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# Magnetic transitions in single-crystal $Ho_2Co_{17}$ studied in high magnetic fields

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High-field (up to 35 T) magnetization measurements performed at 4.2 K along different crystal axes of hexagonal $Ho_2Co_{17}$ reveal, above 20 T, magnetic transitions for field directions in the hexagonal plane that have not been studied before in this type of intermetallic compound. The magnetization data, analyzed in a two-sublattice model, provide accurate results for the magnetic parameters of this system.

Magnetism in the rare-earth—cobalt intermetallic compounds is of special importance in view of applications. Recently, it has been recognized that the $R_2Co_{17}$ compounds have some advantages with respect to the series $RCo_5$, not at least by the favorable ratio of rare-earth to $3d$ metal. Powdering, sintering, and annealing techniques are of vital importance in order to reach optimal sample quality. Besides these preparation methods a basic understanding of the magnetic parameters is required to arrive at the most promising composition or substitution for technical applications.

As far as the basic properties are concerned, large values for the spontaneous magnetization can lead to maximal values of the remanent magnetic induction, a strong uniaxial magnetic anisotropy is required to reduce the Bloch-wall width and its mobility, a strong coupling between the rare earth and $3d$ moments is necessary to realize the required strong anisotropy for the $3d$ moment and, finally, a strong interaction between the $3d$ moments should exist in order to bring the Curie temperature well above room temperature. According to these requirements, the light rare-earth metals with their parallel coupling between the rare earth and the $3d$ magnetic moments are preferred for applications. A study of the rare-earth—$3d$ coupling parameter, however, is easier to perform on materials with antiparallel coupling, i.e., in the heavy rare-earth—$3d$ compounds.

In order to better understand the magnetic interactions in $R_2M_{17}$ compounds, we developed a technique for preparing bulk single crystals of a number of $R_2M_{17}$ compounds and performed high-field magnetization measurements in fields up to 35 T. In these experiments we observed magnetic transitions that, to our knowledge, have not yet been reported in the literature for this type of intermetallic compound. We illustrate our observations by presenting the magnetization curves of $Ho_2Co_{17}$ at 4.2 K along different crystallographic directions. This compound (hexagonal, $Th_2Ni_{17}$ type of structure) has been chosen since low-field magnetization (Clausen and Nielson[1]) and inelastic neutron scattering data (Clausen and Lebech[2]) on single-crystal samples are already available.

The apparent discrepancy between the values for the anisotropy constants of this compound, as derived from low-field magnetization measurements and from inelastic neutron scattering data, has been resolved by Sarkis and Callen.[3] These authors pointed out that the low-field magnetization is governed by an effective anisotropy constant one order of magnitude smaller than the intrinsic anisotropy constant of the rare-earth magnetic moment. The weak bond of the net magnetic moment to the easy ($b$) direction is a consequence of the canting of the holmium and cobalt moments for field directions not coinciding with the $b$ direction.

Magnetization curves have been obtained on a spherical sample (diameter 3 mm) in the Amsterdam high-field installation. Details of sample preparation, lattice parameters, and density have been published elsewhere (Menovsky and Franse[4]). The high-field installation and the induction method for magnetization measurements have recently been reviewed (Gersdorf *et al.*[5]). Results along the $a$, $b$, and $c$ directions are presented in Fig. 1. The salient features of these curves are as follows:

(a) For fields along the $b$ direction the magnetization is virtually constant up to 21.5 T and takes a value of 33.5 $A\,m^2/kg$; above 21.5 T the holmium and cobalt moments bend out of the field direction.

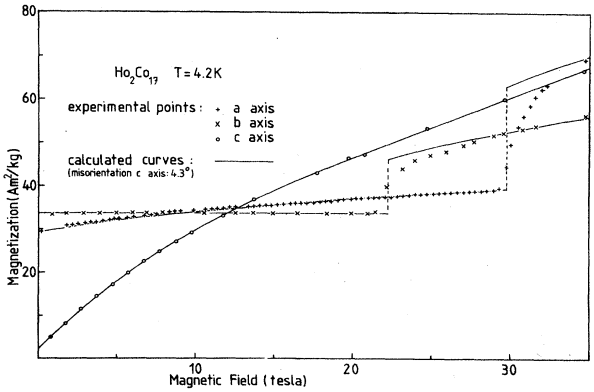


FIG. 1. Magnetization curves along different crystallographic directions of the hexagonal compound $Ho_2Co_{17}$; the full curves represent the calculated magnetization curves in a two-sublattice model with a free-energy expression given by Eq. (1) and with values for the magnetic parameters as given in the text.

 

(b) For fields along the $a$ direction the magnetization starts with a value of $\cos(\pi/6)$ times that along the $b$ direction and increases to a value 1.15 times this starting value at the transition field of 29.5 T; the susceptibility above the transition is about 20 times larger than below.

(c) For fields along the $c$ direction a continuous increase of the magnetization with increasing field is observed.

Assuming a two-sublattice model to be valid, we have to introduce for a proper description of the magnetization curves, besides the sublattice magnetic moments ($M_R$ and $M_T$) and the antiparallel exchange coupling parameter ($n_{RT}$), a number of crystal-field parameters adding up to 8 at restricting the crystal-field terms up to degree 6. The following free-energy expression has been used:

$$E = E_m + E_a + E_{\text{exch}} , \tag{1}$$

where $E_m$ represents the magnetostatic energy of the two-sublattice magnetizations $M_R$ and $M_T$ in the applied field: $E_m = -\mathbf{M}_R \cdot \mathbf{B} - \mathbf{M}_T \cdot \mathbf{B}$. The anisotropy energy $E_a$ is expressed as

$$E_a = \sum_i K_1^i \sin^2\theta_i + K_2^i \sin^4\theta_i + K_3^i \sin^6\theta_i + K_4^i \sin^6\theta_i \cos 6\phi_i \tag{2}$$

with $i = T$ or $R$ and where $\theta_i$ and $\phi_i$ are the polar and azimuthal angles of the direction of the magnetization ($M_T$ or $M_R$) with respect to the $c$ axis and $a$ axis, respectively; the last term in Eq. (1) is the exchange energy term and is expressed in a molecular-field type of description as $n_{RT}\mathbf{M}_R \cdot \mathbf{M}_T$. With the appropriate set of values for the parameters in Eq. (1), not only the transition fields, but also the types of transitions have to be understood. We started our analysis by concentrating on the type of transition. At the $b$-axis transition the symmetry could in principle be broken either in the $b$-$c$ plane or in the $b$-$a$ plane. In the latter case the transition will be, in general, first order, whereas the data in Fig. 1 show a smooth behavior. Therefore, we concentrated on a $b$-$c$ break. The behavior in the $a$ direction below the transition is obviously a rotation in the $a$-$b$ plane. The susceptibility above the transition is about 20 times larger than below and also in this case we have been looking for symmetry breaks in the $c$ direction.

In spite of elaborate computational efforts, however, no satisfying set of parameters has been found that reproduces the measured magnetization curves along the $a$ and $b$ directions under the assumption of a symmetry break in the $c$ direction. It turns out that for most parameter sets the $a$ transition occurs at about 1.15 times the $b$-transition field, which follows from the following reasoning: At the magnetic transitions the external field destabilizes the configuration in which the magnetic moments are bound to the basal plane by the crystal field and the exchange coupling; for field directions along the $a$ axis only $\cos(\pi/6)$ times the external field is effective as a destabilizing factor.

The experimental value for this ratio of transition fields amounts to 1.38 and is far too large. The only way out is a situation in which the cobalt moment has a strong preference for the $(a,b)$ plane and the holmium moment for the $c$ direction. This situation is quite different from physical expectations and does not reproduce the magnetization curves either. We have to conclude that our assumption of a symmetry break in the $c$ direction is not compatible with a description of the measured magnetization curves on the basis of the free-energy expression introduced in Eq. (1).

In our second analysis we disregarded the detailed behavior of the magnetization curves above the transition and concentrated on the values of the transition fields and the magnetization values below those fields. We obtained an almost perfect description in this field range by assuming that the sublattice moments strictly rotate in the basal plane and by considering the free-energy expression for different moment configurations in this plane. Above the transition fields the magnetization data along the $a$ and $b$ axes approach to the solid curves in Fig. 1 which have been calculated on the basis of the fit of the magnetization data below the transition fields.

For an external field $\mathbf{B}$ applied along the easy $b$ axis, for instance, Eq. (1) can now be written as

$$E = -M_R B \cos\alpha - M_T B \cos\beta + K_4^R \cos 6\alpha + K_4^T \cos 6\beta + n_{RT} M_R M_T \cos(\alpha+\beta) \tag{3}$$

with $\alpha$ and $\beta$ the angles between the sublattice magnetizations $\mathbf{M}_R$ and $\mathbf{M}_T$ and the external field $\mathbf{B}$, respectively. Two different moment configurations for fields applied along one specific $b$ axis are shown in Fig. 2.

In this analysis we neglected the basal plane anisotropy of the cobalt sublattice and took the sublattice moments of the holmium and cobalt equal to 83.9 and 117.5 A m$^2$/kg, respectively. These latter numbers correspond with magnetic moments of $10\mu_B$ for the holmium atoms and $1.65\mu_B$ for the cobalt atoms. The antiparallel coupling parameter $n_{RT}$ and the in-plane anisotropy constant to $K_4^{\text{Ho}}$ follow from the magnetization curve along the $a$ axis below the transition field and amount to 0.55 T·kg/A m$^2$ and 190 J/kg, respectively. A change in spin configuration follows from the free energy at a field of 29.8 T with the above given values for the parameters. A similar energy consideration applied to the $b$ axis results in a transition field of 22.3 T. The calculated magnetization curves along the $a$ and $b$ axes with the above given values for the parameters $M_R$, $M_T$, $K_4^{\text{Ho}}$, and $n_{RT}$ are represented by the full curves in Fig. 1.

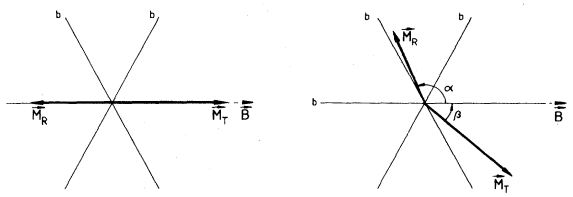


FIG. 2. Two different moment configurations for the external field $\mathbf{B}$ applied along one specific (easy-magnetization) $b$ axis in the basal plane of a hexagonal easy-magnetization-plane ferrimagnetic compound. The two configurations apply to fields below 22.3 and at 26 T, respectively.

The magnetization curve along the $c$ axis has been fitted independently. Values for the anisotropy constants $K_1^{Co}$, $K_2^{Co}$, and $K_3^{Co}$ were taken to be equal to the values reported for $Y_2Co_{17}$ (Hoffer and Strnat[6]). In order to obtain the best fit to the $c$-axis magnetization data we had to assume a misorientation of 4.3° between the $c$ axis and the field direction.

The parameter $n_{RT}$ was found to be equal to the value derived from the magnetization curve along the $a$ axis. For the remaining anisotropy constants we derived the following results: $K_1^{Ho} = -1750$ J/kg, $K_2^{Ho} = 640$ J/kg, $K_3^{Ho} = 0$ J/kg.

The full curve in Fig. 1 represents the best fit to the measuring points with the above given values of the parameters. Except for the value of $K_2^{Ho}$, our values for the anisotropy constants are in good agreement with the results of inelastic neutron scattering experiments: $K_1^{Ho} = -1800$ J/kg, $K_2^{H_0} = K_3^{H_0} = 0$, $K_4^{H_0} = 200$ J/kg.[7]

The calculated curves perfectly follow the experimental data except for the field region just above the transition fields. These deviations possibly point to magnetoelastic effects connected with a distribution of stresses that induces a symmetry-breaking perturbation, quadratic in field (see for instance, Ref. 8). Crystal growth by the Czochralski method proceeds with large temperature gradients over the sample by which mechanical stresses can easily be introduced. No additional annealing has been applied to our as-grown samples in order to reduce stresses.

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