

Magnetization measurement of the Co moment induced by the molecular field in $Y_{1-t}Gd_tCo_3$

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1994 J. Phys.: Condens. Matter 6 11119

(<http://iopscience.iop.org/0953-8984/6/50/019>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.179

The article was downloaded on 13/05/2010 at 11:35

Please note that [terms and conditions apply](#).

Magnetization measurement of the Co moment induced by the molecular field in $Y_{1-t}Gd_tCo_3$

H Aruga Katori†, T Goto†, I Yu Gaidukova†, R Z Levitin‡, A S Markosyan‡, I S Dubenko§ and A Yu Sokolov§

† Institute for Solid State Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo 106, Japan

‡ Physics Department, M V Lomonosov Moscow State University, 119899 Moscow, Russia

§ Moscow Institute of Radio Engineering Electronics and Automation, 117454 Moscow, Russia

Received 17 June 1994, in final form 6 September 1994

Abstract. The magnetization of polycrystalline $Y_{1-t}Gd_tCo_3$ with $0 \leq t \leq 1$ has been measured up to 40 T, and the effect of the molecular field from the Gd sublattice on the itinerant metamagnetism of the Co sublattice is studied. The compounds with $t < 0.25$ show a field-induced metamagnetic transition of the Co sublattice. A transition from the collinear magnetic structure to the non-collinear structure is observed in compounds with $0.50 \leq t \leq 0.55$. The spontaneous Co moment, which is evaluated from the magnetization curve, exhibits a stepwise increase at $t \simeq 0.25$ when the Gd concentration is increased. This means that the metamagnetic transition of the Co sublattice is induced by the molecular field from the Gd sublattice in $Y_{1-t}Gd_tCo_3$. The average intersublattice molecular-field coefficient λ_{GdCo} between the Gd and Co sublattices is estimated to be $\lambda_{GdCo} = 40.9 \pm 0.4$ T formula units/ μ_B ; the corresponding exchange coupling parameter J_{GdCo} is $J_{GdCo} = -(1.64 \pm 0.03) \times 10^{-22}$ J.

1. Introduction

It is well known that the spontaneous Co moment in Y–Co compounds depends on the Y content in formula unit (FU). As the molar fraction of Y increases, the spontaneous Co moment decreases and vanishes in YCo_2 . YCo_3 has the smallest spontaneous Co moment in the Co-rich Y–Co compounds except for YCo_2 . However, an increment in the Co moment in YCo_3 is induced by a magnetic field. The magnetization of YCo_3 has been measured in ultra-high magnetic fields up to 110 T and enhancement of the Co moment has been observed [1]. Successive field-induced transitions occur at $H_{M1} = 60$ T and $H_{M2} = 82$ T. After the second transition, the Co moment is almost saturated at $1.22\mu_B/Co$.

The value of the Co moment in RCo_3 compounds, where R is a magnetic rare earth, depends on the molecular field from the R sublattice and it is larger than in YCo_3 [2, 3]. When Y in YCo_3 is replaced by R atoms, therefore, the value of the Co moment is expected to depend on the R concentration. In the present study, we focus on the magnetization of $Y_{1-t}Gd_tCo_3$ in order to investigate the enhancement of the Co moment due to the molecular field from the R sublattice. The reasons why we chose Gd as the R atom are as follows: the anisotropy of the Gd^{3+} ion is negligible, the 4f shell is half-filled and there is no orbital contribution to the magnetic moment.

RCo_3 compounds have a rhombohedral crystal structure of the $PuNi_3$ type with space group $R\bar{3}m$. This structure can be regarded as a consecutive arrangement of the alternating

RCO_5 and RCO_2 structure blocks along the c axis in the ratio 1:2. The R atoms occupy two non-equivalent sites of $\text{R}_\text{I}(3a)$ and $\text{R}_\text{II}(6c)$, whereas the Co atoms have three sites, namely $\text{Co}_\text{I}(3b)$, $\text{Co}_\text{II}(6c)$ and $\text{Co}_\text{III}(18h)$.

YCo_3 is a collinear ferromagnet. The easy direction of magnetization is along the c axis [4]. The anisotropy field is estimated to be 8.0 T [4]. The value of the spontaneous Co moment in YCo_3 depends slightly on the stoichiometry; the average Co moments observed from the experimental results vary from $0.5\mu_\text{B}$ [4–6] to $0.6\mu_\text{B}$ [1]. On the other hand, GdCo_3 is a collinear ferrimagnet with a uniaxial anisotropy along the c axis [7]. Because of the large molecular field acting on the Co sublattice, the Co moment in GdCo_3 has a value of about $1.2\mu_\text{B}/\text{Co}$ [3, 7, 8].

We have measured the high-field magnetization of polycrystalline $\text{Y}_{1-t}\text{Gd}_t\text{Co}_3$ with $0 \leq t \leq 1$. From the magnetization of each compound, we have obtained the t -dependence of the spontaneous Co moment. We have also evaluated the intersublattice molecular-field coefficient λ_{GdCo} between the Gd and the Co sublattices, and the exchange coupling parameter J_{GdCo} .

2. Experimental procedure

Polycrystalline samples of the $\text{Y}_{1-t}\text{Gd}_t\text{Co}_3$ system were prepared by induction melting the constituents in an argon atmosphere with a water-cooled copper crucible under quasi-levitation conditions. The ingots were annealed at 1050 °C for 24 h in vacuum for homogenization. X-ray diffraction analysis indicates that the main phase in samples has the PuNi_3 -type crystal structure but some samples contain a small amount (about 3%) of impurities.

Magnetization measurements were performed at 4.2 K in pulsed magnetic fields up to 40 T, which were produced by a wire-wound magnet with a duration time of 12 ms. The magnetization was measured for powdered samples using an induction method with well balanced pick-up coils.

3. Experimental results

The magnetization of polycrystalline $\text{Y}_{1-t}\text{Gd}_t\text{Co}_3$ is shown in figures 1–3. For each compound a rapid increment in magnetization is found in weak external fields. When the external field exceeds the anisotropy field, both the Co and the Gd moments become parallel or antiparallel to the external field and the magnetization increases linearly. In this section, we explain the characteristics of the magnetization curves in detail.

The magnetization curves of $\text{Y}_{1-t}\text{Gd}_t\text{Co}_3$ with $0.00 \leq t \leq 0.25$ are shown in figure 1. A metamagnetic transition is observed below 40 T for $0.15 \leq t < 0.25$. It corresponds to the transition at $H_{\text{M}2} = 82$ T in YCo_3 . We determine the transition field $H_{\text{M}2}(t)$ as the field where the derivative dM/dH of magnetization with respect to the field has a maximum. $H_{\text{M}2}(t)$ decreases with increase in t .

Figure 2 shows the magnetization curves of $\text{Y}_{1-t}\text{Gd}_t\text{Co}_3$ with $0.30 \leq t \leq 1.00$. No metamagnetic transition appears for these compounds. However, we observed a rapid increase in the magnetization at high fields for the compounds with $t = 0.40$ and 0.60 . We measured the magnetization of the compounds with $t \simeq 0.5$ in more detail and the results are shown in figure 3. The magnetization curve of each compound indicates the occurrence of the transition from the collinear magnetic structure to the non-collinear structure. The

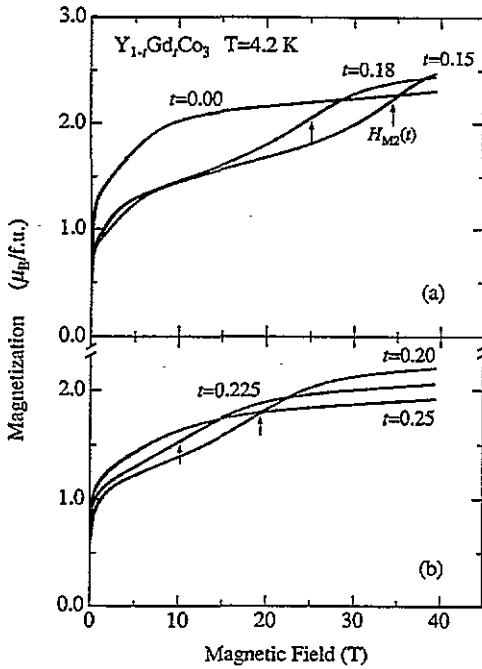


Figure 1. Magnetization of $Y_{1-t}Gd_tCo_3$ with $0.00 \leq t \leq 0.25$. The arrows indicate the metamagnetic transition field $H_{M2}(t)$.

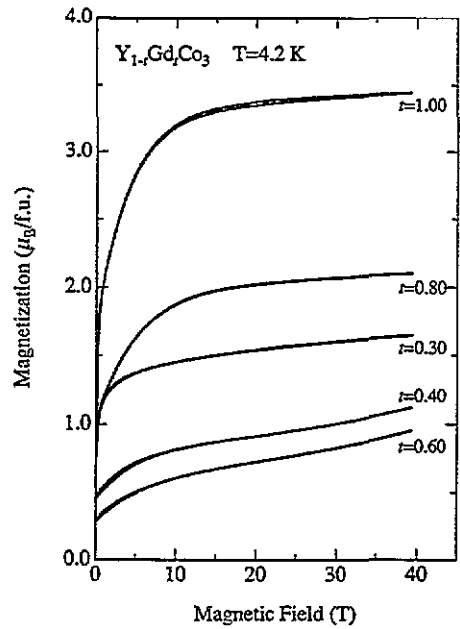


Figure 2. Magnetization of $Y_{1-t}Gd_tCo_3$ with $t = 0.30, 0.40, 0.60, 0.80$ and 1.00 .

magnetization in the collinear phase is not constant but increases with increasing field, since in the powdered samples there are magnetic moments whose easy directions are not parallel to the external field and some of them start to cant below the transition field. The magnetization curves in the collinear phase and in the non-collinear phase are fitted by two straight lines, and the critical field $H_c(t)$ is determined as their intersection.

We summarize the t -dependence of $H_{M2}(t)$ in figure 4. $H_{M2}(t)$ decreases linearly with the increase in t . The critical concentration t_{c2} at which the metamagnetic transition vanishes is estimated as 0.26. This figure indicates that the molecular field acting on the Co sublattice from the Gd sublattice increases in proportion to t and exceeds 82 T at t_{c2} .

We estimate the spontaneous magnetization $M_S(t)$ at zero field by extrapolation of the linear part of magnetization appearing in fields lower than $H_{M2}(t)$ or $H_c(t)$. We show the t -dependence of $M_S(t)$ in figure 5. $M_S(t)$ exhibits a stepwise increase at $t \simeq 0.25$. Since we expect the Gd moment M_{Gd} in $Y_{1-t}Gd_tCo_3$ to be independent of t , it is attributed to the discontinuous t -dependence of the Co moment $M_{Co}(t)$. Above $t \simeq 0.25$, $M_S(t)$ decreases and becomes zero at $t_{comp} \simeq 0.52$, which is a ferrimagnetic compensation point at 4.2 K, and then increases for $t > t_{comp}$. For $t < t_{comp}$ the Co sublattice magnetization $3M_{Co}(t)$ in formula units is larger than the Gd magnetization tM_{Gd} , whereas, for $t > t_{comp}$ $3M_{Co}(t)$ is smaller than tM_{Gd} . Therefore, $M_S(t)$ can be expressed as

$$M_S(t) = \begin{cases} 3M_{Co}(t) - tM_{Gd} & \text{for } \begin{cases} t < t_{comp} \\ t > t_{comp}. \end{cases} \end{cases} \quad (1a)$$

$$\begin{cases} t < t_{comp} \\ t > t_{comp}. \end{cases} \quad (1b)$$

From figure 5 we find that the dependence of $M_S(t)$ on t for $t > t_{comp}$ is well described by $M_S(t) = 7.00t - 3.67$. This means that, for $t > t_{comp}$, both $M_{Co}(t)$ and M_{Gd} are independent

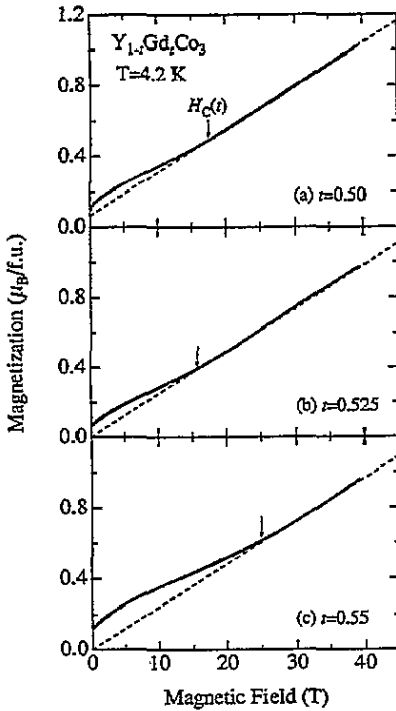


Figure 3. Magnetization of $Y_{1-t}Gd_tCo_3$ with $t = 0.50, 0.525$ and 0.55 . The arrows indicate the transition field $H_C(t)$: ---, linear extrapolations of the magnetization curve above $H_C(t)$.

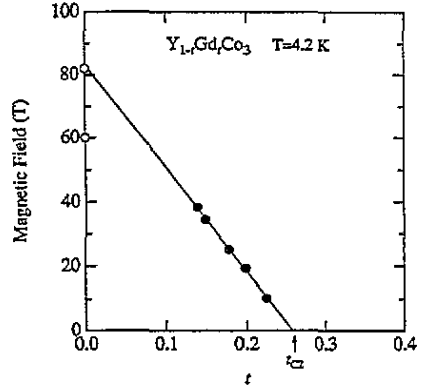


Figure 4. Gd concentration dependence of the transition field $H_{M2}(t)$ (●): ○, metamagnetic transition fields determined by the previous ultra-high-field magnetization measurement; —, result of fitting to equation (5).

of t , $M_{Co}(t) = 1.22\mu_B$ and $M_{Gd} = 7.00\mu_B$. This value of M_{Gd} is in good agreement with the moment of Gd^{3+} with a ground state given by the Hund rules. In figure 6, we show the values of $M_{Co}(t)$ estimated by using equation (1) with $M_{Gd} = 7.0\mu_B$.

4. Discussion

4.1. The magnetic moment of the Co sublattice

From the present investigation, we found that substitution of Gd for Y in YCo_3 leads to enhancement of the spontaneous Co moment, which exhibits a stepwise increase at $t \simeq 0.25$ as shown in figure 6. Such a stepwise increase in the Co moment is also found in YCo_3 when an external field is imposed [1]. It should be noted that the increment of the Co moment at $t \simeq 0.25$, which is estimated to be $0.27\mu_B/Co$, is almost the same as the corresponding increment at H_{M2} in the magnetization of YCo_3 . These results reveal that the metamagnetic transition of the Co sublattice induced by a magnetic field also occurs in $Y_{1-t}Gd_tCo_3$ owing to the molecular field from the Gd sublattice. The concentration $t \simeq 0.25$ agrees with t_{c2} . This means that the strong magnetic state of the Co sublattice, which is realized above $H_{M2}(t)$ for $t < t_{c2}$, is stabilized even without an external field for $t > t_{c2}$.

In the magnetization of YCo_3 a broad transition at H_{M1} is observed and the increment in the Co moment due to this transition is about a half of the increment at H_{M2} . In

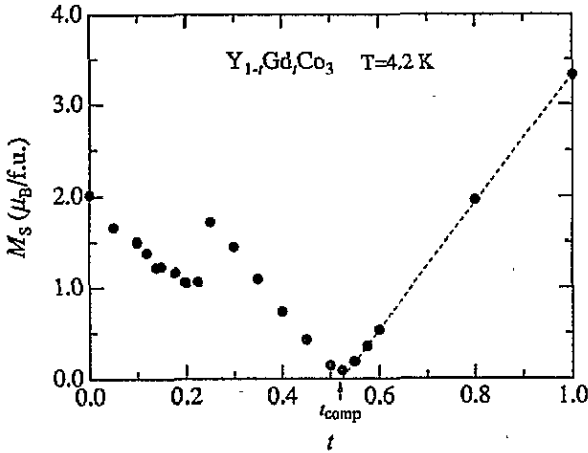


Figure 5. Gd concentration dependence of the spontaneous magnetization $M_S(t)$: ----, result of fitting to equation (1b).

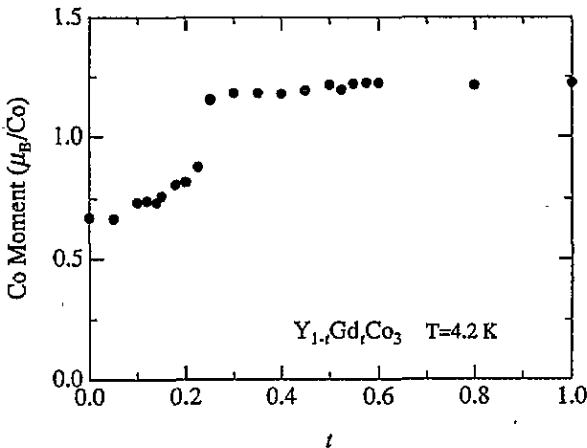


Figure 6. Gd concentration dependence of the spontaneous Co moment $M_{Co}(t)$, which is obtained using equation (1).

the magnetization of $Y_{1-t}Gd_tCo_3$, however, the transition at $H_{M2}(t)$ becomes broad as t increases owing to the inhomogeneous molecular field from the Gd sublattice and the transition corresponding to that at H_{M1} was not observed. We cannot observe a clear increase in the Co moment corresponding to the metamagnetic transition at H_{M1} in figure 6.

We summarize the above results as a field versus Gd concentration phase diagram in figure 7. The transition field from the collinear phase to the non-collinear phase for $t = t_{comp}$ is estimated at 10 T. If the Co sublattice magnetization is equal to the Gd magnetization, the transition field should be zero, which will be explained in the next section. We consider that the anisotropy of the Co sublattice increases the transition field.

4.2. The intersublattice exchange interaction between the Gd and Co sublattices

High-field magnetization measurements give useful information about the intersublattice

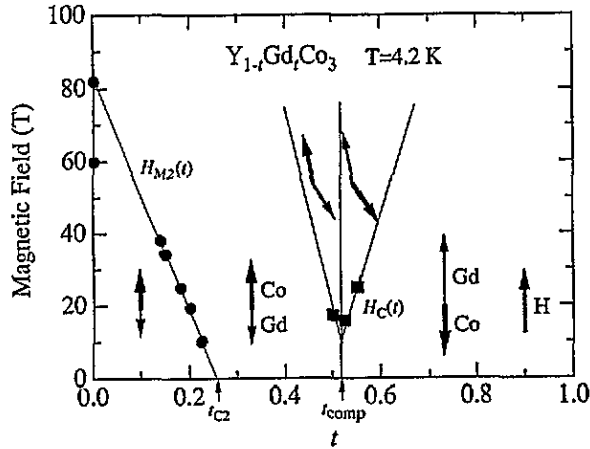


Figure 7. Field versus Gd concentration phase diagram of $Y_{1-t}Gd_tCo_3$: ●, metamagnetic transition field; ■, transition field from the collinear magnetic structure to the non-collinear structure; —, guide to the eye. The schematic magnetic configurations are indicated. The lengths of the arrows indicate the relative magnitudes of the Co and Gd moments.

exchange interaction. In the intermetallic compounds consisting of a heavy rare earth and a 3d transition metal, there exists a two-sublattice collinear ferrimagnetic state in which the magnetization M_d of the itinerant d subsystem is antiparallel to the magnetization M_f of the localized f subsystem. The field dependence of the magnetization of the two-sublattice ferrimagnet has been analysed on the basis of the molecular-field approximation where the effect of the magnetocrystalline anisotropy is neglected [9]. If the external magnetic field is applied to the ferrimagnetic state, a transition from the collinear magnetic structure to the non-collinear structure occurs at a certain field H_c given by

$$H_c = \lambda_{fd} |M_f - M_d| \quad (2)$$

where λ_{fd} is an intersublattice molecular-field coefficient of the f-d exchange interaction. In the non-collinear phase, the total magnetization $M(H)$ parallel to the external magnetic field H is proportional to H :

$$M(H) = H/\lambda_{fd}. \quad (3)$$

In $Y_{1-t}Gd_tCo_3$, $H_c(t) < 40$ T for $0.50 \leq t \leq 0.55$ and we can estimate the value of the coefficient λ_{GdCo} using equations (2) and (3). Figure 3 shows that the zero-field extrapolation of the magnetization curve in the non-collinear phase passes through the origin for the compounds with $t = 0.525$ and 0.55 . The compound with $t = 0.50$ may contain some magnetic impurities. In order to eliminate the impurity effect, we shift the magnetization curve so that it has a zero-field extrapolation point at the origin. We show H_c as a function of the magnetization $M(H_c)$, at H_c , in figure 8. H_c has a linear relation with $M(H_c)$ and we obtain $\lambda_{GdCo} = 40.5$ T FU/ μ_B from equation (2), if we assume that the value $|M_f - M_d|$ is given by $M(H_c)$. On the other hand, using equation (3) we can estimate the value of λ_{GdCo} from the slopes of the magnetization curves in the non-collinear phase: $\lambda_{GdCo} = 40.9$ T FU/ μ_B , 40.7 T FU/ μ_B and 41.5 T FU/ μ_B for the compounds with

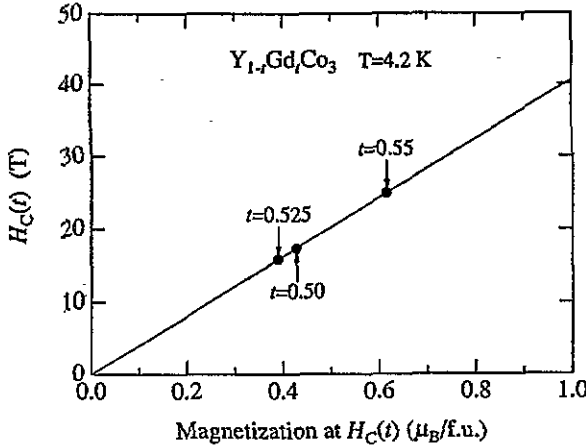


Figure 8. Critical field $H_c(t)$ as a function of the magnetization at $H_c(t)$: —, result of fitting to equation (2).

$t = 0.50, 0.525$ and 0.55 , respectively. Two different methods give almost the same values of λ_{GdCo} .

The value of λ_{fd} can be estimated also from the metamagnetic transition field. In RCo_3 compounds, the Co sublattice is subject to an effective field H_{eff} which is a sum of the external field H and the molecular field H_{mol} :

$$H_{eff} = H + H_{mol} \tag{4}$$

where $H_{mol} = \lambda_{fd}tM_f$. For $Y_{1-t}Gd_tCo_3$ with $t < t_{c2}$, therefore, the decrease in $H_{M2}(t)$ relates to the increase in the molecular field acting on the Co sublattice and $H_{M2}(t)$ can be written as

$$H_{M2}(t) = H_{M2}(0) - \lambda_{GdCo}tM_{Gd}. \tag{5}$$

Figure 4 shows that equation (5) holds for $Y_{1-t}Gd_tCo_3$. By using the value of $M_{Gd} = 7.00\mu_B$, we obtain $\lambda_{GdCo} = 45.3 \text{ T FU}/\mu_B$. This value is larger than the values obtained using equations (2) and (3). A neutron diffraction study on YCo_3 reveals that the spontaneous Co moment depends on the sites: $0.73\mu_B, 0.94\mu_B$ and $0.47\mu_B$ for the 3b, 6c and 18h sites, respectively [10]. On the basis of this result, it has been proposed that the Co moments on the 3b and 6c sites exhibit a metamagnetic transition at H_{M1} and the Co moment on the 18h site at H_{M2} [1]. We conclude that λ_{GdCo} averaged over three sites is $40.9 \pm 0.4 \text{ T FU}/\mu_B$ and λ_{GdCo} for the 18h site is $45.3 \text{ T FU}/\mu_B$.

If only the neighbouring interactions are taken into account, λ_{GdCo} is given by using a Gd-Co exchange coupling parameter J_{GdCo} as

$$\lambda_{GdCo} = -J_{GdCo}Z_{GdCo}(g_{Gd} - 1)/g_{Gd}\mu_B^2 \tag{6}$$

where $g_{Gd} = 2$ is the g -factor of the Gd ion and Z_{GdCo} is the number of nearest-neighbour Co atoms of the Gd atom. For $GdCo_3$, Z_{GdCo} averaged over three Co sites is $\frac{14}{3}$ and Z_{GdCo} for the 18h Co site is 5. We obtain $J_{GdCo} = -(1.63 \pm 0.01) \times 10^{-22} \text{ J}$ by using $\lambda_{GdCo} = 40.9 \pm 0.4 \text{ T}$

FU/μ_B and $Z_{GdCo} = \frac{14}{3}$, and $J_{GdCo} = -1.68 \times 10^{-22}$ J by $\lambda_{GdCo} = 45.3$ T FU/μ_B and $Z_{GdCo} = 5$. We conclude that $J_{GdCo} = -(1.64 \pm 0.03) \times 10^{-22}$ J.

The f-d exchange interactions are estimated for various intermetallic compounds consisting of magnetic rare earth and 3d transition elements by paramagnetic susceptibility, magnetization, inelastic neutron scattering measurements and so on. One of the direct methods providing reliable data is a magnetization measurement in a high magnetic field. In this paper we have estimated the exchange coupling parameter in $GdCo_3$ for the first time on the basis of magnetization measurements. From the previous analysis of paramagnetic susceptibility of $GdCo_3$ it has been estimated that $J_{GdCo} = -1.75 \times 10^{-22}$ J [11]. Our present result is consistent with this value. J_{GdCo} was also obtained from the results of the magnetization measurements for $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$, which is a RCO_2 type; $J_{GdCo} = -1.98 \times 10^{-22}$ J [12] and $-(1.78 \pm 0.06) \times 10^{-22}$ J [13]. These results imply that J_{GdCo} hardly depends on whether the structure is RCO_3 type or RCO_2 type. It is reasonable because the RCO_3 structure consists of the consecutive arrangement of RCO_5 and RCO_2 structure blocks.

The R-dependence of J_{RCO} has been well studied for RCO_2 . The results deduced from the paramagnetic susceptibility measurements shows that the values of J_{RCO} in light-rare-earth RCO_2 compounds are larger than those in heavy-rare-earth compounds [14]. On the other hand, the values of J_{RCO} in RCO_3 -type compounds have been evaluated only for $NdCo_3$ and $GdCo_3$ so far. From the Nd concentration dependence of two metamagnetic transition fields in $Y_{1-x}Nd_xCo_3$, J_{NdCo} was estimated to be 2.0×10^{-22} J [15, 16]. Since this value is greater than the present results for $GdCo_3$, we expect that light-rare-earth compounds have larger values of J_{RCO} than heavy-rare-earth compounds not only for RCO_2 but also for RCO_3 . Further experiments are required to verify this conjecture.

Acknowledgments

This work was partially supported by the Russian Fundamental Research Foundation under project 93-02-2006. One of the authors (RZL) is grateful to Yamada Science Foundation for financial support for a stay in Japan.

References

- [1] Goto T, Aruga Katori H, Sakakibara T and Yamaguchi M 1992 *Physica B* **177** 255
- [2] Bartashevich M I, Goto T and Yamaguchi M 1992 *J. Magn. Magn. Mater.* **111** 83
- [3] Bartashevich M I, Goto T, Yamaguchi M, Yamamoto I and Sugaya F 1993 *Physica B* **190** 315
- [4] Bartashevich M I, Goto T, Yamaguchi M, Yamamoto I and Andreev A V 1992 *Solid State Commun.* **82** 201
- [5] Burzo E and Seitabla D 1981 *Solid State Commun.* **37** 663
- [6] Shcherbakova Ye V, Yermolenko A S and Korolev A V 1986 *Phys. Met. Metallorg. (USSR)* **62** 77
- [7] Katayama T and Shibata T 1981 *J. Magn. Magn. Mater.* **23** 173
- [8] Burzo E 1972 *Phys. Rev. B* **6** 2882
- [9] Clark A E and Callen E 1968 *J. Appl. Phys.* **39** 5972
- [10] Tasset F 1975 *Thesis* Grenoble University
- [11] Duc N H, Hien T D and Givord D 1992 *J. Magn. Magn. Mater.* **104-7** 1344
- [12] Ballou R, Gamishidze Z M, Lemaire R, Levitin R Z, Markosyan A S and Snegirev V V 1993 *J. Magn. Magn. Mater.* **118** 159
- [13] Goto T, Aruga Katori H, Kouki K, Levitin R Z, Markosyan A S and Gamishidze Z M 1994 *Physica B* **201** 131
- [14] Duc N H, Hien T D, Brommer P E and Franse J J M 1992 *J. Magn. Magn. Mater.* **104-7** 1252
- [15] Goto T, Kouki K, Bartashevich M I, Aruga Katori H, Yamaguchi M, Yamamoto I and Sugaya F 1994 *Physica B* **193** 10
- [16] Kouki K, Bartashevich M I, Goto T, Aruga Katori H and Yamaguchi M 1994 *Physica B* **201** 143