Isotropic and anisotropic magnetoelastic interactions in heavy and light RCo$_2$ Laves phase compounds

Isotropic and anisotropic magnetoelastic interactions in heavy and light RCo$_2$ Laves phase compounds

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Abstract. The thermal expansion and anisotropic magnetostriction of the RCo$_2$ Laves phases were studied in the temperature range 4–500 K using the x-ray powder diffraction method. In the heavy RCo$_2$ the magnetic moment of the itinerant d electron subsystem derived from the magnetovolume effect was found to fit well with the magnetization curve of YCo$_2$. A pronounced paraprocess above the metamagnetic transition has been observed when increasing the f–d exchange field. The type and temperature variation of the distortion of the cubic unit cell of the three compounds PrCo$_2$, NdCo$_2$ and SmCo$_2$ have been studied in detail. The corresponding magnetostriction constants λ_{111} or λ_{100} were calculated. At 4 K the following values have been obtained: PrCo$_2$, tetragonal distortion, easy axes $\langle 100 \rangle$, $\lambda_{100} = -3.4 \times 10^{-3}$; SmCo$_2$, rhombohedral distortion, easy axes $\langle 111 \rangle$, $\lambda_{111} = -4.6 \times 10^{-3}$; NdCo$_2$, tetragonal distortion and easy axes $\langle 100 \rangle$ for 42 K $< T < T_c$, orthorhombic distortion and easy axes $\langle 110 \rangle$ for $T < 42$ K with $|\lambda_{111}| = 1.9 \times 10^{-3}$ and $\lambda_{100} = -4.0 \times 10^{-3}$.

1. Introduction

Much attention is paid to the rare earth (RE) Laves phase compounds with cobalt RCo$_2$ for several reasons; the d magnetism instability and giant magnetostriction, both volume and anisotropic, which have been intensively studied during the last fifteen years [1–3]. The simple magnetic as well as crystallographic structures make possible a clear interpretation of most experiments carried out on the RCo$_2$ compounds.

The magnetic instability causes an itinerant electron metamagnetism of the d-electron subsystem in RCo$_2$, i.e. a field-induced magnetic phase transition from the paramagnetic to ferromagnetic state. In the compounds with non-magnetic rare earths, YCo$_2$ and LuCo$_2$, which are itinerant Pauli paramagnets, the transition was recently observed by direct $M(H)$ measurements at critical fields H_M equal to 69.5 T and 74 T, respectively [4]. With magnetic rare earths these compounds are collinear ferrimagnets, the itinerant d subsystem being ordered due to the f–d intersublattice exchange magnetic field H_{fd}, which exceeds H_M. A number of experiments were carried out in different substituted RCo$_2$ compounds with the aim of outlining the metamagnetic character of the d subsystem, and the results were well explained using the simple phenomenological model of itinerant metamagnetism based on the Landau theory [2,5].

Due to a large magnetovolume effect which accompanies the magnetic order of itinerant electron systems, the thermal expansion measurements serve a useful tool to study the d-electron magnetism in the RCo$_2$ compounds. These measurements are to some extent

complementary to the magnetic measurements. The value of the magnetovolume anomaly given by: $\omega_s = (V_m - V_0)/V_0$ (where V_m and V_0 denote the crystal volume in the magnetic and non-magnetic state, respectively) can be related in a first approximation to the d-electron magnetic moment μ_d by

$$\omega_s = kC\mu_d^2. \tag{1}$$

Here k is the isotropic compressibility and C is the magnetovolume coupling constant. Although equation (1) does not include a possible contribution to ω_s caused by the RE subsystem, it is generally accepted that with this expression the variation of the d magnetic moment μ_{Co} in RCo$_2$ can satisfactorily be described [2].

In heavy RCo$_2$, there are some doubts as to whether μ_{Co} takes a value around $1\mu_B$, which clearly does not coincide with the μ_{Co} value of YCo$_2$ and LuCo$_2$ deduced from magnetization measurements at fields higher than H_M (about 0.5 μ_B/Co) [4]. It seems that above the metamagnetic phase transition the compounds are still far from being saturated. The agreement of μ_{Co} data on the heavy RCo$_2$ compounds with the high-field magnetization data on YCo$_2$ is poor; therefore the data do not allow one to predict the behaviour of YCo$_2$ in the ferromagnetic state above H_M correctly [4, 6].

The RCo$_2$ compounds, as well as other cubic Laves phases RMe$_2$ (with Me = Fe, Ni and Al) exhibit an anisotropic magnetostriction within the ordered state, which at 4.2 K exceeds the value of 10^{-3} [2, 7–9]. The magnetostriction constants obtained by both magnetic and x-ray methods performed on the RFe$_2$ compounds are in good agreement [10]. Unfortunately, there are no measurements available for the field-induced deformation carried out on single crystals of RCo$_2$. This is because the preparation of RCo$_2$ single crystals large enough for this kind of experiments is very difficult.

On the other hand, x-ray measurements on powdered samples enable us to determine the anisotropic magnetostriction constants by measuring the lattice parameters in the magnetic ordered state. For a cubic crystal, the magnetostriction in any direction given by the direction cosines β_i can be expressed by

$$\lambda = (3/2)\lambda_{100} \sum_i \left(\alpha_i^2 \beta_i^2 - 1/3\right) + 3\lambda_{111} \sum_{i<j} \alpha_i \alpha_j \beta_i \beta_j \tag{2}$$

where α_i represent the direction cosines of the magnetization. Here the magnetostriction constants λ_{100} (:=λ_{001}) and λ_{111} are defined as the deformations along the $\langle 001 \rangle$ and $\langle 111 \rangle$ directions with the magnetization parallel to $\langle 001 \rangle$ and $\langle 111 \rangle$, respectively. The spontaneous magnetoelastic distortion can be connected with the magnetostriction constants by equation (2) taking α_i as the easy axis of magnetization and β_i as the direction $\langle hkl \rangle$ corresponding to the measured x-ray reflection peak. In RCo$_2$ with heavy rare earths, the magnetostriction constant λ_{111} was found to be completely determined by the RE contribution and follows well the single-ion model predictions, whereas in λ_{100} a large contribution from the Co subsystem was observed by experiments on GdCo$_2$ [11].

In the present investigation we have studied the thermal expansion of nearly all the RCo$_2$ compounds and the behaviour of the d magnetic moment as a function of the effective f–d exchange field H_{eff} has been discussed. We have extended the studies of the anisotropic magnetostriction to light RCo$_2$ with R = Pr, Nd and Sm. TmCo$_2$ which has the lowest value of T_c (about 4 K) in the RCo$_2$ series was not studied in detail until now. Therefore we also studied the behaviour of TmCo$_2$ by investigating the related pseudobinaries Tm$_{1-x}$Gd$_x$Co$_2$, in which the Gd contribution to the anisotropic magnetostriction is negligible compared to the Tm contribution.

The crystal cell parameters of polycrystalline RCo$_2$ samples were studied in the temperature range 5–500 K. A conventional Siemens D500 x-ray powder diffractometer with Co Kα radiation was used for scanning the {440} and {222} reflection peaks. For calibration Ge powder was used as internal standard. Details of the sample preparation have already been given elsewhere; therefore we do not describe them here, instead referring the reader to [2, 7].

2. Experimental results

2.1. Heavy rare-earth compounds

In figure 1 the temperature dependence of the normalized (to 300 K) cubic lattice parameter of the heavy RCo$_2$ compounds (R = Y, Gd, Tb, Dy, Ho, Er, Lu) is presented. The results are in good agreement with those obtained in earlier investigations using different experiments [2]. They clearly show the occurrence of positive magnetovolume anomalies below the corresponding Curie temperature. For comparison the $a(T)$ dependences for the two isostructural non-magnetic RCo$_2$ compounds YCo$_2$ and LuCo$_2$ are included in this figure. As can be seen, these paramagnetic compounds fit well the uniform behaviour of the whole series for $T > T_c$. The important conclusion, which can be drawn from figure 1, is that there is a clear diminution of the magnetovolume anomaly at 4 K when going from Gd towards Er.
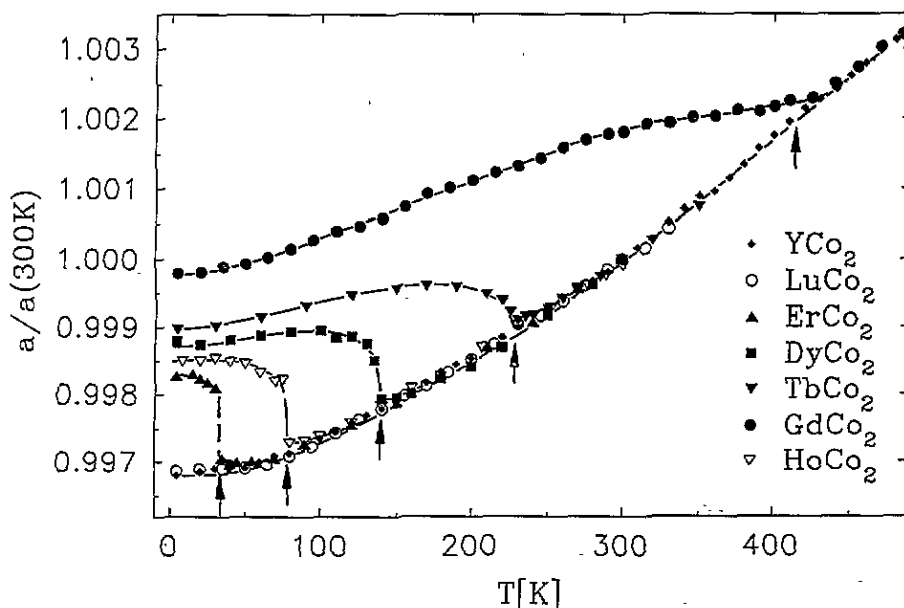

Figure 1. The normalized (to 300 K) cubic lattice parameter of the heavy RCo$_2$ compounds as a function of temperature. Below T_c the average value of $a = \sqrt[3]{V}$ (V is the unit cell volume) is drawn. The arrows indicate the Curie temperatures.

It should be noted that there is a large anisotropic deformation of the cubic unit cell in all these compounds observable which is in agreement with our previous results, both in the sign and value (for details see [2, 7]).
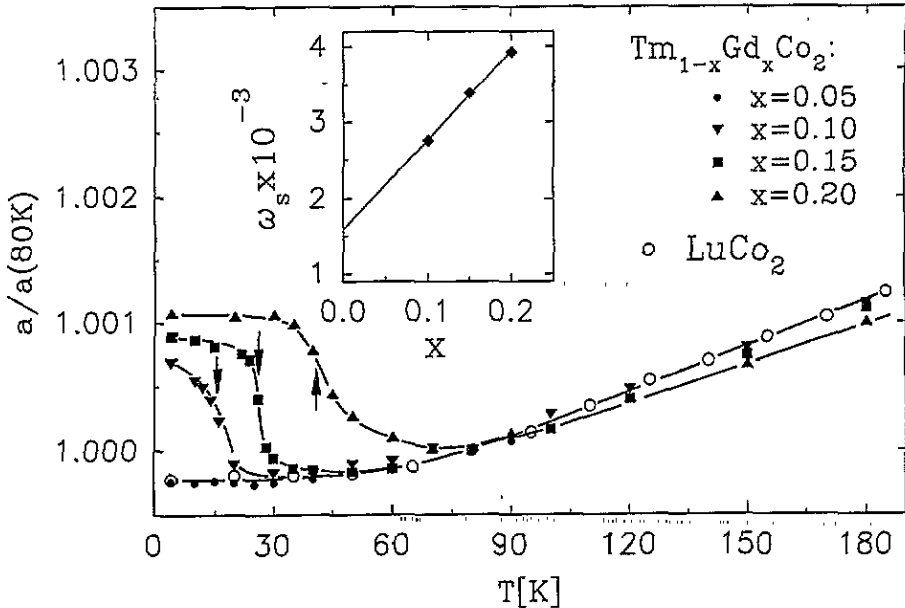
Figure 2. The normalized cubic lattice parameter of the $Tm_{1-x}Gd_xCo_2$ compounds as a function of temperature. Below T_c (arrows) the average value of $a = \sqrt[3]{V}$ (V is the unit cell volume) is drawn. The inset shows the concentration dependence of the magnetovolume effect.

In figure 2 the temperature dependence of the lattice parameters of the pseudobinary $Tm_{1-x}Gd_xCo_2$ compounds is shown. The volume anomaly existing below T_c increases with increasing Gd concentration, i.e. it exhibits a pronounced dependence on the RE atomic number. The inset in figure 2 shows the concentration dependence of ω_s for the $Tm_{1-x}Gd_xCo_2$ system at 4 K. The linear extrapolation gives, for $TmCo_2$, a value of $\omega_s = 1.6 \times 10^{-3}$. At low temperatures, when the long-range magnetic order occurs in this Tm-based compound, a rhombohedral distortion of the cubic unit cell appears, indicating the [111] direction as the easy axis of magnetization. Figure 3 shows the temperature dependence of the lattice parameters of these pseudobinary systems in the hexagonal description versus temperature. As can be seen, the degree of distortion diminishes with increasing temperature.

2.2. Light rare-earth compounds

The temperature dependence of the lattice parameters of $PrCo_2$, $NdCo_2$ and $SmCo_2$ is shown in figures 4–6. Below T_c all the compounds exhibit a large increase of the volume, however this effect is smaller than that of the heavy RCo_2 compounds (see figure 1). This can be explained by the lower values of the Co moment observed in the light RCo_2 compounds. As in the case of the heavy RCo_2 compounds, the crystal symmetry is lowered below the Curie temperature indicating large anisotropic magnetoelastic interactions. The character of the unit cell distortion is determined by the orientation of the easy axis of magnetization in all cases.

In $SmCo_2$ with the $\langle 111 \rangle$ easy axes, the distortion is rhombohedral and the value of the magnetostriction constant λ_{111} derived from equation (2) achieves -4.6×10^{-3} at 4 K. In contrast, $PrCo_2$ shows a tetragonal distortion in the ordered state in accordance with the easy
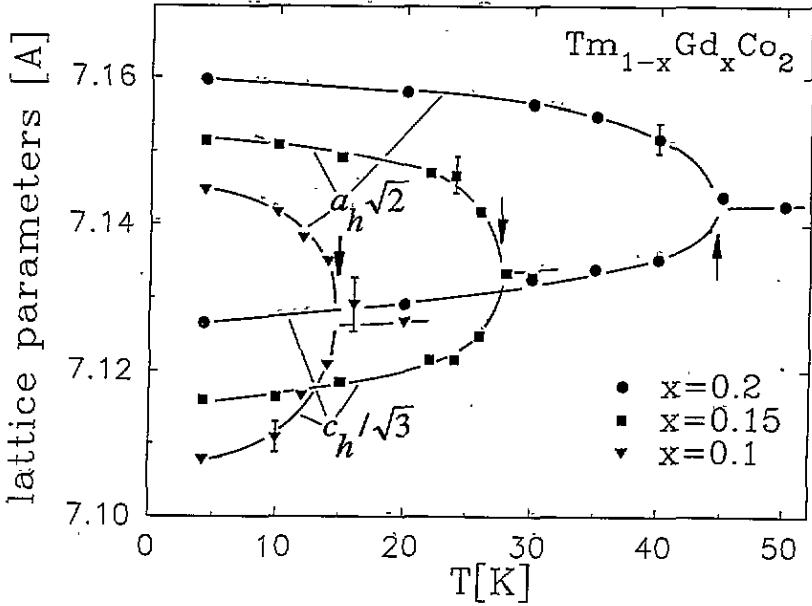
Figure 3. The temperature dependence of the lattice parameters of the distorted unit cell of $Tm_{1-x}Gd_xCo_2$ below T_c (arrows) in the hexagonal description.

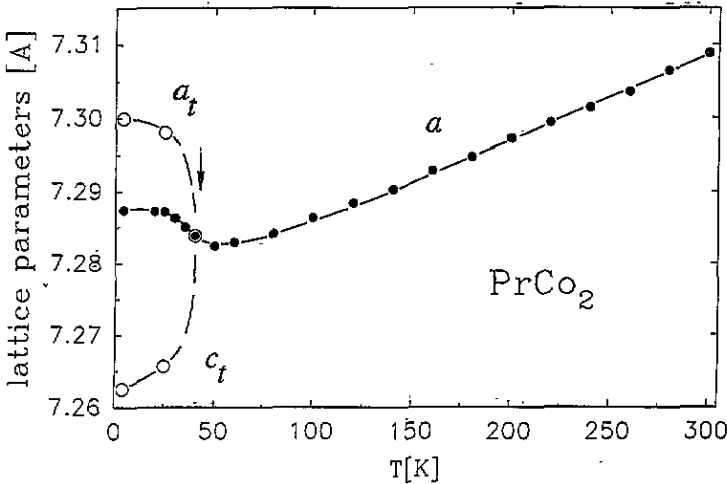**Figure 4.** The temperature dependence of the cubic (a) and tetragonal (a_t and c_t) lattice parameters of $PrCo_2$. Below T_c (arrow) the average value of $a = \sqrt[3]{V}$ is drawn (full circle).

axes $\langle 100 \rangle$. The low-temperature magnetostriction constant λ_{100} derived from equation (2) is -3.4×10^{-3}.

In $NdCo_2$, the easy axis below T_c is of the $\langle 100 \rangle$ type; however, it changes to $\langle 110 \rangle$ at about 42 K. Accordingly, we observed a tetragonal distortion in this compound in the region $42 < T < T_c$, which turns into an orthorhombic one below 42 K. In the orthorhombic phase
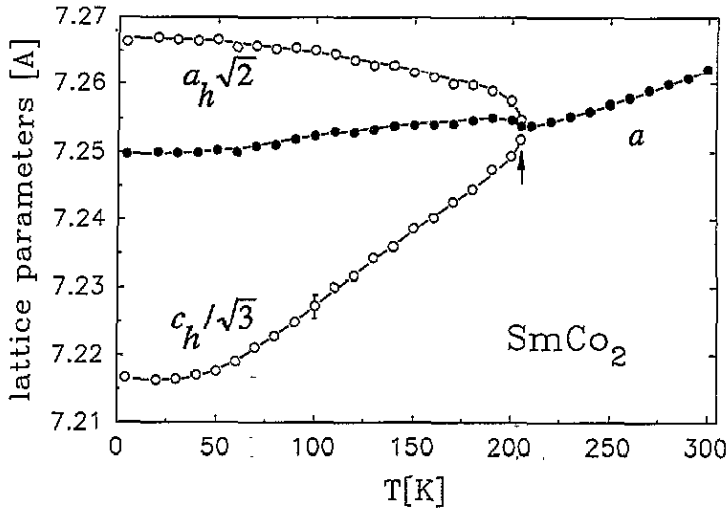
Figure 5. The temperature dependence of the cubic (a) and hexagonal (a_h and c_h) lattice parameters of SmCo$_2$. Below T_c (arrow) the average value of $a = \sqrt[3]{V}$ is drawn (full symbols).
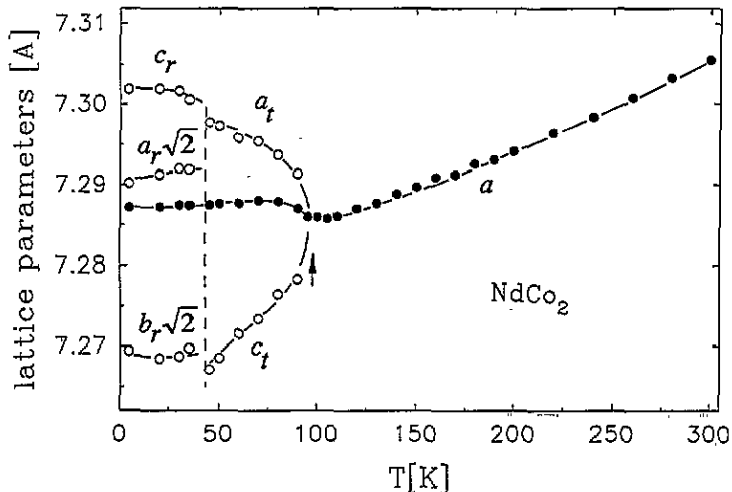

Figure 6. The temperature dependence of the cubic (a), tetragonal (a_t and c_t) and orthorhombic (a_r, b_r and c_r) lattice parameters of NdCo$_2$. Below T_c (arrow) the average value of $a = \sqrt[3]{V}$ is drawn (full symbols).

it is possible to evaluate both magnetostriction constants λ_{111} and λ_{100} from equation (2) in the form expressed by the orthorhombic lattice parameters

$$\lambda_{100} = \frac{2\sqrt{2}}{3c_r}(a_r + b_r - c_r\sqrt{2}) \qquad \lambda_{111} = \frac{2\sqrt{2}}{3c_r}(a_r - b_r). \tag{3}$$

At 4 K we found the following values: $|\lambda_{111}| = 1.9 \times 10^{-3}$ and $\lambda_{100} = -4.0 \times 10^{-3}$. However, because of the symmetrical splitting of the $[\bar{4}40]$ and $[4\bar{4}0]$ reflection peaks and their equal intensities, it was impossible to determine the sign of λ_{111}. We can assume

though that it is positive—similar to that of NdFe$_2$ [10]. Figure 7 shows the temperature variation of λ_{111} and λ_{100} for these three compounds. The discontinuity of λ_{100} of NdCo$_2$ at the spin reorientation is caused by a change of the absolute value of the magnetization, as is shown in [12].
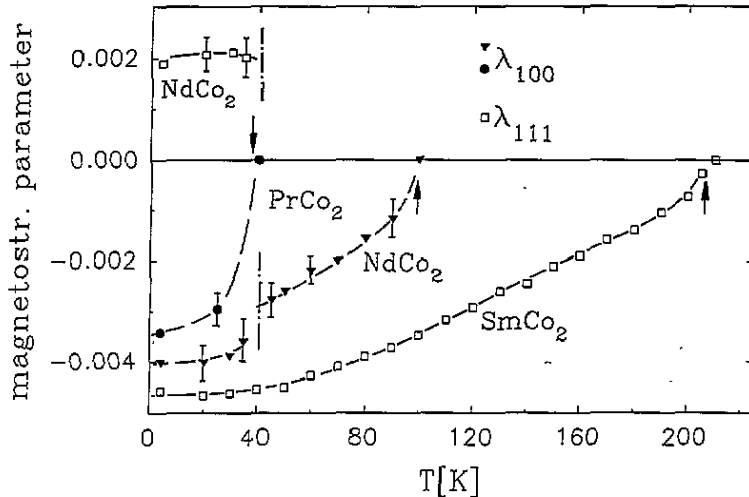

Figure 7. The anisotropic magnetostriction constants λ_{100} (full symbols) and λ_{111} (open symbols) of PrCo$_2$, NdCo$_2$ and SmCo$_2$ as a function of temperature. The arrows indicate the Curie temperatures.

3. Discussion

The results obtained from x-ray measurements enable us to determine the magnetic moment μ_{Co} of the Co atoms in the heavy RCo$_2$ compounds and its variation as a function of the f–d exchange field. As the values of μ_{Co} determined by different methods vary in a wide range between 0.7 and 1.3 μ_B [13–20], for calibration we used the magnetization data on GdCo$_2$, for which all the previously performed experiments give approximately the same value (1.03–1.07 μ_B, with $\mu_{Gd} = 7\ \mu_B$) [21–24]. Thus the magnetoelastic coefficient kC in equation (1) was taken to be equal to 8.14 $\times 10^{-3}\ \mu_B^{-2}$/Co.

The molecular field H_{fd} acting on the d-electron subsystem RCo$_2$ can be taken as $\lambda_{fd}\mu_R$. There is, however, some uncertainty in the values of λ_{fd} (or I_{fd}) given in different works, which arises from several fitting parameters used in calculations [25–27]. Here we used the recent data on the molecular field coefficients λ_{fd} found for ErCo$_2$ and GdCo$_2$ from the observations of field-induced non-collinear structures and metamagnetic transitions in ErCo$_2$ ($\lambda_{fd} = 20\ $T/$\mu_B^{-1}$) and in the Y$_{1-t}Gd_t$(Co$_{1-x}Al_x$)$_2$ compounds ($\lambda_{fd} = 64\ $T/$\mu_B$) [28, 29]. This method does not need any additional adjustable parameter and gives high accuracy for λ_{fd}. As the exchange integrals I_{fd} of these two components have proven to be equal to each other (-1.97×10^{-22} J), we used the same value for the whole series with heavy rare earths from Gd to Er, and taken μ_R to be gJ. However, smaller values can be observed mainly in Tm-based compounds [30, 31]. For the pseudobinary Tm$_{1-x}$Gd$_x$Co$_2$ compounds we calculated H_{fd} using the simple molecular field approach

$$H_{fd} = (1 - x)\lambda_{TmCo}\mu_{Tm} + x\lambda_{GdCo}\mu_{Gd}. \tag{4}$$

The values of μ_{Co} versus H_{fd} thus obtained are depicted in figure 8 together with the magnetization curve of YCo_2 measured under ultra-high pulsed magnetic fields [4]. For comparison μ_{Co} from single-crystal magnetization data on $ErCo_2$, $HoCo_2$ and $TbCo_2$ [16, 17, 20] are included in that figure. The x-ray data correspond well with the direct magnetization measurements on YCo_2. As can be seen, the $M(H)$ curve of YCo_2 shows a large paraprocess contribution. We refer the progressive increase of μ_{Co} for the RCo_2 compounds towards $GdCo_2$ also to that paraprocess.
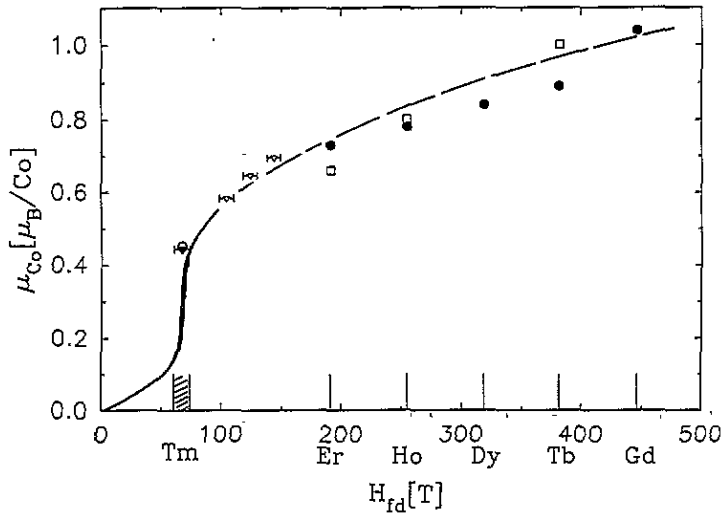

Figure 8. The variation of the d-electron magnetic moment μ_{Co} versus H_{fd} derived from x-ray powder diffraction data of RCo_2 (full circles) and $Tm_{1-x}Gd_xCo_2$ (open triangles). The full triangle shows the extrapolated value for $TmCo_2$, the open circle is the value for $Tm(Co_{0.88}Al_{0.12})_2$ [27]. The open squares represent single-crystal magnetization data taken from the literature [16, 17, 20]. The solid line represents the metamagnetic transition in the magnetization curve of YCo_2 [4]. The broken line is drawn as a guide for the eyes.

Note that μ_{Co} for $TmCo_2$ evaluated from the extrapolated value of ω_s (see the inset in figure 2) should be considered as the upper limit, i.e. its real value cannot exceed $0.44\mu_B$. The open circle in that figure shows μ_{Co} derived from magnetization measurements [30] of the $Tm(Co_{0.88}Al_{0.12})_2$ compound in which the d magnetic moment is stabilized by the Al substitution. This value is in good agreement with the estimated μ_{Co} for $TmCo_2$. The comparison with the magnetization curve of YCo_2 clearly shows that the molecular field H_{fd} in $TmCo_2$ is very close to the critical field of the metamagnetic transition. It should be noted that in equation (1) the spin fluctuation contribution to ω_s is neglected; furthermore the molecular field approximation used for the calculation of H_{fd} is also an essential simplification. In our opinion the observed agreement between x-ray and magnetic data is due to a temperature variation of the paramagnetic spin fluctuation amplitude which is constant for the whole heavy RCo_2 series. This allowed us to derive ω_s from the normalized $a(T)$ dependences without using the $a(T)$ behaviour of non-magnetic isostructural compounds.

Now we consider the anisotropic magnetoelastic properties of the studied compounds. For heavy RCo_2 it was shown that the magnetoelastic distortion along the $\langle 111 \rangle$ axes is entirely determined by the rare-earth ions, i.e. the Co contribution is negligible [2]. The

rhombohedral distortion observed in the $Tm_{1-x}Gd_xCo_2$ systems (the Gd^{3+} ions are in the S state with $L = 0$), should be due to the Tm contribution. Below T_c the exchange interaction tends to align the strongly asymmetrical 4f shells of the Tm^{3+} ions (via L–S coupling) giving rise to an anisotropic lattice response. We can estimate λ_{111} for $TmCo_2$ by a linear extrapolation of λ_{111} of the Gd substituted compounds towards $x = 0$. This procedure (figure 9) gives a value of $\lambda_{111} = -4.2 \times 10^{-3}$ for $TmCo_2$.
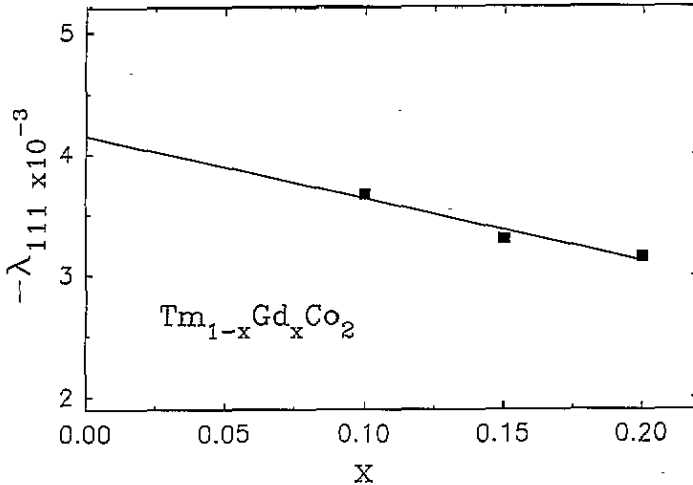

Figure 9. The concentration dependence of λ_{111} of the $Tm_{1-x}Gd_xCo_2$ compounds at 4 K.

In the scope of the single-ion model one can represent the magnetostrictive constants of a cubic crystal by the elastic (C_{11}, C_{12}, C_{44}) and magnetoelastic (B_1, B_2) coupling constants in the form

$$\lambda_{100} = \frac{2}{\sqrt{6}} \frac{B_1}{C_{11} - C_{12}} \langle O_2^0 \rangle_T \qquad z - \text{axis} \parallel M \parallel \langle 001 \rangle$$

$$\lambda_{111} = \frac{B_2}{4C_{44}} \langle O_2^0 \rangle_T \qquad z - \text{axis} \parallel M \parallel \langle 111 \rangle. \tag{5}$$

Here $\langle O_2^0 \rangle_T$ is the thermal averaged Stevens operator O_2^0. Considering B_i to be proportional to $\langle r_{4f}^2 \rangle \alpha_J$ and assuming $\langle O_2^0 \rangle_{0\,K} = \langle M_j = J | O_2^0 | M_J = J \rangle = J(2J - 1)$ (saturated value) one can express the magnetostriction constants in the form [32]

$$\lambda_{100, 111} = D_{1,2} \langle r_{4f}^2 \rangle \alpha_J J(J - \tfrac{1}{2}) \tag{6}$$

where D_1, D_2 are coefficients that are approximately the same for all RCo_2.

Equation (6) was successfully used in [2] for the analysis of λ_{111} in the heavy RCo_2 compounds. Its variation with RE was found to follow the single-ion prediction. The sign changes from positive to negative between $HoCo_2$ and $ErCo_2$ due to the change of the sign of the Stevens coefficient α_J. Although for $TmCo_2$ the crystal field splitting of the Tm^{3+} ion levels cannot be considered as a small effect compared to the exchange field, one can see that the estimated value for $TmCo_2$ is in good agreement (with respect to its sign and value) with the single-ion prediction for the heavy RCo_2 compounds according to equation (6) [2].

The same limitation for $TmCo_2$ holds for the light RCo_2 compounds, in which the ground state also cannot be considered to be $|M_J = J\rangle$. There the situation is even more complex because of mixing effects from the higher J multiplets. Note that the easy axes in $PrCo_2$, $NdCo_2$ and $SmCo_2$ do not coincide with those of their RFe_2 analogues in the low-temperature region; $NdCo_2$ and $SmFe_2$ exhibit spin reorientation phase transitions. However, some qualitative comparisons are worthwhile. Also, in the case of the light RCo_2 λ_{111} seems to follow the single-ion model better than λ_{100}, $\lambda_{111}(SmCo_2)/\lambda_{111}(NdCo_2) = -2.4$ (-2.8 according to formula (6)) and $\lambda_{100}(PrCo_2)/\lambda_{100}(NdCo_2) = 0.85$ (3.07 according to formula (6)). The ratio for λ_{111} is close to that for $SmFe_2$ ($\lambda_{111} = -4.9 \times 10^{-3}$ [33]) and $NdFe_2$ ($\lambda_{111} = 2.0 \times 10^{-3}$) [10]. One can assume that in RCo_2 the contribution from higher multiplets of the RE^{3+} ions to λ_{111} is smaller than that to λ_{100}. On the other hand, the Co contribution to λ_{100} can also cause the agreement of λ_{100} to be less good than that of λ_{111}.

4. Conclusion

The magnetovolume effect of the heavy RCo_2 compounds follows well the regularities in the variation of the magnetic moment of the itinerant d subsystem, μ_{Co}, including its metamagnetic behaviour. The magnetization curve for YCo_2 fits the data derived from the x-ray measurements, thus confirming the large paraprocess of M_d after the metamagnetic transition has occurred.

Like the heavy RCo_2 compounds, the light RCo_2 Laves phases also exhibit a large anisotropic magnetostriction at low temperature, which causes a lattice distortion according to the easy axis direction. The magnitude and character of the distortions allow us to attribute the anisotropic magnetoelastic interaction to the single-ion mechanism of the crystal field effect on the R^{3+} ion.

Acknowledgments

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