

High-field moment reorientation in $\text{Er}_2\text{Co}_{17}$

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The occurrence of a field-induced transition is reported for the first time in a ferrimagnetic heavy-rare-earth R_2T_{17} compound with easy-axis anisotropy. At 40 T, a transition occurs in $\text{Er}_2\text{Co}_{17}$ from the collinear ferrimagnetic structure to a canted structure and, with increasing field, the forced-ferromagnetic state is gradually approached. The $4f$ - $3d$ interaction in this compound has separately been investigated by measurement of the free-powder magnetization. The observed magnetic isotherms have been analyzed within a mean-field model, describing the competing interactions in the compound.

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I. INTRODUCTION

In the last decades, the magnetism of rare-earth intermetallic compounds of the series $R_2\text{Co}_{17}$ and $R_2\text{Fe}_{17}$ has been extensively studied, particularly after single crystals became available. In these compounds, as generally in R - T (transition-metal) compounds, the strongest interaction is the $3d$ - $3d$ interaction which primarily determines the Curie temperature. The $4f$ - $4f$ interaction is very weak and can be neglected, although its effect contributes to a characteristic variation of the Curie temperature with the R element in an isostructural series. The $4f$ - $3d$ interaction, although much weaker than the $3d$ - $3d$ interaction, is of special importance since by this interaction the strongly anisotropic R -sublattice magnetization is coupled to the much less anisotropic T -sublattice magnetization. In this way, some of the R - T compounds exhibit large anisotropies even at room temperature, one of the prerequisites for potential application of ferromagnetic R - T compounds (in which R is a light rare-earth element) as permanent-magnet material.

$R_2\text{Co}_{17}$ and $R_2\text{Fe}_{17}$ compounds in which R is a heavy-rare-earth element are ferrimagnetic and not appropriate as permanent-magnet material. The exchange interaction between the $4f$ and $3d$ electrons is usually represented by the molecular field by which the R - and T -sublattice moments are coupled. For Fe- or Co-rich R - T compounds, the values for the molecular field are typically of the order of 100 T, so that large magnetic fields are needed to induce changes in the magnetic configurations of the two magnetic sublattices in these compounds. Evidently, to investigate the strength of the R - T interactions by magnetization measurements, ferrimagnetic compounds are best suited because the strict antiparallel configuration of the moments will be affected by a sufficiently high magnetic field and noncollinear configurations may be induced.

The $R_2\text{Co}_{17}$ and $R_2\text{Fe}_{17}$ compounds in which R is one of the heavy-rare-earth elements Tb, Dy, Ho, Er, or Tm crystallize in the hexagonal $\text{Th}_2\text{Ni}_{17}$ (2:17H) type of structure and for the vast majority of these ferrimagnetic compounds ($\text{Er}_2\text{Fe}_{17}$, $R_2\text{Co}_{17}$, and $R_2\text{Fe}_{17}$ compounds with R =Tb, Dy, and Ho), the preferred moment direction is located in the

(hexagonal) basal plane. If a sufficiently large magnetic field is applied along one of the main crystallographic directions in the basal plane, field-induced transitions will occur before the forced-ferromagnetic state is reached. Observation of these transitions, which are primarily based on the interplay between the strength of the applied field and the strength of the R - T interaction, requires high magnetic fields, in the approximate field range of 20–250 T. Transitions of this type have for the first time been found in $\text{Ho}_2\text{Co}_{17}$ (Ref. 1) and later in several other compounds.²

In the remaining ferrimagnetic $R_2\text{Co}_{17}$ and $R_2\text{Fe}_{17}$ compounds, i.e., $\text{Er}_2\text{Co}_{17}$, $R_2\text{Co}_{17}$, and $R_2\text{Fe}_{17}$ compounds with Tm, the hexagonal c axis is the easy moment direction and only one transition will occur before the forced-ferromagnetic state is reached. These transitions, that have been estimated by means of mean-field calculations to require fields in excess of 40 T,³ have thus far not been observed. In the present paper, such a transition in an easy-axis system, predicted to occur in $\text{Er}_2\text{Co}_{17}$ at about 40 T,^{3,4} is reported for this compound.

II. EXPERIMENT

The compound $\text{Er}_2\text{Co}_{17}$ was prepared by arc-melting stoichiometric amounts of the constituent elements Er and Co in a high-purity argon atmosphere. The purity of the starting materials was 99.9% or higher. The ingot was remelted at least three times for homogenization. The ingot was then wrapped in molybdenum foil and sealed in a quartz tube and annealed at 1373 K for 72 h under protection of argon atmosphere, followed by water quenching. Despite the annealing in inert-gas atmosphere, the annealed ingot exhibited slight traces of oxidation which were carefully removed. X-ray powder diffraction (XRD) with $\text{Cu-K}\alpha$ radiation was used to check the hexagonal $\text{Th}_2\text{Ni}_{17}$ (2:17H) structure and to determine the lattice parameters $a=0.8314$ nm and $c=0.8125$ nm.

Fine powder of $\text{Er}_2\text{Co}_{17}$ was prepared in an agate mortar by hand under protection of alcohol. Since the material is very brittle and easy to be milled into fine powder, we did not anneal it after milling to remove imperfections possibly

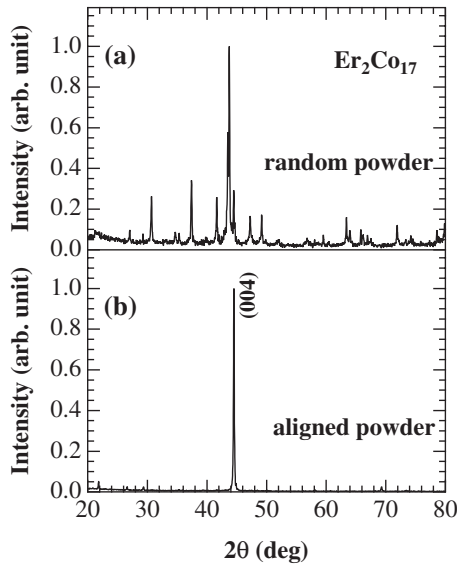


FIG. 1. XRD patterns of (a) randomly oriented powder and (b) magnetically aligned powder of $\text{Er}_2\text{Co}_{17}$.

introduced by the milling. The size of the particles was measured by means of an optical microscope and found to be around $20\text{--}30\ \mu\text{m}$. The average grain size in the annealed material was found to be about $80\text{--}120\ \mu\text{m}$. Inspection of the particles under the microscope did not reveal any oxidation. Magnetically aligned samples were prepared by mixing the powder particles with epoxy resin, putting the mixture in a plastic tube, and letting the mixture in the tube solidify in a field of about 1 T. The cylinder axis of the tube was positioned parallel to the field direction, so that the cylindrical axis corresponds to the easy magnetization direction and the direction perpendicular to the axis is the hard magnetization direction. The XRD patterns presented in Fig. 1 illustrate the effect of the magnetic alignment.

The high-field magnetization measurements have been performed by using a nondestructive pulse magnet installed at the Center for Quantum Science and Technology under Extreme Conditions (KYOKUGEN) at Osaka University, in pulsed fields up to 55 T with pulse duration of 40 ms. The magnetization was detected by an induction method with a standard pick-up coil system.

III. RESULTS

Figure 2 shows the magnetization at 4.2 K of a magnetically aligned-powder sample of $\text{Er}_2\text{Co}_{17}$ measured in fields up to 55 T applied parallel to the alignment direction, which is equivalent to the crystallographic c -axis direction. Below 40 T, the compound is in the ferrimagnetic state, the moments of the Co and the Er sublattice being aligned parallel and antiparallel, respectively, to the applied field. The small increase of the magnetization in this field region is not observed in a measurement on a single crystalline sphere⁵ and is therefore not intrinsic. The increase of the magnetization in the field region before the magnetization jump should be associated with the less perfect circumstance that, in our experiments, we are dealing with aligned-powder particles. The

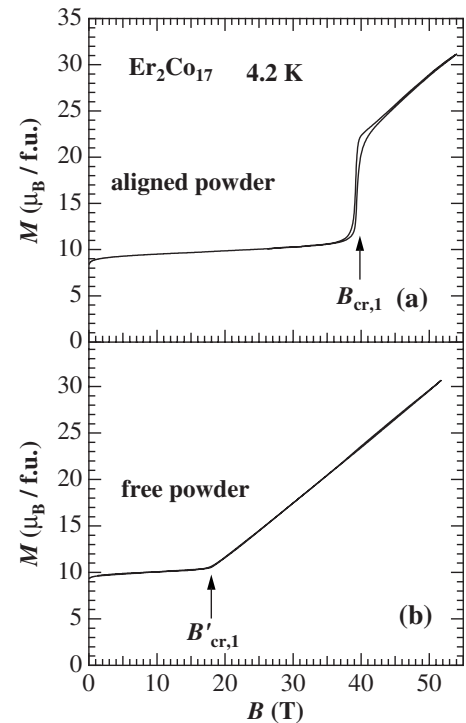


FIG. 2. Magnetization of $\text{Er}_2\text{Co}_{17}$ measured at 4.2 K on (a) powder magnetically aligned in the direction of easy magnetization, with field applied in the alignment direction, and (b) on powder particles free to rotate in the sample holder.

moment in the ferrimagnetic state is most closely represented by the moment of about $10.5\ \mu_B/\text{f.u.}$ measured around 35 T, just before the magnetization jump, where the strong applied field has removed to a large extent the effect of the smaller contribution to the magnetization of imperfectly aligned powder particles. If we assume the Er magnetic moment in $\text{Er}_2\text{Co}_{17}$ to be equal to the free-ion value of $9\ \mu_B$ and to be aligned fully antiparallel with the Co-sublattice moment, this leads to about $28.5\ \mu_B$ for the magnetic moments of the Co sublattice, i.e., $1.68\ \mu_B/\text{Co}$ atom which, given the uncertainty of a few percent in the presently established value of the established Co-sublattice moment, is in good agreement with the value of $1.65\ \mu_B/\text{Co}$ atom reported by Matthaehi *et al.*⁶ for the Co moment in Y_2Co_{17} .

At $B_{\text{cr},1} = 40$ T, a first-order field-induced transition takes place from the state in which the sublattice moments are aligned perfectly antiparallel, with the resultant moment in the c -axis direction, to a state in which they are canted with respect to the c axis as depicted in Fig. 3(a). With further increasing field, an approximately linear increase of the magnetization is observed, corresponding to gradual bending of the two sublattice moments into the c -axis direction. The forced-ferromagnetic state will be reached at a critical-field value $B_{\text{cr},2}$ which is not experimentally accessible.

Also in Fig. 2, the magnetic isotherm at 4.2 K is shown for the powder particles used for magnetic alignment but now free to orient themselves in the sample holder in the applied field. This so-called free-powder technique has been intensively applied to determine the intersublattice-coupling strength in many series of ferrimagnetic R (heavy-rare-earth)

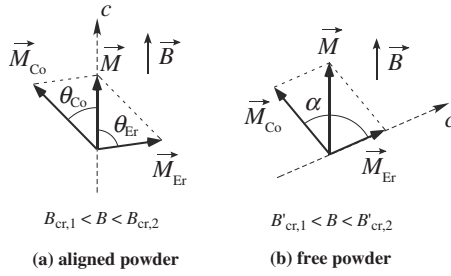


FIG. 3. Schematic representation of the Er- and Co-sublattice moments configuration for the mean-field calculation in (a) an aligned- and (b) a free-powder sample. θ_{Er} and θ_{Co} are the angles of \vec{M}_{Er} and \vec{M}_{Co} , respectively, with the c -axis direction. α is the angle between \vec{M}_{Er} and \vec{M}_{Co} .

- T (transition-metal) compounds.⁷ Although an equivalent and more accurate measurement has been reported by Verhoef *et al.*^{8,9} on a single-crystalline sphere that was free to rotate in the applied field, we have also measured the free-powder magnetization in order to obtain information on the presently used powder particles.

Similar to the magnetization of the aligned sample, also the magnetization of the free-powder particles exhibits a small increase of the magnetization in the low-field region which, on the basis of the ferrimagnetic ground state, is not expected and should not be regarded as intrinsic. This field dependence of the magnetization is absent in the magnetization of the free single crystalline sphere.^{8,9} This indicates that the powder particles used in the present study, although their average size of 20–30 μm is appreciably smaller than the grain size of 80–120 μm , may not be perfectly single crystalline. This would also explain the similar field dependence of the magnetization of the magnetically aligned sample in which, in addition, also the inevitable imperfect alignment of the single-crystalline particles will play a role.

In accordance with the result for the aligned sample, a value for the ferrimagnetic moment of $10.5\mu_B/f.u.$ is derived from the low-field part of the magnetization curve. At $B'_{\text{cr},1} = 18$ T, the perfect antiparallel configuration of the Er- and Co-sublattice moments is broken and, with further increasing field, the powder particles will rotate into a minimum-energy direction in which the direction of the resultant magnetization of the particle will coincide with the direction of the applied field as shown in Fig. 3(b). At a second critical field $B'_{\text{cr},2}$, which cannot be reached experimentally, both sublattice moments will be directed into the direction of the applied field.

IV. DISCUSSION

The magnetic isotherm of Er₂Co₁₇ at 4.2 K, as presented in Fig. 2, has been obtained with the magnetic field applied along the direction of the magnetic alignment. Since the c -axis direction is the preferred magnetization direction in Er₂Co₁₇, the observed magnetization behavior of the magnetically aligned powder can be considered to represent the magnetization behavior along the crystallographic c axis. The isotherm has been analyzed in terms of a mean-field

description in which the compound is assumed to consist of two magnetic sublattices with magnetic moments M_{Er} and M_{Co} . In a mean-field description, the free energy of Er₂Co₁₇ is represented by

$$E = E_{\text{Er}}^{\text{an}}(\theta_{\text{Er}}, \phi_{\text{Er}}) + E_{\text{Co}}^{\text{an}}(\theta_{\text{Co}}, \phi_{\text{Co}}) + n_{\text{ErCo}} \vec{M}_{\text{Er}} \cdot \vec{M}_{\text{Co}} - (\vec{M}_{\text{Er}} + \vec{M}_{\text{Co}}) \cdot \vec{B}. \quad (1)$$

The first two terms represent the anisotropy energies of the Er and the Co sublattice, respectively. In the present analysis, we consider the Er-anisotropy constants up to fourth order and, since the Co anisotropy is much smaller, only the second-order Co-anisotropy constant,

$$E_{\text{Er}}^{\text{an}} = K_1^{\text{Er}} \sin^2 \theta_{\text{Er}} + K_2^{\text{Er}} \sin^4 \theta_{\text{Er}},$$

$$E_{\text{Co}}^{\text{an}} = K_1^{\text{Co}} \sin^2 \theta_{\text{Co}}. \quad (2)$$

θ_{Er} and θ_{Co} are the polar angles of \vec{M}_{Er} and \vec{M}_{Co} as shown in Fig. 3(a). Since we are considering a magnetization process in which the magnetic field is applied along the c axis, the anisotropy terms involving the azimuthal angles ϕ_{Er} and ϕ_{Co} of \vec{M}_{Er} and \vec{M}_{Co} , respectively, have not been taken into consideration. The third term in Eq. (1) describes the $3d$ - $4f$ exchange interaction and contains the intersublattice-molecular-field coefficient n_{ErCo} . The last term in Eq. (1) represents the Zeeman energy in an applied field B .

Information on the approximate value of the intersublattice molecular-field coefficient n_{ErCo} in the presently studied Er₂Co₁₇ sample can be derived in a straightforward manner^{8–10} from the free-powder magnetization depicted in Fig. 2(b). In general, at low temperatures, the R -sublattice anisotropy strongly dominates the T -sublattice anisotropy in R - T compounds. In the magnetization process of a single crystal that can freely rotate in the applied field, under the assumption that the T -sublattice anisotropy is zero, the R sublattice will be in its easy direction for all possible moment orientations of the two sublattices. Instead of a single crystal, also polycrystalline material, if powdered to a sufficiently small particle size, can be used because the powder can be considered as a collection of single crystals, leading to the same result as one single crystal. In this simple case, where no change in anisotropy energy is involved in the magnetization process, Eq. (1) can be simplified to

$$E = n_{\text{ErCo}} M_{\text{Er}} M_{\text{Co}} \cos \alpha - MB \quad (3)$$

with M given by

$$M = \sqrt{M_{\text{Er}}^2 + M_{\text{Co}}^2 + 2M_{\text{Er}}M_{\text{Co}} \cos \alpha}. \quad (4)$$

The equilibrium directions of \vec{M}_{Er} and \vec{M}_{Co} can be determined by minimizing the free energy in Eq. (3) with respect to α [see Fig. 3(b)]. At low fields, the configuration of the moments of the two sublattices is perfectly antiparallel and the magnetization is $M = M_{\text{Co}} - M_{\text{Er}}$. Above a first critical field,

$$B'_{\text{cr},1} = n_{\text{ErCo}}(M_{\text{Co}} - M_{\text{Er}}), \quad (5)$$

the moments start to bend towards each other and the magnetization is represented by

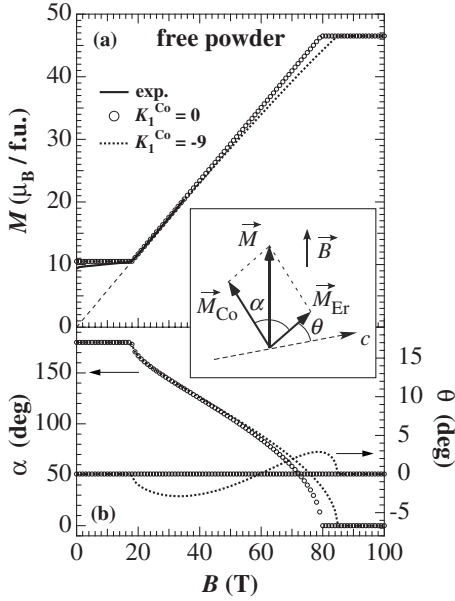


FIG. 4. Calculations of the free-powder magnetization. (a) Magnetization of $\text{Er}_2\text{Co}_{17}$, as measured at 4.2 K (solid line), and as calculated for $K_1^{\text{Co}}=0$ K/f.u. (open circles) and $K_1^{\text{Co}}=-9$ K/f.u. (dotted line). Dashed line shows the straight-line fit up to 35 T. (b) Calculated field dependencies of the angles α and θ . Inset: schematic representation of the moments configuration in a free-powder sample with nonzero K_1^{Co} .

$$M = n_{\text{ErCo}}^{-1} B, \quad (6)$$

which allows for a straightforward determination of the parameter n_{ErCo} . Beyond a second critical field, $B'_{\text{cr},2} = n_{\text{ErCo}}(M_{\text{Co}} + M_{\text{Er}})$, forced-ferromagnetic alignment of the two sublattice moments is reached, corresponding to a magnetization equal to $M = M_{\text{Co}} + M_{\text{Er}}$.

The analysis of the free-powder data in terms of the above description can only be carried out with limited accuracy. The curvature of the magnetization below 18 T demonstrates, as discussed in the preceding section, that the powder particles are not perfectly single crystalline. Also the linear part of the magnetization persists only up to 35 T. Above this field value, slight upward curvature of the magnetization and very small hysteresis are observed, which are very likely related to the transition observed in the aligned sample at 40 T, indicating again that the particles are not perfectly single crystalline. Figure 4(a) shows the straight-line fit to the data up to 35 T which, in accordance with relation $M = n_{\text{ErCo}}^{-1} B$, passes through the origin. This fit provides $n_{\text{ErCo}} = 1.71$ T f.u./ μ_B , which, trivially, is the same value as that obtained when we insert into Eq. (5) the values $B'_{\text{cr},1} = 18$ T and $M_{\text{Co}} - M_{\text{Er}} = 10.5 \mu_B/\text{f.u.}$, the latter value taken identical to the value of the magnetization at $B'_{\text{cr},1}$.

In the above analysis, zero Co anisotropy has been assumed which of course is an approximation. We will now discuss in which way the above determined value $n_{\text{ErCo}} = 1.71$ T f.u./ μ_B is affected by the presence of nonzero Co anisotropy. Because the value of K_1^{Co} in $\text{Er}_2\text{Co}_{17}$ is not known, we have assumed its value to be equal to K_1^{Co} in

Y_2Co_{17} , for which we have taken -9 K/f.u., an average of values reported in the literature.^{5,6,11} In the case of nonzero Co anisotropy, an additional term enters Eq. (6) which, if Er- and Co-anisotropy constants are considered up to second order, becomes^{10,12}

$$\frac{B}{M} = n_{\text{ErCo}} - \frac{2K_1^{\text{Er}}K_1^{\text{Co}} \cos \alpha}{M_{\text{Er}}M_{\text{Co}}K} \quad (7)$$

with

$$K = \sqrt{(K_1^{\text{Er}})^2 + (K_1^{\text{Co}})^2 + 2K_1^{\text{Er}}K_1^{\text{Co}} \cos 2\alpha}.$$

The expression for the first critical field ($\alpha = \pi$) then reads

$$B'_{\text{cr},1} = (n_{\text{ErCo}} + \Delta)(M_{\text{Co}} - M_{\text{Er}}) \quad (8)$$

with

$$\Delta = \frac{2K_1^{\text{Er}}K_1^{\text{Co}}}{M_{\text{Er}}M_{\text{Co}}(K_1^{\text{Er}} + K_1^{\text{Co}})}.$$

It can be seen that the presence of the additional term Δ affects the value of n_{ErCo} as determined by using Eq. (8). The value of Δ is proportional to K_1^{Co} and further depends on the ratio $K_1^{\text{Co}}/K_1^{\text{Er}}$. For $K_1^{\text{Co}} = -9$ K/f.u., $K_1^{\text{Er}} > 0$, as is the case in $\text{Er}_2\text{Co}_{17}$, and for $|K_1^{\text{Co}}/K_1^{\text{Er}}| < 0.2$, which safely can be assumed to be fulfilled, $\Delta = -0.06$ T f.u./ μ_B is found. This implies that for $K_1^{\text{Co}} = -9$ K/f.u., compared to the analysis in which zero Co anisotropy was assumed, a 3.5% larger value of n_{ErCo} , equal to 1.77 T f.u./ μ_B is found. In Fig. 4(a), also the magnetization curve calculated for $K_1^{\text{Co}} = -9$ K/f.u., $|K_1^{\text{Co}}/K_1^{\text{Er}}| < 0.2$, and $n_{\text{ErCo}} = 1.77$ T f.u./ μ_B is shown. It can be seen that, in the field region up to 35 T, the difference with the curve for $K_1^{\text{Co}} = 0$ K/f.u. is very minor, making the free-powder magnetization in this field region only suited for determination of an approximate value of n_{ErCo} and not for a determination with satisfactory precision of the Co anisotropy. At much higher fields, upon approaching the forced-ferromagnetic state at $B'_{\text{cr},2}$, the zero and nonzero Co anisotropy applied in the two calculated curves lead to distinctly different magnetization behavior. In particular, the presence of a negative K_1^{Co} causes that forced-ferromagnetic state to be reached at a higher value of $B'_{\text{cr},2}$, which is also seen in Fig. 4(b) showing that the angle α [see Fig. 3(b)] between the Er- and Co-sublattice moments closes at a higher field value. If Co anisotropy plays a role in the magnetization process, the Er-sublattice moment will no longer remain directed along the easy c direction [as depicted in Fig. 3(b)] but will deviate by an angle θ from its preferred c -axis direction (see inset in Fig. 4). Because the Er anisotropy is dominant, the angle θ remains limited to a few degrees [Fig. 4(b)].

On the basis of the above considerations regarding the analysis of the free-powder magnetization, also taking into account that the used powder particles are not perfectly crystalline or otherwise imperfect, we derive from the free-powder magnetization of $\text{Er}_2\text{Co}_{17}$ that the intersublattice molecular-field coefficient in this compound is equal to $n_{\text{ErCo}} = 1.8 \pm 0.1$ T f.u./ μ_B .

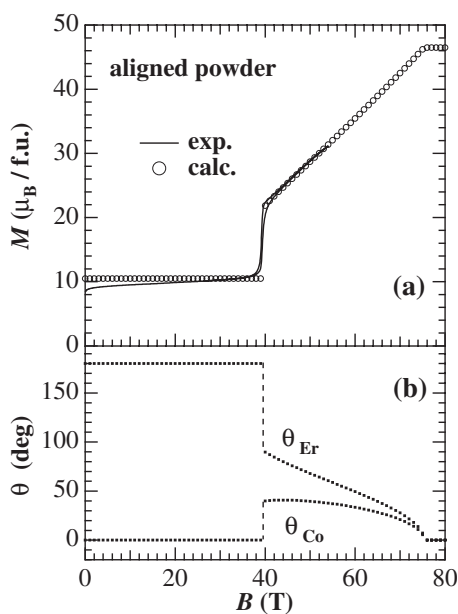


FIG. 5. Calculation of aligned-powder magnetization. (a) Magnetization of Er₂Co₁₇, as measured at 4.2 K (solid line), and as calculated (open circles). (b) Calculated field dependencies of the angles θ_{Er} and θ_{Co} of the Er- and Co-sublattice moments, respectively, with the c -axis direction [see Fig. 3(a)].

By minimizing the free energy in Eq. (1), the equilibrium directions of \vec{M}_{Er} and \vec{M}_{Co} , i.e., the angles θ_{Er} and θ_{Co} with respect to the applied-field direction, can be determined as a function of the applied field. Figure 5 shows that, using $M_{\text{Co}}=28.5\mu_B/\text{f.u.}$, $M_{\text{Er}}=18\mu_B/\text{f.u.}$, and $K_1^{\text{Co}}=-9\text{ K/f.u.}$, a very satisfactory fit to the observed magnetization curve is obtained with the following values for the free parameters, $n_{\text{ErCo}}=1.83\text{ T f.u.}/\mu_B$, $K_1^{\text{Er}}=110\text{ K/f.u.}$, and $K_2^{\text{Er}}=-30\text{ K/f.u.}$ The accuracy of n_{ErCo} is estimated to be about 2%. The n_{ErCo} value is consistent with the more approximate value $n_{\text{ErCo}}=1.8\pm 0.1\text{ T f.u.}/\mu_B$ obtained in the free-powder experiment and is also in reasonable agreement with the value of $1.91\text{ T f.u.}/\mu_B$ derived from a free-single-crystal magnetization measurement by Verhoef *et al.*^{8,9} The accuracies of the anisotropy constants K_1^{Er} and K_2^{Er} , as obtained in the fitting process, are estimated to be about 10%. The obtained value $K_1^{\text{Er}}=110\text{ K/f.u.}$ strongly deviates from the value of 225 K/f.u. obtained by Sinnema⁵ from analysis of the magnetization of a Er₂Co₁₇ single crystal measured in fields up to 35 T. Since the present knowledge of the occur-

rence of the transition at 40 T strongly reduces the range of possible K_1^{Er} values, the value of 110 K/f.u. should be considered as more realistic. For K_2^{Er} , the same value of -30 K/f.u. as reported by Sinnema⁵ has been used. For K_1^{Co} , the same value of -9 K/f.u. has been taken as used in the analysis of the free-powder magnetization.

For enabling a direct comparison of the magnetic-coupling strength between the Er and Co moments in Er₂Co₁₇ with the strengths of the coupling of R and T moments in other compounds, it is needed to transform the n_{ErCo} value into J_{ErCo} , the coupling between two nearest-neighbor spins appearing in the Heisenberg Hamiltonian. The relation between n_{ErCo} and J_{ErCo} is given by

$$Z_{\text{ErCo}}J_{\text{ErCo}} = \frac{g_{\text{Er}}}{(g_{\text{Er}} - 1)} \mu_B^2 N_{\text{Co}} n_{\text{ErCo}} \quad (9)$$

with Z_{ErCo} the number of nearest Co neighbors of an Er atom, which equals 19 in the Th₂Ni₁₇ type of structure, N_{Co} (=17) the number of Co atoms per formula unit and g_{Er} (=6/5) the Landé factor of Er. Inserting in Eq. (9) the value $n_{\text{ErCo}}=1.83\pm 0.03\text{ T f.u.}/\mu_B$, we find $J_{\text{ErCo}}/k_B=6.6\pm 0.1\text{ K}$, a slightly smaller value than the value of 6.9 K assessed for Er₂Co₁₇ by Liu *et al.*⁷

V. CONCLUSIONS

The occurrence of a field-induced transition to a noncollinear configuration is reported in a heavy-rare-earth R_2T_{17} compound with easy-axis anisotropy. The transition is observed in a magnetically aligned-powder sample of Er₂Co₁₇ and has been analyzed in a mean-field model. The parameters involved in the competing terms of the free energy, comprising the $3d-4f$ interaction, the magnetic anisotropies and the Zeeman energy, have been determined. The results obtained also illustrate that, if single crystals are not available, very satisfactory information can be obtained on magnetically aligned powder.

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