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Rare-earth—gallium—iron glasses.

II. Anomalous magnetic hysteresis in alloys based on Pr, Nd, and Sm

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Giant magnetic coercivity is reported in several metallic glasses of the form $(R_{80}G_{20})_{100-x}Fe_x$ where R represents Pr, Nd, or Sm, G represents Ga or Au, and $15 \leq x \leq 30$. An unusual temperature variation of the coercive field is observed showing peaks at intermediate temperatures (≈ 90 K). In contrast to similar glasses based on heavy rare-earth metals, these glasses exhibit significant chemical short-range order and even phase separation as is shown by the Mössbauer effect and other measurements. The results are consistent with a recent theory which predicts that large coercivity can result from the presence of site-to-site variations in magnetic properties.

I. INTRODUCTION

Magnetic properties of metallic glasses based on heavy rare earths have been studied relatively extensively over the past few years.¹⁻⁵ One reason for this is the relative ease of interpretation of the experimental results of magnetic measurements of heavy-rare-earth-based glasses as compared to metallic glasses based on light rare earths. This may be because the $4f$ wave functions responsible for magnetic properties of light rare earths are less deeply "buried" within the ion than in the heavy rare earths and thus crystal-field (CF) effects become important.⁶⁻⁸ Fert and Campbell⁹ have shown that the general quadrupole electric field Hamiltonian,

$$H_{CF} = \alpha J_x^2 + \beta J_y^2 + \gamma J_z^2, \quad (1)$$

is necessary to adequately explain magnetic behavior in these systems. Here x , y , and z are local principal axes for each ion. Thus, a mixing of crystal-field states may complicate the magnetic behavior of metallic glasses based on light rare earths, especially for non-Kramers ions.^{10,11} Results of these effects might be seen in the reduction of the initial susceptibility and a reduction of the "spontaneous" moment obtained by high-field measurements to a value below that expected, that is, a moment of $\frac{1}{2}gJ$ per magnetic ion if the structure is asperomagnetic. Recently, there has been considerable evidence that light-rare-earth-based metallic glasses with Fe form materials with magnetic ordering temperatures higher than heavy rare earths with equivalent de Gennes factors.^{12,13} This is attributed to the size of the lanthanide ion.¹⁴ Recently, we have reported results of magnetic measurements on the glasses $(Pr_{80}Ga_{20})_{100-x}Fe_x$ with $0 \leq x \leq 30$, which show unusually large coercive forces and other anomalies.¹⁵ Other workers have reported on Fe-rich Nd and Pr-based alloys with similar findings, but with smaller low-temperature coercivities.¹⁶⁻²⁰ In this paper we expand our study of the above alloys to include Nd-based alloys of the same stoichiometry as above as well as the Sm-based system $(Sm_{80}Ga_{20})_{85}Fe_{15}$. We report results of high-field magnetization, magnetic susceptibility, and x-ray diffraction, as well as some ⁵⁷Fe

Mössbauer-effect results.

The results of these measurements are interpreted in terms of a "fluctuation" model of magnetic hardness proposed by Paul.²¹ Here the term "fluctuation" is taken to be a site-to-site variation in magnetic properties such as anisotropy and exchange. Thus variations in the concentration of magnetic ions from region to region in a sample would cause site-to-site variations in magnetic properties and lead to magnetic hardness within this model. The temperature dependence of H_c , the intrinsic coercive force, is shown to be consistent with a theory of thermal activation of coherent rotations of single-domain particles presented by Gaunt.^{22(a)} Finally, the presence of a *peak* in H_c vs T , as well as shifts of the low-temperature magnetization loops, are explained in terms of the presence of more than one magnetic phase in these materials. It is shown that the magnetic properties of this series of alloys are dominated by the presence of a phase separation into Fe-rich and Fe-deficient regions. The presence of giant intrinsic magnetic hardness, observed in many of the anisotropic alloys, is seen to be due to the heterogeneous nature of the multiphase systems involved.

This is a companion paper to another, hereafter denoted as paper I.^{22(b)} The glasses of paper I contained similar constituents except that the rare earths were all *heavy* rare earths. Most of the experimental techniques and sample-preparation methods in this paper are described in I. However, the technique used in interpreting the very broad ⁵⁷Fe Mössbauer spectra observed in Fe-containing magnetic amorphous materials was developed by Window.²³

II. RESULTS

A. Pr-based glasses

dc susceptibility for $Pr_{80}Ga_{20}$ is shown in Fig. 1. Least-squares fitting to the Curie-Weiss law gave $\Theta \approx 6$ K and $P_{eff} \approx 3.37\mu_B/Pr$ atom. This suggests positive exchange between Pr atoms and normal paramagnetic behavior above about 10 K with only a slight reduction of moment expected below $3.58\mu_B$. Experiments of Faraday

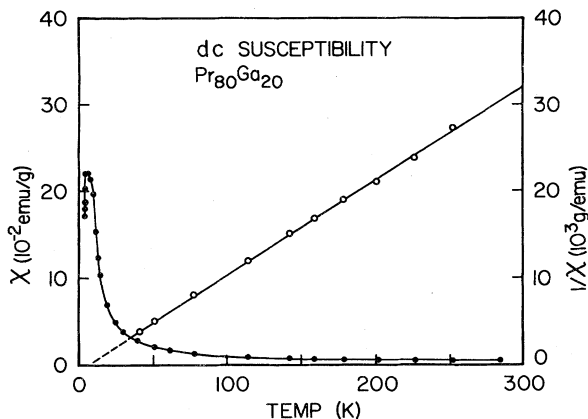


FIG. 1. dc magnetic susceptibility on $\text{Pr}_{80}\text{Ga}_{20}$. The inverse susceptibility is shown by open circles.

and ac magnetic susceptibility show a rapid development of a high-temperature peak as the Fe concentration increases beyond 10 at. %. A low-temperature peak still appears, but a second peak centered at $\approx 375\text{--}450$ K develops in the Pr-based alloys.¹⁵ Thus, there is strong evidence for *two magnetic transitions* in all the alloys containing more than 10 at. % Fe. This observation is consistent with suggestions of Heiman and Lee¹³ in which light-rare-earth-based glasses with Fe form high-temperature magnetic phases.

The spontaneous moment at 4.2 K, for $\text{Pr}_{80}\text{Ga}_{20}$ as determined by fits to the law of an approach to saturation (see paper I), is 35 emu/g or about 31% of the expected moment for aligned Pr^{3+} spins. This suggests a large degree of antiparallel coupling. Measurements of the high-field magnetization of the $x > 10$ alloys shows a more complex behavior with low-temperature hysteresis loops being narrower with less remanence than higher-temperature loops. Figure 2 shows a series of loops at different temperatures for the $x = 20$ alloy. This figure indicates that the magnetic state at 4.2 K is spin-glass-like with small remanence and no saturation in high fields. As T is raised above ≈ 25 K the remanence increases, H_c dramatically increases, and the magnetization seems to be nearly saturated at 80 kOe by the time T is above 100 K. Similar results are obtained for other Fe concentrations ($x > 10$). That this is a product of the presence of an amorphous state is shown in Fig. 3, where measurements on polycrystalline $(\text{Pr}_{80}\text{Ga}_{20})_{70}\text{Fe}_{30}$ are compared to the amorphous materials of the same composition. At no temperature does the polycrystalline sample show significant magnetic hardness. If the amorphous samples are cooled to 4.2 K in the presence of a large magnetic field (80 kOe), large shifts of hysteresis loops along the magnetization axis are observed. It is interesting that the maximum magnetization shift observed (μ_{sh}) for different field-cooling experiments is roughly equal to the expected fully aligned Fe moment for each alloy. The extrapolated spontaneous moment and the temperature dependence of μ_{sh} for $x = 20$ are shown in Fig. 4. In all the alloys with $x > 10$, μ_{sh} disappears above 100 K. Perhaps the most interesting and important feature of these materials is the anomalous temperature dependence, as well as magnitude

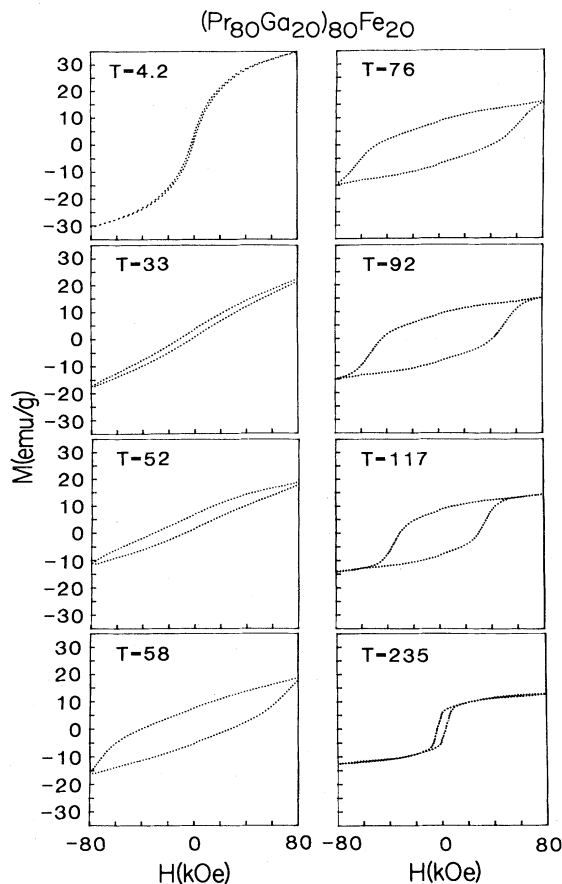


FIG. 2. Various magnetization loops for $(\text{Pr}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$.

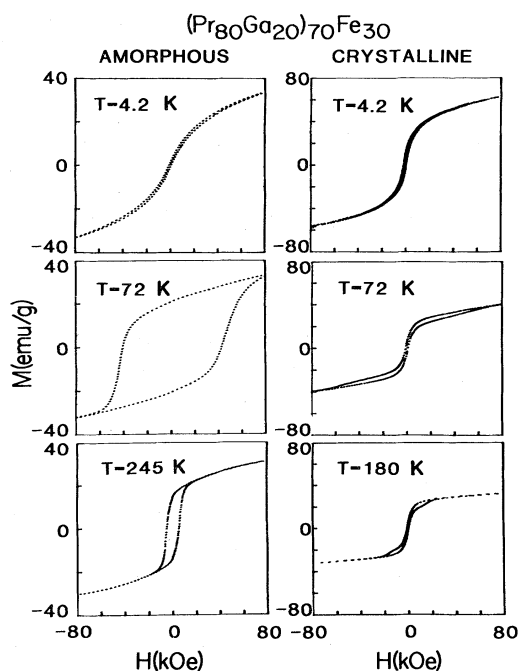


FIG. 3. Magnetization loops for (a) amorphous and (b) crystalline $(\text{Pr}_{80}\text{Ga}_{20})_{70}\text{Fe}_{30}$.

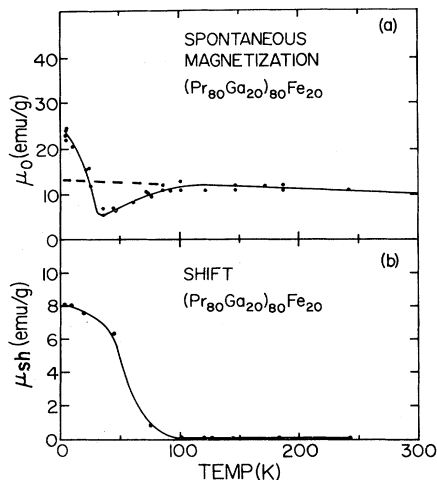


FIG. 4. (a) Spontaneous magnetization of $(\text{Pr}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$. (b) Shift of magnetization loops after field cooling.

of the intrinsic coercive field, H_c . Results of measurements of H_c vs T for $(\text{Pr}_{80}\text{Ga}_{20})_{100-x}\text{Fe}_x$, with $x = 10, 15,$ and 25 , are shown in Fig. 5. The samples were cooled with the field off and the temperature was increased for the measurements. Results are different for field-cooled loops. The peaks are typical of these materials and seem to rapidly develop as the concentration of Fe is increased beyond $x = 10$.

^{57}Fe Mössbauer-effect measurements were made on the $x = 20$ glass from 4.2 to 300 K and on other glasses at 300 K. Figures 6 and 7 show measurements at 300 K for $x = 10$ and 30, respectively. In these figures the solid circles represent actual data points, the solid line represents a fit to the raw spectrum using the Window technique,²³ and the $P(H)$ -vs- H curves represent best fits to the data

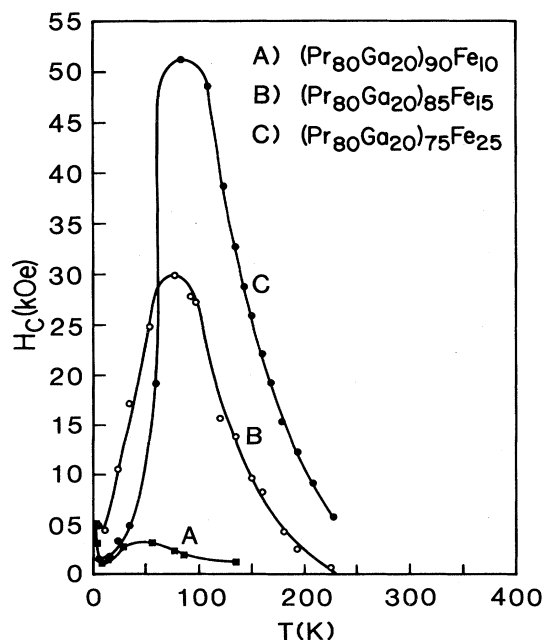


FIG. 5. H_c data for various Pr-based glasses.

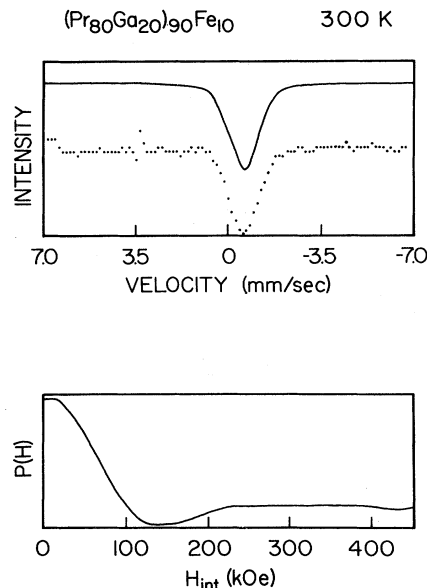


FIG. 6. Mössbauer results on $(\text{Pr}_{80}\text{Ga}_{20})_{90}\text{Fe}_{10}$. Raw spectrum and fit (top), and $P(H)$ distribution obtained (bottom). $T = 300$ K.

and give the probability of internal field H . The cross-hatched low-internal-field peaks are due to the nonmagnetic central portion of the Mössbauer spectrum and represent the program's attempt to fit this central maxima to a six-line spectrum. Using the Gubbens formula,²⁴

$$\mu_{\text{Fe}} = \frac{\mu_B H}{149 \text{ kOe}}, \quad (2)$$

we obtain $\mu_{\text{Fe}} = 1.69\mu_B$ for $x = 30$, and 0 for the $x = 10$ glass. For $x = 20$, μ_{Fe} is seen to be about $1.60\mu_B$ at 300 K, while at $T = 4.2$ K, μ_{Fe} is found to be about $2\mu_B$.

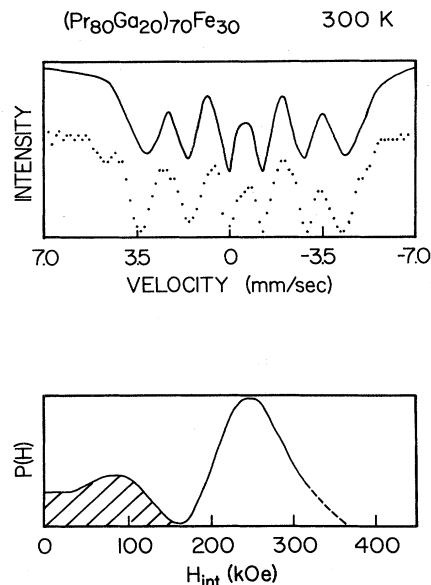


FIG. 7. Mössbauer results on $(\text{Pr}_{80}\text{Ga}_{20})_{70}\text{Fe}_{30}$. Raw spectrum and fitted spectrum (top), and $P(H)$ distribution obtained (bottom). $T = 300$ K.

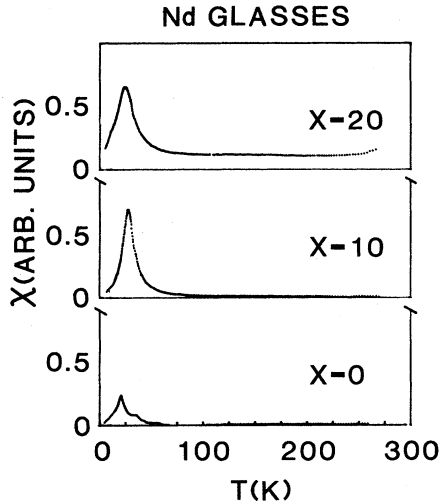
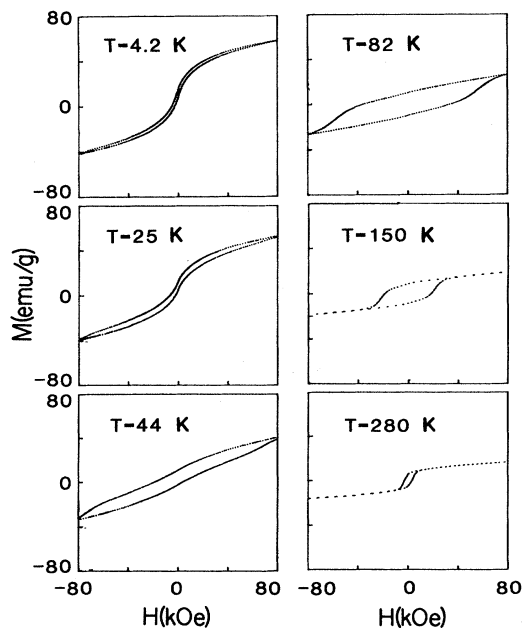
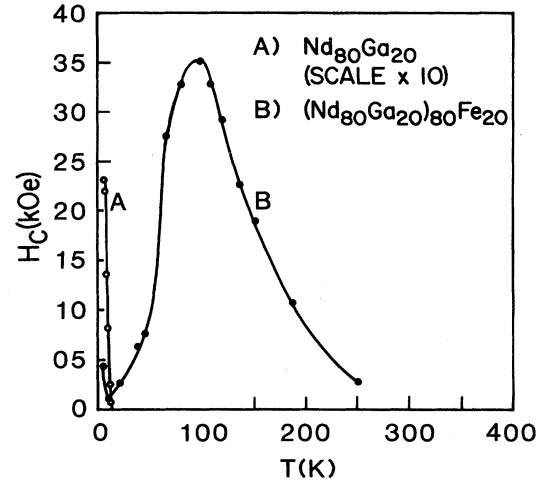


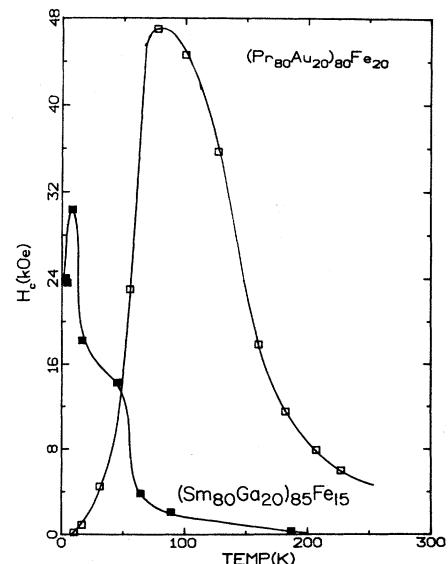
FIG. 8. ac susceptibility on Nd glasses.

B. Nd- and Sm-based glasses

Results on Nd-based alloys are surprisingly similar to corresponding Pr alloys. Figure 8 shows results of ac-susceptibility measurements of Nd alloys with $x=0, 10,$ and 20 . As with Pr-Gr-Fe alloys, a second peak is apparently being approached slightly above room temperature (note the nonzero susceptibility well above the lower-temperature peak for the $x=20$ data). The low-temperature peak is observed in dc measurements at 14 K for $(\text{Nd}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$, and ordinary Curie-Weiss behavior is observed above this temperature. High-field-magnetization measurements are also similar to Pr data and show

FIG. 9. Various magnetization loops for a Nd-based glass showing development of giant coercive fields as T is raised.FIG. 10. H_c vs T for Nd-based glasses.

dramatic increases in H_c as T is raised above the lower ordering temperature. Figure 9 shows vibrating-sample-magnetometer results for $(\text{Nd}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$ at different temperatures. One interesting observation is the fairly constant slope of high-field magnetization which increases as T is lowered from room temperature to about 80 K. Figure 10 shows H_c vs T for two Nd alloys; the $x=20$ alloys has a peak similar to the Pr alloys at about 100 K. One difference is the small rise in H_c that develops below 10 K. A Mössbauer measurement on the $x=20$ glass was performed at 300 K. As with the $x=20$ Pr alloys, a six-line spectrum is observed in the raw data with some enhancement of the central peak above the expected intensity of one-third of the intensity of the first and sixth peaks. The $P(H)$ distribution shows a peak very similar to the $x=20$ Pr glass with $\mu_{\text{Fe}} \approx 1.60\mu_B$ from the Gubbens formula. Many other experiments were performed

FIG. 11. H_c -vs- T data for $(\text{Sm}_{80}\text{Ga}_{20})_{85}\text{Fe}_{15}$ and $(\text{Pr}_{80}\text{Au}_{20})_{80}\text{Fe}_{20}$.

on other alloys based on the light rare earths. Figure 11 shows H_c vs T for the alloys $(\text{Pr}_{80}\text{Au}_{20})_{80}\text{Fe}_{20}$ and $(\text{Sm}_{80}\text{Ga}_{20})_{85}\text{Fe}_{15}$. In each case, significant magnetic hardness is observed at low temperatures, field-cooling effects, such as shifts along the magnetization axis, are observed, and a relatively linear high-field magnetization is observed, suggesting some type of approach to saturation. If a transition metal other than Fe is introduced in the Pr-based alloys, for instance, Mn, Cr, Co, Ni, or Cu, the magnetic hardness is not observed, high-temperature peaks in the magnetic susceptibility disappear, and regular Curie-Weiss behavior is observed at all temperatures above the $\text{Pr}_{80}\text{Ga}_{20}$ ordering temperature. Thus it appears to be the presence of Fe that produces the magnetic hardness within this entire series of alloys.

III. DISCUSSION

A. Phase separation

It has been shown by several authors that a chemical separation of amorphous phases takes place in some amorphous alloys. Cargill *et al.* discuss chemical short-range order in metallic glasses²⁵ and show some alloys in which it occurs. Tenhover²⁶ and Kim *et al.*²⁷ have found chemical short-range order in $\text{Y}_{66}(\text{Fe}_x\text{Mn}_{1-x})_{34}$ and Pb-Au-Sb alloys. Careful analysis of partial pair distribution functions derived from x-ray diffractograms on $(\text{Pr}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$ and other alloys along this series has shown that these glasses have a nonrandom distribution of atoms even though they are amorphous. Furthermore, results of selected-area diffraction and selected-area fluorescence have been performed which give clear evidence of a phase separation in these glasses. This has been reported elsewhere.²⁸ The Mössbauer results support this finding for all the alloys. The results of fitting these spectra using the Window technique give $P(H)$ spectra which show both a nonmagnetic (or paramagnetic) Fe peak as well as a high-field peak from nuclei experiencing strong hyperfine effective fields. It is reasonable to assume that Fe-rich regions have magnetic Fe atoms and regions where Fe atoms have too few Fe near neighbors, or which may have destabilized moments because of local environment effects, contribute to the "nonmagnetic" central portion of the Mössbauer spectrum.²⁹ The relatively high ordering temperatures, up to ≈ 450 K as indicated by dc-susceptibility measurements as well as Mössbauer results, are not unexpected for light-rare-earth iron alloys. Several authors, who studied Fe-rich light-rare-earth alloys,¹⁶⁻¹⁹ find ordering temperatures higher than room temperature. Heiman and Kazama, and Heiman and Lee, conclude that the size of the rare-earth atom is a significant factor in determining the magnetic properties of these materials by affecting the Fe-Fe exchange interaction.³⁰ Thus, the ordering temperatures observed for these alloys (from about 275 to 450 K) are as would be expected for phase-separated systems.

B. Giant coercivities

The magnetic hardness of these materials is perhaps their most interesting property because of possible

development as permanent magnets. Croat *et al.*^{18,20} found a coercivity of 62 kOe for $\text{Pr}_{45}\text{Fe}_{55}$ at 20 K, compared to our result of ≈ 63 kOe for $(\text{Pr}_{80}\text{Ga}_{20})_{70}\text{Fe}_{30}$ at about 100 K. They also find $H_c \approx 52$ kOe at 20 K for $\text{Nd}_{50}\text{Fe}_{50}$, whereas we find 44 kOe at 90 K for $(\text{Nd}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$. As far as we are aware, a coercive field of 63 kOe at 100 K is the largest H_c reported for any metallic glass at 100 K (and perhaps at any temperature).

In order to find the Fe contribution to the reversible magnetization in the $x=20$ glass, we use results of the Mössbauer measurements and hyperfine-field distributions. Taking the center of the magnetic peak as H_{int} and using the Gubbens formula, we obtain $\bar{\mu}_{\text{eff}} \approx 1.6\mu_B$ (300 K) and $2\mu_B$ (4.2 K). Since $\approx 65\%$ of the atoms contribute [as measured from the relative areas of the nonmagnetic to the magnetic portion of $P(H)$], this leads to magnetization values of 13.8 and 11 emu/g at 4.2 and 300 K, respectively, as shown in Fig. 4 by the dashed line drawn between these points. Since the measured spontaneous magnetizations at different temperatures lie along this line, this clearly suggests that it is Fe alone that contributes to reversible magnetization above ≈ 100 K. Similar results were obtained for all other alloys in which Mössbauer measurements have been made. To explain the very large anisotropy field that the Fe spins must be experiencing in order to produce such giant coercivities, it is possible that the Fe spins couple to the lattice through exchange anisotropy with the speromagnetic Pr subnetwork.³¹

The theory of Paul²¹ as discussed earlier, although designed to consider domain-wall motion in bulk materials, might be applicable to some regions of these materials (Fe rich). Figure 12 shows the inverse high-field susceptibility plotted against temperature for some glasses. These plots suggest that there is a paramagnetic contribution to the high-field magnetization since a Curie-Weiss-like behavior,

$$1/\chi_H \approx C^{-1}(T - \Theta), \quad (3)$$

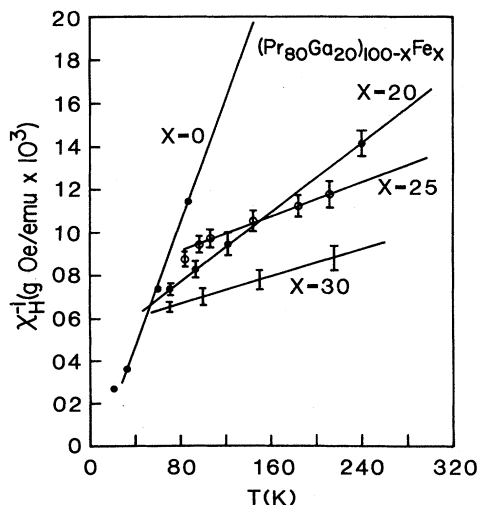


FIG. 12. Inverse high-field susceptibility for selected Pr glasses.

is observed. This suggests that significant regions within these may be paramagnetic, possibly with compositions near that of paramagnetic $\text{Pr}_{80}\text{Ga}_{20}$. This has been observed in other materials, in some Alnico magnets, for instance, and some microscopic regions become paramagnetic as the temperature is raised, leading to increased magnetic hardness.³² Paul has found, in the limit that domain walls impinge on nonmagnetic "inclusions," that

$$H_c \approx 2K/M_0, \quad (4)$$

and the theoretical limit for coherent rotations of spins is reached.³³ Evaluated for $x=20$ in this limit, with $M_0 \approx 300 \text{ emu/cm}^3$ for magnetic regions (assuming the magnetic regions to have the composition $\text{Pr}_{50}\text{Fe}_{50}$) and $K \approx 1.5 \times 10^7 \text{ erg/cm}^3$ (this value was obtained from $\text{Pr}_{45}\text{Fe}_{55}$ of Ref. 17 by the magnetization-area method of Sec. III of paper I), the value obtained is $H_c \approx 100 \text{ kOe}$. This is close to the 90 kOe extrapolated from plotting experimental $H_c^{1/2}$ vs T .

The theory of Callen *et al.*³⁴ (discussed in paper I) treats the magnetic properties of "microdomain" systems. Stoner and Wohlfarth studied a system of noninteracting single-domain particles experiencing random uniaxial anisotropy barriers.³² Callen *et al.* extend this theory by including an exchange-enhanced effective field to allow for interactions between single-domain particles at $T=0$. Figure 11 of paper I shows some predictions for this theory. For a fairly large reduced remanence, ≈ 0.9 (as is the case for the $x > 10$ materials above 100 K), the theory requires a small d (d is the ratio of anisotropy to ex-

change). For $d \leq 1$ the coercive field should be very small. If the theory of Gaunt²² is applied, which extended the microdomain model of Callen *et al.* to finite temperatures, a prediction of H_c vs T can be made. Furthermore, d is expected to decrease as T is increased, which would allow for reduced remanences greater than 0.5. Figure 13 shows the results of fitting experimental data to the theory (see Ref. 22 and references therein). In the region above the peak in H_c the fits are fairly good. For the $x=25$ glass the quantity $KV=1.5 \times 10^{-12} \text{ erg}$, where K is the uniaxial anisotropy and V is the reversal volume and $H_c(T=0) \approx 150 \text{ kOe}$. If M_0 is reasonably taken to be 300 emu/cm^3 , then, upon taking the coherent rotation limit, the minimum K expected is

$$K_{\min} = 0.5(300 \text{ emu/cm}^3)150 \text{ kOe} = 2.25 \times 10^7 \text{ erg/cm}^3. \quad (5)$$

For randomly oriented single-domain particles this gives a volume of 67000 \AA^3 (a volume of about 10 atoms on a side). This value is larger than previously reported values, but the fits are quite good. These fits tend to suggest that the thermal activation of interacting single-domain particles over spin-reversal barriers dominates the temperature dependence of H_c .

C. Anomalies in hysteresis loops

Upward shifts of the magnetization loops after field cooling were observed on all of the extremely hysteretic materials. Figure 4 shows the temperature dependence of the shift for $(\text{Pr}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$. These loops were also characterized by not closing in high fields. In materials such as these a distribution of anisotropy fields may be expected. As T is lowered the distribution will shift to higher values and a portion of the distribution may move beyond the available reversing magnetic field (in our case, $\approx 80 \text{ kOe}$). At this point the loops will become "open" and the value of the total reversible magnetization will begin to decrease as some spins "freeze out." If the process continues, all of the spins may freeze out of the reversible magnetization process and no measurement would be possible (i.e., $M=0$). If a field is applied at high temperatures, a preferred alignment of spins may "freeze in" as T is lowered and obviously give rise to an irreversible component of M . This will cause a shift of the magnetization, μ_{sh} , in the direction of the applied field. Thus, as T is lowered below T_F (the spin-freezing temperature) the magnetization loops become narrower and at the same time μ_{sh} will increase. Thus the following relation holds for nearly square loops:

$$\frac{1}{2} \Delta M_R + \mu_{\text{sh}} = M_0, \quad (6)$$

where ΔM_R is the width of the magnetization loop as measured on the magnetization axis as $H=0$. The mechanism by which Fe atoms are frozen in orientation could be exchange anisotropy since there is no reason in these materials to assume that Fe atoms can directly spin-orbit couple to the lattice. It has been suggested, however, that even for ions with a "quenched" orbital angular momentum it may be possible for the spin-orbit coupling to mix an orbital term into the ground state.³⁴ The peak in H_c vs

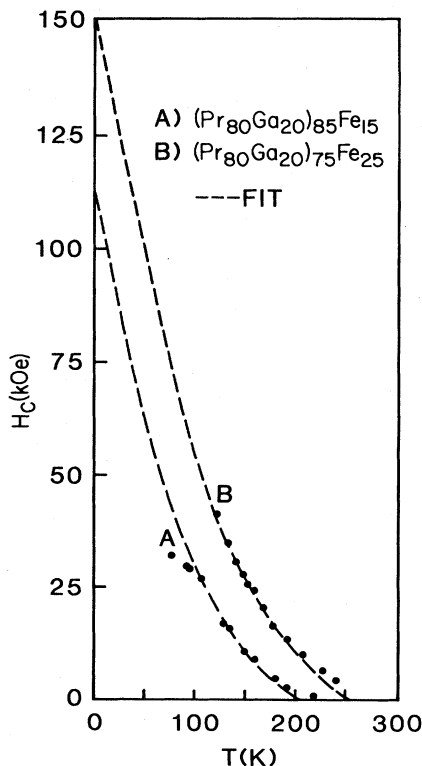


FIG. 13. Fits to Gaunt's theory for H_c -vs- T data on selected Pr glasses.

T follows from the above discussion. In a homogeneous single-phase material the magnetization loop would eventually approach $M(H)=\text{const}$ if the above situation occurred. This would just be the situation where all the spins were completely frozen so an application of a field could not reverse any spin. Thus the traversed loop would be a straight line at M_0 , the value of the frozen-in spontaneous moment. If only *some* of the spins are completely frozen in orientation for a square-loop material, the loop height would be reduced but the measured coercive force would be unchanged (for this simple model there are only two possible anisotropy fields—one is infinite for *frozen* spins and the other causes the coercive-field “jump” at the vertical portion of the square loops). If, however, a portion of the sample is *paramagnetic* with susceptibility χ , then the following relation holds:

$$M = M(H) + \chi H, \quad (7)$$

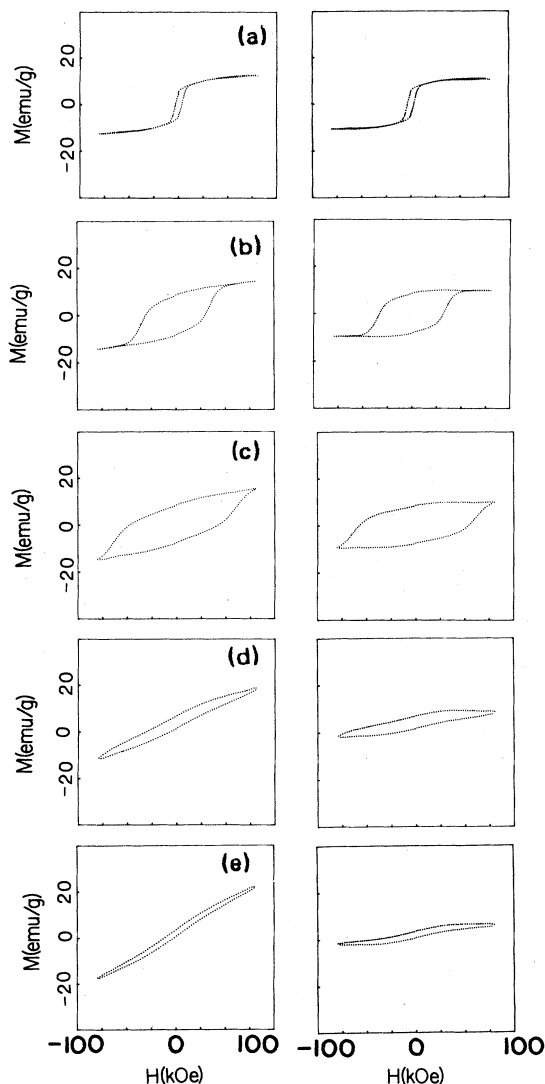


FIG. 14. Results of subtracting paramagnetic susceptibility from magnetization loops for a Pr-based glass. The temperatures of the loops are (a) $T=235$ K, (b) $T=117$ K, (c) $T=76$ K, (d) $T=52$ K, and (e) $T=33$ K.

which would give the square loop a slope χ (but not the vertical “jumps” at H_c). If $\chi H_{\text{max}} \geq M_R$, for *unshifted square loops*, H_c will be reduced simply because the loop “tilted” to below $M=0$ at H_c . In the materials studied here, the spontaneous moment is small for $x=20$ with $M_0 \approx 10$ emu/g. An estimate of the amount of paramagnetic Pr from low-temperature measurements and paramagnetic Fe from Mössbauer results gives $\chi(80 \text{ K}) \approx 9 \times 10^{-5}$ emu/gOe. Thus, paramagnetism can contribute ≈ 7 emu/g at 80 kOe (at 80 K) and larger values as T is lowered. Figure 14 shows the results of subtracting the paramagnetic contribution (constrained to Curie-Weiss behavior) from various high-field-magnetization loops. The final loops thus give a measure of the magnetically hard portions of the sample. In this case H_c need *not* turn over below ≈ 100 K. A monotonic increase (at least to 80 kOe) is seen in H_c for the loops which have the paramagnetic contribution subtracted. Here, only a *linear* susceptibility has been subtracted. Obviously, a Brillouin function more appropriately characterizes χ at lower temperatures and would likely give rise to some of the observed *curvature* in $M(H)$. Croat *et al.*^{18,20} have found a similar turnover of H_c in $\text{Nd}_{55}\text{Fe}_{45}$ and have attributed it to a variation of the anisotropy of Nd as a function of T . While this explanation certainly is possible, especially for light rare earths, the above explanation should be considered. It should be noted that the peak in the coercivity is an intrinsic property of the two-phase nature of these materials if the paramagnetic contribution to $M(H)$ is sufficiently large. Also, the magnetic hardness of the Fe-rich phase may be reduced or destroyed if the paramagnetic phase were not present.

IV. SUMMARY AND CONCLUSIONS

Perhaps the most general feature of this group of materials is the apparent presence of chemical short-range order. All of the magnetic alloys show evidence of this in their initial susceptibilities by the presence of two well-defined magnetic transitions at widely different temperatures. An x-ray analysis in terms of pair distribution functions suggests significant chemical short-range order and a phase separation is shown by electron microscopy on $(\text{Pr}_{80}\text{Ga}_{20})_{80}\text{Fe}_{20}$. The presence of Ga does not seem to be required since its replacement by Au resulted in slight change in the magnetic properties. The presence of Fe, however, is critical. If Fe is replaced by Ni or Co or other transition metals the high-temperature magnetic phase disappears, leading to paramagnetism at room temperature. The available evidence seems to suggest, therefore, that it is a general property of these amorphous alloys of light rare earths and Fe, for $10 \leq x \leq 30$, to form phase-separated systems which are segregated into Fe-rich and Fe-deficient regions.

The magnetic properties of the anisotropic light rare earths containing Fe are dominated by the presence of at least two magnetic phases. Recently, there have been several publications which predict that giant intrinsic magnetic hardness may be due to the heterogeneous nature of the microscopic magnetic and structural properties of disordered materials. These theories predict, for instance, strong domain-wall pinning at grain boundaries or

phase interfaces,²¹ or the increase of energy barriers to coherent rotation.²² The evidence presented above suggests that the glasses under study here consist of a mixture of a magnetic phase and a paramagnetic one. Within the theory of Paul, domain walls may be pinned to their coherent rotation limit at a magnetic-nonmagnetic boundary. This is consistent with the expected value obtained by an extrapolation of H_c data to $T=0$, which was about 150 kOe for $(\text{Pr}_{80}\text{Ga}_{20})_{75}\text{Fe}_{25}$. However, approximately this same limit would be reached naturally for noninteracting single-domain particles; thus the distinction between "domain-wall pinning" and "coherent rotation processes" seems to be lost. The excellent fits to the temperature dependence of H_c assuming activation of single-domain particles over anisotropy barriers support a microdomain model such as that of Callen *et al.*³⁴ The observed remanence ratio, however, suggests strong microdomain interactions and should, within the theory of Callen *et al.* give weak or zero coercive fields. This is definitely *not* observed. The extension of this theory by Gaunt to finite temperatures allows for larger reduced remanence and good fits of H_c vs T to experimental data are obtained. However, the two-phase nature of these alloys (leading to large H_c values) and the unavailability of a very large magnetic field did not permit fits to be made to data below 100 K. It would be interesting to extend

these measurements to above 150 kOe since very large coercive fields may be obtained in some cases. Whatever the exact mechanism for spin reversal is, it is likely that a significant part of the volume of these materials are boundary regions separating paramagnetic and magnetically ordered regions. Thus, these materials may be representative of a class of phase-separated metallic glasses in which the magnetic heterogeneity is enhanced because of the widely differing magnetic character of the different regions. It seems likely that, if M_0 could be increased within the framework of these "enhanced-fluctuation materials," good permanent magnet materials containing Fe might be developed with coercive fields approaching the Stoner-Wohlfarth coherent rotation limit of $H_c=2K/M_0$ at low temperatures. Work along these lines has already shown great promise.³⁵

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