Effect of magnetic anisotropy on the temperature dependence of mangetization of some compounds of the RCo_5 type

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The temperature dependence of the spontaneous magnetization and of the position of the easy magnetization axis of compounds $TbCo_{5.1}$ and $DyCo_{5.2}$ are investigated. An anomalous change in magnetization is observed in the region of the easy axis-easy plane transition. An explanation of the change based on high anisotropy of the rare earth sublattice is proposed and yields semiquantitative agreement with the experiments.

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INTRODUCTION

Rare-earth magnets have a rather large magnetic-anisotropy energy ($\sim 10^8$ erg/cm^3), exceeding by two or three orders the corresponding value for magnets of the iron group. In many cases it becomes even comparable with the exchange energy and this, generally speaking, should lead to a number of essential peculiarities in the magnetic properties (e.g., in the behavior of the domain walls [1] and in the value of the spontaneous magnetization at T = 0 [2]).

We call attention in this paper to the existence of interesting effects of the influence of magnetic anisotropy on the temperature dependence of the spontaneous magnetization. Favorable conditions for their observations obtain in rare-earth metal compounds of the type RCo_3. They have not only a high isotropy, but also (for R = Pr, Nd, Tb, Dy, and Ho, see, e.g., [3]) temperature transitions of the easy plane—easy axis type.1) In the latter case, as we shall show, a jump is observed in the spontaneous magnetization of the crystal, and reflects in pure form the influence of the anisotropy on the magnitude of the spontaneous magnetization.

SAMPLES AND EXPERIMENTAL PROCEDURE

We investigated the compounds $TbCo_{5.1}$ and $DyCo_{5.2}$, the composition of which was set in accord with the results of [4]. The alloys were fused by the induction method in alundum crucibles in an argon atmosphere. After an homogenizing annealing at 1200° C and rapid cooling, the alloys turned out to be single-phase with structure of the $CaCu_5$ type, as revealed by the metallographic and x-ray diffraction data. Spherical single-crystal samples of 2 mm diameter were cut from individual grains of the ingots. They were used to study the dependence of the position of the easy magnetization axis on the temperature in a rotating magnetometer. The same samples were used to investigate the temperature dependence of the spontaneous magnetization with the aid of a vibration magnetometer. In the latter case, at each temperature the sample was mounted with the easy-magnetization direction along the magnetic-field direction. The magnetization was measured in fields 4 and 9 kOe sufficient to produce technical saturation in the sample.

The samples for the neutron-diffraction investigations were pressed from powder with particle dimensions $d < 100\ \mu$. The neutron diffraction patterns were obtained with a diffractometer mounted on one of the horizontal channels of an IVV-2 reactor, in a beam of neutrons with wavelength $\lambda = 1.075$ Å. In all the investigations, the temperature was monitored with copper-constantan and copper-copper + iron thermocouples.

MAGNETIC MEASUREMENTS

Figure 1 shows the variation of the angle θ_0 between the easy-magnetization direction and the c axis of the samples $TbCo_{5.1}$ and $DyCo_{5.2}$ as a function of the temperature, for two values of the magnetic field, 8 and 26 kOe. It is seen that in the case of $TbCo_{5.1}$ the transition from the easy axis anisotropy to the easy plane anisotropy occurs in a narrower temperature interval than in the case of $DyCo_{5.2}$. The temperature limits of the transitions are in reasonable agreement with the results of Okamoto et al. [5]; it must be taken into account here that our samples had a somewhat different composition than theirs. There is a noticeable dependence of θ_0 on the magnetic field intensity, a fact already mentioned earlier [6].

Figures 2a and 2b show the temperature dependence of the magnetic moment μ per formula unit for the samples $TbCo_{5.1}$ and $DyCo_{5.2}$. The general character of the dependence is due to the ferrimagnetic structure of the investigated compounds, and was discussed earlier. [5] Attention should be called to the anomaly of the temperature dependence of the magnetization in the region where the easy-magnetization direction goes over from the basal plane to the c axis. It is seen that the magnetic moments of both compounds increase strongly here, and characteristic kinks of the μ(T) curves are observed at the boundaries of the regions. This behavior of the magnetization was not noted in [5], apparently because the function μ(T) was not investigated in sufficient detail.

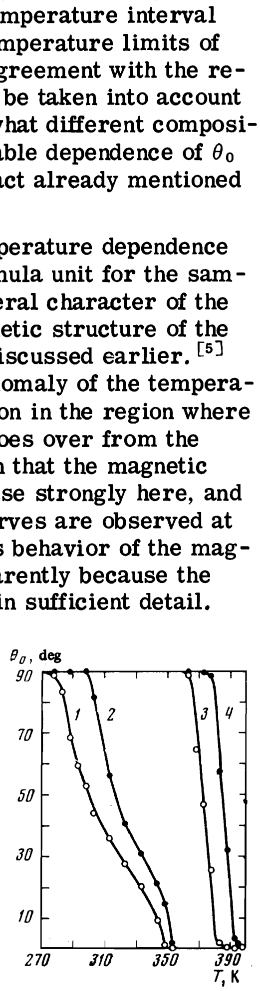

FIG. 1. Temperature dependence of the angle θ_0 between the easy-magnetization direction and the c axis in the single crystal $DyCo_{5.2}$ (1, 2) and $TbCo_{5.1}$ (3, 4). The magnetic-field intensity in the measurements is equal to 8 kOe (dark circles) and 26 kOe (light circles).

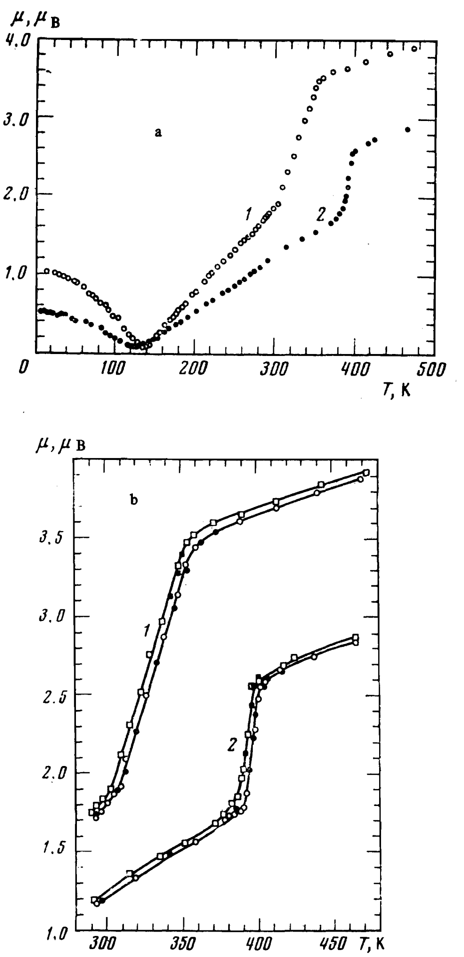

FIG. 2. Temperature dependence of the magnetic moment per formula unit of the compounds $DyCo_{5.2}$ (curves 1) and $TbCo_{5.1}$ (curves 2). a) Curves plotted in 9-kOe field; b) curves plotted in 9-kOe field (squares) and 4-kOe field (circles). Light symbols—measurements with increasing temperature, dark—with decreasing temperature.

As seen from Fig. 2, the transition from the easy plane to the easy axis is accompanied in these compounds by an increase of the magnetization, amounting from 0.6 to 1.6 Bohr magnetons per formula unit for $TbCo_{5.1}$ and $DyCo_{5.2}$, respectively.

NEUTRON DIFFRACTION DATA

We investigated by neutron diffraction only the compound $TbCo_{5.1}$. The diffraction pattern obtained at 4.2°K contains, in the interval of the angle 2θ to 38°, eight groups of reflections, labeled in accord with three-digit system of indices as (100), (0001), (101), (110), (200 + 100), (002), (102), and (201). The ratio of the reflection intensities is close to that obtained by Lemaire and Schweizer.[7] Starting from the magnetic-structure model shown in Fig. 3, the best agreement between the calculated and experimental intensities was obtained at the following values of the magnetic moments for 4.2°K:

$$\mu_{Tb}=8.35\mu_B,\ \mu_{CoI}=1.55\mu_B,\ \mu_{CoII}=1.67\mu_B.$$

The values of the form factor for Tb were taken from the paper of Cable et al.[6], and those for Co from Moon's paper.[9]

The most convenient for a detailed study of the transition of the easy-magnetization axis are the reflections (100) and (001), since their intensities are high enough, they contain a negligibly small nuclear contribution, and have a strongly pronounced dependence on the orientation of the magnetic moments relative to the crystal axes. The vector of the magnetic interaction is determined by the relation

$$\mathbf{q}=\boldsymbol{\varepsilon}(\boldsymbol{\varepsilon}\mathbf{m})-\mathbf{m},$$

where ϵ is the scattering vector, $\mathbf{m}$ is a unit vector in the direction of the magnetic moment of the sublattice. Its modulus for the (001) reflection decreases from 0 to 1 when the orientation of the magnetic moments in the unit cell (Fig. 3) changes from the direction along the a axis to the c axis, whereas for the (100) reflection it increases by $\sqrt{2}$ times. Since the intensity of the reflection is proportional to q^2, the transition of the direction of the easy magnetization from the basal plane to the c axis should lead to a doubling (at equal magnetic moments of the sublattices) of the intensity of the magnetic scattering from the (100) plane and to its vanishing from the (001) plane.

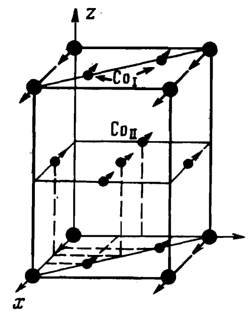

FIG. 3

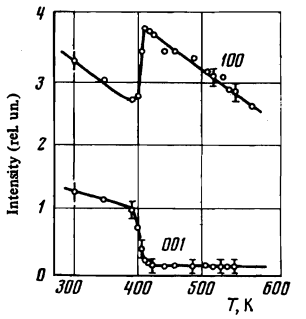

FIG. 4

FIG. 3. Model of magnetic structure of $TbCo_{5.1}$ at 4.2°K. Large circles—Tb atoms, small—Co atoms in two positions, Co_I and Co_{II}.

FIG. 4. Temperature dependence of the intensity of the neutron-diffraction reflections (100) and (001) for the compound $TbCo_{5.1}$.

Figure 4 shows the temperature dependence of the intensities of the reflections (100) and (001) at 300—600°K. It is seen that in the interval 390—410°K there is an abrupt increase of the intensity of the (100) reflection and a decrease of that of (001); this, in accord with the foregoing, indicates rotation of the easy-magnetization axis. The small shift of the region of this rotation towards higher temperatures in comparison with the magnetic-measurement data may be due to the fact that during the neutron-diffraction investigations there was no external magnetic field to influence, as noted above, the temperature of the easy axis—easy plane transition. Above 410°K, the reflection (100) decreases at approximately the same rate as in the interval 80—390°K.

Using the data of Lemair and Shweizer[7] on the temperature dependence of μ_{CoI} and μ_{CoII}, we calculated the magnetic moment of the terbium subsystem at different temperatures from the intensities of the reflections (100) and (001). The estimates show that the decrease of the intensity of the (100) reflection is much less than expected under the condition when the magnetic moment of the Tb sublattice obeys the same variation law in the intervals 80—390 and 490—410°K. According to the data of[7], the rate of change of the magnetic moment of the rare-earth ion at 380°K is approximately 0.01 μ_B/°K, which should have decreased μ_{Tb} by 0.2 μ_B when the temperature was increased from 390 to 410°K, whereas the decrease calculated by us is not less than 0.8 μ_B. This agrees with the change of the combined magnetic moment as obtained from the magnetic measurements. The anomalous decrease of the magnetization of the terbium sublattice explains the indicated decrease of the intensity of the (100) reflection at 410°K in comparison with the calculated one.

THEORY

In addition to the usual role of the magnetic-moment orientation in the crystal, the magnetic crystal anisotropy, characterized by the constant K, can be also the cause of two more radical effects. The latter, however, are quite small (of the order of K/I, where I is the exchange-interaction energy) and are difficult to observe in magnets of the iron group. One of these effects is the change of the spontaneous magnetic moment itself. The physical mechanism of this phenomenon can be particularly clearly understood by considering the limiting case of very large K ($K \gg I$). From the point of view of microscopic representations, we are dealing here with the case of a strong crystalline field, the theory of which has been well developed for the paramagnetic state. Allowance for the crystalline field leads to a separation of the atomic states with a given value of the magnetic moment, into states that have the symmetry of the crystal and have, generally speaking, different values of the magnetic moment. This effect, in particular, can lead to complete vanishing (freezing) of the magnetic moment, if the energywise lowest level turns out to be nonmagnetic (singlet) after the splitting by the crystal field.

Of course, quenching in pure form does not occur for the actually realized case $K \lesssim I$. However, it is necessary here, too, to take into account the restructuring of the energy spectrum under the influence of the crystal field; this restructuring, in full analogy with the quenching effect, leads to a change of the nominal value of the magnetic moments in states corresponding to the symmetry of the crystal potential.

The mathematical procedure reduces to diagonalization of the energy operator, which includes an exchange term and a crystal-field term. The influence of the crystal field manifests itself here in the change of the value of the magnetic moment in the ground state (and this determines its value at T = 0) as well as in the change of the energy (and of the values of the magnetic moment) of the excited states (this affects the temperature dependence of the magnetization). For a complete description of the magnetic properties it is necessary to take into account both indicated factors. However, diagonalization of the crystal potential for real systems with large values of the total angular-momentum quantum numbers J is a very laborious task. Some cases were already calculated earlier.[10] In this paper we confine ourselves only to consideration of the second factor, which determines the change of the temperature dependence of $\mu(T)$ owing to the influence of the magnetic anisotropy. As we shall show, even this calculation makes it possible to explain qualitatively, and to some degree also quantitatively, the experimental data. The difference between theory and experiment indeed gives an idea of the magnitude of the first factor, which is connected with diagonalization.

A theoretical analysis of the compounds RCo_5 can be based on the Hamiltonian

$$\mathscr{H}_0 = -\sum_{i,i'} I_R(\mathbf{R}_i-\mathbf{R}_{i'})\mathbf{J}_i\mathbf{J}_{i'} - \sum_{jj'} I_{Co}(\mathbf{R}_j-\mathbf{R}_{j'})\mathbf{S}_j\mathbf{S}_{j'} - \sum_{ij} I_{RCo}(\mathbf{R}_i-\mathbf{R}_j)\mathbf{J}_i\mathbf{S}_j + K_R\sum_i (J_i^z)^2 - K_{Co}\sum_j (S_j^z)^2, \tag{1}$$

where I_R and I_{Co} are parameters characterizing the exchange in the rare-earth and cobalt sublattices, I_{RCo} is the interlattice exchange parameter, K_R and K_{Co} are the second-order anisotropy constants per ion of the rare-earth and cobalt sublattices, $\mathbf{J}_i$ and $\mathbf{S}_j$ are the operators of the total and spin angular momenta of the rare-earth and cobalt ions, respectively, J_i^z and S_j^z are their z-projections, and $\mathbf{R}_i$ and $\mathbf{R}_j$ are the radius vectors of the ions.

Even this Hamiltonian is written with several simplifying assumptions. The most essential are the assumptions that the Co sublattice can be described in the Heisenberg model (and not in the collectivized model), and that only second-order anisotropy constants are taken into account. As already indicated,[3] the experimental data give grounds for assuming that the exchange inside the rare-earth (RE) sublattice is much smaller than the exchange between the sublattices, and the latter is smaller than the exchange within the Co sublattice. This allows us to neglect the first term in (1) and to change over to the Hamiltonian for the RE ions:

$$\mathscr{H}_i = -I\sum_i \mathbf{n}\mathbf{J}_i + K_R\sum_i (J_i^z)^2. \tag{2}$$

Here I is a parameter characterizing the exchange field produced at the RE ion by the Co sublattice, and n is a unit vector in the direction of its magnetization. The transition to the Hamiltonian (2), generally speaking, presupposes that the characteristics of the Co sublattice (as well as the temperature dependences of the magnetization and of the anisotropy) are already known. These characteristics can be obtained approximately from experiments with compounds such as $LaCo_5$ and YCo_5; they can be obtained more accurately by neutron-diffraction methods, but, since this has not yet been done, it is necessary to use the first method, and this makes the accuracy of the quantitative results seriously worse.

We consider the Hamiltonian (2) in the case $K_R > 0$ (as is the case for R = Pr, Nd, Tb, Dy, Ho), i.e., when the RE sublattices has an anisotropy of the easy plane type. If $\mathbf{n} \parallel z$ (the magnetization is along the difficult axis) and $K_R < I/(2j-1)$, then the ground state is the one with the maximum projection $\mathbf{j}\cdot\mathbf{n}$ on the z axis, i.e., as $T \to 0$ the magnetization per atom reaches its maximum value $g\mu_B j$. The excited states, however, lie closer to the ground state than in the case when there is no anisotropy, since allowance for the anisotropy leads to a shift of each of the initial equidistant levels upward by an amount $K_R m^2$, see Fig. 5. Thus, the lower levels come closer together, and the upper levels move apart. Naturally, this effect leads to a more rapid decrease of the magnetization with temperature in comparison with the Brillouin curve.

If $\mathbf{n} \perp z$, the picture is somewhat different. Now (as well as at any nonzero angle between z and $\mathbf{n}$) the states with definite projection of the angular momentum on $\mathbf{n}$ are not eigenstates of the Hamiltonian, and the ground

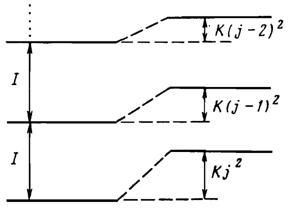

FIG. 5. Change of energy levels under the influence of the magnetic anisotropy.

state (as well as any other state) will be a superposition of these states. Therefore even as $T \rightarrow 0$ the magnetization per atom will be smaller than $g\mu_B j$. It is obvious, however, that allowance for the influence of the anisotropy on the position of the levels will cause, in contrast to the case $\mathbf{n} \perp z$, the lower levels to move apart and the upper ones to come closer together. This means that the decrease of the average angular momentum with temperature will be slower than given by the Brillouin function.

Thus, when the direction of **n** changes from $\mathbf{n} \perp z$ to $\mathbf{n} \parallel z$, the magnetization should increase at low temperatures and decrease at high ones (the boundary is determined by the value of T at which the magnetization curves corresponding to $\mathbf{n} \parallel z$ and $\mathbf{n} \perp z$ intersect).

A more accurate solution of our problem, connected as indicated above with diagonalization of matrices of order $(2J + 1)(2J + 1)$, will become meaningful only if the parameters of the Co sublattice become known with sufficient accuracy.

We used for the calculation the well known methods of the molecular-field theory, i.e., we neglected the level mixing, and took into account only the change of their relative position on account of the anisotropy. In this case it is easy to obtain the following relations for the free energy of the crystal:

$$F=-A\cos(\theta_R-\theta_{Co})+k_R\cos^2\theta_R-k_{Co}\cos^2\theta_{Co}, \tag{3}$$

$$A=N_R I\langle m\rangle, \qquad k_R=N_R\frac{K_R}{2}[3\langle m^2\rangle-j(j+1)], \tag{3a}$$

$$\langle m^l\rangle=\sum_{m=-j}^{j} m^l\exp\left\{\left[Im\cos(\theta_R-\theta_{Co})+\frac{K_R}{2}m^2(1-3\cos^2\theta_R)\right]\frac{1}{k_BT}\right\}\Big/\sum_{m=-j}^{j}\exp\left\{\left[Im\cos(\theta_R-\theta_{Co})+\frac{K_R}{2}m^2(1-3\cos^2\theta_R)\right]\frac{1}{k_BT}\right\}. \tag{3b}$$

Here k_{Co} is the anisotropy constant per unit volume of the Co sublattice; θ_R and θ_{Co} are the angles between the z axis and the magnetizations of the RE and Co sublattices; N_R is the number of RE ions per cm^3; m is the projection of the angular momentum of the RE ion on the direction of the magnetization vector of the RE sublattice. Thus, in the molecular field method the jump of the magnetization for the RE sublattice when the (common) easy direction goes over from the basal plane to the c axis is always negative and vanishes as $T \rightarrow 0$.[2)] This is the natural consequence of dispensing with diagonalization of (2).

Magnets with free energy (3) have already been considered before.[3] As shown in that reference, the transition of the common easy direction from the basal plane to the c axis is connected with a rapid decrease of k_R with rising temperature (the decrease of k_{Co} is much slower, since the exchange within the Co sublattice is much larger than the exchange between the sublattices). Therefore, even though we do have $k_R > k_{Co}$ at $T = 0$, the sign of this inequality is reversed at sufficiently high temperatures. With increasing temperature, at a certain value T_1 ($k_R(T_1) > k_{Co}(T_1)$) the arrangement of the angular momenta of the sublattices in the basal planes ceases to be energywise favored, and further increase of the temperature causes them to rotate towards the c axis; the rotation is accompanied also by a change of the angle between them. The transition terminates at $T = T_2$ ($k_R(T_2) < k_{Co}(T_2)$), when the angular momenta are directed along the c axis. The formula

$$I\langle m\rangle_{1,2}=\left|\frac{K_R[3\langle m^2\rangle_{1,2}-j(j+1)k_{Co}}{{}^1/_2N_RK_R[3\langle m^2\rangle_{1,2}-j(j+1)]-k_{Co}}\right|, \tag{4}$$

which relates the anisotropy and exchange constants and the transition temperature limits T_1 and T_2 (at which the averaging in (4) should be carried out) was derived earlier.[3] We note that if only one anisotropy constant is taken into account for each sublattice, but the exchange between the sublattices is constant, then complete anisotropy constants appear, not only of second order, but also of higher orders. This is clear already from the fact that even at $k_R = k_{Co}$ the sublattice magnetization vectors can rotate freely only if they are parallel, i.e., the exchange between the sublattices is infinitely large. Allowance for the sublattice anisotropy constants of higher orders leads to large mathematical difficulties and can hardly lead to a substantial change in the result, at least in those regions near the transition boundaries, which are essential for the derivation of (4), and where the difference between the sublattice anisotropy constants is large enough. To estimate the jump of the magnetization we therefore used formula (4). In the calculation we made the simplifying assumption that the magnetization and the anisotropy of the Co sublattice do not change when the temperature rises from T_1 to T_2.

The system of two equations in (4) constitutes, at given k_{Co}, T_1, and T_2, a system of complicated transcendental equations in K_R and I. In the zeroth approximation we can solve each of the equations of this system separately for a given I, neglecting in (3b) the dependence on K_R (the Brillouin case). The solutions for I_{Tb} = 105, 120, and 135° are listed in the table (for the remaining elements, the parameters I are connected with I_{Tb} by the De Gennes rule). The table lists also the magnetization jumps calculated from formula (3) at $K_R = K_1$, $K_R = (K_1 + K_2)/2$, $K_R = K_2$, where K_1 and K_2 are the values of K_R obtained from (4) at $T = T_1$ and $T = T_2$, respectively. We see that at $I_{Tb} \approx 110°$, $K_{Tb} \approx 5°$; $I_{Dy} \approx 65°$, and $K_{Dy} \approx 4$–$5°$ the values of the magnetization jump coincide with the experimental ones. Theory, however, predicts in this approximation a somewhat broader transition region than is observed in experiment.

A more consistent calculation, with account taken of K_R in (3b), should decrease in principle the width of the transition region, inasmuch as the $k_R(T)$ curves lies higher than the Brillouin curve at the same values of K_R and I when the angular momenta of the sublattice lie in the plane of the basis, and higher than the Brillouin curve

TABLE I. Values of the anisotropy constants K_1 and K_2 obtained from (4) at $T = T_1$ and $T = T_2$, and of the magnetization jump $\Delta\mu$ in the plane-to-axis transition in accord with formula (3b) at a given I.

$TbCo_{5,1}$, (k_{Co} = 30) $\Delta\mu_{exp}$ = 0.8				$DyCo_{5,2}$, (k_{Co} = 35) $\Delta\mu_{exp}$ = 1.6				$NdCo_5$, (k_{Co} = 400)		
I	K_1	K_2	$\Delta\mu_{theor}$	I	K_1	K_2	$\Delta\mu_{theor}$	I	K_1	K_2
105	5.7	4.1	0.92 0.81 0.70	70	6.7	5.9	2.30 2.20 2.10	57.3	73.6	14
120	4.4	3.5	0.75 0.68 0.63	80	3.4	2.8	1.48 1.40 1.32	65.5	36.9	12.3
135	3.7	3.0	0.65 0.59 0.54	90	2.8	2.4	1.31 1.25 1.20	73.6	23.8	11

The upper values of $\Delta\mu_{theor}$ correspond here to $K_R = K_1$, the lower ones to $K_R = K_2$, and the middle ones to $K_R = (K_1 + K_2)/2$. The values of the exchange constants and of the anisotropy constants are given in deg/atom of R, and the values of the magnetization jumps are given in μ_B/atom of R.

when they are directed along the c axis. This leads, naturally, to a raising of T_1 and a lowering of T_2. At reasonable values of K_R and I it is impossible to obtain in the theory values of T_1 and T_2 that coincide with the experimental ones, since this effect leads to too strong a narrowing of the transition region or even to a "spilling" over the boundaries, i.e., to the condition $T_1 > T_2$. Thus, calculation by means of more exact formulas leads to worse agreement with experiment. However, since allowance for the diagonalization should lead to an overall lowering of the $k_R(T)$ curve at low temperatures, and hence to a decrease of T_1, it must be assumed that when the diagonalization of the $(2J + 1)(2J + 1)$ matrix of the initial Hamiltonian is taken into account the agreement with experiment should improve.

It must also be taken into account that neglect of the diagonalization is not the only cause of deterioration of the agreement between theory and experiment. From among the other causes, it suffices to point out the possible change of the anisotropy constant of the Co sublattice in RCo_5 as a function of the value of R and (albeit much smaller) jump in the values of the magnetization and of the anisotropy constant, which should take place also in the Co sublattice in the region of the transition.

The effect described here is of interest not only in itself, but also as a method of determining the sublattice anisotropy constants, an important factor in the elucidation of the nature of the anisotropy in these compounds. For a more accurate determination of these constants, the following are necessary: 1) a detailed investigation of the magnetization and the anisotropy of the Co sublattice by neutron-diffraction methods, and possibly by NMR and FMR methods; 2) an exact determination of the magnetization and anisotropy of the RE sublattice from (2) with the experimental data on the Co sublattice taken into account.

[1)] This transition does not proceed to conclusion in the case of R = Pr and Ho, but a rotation of the easy-magnetization axis towards the basal plane is observed when T is lowered.

[2)] Naturally, it is assumed that $K_R < I/(2j - 1)$.

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