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Magnetic Structure of an Amorphous Rare-Earth Transition-Metal Alloy

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^{161}Dy Mössbauer spectra show that the dysprosium moments in amorphous $\mathrm{DyCo}_{3.4}$ are strongly coupled to the local crystal field axes. The magnetic structure is one in which the cobalt "sublattice" is strongly ferromagnetic, and the dysprosium moments are almost randomly distributed in directions between antiparallel and perpendicular to the cobalt.

Although ferromagnetism in amorphous alloys is well known,[1] the rich variety of more complex magnetic structures which can occur in amorphous solids is just now becoming apparent. Amorphous rare-earth transition-metal alloys in particular are much studied at present.[2-6] The magnetic structures in these alloys will be strongly influenced by the local anisotropy field at the rare-earth sites, just as in their crystalline counterparts. Harris, Plischke, and Zuckermann[7] have suggested that this field, determined by the spatial configuration of the neighboring atoms, will be random in direction and comparable in magnitude to the exchange field experienced by the rare-earth ions. In this Letter we present direct evidence that the directions of the rare-earth moments are closely correlated with the directions of the local anisotropy field in amorphous $\mathrm{DyCo}_{3.4}$. The magnetic structure [Fig. 3(b)] is deduced from Mössbauer spectra and magnetization measurements. Finally we outline a classification scheme for magnetic order in

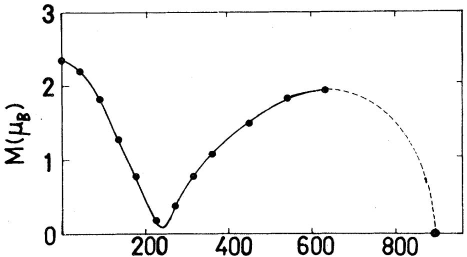

FIG. 1. Temperature dependence of the spontaneous magnetization (i.e., the resultant moment) of amorphous $DyCo_3$.

amorphous solids.

Amorphous films of composition $Dy_{23}Co_{77}$ and $Dy_{23}Co_{74}Fe_3$ were prepared by dc sputtering. Their magnetization could be saturated easily at 4.2 K if the magnetic field was applied in the plane of the film but the perpendicular direction was much harder and required a field ~ 10 kOe for saturation.

The temperature dependence of the moment per $DyCo_{3.4}$ unit is shown in Fig. 1. The Curie temperature, roughly 900 K, is double that of crystalline $DyCo_3$,[8] but it is difficult to measure exactly, because of crystallization. There is a characteristic minimum in the moment at the compensation temperature, 230 K. We will distinguish the *resultant* moment of a domain from the *net* moment of the cobalt or dysprosium "sublattices." The moment actually measured is the sum of the resultant moments of the domains. Upon saturation it gives the resultant moment directly. The near disappearance of the resultant moment at 230 K is explained by the cancelation of the net moments of the two sublattices. The net moment of a sublattice in turn depends on the magnitudes and directions of the individual atomic moments. Such information cannot be obtained from magnetization measurements, but may be deduced from Mössbauer spectra.[9]

The cobalt sublattice in $Dy(Co_{0.96}Fe_{0.4})_{3.4}$ was tagged with pure ^{57}Fe, whose moments are expected to couple strongly with the cobalt, thus giving information about the direction, but not the magnitudes of the cobalt moments. The spectra were well defined even at 300 K, like the one in Fig. 2(a). Average hyperfine parameters were derived from least-squares fits to the data. They are listed in Table I, together with the full width at half-maximum of their distributions. The fraction I_0 of the total absorbtion in the $\Delta m = 0$ transitions is also given: $I_0 = \frac{1}{2}\langle \sin^2\theta \rangle$, where θ is the angle between the atomic moments and the γ direction, usually arranged to be perpendicular to the film. In that case $\langle \sin^2\theta \rangle = 0.98 \pm 0.05$, which indicates that all the iron moments lie in the plane of the film. Moreover, the moments were shown to be ferromagnetically coupled within the plane by saturating the magnetization in the plane and taking spectra with several different angles between the γ direction and the applied field.

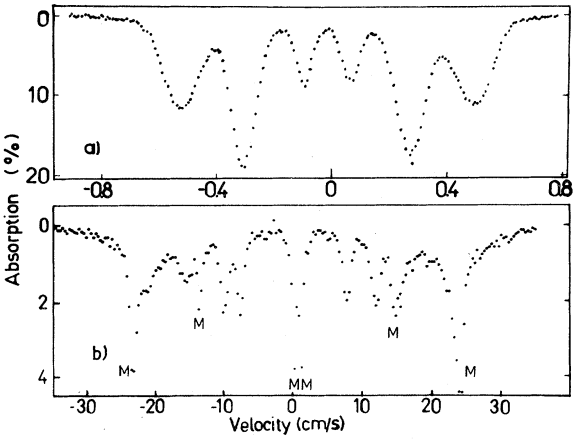

FIG. 2. (a) ^{57}Fe Mössbauer spectra of amorphous $Dy(Co_{0.96}Fe_{0.4})_{3.4}$. (b) ^{161}Dy Mössbauer spectra of amorphous $DyCo_{3.4}$.

The ^{161}Dy spectra [Fig. 2(b)] give a quite different result for the rare-earth sublattice. Well-defined spectra were observed only near liquid-helium temperature. At higher temperatures, the dysprosium moment falls off rapidly and inhomogeneously due to the weak, inhomogeneous exchange coupling with the cobalt. However, at 4.2 K, the hyperfine field is typical of dysprosi-

TABLE I. Hyperfine interactions in amorphous $DyCo_3$ at 4.2 K. Distribution widths are given in parentheses.

Composition	H_{hf} (MOe)	e^2qQ (cm/sec)	I_0
$^{161}DyCo_{3.4}$	5.91 (0.06)	10.0 (1.9)	0.49 ± 0.02
$Dy(Co_{0.96}{}^{57}Fe_{0.4})_{3.4}$	0.312 (0.028)	0.001 (0.164)	0.37 ± 0.04

um in a $|\frac{15}{2}\rangle$ ground state, and its relative distribution is only 1%. The spectrum resembles that of a powder sample of amorphous $\mathrm{DyFe_2}$, reported recently by Forester *et al.*[4] It differs from that of crystalline $\mathrm{DyCo_3}$[10] in that the hyperfine field is marginally smaller, and that the lines whose positions depend on the quadrupole interaction (lines not marked "*M*" on the figure) are much broader. The fit gives $\langle \sin^2\theta\rangle = 0.74 \pm 0.08$, almost what would be expected if the dysprosium moments were distributed over all directions with equal probability (0.67).

The electric field gradient at the nucleus may be written in the usual form as

$$eq = eq_1(1-R) + eq_2(1-\gamma_\infty),$$

where the first term is due to the $4f$ shell, and the second is due to conduction electrons and distant charges. The first term gives a quadrupole interaction of 14.1 mm/sec for the $|\frac{15}{2}\rangle$ state. The total quadrupole interaction in amorphous $\mathrm{DyCo_{3.4}}$ is 10.0 cm/sec with a distribution whose full width at half-maximum was found to be 1.9 cm/sec from the widths of the lines not marked "*M*." In crystalline $\mathrm{DyCo_3}$ the interaction is 12.4 cm/sec.[10] Point-charge calculations by Cochrane, Harris, and Plischke[11] for a random close-packed binary model structure showed that the dominant term in the crystal field is V_2^0, which defines a local symmetry axis. V_2^0 is proportional to the principal component of q_2 in this approximation, so that if the directions of the moment and the crystal field were uncorrelated, the second term would be distributed about zero. This *is* the case for the iron spectra; the first term, due to $3d$ electrons, is negligible and the second is distributed about zero with a width of 0.24 mm/sec. However, in the dysprosium spectra the contribution of the second term is even larger than in the crystalline alloy. Furthermore it is large compared with the distribution of the quadrupole interaction, which indicates that *the dysprosium moment directions are strongly correlated with the local symmetry directions.* The two directions should be almost perpendicular.

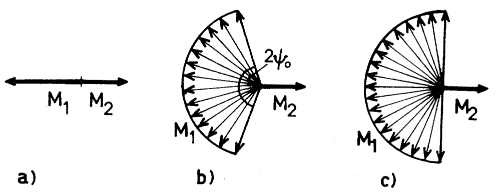

FIG. 3. Magnetic structures which result when the last term in Eq. (1) is dominant, and the ratio of the first to the fourth terms is (a) ≪1; (b) ~1 or (c) ≫1. 1 and 2 refer to the rare-earth and transition-metal sublattices. (a) is ferrimagnetic, (b) and (c) are sperimagnetic.

We now turn to the problem of the magnetic structure at 4 K. The following elements of the solution have been established: (i) antiparallel coupling of the net dysprosium and cobalt sublattice moments; (ii) the cobalt moments lie essentially in the plane of the film; (iii) the dysprosium moments are strongly correlated with the local crystal field axis, which varies randomly in direction; and (iv) they are distributed in direction so that $\langle \sin^2\theta\rangle = 0.74 \pm 0.08$. The magnetic structure illustrated in Fig. 3(b) fulfills all these requirements. In this solution, the dysprosium moments are uniformly distributed in the directions lying within a cone of half angle $\psi_0 = 70°$ whose axis is antiparallel to that of the collinear cobalt sublattice.

The net moment of the dysprosium sublattice is $M(\mathrm{Dy})\frac{1}{2}(1+\cos\psi_0)$; $M(\mathrm{Dy}) = 10\mu_B$ for the $|\frac{15}{2}\rangle$ state. The measured resultant moment is $2.3\mu_B$ per $\mathrm{DyCo_{3.4}}$ formula unit. Therefore the net moment of the cobalt is $(1.3 \pm 0.4)\mu_B$/atom. The errors correspond to those on $\langle \sin^2\theta\rangle$. For comparison, the cobalt moment is $(0.8 \pm 0.2)\mu_B$ in crystalline $\mathrm{DyCo_3}$[8] and $1.65\mu_B$ in amorphous $\mathrm{GdCo_{3.5}}$,[5] assuming a collinear structure [Fig. 3(a)].

The magnetic structure of amorphous $\mathrm{DyCo_{3.4}}$ may be understood by comparing the magnitudes of the terms in the Hamiltonian (cf. Ref. 7)

$$-\mathcal{H} = \sum_i V_{1_i} + \sum_j V_{2_j} + \sum_{i>i'} \mathcal{J}_{11}(r)\vec{J}_i\cdot\vec{J}_{i'} + \sum_{ij} \mathcal{J}_{12}(r)\vec{J}_i\cdot\vec{J}_j + \sum_{j>j'} \mathcal{J}_{22}(r)\vec{J}_j\cdot\vec{J}_{j'}, \tag{1}$$

where the first two terms represent the anisotropy and the last three the exchange. i and j run over the rare-earth (1) and transition-metal (2) sublattices, respectively. For cobalt, intrasublattice exchange is the dominant term. The zero average quadrupole interaction shows that $J_2\sum_j \mathcal{J}_{22}(r)J_j \gg V_2$, and a molecular-field fit to the magnetization curve gives

$$\textstyle\sum_j \mathcal{J}_{22}(r)J_j \sim 30\sum_i \mathcal{J}_{12}(r)J_i.$$

For dysprosium, anisotropy is dominant: V_1

$\sim 3J_1 \sum_j \mathcal{J}_{12}(r) J_j$.

The structures sketched in Fig. 3 will appear near $T=0$ when the last term of Eq. (1) dominates and the second and third terms can be neglected. (a) corresponds to $V_1 \simeq 0$ ($GdCo_3$), (b) to $DyCo_{3.4}$, and (c) to the extreme anisotropy limit where the only effect of exchange on the rare earth is to make it choose the easy direction most nearly antiparallel to the transition-metal sublattice. At higher temperatures the cone will close up because of the sharp decrease of the anisotropy $V_1 \propto J_1^3$. If the last term does not dominate the other two, the transition-metal sublattice will also have a noncollinear arrangement.

Finally we place the above structures in the context of other types of magnetic order known in amorphous solids. *Ferromagnetism* occurs in metals[1] and insulators[12] when the exchange is positive and its distribution is not too wide compared to its average value.[13] Collinear *antiferromagnetism* has not been positively identified in any homogeneous amorphous solid,[14] but *speromagnetic* ordering[15,13] may occur instead when the distribution of exchange is very broad or centered about a negative value. This is a structure where the moments each have a finite time average, but are randomly distributed in direction in such a way that the spin-spin correlations average to zero beyond at most a few interatomic distances. Speromagnetism may also occur where the anisotropy dominates the exchange, in pure amorphous rare-earth metals, for example.

Ferrimagnetism is possible when two sublattices can be defined chemically [Fig. 3(a)]. By analogy with the previous structures, those with the moments in one (or both) sublattices distributed randomly in direction according to a probability $P(\psi)$ may be termed *sperimagnetic*. The prefix means "scattered in direction."[15]

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