

Remanent magnetization of a rare-earth spin-glass: $(La, Gd)Al_2$

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(Received 19 December 1978)

The thermoremanent and isothermal remanent magnetization of $(La_{1-x}Gd_x)Al_2$, as well as its time, temperature, magnetic field, and concentration dependence have been studied. The saturated remanent magnetization σ_{rs} follows scaling laws up to $x=4$ at. % Gd. The general relation between time and temperature dependence of σ_{rs} , based on a model of spin regions, is shown to hold in this system. An explanation for the occurrence of a maximum in the field dependence of the thermoremanence is suggested.

I. INTRODUCTION

The existence of a remanent magnetization in spin-glasses is well established experimentally. Studies on remanent properties have been reported for a variety of substances: for the canonical spin-glasses $AuFe$ and $CuMn$,¹⁻⁶ for $PtMn$,⁷ amorphous $(La, Gd)_{0.8}Au_{0.2}$ ($La_{0.8}Au_{0.2}$ as host with Gd impurities replacing La),⁸ and also for the insulating spin-glass-like $(Sr, Eu)S$.^{9,10} Also the concentration regime over which remanent effects are observed is quite large, extending from moderate or high concentrations of magnetic atoms (≥ 20 at. %) to very dilute spin-glasses, e.g., 0.05 at. % in $AuFe$.⁵ The description of these experiments can be based on the application of Néel's work on rock magnetism¹¹ to spin-glasses as done by Tholence and Tournier.⁵

The main assumption of this phenomenological theory is the existence of well-defined "spin regions" or "spin clouds", which can relax between equilibrium orientations. (The term "region" will be used throughout this paper in order to distinguish these magnetic structures from chemical or magnetic clusters.) This relaxation leads to the experimentally observed time-dependent magnetic behavior below the freezing temperature, e.g., difference in long-time and short-time susceptibility,^{5,6,12} and a remanent magnetization decaying slowly with time. More specifically, the model assumes that within one region the impurity spins are coupled with each other by the RKKY (Ruderman-Kittel-Kasuya-Yosida) interaction. The energy barrier between the different orientations of one region should be due to the dipolar interaction.⁹ However, there exists a major difficulty in the application of rock magnetism to spin-glasses. In the Néel model¹¹ magnetic ordering inside a magnetic grain is attained at an ordering temperature T_N well above the blocking temperature T_B . In

the spin-glass case the distinction between the blocking temperature of a spin region and its "ordering temperature" is difficult. Although RKKY coupled pairs, triplets, etc. form already well above the freezing temperature T_f as defined by the maximum in the ac susceptibility and contribute largely to the specific heat,¹³⁻¹⁵ isolated magnetic atoms become progressively "ordered", i.e., members of regions as the temperature is lowered even to values below T_f .

As in rock magnetism the remanent magnetization depends strongly on the "history" of the sample. The isothermal remanent magnetization (IRM) σ_r^i , is obtained by cooling the sample through T_f in zero applied field H , turning H on for a fixed time and then measuring σ_r^i . For thermoremanent magnetization (TRM) σ_r^{th} , one applies H at $T > T_f$ and cools through T_f leaving the magnetic field on. Since in this case all regions could be turned in the direction of H before being blocked, σ_r^{th} is always larger than σ_r^i .

The remanent properties of spin-glasses have begun to be of interest also to theorists. In particular, Monte Carlo calculations based on the Edwards-Anderson spin-glass model have been able to reproduce most of the experimentally observed remanent effects.^{16,17}

In this article we report measurements of the thermoremanent and isothermal remanent magnetizations and their field, time, temperature, and concentration dependence in a crystalline rare-earth spin-glass. For this investigation the system $(La, Gd)Al_2$ seemed particularly well suited not only because it has been previously established as a spin-glass,^{12,18} but also because in this system, the frequency dependence of T_f , a consequence of the model of spin regions could be experimentally verified.¹⁹ It is the aim of this article to stress once more the importance of remanent effects in spin-glasses and furthermore to

classify them according to what extent they are generally obeyed or else depend on specific assumptions of the particular distribution of energy barriers.

II. RESULTS

A. Magnetic-field dependence of the remanent magnetization

Figure 1 shows the thermoremanent magnetization σ_r^{th} and the isothermal magnetization σ_r^i as a function of the previously applied magnetic field H for the 2 at. % alloy at $T \approx 0.08$ K, i.e., well below the freezing temperature of $T_f = 0.52$ K.¹² Similar curves were observed for the other samples. (The alloys were mostly the same as in Ref. 12 or similarly prepared. The measurements were carried out in a demagnetization cryostat. In order to be well defined, the remanent magnetization was always measured about 30 sec after the field H had been switched off. For taking data of σ_r^i , H was left on for a fixed time of 2 min, because σ_r^i was noted to evolve slowly towards σ_r^{th} when the field was left on for a long time.)

In small fields, the TRM σ_r^{th} is always larger than the IRM σ_r^i . Both σ_r^{th} and σ_r^i saturate to the same value σ_{rs} as observed in the canonical spin-glasses. However, the field in which saturation is reached is rather low (≈ 1 kOe) when compared to $3d$ spin-glasses.⁵ The inset of Fig. 1 shows σ_{rs} for different concentrations, always measured at temperatures between 0.06 and 0.08 K. σ_{rs} varies linearly with x up to 4 at. %, this "scaling law" being expected from the model⁵ and showing that the remanent effects are not due to chemical clustering or precipitation. Since the 2 at. % alloy falls well within the scaling regime, all further investigations were also carried out on this sample. At higher concentrations, σ_{rs} grows faster

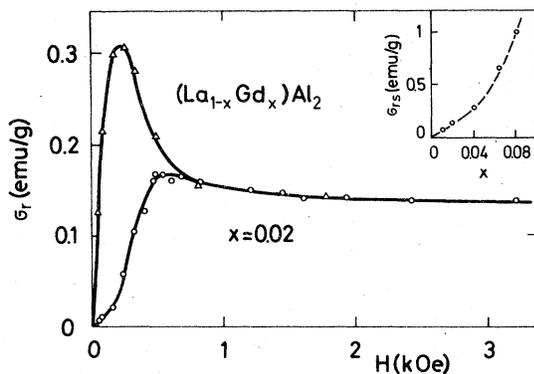


FIG. 1. Isothermal remanent magnetization (IRM), σ_r^i (circles) and thermoremanent magnetization (TRM), σ_r^{th} (triangles) as a function of the previously applied magnetic field H for $(\text{La}_{1-x}\text{Gd}_x)\text{Al}_2$, $x=0.02$, measured at $T \approx 0.08$ K. Inset shows the saturated remanent magnetization, σ_{rs} as a function of the concentration x .

hinting at ferromagnetic quasiclusters composed of nearest and next-nearest Gd neighbors.¹²

The behavior of σ_r^{th} in the magnetic fields below the saturation field is quite peculiar: it rises steeply in very low fields and has a maximum whose value is twice as large as σ_{rs} . In AuFe , the maximum of σ_r^{th} is much less pronounced, i.e., only about 10% above σ_{rs} .⁵ The only other system known to us which shows similar anomalies in σ_r^i is the insulating spin-glass-like system $(\text{Sr},\text{Eu})\text{S}$.¹⁰

B. Time dependence of the remanent magnetization

Figure 2 shows the saturated remanent magnetization σ_{rs} as function of time on a logarithmic scale for different temperatures. σ_{rs} decreases slowly with time, as known from other spin-glasses.^{3,9} However, due to the limited time span of the experiment, the exact functional variation of σ_{rs} with time is not unambiguous. Whereas Fig. 2 suggests $\sigma_{rs} \sim 1 - \log t$, any slowly varying function $\sigma_{rs} \sim t^{-\alpha}$ with $\alpha \ll 1$ can be fit to the data. This is clearly seen from Fig. 3, where the data of Fig. 2 are shown as $\log \sigma_{rs}$ vs $\log t$. This plot too yields approximately straight lines. This ambiguity has been noted before by Binder¹⁶ and Prejean.²⁰

The slope S of the σ_{rs} vs $\log t$ curves depends on temperature. A maximum of S around $T \approx 0.25$ K is inferred from Fig. 2. Below 0.2 K, the time dependence of σ_{rs} is rather small and could not be resolved accurately in the demagnetization cryostat. It should be noted that above that temperature the time decay is quite fast when compared to, e.g., AuFe .⁹

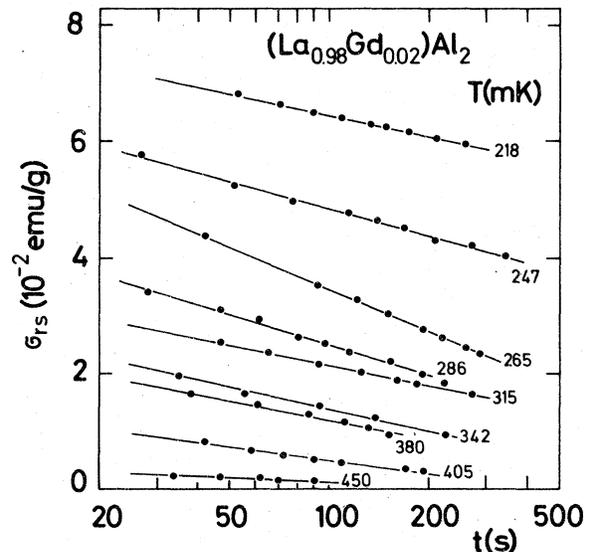


FIG. 2. Saturated remanent magnetization σ_{rs} as a function of time (logarithmic) for $(\text{La}_{0.98}\text{Gd}_{0.02})\text{Al}_2$ at various temperatures.

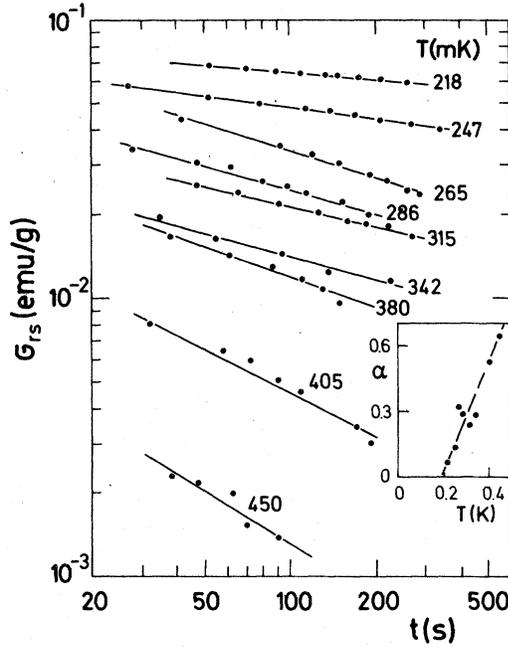


FIG. 3. Same data as Fig. 2 plotted as $\log \sigma_{rs}$ vs $\log t$. The inset shows the slope α of these curves.

C. Temperature dependence of the remanent magnetization

Figure 4 shows the thermal variation of σ_{rs} , each point being taken after the magnetic field has been switched off for about 30 sec (values partly extrapolated from Fig. 2). Whereas $\sigma_{rs}(T)$ decays approximately exponentially for $T \leq T_f \approx 0.5$ K, it is clearly nonexponential below 0.2 K. At lower temperatures it levels off as can be seen from the inset of Fig. 4 where the values $\sigma_{rs}(T)$ are plotted on a logarithmic scale. The temperature where the leveling-off of $\sigma_{rs}(T)$ occurs is about the same at which the time dependence of σ_{rs} becomes very small as mentioned in Sec. II B. We note that the thermal variation of the maximum of the TRM, $\sigma_{r,max}^{th}$ more closely resembles an exponential, as can be seen from the inset of Fig. 4.

III. DISCUSSION

In the model of Tholence and Tournier⁵ the relaxation of a spin region between two equilibrium orientations separated by an energy barrier E_a is assumed to be described by an Arrhenius law

$$t = \tau_0 e^{E_a/k_B T}, \quad E_a = \ln \left(\frac{t}{\tau_0} \right) k_B T, \quad (1)$$

where τ_0 is some intrinsic time constant. The energy barrier possibly has its origin in the anisotropic mag-

netic dipolar interaction.⁹ This implies that a particular region is blocked at a temperature T_B for which the relaxation time t becomes equal to the measuring time τ_m

$$\tau_m = \tau_0 e^{E_a/k_B T_B} \quad (2)$$

In the case of a distribution of E_a the regions with the largest E_a are blocked at the highest temperature. This yields a maximum in the ac susceptibility χ at the "freezing temperature" T_f . From Eq. (2) it is apparent that even in the case of a distribution $p(E_a)$ of E_a χ and T_f should depend on frequency. Whereas the depression of χ with increasing frequency has been known for some time in micromagnetic CuMn,²¹ the shift of T_f has been observed recently in a number of spin-glasses: in dilute $(La,Gd)Al_2$,¹⁹ in insulating $(Sr,Eu)S$,^{10,22,23} and in rather concentrated alloys of $AuFe$,^{24,25} as well as in thin films of $AuCo$.²⁶ Although measurements of the susceptibility of dilute $AgMn$ over a considerable frequency range failed to show a frequency dependence of T_f ,²⁷ its existence in a variety of spin-glasses supports the basic assumption of the model of spin regions with an Arrhenius-type relaxation which governs the magnetic behavior of spin-glasses.

Turning to the remanent magnetization σ_r , we first note that the model provides a quite general relationship between the time and temperature dependence of σ_r .⁵ Let $p(E_a)dE_a$ be the number of regions with an anisotropy energy between E_a and $E_a + dE_a$. Then the corresponding change of the remanent magnetization is

$$d\sigma_r = -\frac{1}{2} M_g(E_a) p(E_a) dE_a,$$

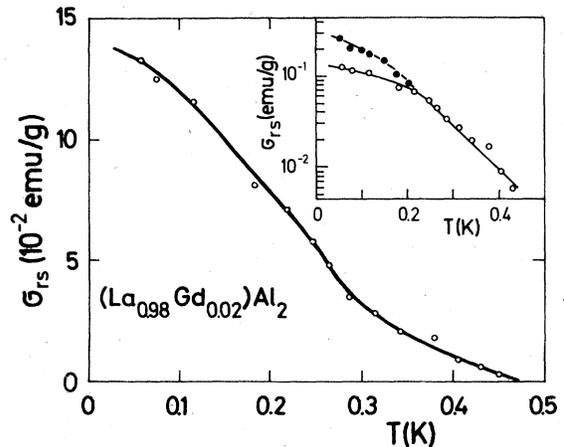


FIG. 4. Saturated remanent magnetization σ_{rs} as a function of temperature T for $(La_{0.98}Gd_{0.02})Al_2$ (data taken for $t = 30$ sec after H was switched off). The inset shows the same data plotted logarithmically (open symbols) together with the maximum thermoremanent magnetization, $\sigma_{r,max}^{th}$ (closed symbols) as a function of T .

where $M_g(E_a)$ is the mean moment of those regions and the factor $\frac{1}{2}$ takes into account the projection of M_g on the direction of field initially applied. By partial differentiation with respect to $\ln t$ and T one obtains with Eq. (1)

$$\frac{\partial \sigma_r}{\partial \ln t} = -\frac{1}{2} M_g(E_a) p(E_a) \frac{\partial E_a}{\partial \ln t}, \quad (3)$$

$$\frac{\partial \sigma_r}{\partial T} = -\frac{1}{2} M_g(E_a) p(E_a) \frac{\partial E_a}{\partial T}. \quad (4)$$

From Eqs. (3) and (4) one deduces immediately⁵

$$\frac{\partial \sigma_r}{\partial \ln t} = \frac{1}{\ln t - \ln \tau_0} T \frac{\partial \sigma_r}{\partial T}, \quad (5)$$

which also holds when replacing \ln by \log_{10} .

We note that the relationship between time and temperature dependence of σ_r holds independent of the specific form of $p(E_a)$ and $M_g(E_a)$. Figure 5 shows a plot of $\partial \sigma_{rs} / \partial \log_{10} t$ vs $T(\partial \sigma_{rs} / \partial T)$ for $(La, Gd)Al_2$. Indeed a roughly linear behavior is observed within the (large) error bars. Hence this plot gives further support to the model. For the slope s of the $\partial \sigma_{rs} / \partial \log_{10} t$ vs $T(\partial \sigma_{rs} / \partial T)$ straight line one evaluates $1/s = 7.5$ decades, hence with $\log_{10} \tau_m \approx 1.5$ decades ($\tau_m = 30$ sec): $\tau_0 \approx 10^{-6}$ sec which is not quite an order of magnitude larger than in noble-metal $3d$ spin-glasses.⁹ As a specific function which satisfies Eq. (5), Prejean²⁰ considers σ_{rs} to be of the form

$$\sigma_{rs}(t, T) = \sigma_{r0} \exp \left[-aT \ln \frac{t}{\tau_0} \right], \quad (6)$$

which yields $\partial \ln \sigma_{rs} / \partial \ln t = \text{const}$ for fixed temperature, i.e., $\sigma_{rs} \sim t^{-\alpha}$. As was pointed out in Sec. II B, it is almost impossible to distinguish experimentally the slowly decaying functions $\sigma_{rs} \sim 1 - \log t$ or $\sigma_{rs} \sim t^{-\alpha}$, cf. Figs. 2 and 3 and Ref. 20. Assuming the validity of Eq. (6), the slope of the $\log \sigma_{rs}$ vs $\log t$ curves (Fig. 3) should be given by $\alpha = aT$.

As can be seen from the inset of Fig. 3, α decreases with decreasing temperature. However, α seems to go to zero at a finite temperature, hinting at very small values of α for low temperatures. From Eq. (5), this very slow relaxation of σ_{rs} should coincide with an only weak temperature dependence of σ_{rs} at low temperatures, as is indeed observed, cf. Sec. II C (inset of Fig. 4).

We note that Eq. (6) is based on the assumption of a Gaussian for the distribution of magnetic moments of the spin regions. In other spin-glasses, such a Gaussian has been inferred from $\sigma_{rs}(T)$. Why then does Eq. (6) or a similar expression describe the data on $(La, Gd)Al_2$ satisfactorily only at higher temperatures, but not for $T \rightarrow 0$? This could be due to the fact that $\sigma_{rs}(T \rightarrow 0)$ does not contain contributions from all regions but only from the "harder" ones.

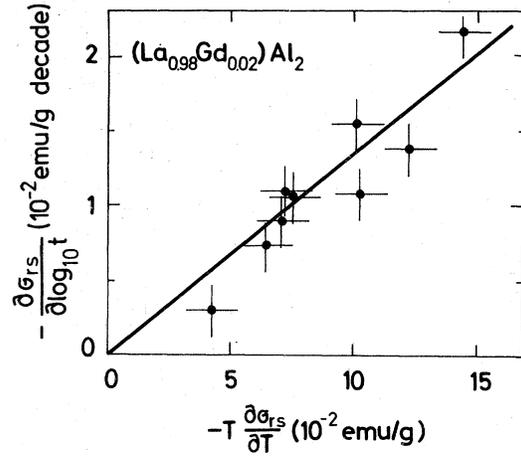


FIG. 5. $-\partial \sigma_{rs} / \partial \log t$ vs $-T \partial \sigma_{rs} / \partial T$ as evaluated from Figs. 2 and 4 for $(La_{0.98}Gd_{0.02})Al_2$. Straight line yields $\tau_0 = 10^{-6}$ sec (see text).

Rather than σ_{rs} , the maximum of the TRM as a function of magnetic field, $\sigma_{r, \text{max}}^{\text{th}}$, in the limit $T \rightarrow 0$ should be taken as a representative of all regions. This idea is supported by the fact that $\sigma_{r, \text{max}}^{\text{th}}(T)$ resembles a bit more closely an exponential decay with temperature and it also could explain why the deviations of σ_{rs} from Eq. (6) are smaller in other spin-glasses,⁹ because there the difference between σ_{rs} and $\sigma_{r, \text{max}}^{\text{th}}$ is rather small (10 to 20%) as compared to $(La, Gd)Al_2$ (a factor of 2). This will be pursued below in the discussion of the field dependence of IRM and TRM.

Usually the saturated remanent magnetization for $T \rightarrow 0$ is thought to be determined by the turning of every region into the direction of applied field on its own anisotropy axis.⁹ Assuming an Ising model, the mean number of uncompensated spins in a region of n spins is $(n)^{1/2}$. With a Gaussian distribution of the magnetic moments of the regions this leads to a remanent magnetization of

$$\sigma_{rs} = \frac{1}{2} N \mu_0 (2n_0/\pi)^{1/2} \quad (7)$$

at $T=0$, where n_0 is the mean number of spins in a region, N the number of regions per unit mass, and μ_0 the magnetic moment of the magnetic ion.⁵ The total saturation magnetization is of course $M_s = N n_0 \mu_0$. Hence, the measurement of σ_{rs} and M_s yields the mean number of spins per region for $T \rightarrow 0$,^{5,9}

$$n_0 = \frac{1}{2\pi} \left(\frac{M_s}{\sigma_{rs}} \right)^2. \quad (8)$$

Table I shows the values of M_s (taken partly from Ref. 12) and σ_{rs} (cf. inset of Fig. 1) for $(La, Gd)Al_2$ alloys, together with the value of n_0 [after Eq. (8)]. The observed linear increase of σ_{rs} with x for $x \leq 4$

TABLE I. Properties of the investigated $(La_{1-x}Gd_x)Al_2$ spin-glass alloys: Freezing temperature T_f , saturated magnetization M_s , saturated remanent magnetization σ_{rs} , and mean number of spins per region n_0 as calculated from σ_{rs} .

x (at. % Gd)	T_f (K) ^a	M_s $\left(\frac{\text{emu}}{\text{g}}\right)$	σ_{rs} $\left(\frac{\text{emu}}{\text{g}}\right)$ ^b	n_0
1	0.25 ^c	2.0	0.07	130
2	0.52 ^c	4.0	0.13	150
4	1.1 ^c	8.0 ^c	0.28	130
6.4	1.8	12.8	0.66	60
8	2.3 ^c	16.0 ^c	1.0	40

^aAs measured by ac susceptibility with $\nu = 16$ Hz.

^bMeasured for $T \rightarrow 0$ ($T = 0.07$ K).

^cTaken from Ref. 12.

at. % leads to a concentration independence of n_0 . The steeper rise of σ_{rs} at higher concentrations leads to an apparent decrease of n_0 . The assumption of a "random-walk" mean moment $\mu_0(2n/\pi)^{1/2}$ is no longer valid because the RKKY interaction in $(La,Gd)Al_2$ is ferromagnetic for nearest and next-nearest neighbors. This leads to quasiclusters in which the spins are aligned parallel and which are of rather small size as compared to n_0 .¹² However, in $(La,Gd)Al_2$ n_0 as found from Eq. (8) should not be identified with the mean number of spins per region for the reason discussed above. Putting $\sigma_{r,\max}^{\text{th}}$ into Eq. (8) yields $n_0' = 30$ instead of $n_0 = 140$ in the scaling regime which is an order of magnitude smaller than the values found for $AuFe$. Since the mean number of spins per region is a rough measure of the strength of the RKKY interaction, the smallness of n_0 in $(La,Gd)Al_2$ reflects the smallness of this interaction in rare-earth spin-glasses. Similar values of the mean number of spins per region have also been reported for amorphous $(La,Gd)_{0.8}Au_{0.2}$ alloys.⁸

We now turn to the discussion of the magnetic field dependence of the thermoremanent magnetization as depicted in Fig. 1. The pronounced maximum of $\sigma_r^{\text{th}}(H)$ is rather unusual as already mentioned. However, all spin-glasses show weak maxima in the TRM. Experimentally, the major difference between $\sigma_{r,\max}^{\text{th}}$ and the saturated remanence σ_{rs} is that σ_{rs} is more stable when submitted to an external perturbation such as heating or application of an inverse field, and σ_{rs} decays very slowly with time, as has been discussed above. Hence σ_{rs} appears as being stabilized compared to $\sigma_{r,\max}^{\text{th}}$ as if the regions having the lowest blocking temperatures, i.e., fastest relaxation times did not contribute to σ_{rs} . $\sigma_{r,\max}^{\text{th}}$ which presumably contains a broader distribution of regions, decreases exponentially with T over a larger temperature regime and decays faster with time than

σ_{rs} . It even has been observed that sometimes upon rapid cooling σ_{rs} is obtained after a jump from a larger value,²⁸ this also hinting at rapid relaxation of part of the "spectrum" of the regions.

All these properties suggest that after cooling in a large enough field the smallest spin regions are mostly oriented randomly with respect to the applied field. This could possibly be due to a local-field effect of the biggest regions similar to what was proposed by Neel²⁹ for the case of rock magnetism. In the case of a spin-glass where the distribution of blocking temperatures is large, one can imagine that the smallest regions which are blocked still below the temperature of the measurement and hence can be reoriented by the effective field, are submitted to a negative local demagnetizing field originating from large regions and therefore contribute with a negative sign to σ_{rs} when the magnetic field increases. Clearly more detailed experiments on the field dependence of σ_r^{th} are needed in order to test this model.

IV. CONCLUSIONS

The notion of spin regions which can relax over an energy barrier and are progressively blocked as the temperature decreases is a quite useful concept to explain phenomenologically the behavior of irreversible properties of spin-glasses. In particular, the relation between time and temperature dependence of the remanent magnetization can be well understood. This general relation also holds in $(La,Gd)Al_2$ alloys, although this spin-glass differs from the canonical transition-metal spin-glasses: The maximum of $\sigma_r^{\text{th}}(H)$ is very pronounced. A possible explanation for this maximum is the existence of negative (with respect to the applied field) local demagnetizing fields originating from large-spin regions so that small-spin regions give a negative contribution to $\sigma_r^{\text{th}}(H)$ at

higher fields. Hence $\sigma_{r,\max}^{\text{th}}$ rather than σ_{rs} should be taken as representative of *all* spin regions. Only the harder part of the spectrum of regions is contained in σ_{rs} . This could explain the relative stability of $\sigma_{rs}(t, T)$ at low temperatures.

The fact that $\sigma_{r,\max}^{\text{th}}$ is so much larger as compared to σ_{rs} in $(La, Gd)Al_2$ than in the canonical transition-metal spin-glasses can perhaps be understood by the existence of many small regions in the former, because the average number n_0 of spins per region is considerably reduced which is presumably due to the weak RKKY interaction in rare-earth metals. The smallness of the average RKKY interaction in $(La, Gd)Al_2$ has also been extracted from susceptibility and magnetization measurements.¹² It might account for the small value of the freezing temperature in

rare-earth spin-glasses, because the anisotropy energy is expected to be proportional to n_0 . This would also explain the observable frequency dependence of T_f in $(La, Gd)Al_2$.¹⁹ Consistently enough, the fact that time effects are appreciably larger in this system as compared to transition-metal spin-glasses is reflected by a much faster time decay of the remanent magnetization.

ACKNOWLEDGMENTS

We thank F. Steglich and R. Tournier for valuable discussions. One of the authors (H.v.L.) is indebted to the Deutsche Forschungsgemeinschaft which made his stay at Grenoble possible, and to the Centre de Recherches sur les Très Basses Températures for its hospitality.

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