Anisotropic rare-earth spin-glasses

A. Fert, P. Pureur, and F. Hippert

Laboratoire de Physique des Solides, Université de Paris-Sud, F-91405 Orsay, France

K. Baberschke and F. Bruss*

Institut für Atom and Festkörperphysik, Freie Universität Berlin, Boltzmannstrasse 20, D-1000 Berlin 33, Germany

We present magnetic measurements (ac and dc susceptibilities and magnetization) on YEr and YGd single crystals. The longitudinal (i.e., along the c axis) susceptibility of YEr shows a typical spin-glass-like cusp, whereas no cusp is observed in the transverse directions. This suggests that only the longitudinal spin components are frozen, in agreement with the theoretical predictions of Cragg and Sherrington for a spin-glass with a large uniaxial crystal field. The YEr system appears to represent an almost perfect Ising type of spin-glass. Surprisingly the YGd system is also definitely anisotropic. A spin-glass-like cusp is observed only for the transverse susceptibility. This anisotropy cannot be ascribed to the crystal field and we discuss its possible origins. Finally, we present some data on the remanence properties of the YEr and YGd alloys.

I. INTRODUCTION

The properties of the spin-glass state have been extensively studied in cubic systems and rarely in crystals of uniaxial symmetry. However, studies in uniaxial spin-glass systems should be of great interest because, depending on the sign and the magnitude of the uniaxial crystal field acting on the magnetic moments {i.e., $H_{CF} = B_{20}O_{20} \equiv B_{20}[3J_z^2 - J \cdot (J+1)]$} these systems can be *Ising-like* (for B_{20} negative and much larger than kT_g), *XY-like* (for B_{20} positive and much larger than kT_g) or *Heisenberg-like* (for B_{20} much smaller than kT_g).

Some theoretical aspects of these situations have been developed by Ghatak and Sherrington[1] a few years ago. More recently, Cragg and Sheerington[2] have extended the Sherrington-Kirkpatrick (SK) model[3] of spin-glasses to the case of a system of spins in a uniaxial crystal field. In particular, this theory predicts spin-glass ordering for the longitudinal (i.e., parallel to the crystal-field axis) spin components and not for the transverse ones when the crystal-field constant B_{20} is negative and large enough with respect to the exchange coupling.

Few experimental studies of uniaxial spin-glasses performed on single crystals exist. There have been investigations of $(Ti_{1-x}V_x)_2O_3$ (Ref. 4) and $(Ni_{1-x}Fe_x)_{1-\delta}S$ (Ref. 5) single crystals revealing spin-glass-like maxima of the susceptibility only in certain lattice directions or different maxima in different lattice directions. However, these spin-glass systems are not very simple, the origin of the magnetic interactions is not well known in $(Ti_{1-x}V_x)_2O_3$, while in $(Ni_{1-x}Fe_x)_{1-\delta}S$ it is supposed that the magnetic moments are developed on pairs of iron impurities. Recently, Albrecht *et al.*[6] investigated the mag-

netic properties of ZnMn, CdMn, and MgMn single crystals. ZnMn shows an Ising-like behavior with a spin-glass maximum of the susceptibility only for longitudinal fields, CdMn shows a XY-like behavior and MgMn present isotropic properties. In this paper we report on an investigation of the spin-glass properties of YEr and YGd single crystals.[7] Up to now the magnetic properties of yttrium—rare-earth alloys have been investigated only in polycrystals.[8-10] A spin-glass behavior has been identified at low concentrations and a transition to helical ordering has been suggested at higher concentrations (above 2.8 at.% in YGd, above 5 at.% in YTb).[9]

II. YEr ALLOYS

We first focus on the YEr system. Its crystal-field parameters have been determined by Touborg[11] from measurements of the paramagnetic susceptibility of very dilute alloys and neutron scattering experiments. They are

$$B_{20} = -2.8 \times 10^{-1} \text{ K}, \quad B_{40} = 6.0 \times 10^{-4} \text{ K} ,$$

$$B_{60} = 2.48 \times 10^{-5} \text{ K}, \quad B_{66} = 2.76 \times 10^{-4} \text{ K} .$$

The coefficients B_{40}, B_{60}, and B_{66} are much smaller than B_{20}. If they could be neglected, the ground state would be the doublet $|\frac{15}{2}, \pm\frac{15}{2}\rangle$ and the system would be Ising-like in the low-temperature limit. When the coefficients B_{40}, B_{60}, and B_{66} are taken into account, the actual ground state corresponds to the following combination[11]:

$$0.966|\tfrac{15}{2}, \pm\tfrac{13}{2}\rangle - 0.228|\tfrac{15}{2}, \pm\tfrac{1}{2}\rangle + 0.122|\tfrac{15}{2}, \mp\tfrac{11}{2}\rangle .$$

This ground state is predominantly a $|\frac{15}{2}, \pm\frac{13}{2}\rangle$ state with a small admixture of $|\frac{15}{2}, \pm\frac{1}{2}\rangle$ and $|\frac{15}{2}, \pm\frac{11}{2}\rangle$ states. It lies at 27 K below the lowest of the other eigenstates and the overall splitting by crystal fields amounts to 123 K. In spite of the complications arising from the fourth- and sixth-order terms, the one-ion behavior observed in dilute alloys is not very different from what it would be with only quadratic terms, i.e., the one-ion susceptibility at low temperatures is much larger in longitudinal fields ($\vec{H}$ along c) than in transverse fields ($\vec{H}$ in the basal plane).[11] Equally, the transverse susceptibility is almost isotropic when the magnetic field rotates in the basal plane.[11] Thus we believe that it is reasonable, in first approximation, to forget the effects of the fourth- and sixth-order terms and to consider that the YEr system can be used to test the predictions of the theoretical models of spin-glasses with uniaxial quadratic anisotropy. Because the ground-state isolation is relatively large ($\sim$27 K), we can also anticipate that the YEr system is probably not very far from the Ising limit in the temperature range of the spin-glass
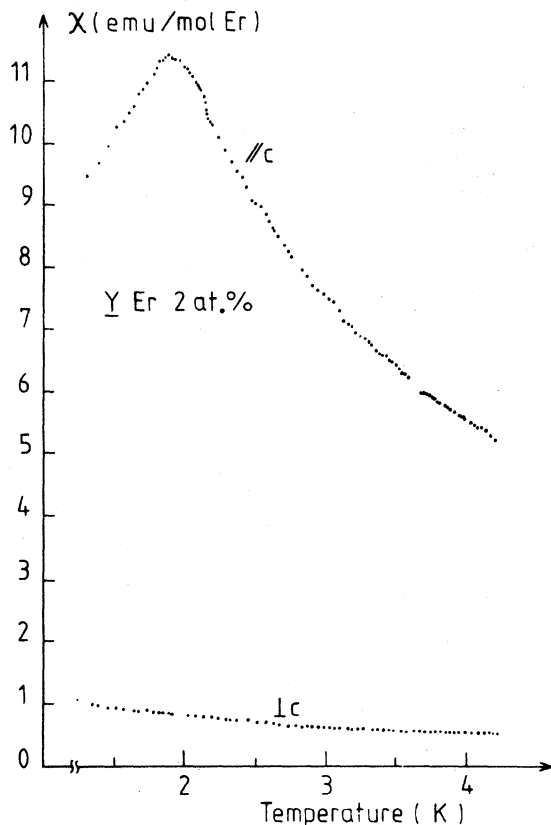

FIG. 1. ac magnetic susceptibility of YEr 2 at.% for fields along the c axis and along the b axis (measurements along the a axis give the same results as along the b axis). The susceptibility of pure yttrium has been subtracted. The measurements have been performed in 1 G at 120 Hz.
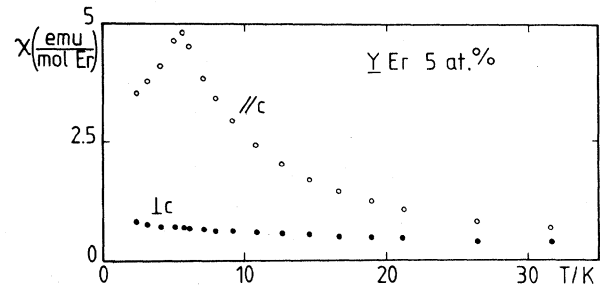

FIG. 2. dc magnetic susceptibility of YEr 5 at.% measured with a vibrating sample magnetometer in $H = 100$ G parallel or perpendicular to the c axis.

state for moderate concentrations ($T_g \sim 1$ K/at.%).

We show in Figs. 1 and 2 the longitudinal and transverse susceptibilities of two YEr alloys as a function of temperature (the longitudinal or transverse susceptibilities of pure Y have been subtracted). The longitudinal susceptibility shows a spin-glass typical cusp at $T_g = 1.9$ K for $c = 2$ at.% and $T_g = 5.6$ K for $c = 5$ at.%. In contrast, the transverse susceptibility increases continuously when the temperature decreases and there is no evidence of any event at T_g. As mentioned in our introduction, recent calculations[2] predict spin-glass ordering only for the longitudinal-spin components when the crystal-field coefficient B_{20} is negative and large enough with respect to the exchange interaction, while the transverse Edwards-Anderson parameter would remain zero down to $T = 0$. Our experimental results, with a susceptibility cusp in longitudinal fields but not in transverse fields, should correspond to this situation. A similar behavior has been observed by Albrecht et al.[6] in ZnMn alloys and by Roux-Buisson and Coey[5] in some $(Ni_{1-x}Fe_x)_{1-\delta}S$ alloys.

In longitudinal fields the YEr alloys exhibit the typical remanence properties of spin-glasses. We show in Fig. 3 the thermo-remanent and irreversible remanent magnetization (TRM and IRM) curves of
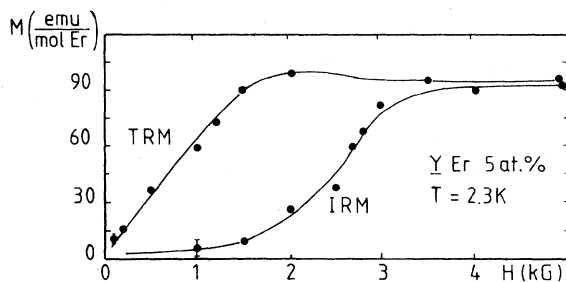

FIG. 3. Thermoremanent and irreversible remanent magnetization for a YEr 5 at.% crystal at $T = 2.3$ K ($T/T_g = 0.4$) and for fields along the c axis. The solid lines are guides for the eyes through the experimental points.

YEr 5 at.% These curves look like those observed in classical spin-glasses such as CuMn or AuFe. However the magnitude of the remanent magnetization with respect to the reversible magnetization in the same fields is smaller than in CuMn or AuFe. We could not observe any remanence effect in transverse fields.

III. YGd SYSTEM

The crystal field of the YGd system, in contrast to that of YEr, is very small. The coefficient B_{20}, derived from EPR data on dilute alloys by Weimann and Elschner,[12] is

$$B_{20} = -7.22 \times 10^{-3} \text{ K} \ .$$

This gives an overall crystal-field splitting of 0.26 K. The effects of such a small crystal field on the spin-glass properties can be anticipated to be negligible, at least in our alloys which have a freezing temperature higher than 2.1 K. It can be noted, for example, that the crystal effects are negligible in the MgMn alloys investigated by Albrecht et al.,[6] although the freezing temperature of these alloys is larger than their overall splitting by only a factor of 2. However, our susceptibility measurements on YGd single crystals show striking anisotropy effects. The susceptibility of YGd 1 at.% (Fig. 4) and 2.3 at.% (Fig. 5) show a typical spin-glass cusp for $\chi_\perp$ at $T = 2.1$ and 7.0 K, respec-
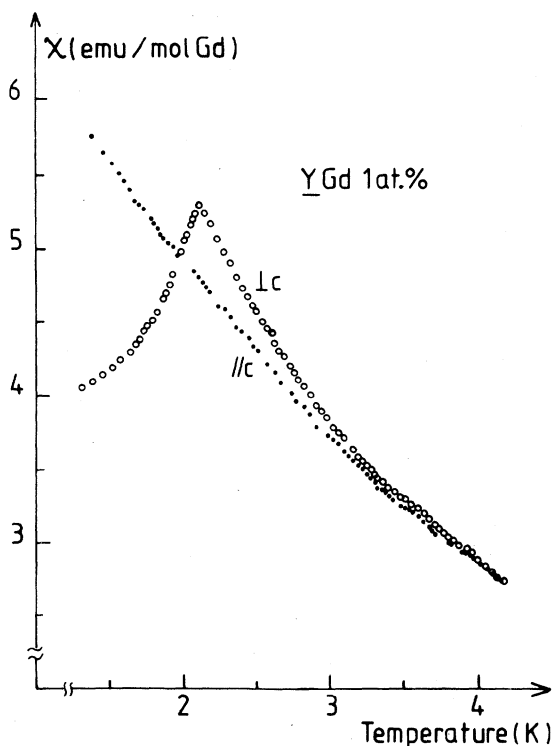
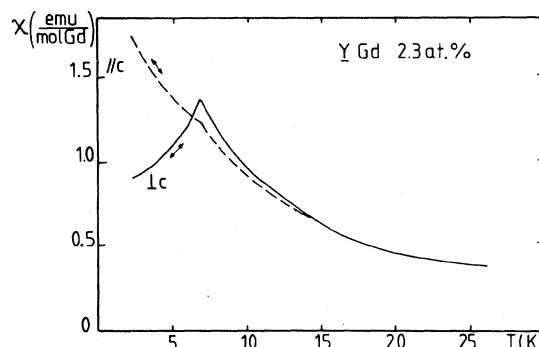FIG. 5. dc susceptibility of YGd 2.3 at.% in $H = 100$ G. As indicated by the arrows, there is no significant difference between FC and ZFC measurements. The small cusp in $\chi_\parallel$, which is not observed for YGd 1 at.%, could be due to a small misorientation of the crystal (admixture of a small transverse component).

tively. In contrast the longitudinal susceptibility monotonously increases for decreasing temperature. The susceptibility is isotropic at high temperatures (at least after subtracting the susceptibility of pure yttrium). When the temperature decreases, $\chi_\perp$ increases more rapidly than $\chi_\parallel$. Then, below T_g, $\chi_\perp$ decreases and becomes smaller than $\chi_\parallel$. These results are very striking. The anisotropy cannot be ascribed to the crystal field, first because the overall crystal-field splitting is very small, and then because, anyway, the negative sign of B_{20} would lead to a larger longitudinal susceptibility above T_g. The concentration dependence of the anisotropy is also inconsistent with a crystal-field effect. Therefore, the anisotropy of the YGd alloys must be ascribed to another sort of spin-lattice coupling. A first possibility is a coupling of the Gd moments with spin-density waves in a conduction band with a significant spin-orbit coupling (a coupling with spin-density waves has been already proposed to account for a transition from spin-glass ordering to helical ordering at higher concentration[8,9]). Another explanation could be the existence of anisotropic interactions between the Gd moments. More specifically the large spin-orbit coupling of the $5d$ outer electrons of Gd should give rise to significant Dzyaloshinsky-Moriya—type interactions[13] and, consequently, to a concentration-dependent pseudo-crystal field.

Another definite difference between the YEr and YGd alloys concerns the remanence properties. The remanent magnetization of YGd is much smaller than that of YEr. As shown in Fig. 5 the dc susceptibility (magnetization at 100 G) is, within our experimental uncertainty, almost reversible and independent of field cooling (FC) or or zero field cooling (ZFC). We could not draw accurately TRM and IRM curves such as those of Fig. 3 for YEr. It turns out the yttrium-rare-earth system definitely departs from

FIG. 4. Same caption as Fig. 1 but for YGd 1 at.%.

the classical transition-metal impurity spin-glasses. In the latter the remanence properties appear to be independent of the anisotropy properties; i.e., they are similar for alloys with weak spin-lattice couplings (CuMn,AgMn) and for alloys with strong spin-lattice couplings (AuFe). In the yttrium-rare-earth system, the remanence properties seem to be associated with the existence of strong spin-lattice couplings (YEr).

IV. CONCLUSIONS

YEr: The existence of a cusp in the longitudinal susceptibility and not in the transverse seems to be in agreement with the calculations of Cragg and Sherrington, predicting spin-glass ordering only for longitudinal spin components in the presence of a strong enough uniaxial crystal field. The YEr should be close to an Ising spin-glass system. On the other hand, YTb or YDy, for which the crystal-field coefficient B_{20} is positive, should be close to the XY-like

spin-glass systems. Recent measurements have confirmed that YTb shows a maximum of susceptibility only for transverse field.[14]

YGd: The properties of YGd exhibit a surprising anisotropy which cannot be ascribed to the crystal field (too small and having the wrong sign). The anisotropic spin-glass properties of YGd could be due to a coupling with spin-density waves in the conduction band of Y or to anisotropic interactions between the Gd moments. We also found that the YGd alloys present very weak remanence effects.

Note added in proof. Theoretical results of Ref. 2 have also been found by S. A. Roberts and A. J. Bray, J. Phys. C **15**, L527 (1982).

*Now at Schoeller Elektronik D-3552 Wetter 1, Germany.

[1]S. K. Ghatak and D. Sherrington, J. Phys. C **10**, 3149 (1977).

[2]D. M. Cragg and D. Sherrington (unpublished).

[3]D. Sherrington and S. Kirkpatrick, Phys. Rev. Lett. **35**, 1792 (1975).

[4]J. Dumas, C. Schlenker, J. L. Tholence, and R. Tournier, Phys. Rev. B **20**, 3913 (1979).

[5]H. Roux-Buisson and J. M. D. Coey, J. Phys. F **10**, 275 (1980).

[6]H. Albrecht, E. F. Wassermann, F. T. Hedgcock, and P. Monod, Phys. Rev. Lett. **48**, 819 (1982).

[7]Part of the results have been communicated: F. Bruss and

K. Baberschke, Verh. Dtsch. Phys. Ges. **6**, 186 (1979). We would like to thank Dr. Ghatak for discussions in the early state of the experiments.

[8]B. V. B. Sarkissian and B. R. Coles, Commun. Phys. **1**, 17 (1976).

[9]B. V. B. Sarkissian, J. Phys. F **7**, L139 (1977).

[10]S. P. McAlister, J. Appl. Phys. **52**, 1769 (1981).

[11]P. Touborg, Phys. Rev. B **16**, 1201 (1977); P. Touborg, thesis (Odense University, 1979) (unpublished).

[12]G. Weimann and B. Elschner, Z. Phys. **261**, 85 (1973).

[13]A. Fert and P. M. Levy, Phys. Rev. Lett. **44**, 1538 (1981).

[14]P. Pureur and F. Hippert (private communication).