The effect of *f-d* interaction on the magnetic state of *d*-subsystems in the itinerant magnets $Y(Co_{1-x}Al_x)_2$; investigation of the compounds $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$

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In order to investigate the effect of the *f-d* exchange interaction on the magnetic state of *d*-subsystems in the itinerant metamagnets $Y(Co_{1-x}Al_x)_2$, we have measured the magnetization and magnetic susceptibility of the intermetallic compounds $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ (for $0 \leqslant t \leqslant 0.2$, $x = 0.0, 0.05, 0.07, 0.085$, and 0.105). We have observed that in the compounds with high aluminum content ($x \geqslant 0.05$) the field at which the metamagnetic transition occurs decreases with increasing gadolinium content, eventually leading to a ferromagnetic state, whereas in the compounds with low aluminum content ($x \leqslant 0.05$) introduction of aluminum causes the metamagnetic transition field to increase and leads to formation of a cluster spin glass state. We have constructed the magnetic phase diagrams as well. Most of our results can be described using a molecular-field model that takes into account the presence of both itinerant *d*-subsystems and localized *f*-subsystems in these intermetallics.

INTRODUCTION

In order to shed light on the problem of why subsystems of itinerant *d*-electrons in intermetallic compounds are magnetically unstable, much attention has been focused on the Laves phases RCo_2 (with type C15 fcc crystal structure), where R is a rare earth or yttrium.[1] In these compounds, a *d*-electron subsystem forms as a result of *3d-5d* hybridization (*3d-4d* in the case of YCo_2) of the cobalt and rare earth *d*-electrons. For this subsystem, the density of *d*-states at the Fermi level and the magnitude of *d-d* exchange are such that the criterion for appearance of itinerant ferromagnetism (i.e., the Stoner criterion) is not fulfilled.[2,3] Because of this, the compounds with nonmagnetic rare earths, i.e., yttrium and lutetium, are exchange-enhanced itinerant paramagnets.[4]

An important feature of the magnetic behavior of YCo_2 and $LuCo_2$ is the fact that these compounds are metamagnets, i.e., they convert discontinuously to a ferromagnetic state at a field H_M, the so-called metamagnetic transition field[2] (this is referred to as the phenomenon of itinerant metamagnetism). This transition, which has only recently been observed experimentally,[5,6] takes place in fields $H_M \sim 10^6$ Oe, and is explained by peculiarities in the energy dependence of the density of states $N(E)$ of the *d*-electrons near the Fermi level which cause $N(E)$ to increase in a magnetic field.[3]

In the RCo_2 intermetallics with magnetic rare earths, both magnetic subsystems—the localized 4*f* electrons of the rare earths and the itinerant *d* electrons—are magnetically ordered.[7] In this case, the magnetic ordering of the itinerant subsystem is extrinsic, and is due to a magnetizing molecular field $H^d_{\rm mol}$ acting on the *d*-subsystem. This field, which arises from the *f*-subsystem of the magnetic rare earth atoms, is larger than the metamagnetic transition field H_M for all R.[2,8] If we dilute the rare earth *f*-subsystem with nonmagnetic yttrium, we can decrease this molecular field $H^d_{\rm mol}$ acting on the itinerant subsystem, and follow the influence of *f-d* exchange on the properties of the *d*-subsystem. Such investigations have been carried out on the (RY)Co_2 compounds for R = Gd, Tb, Dy, Ho, and Er.[9-17] According to these investigations, magnetic instability of the *d*-subsystem arises in ranges of rather high rare earth concentrations (more than 20 at.%). However, up to now the effect of small impurity concentrations of rare earths on the properties of *d*-subsystems has been left almost unstudied, except for Ref. 18, in which the properties of the system $(Y_{1-t}Gd_t)Co_2$ were analyzed for gadolinium concentrations *t* smaller than 0.20. There it was shown that these compounds are cluster spin glasses.

Comparatively recently it has been observed that partial replacement of the cobalt by aluminum in YCo_2 leads to a decrease in the metamagnetic transition field H_M, and to the appearance of weak itinerant ferromagnetism in the $Y(Co_{1-x}Al_x)_2$ system for $x \geqslant 0.12$.[19] This variation in the properties of these compounds with aluminum replacement can be understood within the framework of the itinerant magnetism model by an increase in the density of states as the Fermi level as the cobalt is replaced by aluminum. Thus, in the $Y(Co_{1-x}Al_x)_2$ system the magnetic stability of the *d*-subsystem increases as the aluminum content increases,

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which can significantly modify the way the impurity rare earths affect the *d*-subsystem.

At present, studies of this kind are nonexistent; therefore, in this paper we have undertaken to investigate the magnetic properties of the system $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ in order to clarify the effect of *f-d* exchange interactions on the behavior of the itinerant magnetic system $Y(Co_{1-x}Al_x)_2$ for small concentrations of localized impurities. We will limit ourselves to that range of aluminum concentrations ($0 \leqslant x \leqslant 0.105$) for which the original $Y(Co_{1-x}Al_x)_2$ compound is a paramagnet, undergoing a metamagnetic transition in the field. Compounds with gadolinium were chosen because Gd^{3+} is an *S*-state ion with small anisotropy, so that we need to take into account only the isotropic exchange interaction when interpreting the results.

THEORETICAL MODEL

Our theoretical discussion of the fundamental magnetic properties of the $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ compounds under discussion here can be carried out using the mean field model. We note at the outset that this model is approximate, in that it does not take into account either spin fluctuations in the itinerant *d*-subsystem or magnetic clusters which can form for small gadolinium concentrations. Therefore our model will apply only to uniform systems with long-range magnetic order (i.e., ferro- or ferrimagnetism), so that a description of spin glasses and other inhomogeneous states is outside its framework.

Following the approach of Refs. 2 and 16, we will treat the itinerant *d*-subsystem in these compounds within the Wohlfarth model of weak itinerant ferromagnetism. Then, taking into account the *f-d* exchange interaction, we can write the equation of state for the itinerant subsystem in the form

$$\alpha \mathbf{M}_d + \beta \mathbf{M}_d^3 + \gamma \mathbf{M}_d^5 = \mathbf{H}_{\text{eff}}^d, \tag{1}$$

where $\mathbf{M}_d$ is the magnetization of the *d*-subsystem and $\mathbf{H}_{\text{eff}}^d$ is the effective field acting on this subsystem:

$$\mathbf{H}_{\text{eff}}^d = \mathbf{H} + t\lambda_{fd}\mathbf{M}_f, \tag{2}$$

where $\mathbf{H}_{\text{mol}}^d = t\lambda_{fd}\mathbf{M}_f$ is the molecular field acting on the *d*-subsystem, λ_{fd} is the molecular field coefficient for the *f-d* exchange interaction (according to Ref. 7, λ_{fd} is negative for compounds with heavy rare earths), and $\mathbf{M}_f$ is the magnetization of the *f*-subsystem (calculated for a single *f*-ion).

It was shown in Ref. 20 that the itinerant system is paramagnetic if

$$\alpha > 0, \tag{3a}$$

while itinerant metamagnetism appears when we have

$$\beta < 0, \ \gamma > 0 \tag{3b}$$

and when a certain relation holds between α, β, and γ.

The magnetization of the *f*-subsystem $\mathbf{M}_f$ can be written

$$M_f = \mu_f B_{7/2}[(\mu_f/k_B T)|\mathbf{H}_{\text{eff}}^f|], \tag{4}$$

where μ_f is the saturation magnetic moment of the *f*-subsystem (for gadolinium $\mu_f = 7\mu_B/\text{atom}$) and the effective field acting on the *f*-subsystem equals

$$\mathbf{H}_{\text{eff}}^f = \mathbf{H} + \lambda_{fd}\mathbf{M}_d + t\lambda_{ff}\mathbf{M}_f \tag{5}$$

(λ_{ff} is the molecular-field coefficient for the *f-f* exchange interaction).

Ground state

In this model the ground state is ferromagnetic, because the exchange interaction between the *f*- and *d*-subsystems is negative ($\lambda_{fd} < 0$) in compounds with heavy rare earths. In this case, as we have already noted, magnetic ordering in the *d*-subsystem is extrinsic, i.e., it appears as a result of the *f-d* exchange interaction, whereas in the *f*-subsystem magnetic ordering is intrinsic if $\lambda_{ff} \neq 0$, and appears even if $\lambda_{dd} = 0$.

Depending on the value of the ratio H_M/H_{mol}^d, we distinguish two cases.

1. If the effective molecular field H_{mol}^d acting on the *d*-subsystem is smaller than the critical field H_M for the metamagnetic transition, the *d*-subsystem remains in a magnetized paramagnetic phase. In what follows we will refer to this state as the weakly ferrimagnetic phase. We can write the spontaneous magnetization of the compound in this phase at 0 K in the form

$$M = (-\chi_d\lambda_{fd} - 1)tM_f, \tag{6}$$

where the susceptibility of the *d*-subsystem is $\chi_d = 1/\alpha$.

2. The case $H_{\text{mol}}^d > H_M$, i.e., when the *d*-subsystem is ferromagnetic at low temperatures; we will call this the strongly ferrimagnetic phase. In this phase, at 0 K we have

$$M = M_d(H_M) + (-\chi_d'\lambda_{fd} - 1)tM_f - \chi_d' H_M, \tag{7}$$

where the quantities $M_d(H_M)$, H_M, and for fields $H > H_M$ the susceptibility of the *d*-subsystem $\chi_{d'}$ can be written in terms of the coefficients α, β, and γ.[20]

The critical concentration of rare earths t_K at which a transition occurs in the itinerant subsystem from the paramagnetic to the ferromagnetic state, i.e., a transition of the *f-d* system from the weakly ferrimagnetic to the strongly ferrimagnetic phase, is given by the relation

$$t_K = H_M/(-M_f\lambda_{fd}). \tag{8}$$

The magnetic phase transition temperature

Depending on the characteristics of the *d*-subsystem, the transition from the magnetically ordered to the paramagnetic state can be a phase transition of either second or first order. If the *d*-subsystem is in the paramagnetic phase in its ground state ($H_{\text{mol}}^d < H_M$), then the *f-d* magnetic system undergoes a second-order phase transition at the Curie point, and the transition temperature of the system is given by the expression

$$T_c^{\text{II}} = \frac{g^2 J(J+1)\mu_B^2}{3k_B} t\,(\lambda_{fd}^2 \chi_d|_{T=T_c^{\text{II}}} + \lambda_{ff}). \tag{9}$$

An analogous situation is encountered when the *d*-subsystem is ferromagnetic in its ground state ($H_{\text{mol}}^d > H_M$); however, the metamagnetic transition itself is smeared out at the Curie temperature.

If, however, the metamagnetic character of the magnetization of this subsystem is preserved up to the Curie temperature, then the transition to the paramagnetic state is a first-order phase transition, and its temperature can be determined from the expression:[15]

$$\frac{H_M{}^{\mathrm{I}}}{-t\lambda_{fd}}\bigg|_{T=T_c{}^{\mathrm{I}}}=\mu_f B_J\left\{\frac{\mu_f}{kT_c{}^{\mathrm{I}}}\left(\lambda_{fd}M_d H_M\,|_{T=T_c{}^{\mathrm{I}}}\right.\right.$$

$$\left.\left.+H_M\frac{\lambda_{ff}}{\lambda_{fd}}\bigg|_{T=T_c{}^{\mathrm{I}}}\right)\right\}. \qquad (10)$$

Thus, the character of the magnetic phase transition (i.e., a first- or second-order transition) depends on whether or not this temperature is larger than the Curie temperature.

Behavior in a magnetic field

For metamagnetic compounds, the behavior of the magnetization in a magnetic field can be analyzed using Eqs. (1) and (2). If the magnetization of the d-subsystem exceeds that of the f-subsystem ($M_d > tM_f$), then the magnetic moment of the d-subsystem is oriented parallel to the external field, while the moment of the f-subsystem is antiparallel to the latter; here the f-d exchange interaction leads to a decrease in the metamagnetic transition field [see Eq. (2)]:

$$H_M(t)=H_M(0)-(-t\lambda_{fd}M_f) \qquad (11)$$

(we recall that $\lambda_{fd}<0$).

For $M_d < tM_f$, however, the magnetization of the f-subsystem is oriented along the field. Then application of an external field causes the d-subsystem to be initially demagnetized, and then to be magnetized parallel to this field. Thus, in this case the f-d interaction leads to an increase in the metamagnetic transition field:

$$H_M(t)=H_M(0)+(-t\lambda_{fd}M_f). \qquad (12)$$

In fields smaller than the metamagnetic transition field, we can treat the susceptibility of the d-subsystem as approximately field independent. The conditions under which introduction of magnetic rare earths will bring about a decrease in the metamagnetic transition field are obtained from (6):

$$-\chi_d\lambda_{fd}=1. \qquad (13)$$

We note the approximate character of condition (13), which was obtained in the approximation that the d-subsystem susceptibility is field-independent in fields $H<H_M$; furthermore, it includes only collinear two-sublattice structures without considering the possibility that the sublattices could split into sub-sublattices.

In the strongly ferrimagnetic phase, it is well known that a noncollinear magnetic structure should appear in an external field.[21] The field interval where this structure exists is

$$H_{\mathrm{k}1}\leqslant H\leqslant H_{\mathrm{k}2}, \qquad (14)$$

where

$$H_{\mathrm{k}1}=-\lambda_{fd}|M_f-M_d|,$$
$$H_{\mathrm{k}2}=-\lambda_{fd}|M_f+M_d|. \qquad (15)$$

In the noncollinear phase, the magnetization depends on the field linearly:

$$M_{\mathrm{hk}}=-\frac{H}{|\lambda_{fd}|}. \qquad (16)$$

Thus, these considerations show that in R-3d intermetallics with metamagnetic itinerant subsystems it is possible to observe new effects at small concentrations of magnetic rare earths, caused by the mutual interaction of the two subsystems. Experimental investigation of these effects allows us to understand more deeply the peculiarities of magnetism in such compounds, and to clarify the limits of applicability of the simple model proposed here.

EXPERIMENTAL DETAILS

Polycrystalline samples of the intermetallic compounds $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$, where $0\leqslant t\leqslant 0.20$ and $x = 0.0, 0.05, 0.07, 0.085$, and 0.105, were smelted in an induction furnace under an atmosphere of spectrally pure argon in a water-cooled copper crucible under quasilevitated conditions. The ingots so obtained were homogenized over the course of a week at a temperature of 850 °C. X-ray phase analysis showed that the samples were single-phase, with C15 cubic crystal structure.

The magnetization at 4.2 K was measured in pulsed magnetic fields up to 270 kOe using the induction method. In the interval 2 to 200 K, the magnetization was measured in a static magnetic field up to 75 kOe using both the sample extraction method and a vibrating magnetometer. We measured the magnetic susceptibility over this temperature interval in weak AC fields. The crystal lattice parameters were determined in the temperature range from 5 to 300 K using the x-ray method.

EXPERIMENTAL RESULTS AND DISCUSSION

1. Magnetic properties at 4.2 K

Our measurements showed that the low temperature magnetic properties of the system $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ depend strongly on the aluminum content. The compounds we studied can be divided into two groups with different magnetic behavior. One group consisted of compounds with low aluminum content, the other the remaining high-Al compounds.

a) Systems with low aluminum content ($x=0.00$ and 0.05). The original $Y(Co_{1-x}Al_x)_2$ compounds for these systems are itinerant metamagnets, with metamagnetic transition fields H_M equal to 690 kOe ($x = 0.00$) and 450 kOe ($x = 0.05$) respectively.[19,22]

In Fig. 1 we show as an example the field dependence of the magnetization at 4.2 K for certain compounds belonging to the system $Y_{1-t}Gd_t(Co_{0.095}Al_{0.05})_2$. It is clear that for small replacements by gadolinium ($t<0.12$) there is no spontaneous magnetization. Increasing the gadolinium content leads to increased weak-field susceptibility; the magnetization curves of the compounds with Gd become nonlinear, and exhibit a tendency toward saturation in strong fields. Compounds with Gd content $t\geqslant 0.12$ possess a spontaneous moment. The value of this spontaneous moment decreases as the gadolinium content increases, passing through a minimum at $t_{\mathrm{comp}}\approx 0.17$–$0.18$ and then increasing once more (i.e., this concentration marks a balance point with respect to composition). In compounds near t_{comp} we observed kinks in the magnetization curves, which are characteristic of a transition from a collinear ferrimagnet to the noncollinear phase.

In the system $(Y_{1-t}Gd_t)Co_2$, there was no spontaneous magnetic moment over the entire gadolinium concentration interval $0\leqslant t\leqslant 0.12$ we investigated, and the magnetiza-

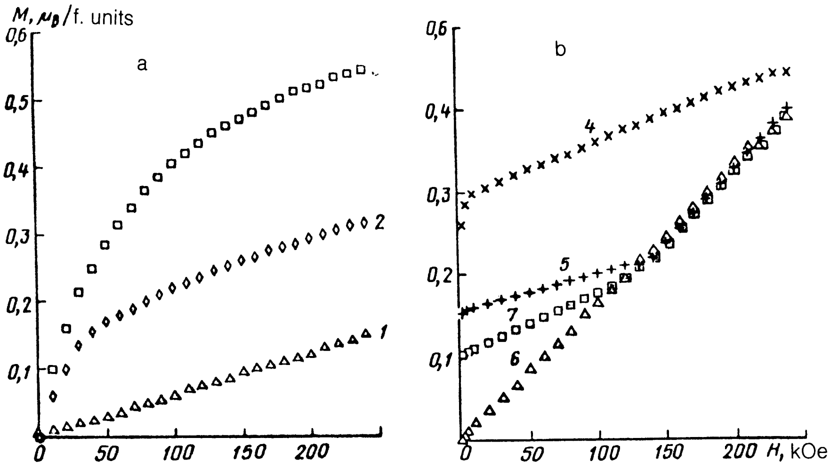

FIG. 1. Magnetization curves for the compounds $(Y_{1-t}Gd_t)(Co_{0.915}Al_{0.085})_2$ at 4.2 K and $t = 0.0$ (*1*), 0.02 (*2*), 0.04 (*3*), 0.10 (*4*), 0.15 (*5*), 0.18 (*6*), 0.20 (*7*).

tion curves did not differ qualitatively from magnetization curves of the compounds from the $Y_{1-t}Gd_t(Co_{0.95}Al_{0.05})_2$ system (see Fig. 1). Ferromagnetic order exists in this system for $t > 0.15$–0.20 (Refs. 18, 23).

b) Systems with high aluminum content ($x = 0.07$, 0.085, and 0.105). The properties of these systems are qualitatively analogous for various aluminum concentrations; therefore we will discuss in detail only the one with $x = 0.085$.

Figure 2 shows the magnetization curves of several compounds of the $Y_{1-t}Gd_t(Co_{0.915}Al_{0.085})_2$ system at 4.2 K. It is clear that the original compound $Y(Co_{0.915}Al_{0.085})_2$ is an itinerant metamagnet with a critical magnetic transition field $H_M = 225$ kOe. As the gadolinium concentration increases, the metamagnetic transition field H_M decreases, and for concentrations $t \gtrsim 0.04$ these compounds possess a spontaneous magnetization. At comparatively small gadolinium concentrations ($0.04 \lesssim t \lesssim 0.06$) the spontaneous magnetization is small in the magnetically ordered region, and application of a field leads to a metamagnetic transition from a weakly ferrimagnetic to a strongly ferrimagnetic state. For larger gadolinium content ($t \gtrsim 0.06$) metamagnetic transitions are not observed: these compounds are in the strongly ferrimagnetic state even at zero field. The saturation magnetization decreases with increasing gadolinium content up to $t_{\text{comp}} \approx 0.17$–$0.18$; further increasing t causes it to increase once more. As with the compounds with low aluminum content, near this concentration a transition is observed from the collinear ferrimagnetic phase to the noncollinear phase in an external magnetic field.

A decrease in the metamagnetic transition field is also observed in the other systems, i.e., $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ with high aluminum content ($x = 0.07$ and 0.105), as the gadolinium content increases. This decrease is followed by the appearance of a weakly ferrimagnetic phase, which is then replaced by a strongly ferrimagnetic phase. The only difference is that increasing the aluminum content causes the metamagnetic transition field in these compounds to increase, and the concentration region where the paramagnetic and weakly ferromagnetic metamagnetic phases can exist to shrink. At the same time, the values of the magnetization and gadolinium concentrations at which compensation of the magnetic moments of the *f*- and *d*-subsystems is observed are close for all these systems, although with certain differences. Furthermore, the spontaneous magnetization of the low-aluminum-content system with $x = 0.05$ is also close to the magnetization of those compounds corresponding to systems with larger amounts of aluminum ($x = 0.07$, 0.085, and 0.105).

All of these features are easy to see in Figs. 3 and 4, where we have plotted the basic characteristics of all the

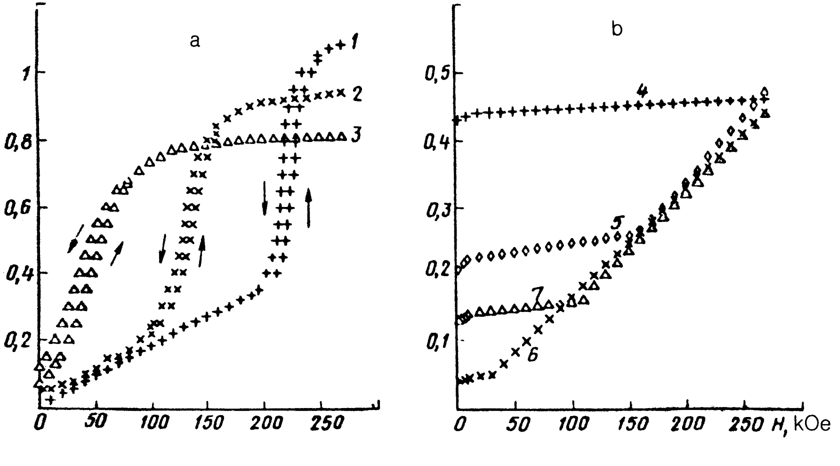

FIG. 2. Magnetization curves of the compounds $(Y_{1-t}Gd_t)(Co_{0.95}Al_{0.05})_2$ at 4.2 K and $t = 0.0$ (*1*), 0.04 (*2*), 0.1 (*3*), 0.12 (*4*), 0.15 (*5*), 0.18 (*6*), 0.20 (*7*).

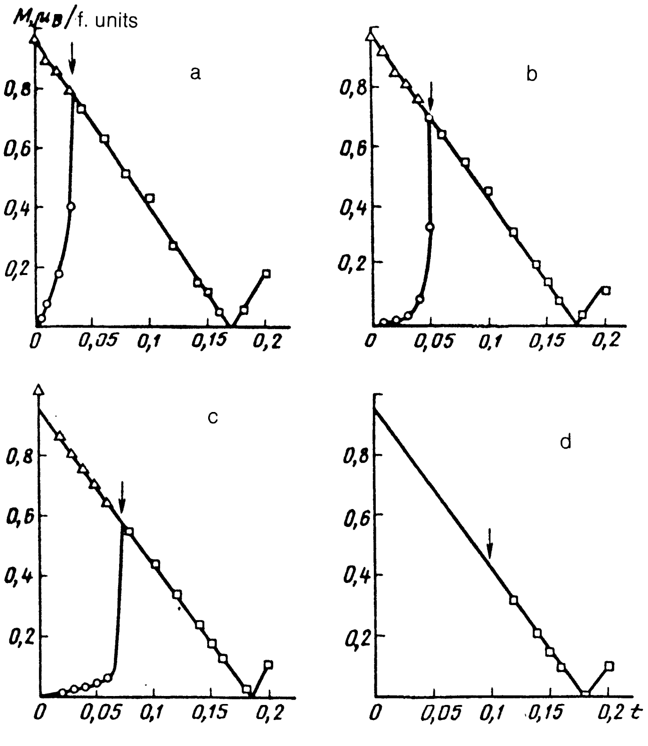

FIG. 3. Dependence of the magnetization on gadolinium content t for various compounds from the system $(Y_{1-t}Gd_t)(Co_{1-x}Al_x)_2$ at 4.2 K. ○—spontaneous magnetization of the weakly ferrimagnetic compounds, △—magnetization of the weakly ferrimagnetic compounds in a field of 270 kOe, □—spontaneous magnetization of the strongly ferrimagnetic compounds. $x = 0.105$ (a), 0.075 (b), 0.07 (c), 0.05 (d).

systems under discussion here at 4.2 K. In Fig. 3 we present data on the magnetization as a function of gadolinium concentration for systems with various x values. Here we show the spontaneous magnetizations of weakly ferrimagnetic and strongly ferrimagnetic samples, as well as magnetizations in a field of 270 kOe for samples with metamagnetic transitions. In Fig. 4 we show the measured dependence of the metamagnetic transition field on gadolinium content for systems with various aluminum contents. A comparison of the experimental data with calculations for *f-d* magnetic systems show that they agree in most cases, at least qualitatively.

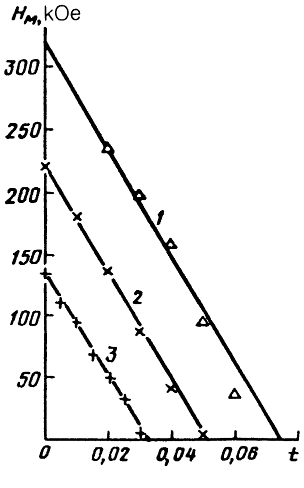

FIG. 4. Dependence of the metamagnetic transition field on gadolinium concentration t for the system $(Y_{1-t}Gd_t)(Co_{1-x}Al_x)_2$: $x = 0.07$, 0.085 (2), 0.105 (3). The straight line is calculated using Eq. (11), and the points are experimental data.

First of all, in the majority of cases a transition from a paramagnetic phase with metamagnetism to a weakly ferrimagnetic metamagnetic phase occurs as the gadolinium content increases, followed by a strongly ferrimagnetic phase (see Fig. 3), as predicted by theory.

Secondly, and also predicted by theory, the metamagnetic transition fields of the high-aluminum systems ($x = 0.07$, 0.085, and 0.105), in which the susceptibility of the *d*-subsystem is large, decrease almost linearly with the gadolinium concentration (Fig. 4).

Thirdly, no metamagnetic transitions are observed in the systems with low aluminum content ($x = 0.05$ and 0.00). This could be related to the small susceptibility of the *d*-subsystem, which in turn is associated with the fact that the magnetic moment of the *f*-subsystem in these compounds is oriented along the field.

For a quantitative comparison of the experimental data with the theoretical calculations it is necessary to know λ_{fd}, the molecular field coefficient for *f-d* exchange. We determined the latter using Eq. (15) and the experimental values of the field for a transition to the noncollinear phase H_{k1} and the magnetization M_{nc} in this field, and also by using Eq. (16) and the field dependence of the magnetization in the noncollinear phase. It turns out that within the limits of experimental error the coefficient λ_{fd} is independent of the aluminum and gadolinium contents, and equals

$$\lambda_{fd} = -(640 \pm 20)\ \text{kOe}/\mu_B\ \text{f. units}$$

(this question is treated in more detail in Ref. 24).

Once we know λ_{fd}, we can use Eq. (8) to calculate the critical concentration of gadolinium t_K at which a transition occurs from the weakly ferrimagnetic to strongly ferrimagnetic states. The computed values of t_K are shown in Fig. 3 by arrows; as is clear from this figure, agreement with the experimental data is satisfactory.

In Fig. 4 we compare experimental data for the dependence of the metamagnetic transition field at 4.2 K on gadolinium concentration t in systems having various aluminum concentrations x with theoretical calculations using Eq. (11). It is clear that good agreement is observed between experiment and theory. In these calculations we assumed that the *f*-subsystem is saturated, i.e., $M_f = \mu_f = 7\mu_B$. The conclusion that this subsystem is saturated at 4.2 K in the metamagnetic transition field $H_M(t)$ follows from computation of the magnetization of the *f*-subsystem using Eqs. (4) and (5). The fact is that at 4.2 K M_f comes to approximately 0.9 of μ_f even in an effective field of 20 kOe. Estimates show that only in the most favorable cases (compounds with $x = 0.07$ and $t = 0.01$) does H^f_{eff} amount to several kilooersteds in the metamagnetic transition field $H_M(t)$. For all the other metamagnetic states it is considerably larger than 20 kOe. This is even more true when the compounds are in their strongly ferrimagnetic phase.

As was shown above, the behavior of the magnetization in a field should change due to the increase in the *d*-subsystem susceptibility with increasing aluminum concentration: for high aluminum concentrations, increasing the gadolin-

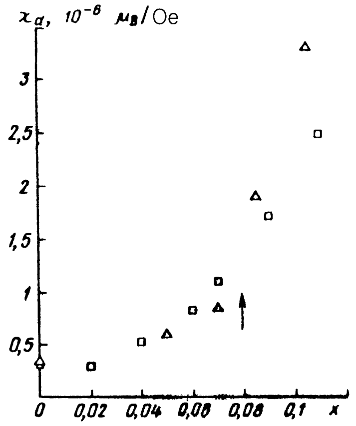

FIG. 5. Dependence of the d-subsystem susceptibility for the compounds $Y(Co_{1-x}Al_x)_2$ on aluminum concentration x at 4.2 K using our data (△) and the data of Ref. 22 (□). The arrow denotes the critical aluminum concentration at which the magnetization of the rare earth and d-subsystems become equal, as calculated from Eq. (13).

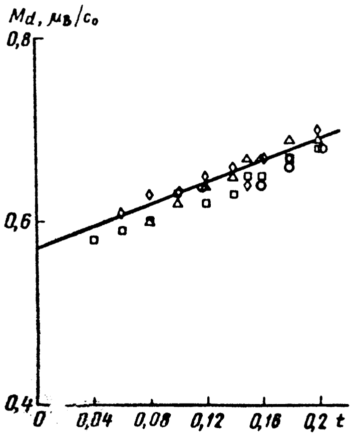

FIG. 6. Magnetic moment of the d-subsystem for the compounds $(Y_{1-t}Gd_t)(Co_{1-x}Al_x)_2$ as a function of gadolinium content t at 4.2 K. $x = 0.105$ (□), 0.075 (◇), 0.07 (×), 0.05 (○).

ium content leads to a decrease in the metamagnetic transition field (Eq. (11)), while for low aluminum concentrations it causes the latter to increase (Eq. (12)). The critical value of the susceptibility is given by Eq. (13). In Fig. 5 we show data on how the susceptibility for compounds in the $Y(Co_{1-x}Al_x)_2$ system (i.e., the susceptibility of the d-subsystem without gadolinium) depends on aluminum concentration, according to the data of our paper and that of Ref. 22. In this figure the arrow denotes the critical concentration at which there is a change in the way the gadolinium affects the metamagnetic transition field (in these estimates we have assumed that the gadolinium subsystem is saturated in the metamagnetic transition field). It is clear that the critical concentration equals 0.08 according to the theoretical estimates, whereas experimentally we observe a transition from one regime to another as we go from the composition $x = 0.05$ to $x = 0.07$. The experimental and theoretical values are therefore close, although with certain differences, possibly connected with the fact that we did not include the field dependence of the d-subsystem susceptibility in the calculations.

Figure 6 shows the behavior of the magnetic moment of the d-subsystem on gadolinium content in compounds that are in the strongly ferrimagnetic state, as calculated from the saturation magnetization assuming that the f-subsystem is saturated. It is clear that M_d increases linearly as the gadolinium composition is increased, which is due to an increase in the effective field acting on the d-subsystem. In this case, the data for systems with various aluminum contents lie on a single line within the limits of experimental error, indicating that the susceptibility χ' is composition independent in the ferrimagnetic state. From these data, using Eq. (7) we find that the susceptibility $\chi' = 1.2 \pm 0.2 \cdot 10^{-7}$ μ_B/Co atom, and determine by linear extrapolation that the moment of the d-subsystem M_d is equal to $1.1 \pm 0.2\,\mu_B$/Co atom in the compound $GdCo_2$. This value is in good agreement with the experimental value of M_d for $GdCo_2$ taken from Ref. 9.

Thus, the simple theory we have developed describes much of the experimental behavior of the system $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$. Nevertheless, it cannot explain the experimental magnetization curves of compounds with low aluminum and gadolinium contents. The magnetization curves for these compounds are nonlinear, and the weak-field susceptibility is considerably larger than for that of compounds with high aluminum content (compare Figs. 1 and 2). Previously it was shown that a cluster spin glass state appears in $(Y_tGd_{1-t})Co_2$ at low gadolinium concentrations ($t \lesssim 0.20$).[18] The fact that behavior analogous to the system $(Y_tGd_{1-t})Co_2$ is seen in the system with $x = 0.05$ suggests that a spin-glass state appears at low temperatures in this system as well.

2. Temperature characteristics. Let us now consider how the magnetic characteristics of the compounds $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ behave at various temperatures. The majority of compounds we studied had characteristic anomalies in the temperature dependence of the initial magnetic susceptibility at low temperatures, allowing us to identify the magnetic phase transition temperatures.

For those compounds that we claim harbor spin glasses, these anomalies take the form of "kinks" in the susceptibility, which is characteristic for a point where a spin glass is frozen in. The nonlinearity of the magnetization curves for compounds in the systems with $x = 0.00$ and 0.05, which are in the spin glass state at low temperatures, decreases with increasing temperature, and above the freeze-in temperature of the spin glass the magnetization curves become linear. When we process the magnetization curves for these compounds using the Belov–Arrot method, we find behavior characteristics of the spin glass state, i.e., nonlinear dependence of H/M on M^2 and zero spontaneous magnetization over the entire measurement temperature range.

For magnetically ordered compounds, the Curie temperature determined by the Belov–Arrot method is close to the Curie temperature found from measuring the magnetic susceptibility in a weak AC field.

In Fig. 7 we show the dependence of the magnetic phase transition temperature on gadolinium concentration t for systems with various aluminum compositions x. It is clear that the transition temperatures increase with increasing t, and that this increase takes place more rapidly as the aluminum concentration rises. In the compounds with higher aluminum content ($x = 0.07$, 0.085, and 0.105) this transition

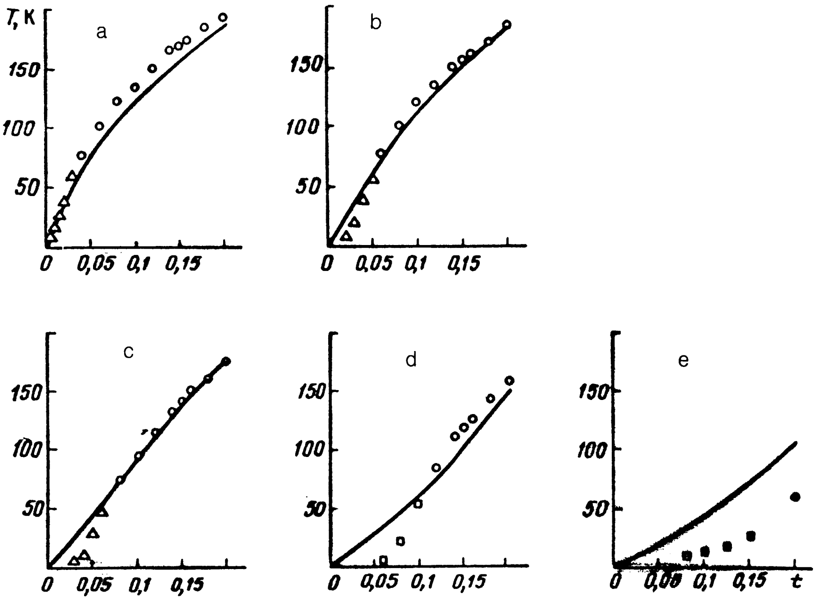

FIG. 7. Magnetic phase transition temperatures of compounds from the system $(Y_{1-t}Gd_t)(Co_{1-x}Al_x)_2$ as a function of the gadolinium concentration t. ◇—the transition to the strongly ferromagnetic phase, □—transition to the weakly ferrimagnetic phase, △—transition to a spin glass state. The curve is calculated theoretically using Eq. (9).

is from the weakly ferrimagnetic to the paramagnetic state for small t, whereas for large t it goes from the strongly ferrimagnetic to the paramagnetic state. In the compounds with small aluminum content ($x = 0.00$ and 0.05), a transition is observed to the spin glass state as the temperature decreases, while the system with $x = 0.085$ becomes strongly ferrimagnetic as the gadolinium content increases.

As we pointed out earlier, the transition from the magnetically ordered state to the paramagnetic state can be either a first-order or second-order transition in the strongly ferrimagnetic samples. In the first case, the paramagnetic and magnetically ordered phases can coexist in a transition region, as occurs in the system $Lu(Co_{1-x}Al_x)_2$.[25,26] In these compounds, determining the type of magnetic transition by using the temperature dependence of global characteristics (magnetization, susceptibility, etc.) becomes difficult because the content of the various phases changes with temperature.[26] Previously it was established by NMR investigations that the system $Y(Co_{1-x}Al_x)_2$, in contrast to $Lu(Co_{1-x}Al_x)_2$, is homogeneous in the transition region,[27] and that magnetic ordering in this compound takes place via a second-order transition. However, since a strongly ferrimagnetic phase appears in the systems $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$, we cannot dismiss the possibility that the nature of the magnetic transition in these compounds can change from second order to first order. Our investigations of thermal broadening by the x-ray method, which allows us to observe the coexistence of various phases (see Ref. 26), showed that the magnetic phase transition was second-order in all the samples we investigated, including the strongly ferrimagnetic samples. This is apparent in Fig. 8, where we show the temperature dependence of the crystal lattice parameter for several compounds: we did not observe coexistence of paramagnetic and magnetically ordered phases at the Curie point, there was no jump in the lattice parameter, and the magnetic anomaly in $a(T)$ was observed to increase monotonically with decreasing temperature.

Our calculations of the Curie temperatures for the systems $Y_{1-t}Gd_t(Co_{1-x}Al_x)_2$ were based on Eq. (9) for second order phase transitions, data on the temperature dependence of the susceptibility of d-subsystems taken from Ref. 22, and a value $\lambda_{ff} = 136$ kOe/μ_B taken from Ref. 7. These data are shown in Fig. 7. It is clear that the computed and experimental functions $T_c(t)$ are in semiquantitative agreement for large values of t. Considerably larger disagreements are observed between the experimental and theoretical dependences of the phase transition temperatures on gadolinium concentration for low gadolinium content, especially for the case of the spin glass state, where the system is significantly inhomogeneous and the crude mean-field approximation is inapplicable. Furthermore, an exact calculation of the Curie temperature must include spin fluctuations in the d-subsystem, which dominate the behavior of itinerant metamagnets.[29]

The temperature behavior of the spontaneous magnetization of the ferrimagnetic states (see Fig. 9, where we show the functions $M_s(T)$ for several compounds from the system with $x = 0.85$) is very interesting. If we are far from the compensation point, as measured from the composition

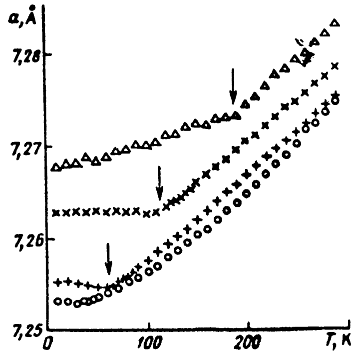

FIG. 8. Temperature dependence of the crystal lattice parameter of several compounds of the system $(Y_{1-t}Gd_t)(Co_{0.915}Al_{0.085})_2$: $t = 0.03$ (O), 0.04 (+), 0.10 (×), 0.20 (△). The arrows show the magnetic ordering temperature.

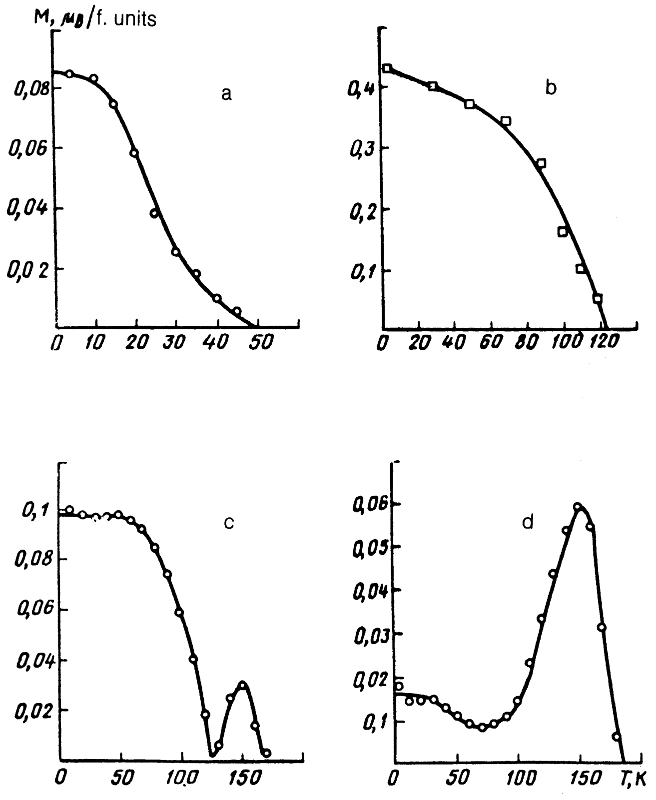

FIG. 9. Temperature dependence of the spontaneous magnetization of several compounds $(Y_{1-t}Gd_t)(Co_{0.915}Al_{0.085})_2$:$t = 0.04$ (a), 0.10 (b), 0.16 (c), 0.18 (d).

t_{comp} (the magnetization of the compounds is large), the spontaneous magnetization falls off monotonically as the temperature increases (see Figs. 9a, 9b). The magnetization of compounds located near t_{comp} depends anomalously on temperature (Figs. 9c and 9d). In this case, compounds located to the left of the compensation point with respect to composition ($t < t_{comp}$) exhibit a magnetic compensation temperature. From these data we may conclude that the temperature dependences of the f and d-subsystem magnetizations are almost identical (so that anomalous temperature dependence of the spontaneous magnetization is observed only if the magnetization of both subsystems are close to one another), and that the magnetization of the d-subsystem decreases more rapidly as the temperature increases than that of the f-subsystem. Therefore a compensation point is observed only for $t < t_{comp}$, when $M_d(0) > tM_f(0)$.

CONCLUSION

Thus, in *f-d* systems with magnetically unstable itinerant *d*-subsystems, the *f-d* exchange interaction can lead to a transition from one magnetic state to another in the *d*-subsystem, along with various effects associated with it. In the paramagnetic $Y(Co_{1-x}Al_x)_2$ compounds, the *f-d* exchange that appears when yttrium is partially replaced by gadolinium can either increase or decrease the critical field for the metamagnetic transition, depending on the aluminum concentration. For large concentrations of gadolinium, when the *f-d* exchange interaction causes the *d*-subsystem to enter the magnetically ordered state, phenomena are observed in this system that are characteristic of saturated collinear ferrimagnets: magnetic compensation of sublattices, noncollinear magnetic structures induced by an external field, etc.

The experimental results we have obtained can in many cases be described theoretically using the molecular-field approximation. The model we present here cannot be used to describe the properties of highly dilute compounds, in which the conditions for applicability of the mean-field approximation are violated, and whose description requires that we take into account the possible formation of magnetic clusters. Furthermore, it should be kept in mind that spin-density fluctuations in the *d*-subsystem play a significant role in altering the magnetic behavior in the compounds under investigation at high temperatures. For a more precise description of the magnetic properties of this class of intermetallics a theory is required that fully takes into account all of these factors.

It is with deep sadness that we report that our friend and colleague Rémy Lemaire was recently and tragically struck down in an automobile accident.

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