

Influence of the f - d exchange interaction on the magnetic state of the itinerant d subsystem and field-induced magnetic phase transitions in the intermetallic compounds $Y_{1-t}Gd_tCo_3$

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The magnetization of the system $Y_{1-t}Gd_tCo_3$ ($0 \leq t \leq 1$) in pulsed magnetic fields up to 40 T was investigated. The dependence of the magnetic moment of the cobalt subsystem on the gadolinium concentration was determined experimentally. An anomalous increase of $M_{Co}(t)$ was observed at $t_2 \approx 0.24$ and possibly also at $t_1 \approx 0.1$. For compounds with $t < 0.225$ the magnetization undergoes a diffuse field-induced metamagnetic transition. The peculiarities of the concentration and field dependences of the magnetization can be explained on the basis of a model that takes into account the magnetic instability of the itinerant d subsystem of cobalt. They are determined by the transition of the cobalt subsystem in an external field (or the internal f - d exchange interaction field) from a weakly magnetic state into an intermediate and strongly magnetic state. The molecular-field f - d exchange interaction constants were determined. The H -versus- t magnetic phase diagrams of the systems $Y_{1-t}Gd_tCo_3$ and $Y_{1-t}R_tCo_3$ with heavy rare earths are constructed. © 1994 American Institute of Physics.

1. INTRODUCTION

The peculiarities of the band structure of magnetic materials based on $3d$ metals often result in magnetic instability—a transition from one magnetic state to another accompanying a change of the external parameters (external magnetic field, pressure, and temperature) and internal parameters (exchange field, stoichiometry, and so on). The most carefully studied manifestation of magnetic instability is itinerant metamagnetism—a transition in a magnetic field from a paramagnetic or weakly ferromagnetic state into a strongly ferromagnetic state in compounds based on YCo_2 , $LuCo_2$, and also $ThCo_5$ (a review is given Ref. 1, where the original works are cited).

Magnetic instability was recently observed in the intermetallic YCo_3 , which has a $PuNi_3$ -type orthorhombic structure (space group $R\bar{3}m$).² The magnetic properties of the compound YCo_3 and the isostructural rare-earth compounds RCo_3 are summarized in Ref. 3. The compound YCo_3 is an itinerant ferromagnet, in which the d band is formed as a result of hybridization of the $3d$ electrons of cobalt with the $4d$ electrons of yttrium (or $5d$ electrons of the rare earth in the compounds RCo_3). According to data given in different works, the Curie temperature of this compound ranges from 264 to 320 K, and the total magnetic moment ranges from $1.2 \mu_B$ to $1.75 \mu_B$ per formula unit (FU).³ This large spread in the values of the basic magnetic characteristics could be caused by stoichiometric differences between the samples studied in different experiments.

Investigations of the magnetization of YCo_3 in super-strong magnetic fields of up to 110 T have revealed two metamagnetic transitions, as a result of which the magnetic moment M_{Co} of the d subsystem of cobalt increases to $3.69 \mu_B/FU$, in fields of 60 T and 82 T.⁴ These magnetic transitions are interpreted as transitions, occurring in a magnetic field, of the magnetically unstable itinerant cobalt subsystem from a weakly magnetic state into an intermediate and then a strongly magnetic state. Band-structure calculations confirm the magnetic instability of the d subsystem in YCo_3 .⁵

There is another possibility for observing metamagnetic transitions in YCo_3 -type compounds. This possibility is associated with the effect of the effective f - d exchange interaction, arising when a magnetic rare-earth R is substituted for nonmagnetic yttrium, on the itinerant magnetic d subsystem of the compounds $Y_{1-t}R_tCo_3$. In such compounds a second magnetic subsystem, formed by the localized $4f$ electrons of the rare earth, coexists with the itinerant magnetic d subsystem. The molecular field exerted on the d subsystem by the f subsystem is proportional to the concentration t of the rare earth:

$$H_{mol}^d = \lambda_{f-d} t \mu_R, \quad (1)$$

where μ_R is the magnetic moment of the rare earth and λ_{f-d} is the molecular-field f - d exchange interaction constant. Therefore, by varying the concentration t of the rare earth the itinerant subsystem can be transferred from a weakly magnetic state into a strongly magnetic state and anomalies re-

sulting from the metamagnetic character of the magnetization of this subsystem can be revealed in the concentration dependence of the magnetization of this subsystem.

Substituting a rare earth for yttrium should also change the external fields H in which metamagnetic transitions occur, since the effective field acting on the d subsystem depends on the concentration of the rare earth:

$$H_{\text{eff}}^d = |\lambda_{f-d} t \mu_R + \mathbf{H}|. \quad (2)$$

Anomalies appearing in the concentration dependence of the spontaneous magnetization of the system $Y_{1-t}Nd_tCo_3$ as a result of a transition of cobalt from one magnetic state into another were observed in Ref. 6. The metamagnetic-transition fields in this system also depend on the neodymium concentration.

In the present work we investigated the magnetic characteristics of intermetallides of the system $Y_{1-t}Gd_tCo_3$. In $GdCo_3$ the magnetic moment of the cobalt subsystem is $4\mu_B/FU$,³ i.e., it is considerably larger than in YCo_3 , and it is greater than the magnetization of YCo_3 , even in fields of 100 T. Hence it can be concluded that the cobalt subsystem in $GdCo_3$ is in a strongly ferromagnetic state.

Gadolinium was chosen as the substituent primarily because it is in a S state. Its orbital angular momentum is zero, and therefore the effect of the crystal field on the magnetic characteristics of the rare-earth subsystem can be ignored. In particular, the magnetic moment of this subsystem can be determined without any difficulty: $\mu_{Gd} = 7\mu_B/\text{atom}$.

Our best present understanding is that the molecular-field $f-d$ exchange interaction constant can be represented in the form

$$\lambda_{f-d} \sim I_{f-d} \frac{\mu_R^s}{\mu_R}, \quad (3)$$

where μ_R^s and μ_R are, respectively, the spin moment and the total moment of the rare earth, and I_{f-d} is the $f-d$ spin exchange constant, which, to a first approximation, is the same for all heavy rare earths. The effective field acting on the d subsystem is found to be strongest for gadolinium, i.e., by substituting gadolinium for yttrium it is possible to investigate the behavior of the cobalt subsystem in a much wider range of effective fields than in the previously studied system $Y_{1-t}Nd_tCo_3$. This is the second reason for studying the system $Y_{1-t}Gd_tCo_3$.

Finally, the third reason for choosing gadolinium is that, in contrast to the system $Y_{1-t}Nd_tCo_3$, where the Nd and Co magnetic moments are parallel to one another, in $Y_{1-t}Gd_tCo_3$ and Gd and Co magnetic moments are antiparallel ($\lambda_{f-d} < 0$), i.e., these compounds are ferrimagnetic, and a noncollinear magnetic structure should arise in them in fields $H_{c1} < H < H_{c2}$.

In the exchange approximation

$$H_{c1} = \lambda_{f-d} |M_{Co} - t \mu_{Gd}|, \quad (4)$$

$$H_{c2} = \lambda_{f-d} (M_{Co} + t \mu_{Gd}),$$

and the magnetic susceptibility in the noncollinear phase is

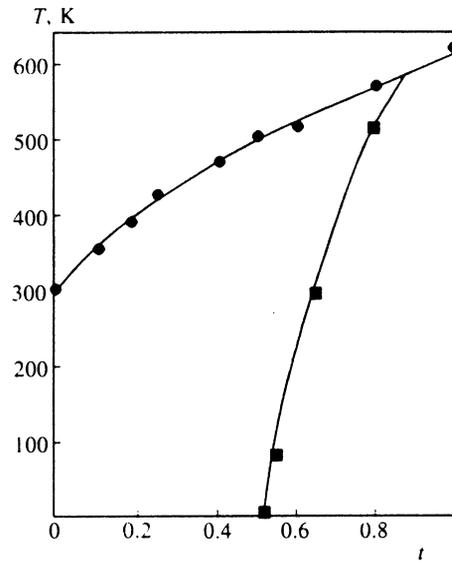


FIG. 1. Concentration dependence of the Curie temperature T_C (●) and the magnetic compensation temperature T_{comp} (■) of the compounds $Y_{1-t}Gd_tCo_3$.

$$X = \frac{1}{\lambda_{f-d}}. \quad (5)$$

Investigations of the field-induced noncollinear structure make it possible to determine independently the molecular-field $f-d$ interaction constant λ_{f-d} and to compare its value to that obtained from measurements of the metamagnetic phase transitions in the d subsystem in the substituted compounds $Y_{1-t}Gd_tCo_3$.

2. SAMPLES AND MEASUREMENT PROCEDURE

Polycrystalline samples of the intermetallides $Y_{1-t}Gd_tCo_3$ were produced from the corresponding metals in an induction furnace (using an argon atmosphere) under conditions of quasilevitation of the melt. The samples were annealed for 24 h in the dynamic vacuum regime at a temperature of 1050 °C. X-ray structural analysis revealed traces of an impurity phase, in addition to the main $PuNi_3$ -type phase, in the synthesized samples. Magnetic measurements showed, however, that the contribution of this phase to the magnetization is small (less than $0.1\mu_B/FU$; see discussion below).

The magnetization was measured by the induction method in pulsed magnetic fields of up to 40 T at 4.2 K and up to 25 T in the temperature range 4.2–300 K. The Curie temperatures were determined according to the magnetic-susceptibility anomalies in weak alternating fields.

3. EXPERIMENTAL RESULTS

Figure 1 displays the T -versus- t phase diagram of the system $Y_{1-t}Gd_tCo_3$. It is obvious that the Curie temperature T_C increases with the gadolinium concentration: linearly for high gadolinium content t and nonlinearly small t . We note that our values of T_C for YCo_3 and $GdCo_3$ are close to the

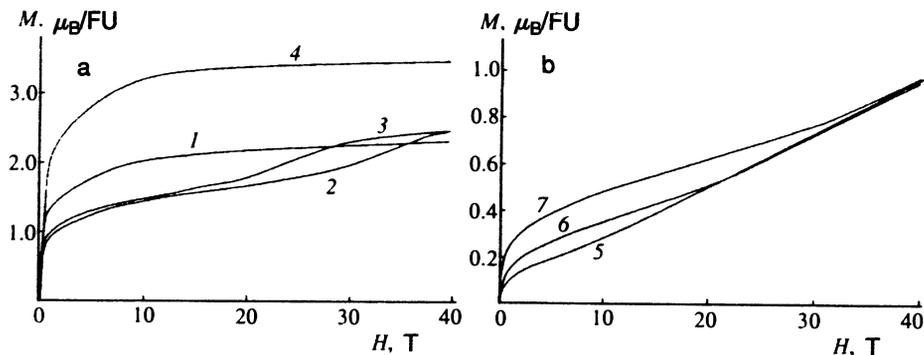


FIG. 2. $Y_{1-t}Gd_tCo_3$ magnetization curve at 4.2 K for some characteristic compositions: a) $t=0$ (1), 0.15 (2), 0.2 (3), and 1 (4); b) $t=0.525$ (5), 0.55 (6), and 0.575 (7).

published values.³ The figure also displays values of the magnetic-compensation temperature T_{comp} , at which the magnetization of the d magnetic subsystem equals that of the f magnetic subsystems. It is obvious that magnetic compensation is observed only over a comparatively narrow concentration range ($0.5 \leq t \leq 0.8$)¹. Hence it follows that the temperature dependence of the magnetization of the f magnetic subsystem is virtually identical to that of the magnetization of the d magnetic subsystem.

Figures 2a and b display magnetization curves for some characteristic $Y_{1-t}Gd_tCo_3$ compounds at 4.2 K. In fields of up to approximately 150 kOe magnetization occurs in a non-linear manner. This is explained by the large magnetic anisotropy of cobalt in these compounds, as shown previously on the basis of measurements of the magnetization of YCo_3 and $GdCo_3$ single crystals.^{3,7} The magnetization of most samples increases linearly in stronger fields and the increase is determined by the paraprocess.

It is interesting that anomalies are observed in magnetization curves of some of the mixed compounds in strong fields. For example, the magnetization of compounds with a low gadolinium concentration ($t \leq 0.225$) undergoes a diffuse metamagnetic transition (Fig. 2a). The metamagnetic transition field H_{m2} as a function of the gadolinium concentration is displayed in Fig. 3.

For compounds close to the concentration compensation point ($t_{comp} = 0.525$), the magnetization increases almost linearly in fields above a critical value (Fig. 2b). This is characteristic of a transition into a noncollinear ferrimagnetic phase.

Figure 4 (curve 1) displays the concentration dependence of the saturation magnetization M_s of the system $Y_{1-t}Gd_tCo_3$ obtained at 4.2 K by linear extrapolation of the strong-field magnetization to zero field. This curve contains two features: a small magnetization anomaly near $t \approx 0.10$ and a large, sharp increase at $t \approx 0.24$. The features observed in the concentration dependences $M_s(t)$ at 4.2 K are also present at higher temperatures. This can be seen in Fig. 5, which displays the curve $M_s(t)$ at 83 K.

The magnetic susceptibility of the compounds studied at 4.2 K in fields above the magnetic saturation field (susceptibility of the paraprocess) is shown in Fig. 4b. The susceptibility is obviously a nonmonotonic function of the gadolinium concentration. Two peaks of the magnetic susceptibility are observed: one large and asymmetric peak at

$t=0.225$ (this peak could be two peaks which have merged) and a second, wider but smaller peak, at $t=0.5$, i.e., near the compensation point.

4. DISCUSSION

In the collinear ferrimagnetic ordering model the spontaneous magnetization of the compounds $Y_{1-t}Gd_tCo_3$ can be represented in the form

$$M_s = \pm M_{Co} \mp t \mu_{Gd}, \quad (6)$$

where the upper signs refer to the concentration range $t < t_{comp}$ and the lower signs refer to the range $t > t_{comp}$. Since at 4.2 K $\mu_{Gd} = 7 \mu_B$, we constructed the concentration dependence of the d -subsystem magnetic moment M_{Co} (Fig. 4a, curve 2). This curve shows that the total magnetic moment of the d subsystem is a nonmonotonic function of the concentration and it increases near $t_1 \approx 0.1$ and $t_2 \approx 0.24$. Thus, our investigations show that the metamagnetic behavior observed in the itinerant d subsystem of the compound YCo_3 in superstrong magnetic fields⁴ also remains when this subsystem is magnetized by the effective f - d exchange interac-

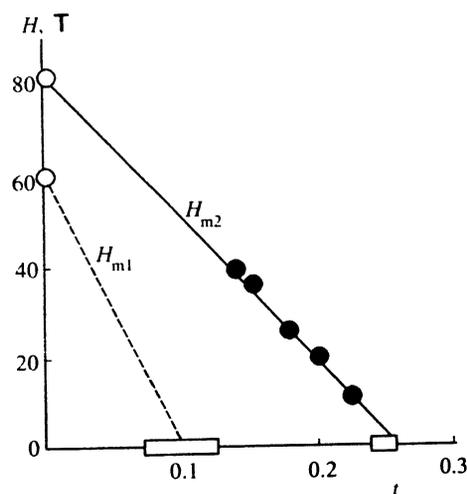


FIG. 3. Concentration dependence of the metamagnetic-transition fields H_{m1} and H_{m2} in the system $Y_{1-t}Gd_tCo_3$: ●—our values of H_{m2} , ○—values of H_{m1} and H_{m2} from Ref. 6, the rectangles represent the critical concentrations t_1 and t_2 .

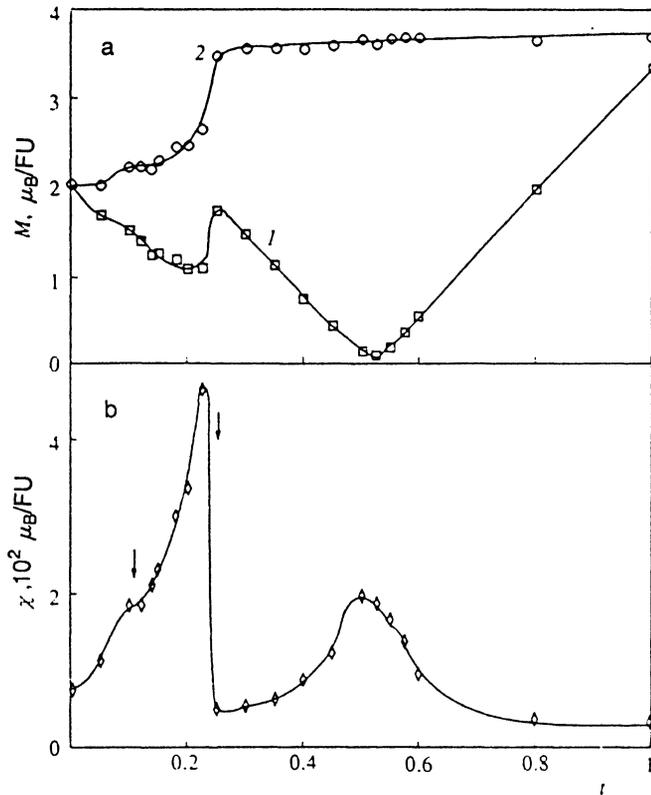


FIG. 4. a) Concentration dependence of the saturation magnetization $M_s(t)$ (curve 1) and magnetic moment $M_{Co}(t)$ of the d subsystem (curve 2) of $Y_{1-t}Gd_tCo_3$ at 4.2 K. b) Susceptibility χ of the paraprocess as a function of the gadolinium concentration t in the system $Y_{1-t}Gd_tCo_3$ at 4.2 K. The arrows mark the critical concentrations t_1 and t_2 for which anomalies are observed in the concentration dependence $M_{Co}(t)$ in the absence of a field.

tion field in the substituted compounds $Y_{1-t}Gd_tCo_3$. The magnetization anomalies of the cobalt subsystem in YCo_3 an external field,⁴ ($(\Delta M_{Co})_{H=60T} = 0.48 \mu_B/FU$ and $(\Delta M_{Co})_{H=82T} = 1.05 \mu_B/FU$) and in the effective field in $Y_{1-t}Gd_tCo_3$ as functions of the concentration ($(\Delta M_{Co})_{t_1} = 0.2 \mu_B/FU$ and $(\Delta M_{Co})_{t_2} = 1.2 \mu_B/FU$) are close to one another. We note that the parameters of the first transition cannot be determined precisely because of the weakness of the magnetization anomaly in the cobalt subsystem at $t \approx 0.1$, the fact that the anomaly is spread over a wide concentration range, and the above-noted presence of an impurity phase in the experimental samples. The values given above for these parameters are therefore only approximate.

The peaks in the susceptibility of the paraprocess near the metamagnetic transitions in the effective molecular field in the system $Y_{1-t}Gd_tCo_3$ are also associated with the metamagnetic increase of the magnetic moment of cobalt with increasing gadolinium concentration. Here, the fact that because the gadolinium is distributed randomly in the substituted compounds the metamagnetic transitions in the effective field are much more diffuse than transitions in YCo_3 in an external field; this is what cause the broadening of the susceptibility peaks of the paraprocess in $Y_{1-t}Gd_tCo_3$.

The nonlinear increase of $T_C(t)$ (see Fig. 1) may be associated with the change occurring in the magnetic state of

the d subsystem as the gadolinium concentration changes. This behavior indicates that the magnetic instability of the d subsystem is also significant near the Curie point.

The concentration range near the concentration compensation point t_{comp} requires a separate analysis. According to Fig. 4, the anomalous increase of the spontaneous magnetic moment of cobalt in this region is small, if it exists at all, and it falls within the range of accuracy of the magnetization measurements. However, the large peak in susceptibility of the paraprocess near t_{comp} confirms the anomalous magnetic behavior of the substituted compounds in this concentration region. It has not been ruled out that in $Y_{1-t}Gd_tCo_3$ a third transition occurs in the effective field at concentration $t \approx 0.5$. It is also entirely possible that the observed anomalies are associated with other effects, in particular, the fact that phenomena caused by the appearance of a noncollinear magnetic structure in a field become important near the compensation point (see below). Although the experimental data were analyzed for fields below H_{c1} , in which a noncollinear magnetic structure arises, because of the random distribution of gadolinium the noncollinear phase can also contribute to the susceptibility in fields $H < H_{c1}$, giving rise to the peak near t_{comp} . Therefore, we can talk about a third metamagnetic transition only tentatively, and additional investigations are required to prove its existence unequivocally.

We now consider the behavior of the magnetization of the compounds $Y_{1-t}Gd_tCo_3$ in a magnetic field. As we have already noted, in some samples of the experimental system a metamagnetic transition is observed with gadolinium concentrations less than the critical concentration t_2 for the transition into the strongly magnetic state (see Fig. 2). Comparing the magnitudes of the magnetization jumps and the metamagnetic transition fields in the substituted compounds $Y_{1-t}Gd_tCo_3$ and YCo_3 shows that this transition corresponds to a transition of cobalt in the field H_{m2} from the intermediate into the strongly magnetic state. The molecular-field f - d exchange interaction constant can be determined from the data on the concentration dependence of H_{m2} (Fig. 3). The effective field acting on the itinerant d subsystem is determined by Eq. (2), and since in the concentration range of the metamagnetic transition the magnetic moment of the itinerant d subsystem is greater than the magnetic moment of the gadolinium subsystem and it is oriented parallel to the field,

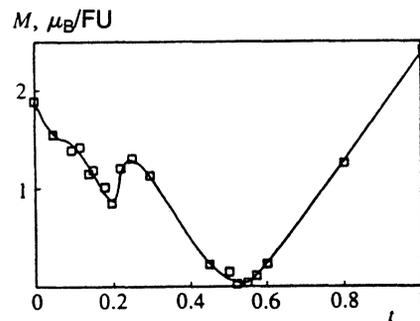


FIG. 5. Concentration dependence of the $Y_{1-t}Gd_tCo_3$ saturation magnetization $M_s(t)$ at 83 K.

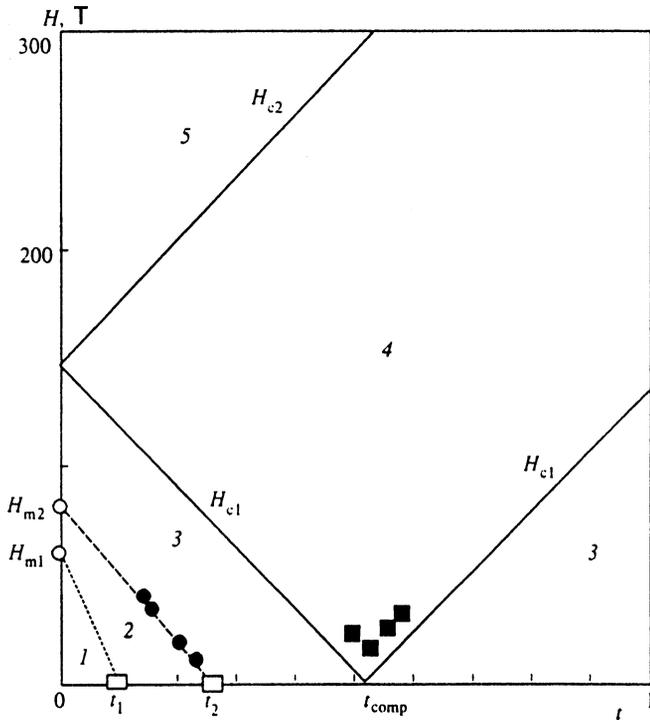


FIG. 6. Computed H -versus- t magnetic phase diagram for $Y_{1-t}Gd_tCo_3$. The circles and squares represent the experimental data. 1—Weakly ferrimagnetic collinear phase, 2—intermediate ferrimagnetic collinear phase, 3—strongly ferrimagnetic collinear phase, 4—noncollinear phase (cobalt in a strongly magnetic state), and 5—strongly ferromagnetic phase.

the effective field can be represented in the form

$$H_{\text{eff}}^d = H + |\lambda_{f-d}| t \mu_{Gd}. \quad (7)$$

Therefore, the metamagnetic-transition field should decrease linearly with increasing gadolinium concentration:

$$H_{m2}(t) = H_{m2}(0) - |\lambda_{f-d}| t \mu_{Gd}. \quad (8)$$

This agrees with the experimental data (Fig. 6), where we obtain $\lambda_{f-d}^{(2)} = 45 \text{ T}/\mu_B$ per formula unit.

Although, as we have noted above, we were not able to observe metamagnetic transitions from the weakly ferrimagnetic state into an intermediate phase in the field H_{m1} (possibly because the change in the cobalt magnetic moment at this transition is small and the transition is spread over a large range of fields), we can estimate approximately the molecular-field constant also from the field of the metamagnetic transition into this phase in pure YCo_3 (Ref. 4) and the critical gadolinium concentration t_1 at which a transition is observed into an intermediate phase in zero magnetic field. Estimates give $\lambda_{f-d}^{(1)} \approx 80 \text{ T}/\mu_B$ per formula unit, i.e., much greater than the value found from the transition into the strongly ferrimagnetic phase.

The molecular-field f - d exchange interaction constant can also be determined from the magnetization curves for compounds with t close to the composition t_{comp} at which transitions into the noncollinear ferrimagnetic phase are observed in strong fields (see Fig. 2b). Using the values of the susceptibility in the noncollinear phase and the critical field

H_{c1} for compounds near the compensation point, we find from the formulas (4) and (5) $\lambda_{f-d}^{\text{eff}} = 41 \text{ T}/\mu_B$ per formula unit.

In summary, we have found that different experimental data yield different values of λ_{f-d} . This could be due to the fact that in the crystal structure of the compounds $Y_{1-t}Gd_tCo_3$ cobalt can occupy three types of crystallographically nonequivalent positions. For this reason, it is natural to associate the two metamagnetic transitions observed in YCo_3 in an external field as well as the two anomalies in the concentration dependences of $M_{Co}(t)$ in $Y_{1-t}Gd_tCo_3$ with the change in the magnetization of cobalt in two of the three different cobalt positions. Strictly speaking, for compounds of the type $Y_{1-t}Gd_tCo_3$, a model that takes into account three local itinerant d subsystems corresponding to the three nonequivalent cobalt positions should be used. This approach is also dictated by neutron scattering data,⁸ according to which in YCo_3 the magnetic moments of cobalt in nonequivalent positions are considerably different from one another ($0.55\mu_B$ in the $3b$ positions, $0.83\mu_B$ in the $6c$ positions, and $0.4\mu_B$ in the $18h$ positions). In this case, the f - d exchange interaction is described by three different molecular-field constants λ_{f-d}^i ($i=1,2,3$), which in the nearest-neighbor approximation are related to the corresponding exchange-interaction integrals

$$\lambda_{f-d}^i = \frac{Z_i}{N_{d_i}} I_{f-d}^i \frac{2(g_R - 1)}{g_R}, \quad (9)$$

where g_R is the g -factor of the rare earth, Z_i is the number of rare-earth atoms in the first coordination sphere of the d atom at the i th position, and N_{d_i} is the number of such atoms per formula unit,⁹ and even if it is assumed that the f - d spin exchange parameters I_{f-d}^i are identical for the three nonequivalent positions, different values of λ_{f-d}^i should be obtained because the number of nearest neighbors is different for different nonequivalent positions (three for the $3b$ positions, four for the $6c$ positions, and six for the $18h$ positions). Investigations of field-induced noncollinear magnetic structures yield, however, an average value λ_{f-d} over the three nonequivalent positions.

5. MAGNETIC PHASE DIAGRAMS OF $Y_{1-t}R_tCo_3$

The data obtained make it possible to construct the complete H -versus- t magnetic phase diagram for the system $Y_{1-t}Ge_tCo_3$. As we have already mentioned, two types of magnetic phase transitions are possible in this system and in the compounds $Y_{1-t}R_tCo_3$ with other heavy rare earths: metamagnetic transitions of the cobalt d system from a weakly magnetic into an intermediate state in an effective field H_{m1} and from an intermediate into a strongly magnetic state in an effective field H_{m2} as well as transitions from the ferrimagnetic phase into a noncollinear ferrimagnetic phase in the field H_{c1} and from the noncollinear phase into the ferromagnetic phase in the field H_{c2} [see Eq. (4)].

According to the experimental data, $H_{m1}, H_{m2} < H_{c1}$ in $Y_{1-t}Gd_tCo_3$ for all t , and for this reason the transition into the noncollinear phase in the field H_{c1} occurs from the

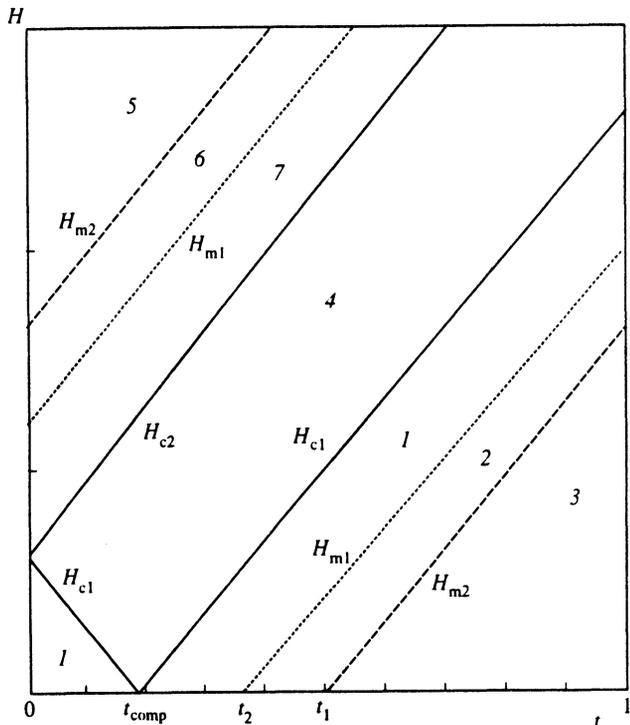


FIG. 7. Qualitative H -versus- t magnetic phase diagram for $Y_{1-t}R_tCo_3$, $R=Tb, Dy, Ho, Er,$ and Tm . 1—Weakly ferrimagnetic collinear phase; 2—intermediate ferrimagnetic collinear phase; 3—strongly ferrimagnetic phase; 4—noncollinear phase (cobalt in a weakly ferromagnetic state), 5—strongly ferromagnetic phase, 6—intermediate ferromagnetic phase, and 7—weakly ferromagnetic phase.

strongly ferrimagnetic phase. The complete H -versus- t phase diagram for this system has the form shown in Fig. 6.

In the mixed compounds $Y_{1-t}R_tCo_3$ with other heavy rare earths, however, a different situation is possible when the metamagnetic-transition fields H_{m1} and H_{m2} are higher than the field H_{c1} of the transition into the noncollinear ferrimagnetic phase as $t \rightarrow 0$. In the simplest case, if it is assumed that λ_{f-d} has the same value in the nonequivalent cobalt positions, then the H -versus- t phase diagram for such compounds has the form shown in Fig. 7. The transition into

the noncollinear phase occurs from the weakly ferrimagnetic state. The spontaneous intermediate and weakly ferrimagnetic states arise for $t > t_{comp}$. Since in this case the cobalt subsystem is oriented antiparallel to the field, for $t > t_{comp}$ demagnetization of the d subsystem should be observed: transitions from the strongly ferrimagnetic into the intermediate and weakly ferrimagnetic state. For fields $H > H_{c2}$, when the magnetic moments of the d and f subsystems are parallel to one another (the system is ferromagnetic), the reverse transition of the d subsystem from a weakly magnetic state into an intermediate and strongly magnetic state occurs. Estimates made on the basis of Eqs. (2) and (4) under the assumption that the exchange parameter I_{f-d} is the same for all rare earths show that phase diagrams of this type should be observed for the mixed systems $Y_{1-t}R_tCo_3$, where $R=Tb, Dy, Ho, Er,$ and Tm . We note that in some cases the phase diagram could become much more complicated if the fact that λ_{f-d}^i assumes different values in different nonequivalent positions is taken into account. Experimental investigations of the phase diagrams of some systems of this type are in progress.

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¹We note that at the magnetic-compensation composition t_{comp} , the spontaneous magnetization is different from zero and equals approximately $0.09\mu_B/FU$. This value can be taken as the maximum magnetization of the impurity phases.

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