavior. This would lead to an overestimation of J_{sf} if one tries to calculate J_{sf} from V_o^* . Nevertheless, we think that V_o^* is mainly RKKY in nature, since the ratio of the remanent and saturation magnetization at 0°K :

$(M_{rs}(0)/M_\infty(0) = 5.10^{-2}$ for $x < 32$) is a measure of the ratio of the amplitude of the RKKY and dipolar couplings.⁸

Our analysis of magnetization data at intermediate and high fields (at low field, the remanent magnetization cannot be neglected) can be summarized for the $x < 32$ alloys in the following way. A given Gd atom forms a ferromagnetic cluster with its Gd first neighbors. From linear extrapolation of structural studies³², the Gd-Gd coordination number CN is $x/10$, so that CN is 8 in $\text{Gd}_{80}\text{Au}_{20}$. Thus the number of Gd atoms in a cluster is defined by $Z = 1 + \frac{x}{10}$, to give individual Gd atoms in the dilute limit. These ferromagnetic clusters behave at intermediate and high fields (H larger than a few kG) like single spin-entities of concentration $x^* = x/Z$, having a spin $S^* = S Z$; they are coupled by a $1/r^3$ interaction mainly of RKKY type with an effective amplitude $V_o^* = V_o/Z$.

(iii) Freezing temperatures

One can determine the freezing temperature T_M of concentrated spin-glasses in the cluster mean-field formalism by taking the transformation $S \rightarrow S^*$ and $J_{ij} \rightarrow J_{ij}^*$. Then T_M will have the same form as expression (1). To go one step further if the RKKY interaction among the clusters is responsible for the freezing-in phenomena, one can proceed to expression (5) by substituting V_o^* for V_o . Then it is not difficult to see that

$T_M \propto x$ under the condition $x^* S^* = x S$ and $V_O^* S^* = V_O S$. In fact such linear dependence of T_M on x is observed for $x < 12$ at.% in our alloys. For higher concentrations, our simple model cannot yield accurate values of T_M . Probably one then has to take into account the distribution in cluster size. Figure 1 indicates that $T_M \propto x^p$ with $p = 1.3$ for $16 < x < 40$. What we have shown is that in the case of ferromagnetic clusters, the variation of T_M with x is approximately linear. However if some of the strongly correlated spins inside a cluster are misaligned, T_M would vary slower than x according to the transformed version of (5). That is $V_O^* S^* = V_O S$ and T_M determined by V_O^* within the RKKY picture. The misalignment can be caused, for example, by antiferromagnetic couplings of the spins or by crystal fields. In canonical spin-glasses such as AgMn, CuMn, AuCr, AuFe,... etc., it was found² that $T_M \propto x^p$ ($0.55 < p < 0.75$) for $1 < x < 10$. Our discussion so far might offer an alternative explanation for the $T_M(x)$ variation other than the RKKY damping approach.^{26,28} Within our model T_M is given by the general form

$$T_M = 2.28 Q x k_B^{-1} \frac{q(Z)}{Z} S^2 V_O d^{-3} \quad (11)$$

With the effective concentration of clusters $x^* = x/Z$ where Z is now the number of spins inside a cluster, and the effective size of the clusters $S^* = q(Z) < Z$ due to partial misalignment of spins. Of course Z is also a function of x . It can be seen from (11) that T_M varies more slowly than x once the clusters are formed. However, one should be cautious to interpret the results in canonical spin-glasses in terms of our simple model. It is not clear that the interaction between the clusters in all these alloys are of the RKKY type. However, if one adopts the

picture of clusters frozen in a dilute spin-glass matrix, then the interaction between the clusters mediated by the matrix presumably has the RKKY form. It would be informative to study the variation of x^* and S^* as a function of x in concentrated canonical spin-glasses. Unfortunately, to our knowledge, such analysis has not been made yet.

Finally, it should be mentioned that our result for T_M is valid as long as relation (8) holds. This requires that the distribution of local field created by clusters be random or equivalently the size of the clusters be small. The latter condition is satisfied in not too concentrated alloys where the remanent magnetization is at most a few per cent of the total magnetization. This point will be discussed in greater detail in the next section.

(iv) Remanent magnetization

Our purpose in this section is to present more complete experimental data on remanent phenomena together with an analysis coherent with that proposed for high-field magnetization results. In a preliminary report⁷, we have shown that the phenomenological model of Tholence and Tournier⁸ forged for dilute spin-glasses can account for the main features of the saturated remanent magnetization in our concentrated spin-glasses, namely, its order of magnitude at 0°K and its temperature and concentration dependence. In particular, the scaling behavior observed for the remanence $M_{rs}(T) = M_{rs}(0) \exp(-\alpha T/x)$ seemed to be contradictory to the fact that the scaling laws for the magnetization $M/x = f(H/x, T/x)$ do not hold anymore for $x > 1$ alloys. This apparent discrepancy can be explained within our CMFA model.

The Tholence-Tournier picture for spin-glasses is that of an assembly of independent clouds containing n_o individual spins on average. This number may be determined consistently from $M_{rs}(o)$ and calculated from the ratio of the amplitude of RKKY and dipolar couplings. We show that the phenomenology of this model remains basically unchanged under the transformation of individual spins into ferromagnetic rigid clusters in the sense of the previous section.

Our concentrated spin-glasses are an assembly of independent clouds containing n_o ferro-clusters, each cluster being made of about Z spins. Similar to reference 8

$$n_o = \frac{1}{2\pi} (M_{\infty}(o))^2 \quad (12)$$

The half-width of the RKKY field distribution Δ^* is given by expression (4) modified for clusters. For the dipolar field-width Δ_d^* , one can carry out the same derivation as for V_o^* in (8) to obtain a similar expression for the effective strength of the inter-clusters dipolar interaction. Hence it is easy to see that n_o remains constant and proportional to V_o :

$$n_o = \Delta^* / \Delta_d^* \propto V_o \quad (13)$$

even for concentrated alloys. At lower concentrations, $\Delta \propto x$. For higher concentrations, Δ^* increases linearly with x^* . Finally, by following the Tholence-Tournier model modified for clusters, one obtains:

$$M_{rs}(T)/x = \frac{M_{rs}(o)}{x} \exp(-\alpha^* T/x^p) \quad (14)$$

where α^* is independent of x and p is the exponent defined by the concentration dependence of T_M ($T_M \propto x^p$). Thus the scaling law for the remanence relies on the fact that $M_{rs}(0) \propto x$ and $T_M \propto x$. The former condition is valid through equations (12) and (13) when n_0 is reduced to depend on V_0 only. This again depends on the constraint of non overlapping small clusters. Thus the first condition automatically limits x to less than 32 at.%. Over the region $16 < x < 32$, $T_M \propto x^{1.3}$. This affects weakly the scaling behavior. This accounts for the apparent universality of the remanence for the alloys up to 32 at.% Gd as observed in reference 7. Additional data on the $x = 24$ and 40 at.% alloys are shown in figure 12. Strong deviations from universality are observed for $x = 40$ at.%. This is consistent with the magnetization measurements, since for this concentration the magnetic isotherms fail to scale with each other.

From intercept at $T = 0$ in figure 12, we obtain $N_{rs}(0)$. From equation (12), n_0 is equal to 50 by calculating Δ^* and Δ_d^* for different x^* values, one obtains from equation (13) $n_0 \approx 40$. This consistency together with the scaling behavior eliminates the assumption that the remanence would arise from structural inhomogeneities. Instead, the remanence phenomena are an intrinsic property relying on a complex balance between dipolar and RKKY forces. By contrast, the freezing-in phenomena are governed mainly by the RKKY coupling in metallic systems without crystal field effects. Thus, there is no a priori reason that the remanent magnetization would vanish at $T = T_M$. Actually, in our system $M_{rs}(T_M)/M_{rs}(0) = 0.5$. Similar thermal dependence of the remanence has been observed in crystalline V-Fe alloys.³⁸

For a $x = 32$ alloy, a cloud of 40 to 50 clusters each of them containing about 4 Gd spins corresponds roughly to 20 shells of atoms

Fig. 12. Reduced saturated remanent magnetization (log scale) versus reduced temperature for the $x = 16, 24, 32$ alloys. Departures from the scaling behavior are observed for the $x = 40$ alloys.

surrounding a given Gd atom. This is comparable in size with the regions of 35 to 50 Å over which ferromagnetic spin-waves would be excited according to specific heat measurements on amorphous GdAl_2 .³⁹ Similarly, ESR experiments⁴⁰ on the same amorphous GdAl_2 exhibited a value of 86 Å for $2\pi \Lambda_0$, this parameter being a measure of the range over which exchange holds the spins strongly coupled against demagnetizing forces. Phenomenologically, these ferromagnetic single domains seem to correspond rather well to the magnetic clouds within the Tholence-Tournier model for remanence in spin-glasses. For a dilute system the spins in a given cloud are assumed to be perfectly aligned (ferromagnetically for Gd) along an uniaxial anisotropy field dipolar in origin. A potential barrier is created which defines the size of the cloud when the dipolar field on individual spins inside the cloud is equal to or larger than the RKKY field arising from spins outside the cloud. Our magnetization measurements lead to a more complex picture for concentrated spin-glasses. According to our definition for cluster and cloud, only a few spins (1 to 4, depending on Z) are strongly coupled to constitute a ferromagnetic cluster. However, the clusters inside a cloud are not totally misaligned; thus a resulting moment exists which experiences the anisotropy field created mainly by the anisotropic part of the dipolar interaction. The size of the clouds would be determined by a balance between the intracloud interactions (mainly dipolar) and the interactions (mainly RKKY) arising from all the clusters outside the cloud. Such an oversimplified picture qualitatively accounts for the remanence phenomena and their concentration - temperature variation. It does not pretend to describe in detail the intracloud and intercloud interactions.

V. SUMMARY AND CONCLUSION

Magnetization and initial susceptibility for amorphous La-Gd-Au alloys in the dilute range ($x < 1$ at.% Gd) were shown to obey the classical scaling laws characterizing long range interactions in $1/r^3$ for dilute crystalline spin-glasses. On the other hand, the strength of RKKY coupling was determined from approach to saturation. This parameter was found to be basically the same in magnitude as in equivalent crystalline systems. Thus two distinct problems were raised. One problem concerns the amplitude itself of the RKKY interaction within the limits of extreme dilution and infinite mean-free-path. Another difficulty is that of the spatial extent of the RKKY oscillatory term in an amorphous medium. We were led to question the model prediction of an exponential damping of RKKY coupling due to finite mean-free-path. Our results suggest that the spin-correlation length of the eigenstates in a disordered system may be larger than the mean-free-path calculated from residual resistivity. Both these problems await further theoretical investigations.

Proportionality between the freezing temperatures T_M/x of dilute metallic spin-glasses and the amplitude V_0 of RKKY coupling was established and an explicit equation was derived which accounts for T_M/x to better than an order of magnitude when V_0 is determined from approach to saturation. This relation holds for dilute crystalline and amorphous systems provided that dipolar interaction and crystal field are negligible as compared with V_0 .

The classical scaling laws for magnetization and remanence in dilute spin-glasses are shown to be valid for our concentrated spin-glasses containing up to about 32 at.% Gd within our cluster-and-cloud

description. A cluster is built up by first Gd neighbors around a given Gd atom. These Z Gd atoms are strongly coupled by short-range interaction. They behave like individual spin-entities having a spin $S^* = SZ$. Their concentration is $x^* = x/Z$. They are coupled by $1/r^3$ long-range interactions mainly RKKY in nature, the amplitude of which is related to the RKKY parameter for dilute alloys by $V_o^* = V_o/Z$. The invariance $V_o S = V_o^* S^*$ accounts for the fact that the freezing temperature is roughly proportional to x. The conduction electrons are scattered off these clusters. The oscillatory nature of the RKKY coupling between clusters may explain the Kondo-like behavior of zero-field resistivity and magnetoresistivity. For alloys containing up to 32 at.% Gd, the amplitude of RKKY coupling is large as compared with that of dipolar interaction. The ratio of these amplitudes is correlated to the square of the ratio of saturated magnetization over remanence at 0°K. This proportionality, together with the fact that the thermal variation of the reduced remanence scales with T/x, is interpreted within the Tholence-Tournier model modified for clusters. Thus the remanence phenomena imply the existence of magnetic clouds made of 40 - 50 clusters. The size of these clouds is governed by an interplay of RKKY and dipolar interactions between clusters. The exact nature and balance of intra-cloud and intercloud interactions is not fully understood.

For alloys containing $32 < x < 56$ at.% Gd, spin-glass phenomena are still observed, but the concentration and temperature dependence of magnetization and remanence do not exhibit the aforementioned regularities. According to magnetization measurements, the cluster size becomes much larger than the number of Gd first neighbors. The condition of non-overlapping clusters is not satisfied any more, indicating a trend for

clusters to percolate. Finally, the $56 \leq x \leq 80$ alloys are good ferromagnets in the conventional sense. Curie temperature and saturation magnetization at 0°K depend linearly on the content of non-magnetic elements (Au + La), according to a dilution law giving for pure amorphous Gd a Curie temperature of $250 (\pm 15)^\circ\text{K}$ and a moment of $7.15 (\pm 0.15) \mu_B$ per Gd atom.

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