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Magnetic anisotropy of rare-earth—transition-metal compounds

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The exchange coupling between rare-earth (R) and transition-metal (T) moments in R_xT_y compounds is not so large as to hold the R and T moments rigidly parallel (for light rare-earth elements) or antiparallel (for heavy rare-earth elements); ionic magnetocrystalline anisotropy energy can be comparable to this intersublattice exchange. Anisotropy torques and/or an external field can induce canting between the sublattice moments. We show that this canting, even when small, can reduce the macroscopic effective anisotropy by a very large amount—more than an order of magnitude—from the intrinsic sublattice anisotropies. Consideration of the effects of canting reconcile hitherto conflicting reports, by spin-wave scattering and moment measurements, of the magnetic anisotropies of Ho_2Co_{17} and Ho_2Fe_{17}. One conclusion of our analysis is that since a huge degeneration of macroscopic magnetic anisotropy, and hence of the coercivity, comes from sublattice flexing, it seems to follow that to increase coercivity one should be trying to increase R-T exchange; the ionic magnetic anisotropy is already greater than its effective value.

I. INTRODUCTION

The lanthanide-rare earth (R)$-3d$ transition-metal (T) intermetallic compounds[1] comprise a dozen structures ranging from the rare-earth-rich R_3T to the transition-metal-rich $R\mathrm{Mn}_{12}$. These compounds are useful because they combine the large magnetic interaction and high Curie temperature of the transition metal with the large anisotropy and magnetostriction of rare-earth elements. The permanent magnets $SmCo_5$ and $Sm_2(CoFe)_{17}$, their complex derivatives,[2] and the magnetostrictive $TbFe_2$ (Ref. 3) come to mind.

Generally speaking, the exchange interaction between the transition-metal spins themselves (T-T) is large and ferromagnetic, the coupling of the transition-metal spin to the rare-earth spin (R-T) is much smaller, and the exchange coupling between the rare-earth elements (R-R) is so small as to be negligible. Since the magnetic moments of the light rare-earth elements ($g < 1$) are parallel to their spin angular momenta, while those of the heavy rare-earth elements are antiparallel to their spin angular momenta, a consequence of the negative RT exchange is that the light rare-earth moments align ferromagnetically with the T moments, while the heavy rare-earth moments align antiferromagnetically to the T moments. For crystal-field reasons, something more happens in the Sm compounds[4]; for example, the moment of $SmCo_5$ is less than that of YCo_5, while both $PrCo_5$ and $NdCo_5$ have larger moments than the yttrium compound.[5] We point this out here because Sm plays a unique role in permanent magnet formulations.

We shall discuss the interplay between the sublattice anisotropies, the intersublattice RT exchange, the sublattice moments, and the external field. A major conclusion of this paper will be that under common circumstances, the macroscopic magnetic anisotropy is very much less than the sum of the sublattice anisotropies. The origin of this is sublattice canting, which can cause the moment to point off axis, change its magnitude, and give it an unusual temperature dependence and a nonsaturating field dependence. Under these circumstances, the macroscopic magnetic anisotropy will not conform to any simple field or temperature dependence.

Some of these effects have already been reported. Rinaldi and Pareti[6] pointed out that in the RT compounds the intersublattice exchange is not strong enough to enforce complete alignment. Those authors focused on systems in which the two sublattices have magnetic anisotropies of different signs so that the intrinsic anisotropies induce canting of the sublattice moments. They show that this can point the net moment off at an unexpected angle and can induce large higher-order anisotropy constants as the two sublattices change their relative angle as the moment is rotated around by an external magnetic field. Lastly, Rinaldi and Pareti show that these circumstances can lead to the so-called first-order magnetization process,[7] a jump in the magnetization curve, as observed.[8]

Noncollinearity of sublattices is in itself not a novel concept.[9] We think that what is useful here is the application of this concept to a number of hitherto unexplained phenomena in the rare-earth—transition-metal compounds.[10] We hope that this understanding will assist in the formulation of better permanent magnets and magnetostrictive materials.

II. Ho_2Co_{17} AND Ho_2Fe_{17}

Our interest was piqued by the following history. In 1976, Miller, D'Silva, and Rodriquez[11] reported measurement of K_1, the energy to rotate the magnetization of Ho_2Co_{17} from the easy plane to the hard c axis. Ho_2Co_{17} is a uniaxial crystal and the free energy can be expanded in the magnetization angles θ from the c axis and ϕ in the basal plane:

$$\begin{aligned}F(\theta,\phi)=K_0(T)-K_1(T)\sin^2\theta\\ -K_2(T)\sin^4\theta-K_3(T)\sin^6\theta\\ -K_4(T)\sin^6\theta\cos6\phi\ .\end{aligned}\tag{2.1}$$

The negative signs in Eq. (2.1) have been chosen to make the plane easy with the K_i positive; this is not the customary convention, but is convenient here and avoids a lot of minus signs in later formulas.

The method by which Miller *et al.* determined K_1 (and other coefficients)—measuring the component of the magnetization M induced along the hard c axis by an external field in that direction—is a standard one. For simplicity let us ignore coefficients higher than K_2 and the effect of demagnetization (which produces an effective K_1). In a magnetic field H along the c axis, the free energy is then

$$F=K_0-K_1\sin^2\theta-K_2\sin^4\theta-MH\cos\theta\ .\tag{2.2}$$

Minimizing with respect to the angle of the magnetization, one finds the component of the magnetization along the field for small fields (and small deviations from the easy direction) to be

$$M\cos\theta\sim\frac{M^2H}{2\mathscr{K}}+\frac{M^4K_2}{4\mathscr{K}^4}H^3+\cdots\ ,\tag{2.3}$$

where the ellipsis represents terms of higher order. Here we have defined

$$\mathscr{K}\equiv K_1+2K_2\ ,\tag{2.4}$$

a combination which recurs. Implicit in this is the assumption that the magnetization in the plane is the same as when one tilts it up toward the c axis. We shall see that this is not right.

Three years later, at the 1979 International Conference on Magnetism in Munich, Clausen and Lebach[12] reported inelastic neutron scattering studies of spin-wave excitations in Ho_2Co_{17} and Ho_2Fe_{17}. Clausen and Lebach observed three spin-wave bands, one nondispersive and two with significant curvatures and with minima at Γ. Because of their simple structures, the three branches can be fitted with six constants—two each—describing their gaps and curvatures. The flat rare-earth branch means that the rare-earth—rare-earth exchange interaction is negligibly small, as is known from other evidence. From the curvatures of the other branches, Clausen finds T-T ferromagnetic exchange interactions which, using mean-field theory, are in satisfying agreement with the observed Curie temperatures of Ho_2Co_{17} and Ho_2Fe_{17}. In both compounds the R-T interaction is antiferromagnetic and much smaller than that between the transition-metal ions: $|J_{RT}|/J_{TT}\sim0.0064$ for Ho_2Co_{17}. (In the iron compound J_{TT} is smaller and anisotropic, but the ratio of $|J_{RT}|$ to J_{TT} is 0.02 or 0.03, three times larger than in the cobalt compound, but still small.) These results conform with normal expectation. For example, Steiner's[13] studies of $R_2(Co_{1-x}Fe_x)_{17}$ show the Curie temperatures of all the R_2Co_{17} compounds to be high, about 1200 K, and much the same for all rare-earth elements (hence $|J_{RT}|$ must be much less than J_{TT}), while in R_2Fe_{17} compounds, the Curie temperatures are about $\frac{1}{4}$ to $\frac{1}{3}$ those of the cobalt compounds and vary somewhat from one rare-earth element to another.

The observed gaps at Γ are related to the site anisotropies of the rare-earth element and transition metal. (Clausen emphasizes that although a band model would be more appropriate for the transition metal, it is simpler to perform the analysis in terms of an effective localized transition-metal moment with single-ion site anisotropy.) From the spin-wave gaps Clausen arrives at expectations of what the macroscopic anisotropies should be. His data appear to suggest that K_1 should be an order of magnitude larger than Miller *et al.*[11] had measured.

Miller *et al.* had done their moment measurements at 77 K; Clausen did his neutron diffraction studies at 4.2 K. To be sure that the large difference in K_1 was not due to the difference in temperature—implausible but possible—Clausen and Nielsen[14] repeated the moment measurements, but at 4.2 K. They again found results not too different from the 77 K magnetization results, and still an order of magnitude less than what the spin-wave gaps seemed to demand. Why?

III. THE TWO SUBLATTICE MODEL

In the R_2T_{17} compounds the 17 transition-metal spins have a combined moment exceeding that of the two rare-earth elements. Holmium, being a heavy rare-earth element, has its moment antiparallel to the cobalt or iron moment. In both Ho_2Co_{17} and Ho_2Fe_{17}, both the Ho and the transition-metal moment prefer the basal plane. [This is not true at intermediate pseudobinary fractions of Co and Fe on the T sites, the region of interest to Rinaldi and Pareti.[6] One of us has recently discussed the easy direction of magnetization in the $R_2(Co_{1-x}Fe_x)_{17}$ compounds as the composition x and the temperature are varied.[15]] In the absence of a magnetic field both M_R and M_T, the two sublattice moments, lie in the basal plane and point antiparallel to each other (apart from possible effects of the individual sixfold anisotropies in the plane), to minimize their anisotropy and exchange energy. But for a nonzero magnetic field along the c axis, the dominant M_T is rotated up toward the field and so the smaller M_R is induced by the antiferromagnetic exchange to rotate downward. (Of course ultimately, when the external field exceeds the exchange field, both sublattice moments are pulled up toward H.) But the external field and the anisotropy of the rare-earth element both oppose this, and so M_R does not rotate down as much as M_T swings up. Canting is induced. Even at the smallest fields, and with small deviations from antiparallel alignment, this induced canting reduces the effective magnetic anisotropy by an order of magnitude.

Let K_{1R} and K_{2R} be the first and second anisotropy constants (per unit volume) of the rare-earth element and K_{1T} and K_{2T} be those of the transition metal. In a field H along c, M_T swings up to angle ψ and M_R down to angle θ, measured from the c axis, as in Fig. 1. The energy density of each sublattice is then

$$\epsilon_R = -M_R H\cos\theta - K_{1R}\sin^2\theta - K_{2R}\sin^4\theta + \lambda M_R M_T\cos(\psi+\theta)\ , \tag{3.1}$$

$$\epsilon_T = -M_T H\cos\psi - K_{1T}\sin^2\psi - K_{2T}\sin^4\psi + \lambda M_R M_T\cos(\psi+\theta)\ . \tag{3.2}$$

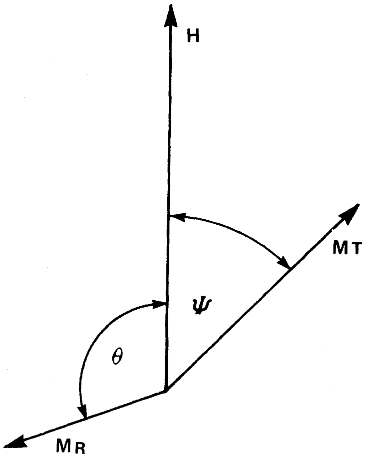

FIG. 1. Direction of rare-earth element magnetization M_R and transition-metal magnetization M_T relative to external field H.

λ is the intersublattice Weiss field constant, and we have dropped all but the two lowest-order anisotropy terms of Eq. (2.1); later on we shall consider higher-order terms. Minimizing Eq. (3.1) with respect to θ and Eq. (3.2) with respect to ψ gives

$$0 = M_R H\sin\theta - K_{1R}\sin2\theta - 2K_{2R}\sin^2\theta\sin2\theta - \lambda M_R M_T\sin(\psi+\theta)\ , \tag{3.3}$$

$$0 = M_T H\sin\psi - K_{1T}\sin2\psi - 2K_{2T}\sin^2\psi\sin2\psi - \lambda M_R M_T\sin(\psi+\theta)\ . \tag{3.4}$$

These equations can be solved on a computer to find the two angles as a function of H and the several parameters. But it is edifying to examine the small-field and small-angle behavior analytically. We introduce the small angles

$$\xi \equiv \theta - \frac{\pi}{2},\quad \eta \equiv \frac{\pi}{2} - \psi\ , \tag{3.5a}$$

so that

$$\delta \equiv \pi - (\theta+\psi) = \eta - \xi \tag{3.5b}$$

is the deviation from antiparallel alignment of the two moments and ξ and η are the deviations of each moment from the basal plane. Up to linear terms in these small angles, Eqs. (3.3) and (3.4) become

$$\eta = \frac{\left[1 + \dfrac{2\mathcal{K}_R}{L}\right] M_T - M_R}{2\left[\mathcal{K}_R + \mathcal{K}_T + 2\left[\dfrac{\mathcal{K}_R\mathcal{K}_T}{L}\right]\right]} H\ , \tag{3.6}$$

$$\xi = \frac{M_T - \left[1 + \frac{2\mathcal{K}_T}{L}\right] M_R}{2\left[\mathcal{K}_R + \mathcal{K}_T + 2\left[\frac{\mathcal{K}_R\mathcal{K}_T}{L}\right]\right]} H , \quad (3.7)$$

and

$$\delta = \frac{\mathcal{K}_R M_T + \mathcal{K}_T M_R}{L(\mathcal{K}_R + \mathcal{K}_T) + 2\mathcal{K}_R\mathcal{K}_T} H . \quad (3.8)$$

Here we have defined for convenience

$$L \equiv \lambda M_R M_T , \quad (3.9a)$$

and again

$$\mathcal{K}_R = K_{1R} + 2K_{2R} \quad (3.9b)$$

and $\mathcal{K}_T$ similarly, as in Eq. (2.4) for the macroscopic effective anisotropy. We see that the canting angle is linear in H.

The component of the magnetization along the c axis is

$$M_H = M_T\cos\psi + M_R\cos\theta \cong M_T\eta - M_R\xi ,$$

$$M_H = \frac{\frac{M^2}{2} + \frac{K_R M_T{}^2 + \mathcal{K}_T M_R{}^2}{L}}{\mathcal{K}_R + \mathcal{K}_T + 2\left[\frac{\mathcal{K}_R\mathcal{K}_T}{L}\right]} H . \quad (3.10)$$

Here we have identified

$$M = M_T - M_R , \quad (3.11)$$

the net ferrimagnetic magnetization when the two sublattices are antialigned in the basal plane. Forcing Eq. (3.10) into the rigid-moment rotation mold of Eq. (2.3), we find that to lowest order in the magnetic field the effective anisotropy constant is

$$\mathcal{K}_{\text{eff}} = (K_1 + 2K_2)_{\text{eff}} = \frac{\mathcal{K}_R + \mathcal{K}_T + \frac{2\mathcal{K}_R\mathcal{K}_T}{L}}{1 + \frac{2(\mathcal{K}_R M_T{}^2 + \mathcal{K}_T M_R{}^2)}{LM^2}} . \quad (3.12)$$

Formula (3.12) behaves properly in the limit of large intersublattice exchange; as J_{RT} (and λ and L) becomes large and the magnetization stiffens, the effective anisotropy trends to the sum of the sublattice anisotropies. Formula (3.12) appears to suggest that $\mathcal{K}_{\text{eff}}$ goes to zero when $M_T = M_R$. It is rather that the whole rigid-rotation approach is unreasonable in that case; the experimentalist would never have applied Eq. (2.3) to an antiferromagnet.

Now let us apply formula (3.12) to Ho_2Co_{17} and Ho_2Fe_{17}. The spin-wave measurements[12] give most, but not all, of the parameters. The two sublattice moments are taken as gJ for the rare-earth element, with the balance assigned to the Co and Fe to fit the measured net moment. The intersublattice exchange constant J_{RT} is, from the measured curvatures, -0.14 meV for Ho_2Co_{17} and -0.12 meV for Ho_2Fe_{17}. These convert to the intersublattice exchange energies listed in Table I.

From the nature of the three branches observed Clausen assigns one branch to the 34 Co or Fe atoms in the unit cell. One nondispersive triply degenerate branch is clearly an R branch, and the remaining branch is a nondegenerate R branch (there are 4 R atoms in the unit cell). Thus there is one gap energy to employ to fix the transition-metal anisotropy and two gaps for the anisotropy of the holmium. Equation (2.1), up the sixth-order terms, contains four coefficients, and each sublattice T and R has its own set of four. To make the problem tractable Clausen terminates the anisotropy expansion, Eq. (2.1) of the transition metal at the leading

TABLE I. Magnetic anisotropy of Ho_2Co_{17} and Ho_2Fe_{17}. All energies are to be multiplied by 10^7 ergs/cm^3. The fourth and fifth columns contain the spin-wave data reported by Clausen (Ref. 12). The last column contains data reported by Clausen and Nielsen (Ref. 14). A different breakdown into K_{1R} and K_{2R} contributions, equally consistent with Clausen's spin-wave data produces, by Eq. (3.12) for Ho_2Fe_{17}, a value $\mathcal{K}_{\text{eff}} = 1.85 \times 10^7$ ergs/cm^3, and also improves the calculated $K_{2\text{eff}}$. See text following Eq. (3.13).

	$\frac{M_T^2}{(M_T - M_R)^2}$	$\frac{M_R^2}{(M_T - M_R)^2}$	L	K_{1R} (spin wave)	K_{1T} (spin wave)	$K_{1R} + K_{1T}$ (spin wave)	$\mathcal{K}_{\text{eff}}$ [calculated by Eq. (3.12)]	$\mathcal{K}_{\text{eff}}$ (by moment measurement)
Ho_2Co_{17}	11.53	5.72	19.7	16.4	14.4	30.8	1.92	1.48
Ho_2Fe_{17}	4.76	1.49	30.3	20.6	0	20.6	2.76	2.15

$l=2$ term, fixing its value. The observed gap converts to $K_{1T}=14.4\times10^7$ ergs/cm^3 for the cobalt and zero for iron. For the Ho there are two measured gaps and four coefficients. Clausen assumes that K_{2R} and K_{3R} are zero and finds K_{1R} and K_{4R}, the coefficient determining the sixfold anisotropy in the plane. The values thus found are listed in Table I. The holmium anisotropy, presumably of single-ion origin, should be the same in the Co and Fe compounds. Perhaps the actual small difference is due to lumping some of K_2 into K_1 in the cobalt case, or merely to the very large error bars on the measured gap, and hence on the determination of K_{1R} in the case of Ho_2Fe_{17}.

Clausen assumed that the two sublattices rotate rigidly antiparallel. Were this so, the macroscopic anisotropy energy density would be the sum of the contributions from the two sublattices, the sixth column of Table I, something like 30 and 20×10^7 ergs/cm^3 in the two materials. Canting, embodied in Eq. (3.12), reduces this by a factor of 10 or 15 to the values entered in the next to last column of Table I. It will be seen that the softened K's are in fact close to those reported by moment measurements shown in the last column [and calculated by the experimentalist by using Eq. (2.3)].

It will be evident to the reader that the $\mathscr{K}_{\rm eff}$ of Eq. (3.12) will not have the usual temperature or magnetic field dependence,[16] and not merely because of the mixing in of K_2 with K_1. But the intrinsic sublattice K_{iR} and K_{iT} should have $\hat{I}_{l+1/2}$ dependence on their sublattice magnetizations.

Clausen and Nielsen's[14] magnetization measurements show a difference between the behaviors of Ho_2Co_{17} and Ho_2Fe_{17}. Up to the fields that were available (10 or 15 kOe) the induced c-axis moment is linear in field for the Co compound but displays a decided negative curvature, an H^3 term, for the Fe compounds. Clausen and Nielsen use an expression equivalent to the cubic term of Eq. (2.3) to measure K_2 in Ho_2Fe_{17}, and their measured value is entered in Table II. But this is, of course, subject to reinterpretation in terms of sublattice canting. Canting creates two contributions to the $l=4$ and higher-order harmonics. A direct result of sublattice bending will be a reduction of $K_{2\,\rm eff}$ from its intrinsic sublattice contributions, as with $K_{1\,\rm eff}$. Another and more harmonious contribution is the echoing in K_2 of the modulation of canting induced by K_1. The latter is the point of the work of Rinaldi and Pareti.[6] As the net moment is rotated around by the magnetic field, the canting angle between the two sublattices will, because of the intrinsic K_1 of either or both sublattices, breathe in and out with the full crystal symmetry. This symmetric modulation of the canting will generate higher harmonics—K_2 and K_3, echoes of K_1—whether or not these are present intrinsically. These overtones will not have the normal temperature or field dependence[16] any more than will $K_{1\,\rm eff}$. In fact, an indication that canting is complicating the anisotropy is the presence of more high-order harmonics than one usually encounters, and their unconventional temperature and field dependence.

Rinaldi and Pareti give an approximation formula for canting-induced higher-order anisotropies. They show that in systems in which one sublattice prefers the plane and the other the axis [this is not the case in Ho_2Co_{17} or Ho_2Fe_{17}, but does obtain at intermediate pseudobinaries of Co and Fe, such as $Ho_2(Co_{0.6}Fe_{0.4})_{17}$] the induced canting angle is large enough so that its modulation makes a large contribution to $K_{2\,\rm eff}$. This can cause the magnetization to lie off at some intermediate angle out of the plane, and can cause large discontinuous jumps in the magnetization curve.[7,8]

We have also developed an expression for the effective coefficient $K_{2\,\rm eff}$ for small anisotropy exchange. The derivation is long and we do not give it here. Comparing our two-sublattice H^3 term to that of Eq. (2.3), we find

TABLE II. Magnetic anisotropy of Ho_2Co_{17} and Ho_2Fe_{17}. The $l=4$, K_2 coefficient. All energies are to be multipled by 10^7 ergs/cm^3. K_{1R} and K_{2R} are underdetermined by the spin-wave gap measurement. An assignment of these constants consistent with the measured gap yields $K_{2\rm eff}=-1.79\times10^7$ ergs/cm^3, and also improves the calculated $\mathscr{K}_{\rm eff}$. See text following Eq. (3.13).

	L	$\frac{M_R}{\lvert M\rvert}$	$\frac{M_T}{M}$	$\mathscr{K}_{1R}$ (spin wave)	$\mathscr{K}_{1T}$ (spin wave)	K_{2R} (as assumed by Clausen)	K_{2T} (as assumed by Clausen)	$K_{2\rm eff}$ [calculated by Eq. (3.12)]	$K_{2\rm eff}$ measured
Ho_2Co_{17}	19.7	2.39	3.40	16.4	14.4	0	0	-0.04	
Ho_2Fe_{17}	30.3	1.22	2.18	20.6	0	0	0	-0.27	-1.75

$$K_{2\,\mathrm{eff}} = \left[L + \frac{2M_R^2}{M^2}\mathscr{K}_T + \frac{2M_T^2}{M^2}\mathscr{K}_R \right]^{-4}$$

$$\times \left[K_{2T}\left[L + 2\mathscr{K}_T\frac{M_T}{M} \right]^4 + K_{2R}\left[L - 2\mathscr{K}_T\frac{M_R}{M} \right]^4 - \left[\frac{2L}{M^2} \right](M_R\mathscr{K}_T + M_T\mathscr{K}_R)^2\right.$$

$$\left.\times \left[L + \frac{M_T}{M}\mathscr{K}_R - \frac{M_R}{|M|}\mathscr{K}_T \right]^2 \right]. \tag{3.13}$$

Note that expression (3.13) is symmetric with respect to the two sublattices, including terms linear in M. We see that as $L \rightarrow \infty$ and the moment rigidifies, $K_{2\,\mathrm{eff}} \rightarrow K_{2T} + K_{2R}$ as it should. The first two terms in the large square brackets represent the softened direct contributions of the individual K_2's to the effective K_2. Though the form of these terms is different from (3.12), the idea is the same. The third term, the product of several factors, is the overtone, the contribution Rinaldi and Pareti discuss. It goes to zero as L^{-1}; as L increases, it straightens its hold on the two moments and breathing is suppressed. Note that this contribution arises from $K_1 + 2K_2$ of the individual sublattices, and will be present even when the sublattice $K_2 = 0$. In fact, $K_2 = 0$ is the assignment assumed by Clausen and listed in our Table II. We see that the harmonic of K_{1R} into $K_{2\,\mathrm{eff}}$ contributes only -0.3×10^7 ergs/cm^3, compared to the measured -1.75×10^7 ergs/cm^3 of Ho_2Fe_{17}. Most of $K_{2\,\mathrm{eff}}$ must come from intrinsic sublattice K_{2R} and K_{2T} contributions.

Clausen, for want of more complete data, made the sensible simplest choice. An assignment equally consistent with the measured spin-wave gap of Ho_2Fe_{17} is $K_{R1} = 98.4\times10^7$ ergs/cm^2 and $K_{R2} = -47.0\times10^7$ ergs/cm^3. This choice gives $K_{\mathrm{eff}} = 1.85\times10^7$ ergs/cm^3 (compared to the measured 2.15×10^7 ergs/cm^3; a 14% error) and $K_{2\,\mathrm{eff}} = -1.79\times10^7$ ergs/cm^3 (compared to the measured -1.75×10^7 ergs/cm^3; a 5% error). $K_{1\,\mathrm{eff}}$ is then 5.43×10^7 ergs/cm^3 (compared to Clausen's estimated 5.65×10^7 ergs/cm^3; a 4% error).

We have also calculated the sixfold anisotropy in the basal plane, the K_4 coefficient of Eq. (2.1). Again we follow the rotation of the two sublattices, but now under torque by a magnetic field in the basal plane. We find the effective coefficient induced by the intrinsic sublattice sixfold anisotropies K_{4R} and K_{4T} to be

$$K_{4\mathrm{eff}} = \frac{L(K_{4R}+K_{4T}) + 36K_{4R}K_{4T}}{L + 36\dfrac{M_T^2}{M^2}K_{4R} + 36\dfrac{M_R^2}{M^2}K_{4T}}\,. \tag{3.14}$$

As $L \rightarrow \infty$, $K_{4\mathrm{eff}} \rightarrow K_{4R} + K_{4T}$ as it should. Table III contains Clausen's spin-wave data, Clausen and Nielsen's measured K_4, and the $K_{4\mathrm{eff}}$ we find by Eq. (3.14). Moment flexing reduces the power of the intrinsic sublattice anisotropy by a factor of 43 in the Co case, and a factor of 15 in the Fe case. Our results are within a factor of the experimental findings. Better agreement will have to wait upon more accurate determination at the intrinsic sublattice anisotropy coefficients.

IV. APPLICATION TO OTHER COMPOUNDS

In deriving the expressions for the effective-anisotropy coefficients, we took it that both sublattices preferred the basal plane, as in Ho_2Co_{17} and Ho_2Fe_{17}. On the other hand, it is clear that the ef-

TABLE III. Sixfold basal plane anisotropies of Ho_2Co_{17} and Ho_2Fe_{17}. All energies are to be multipled by 10^7 ergs/cm^3.

	L	$\left[\frac{M_T}{M}\right]^2$	$\left[\frac{M_R}{M}\right]^2$	K_{4R}	K_{4T}	$K_{4\mathrm{eff}}$ [calculated by Eq. (3.14)]	$K_{4\mathrm{eff}}$ (by moment measurement)
Ho_2Co_{17}	19.7	11.53	5.72	2.0	0	0.046	0.066
Ho_2Fe_{17}	30.3	4.76	1.49	2.5	0	0.17	0.085

fects we are describing will be largest when the canting angle and its harmonic modulation is largest. This will be the case when one sublattice prefers the plane, the other the axis. This was recognized by Rinaldi and Pareti, who applied their analysis to an intermediate pseudobinary of Co and Fe [the transition-metal subsystem prefers the axis in $R_2(\mathrm{Co}_{1-x}\mathrm{Fe}_x)_{17}$ for $0.05 < x < 0.5$] with a planar rare-earth element, such as Pr. The oppositely conflicted situation would be $\mathrm{Tm_2Co_{17}}$ or $\mathrm{Tm_2Fe_{17}}$, since Tm prefers the axis (as do Sm, Er, and Yb) and the pure transition metals prefer the plane.

And so it is useful to compare all the possibilities—both anisotropies planar, both axial, and one of each—to see how big the effects can be. There is another bifurcation: In the light rare-earth elements the rare-earth moment M_R is parallel to M_T, and for these ferromagnetic materials there are again the several possibilities for the signs of K_R and K_T. But the situation is not as complicated as it seems. When one repeats the calculations of Sec. III, setting both vectors $\vec{\mathrm{M}}_R$ and $\vec{\mathrm{M}}_T$ in the first quadrant in Fig. 1, and replacing the exchange coupling of the sublattices in Eqs. (3.1) and (3.2) by $-\lambda M_R M_T \cos(\theta-\psi)$ so that λ and L are still defined as positive quantities, one finds the same result as for antiparallel coupling. To apply formulas (3.12) and (3.14) to either antiparallel or parallel coupling, one always takes L as a positive quantity. The only difference in application to the two couplings is in the interpretation of M, the macroscopic moment. For the ferrimagnetically coupled heavy rare-earth elements M is the (magnitude of the) difference in sublattice moments; for the ferromagnets M is the sum of the sublattice moments $M = M_R + M_T$. This change has a large quantitative impact. Since M occurs quadratically in Eq. (3.12), and multiplied by L, the much larger magnetization which results from ferromagnetic alignment has the same sort of effect as stiffening the interlattice exchange—it makes the effective anisotropy more like the sum of the sublattice anisotropies. Thus for permanent magnets ferromagnetic coupling is preferable not only on the score of larger remanence and induction, but for larger effective magnetic anisotropy and coercive force as well.

To apply formula (3.13) to the case of parallel ferromagnetic coupling, one must change the sign of M_R everywhere, both in M and in the linear terms in M_R.

We worked out Sec. III for two planar sublattices. When both sublattices prefer the axis everything follows as before; one simply inserts the appropriate negative signs for K_{1R} and K_{1T}.

A more difficult situation is obtained when one sublattice prefers the axis, one the plane. This is the case already analyzed by Rinaldi and Pareti.[6] In general the solution is complicated and expressions for effective anisotropies cannot be given in closed form. Rinaldi and Pareti do as well as they can; they analyze in terms of the direction of the magnetization itself. For small fields the sublattice moments and the net moment deviate only a small amount from their zero-field orientations, but these three can point far from the axis, the plane, or each other.

There is a regime in which things simplify. Suppose the intersublattice exchange is much larger than the magnitude of the smaller of the two sublattice anisotropies. Then both sublattice moments will be substantially parallel or antiparallel, and will lie as dictated by the sum of the anisotropies—either close to the axis or to the basal plane. We can then proceed as in Sec. III and carry those equations further; we expand to first order in the small K/L and final simple approximate expressions. For example, Eq. (3.12) becomes

$$\mathscr{K}_{\mathrm{eff}} \simeq (\mathscr{K}_R + \mathscr{K}_T) \Big/ \left[1 + \frac{2(\mathscr{K}_R M_T \mp \mathscr{K}_T M_R)^2}{L(\mathscr{K}_R + \mathscr{K}_T) M^2} \right] . \tag{4.1}$$

Equation (4.1) can be used when $\mathscr{K}_R$ and $\mathscr{K}_T$ have opposite signs so long as either $\mathscr{K}_R/L$ or $\mathscr{K}_T/L$ is small. [It can also be used, of course, when both $\mathscr{K}_R$ and $\mathscr{K}_T$ have the same sign, but is then less accurate than is Eq. (3.12).] Equation (4.1) is applicable to both ferro- and antiferromagnetic coupling. The minus sign in Eq. (4.1) is for antiferromagnetic alignment and the plus sign for ferromagnetic sublattice coupling.

A glance at Table I will show that $\mathscr{K}_R/L$ is not small. The intersublattice exchange is not overwhelming in the $R_x T_y$ compounds, while the rare-earth element anistropy is generally large. But while the cobalt anisotropy is large, that of iron is not. For example, Eq. (4.1) applied to $K_{1\mathrm{eff}}$ of $\mathrm{Ho_2Fe_{17}}$ gives the result 2.76×10^7 ergs/cm^3, the same answer obtained by the more cumbersome Eq. (3.12). But Eq. (4.1) gives entirely the wrong answer for $\mathrm{Ho_2Co_{17}}$ because $K_{1\mathrm{Co}}/L$ is not small. The utility of Eq. (4.1) and the equivalent expansions of Eqs. (3.13) and (3.14) is its applicability to cases in which the two sublattices have anisotropies of opposite sign, but where one is small, as in $\mathrm{Sm_2Fe_{17}}$. On the other hand, some cobalt compounds, such as $\mathrm{Sm_2Co_{17}}$ and perhaps $\mathrm{Ho_2(Co_{0.6}Fe_{0.4})_{17}}$, we expect that the two sublattices should point off at large angles from each other and from the symmetry directions. For these, the Rinaldi and Pareti approach is in order.

Lastly, we emphasize that since a huge degeneration of macroscopic magnetic anisotropy, and hence of coercivity, comes from sublattice flexing it seems to follow that to increase coercivity one should be trying to increase the *R*-*T* exchange; the ionic magnetic anisotropy is already greater than its effective value.

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