

## Magnetic-cluster description of spin glasses in amorphous La-Gd-Au alloys

S. J. Poon

*Edward L. Ginzton Laboratory, W. W. Hansen Laboratories of Physics, Stanford University, Stanford, California 94305*

J. Durand

*Laboratoire de Structure Electronique des Solides (ERA 100), Université Louis Pasteur, 67000 Strasbourg, France*

(Received 3 April 1978)

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 from 1.8 to 290°K. Detailed analysis of the data based on a magnetic-cluster description of the spin glass and mictomagnetic alloys ( $x \leq 56$ ) is presented. 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, and Tournier for the magnetization are obtained and presented graphically for the  $x \leq 32$  alloys in which  $M/x = g(H/x^*, T/x)$ , where  $x^*$  is the concentration of clusters. Saturation remanent magnetization is interpreted in terms of the dipolar anisotropy model of Tholence and Tournier. The strength of the Ruderman-Kittel-Kasuya-Yosida interaction  $V_0$  between clusters (or single spins in the dilute alloys) is determined from high-field magnetization data using the Larkin-Smith approach. The freezing temperatures  $T_M$  (defined by susceptibility maxima) of dilute spin glasses in which  $T_m \propto x$  are accounted for rather well, using the experimentally determined values of  $V_0$ . An attempt is made to explain the freezing temperatures of more-concentrated spin glasses in which  $T_M \propto x^{1.3}$  ( $12 \leq x \leq 40$ ). It is also shown that for the  $x \leq 24$  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.<sup>1</sup> 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 experimental<sup>2</sup> and theoretical<sup>3</sup> 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) can be described in a rather quantitative fashion. It is believed that the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction<sup>4</sup> 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.<sup>5</sup> 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 magneto-resistivity results can be attributed to conduction electrons scattered off magnetic clusters coupled by the RKKY interaction.<sup>6</sup> In addition, preliminary data<sup>7</sup> on saturated remanent magnetization on the

same alloys were interpreted in a phenomenological model of uncompensated magnetic clouds.<sup>8</sup> 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 approximation 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 solute. 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. 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.<sup>9</sup> The magnetic-phase diagram was schematically divided into four regions. The "dilute" alloys ( $0.24 \leq x \leq 1$  at.%) were found<sup>5</sup> to be-

have like canonical *spin-glass* systems, in that the magnetization and initial susceptibility data follow the universal curves of the scaling predictions.<sup>10</sup> For the  $1 \leq x \leq 32$  alloys, the classical scaling laws for  $M(H, T)$  are no longer obeyed. However, the reduced saturated remanence  $M_{rx}(T)/x$  scales with  $T/x$ ,<sup>7</sup>  $T_M$  is roughly proportional to  $x$ . According to Ododo and Coles,<sup>11</sup> 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*.”<sup>12</sup> 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.<sup>13</sup> The present paper will focus on the first two magnetic phases. Our magnetic data will be described within the same cluster picture, so that alloys up to  $x \leq 32$  will be called spin glasses.

In this work, concentrated alloys are represented by rigid ferroclusters (i.e., we ignore the internal dynamics of the clusters) as individual spin entities interacting via random forces. By extending the results of Blandin, Souletie, and Tournier in dilute alloys, we attempt to exploit various regularities in concentrated alloys by suitable transformation of variables (concentration and size of spin clusters, interacting forces, ...). The magnitudes of the variables are determined from magnetization measurements. It should be mentioned that recent theoretical treatment of spin glasses using the cluster-interaction picture have been made. In the Monte Carlo results of Binder,<sup>14</sup> 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 dynamic clusters, Soukoulis and Levin<sup>15</sup> also produced a rounded maximum in the specific heat. Remanence in our alloys is interpreted in terms of the magnetic-cloud (containing clusters as individual entities) model.<sup>8</sup>

The role of RKKY interaction on the “freezing temperature” can be investigated by relating its strength  $V_0$  to  $T_M$ . The effect of amorphousness on the strength and range of the indirect exchange interaction are then inferred from results on dilute amorphous spin glasses. The format of this paper is as follows. In Sec. II, experimental procedures are presented. Sec. 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 presents a phenomenological description of magnetization in the concentrated alloys. In Sec. IV B, the role played by the RKKY interaction on the “freezing temperature” in dilute alloys is discussed. Section IV C discusses remanence in a magnetic-cluster-cloud picture. 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. 16. 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  $\sim 4$  cm<sup>2</sup> and thickness of about 40  $\mu\text{m}$ . 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 pattern ( $\text{CuK}\alpha$ ) of the samples are characterized by a broad maximum, the center of which ranges from  $30.7^\circ$  in  $\text{La}_{80}\text{Au}_{20}$  to  $32.8^\circ$  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  $\sim 17$  Å, which is typical of a glassy metal. 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 150 to 200 °C.

Magnetization measurements as functions of magnetic field (up to 75 kOe) and temperature (1.8 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 (3-mm diam) punched from foils. The thermal output controls have an accuracy of  $\sim 0.05$  °K. Magnetic ordering temperatures were observed using a standard ac inductance bridge technique driven at a frequency of 1 kHz.

## 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.

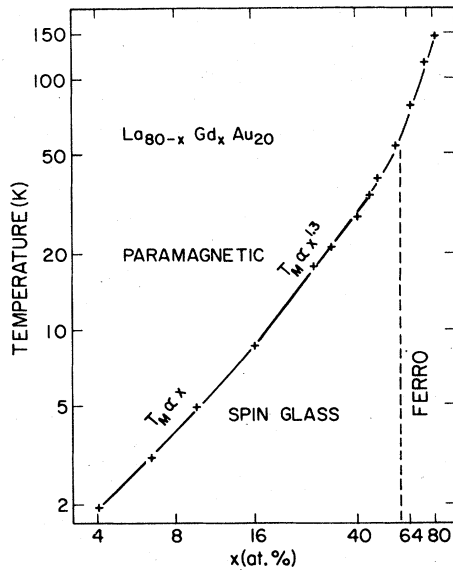


FIG. 1. "Ordering" temperature vs Gd concentration (log-log scale) in amorphous  $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$  alloys. For  $x \leq 56$ -at. %  $T_M$  is the temperature of the cusp in zero-field susceptibility. For  $x > 56$  at. %, the Curie temperature is defined consistently from Arrott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

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  $\chi_0^{-1}(T)$

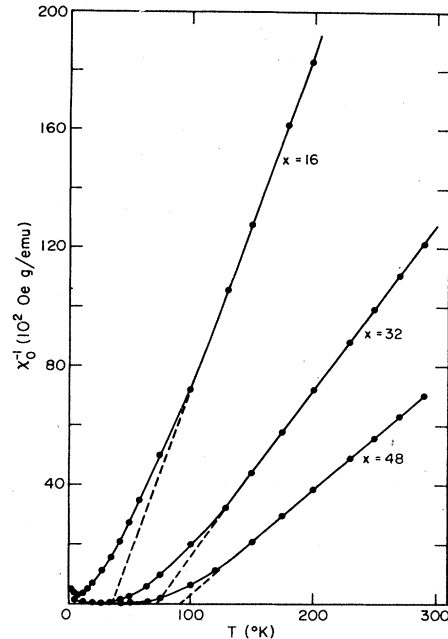


FIG. 2. Inverse susceptibility  $\chi_0^{-1}$  vs temperature for amorphous  $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$  alloys containing 16-, 32-, and 48-at. % Gd.

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  $\Theta_p$ .  $\Theta_p$  [ $\approx (3-5)T_M$ ] 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

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

$x$ (at. %)	$T_M$ (°K)	$\Theta_p^a$ (°K)	$\Theta_s$ (°K)	$M_\infty(0)$ ( $\mu_B/\text{Gd atom}$ )	$\mu_{\text{eff}}^a$ ( $\mu_B/\text{Gd atom}$ )	$x^*$ (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 <sup>a</sup>	155	170	7.28	8.20		
80	150 <sup>a</sup>	165	175	7.00	8.90		

<sup>a</sup> Refer to Curie temperature as determined consistently from Arrott plots and ac measurements.

<sup>b</sup>  $\Theta$  and  $\mu_{\text{eff}}$  are obtained from Curie-Weiss law at high temperature (up to 290 °K).  $M_\infty(0)$  is determined by a  $1/H$  extrapolation.

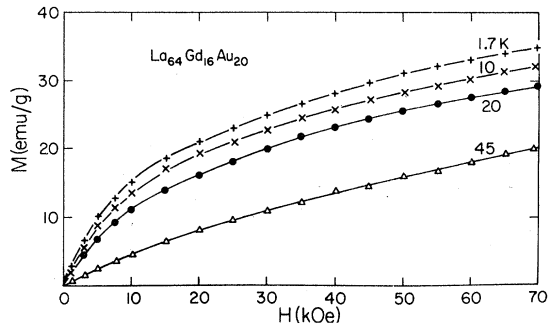


FIG. 3. Magnetization vs applied field for amorphous  $\text{La}_{64}\text{Gd}_{16}\text{Au}_{20}$  at different temperatures.

in their local field below  $T_M$  dissociate 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 in paramagnetic spins. The values of  $T_M$ ,  $\Theta_p$ ,  $\Theta_s$ , and  $\mu_{\text{eff}}$  as a function of  $x$  are listed in Table I.

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 Arrot plots ( $M^2$  vs  $H/M$ ) are obtained. Such plots for two samples ( $x=32, 48$ ) are shown in Fig. 4. Strong departures from linearity at small and high fields at temperatures below  $\Theta_p$  are seen, so that any

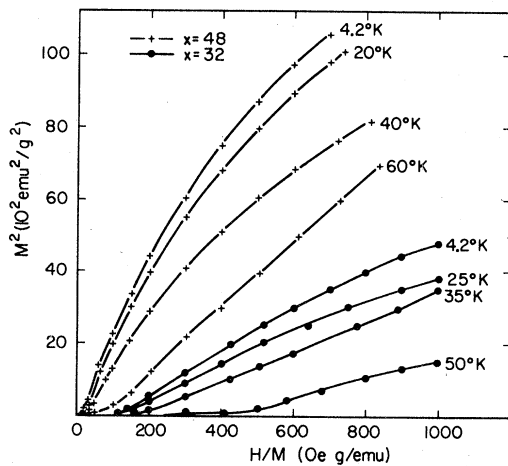


FIG. 4.  $M^2$  vs  $H/M$  at different temperatures for  $x=32$  and 48 alloys.

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 Arrot plots for all  $T < \Theta_p$  also points toward inhomogeneous magnetic interactions.

#### IV. ANALYSIS AND DISCUSSION

##### A. Phenomenological description of concentrated alloys

###### 1. Extended scaling laws of Blandin, Souletie, and Tournier

In this section we attempt to describe the magnetization of our concentrated alloys based on the interaction between clusters. This approach to study the magnetic properties is motivated by the following observations. (a) The magnetoresistivity results can be analyzed in terms of conduction electrons scattered off magnetic clusters coupled by the RKKY interaction.<sup>6</sup> (b) In 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 via random forces. An empirical approach to extract the magnetization-field-temperature relation is by fitting the magnetization data to a modified Brillouin function from which the clusters size(s) is determined.<sup>12</sup> Such approach was exemplified in Ref. 6. Alternatively, a somewhat more quantitative description of magnetization can be obtained by extending the Blandin-Souletie-Tournier (BST)<sup>10</sup> scheme to clusters. In what follows, we will take the latter approach.

We start with the assumption that our spin glasses can be described by a picture of nonoverlapping ferromagnetic clusters (of equal and constant size around  $T_M$ ) interacting via random forces. A trival 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. The RKKY local field  $h_\nu$  acting on a cluster  $\nu$  by the other clusters  $\{\mu\}$  is then given within the Ising model by  $h_\nu = \sum_\mu h_{\nu\mu}$ , with

$$h_{\nu\mu} = \frac{V_0 S^2}{S^*} \sum_{ij} \frac{\cos(2k_F r_{ij} + \Phi)}{r_{ij}^3}, \quad (1)$$

where  $i$  and  $j$  denote spins in clusters  $\nu$  and  $\mu$ , respectively. We have left out the  $(\pm)$  signs in Eq. (1). The sum of the cosine terms determines the

sign of  $h_{\nu\mu}$  since the spins inside a ferromagnetic cluster all align along the same axis. For non-overlapping clusters, 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. To first order approximation, we can combine  $V_0$  and the "average" into a product term  $(V_0^*/R_{\nu\mu}^3)f_{\nu\mu}$ , where  $f$  varies rapidly in sign as a function of  $R_{\nu\mu}$  and  $V_0^*$  is chosen such that the mean-square value of  $f$  equals  $\frac{1}{2}$ . Then Eq. (1) can be rewritten

$$h_\nu = V_0^* S^* \sum_{\mu} \frac{f_{\nu\mu}}{R_{\nu\mu}^3}, \quad (2)$$

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

Equation (2) is similar to the RKKY local field under the transformation  $V_0 \rightarrow V_0^*$ ,  $S \rightarrow S^*$ ,  $r_{ij} \rightarrow R_{\nu\mu}$ , and  $\cos -f$ . It can be viewed as an effective RKKY interaction between clusters. Next, we derive an expression for the effective strength  $V_0^*$  from a statistical argument similar to Ref. 17. We deduce the second moment of the field distribution from two approaches. First, considering the effective interaction  $V_0^*$  between the clusters one obtains  $\Delta^2 \propto x^* S^* V_0^*$ , where  $\Delta$  is the half-width of the molecular-field distribution. 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 one then obtains  $\Delta^2 \propto x^* S^2 V_0^2$ . Comparing the two results one deduces that

$$S^* V_0^* = S V_0. \quad (3)$$

Substituting (3) in (2) reduces the cluster dependent variables in (2) to those contained in the sum  $\sum$ . Returning to the local-field expression in (2) and noticing that the new invariance is  $x^* R_{\nu\mu}^3$ , we can easily extend the BST argument to magnetic clusters.<sup>10,17</sup> Only the magnitude of the moment  $S^*$  depends on the Brillouin function  $B_{S^*}$ . The argument of  $B_{S^*}$  is given by  $x^* S^* (H+h)/T x^*$ , where  $H$  is the external applied field. Then, it follows rather straightforwardly from Ref. 10 that by integrating over the internal field  $h$ , one obtains

$$M/x^* S^* = f(H/x^*, T/x^* S^*), \quad (4)$$

as a scaling law for magnetization. For ferromagnetic clusters  $x^* S^* = xS$  so that (4) becomes

$$M/x = g(H/x^*, T/x). \quad (4')$$

To proceed, we first illustrate (4') based on our magnetization data and determine  $x^*$  (and  $S^*$ ) simultaneously. It is also interesting to determine

the critical concentration at which (4') no longer holds. However, it should be noted that the present scaling law relies on the validity of (3). The latter should be checked by an independent method. This is achieved by determining  $V_0^*$  using the Larkin-Smith method<sup>18,19</sup> modified for clusters. The value is then compared to that predicted by (3) knowing  $S^*$ .

## 2. Magnetization

*a. 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 (4'). For our alloys, we demonstrated previously that the remanence is dipolar in origin.<sup>7</sup> 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 (4') for the total magnetization. In addition, it should be mentioned that the saturated remanent magnetization is about 5% of the saturation magnetization for  $x < 32$  at. % in our alloys.<sup>7</sup> Thus taking  $M$  as the measured magnetization is rather unlikely to introduce any serious uncertainty. In Fig. 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 Fig. 6. In this case, the

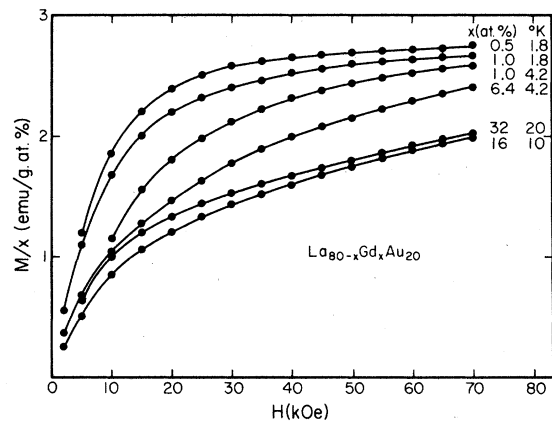


FIG. 5. Reduced magnetization  $M/x$  vs applied field in some amorphous  $\text{La}_{80-x}\text{Gd}_x\text{Au}_{20}$  alloys at different temperatures.

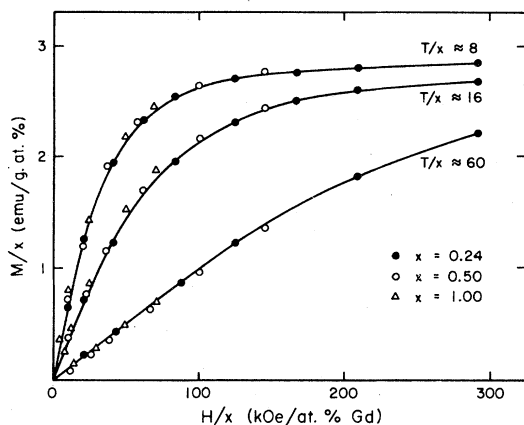


FIG. 6. Reduced magnetization  $M/x$  as a function of reduced magnetic field  $H/x$  at three different temperatures  $T/x$  (from Ref. 5).

scaling law is expected to hold even for  $T \gg T_M$  since 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 to other points on the two curves, it is observed that a 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

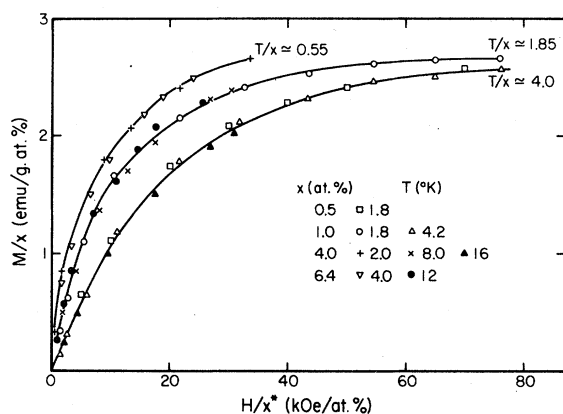


FIG. 7. Reduced magnetization  $M/x$  as a function of reduced magnetic field for clusters  $H/x^*$  at three different reduced temperatures  $T/x$  for  $0.5 \leq x \leq 6.4$  alloys.

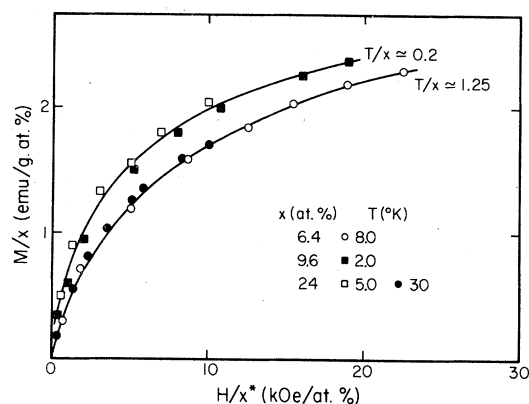


FIG. 8. Modified scaling laws for magnetization in  $6.4 \leq x \leq 24$  alloys.

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, scaling relations are obtained in Figs. 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 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 Fig. 9.

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

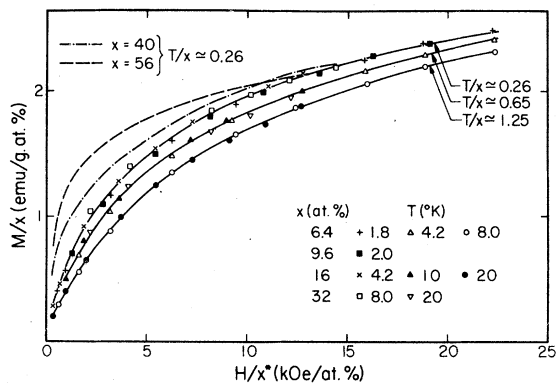


FIG. 9. Modified scaling laws for magnetization in  $6.4 \leq x \leq 32$  alloys. Departures from scaling behavior for alloys containing 40 and 56-at. % Gd are also shown.

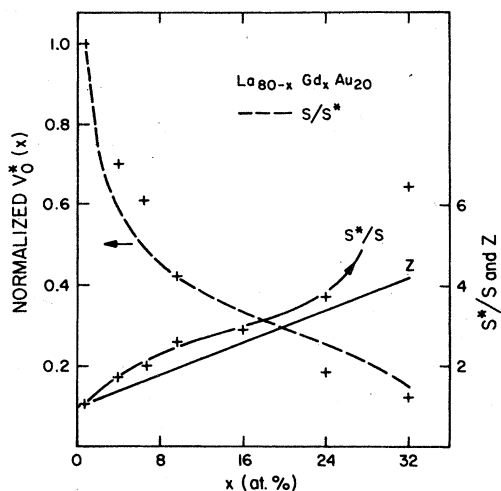


FIG. 10. Number of spins per cluster  $S^*/S$  and effective coupling between clusters  $V_0^*/V$  vs Gd content. The variation of  $S^*/S$  (crosses) is compared with that of  $Z$  (see text) (straight line). The experimental values for  $V_0^*/V$  (crosses) are compared with experimental variation for  $S/S^*$  (dashed line).

included a schematic variation of the first-nearest-neighbors coordination shell (defined by a Gd reference atom and its Gd-Gd nearest neighbors) obtained by the linear extrapolation of radial distribution function (RDF) results<sup>20</sup> on amorphous  $\text{La}_{80}\text{Au}_{20}$ . By comparing the value of  $S^*/S$  and  $Z$ , one can say that for  $x \leq 24$  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 calculations<sup>21</sup> 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 previous analysis.<sup>6</sup> The latter approach is more phenomenological in nature. The maximum number of spins within a cluster for which our simple model is valid equals  $\sim 6$ . The confinement of spins within a cluster constituted by only nearest-neighboring atoms for  $x \leq 24$  is consistent with the assumptions of our model.

b. *Determination of the strength of effective interactions between clusters.* As mentioned beforehand, the consistency of this model can be further checked by investigating Eq. (3). What one has to do is to determine the strength of the effective interaction  $V_0^*$  by an independent method. We adopt the Larkin-Smith<sup>18,19</sup> approach modified for clusters. By taking the transformation  $x \rightarrow x^*$ ,  $S \rightarrow S^*$ ,  $V_0 \rightarrow V_0^*$ , and  $A \rightarrow A^*$  their equation reads

$$M(H, T) = x^* g \mu_B S^* \left( 1 - \frac{2(2S^* + 1)x^* V_0^*}{3g \mu_B H} - \frac{A^* k_B T}{H} \right), \quad (5)$$

for  $g \mu_B H \gg k_B T$ . Using similar plots as in Refs. 5 and 19, one obtains the value  $(2S^* + 1)x^* V_0^* \approx 2S^* x^* V_0^* = 2SxV_0^*$  from which one determines  $V_0^*$ . A typical plot of  $M$  vs  $1/H$  is shown in Fig. 11 for the  $x=4$  alloy. The values of  $V_0^*$  so determined are normalized by  $V_0$  determined for the dilute alloys ( $x \leq 1$ ). They are shown in Fig. 10. It should be mentioned that the physical meaning of the  $A^*$  term is not known up to this point. The values of  $V_0^*$  obtained here are comparable to those obtained previously in magnetoresistivity studies.<sup>6</sup> To test the validity of (3) we plotted (Fig. 10) the curve  $S/S^*$  vs  $x$  determined earlier. It can be seen that relation (3) is reasonably respected. We did not try to determine an effective  $V_0^*$  for  $x > 32$  alloys in which the  $1/H$  term in approach to saturation is more complex in nature. In particular, important contributions may arise from magnetoelastic coupling between internal stresses and spontaneous magnetization as studied by Kronmüller in some amorphous ferromagnets.<sup>22</sup>

Our analysis of magnetization data at intermediate and high fields (at low field, the remanent magnetization cannot be neglected) can be summarized

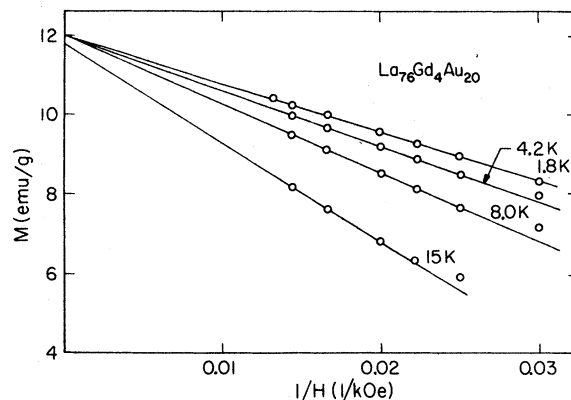


FIG. 11. Magnetization  $M$  as a function of reciprocal magnetic field  $1/H$  for amorphous  $\text{La}_{76}\text{Gd}_4\text{Au}_{20}$  at different temperatures.

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 studies,<sup>20</sup> the Gd-Gd coordination number is  $\frac{1}{10}x$ , so that the coordination number is 8 in  $\text{Gd}_{80}\text{Au}_{20}$ . Thus the number of Gd atoms in a cluster is defined by  $Z = 1 + \frac{1}{10}x$ , 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  $S^* = SZ$ ; they are coupled by a  $1/r^3$  interaction mainly of the RKKY type with an effective amplitude  $V_0^* = V_0/Z$ .

### B. Susceptibility maxima in spin glasses

Although various observed anomalies are taken as the common characteristics of spin glasses,<sup>2</sup> yet the nature and even the existence of a phase transition in this class of alloys remain an open question. The most pronounced anomaly is the low-field ac susceptibility maxima from which a "freezing temperature" is defined. It has been proposed that the latter signifies a cooperative effect of the localized spins interacting via either the RKKY interaction (or the anisotropic part of it<sup>3</sup>) or the anisotropic dipolar interaction.<sup>8</sup> No matter what the origin of the freezing phenomena is, an *a priori* correlation of the spins via the RKKY interaction is involved in the above models. To be specific, in the dipolar model,<sup>8</sup> the size of the clouds is determined from a balance of the RKKY energy and the dipolar energy. Thus, it might be relevant to point out the correlation between the "freezing temperature" and the strength of the RKKY interaction. First, we study the case of dilute spin glasses in which interaction between single spins is considered. A plausible explanation for the concentrated alloys is suggested. Finally we briefly discuss the role of amorphousness on the RKKY interaction as inferred from our results.

In canonical spin glasses, the "freezing-temperature"-concentration relation is observed to be  $T_M \propto x$  for  $x \approx 0.01$  to 0.1 at. %, <sup>2,10</sup> while  $T_M \propto x^p$  ( $0.55 < p < 0.75$ ) for higher concentrations.<sup>2</sup> Attempts to explain the deviation of  $T_M(x)$  from linearity based on the damping of the RKKY interaction<sup>23,24</sup> and the "frustration" theory<sup>24</sup> have been made. Here we will focus on the dilute regime where the scaling laws are observed. The mean-field approximation allows one to write down an empirical relation between the "freezing temperature"  $T_M$  and the strength of the magnetic interaction  $V_0$ . Thus, using dimensional analysis, one guesses that  $k_B T_M/x$  should be proportional to  $QS(S+1)V_0/d^3$ , where  $V_0$  is in  $\text{erg cm}^3$ ,  $Q = \frac{1}{4}, \frac{1}{2}$ ,

and 1 for the simple-cubic, body-centered-cubic, and face-centered-cubic structure, respectively. The above relation is obvious in the ferromagnetic case.  $(T_M^{\text{EXP}}/x)[k_B d^3/QS(S+1)]$  is plotted in Fig. 12 as a function of  $V_0$  for ten alloys.  $T_M^{\text{EXP}}/x$  is the experimentally observed freezing-temperature gradient. With the exception of  $\text{MoFe}$ ,  $V_0$  is determined from the aforementioned Larkin-Smith method.<sup>18,19</sup> The literature references of  $T_M^{\text{EXP}}/x$  and  $V_0$  are cited in the figure. In typical spin-glass theories,<sup>3</sup> one can give an estimate of the numerical constant relating the parameters mentioned above. The dashed lines in Fig. 12 represent such estimates for the Ising and Heisenberg models. It can be seen that the experimental values fall within the predictions of the mean-field values. Figure 12 also illustrates a definite relationship between the strength of the RKKY interaction and the magnitude of  $T_M$ . The  $s$ - $d$  coupling is known to be stronger than the  $s$ - $f$  coupling. The weakness of the indirect exchange interaction due to  $s$ - $f$  coupling originates from the localization of  $f$  electrons. The latter is responsible for the smaller

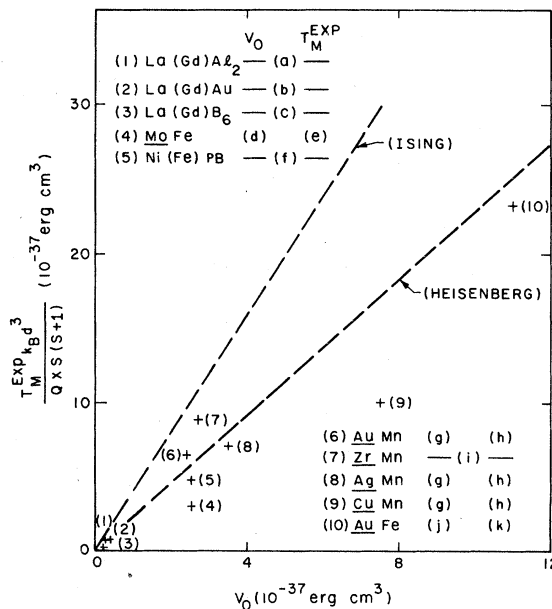


FIG. 12.  $(T_M^{\text{EXP}}/x)[k_B d^3/QS(S+1)]$  as a function of  $V_0$ . The dashed lines are the theoretical Ising and Heisenberg predictions for such plots (f). The references are taken from (a) Ref. 28; (b) Ref. 5; (c) Ref. 30; (d) F. W. Smith and M. P. Sarachik, *Phys. Rev. B* **16**, 4142 (1977); (e) R. Caudron *et al.*, *Physica* **86-88B**, 833 (1977); (f) J. Durand and S. J. Poon, *J. Phys. (Paris)* (to be published); (g) Ref. 19; (h) Ref. 2; (i) H. C. Jones *et al.*, *Phys. Rev. B* **16**, 1177 (1977); (j) F. W. Smith and J. C. Liu, *Solid State Commun.* **26**, 91 (1978); (k) J. L. Tholence and R. Tournier, *J. Phys. (Paris)* **35**, C4, 229 (1974).



$T_M^{\text{Exp}}/x$  values observed in rare-earth compounds. The present analysis indicates that the RKKY interaction is important in determining the freezing temperature in canonical spin glasses. It should be mentioned that we have neglected the influence of crystal field on  $T_M$ . In certain rare-earth alloys, crystal field anisotropy can be as large as the RKKY force and  $T_M$  is affected significantly, while single-ion anisotropy is absent in Gd alloys. Recent experiments<sup>25</sup> and calculations<sup>26</sup> clearly illustrate this point.

For the concentrated La-Gd-Au spin glasses, one can employ the cluster picture as described in Sec. IV A to study the  $T_M(x)$  behavior. The validity of expression (3) (i.e.,  $V_0^*S^* = V_0S$ ) together with the ferromagnetic cluster condition  $x^*S^* = xS$  establishes the relation  $x^*S^*(S^* + 1)V_0^* \simeq xS(S + 1)V_0$ . The latter implies that  $T_M \propto x$ . In fact such a linear dependence of  $T_M$  on  $x$  is observed for  $x \leq 12$  at. % in our alloys. For higher concentrations, our analysis 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  $16 \leq x \leq 40$ . What we have shown is that in the case of RKKY interaction,  $T_M(x)$  is approximately linear for ferromagnetic clusters. However if some of the strongly correlated spins inside a cluster are misaligned,  $T_M$  would vary slower than  $x$ . The latter is clear in the RKKY case since then  $x^*S^* < xS$ , while the relation  $V_0^*S^* = V_0S$  is still valid. The misalignment can be caused, for example, by antiferromagnetic couplings of the spins or by crystal fields. Crystalline alloys containing only S-ion Gd offer an illustration of the above discussion.  $\text{GdAl}_2$  is ferromagnetic ( $T_c \simeq 170$  K),<sup>27</sup> and  $T_M(x)$  in  $\text{La}(\text{Gd})\text{Al}_2$  shows positive deviation from linearity.<sup>28</sup> On the other hand,  $\text{GdB}_6$  is antiferromagnetic ( $T_N \simeq 13$  K),<sup>29</sup> and  $T_M(x)$  in  $\text{La}(\text{Gd})\text{B}_6$  shows negative deviation from linearity.<sup>30</sup> It could be informative to study the variation of  $x^*$  and  $S^*$  as a function of  $x$  in other concentrated spin glasses.

It should be mentioned that our results from  $T_M$  is valid as long as relation (3) 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 percent of the total magnetization. This point will be discussed in greater detail in Sec. IV C.

Finally, we consider the effect of amorphousness on the RKKY interaction. De Gennes<sup>31</sup> pointed out that in disordered alloys, the interaction in its asymptotic form is modified by an exponential factor  $\exp(-r/l)$  where  $l$  is presumably the electronic mean-free path. Both experimental<sup>17,32-34</sup> and the-

oretical considerations<sup>35</sup> are in favor of this prediction. However, present results of La-Gd-Au alloys do not seem to follow the regularities observed in disordered crystalline alloys. The values of  $T_M$  in dilute amorphous La-Gd-Au alloys are of the same order of magnitude as those in crystalline alloys containing Gd.<sup>28,30</sup> In addition, the approach to saturation magnetization follows the scaling law of canonical spin glasses.<sup>5</sup> This suggests that both the strength and range of the  $1/r^3$  interaction are hardly modified at all in our dilute amorphous alloys. De Gennes' result is probably limited to  $l > a$  ( $a$  is the interatomic distance), where the plane-wave description of the electronic states is valid. When  $l \leq a$ , as in our alloys ( $l \simeq 2-3$  Å from a resistivity of 200-300  $\mu\Omega\text{cm}$ ) one probably has to resort to the tight-binding calculation.

### C. 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 report,<sup>7</sup> we have shown that the phenomenological model of Tholence and Tournier<sup>8</sup> 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 the clusters picture discussed beforehand.

The Tholence-Tournier picture for spin glasses is that of an assembly of independent clouds containing  $n_0$  individual spins on average. This number may be determined consistently from  $M_{rs}(0)$  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 Secs. IV A and IV B.

Our concentrated spin glasses are an assembly of independent *clouds* containing  $n_0$  ferroclusters, each *cluster* being made of about  $Z$  spins. Similar to Ref. 8

$$n_0 = (1/2\pi)[M_{\infty}(0)/M_{rs}(0)]^2. \quad (6)$$

The half-width of the RKKY field distribution  $\Delta^*$  is

modified for clusters. For the dipolar field width  $\Delta_d^*$ , one can carry out the same derivation as for  $V_0^*$  in (3) to obtain a similar expression for the effective strength of the interclusters dipolar interaction. Hence it is easy to see that  $n_0$  remains constant and proportional to  $V_0$ :

$$n_0 = \Delta^* / \Delta_d^* \propto V_0, \quad (7)$$

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

$$M_{rs}(T)/x = [M_{rs}(0)/x] \exp(-\alpha^* T/x^p), \quad (8)$$

where  $\alpha^*$  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 Eqs. (6) and (7) when  $n_0$  is reduced to depend on  $V_0$  only. This again depends on the constraint of nonoverlapping small clusters. Thus the first condition automatically limits  $x$  to less than 32 at. %. Over the region  $16 \leq x \leq 32$ ,  $T_M \propto x^{1.3}$ . 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 Ref. 7. Additional data on the  $x=24$  and 40-at. % alloys are shown in Fig. 13. 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 Fig. 12, we obtain  $M_{rs}(0)$ . From Eq. (6),  $n_0$  is equal to 50. By calculating  $\Delta^*$  and  $\Delta_d^*$  for different  $x^*$  values, one obtains from Eq. (7)  $n_0 \approx 40$ . 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. 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.<sup>36</sup>

For a  $x=32$  alloy, a cloud of 40–50 clusters each of them containing about four Gd spins corresponds roughly to 20 shells of atoms surrounding a given Gd atom. This is comparable in size with the regions of 35–50 Å over which ferromagnetic spin waves would be excited according to specific-heat measurements on amorphous GdAl<sub>2</sub>.<sup>37</sup> Similarly, ESR experiments<sup>38</sup> on the same amorphous GdAl<sub>2</sub> exhibited a value of 86 Å for  $2\pi\Lambda_0$ , this pa-

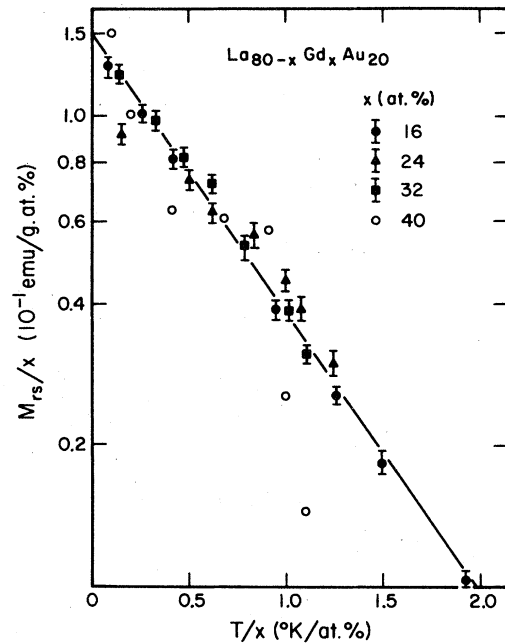


FIG. 13. Reduced saturated remanent magnetization (log scale) vs reduced temperature for the  $x=16$ , 24, 32 alloys. Departures from the scaling behavior for observed for the  $x=40$  alloy.

rameter 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 (1–4, 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 intercloud 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 CONCLUSIONS

Magnetization and initial susceptibility for amorphous La-Gd-Au alloys in the dilute range ( $x \leq 1$ -at. % Gd) were shown to obey the classical scaling laws characterizing long range interactions in  $1/r^3$  for dilute crystalline spin glasses. The strength  $V_0$  of RKKY interactions was determined from approach to saturation magnetization. Both the parameter  $V_0$  and the freezing temperature in our alloys with very short mean path, were found to have the same order of magnitude as those in equivalent crystalline systems. Thus, one is led to question the accepted result that the spatial extent of the indirect exchange interaction is limited to the electronic mean-free path. Further investigations are clearly needed in the case when the mean-free path becomes less than the interatomic spacing.

A definite correlation between the freezing temperature of dilute metallic spin glasses and the amplitude of RKKY coupling was established. This confirms that the indirect exchange interaction plays an important role in the freezing phenomena despite the type of anisotropy model used. The scaling laws for magnetization and remanence in dilute spin glasses are successfully extended to our concentrated spin glasses containing up to about 32-at. % Gd within a 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  $V_0^* = V_0/Z$ . The invariance  $V_0 S = V_0^* S^*$  accounts for the fact that the freezing temperature is roughly proportional to  $x$ . 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 proportional 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 intracloud and intercloud interactions is not fully understood.

#### ACKNOWLEDGMENTS

It is a pleasure to acknowledge the useful advice and encouragement of Professor Pol Duwez. One of us (J.D.) wishes to thank Dr. A. P. Malozemoff for a critical reading of the manuscript and for a fruitful discussion. This work was completed at the California Institute of Technology and was supported by the U. S. Department of Energy Contract No. Y-76-C-03-0822.

<sup>1</sup>See *Amorphous Magnetism II*, edited by R. A. Levy and R. Hasegawa (Plenum, New York, 1977).

<sup>2</sup>J. A. Mydosh, AIP Conf. Proc. **24**, 131 (1974); A. P. Murani, J. Magn. Magn. Mater. **5**, 95 (1977); J. Souletie, J. Phys. (Paris) **39**, C-2, 3 (1978).

<sup>3</sup>K.H. Fischer, Physica **86-88B**, 813 (1977); P. W. Anderson, J. Appl. Phys. **49**, 1599 (1978).

<sup>4</sup>For a review, see, C. Kittel, Solid State Phys. **22**, 1 (1968).

<sup>5</sup>S. J. Poon and J. Durand, Solid State Commun. **21**, 793 (1977); **21**, 999 (1977).

<sup>6</sup>S. J. Poon, J. Durand, and M. Yung, Solid State Commun. **22**, 475 (1977).

<sup>7</sup>S. J. Poon and J. Durand, Commun. Phys. **2**, 87 (1977).

<sup>8</sup>J. L. Tholence and R. Tournier, Physica **86-88B**, 873 (1977); F. Holtzberg, J. L. Tholence, and R. Tournier, in Ref. 1, p. 155; J. L. Tholence *et al.*, J. Phys. C **39**, 928 (1978); H. V. Löhneysen, J. L. Tholence, and R. Tournier, *ibid.* **39**, 922 (1978).

<sup>9</sup>S. J. Poon and J. Durand, in Ref. 1, p. 245.

<sup>10</sup>J. Souletie and R. Tournier, J. Low Temp. Phys. **1**, 95 (1969); A. Blandin, thesis (Paris, 1961) (unpublished).

<sup>11</sup>J. C. Ododo and B. R. Coles, J. Phys. F **7**, 2393

(1977).

<sup>12</sup>P. A. Beck, Trans. AIME **2**, 2015 (1971); J. S. Kouvel, J. Phys. Chem. Solids **21**, 57 (1961); **24**, 795 (1963).

<sup>13</sup>S. J. Poon and J. Durand, Phys. Rev. B **16**, 316 (1977).

<sup>14</sup>K. Binder, Z. Phys. B **26**, 339 (1977).

<sup>15</sup>C. M. Soukoulis and K. Levin, Phys. Rev. Lett. **39**, 581 (1977); and also see comment 13 in the same paper on the use of an average size cluster instead of a distribution of cluster sizes; also private communication.

<sup>16</sup>P. Duwez, in *Progress in Solid State Chemistry* (Pergamon, Oxford, 1966), Vol. 3, p. 377.

<sup>17</sup>J. Souletie, thesis (Grenoble, 1968) (unpublished); J. Souletie, J. L. Tholence, and R. Tournier, in *Proceedings of the Conference on Low Temperature Physics XI*, edited by J. F. Allen, D.M. Finlayson, and D.M. McCall (University, St. Andrews-Scotland, 1968), Vol. 2, p. 1263. The half-width of the molecular-field distribution  $\Delta$  is obtained from  $\Delta^2 = (2Qx/d^3) \int_{R_c}^{\infty} (SV_0/r^3)^2 4\pi r^2 dr$  and  $(16\pi/3)QR_c^2 = d^3$ , where  $Q$  is the structure factor discussed in the text,  $R_c$  is the radius of the sphere containing a magnetic atom, and  $d$  is the lattice parameter.

<sup>18</sup>A. I. Larkin and D. E. Khmel'nitskii, Zh. Eksp. Teor. Fiz. **58**, 1789 (1970) [Sov. Phys. JETP **31**, 958 (1970)];

- A. I. Larkin, V. I. Mel'nikov, and D. E. Khmel'nitskii, *Zh. Eksp. Teor. Fiz.* **60**, 846 (1971) [*Sov. Phys. JETP* **33**, 458 (1971)]. The expression of  $V_0$  as a function of  $\overline{M}(H, T)$  presented in the first paper has to be corrected by a factor of 2 according to the second paper [F. W. Smith (private communication)]. A more exact derivation of high-field expansion has been given recently by K. Matho, *Physica* **86-88B**, 854 (1977).
- <sup>19</sup>F. W. Smith, *Phys. Rev. B* **10**, 2980 (1974); **B 13**, 2976 (1976); **B 14**, 241 (1976).
- <sup>20</sup>J. Logan, *Scr. Metall.* **9**, 379 (1975).
- <sup>21</sup>R. J. Elliott, *J. Phys. Chem. Solids* **16**, 165 (1960); M. F. Sykes and J. W. Issam, *Phys. Rev. A* **133**, 310 (1964); H. Sata and R. Kikuchi, *AIP Conf. Proc.* **18**, 605 (1974).
- <sup>22</sup>H. Kronmüller and J. Ulner, *J. Magn. Magn. Mater.* **6**, 52 (1977).
- <sup>23</sup>U. Larsen, *Solid State Commun.* **22**, 311 (1977); Ref. 1, p. 85.
- <sup>24</sup>W. Kinzel, *Phys. Lett.* **62A**, 362 (1977); W. Kinzel and K. H. Fisher, *J. Phys. F* **7**, 2163 (1977).
- <sup>25</sup>B. V. B. Sarkissian, *J. Phys. F* **7**, L139 (1977); *Physica* **86-88B**, 865 (1977).
- <sup>26</sup>S. K. Ghatak and D. Sherrington, *J. Phys. C* **10**, 3149 (1977); J. H. Chen and T. C. Lubensky, *Phys. Rev. B* **16**, 2106 (1977); R. Harris and D. Zobin, *J. Phys. F* **7**, 337 (1977).
- <sup>27</sup>K. H. J. Buschow, J. F. Fast, A. M. van Diepen, and H. W. de Wijn, *Phys. Status Solidi* **24**, 715 (1967).
- <sup>28</sup>H. V. Löhneysen, J. L. Tholence, and F. Steglich, *Z. Phys. B* **29**, 319 (1978).
- <sup>29</sup>B. R. Coles and D. Griffiths, *Proc. Phys. Soc.* **77**, 243 (1961).
- <sup>30</sup>W. Felsch, *Z. Phys. B* **29**, 203 (1978).
- <sup>31</sup>P. G. de Gennes, *J. Phys. Radium* **23**, 630 (1962).
- <sup>32</sup>A. J. Heeger, A. P. Klein, and P. Tu, *Phys. Rev. Lett.* **17**, 803 (1966).
- <sup>33</sup>F. W. Smith, *Solid State Commun.* **25**, 341 (1978).
- <sup>34</sup>R. Buchmann, H. Falke, H. P. Jablonski, and E. F. Wassermann, *Phys. Rev. B* **17**, 4315 (1978).
- <sup>35</sup>A. Madhukar, *J. Phys. (Paris) C* **4**, 295 (1974); T. Kaneyoshi, *J. Phys. F* **5**, 1014 (1975).
- <sup>36</sup>H. Claus, *Phys. Rev. Lett.* **34**, 26 (1975).
- <sup>37</sup>J. M. D. Coey, S. von Molnar, and R. J. Gambino, *Solid State Commun.* **24**, 167 (1977).
- <sup>38</sup>A. P. Malozemoff and J. P. Jamet, *Phys. Rev. Lett.* **39**, 1293 (1977).