

Phenomenological study of the amorphous $\text{Fe}_{80}\text{B}_{20}$ ferromagnet with small random anisotropy

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(Received 1 November 1989)

The magnetic behavior of some amorphous ferromagnets of composition $\text{Fe}_{80-x}\text{R}_x\text{B}_{20}$ (R being a rare-earth element) is investigated as a function of the external applied magnetic field and temperature using dc magnetic measurements. Random magnetic anisotropy is generated by dilution of rare-earth atoms in the $\text{Fe}_{80}\text{B}_{20}$ ferromagnetic matrix. Hysteresis curves show a quasireversible behavior with very small coercivity and remanence, suggesting a weak random magnetic anisotropy. In the high-applied-field regime the samples show ferromagnetic saturation, and from the M values it is possible to conclude that the light rare-earth atoms (Ce, Nd) are ferromagnetically coupled with the iron atoms, whereas the heavy atoms (Gd, Dy) couple ferrimagnetically to the Fe moments. The temperature dependence of the magnetization has also been studied in the conventional spin-wave framework, and the values obtained for the spin-wave stiffness constant D are close to $100 \text{ meV } \text{Å}^2$, which is typical for this kind of material. In the low-applied-field and low-temperature regime a much more complex behavior is observed as a consequence of the competition between local random anisotropy and exchange interactions. The different dependence on T of the correlation length associated to the local random anisotropy and to the exchange interactions makes possible the existence of different magnetic orderings, but no phase transition is observed between them.

I. INTRODUCTION

Metallic glasses based on $3d$ transition metals are usually soft ferromagnets with small coercivity and are easily saturated in the low-applied-field regime. Amorphous $\text{Fe}_{80}\text{B}_{20}$ is a typical example of this kind of material, which has, in addition to its rather high-saturation induction of 16 kOe, a remarkable magnetostriction λ_s of about $+38 \times 10^6$. It has been shown previously that the dilution of some amount of rare-earth atoms may drastically change these properties.^{1,2}

In the case of rare-earth ions, the $4f$ electronic waves functions are responsible for the formation of localized magnetic moments. The $4f$ electrons occupying an inner shell are strongly localized and retain much of their atomlike character with very little or even no quenching of the orbital angular momentum.

The rare-earth atoms have large spin and orbital moments and large spin-orbit coupling constants; so a strong coupling of the spins to the local environment is expected. A direct consequence of the topological disorder inherent to the amorphous state is the existence of highly irregular local electrostatic fields. By means of the interplay between these crystalline electrostatic fields and the aspherical $4f$ electron clouds of the rare-earth ions, a high magnetocrystalline anisotropy results.

Harris, Plischke, and Zuckerman (HPZ)³ were the first to propose a magnetic model to account for the magnetic properties of these materials. The HPZ hamiltonian is a Heisenberg model in which each spin has the same magnitude but is subjected to a local random anisotropy field:

$$\mathcal{H} = \sum_i V_i - \frac{1}{2} J \sum_i \delta \mathbf{J}(i) \mathbf{J}(i + \delta), \quad (1)$$

where V_i is a local single-ion anisotropy field at site i , $\mathbf{J}(i)$ is the angular momentum operator for the magnetic ion on site i and J is the nearest-neighbor coupling constant of the Heisenberg model. In principle, fluctuations in the value of J should be expected due to the structural disorder of the amorphous alloys, but it is argued that the influence of the random magnetic anisotropy on the magnetic properties of the rare-earth alloys is dominant over those based on the exchange interactions;⁴ however, this argument seems to hold only in the limit of very high fields.⁵

The most commonly used expression for the anisotropy field is⁴

$$\mathcal{H}_{\text{RMA}} = -D \sum_i (J_{z_i})^2, \quad (2)$$

where D is a constant to gauge the random anisotropy strength.

Expression (2) is only meaningful when D is positive. The amorphous nature of the system is described by assuming that the easy axes Z_i are randomly oriented through the sample.

The first studies of rare-earth alloys were done by means of a mean-field treatment of the HPZ hamiltonian and it was believed to possess long-range ferromagnetic or ferrimagnetic order below a finite temperature.^{6,7} Besides these ideas, some theoretical papers pointed out some doubts about the mean-field approximation predictions of the HPZ hamiltonian.^{8,9}

Finally, Aharony and Pytte¹⁰ calculated the magnetic equation of state for random magnetic anisotropy systems and they showed that no long-range magnetic order exists in systems with less than four dimensions.

The effect of an applied magnetic field in a RMA system was first studied by Chudnovsky *et al.*^{11,12} These authors have shown that the magnetic structure of such amorphous magnets with ferromagnetic exchange and RMA depends crucially on the parameter $\Lambda_r = \lambda_r (R_\alpha/a)^2$, where λ_r is the ratio of the random magnetic anisotropy strength to exchange, R_α is the scale of the spatial correlation of the easy axes, and a is the interatomic distance.

Following their notation three characteristic fields may be introduced. (i) The exchange field $H_{\text{ex}} \equiv \alpha M_0/R_\alpha$ where the parameter α is proportional to $J a^2$ and M_0 is the saturation value of the magnetization which depends on the temperature and on the short-range exchange constants. (ii) The random anisotropy field $H_R = \beta_R M_0$, where β_R is proportional to the average random magnetic anisotropy constant D . (iii) The coherent anisotropy field $H_C = \beta_C M_0$, where β_C is proportional to the coherent anisotropy strength D_c .

Depending on the relations between these different terms and the external applied field H a wide variety of magnetic arrangements are found.

In the strong random anisotropy limit, namely $H_R > H_{\text{ex}}$, each spin is almost directed along the local random anisotropy axis at its site. As a consequence the magnetic susceptibility is small and a very large magnetic field is needed to reach a reorientation of the spins in the hemisphere defined by the field. In this case the system

exhibits a finite coercivity and hysteretic behavior.¹³

In the limit of weak random anisotropy ($H_R < H_{\text{ex}}$) the system retains some aspects of its collective behavior when no random anisotropy is present. It shows large magnetic susceptibility and large but finite ferromagnetic-like correlation length. Depending on the strength of the external applied field, H , three different regimes are observed. (i) When $H=0$ the spins are disordered and this state is called correlated spin glass (CSG). The main macroscopic features are the absence of hysteresis and the large zero-field susceptibility. (ii) When an external field is applied an alignment of the spins occurs and if the field is strong enough ($H > H_S \equiv H_R^4/H_{\text{ex}}^3$), aligns the CSG leading to a new state denominated ferromagnet with wandering axes (FWA). This is a slightly noncollinear structure in which the deviation of the magnetization relative to the field changes over the system. (iii) If the applied field is increased further the situation is very similar to the FWA case but now the spins lie even closer to the direction of the applied magnetic field.

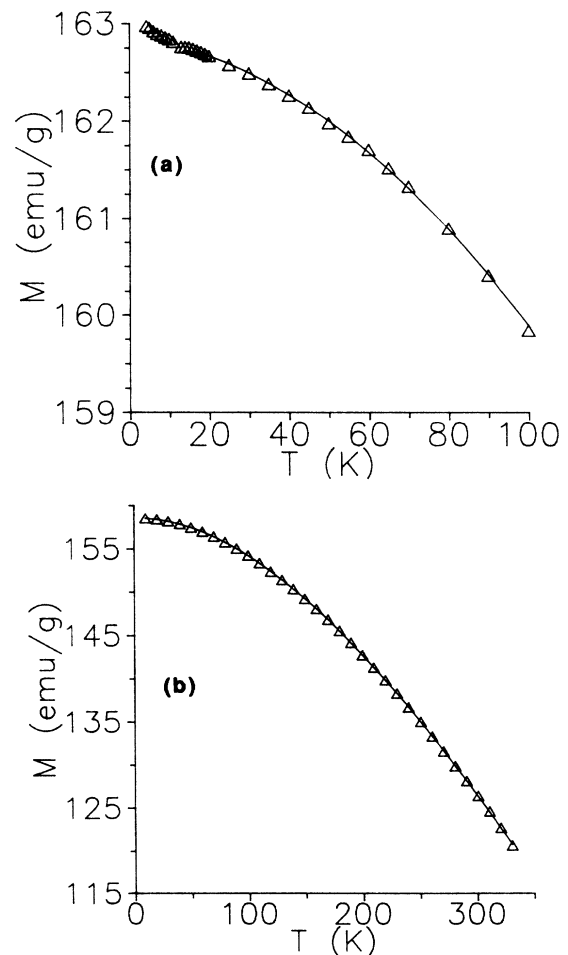


FIG. 1. Variation of the saturation magnetization with temperature at constant applied field for some of the $\text{Fe}_{74}\text{R}_6\text{B}_{20}$ samples: (a)— $R = \text{Ce}$; (b)— $R = \text{Nd}$. Solid line corresponds to the fit following Eq. (3).

TABLE I: Values of the fitted parameters of Eq. (3), spin-wave stiffness constant D , and the effective anisotropy constant K . [Values of the C and A parameters of Eq. (3) have been omitted because they are two or three orders of magnitude smaller than B .]

Sample	M (emu/g)	B ($10^{-5}K^{-3/2}$)	D (meV \AA^2)	D/Tc	K (10^6erg cm^{-3})
Ce	163.(5)	2.2	103	0.43	2.3
Nd	158.(5)	2.0	116	0.47	2.4
Lu	147.(5)	3.6	81	0.42	1.5
Gd	95.(5)	3.5	109	0.39	0.8
Dy	92.(5)				1.3

II. EXPERIMENTAL

A set of ribbons of the composition $(\text{Fe}_{80-x}\text{Re}_x)\text{B}_{20}$ ($X=6$ and $R = \text{Ce, Nd, Gd, Dy, and Lu}$) were produced by the single roller technique in a closed chamber with an He gas protective atmosphere. The melt was quenched on a Cu-Zr wheel (diameter 200 mm). The surface velocity was varied between 37 and 47 m/s. The amorphous state of the ribbons was tested by x-ray but also by optical microscopy, and no evidence of crystalline inclusions was detected.

DC-magnetic measurements of the samples were carried out by using a commercial SHE-SQUID magnetometer with in-plane applied fields up to 50 kOe in the temperature range between 4.2 and 350 K.

III. RESULTS AND DISCUSSION

A. Temperature dependence of the saturation magnetization

The temperature dependence of the saturation magnetization in the high-applied-field regime was studied by measuring the variation of the magnetization in a constant applied field while heating the sample from 4.2 to 250 K. In the case of the Nd sample the study was done up to 350 K to check if some special behavior was present. The experimental data obtained in this way were analyzed in the framework of the classical spin-wave theory. In the presence of an applied field the experimental data should be analyzed by means of the formula¹⁴

$$M(T)/M_0 = 1 - B[F(\frac{3}{2}, t_H)/\zeta(\frac{3}{2})]T^{3/2} - C[F(\frac{5}{2}, t_H)/\zeta(\frac{5}{2})]T^{5/2} - AT^2, \quad (3)$$

where M_0 is the saturation magnetization value at $T=0$ K, $\zeta(\frac{3}{2})$ and $\zeta(\frac{5}{2})$ are the Riemann ζ function, $F(s/t_H)$ is the Bose-Einstein integral function, and t_H is defined by the expression

$$t_H = (Tk_B)/(g\mu_B H_{ap}), \quad (4)$$

where k_B is the Boltzman constant, g is the Lande factor, μ_B is the Bohr magneton, and H_{ap} is the external applied field.

Expression (3) takes into account the effect of the applied field on the spin-wave spectrum first studied by Holstein and Primakoff.¹⁵ The term in T^2 is included to account for possible Stoner-type excitations.

The spin-wave stiffness constant D of the dispersion law is related to the B coefficient of the modified Bloch's law given by Eq. (3) through the expression

$$B = [\zeta(\frac{3}{2})g\mu_B/M_0](k_B/4\pi D)^{3/2}. \quad (5)$$

In Fig. 1 we show the low-temperature behavior for some of the samples. The full line represents the theoretical $M(T)$ curve obtained by using Eq. (3) and the symbols correspond to experimental points. The results obtained by the fitting procedure are summarized in Table I. It is evident from Fig. 1 that for temperatures higher than approximately 15 K the theoretical curve shows a very good agreement with the experimental data points. In the very-low-temperature regime some discrepancies with the theoretical curve are evident especially in Gd and Ce samples. These discrepancies can be explained taking into account the local random anisotropy of the samples and the different dependence on T of the correlation length associated with random anisotropy and exchange interactions. The case of the Dy sample is completely different and shows a $M(T)$ curve with a broad peak, even in external applied field up to 50 kOe, which could be related to some kind of ferrimagnetic ordering (see Fig. 2). The broadening of the peak is due to the noncollinear magnetic structure induced by the local random anisotropy.

From the data included in Table I the spin-wave

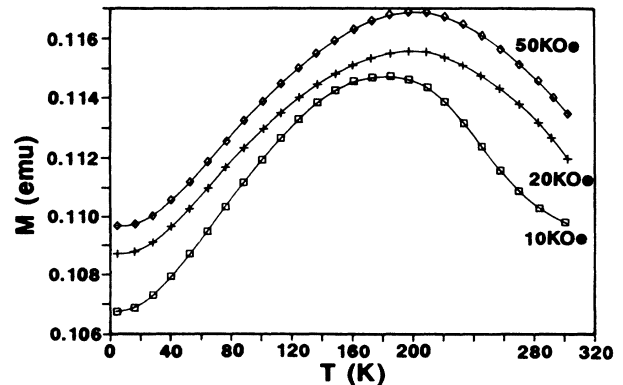


FIG. 2. Thermal variation of the magnetization for $\text{Fe}_{74}\text{Dy}_6\text{B}_{20}$ sample for different values of the external applied field.

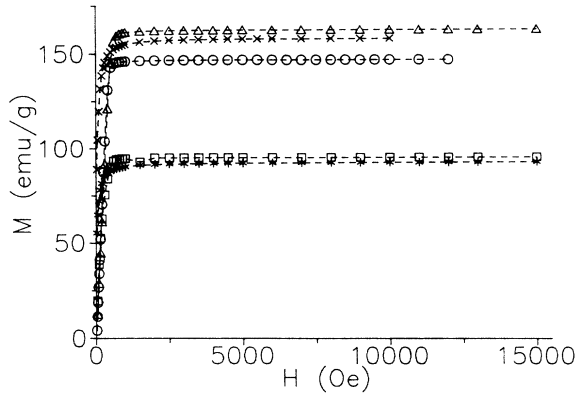


FIG. 3. Isothermal magnetization curves at $T=4.2$ K for $\text{Fe}_{74}\text{Re}_6\text{B}_{20}$ samples: $R = \text{Ce}$ (Δ), $R = \text{Nd}$ (\times), $R = \text{Lu}$ (\circ), $R = \text{Gd}$ (\square), and $R = \text{Dy}$ ($*$).

stiffness constant D could be calculated. The values obtained are close to $100 \text{ meV } \text{\AA}^2$ which is typical for these materials.^{16,17}

B. Magnetization curve

The variation of the magnetization as a function of the applied field is shown in Fig. 3. Comparing the values of the saturation magnetization it is possible to get the following information about the magnetic ordering in each sample. (i) M_0 of the Ce and Nd samples lies above that of the Lu sample indicating a ferromagnetic coupling between the R moments and the Fe moments. The mean moment per R atom is $2.44\mu_B$ per Ce ($2.54\mu_B$) and $1.67\mu_B$ per Nd ($3.62\mu_B$) where the free-ion values are given in parenthesis. It is worth noting that Ce has a magnetic moment which is close to the trivalent state. (ii) M_0 of the Gd and Dy samples lies below that of the Lu sample indicating a ferrimagnetic coupling between Fe and Re moments. The mean moment per Re atom is $8.16\mu_B$ per Gd ($7.94\mu_B$) and $8.74\mu_B$ per Dy ($10.63\mu_B$), respectively.

Regarding now the magnetic moments of the light rare-earth atoms (Ce and Nd) and also those of the heavy rare-earth atoms (Gd and Dy) in these compounds, there is no doubt that the light rare-earth atoms couple ferrimagnetically to the Fe moments whereas the heavy rare-earth atoms couple ferromagnetically.

In Fig. 4 we show representative hysteresis loops for some of the samples. We can observe a quasireversible behavior with no coercivity in the case of the samples containing Lu, Ce, Nd, and Gd. These features were predicted by Chudnovsky *et al.*¹² as one of the fingerprints of the correlated spin-glass state. This state is characterized by a smooth rotation of the magnetization over the total volume of the sample so that the directions of the magnetization are ferromagnetically correlated within regions of size R , much greater than the interatomic distance. The main macroscopic features of a correlated spin glass are the reversible magnetization curve and the large zero-field susceptibility $\kappa \cong \Lambda_R^{-4}$.

In the case of the Dy sample some irreversibility is found and the value of the coercive field is approximately 20 Oe. The different behavior of the sample containing Dy could be understood taking into account that it is the element with the bigger anisotropy. When the random anisotropy is large ($H_R > H_{ex}$), each spin is almost directed along the random anisotropy axis at its site. The "arrow representation" of this magnetic state may be the same as for a spin glass. The magnetic susceptibility is very small ($\chi \cong M_0/H_R$) and the system exhibits a finite coercivity and hysteretic behavior.¹⁹ In this case, due to the strong local random anisotropy the collective behavior is only a secondary phenomenon.

C. Low-field magnetic behavior:

To determine the region of irreversibility, samples were cooled to 4.2 K in the zero applied field (zero-field cooling process — ZFC), then the field was applied and $M_{ZFC}(H)$ measured up to 250 K. The temperature was then reduced to 4.2 K again and the $M_{FC}(H)$ measured in the same applied field [field cooling process — FC].

The low-field magnetization curves obtained in this way are shown in Fig. 5 for the Nd and Ce samples.

The onset of irreversibility starts before the cusp of the peak indicating that there are not individual spins but clusters of spins which are involved in the freezing

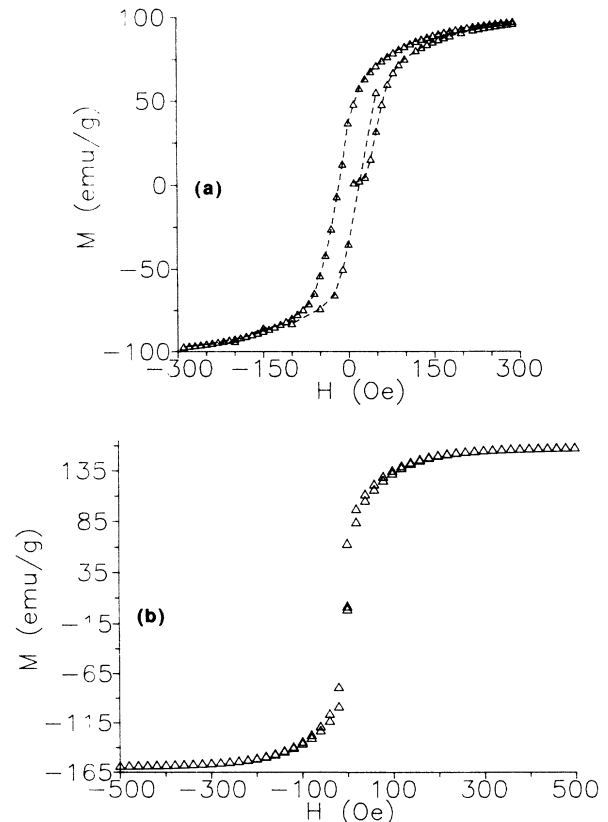


FIG. 4. Hysteresis loops at $T=4.2$ K for some of the samples: Dy (a) and Nd (b).

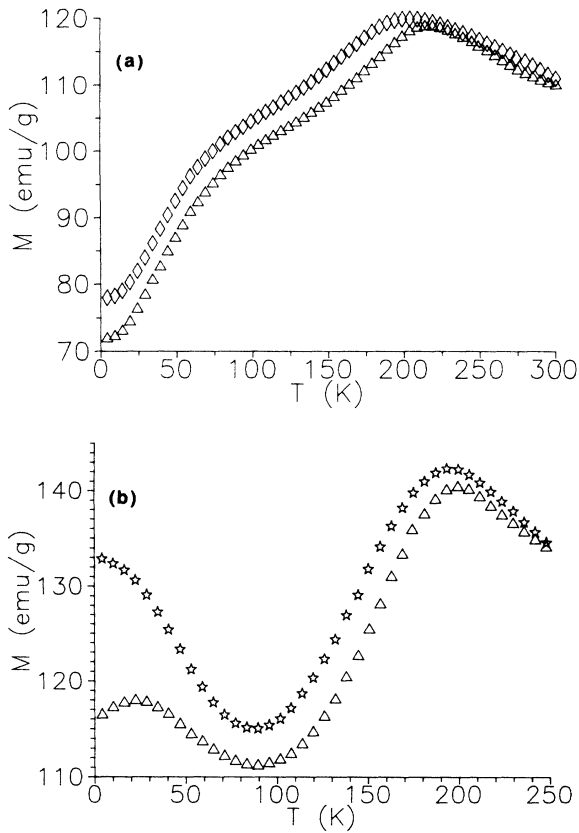


FIG. 5. Low-field magnetization curves obtained in a ZFC-FC process for Nd and Ce samples: (a) Ce [\diamond (F.C.); \triangle (ZFC)]; (b) Nd [\star (FC); \triangle (ZFC)].

phenomenon as has been previously observed in other systems with spin-glass-like behavior.

The broadening of the peak points out the existence of a noncollinear magnetic structure in the samples due to the local random anisotropy. Furthermore, we observe two peaks in the susceptibility — one for high temperatures ($T \approx 150$ – 200 K) and another one in the low-temperature regime. These two peaks may be related with different freezing phenomena. The high-temperature peak could be associated with the freezing of the clusters of spins with local ferromagnetic ordering. As the temperature decreases the influence of the local random anisotropy increases and the size of the regions with local ferromagnetic order becomes smaller and smaller. For temperatures low enough, all the spins almost point along the directions of the local random anisotropy axes and the system displays another spin glass-like transition. The lowest-temperature peak may be associated to this second freezing phenomenon.

In Fig. 6 we show the dependence of the $M(T)$ curves as a function of the applied field strength measured after a process of field cooling. As the intensity of the external applied field increases, the peak moves to lower temperatures following a linear relation $T_{\text{PEAK}} = aH_{\text{AP}} + b$ (see Fig. 7). This variation of the position of the peak also makes it evident that the magnetic ordering of the system

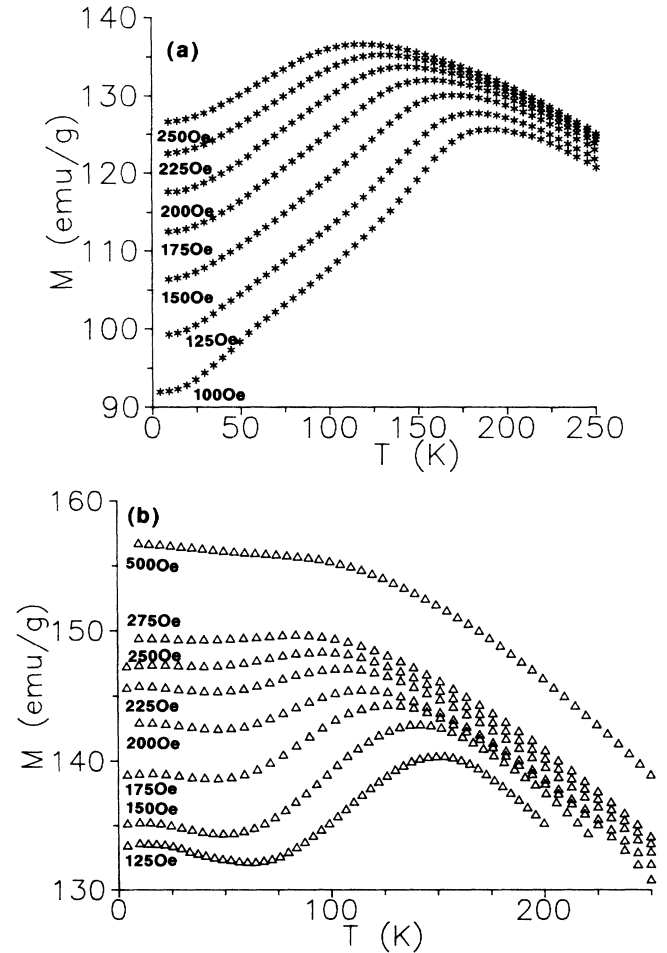


FIG. 6. Thermal variation of the FC magnetization in low-applied-field regime: (a) Ce; (b) Nd.

differs from that corresponding to a canonical spin glass. It is well known that in spin-glass systems the position of the peak in the ZFC curve follows the AT line (Almeida-Thouless line)¹⁸ for Ising spin glasses, or a GT line

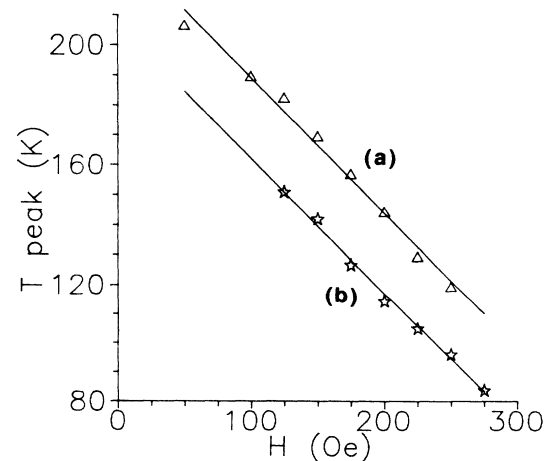


FIG. 7. Variation of the temperature corresponding to the peak position as a function of the external applied field: (a) Ce; (b) Nd.

(Gabay-Toulouse line)¹⁹ for isotropic spin glasses (Heisenberg spins).

Due to the decrease of the correlation length between the local random anisotropy axes when the temperature increases, a crossover from a spin glasslike magnetic phase (strong random anisotropy limit at low temperature) to a correlated spin-glass phase with a large ferromagnetic correlation length and large susceptibility may occur. This crossover is evident in our samples (see Fig. 6). At the same time, a similar crossover induced by increasing the external applied field is observed.

Following the notation of Chudnovsky *et al.* we can characterize the different magnetic states of our samples. In the limit of weak random anisotropy ($H_R < H_{ex}$) the behavior of the system displays some aspects of its collective behavior when no random anisotropy exist. The characteristic correlation length over which the spins retain local ferromagnetic order is given by $R_A(H_{ex}/H_R)^2$.

When an external field H_{AP} is applied, an alignment of the spins occurs and if the field is strong enough ($H_{AP} > H_S \equiv H_R^4/H_{EX}^3$) aligns the CSG, leading to a slightly noncollinear structure in which the deviation of the magnetization relative to the field changes over the

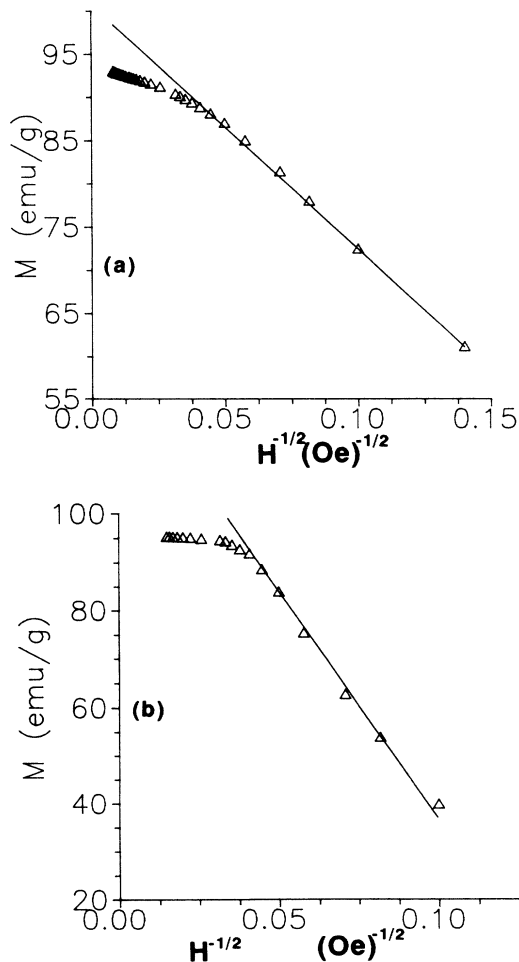


FIG. 8. Magnetization dependence on $H^{-1/2}$. Solid lines correspond to the straight-line fit: (a) Dy; (b) Gd.

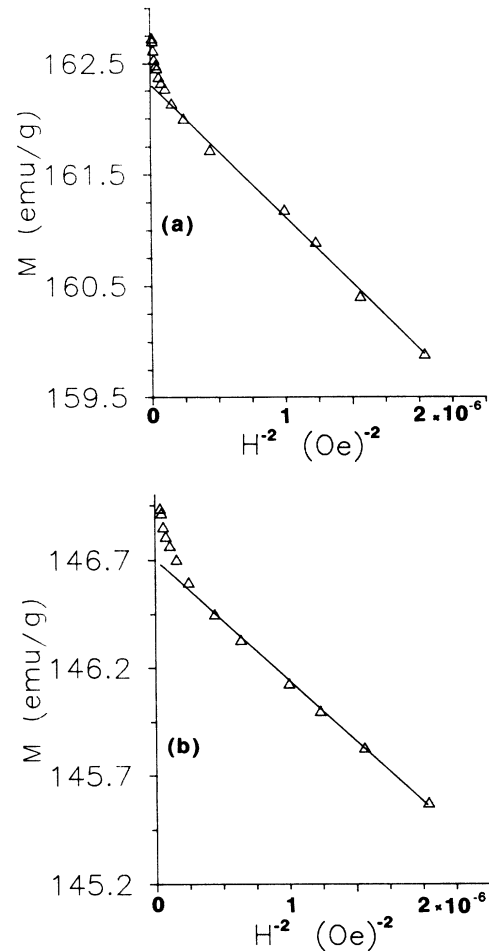


FIG. 9. Variation of the magnetization vs H^{-2} in the intermediate field regime: (a) Ce; (b) Lu.

system. This new magnetic structure is called a ferromagnet with wandering axis (FWA). The approach to magnetic saturation in this state varies as $H^{-1/2}$. We observe this dependence in our samples in the field range from 50 to 500 Oe approximately (see Fig. 8). As we increase the external applied field, the noncollinear structure is closed even more in the direction of the field. Each spin is only slightly tipped from the field direction by the random anisotropy at its site. The magnetization deviation in the law of approach to saturation is proportional to (H_R/H_{AP}^2) , following the expression^{11,20}

$$\delta M/M_0 = \frac{1}{15} [H_R / (H_{AP} + H_{ex})]^2 \quad (6)$$

We find this dependence in our experimental data in the range of applied fields to be from 0.5 to 4–5 kOe (Fig. 9).

In preceding studies^{21,22} a fit of $M(H)$ is shown that follows a $H^{-1/2}$ law over a range of fields from 10 kOe to 50 or even 80 kOe, but it must be pointed out that the required condition that H_{AP} should be bigger than H_S is not satisfied. The author's conclusion is that the values of H_S obtained from the fitting are only an upper bound and that the condition $H_{AP} > H_S$ for the FWA approach to saturation is too general to be true in all the cases.²²

From the slopes of the straight lines in Fig. 9 an effective anisotropy constant K can be calculated^{1,21} and the values obtained are between 1×10^6 and 2×10^6 erg cm⁻³ in rough agreement with early estimates of Cornelison and Sellmyer¹ and Sellmyer and Nafis²¹ in systems with local random anisotropy.

In all the preceding discussion we have made no mention of the coherent anisotropy, but it could have an important influence on the magnetic behavior. The coherent anisotropy may have different origins. One of the most common origins is the tensile stresses in the preparation of the samples in the very fast cooling process or even in the manufacturing process to fix the sample in the experimental setup. The effect of the coherent anisotropy may stabilize the long-range magnetic order in the samples against the influence of the local random anisotropy.²³ In the notation of Chudnovsky's *et al.* notation the coherent anisotropy plays a role completely similar to the external applied field and all the preceding expressions are valid changing H_{AP} by $(H_{AP} + H_C)$. The existence of a considerable amount of coherent anisotropy in the samples could make the changes more reliable in the behavior that we observe with relatively small variations of the external applied field.

D. Scaling analysis

The study and understanding of the phase transition in random and disordered systems is one of the more intriguing problems in condensed matter. In the case of systems with local random anisotropy it is still an open question as to whether the transitions are true phase transitions based in equilibrium statistical mechanics or glass transitions in which the relaxation times smoothly increase as the temperature is lowered and finally become equal to the measuring time at T .

Pioneering work in this field was done by Coey *et al.*²⁴ who attempted to describe the spin freezing at T in the Dy_{0.41}Cu_{0.59} system as a second-order phase transition. Working within the same limitations of strong local random anisotropy Sellmyer and Nafis^{21,25} carried out a scaling analysis of the nonlinear susceptibility as in the spin-glass problem and their results seem to confirm the existence of a phase transition in a strong random magnetic anisotropy system.

Within the limitations of weak random anisotropy some attempts have been made to explore the validity of a phase transition description. In this case of the system $(Dy_x Y_{1-x})Al_2$, a scaling analysis based on equilibrium statistical mechanics has been performed.^{22,26} In this case, the usual ferromagnetic equation of state was considered²⁷

$$M/|t|^\beta = F(H/|t|^{\beta\delta}), \quad (7)$$

Where $F(X)$ is a scaling function; β and δ are the usual critical exponents, and $t = (T - T_F)/T_F$ is the reduced temperature. A very good scaling of the data is obtained in a wide range of fields and temperatures giving a strong indication of a true thermodynamic phase transition.

We have tried to perform a similar scaling study in our samples by using Eq. (7) but no scaling was obtained in any of the samples. Since no spontaneous moment ever develops in the samples, as can be easily shown by representing the magnetization in a typical Arrot plot, the magnetization M cannot be used as a proper order parameter for those compounds. So it is an open question as to whether or not the scaling analysis based on Eq. (7) is meaningful in systems with weak local random anisotropy.

The scaling analysis of the nonlinear susceptibility proposed by Sellmyer and Nafis²⁵ in the strong random anisotropy limit seems to be inadequate for the weak random anisotropy systems studied in the present work. In any case it is not clear as to how to apply this kind of scaling analysis to systems that display reentrant behavior.

IV. CONCLUSIONS

The dilution of some amount of rare-earth atoms in the amorphous ferromagnetic matrix Fe₈₀B₂₀ completely changes the magnetic behavior of the system from ferromagnetism to some new magnetic ordering that displays the main macroscopic features associated with the so-called correlated sping glass state proposed by Chudnovsky *et al.*

In the low-applied-field regime the magnetization curve for all the samples except Dy displays a quasireversible behavior with very small coercivity and remanence suggesting the existence of weak random magnetic anisotropy.

The law of the approach to magnetic saturation follows a $H^{-1/2}$ dependence for low fields and changes to H^{-2} as the external applied field is increased in good agreement with the theory of Chudnovsky *et al.*

At low field and low temperature the system exhibits a crossover from the CSG state or FWA state to a ferromagnetic state as both field and/or temperature are increased.

Even though the magnetization data display a broad peak related with some spin-freezing phenomenon, the scaling study performed with these data by using Eq. (7) does not show any evidence of the nature of the involved transition. Furthermore, the field dependence of the low-field-magnetization peak indicates clearly that the system is not a conventional spin glass.

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