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Magnetic properties of R ions in RCo$_5$ compounds (R = Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er)

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The values of the R-Co exchange field H_{ex} and the crystalline-electric-field parameters A_n^m at the rare-earth (R) site in RCo$_5$ (R = Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er) and $\mathrm{Y}_{1-x}\mathrm{Nd}_x\mathrm{Co}_5$ (x=0.5, 0.6, 0.8, and 1.0) compounds are evaluated by a comparison of calculations with a series of experiments. The experiments provide the following data: temperature dependence of spontaneous magnetization of the compounds and magnetic moment of the rare-earth ion, magnetization curves along the crystallographic axes, and temperature dependence of the easy direction of magnetization. The anisotropies of the R-Co exchange interaction and the magnetic moment of the Co sublattice are taken into account in the calculation. The R-Co exchange field H_{ex} decreases monotonically across the rare-earth series from Pr to Er. For PrCo$_5$, the value of A_2^0 is much smaller than those of the other compounds, which suggests valence fluctuations for the Pr ion. For $\mathrm{Y}_{1-x}\mathrm{Nd}_x\mathrm{Co}_5$ and SmCo$_5$, the absolute value of A_2^0 decreases with increasing temperature.

I. INTRODUCTION

(Sm,Pr)Co$_5$ is considered an excellent material for high-coercivity magnet applications. The magnetic properties of RCo$_5$ (R, a rare earth) compounds have been studied extensively. The compounds with Y, La, Ce, Sm, Gd, and Er constituents have the c axis as the easy axis; those with Nd, Tb, and Dy have the plane normal to the c axis as the easy plane of magnetization below the spin reorientation temperature T_{SR2}, a cone spin configuration between T_{SR1} and T_{SR2}, and finally an easy c axis above T_{SR1}; the compounds of Pr and Ho have a cone spin configuration below T_{SR1} and an easy c axis above T_{SR1} (Fig. 1).

Tatsumoto *et al.*,[1] Okamoto *et al.*,[2] and Klein *et al.*[3] measured the temperature dependence of the spontaneous magnetization $M(T)$, magnetocrystalline anisotropy (MCA) constants $K(T)$, and the cone angle between the easy direction of magnetization (EDM) and the c axis, $\theta_c(T)$, for a series of RCo$_5$ compounds. Greedan *et al.*[4] showed that the magnetic anisotropy of the R ion in RCo$_5$ can be explained on the basis of a single-ion model. Bushow *et al.*[5] and Sanker *et al.*[6] evaluated the values of the exchange field H_{ex} and the crystalline-electric-field (CEF) parameters A_n^m at the Sm site in SmCo$_5$ by fitting the calculation with experimental data on $K_1(T)$. Ermolenko and collaborators[7-15] measured $M(T)$, $\theta_c(T)$, and magnetization curves along the crystallographic axes $M(H)$ for a series of RCo$_5$ and $R_{1-x}R'_x\mathrm{Co}_5$ compounds. They analyzed the magnetization curves measured at 4.2 K on the basis of classical two- or three-sublattice models. Asti *et al.*[16] and Grössinger *et al.*[17] observed the first-order magnetization process (FOMP) in PrCo$_5$, and pointed out that the sixth-order MCA energy term is responsible for the FOMP. Kelarev *et al.*,[18] by means of neutron-diffraction techniques, studied the temperature dependence of the magnetic moment of the R ion, $M_R(T)$, and that of the cone angle between the magnetic moment and the c axis, $\theta_R(T)$, for the Tb, Dy, and Ho compounds. Decrop *et al.*[19] measured the $M(T)$, $M_{\mathrm{Co}}(T)$, $M_{\mathrm{Ho}}(T)$, $\theta_c(T)$, $\theta_{\mathrm{Co}}(T)$, and $\theta_{\mathrm{Ho}}(T)$ curves for the Ho compound, and explained the experiments on the basis of a single-ion model. Alameda *et al.*[20] found the anisotropy of the magnetization in YCo$_5$. Ballou *et al.*[21,22] showed that the anisotropy of the Gd-Co exchange interaction plays an important role in characterizing the $M(H)$ curve of GdCo$_5$.

The purpose of this work is to evaluate the values of H_{ex} and A_n^m, together with M_{Co}, for a series of RCo$_5$ (R=Pr, Nd, Sm, Gd, Tb, Dy, Ho, and Er) and $\mathrm{Y}_{1-x}\mathrm{Nd}_x\mathrm{Co}_5$ (x=0.5, 0.6, 0.8, and 1.0) compounds by fitting our calculations with as much experimental data

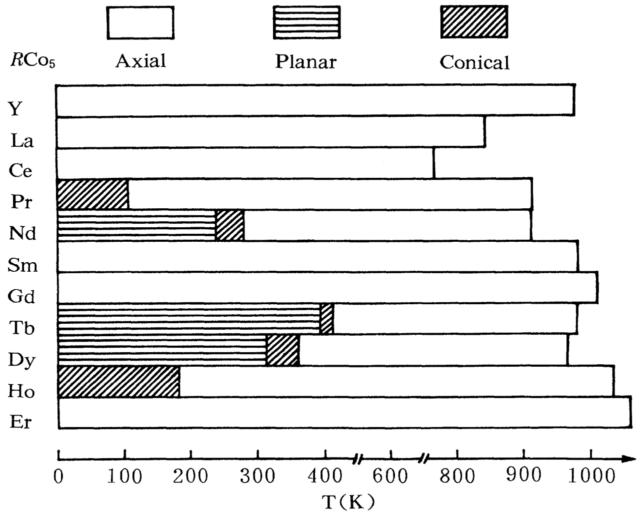

FIG. 1. Temperature variation of the magnetic structure of the RCo$_5$ series.

TABLE I. The fitted values of $2\mu_B H_{\rm ex}$ and A_n^m together with $M_{\rm Co}$ and $K_{1\rm Co}$ in the RCo$_5$ series at T=0 K.

R	$2\mu_B H_{\rm ex}$ (K)	A_2^0 (K)	A_4^0 (K)	A_6^0 (K)	A_6^6 (K)	$M_{\rm Co}$ (μ_B/f.u.)	$K_{1\rm Co}$ (K/f.u.)
Pr	1300	25	−75	250	−600	7.70	45
Nd	750	−1020	0	115	150	7.70	45
Sm	440	−330	−50	0	0	8.33	45
Gd	290					8.55	45
Tb	265	−340	−240	0	0	8.75	44
Dy	235	−425	−50	0	0	8.92	42
Ho	220	−615	−260	−30	0	9.24	37
Er	210	−350	−100	0	0	9.86	30

as available. The anisotropies of the R-Co exchange interaction and the magnetic moment of the Co sublattice are taken into account in the calculation.

II. METHOD OF CALCULATION

The RCo$_5$ compound has a CaCu$_5$-type hexagonal structure with space group $P6/mmm$. The Hamiltonian of the R ion in the compound consists of the spin-orbit coupling interaction, the CEF interaction, the R-Co exchange interaction, and Zeeman energy; that is,

$$\mathcal{H}_R=\lambda \mathbf{L}\cdot\mathbf{S}+\mathcal{H}_{\rm CEF}+2\mu_B\mathbf{S}\cdot H_{\rm ex}+\mu_B(\mathbf{L}+2\mathbf{S})\cdot H \ . \tag{1}$$

The Hamiltonian of the CEF interaction in the ⟨1000⟩ coordinate system with the z axis along the [0001] axis is formulated as

$$\mathcal{H}_{\rm CEF}=A_2^0C_2^0+A_4^0C_4^0+A_6^0C_6^0+A_6^6(C_6^6+C_6^{-6}) \ . \tag{2}$$

Here,

$$C_n^m=\sum_j[4\pi/(2n+1)]^{1/2}Y_n^m(\theta_j,\varphi_j) \ , \quad (n=2,4,6;m=0,\pm 6;|m|\le n) \ , \tag{3}$$

where $Y_n^m(\theta,\varphi)$ are the spherical harmonics, and θ_j and φ_j are the polar angles of the position vector of the jth $4f$ electron. It is assumed that the R ion is triply ionized, and that A_n^m is independent of temperature unless otherwise mentioned. The R-R exchange interaction, which is much weaker than the R-Co exchange interaction, is neglected. Both $H_{\rm ex}$ and $M_{\rm Co}$ are anisotropic, and are represented as[20–22]

$$H_{\rm ex}(\theta_{\rm Co},T)=H_{\rm ex}(T)[1-p'(T)\sin^2\theta_{\rm Co}] \ , \tag{4}$$

$$M_{\rm Co}(\theta_{\rm Co},T)=M_{\rm Co}(T)[1-p(T)\sin^2\theta_{\rm Co}] \ , \tag{5}$$

with $p'(0)=0.020$ (Refs. 21 and 22) and $p(0)=0.037$.[20] $H_{\rm ex}(T)$ is assumed to be proportional and antiparallel to $M_{\rm Co}(T)$, and the values of $M_{\rm Co}(T/T_C)/M_{\rm Co}(0)$ (T_C, the Curie temperature) and $p(T)$ are taken to be those of YCo$_5$.[10,20,23] The relation $p'(T)/p'(0)=p(T)/p(0)$ is used in the calculation. The matrix elements of Eq. (1)

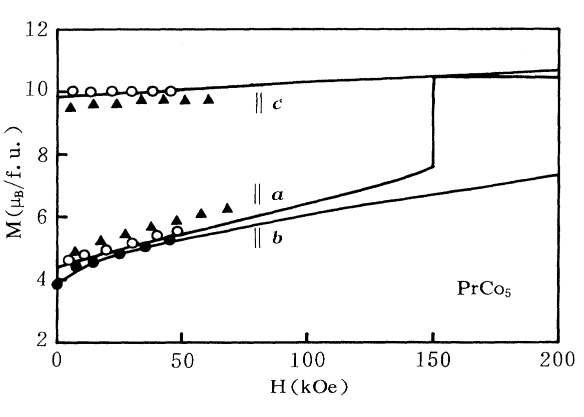

FIG. 2. Magnetization curves along the a, b, and c axes at 4.2 K for the PrCo$_5$ compound. The solid lines represent the calculations. The experimental data are taken from Ermolenko (Ref. 15) (▲) and Andoh *et al.* (Ref. 27) (○,●).

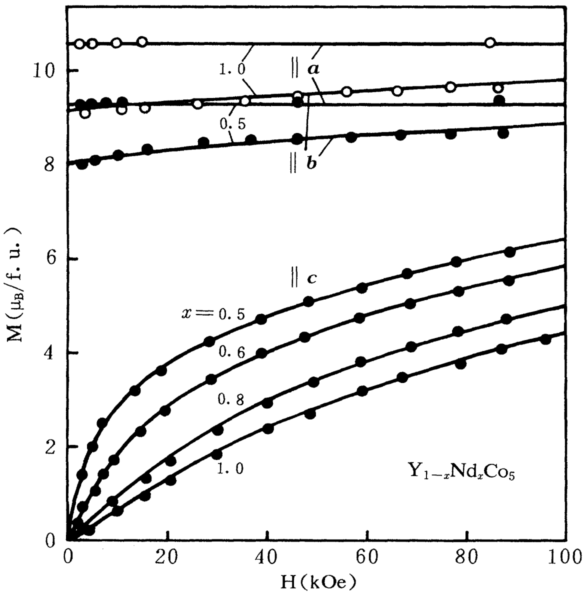

FIG. 3. Magnetization curves along the a, b, and c axes at 4.2 K for Y$_{1-x}$Nd$_x$Co$_5$ (x=0.5, 0.6, 0.8, and 1.0) compounds. The solid lines represent the calculations. The experimental data (●,○) are taken from Ermolenko (Refs. 9 and 11).

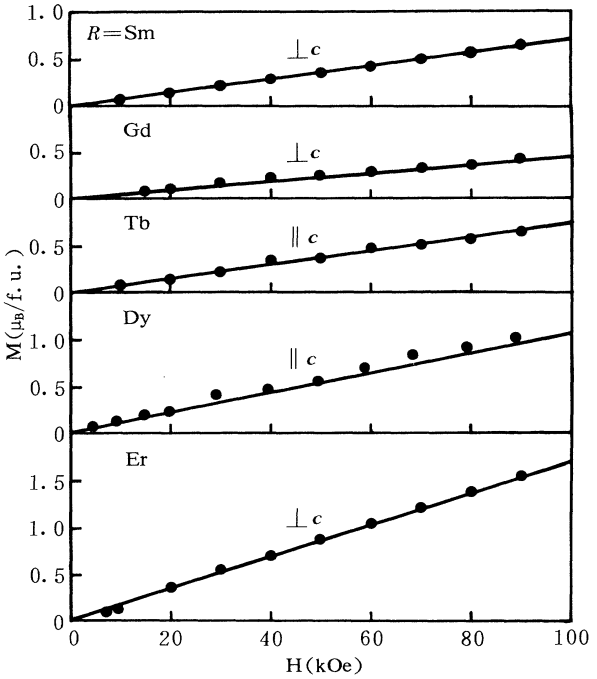

FIG. 4. Magnetization curve perpendicular to the c axis for the Sm, Gd, and Er compounds, and that parallel to the c axis for the Tb and Dy compounds at 4.2 K. The solid lines represent the calculations. The experimental data (●) are taken from Ermolenko *et al.* (Refs. 8–10, 12, and 14).

are calculated by using the irreducible-tensor-operator technique.[24] For a given applied field H and a direction of H_{ex}, the eigenvalues E_n and eigenfunctions $|n\rangle$ $[n=1,2,\ldots,\sum_J(2J+1)]$ are obtained by diagonalizing the $\sum_J(2J+1)\times\sum_J(2J+1)$ matrix of Eq. (1). The diagonalization is carried out within the subspace of the ground J multiplet for the heavy R ions, within the subspace consisting of the ground and the first excited J multiplets for the Pr and Nd ions with $\lambda=610$ and 536 K, respectively, and within the subspace consisting of the ground and the two lowest excited J multiplets for the Sm ion with $\lambda=410$ K.[25] The free energy for the $\text{Y}_{1-x}R_x\text{Co}_{5+y}$ system is given by

$$F(H,H_{\text{ex}},T)=-xk_BT\ln Z+K_{1\text{Co}}(T)\sin^2\theta_{\text{Co}}-M_{\text{Co}}(\theta_{\text{Co}},T)\cdot H\ , \tag{6}$$

where

$$Z=\sum_n \exp(-E_n/k_BT)\ , \tag{7}$$

and $K_{1\text{Co}}$ is the MCA constant of the Co sublattice. $K_{1\text{CO}}(T/T_C)/K_{1\text{CO}}(0)$ is taken as that of YCo$_5$.[20] $K_{1\text{CO}}(0)$ varies linearly with y, and its value for $y=0$ and 3.5 is that of YCo$_5$ (Ref. 20) and Y$_2$Co$_{17}$,[26] respectively. The equilibrium direction of H_{ex} is determined from minimization of the free energy. The magnetic moment of the R ion is given by

$$M_R(T)=\sum_n \mu_n\exp(-E_n/k_BT)/Z\ , \tag{8}$$

where

$$\mu_n=-\mu_B\langle n|(\mathbf{L}+2\mathbf{S})|n\rangle\ . \tag{9}$$

The magnetic moment of the $\text{Y}_{1-x}R_x\text{Co}_{5+y}$ system is

$$M(T)=xM_R(T)+M_{\text{Co}}(\theta_{\text{Co}},T)\ . \tag{10}$$

III. RESULTS AND DISCUSSION

The fitted values of $2\mu_BH_{\text{ex}}$ and A_n^m, together with M_{Co} and $K_{1\text{Co}}$ for each RCo$_5$ compound at T=0 K, are listed in Table I. Figures 2–7 and Table II present the

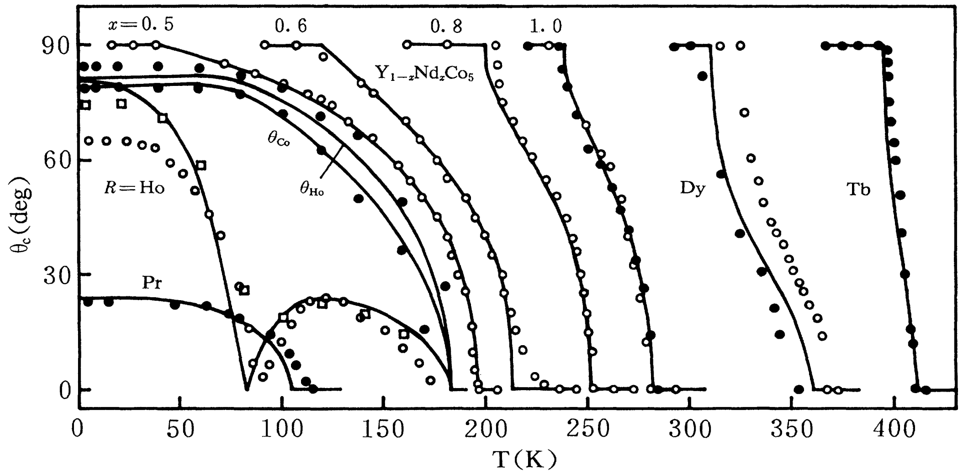

FIG. 5. Temperature dependence of θ_c for the Pr, $\text{Y}_{1-x}\text{Nd}_x$ (x=0.5, 0.6, 0.8, and 1.0), Tb, Dy, and Ho compounds, and those of θ_{Ho} and θ_{Co} for the Ho compound. The solid lines represent the calculations. The experimental data are taken from Ermolenko *et al.* (Refs. 9, 11–15) for R=Pr(●), $\text{Y}_{1-x}\text{Nd}_x$(○), Tb(●) Dy(●), and Ho(○), from Klein *et al.* (Ref. 3) for R=Nd(●), from Tsushima *et al.* (Ref. 28) for R=Dy(○), and from Decrop *et al.* (Ref. 19) for R=Ho(□,●).

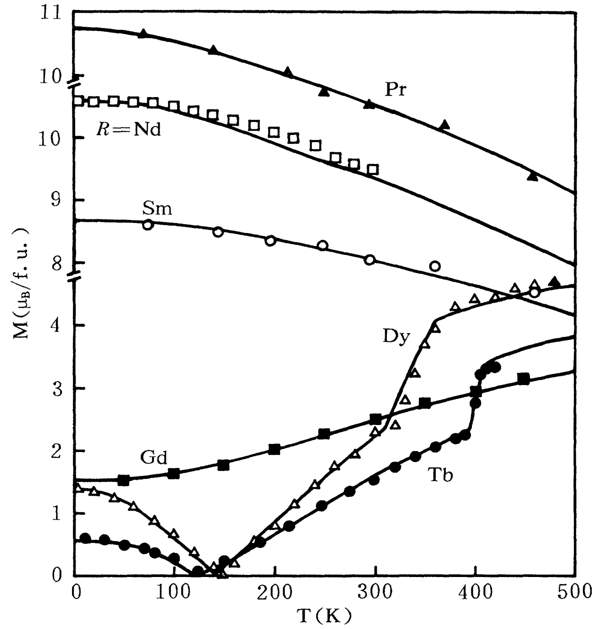

FIG. 6 Temperature dependence of the spontaneous magnetization for the Pr, Nd, Sm, Gd, Tb, and Dy compounds. The solid lines represent the calculations. The experimental data are taken from Klein *et al.* (Ref. 3) for R=Pr(▲) and Sm(○), from Ermolenko *et al.* (Refs. 10–12) for R=Nd(□), Gd(■), and Tb(●), and from Tsushima *et al.* (Ref. 28) for R=Dy(△).

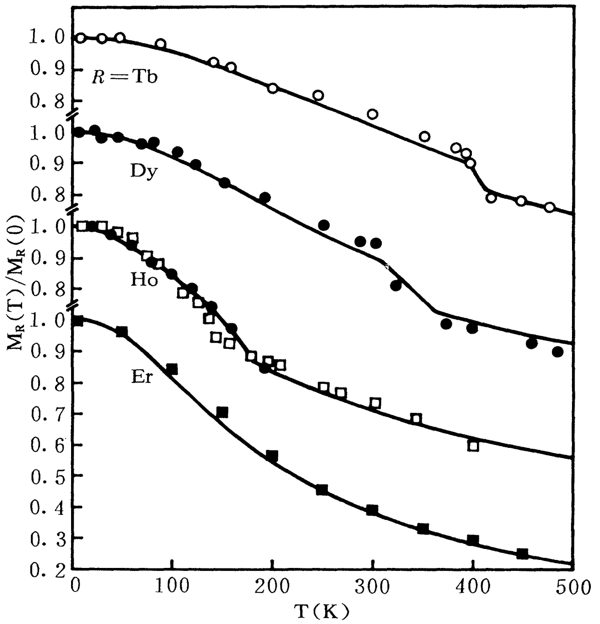

FIG. 7. Temperature dependence of the magnetic moment of the R ion in the Tb, Dy, Ho, and Er compounds. The solid lines represent the calculations. The experimental data are taken from Kelarev *et al.* (Ref. 18) for R=Tb(○), Dy(●), and Ho(□), from Decrop *et al.* (Ref. 19) for R=Ho(●), and from Ermolenko *et al.* (Ref. 14) for R=Er(■).

TABLE II. The calculated and experimental values of the spin reorientation temperatures $T_{\rm SR1}$ (K) and $T_{\rm SR2}$ (K) in the Pr, Nd, Tb, Dy, and Ho compounds.

R	$T^{\rm calc}_{\rm SR1}$	$T^{\rm calc}_{\rm SR2}$	$T^{\rm expt}_{\rm Sr1}$	$T^{\rm expt}_{\rm SR2}$	Reference
Pr	106		102		3
			107		16
			100		29
Nd	282	237	282	236	3
			280	230	11
			280	241	29
Tb	412	394	412	396	12
			414	396	18
			409	400	29
Dy	361	310	355	300	14
			367	325	28
			362	316	29
Ho	183		175		13
			181		18
			190		29

comparison of the calculations with experiment. Figures 2 and 3 show the $M(H)$ curves along the a, b, and c axes at T=4.2 K for PrCo$_5$ and Y$_{1-x}$Nd$_x$Co$_5$ (x=0.5, 0.6, 0.8, and 1.0) compounds. Figure 4 shows the $M(H)$ curve perpendicular to the c axis for the Sm, Gd, and Er compounds and parallel to the c axis for the Tb and Dy compounds at T=4.2 K. Figure 5 shows the temperature dependence of θ_c for the Pr, Y$_{1-x}$Nd$_x$ (x=0.5, 0.6, 0.8, and 1.0), Tb, Dy, and Ho compounds, and those of $\theta_{\rm Ho}$ and $\theta_{\rm Co}$ for the Ho compound. Table II presents the spin reorientation temperatures of the Pr, Nd, Tb, Dy, and Ho compounds. $M(T)$ curves for the Pr, Nd, Sm, Gd, Tb, and Dy compounds are plotted in Fig. 6; and the $M_R(T)/M_R(0)$ curves for the Tb, Dy, Ho, and Er compounds, in Fig. 7. Further comparisons of the calculations with experiment and some discussions are given below.

For the Pr compound, the cone angle θ_c at T=4.2 K is calculated to be 23.8°, which is in agreement with the experimental value of 21°–25°.[27,15,30] Our calculations also reproduce well the FOMP along the a axis at a critical field $H_{\rm cr}$=150 kOe (Refs. 16 and 17) (see Fig. 2). At the critical field, the free energy of the domain with the magnetization along the a axis becomes smaller than that of the domain that persisted below the critical field, and the former domain nucleates and grows through the domain-wall movement which characterizes the FOMP. The sixth-order CEF terms play a critical role in reproducing the FOMP. It can be seen from Table I that A_2^0 of the Pr ion has a positive sign and is much smaller than those of the other R ions. An anomalous decrease in A_2^0 has been also found in Pr$_2$Fe$_{14}$B.[31] It is suggested that the valence of the Pr ion fluctuates. The fact that the lattice constant, a, in the RCo$_5$ series drops anomalously for PrCo$_5$, as well as for CeCo$_5$, would support this interpretation.[32]

For Y$_{1-x}$Nd$_x$Co$_5$ (x=0.5, 0.6, 0.8, and 1.0) compounds, our calculations have shown that it would be impossible to reproduce the experimental $M(T)$, $M(H)$, and $\theta_c(T)$ curves if the parameter A_2^0 were independent of temperature. In fact, the $M(T)$ curve is determined dominantly by the strength of the exchange field $H_{\rm ex}$, and is affected slightly by the parameter A_2^0. The $\theta_c(T)$ curve is characterized mainly by the parameters $H_{\rm ex}$ and A_2^0. The higher-order CEF parameters affect the $\theta_c(T)$ curve in a minor way. Similarly, A_2^0 is the most important parameter in characterizing the main feature of the $M(H)$ curve. Using the value $2\mu_B H_{\rm ex}=750$ K, determined by fitting the $M(T)$ curve, the parameter A_2^0 should be assigned values of about -600 or -1000 K in order to reproduce the main characteristics of the $\theta_c(T)$ or $M(H)$ curves for NdCo$_5$. By allowing A_2^0 to vary with temperature, the temperature dependence of the parameter A_2^0 is deduced from the $\theta_c(T)$ curve for each compound as shown in Fig. 8. It should be noted that $-A_2^0(T)$, which is deduced from the fairly complicated $\theta_c(T)$ curve, decreases simply with increasing temperature. However, the rate of decrease is too fast and difficult to explain. One possible explanation is that the assumption of the Nd ion being triply ionized in the calculation is too crude. The valence of the Nd ion could fluctuate to some extent, which is also supported by the anomalous behavior in the lattice constant of NdCo$_5$.[32]

For the Sm compound, the temperature dependence of the parameter A_2^0 is also deduced by fitting the experimental data on K_1 at various temperatures. The values of $-A_2^0$ should decrease to 290 and 240 K at $T=300$ and 475 K, respectively. A similar temperature dependence of A_2^0 was also found in Pr$_2$Co$_{14}$B and Nd$_2$Co$_{14}$B; Nd$_2$Co$_{14}$B (Ref. 33). The variation to such an extent in the electric-field gradients V_{zz}, which is proportional to the parameter A_2^0, has been observed in a ^{155}Gd Mössbauer measurement on Gd$_2$Fe$_{14}$B.[34] The origin of this temperature dependence of A_2^0 is not yet understood. The magnetic moments of the Sm ion are calculated to be $0.35\mu_B$ and $0.05\mu_B$ at $T=4.2$ and 300 K, respectively, which are comparable to the experimental values of $0.38\mu_B$ and $0.04\mu_B$.[35] The fitted values $2\mu_B H_{\rm ex}=440$ K and $A_2^0=-330$ K are roughly consistent with those estimated by Bushow *et al.*[5] ($2\mu_B H_{\rm ex}=400$ K and $A_2^0=-360$ K), Sanker *et al.*[6] ($2\mu_B H_{\rm ex}=480$ K and $A_2^0=-420$ K), and Givord *et al.*[35] ($2\mu_B H_{\rm ex}=350$ K and $A_2^0=-200$ K).

For the Gd compound, the values of K_1 at $T=300$ and 500 K are calculated to be 31.6 and 33.3 K/f.u., respectively, which are in good agreement with 31.4 and 33.3 K/f.u. from experiment.[7] If p' is assumed to be independent of temperature, i.e.,

$$p'(500\ {\rm K})=p'(0\ {\rm K})=0.020\ ,$$

instead of

$$p'(500\ {\rm K})=p'(0\ {\rm K})p(500\ {\rm K})/p(0\ {\rm K})=0.030\ ,$$

the value of K_1 at $T=500$ K would be calculated to be less than 30.0 K/f.u.

For the heavy-rare-earth compounds, the experimental $M(H)$, $\theta_c(T)$, $M(T)$, and $M_R(T)$ curves shown in Figs. 4–7 can be reproduced well by assuming that A_2^0 is independent of temperature. For the Ho compound, the open circles and squares in Fig. 5 are two independent experimental results for the $\theta_c(T)$ curve as reported by different authors. According to Ermolenko *et al.*,[13] the value of cone angle θ_c, measured in an applied field, depends strongly on the strength of the field. For instance, the value of θ_c at $T=4.2$ K is 65° for $H=9$ kOe (Fig. 5, the open circles), and it is about 82° by extrapolation to $H=0$ kOe. The calculated value of θ_c at 4.2 K is 80.4°. The fitted values of $2\mu_B H_{\rm ex}$ and A_n^m (Table I) are consistent with those reported by Decrop *et al.*[19] ($2\mu_B H_{\rm ex}=200$ K, $A_2^0=-620$ K, $A_4^0=-340$ K, and $A_6^0=-50$ K), although the anisotropies of $H_{\rm ex}$ and $M_{\rm Co}$ had not been taken into account in their work.

Figure 9 shows the variation of the exchange field $2\mu_B H_{\rm ex}$ at $T=0$ K across the *R*Co$_5$ series. The value of $2\mu_B H_{\rm ex}$ decreases monotonically from Pr to Er. A simi-

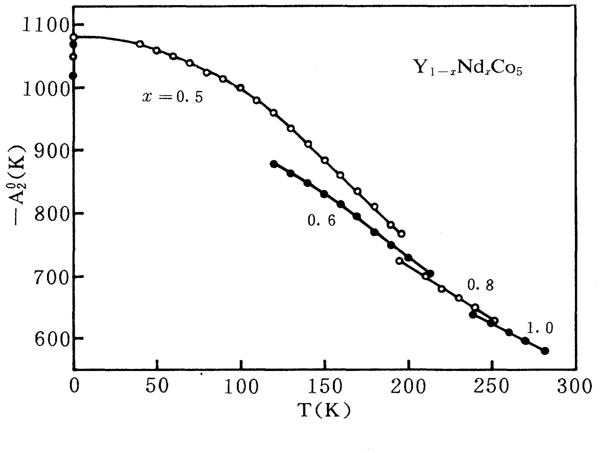

FIG. 8. Temperature dependence of the second-order CEF parameter A_2^0 in Y$_{1-x}$Nd$_x$Co$_5$ [$X=0.5$(○), 0.6(●), 0.8(○), and 1.0(●)] compounds.

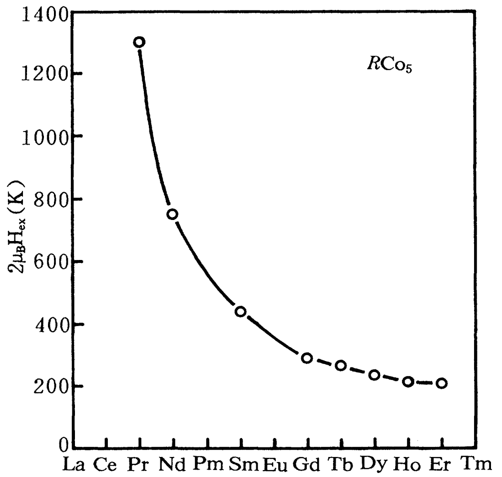

FIG. 9. Variation of the exchange field $2\mu_B H_{\rm ex}$ at $T=0$ K across the *R*Co$_5$ series.

lar behavior has been found in RNi$_5$, R_2Fe$_{14}$B, and some other rare-earth transition-metal compounds.[36,37] Such a variation in the exchange field has been attributed by Belorizky *et al.*[37] to the change of the difference between the orbital radii of the 5d and 4f electrons across a given compound series.

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