

NATURE OF MAGNETIC ANISOTROPY OF DYSPROSIUM. PARAMAGNETIC
SUSCEPTIBILITY OF DYSPROSIUM-GADOLINIUM ALLOYS

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The paramagnetic susceptibility of Dy_xGd_{1-x} alloys is measured in the basal plane and along the hexagonal axis (c axis) of the crystal. It is shown that the susceptibility obeys the Curie-Weiss law, the effective magnetic moment being the same in both directions and the paramagnetic Curie temperatures being different. In this case $\Delta\theta_p = \theta_{pp} - \theta_{pc} > 0$ and increases on increase of the dysprosium content in the alloy. An expression for the paramagnetic susceptibility of a two-component alloy is obtained in the molecular-field approximation, which takes into account, along with exchange energy, also the single-ion anisotropy and anisotropy due to exchange interaction. A comparison of the experimental dependence of $\Delta\theta_p$ on the composition of the alloys with the theoretical dependence shows that the alloy anisotropy is single-ion and is due to interaction between the anisotropic 4f-electron charge cloud and crystalline field of the lattice.

THE question of the nature of the magnetic anisotropy of heavy rare-earth metals (HREM) has not yet been completely answered. According to theoretical estimates, there are two mechanisms capable of explaining the tremendous value of the uniaxial magnetic anisotropy of HREM, namely single-ion anisotropy^[1-4] and anisotropy due to exchange interaction^[5-11].

In an earlier article^[12] we investigated the magnetic anisotropy of dysprosium-gadolinium alloys in a magnetically-ordered state, and concluded that uniaxial magnetic anisotropy of the dysprosium and its alloys with gadolinium is mainly of the single-ion type. However, owing to the large value of the anisotropy, we were unable to investigate alloys that contain more than 50 at.% of dysprosium. In addition, we were unable to take exact account of the magnetoelastic effects, the influence of which on the anisotropy at low temperature is quite large^[13]. A number of workers^[3,4,10,11] have shown that the uniaxial magnetic anisotropy of HREM in the magnetically-ordered state, and the anisotropy of the paramagnetic susceptibility of these substances, are interrelated and that information concerning the magnetic anisotropy can be obtained by investigating the paramagnetic susceptibility of HREM single crystals along different crystallographic directions.

We present here the results of the measurement of the paramagnetic susceptibility of single crystals of dysprosium-gadolinium alloys. These measurements were made to obtain additional information on the nature of the uniaxial magnetic anisotropy of HREM.

SAMPLES. EXPERIMENTAL PROCEDURE

We investigated the magnetic properties of single crystals of Dy_xGd_{1-x} alloys ($x = 0.046, 0.103, 0.189, 0.296, 0.378, 0.49, 0.70, \text{ and } 1.0$). The samples were grown by crucibleless zone melting in the Laboratory for Rare-Metal Alloys of the Metallurgy Institute of the USSR Academy of Sciences. The measurements were made on samples in the form of spheres 2.5–3 mm in diameter. The samples were oriented in the basal plane

and along the hexagonal axis by a magnetic method at room temperature. This is made possible by the large anisotropy of the paramagnetic susceptibility of the investigated alloys. A check by an x-ray method has shown the orientation accuracy to be 1–2%.

A magnetometer of the Dominicali-pendulum type was used to measure the magnetization of single crystals of the alloys in the basal plane in fields up to 15 kOe and in the temperature interval 78–500°K. These data were used to determine the temperature dependence of the susceptibility in the basal plane (χ_{bp}), and also to determine the magnetic-ordering temperature (θ_2) and the temperature of the transition from the antiferromagnetic to the ferromagnetic states (θ_1). In addition, an Akulov anisometer was used to measure the torque acting on the single-crystals of the alloy in fields up to 19 kOe and in the temperature interval 273–390°K. In the magnetic region, the torque is equal to

$$L = \frac{1}{2}(\chi_{bp} - \chi_c)H^2 \sin 2\varphi, \quad (1)$$

where χ_c is the susceptibility along the hexagonal axis and φ is the angle between the field direction and the hexagonal axis. Thus, measurement of the torque makes it possible to find the susceptibility difference $\Delta\chi = \chi_{bp} - \chi_c$ and to determine χ_c with the aid of the measured magnetization χ_{bp} in the basal plane.

Both in the susceptibility measurements and in the determination of $\Delta\chi$ it is necessary to take into account the demagnetizing factor. Simple calculations lead to the following relations between the measured and true values of χ and $\Delta\chi$ (per cm^3):

$$1/\chi = 1/\chi_{\text{meas}} - N, \quad (2)$$

$$\Delta\chi = \Delta\chi_{\text{meas}} / (1 - 4N\chi), \quad (3)$$

where the demagnetizing factor N is equal to $4\pi/3$ for a sphere.

EXPERIMENTAL RESULTS

Figure 1 shows the temperatures θ_1 and θ_2 determined by us for the dysprosium-gadolinium alloys. It

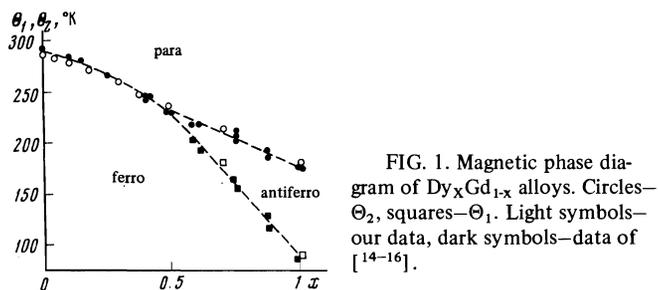


FIG. 1. Magnetic phase diagram of Dy_xGd_{1-x} alloys. Circles— Θ_2 , squares— Θ_1 . Light symbols—our data, dark symbols—data of [14-16].

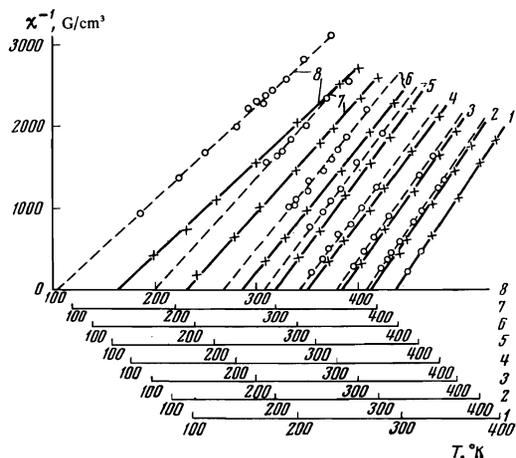


FIG. 2. Temperature dependence of the reciprocal paramagnetic susceptibility of single crystal Dy_xGd_{1-x} alloys: crosses— χ_{bp} , circles— χ_c . The concentrations for the different curves are as follows: 1—0.046, 2—0.103, 3—0.183; 4—0.296, 5—0.378, 6—0.49, 7—0.70, 8—1.0. The temperature scales are shifted for the different samples.

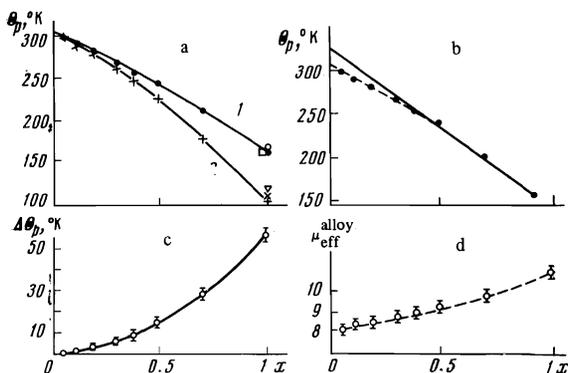


FIG. 3. Properties of Dy_xGd_{1-x} alloys in the paramagnetic region. a—Paramagnetic Curie temperatures; 1— $\Theta_{p.bp}$, 2— $\Theta_{p.pc}$. Points: \bullet , \times —our data; ∇ , \square —[17], \circ , \times —[18]. b—Difference of the paramagnetic Curie temperatures $\Delta\Theta_p = \Theta_{p.bp} - \Theta_{p.pc}$. c—Paramagnetic Curie point $\Theta_{p.poly} = (2/3)\Theta_{p.bp} + (1/3)\Theta_{p.pc}$; dark circles—experimental data, curve—theoretical plot based on (14). d—Effective magnetic moment $\mu_{\text{alloy, eff}}$ of the alloy in units of μ_B (per atom). Points—experimental data, dashed curve—theoretical plot according to (15).

shows also data obtained from measurements on polycrystalline samples in [14-18]. It is seen that the experimental values obtained by different workers are in good agreement.

Figure 2 shows the temperature dependences of the reciprocal paramagnetic susceptibility of dysprosium-gadolinium alloys in the basal plane and along the hexagonal axis. In the investigated temperature interval, the

susceptibility along both directions follows the Curie-Weiss law and is given by (per atom)

$$\chi = \mu_{\text{alloy, eff}} / 3k(T - \Theta_p). \tag{4}$$

Figure 3 shows the characteristic properties of the dysprosium-gadolinium alloys in the paramagnetic region. It is seen from Fig. 3a that Θ_p decreases with increasing dysprosium content in the alloy, with $\Theta_{p.bp} > \Theta_{p.pc}$. As shown in Fig. 3b, $\Delta\Theta_p = \Theta_{p.bp} - \Theta_{p.pc}$ increases with increasing x . Figure 3c shows a plot of $\Theta_{p.poly} = (2/3)\Theta_{p.bp} + (1/3)\Theta_{p.pc}$ against the dysprosium content. As follows from [3,4], this quantity characterizes the paramagnetic Curie temperature of the polycrystal and is not affected by the anisotropy. Figure 3d shows the dependence of the effective magnetic moment of the alloy $\mu_{\text{alloy, eff}}$ on the composition. We note that $\mu_{\text{alloy, eff}}$ is independent of the measurement direction within the limits of the experimental error ($\sim 4\%$).

DISCUSSION OF RESULTS

As shown in numerous studies (see, for example, the review [19]), the magnetic properties of HREM are well described by the model of localized moments. This model was used to investigate in detail the properties of the HREM in the paramagnetic region with allowance for the exchange and magnetic interactions [3,4,18].

There has been much less theoretical study of the properties of solid solutions of REM with one another at temperatures below Θ_p . We know of only one investigation [20] in which the influence of the one-ion anisotropy on the paramagnetic susceptibility of holmium-erbium alloys is analyzed. However, as shown by analysis, the theoretical relations obtained in that paper are valid only when $T \gg \Theta_p$, and can therefore not be used to interpret our data [1].

We consider below the paramagnetic properties of REM alloys in the molecular-field approximation. We first perform the calculation for the isotropic case, and then take into account the influence of the magnetic anisotropy on the paramagnetic susceptibility.

A. Isotropic Case

For an isotropic two-component alloy, the magnetic point of the energy takes the form

$$E = -\frac{1}{2} \sum_{i,j} c_i c_j J_{ij}^0 \mu_i \mu_j - H \sum_i c_i \mu_i. \tag{5}$$

The first term in (5) describe the isotropic exchange interaction [2], and the second term represents the Zeeman energy; μ_i and μ_j are the magnetic moments of the dysprosium atoms ($i, j = 1$) and gadolinium atoms ($i, j = 2$) at the given temperature, and c_i is the concentration of the alloy components:

$$c_1 = x, \quad c_2 = 1 - x. \tag{6}$$

¹For the same reason, the interpretation of the experimental data given in [20] is also incorrect, since these data were obtained at temperatures relatively close to Θ_p .

²This form of the expression is valid for an alloy that is fully disordered structurally [19].

In the molecular-field approximation, the magnetic moment of the alloy (per atom) is

$$\mu_{\text{alloy}} = \sum_i c_i \mu_{i0} B_{J_i} \left(\frac{\mu_{i0}}{kT} H_{i,\text{eff}} \right), \quad (7)$$

where $\mu_{i0} = g_i J_i$ is the magnetic moment of the i -th component of the alloy at 0°K , and the effective field $H_{i,\text{eff}}$ is equal to

$$H_{i,\text{eff}} = -dE / d\mu_i. \quad (8)$$

In the paramagnetic temperature region, the argument of the Brillouin function in (8) is small, the expanding the functions B_{J_i} in a series, we can easily obtain, after straightforward but cumbersome transformations, the following expression for the paramagnetic susceptibility of the alloy (per atom):

$$\left(\frac{d\mu_{\text{alloy}}}{dH} \right)_{H=0} = \chi = \frac{AT - B}{T^2 - DT + F}, \quad (9)$$

where

$$\begin{aligned} A &= \frac{1}{3k} (c_1 \mu_{1,\text{eff}}^2 + c_2 \mu_{2,\text{eff}}^2), \\ B &= \frac{c_1 c_2 \mu_{1,\text{eff}}^2 \mu_{2,\text{eff}}^2}{9k^2} (I_{11}^0 + I_{22}^0 - 2I_{12}^0), \\ D &= \frac{1}{3k} (c_1 \mu_{1,\text{eff}}^2 I_{11}^0 + c_2 \mu_{2,\text{eff}}^2 I_{22}^0), \\ F &= \frac{c_1 c_2 \mu_{1,\text{eff}}^2 \mu_{2,\text{eff}}^2}{9k^2} (I_{11}^0 I_{22}^0 - I_{12}^0{}^2); \end{aligned} \quad (10)$$

The effective magnetic moments $\mu_{i,\text{eff}}$ of the alloy components are equal to

$$\mu_{i,\text{eff}} = g_i [J_i (J_i + 1)]^{1/2}. \quad (11)$$

Formula (9) for the paramagnetic susceptibility differs from the Curie-Weiss law and represents a more general relation, the Néel law for the paramagnetic susceptibility of ferrimagnets (see, for example, [21]). The reason is that the alloy contains different magnetic atoms, the exchange interactions between which are not equal.

The general expression for the susceptibility becomes simpler for HREM alloys. As shown by numerous experimental and theoretical studies (for example, the review [4]), the exchange interaction constant I_0 between the spins of the rare-earth ions is approximately constant in the series of rare-earth elements (with an accuracy on the order of 10%). This circumstance leads to the following connection between the coefficients I_{ij}^0 :

$$I_{ij}^0 = I_0 (g_i - 1) (g_j - 1) / g_i g_j. \quad (12)$$

When these relations are taken into account, we find that $F = 0$ and the susceptibility of the REM alloys can be represented in the form

$$\chi = A(1 - B/TA) / (T - D). \quad (13)$$

It also follows from (12) that $B/AT \ll 1$ for REM alloys in the paramagnetic temperature region, and that the susceptibility of these alloys near the magnetic-ordering temperature can be approximated by the Curie-Weiss law (4)³⁾, where

³⁾Our estimates show that the error due to approximating the paramagnetic susceptibility of dysprosium-gadolinium alloys by the Curie-Weiss law in the temperature interval from Θ_p to 500°K does not exceed 4%.

$$\Theta_p = D = \frac{I_0}{3k} \left[c_1 \mu_{1,\text{eff}}^2 \left(\frac{g_1 - 1}{g_1} \right)^2 + c_2 \mu_{2,\text{eff}}^2 \left(\frac{g_2 - 1}{g_2} \right)^2 \right] \quad (14)$$

$$\begin{aligned} \mu_{\text{alloy,eff}} &= \left[\frac{1}{3k} A \left(1 - \frac{B}{DA} \right) \right]^{1/2} \\ &= (c_1 \mu_{1,\text{eff}}^2 + c_2 \mu_{2,\text{eff}}^2)^{1/2} \left[1 - \frac{I_0 c_1 c_2 \mu_{1,\text{eff}}^2 \mu_{2,\text{eff}}^2}{3g_1 g_2 (c_1 \mu_{1,\text{eff}}^2 + c_2 \mu_{2,\text{eff}}^2) k \Theta_p} \right]^{1/2}. \end{aligned} \quad (15)$$

Let us compare the theoretical relations (14) and (15) with the experimental data for dysprosium-gadolinium alloys. It must be recognized here that the theoretical relations were obtained for the isotropic case, while the experimental data were determined from measurements on single crystals, i.e., they are influenced by the anisotropy. We have therefore compared the theoretical plot of $\Theta_p(x)$ with the theoretical plot of

$\Theta_{p,\text{polycr}}(x)$. These data are shown in Fig. 3c. For the theoretical calculations we used for the parameter I_0 a value determined from the paramagnetic Curie temperature of pure dysprosium. It is seen that the experimental points are close to the theoretical curve, with the exception of the gadolinium-rich compositions ($x \leq 0.3$). This circumstance can be explained by assuming that the gadolinium-gadolinium exchange interaction in the alloys is somewhat smaller (by 5%) than follows from (12).

Figure 3b shows a theoretical plot of $\mu_{\text{alloy,eff}}$ against x in accordance with formula (15). In this case we can disregard the correction for the anisotropy. This follows from our experimental data, since the values of $\mu_{\text{alloy,eff}}$ determined from the measurements along different crystallographic directions are close to one another, and also from theoretical estimates (see below).

As seen from Fig. 3d, there is good agreement between the theoretical and experimental plots of $\mu_{\text{alloy,eff}}$. The experimental values $\mu_{\text{Dy,eff}} = (10.9 \pm 0.4) \mu_B$ and $\mu_{\text{Gd,eff}} = (8.2 \pm 0.3) \mu_B$ are close to the theoretical ones for free trivalent ions ($\mu_{\text{Dy,eff}} = 10.6 \mu_B$, $\mu_{\text{Gd,eff}} = 7.94 \mu_B$). Thus, a comparison of the theoretical and experimental data shows that the assumed theoretical model describes satisfactorily the properties of dysprosium-gadolinium alloys in the paramagnetic region.

B. Allowance for Anisotropy

Let us examine the influence exerted on the paramagnetic properties of REM alloys by the magnetic anisotropy. The energy of uniaxial magnetic anisotropy (per atom of the alloy) can be represented in the form

$$E_A = \frac{1}{2} \sum_{i,j} c_i c_j K_{2ij}^0 (3 \cos \theta_i \cos \theta_j - 1) + \frac{1}{2} \sum_i c_i K_{2i}^0 (3 \cos^2 \theta_i - 1). \quad (16)$$

The first term, which is quadratic in the concentrations of the components, describes here the anisotropy due to the exchange interaction, while the second term, linear in the concentrations, describes the single-ion anisotropy; θ_i is the angle between the direction of the magnetic moment of the i -th component and the hexagonal axis of the crystal, and K_2^0 stands for the first anisotropy coefficients in the expansion of the anisotropy energy in harmonic polynomials.

In the paramagnetic temperature region the constant K_2^0 depends quadratically on the magnetization both for single-ion anisotropy and for anisotropy due to exchange interaction^[22]:

$$K_{2ij}^0 = a_{ij}\mu_i\mu_j, \quad K_{2i}^0 = b_i\mu_i^2. \quad (17)$$

In addition, it is easy to show that at $T > \Theta_p$ the magnetic moments of the alloy components are oriented parallel to the field even in weak fields. Then the anisotropy energy can be written in the form

$$E_A = \frac{1}{2} \left[\sum_{i,j} c_i c_j a_{ij} \mu_i \mu_j + \sum_i c_i b_i \mu_i^2 \right] (3 \cos^2 \varphi - 1), \quad (18)$$

where φ is the angle between the direction of the field and the hexagonal axis.

Comparing the formula for the energy of the magnetic anisotropy (18) with the expression for the isotropic exchange energy (5), we easily see that the anisotropy can be accounted for by replacing the coefficients I_{ij}^0 in (5) by coefficients I_{ij} that depend on the field direction:

$$\begin{aligned} I_{ij} &= I_{ij}^0 - a_{ij}(3 \cos^2 \varphi - 1), \quad i \neq j; \\ I_{ii} &= I_{ii}^0 - (a_{ii} + b_i/c_i)(3 \cos^2 \varphi - 1). \end{aligned} \quad (19)$$

Consequently, in the presence of anisotropy, the paramagnetic susceptibility of the alloy is also described by formula (10), in which I_{ij}^0 are replaced by the quantities I_{ij} , which take the anisotropy into account.

Expression (10) for the paramagnetic susceptibility of an anisotropic alloy can be simplified if it is recognized that the energy of uniaxial anisotropy in an alloy is much less than the isotropic exchange energy. Then, as shown by straightforward but cumbersome calculations, the paramagnetic susceptibility of the anisotropic alloy will follow the Curie-Weiss law (4), with the paramagnetic Curie point and the effective magnetic moment of the alloy dependent on the direction of the measurements:

$$\Theta_p(\varphi) = \Theta_p - \frac{1}{3}(3 \cos^2 \varphi - 1)\Delta\Theta_p, \quad (20)$$

$$\mu_{\text{alloy,eff}}(\varphi) = \mu_{\text{alloy,eff}} - \frac{1}{3}(3 \cos^2 \varphi - 1)\Delta\mu_{\text{eff}}, \quad (21)$$

where Θ_p and $\mu_{\text{alloy,eff}}$ are given by formulas (14) and (16), while $\Delta\Theta_p$ and $\Delta\mu_{\text{eff}}$ are equal to

$$\begin{aligned} \Delta\Theta_p &= \frac{I_0}{3k^2\Theta_p} \left[\sum_{i,j} c_i c_j \frac{(g_i - 1)(g_j - 1)}{g_i g_j} a_{ij} \mu_{i,\text{eff}}^2 \mu_{j,\text{eff}}^2 \right. \\ &\quad \left. + \sum_i c_i \frac{(g_i - 1)^2}{g_i^2} b_i \mu_{i,\text{eff}}^4 \right], \end{aligned} \quad (22)$$

$$\begin{aligned} \Delta\mu_{\text{eff}} &= -\frac{c_1 c_2 \mu_{1,\text{eff}}^2 \mu_{2,\text{eff}}^2}{2k\Theta_p \mu_{\text{alloy,eff}}^2} \left[\sum_{i,j} \left(1 - \frac{\mu_{\text{alloy,eff}}}{3k\Theta_p} \frac{(g_i - 1)(g_j - 1)}{g_i g_j} a_{ij} \right) \right. \\ &\quad \left. + \sum_i \left(1 - \frac{\mu_{\text{alloy,eff}}}{3k\Theta_p} \frac{(g_i - 1)^2}{g_i^2} b_i \right) \right]. \end{aligned} \quad (23)$$

The derived relations enable us to determine from the experimental data the contributions of the different mechanisms to the anisotropy of REM alloys. It follows from (22) that if the anisotropy of the alloy is due to exchange interaction, then $\Theta_p \Delta\Theta_p$ is a quadratic function of the component concentrations, and if the anisotropy is of the single-ion type, then this quantity is a

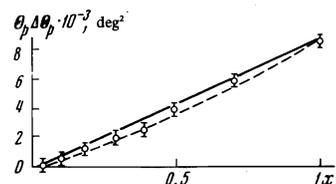


FIG. 4. Dependence of $\Theta_p \Delta\Theta_p$ on the composition of the $\text{Dy}_x \text{Gd}_{1-x}$ alloys. Circles—experimental data. Solid curve—Theoretical plot for single-ion model. Dashed—theoretical plot for single-ion model with allowance for the dependence of the crystal-field parameters on the composition of the alloy.

linear function of the concentration components. Figure 4 shows an experimental plot of $\Theta_p \Delta\Theta_p$ against the compositions of dysprosium-gadolinium alloys. It shows also the theoretical plot of $\Theta_p \Delta\Theta_p$ against x , constructed under the assumption that the anisotropy of the alloys is of the one-ion type. In the plotting of the theoretical curve we used the value $\Delta\Theta_p \text{Dy} = 58^\circ \text{K}$, which we obtained in our work, and the value $\Delta\Theta_p \text{Gd} = -0.7^\circ \text{K}$ from^{[23,4)}. The theoretical and experimental plots are close to each other. A certain difference between the experimental and theoretical plots of $\Theta_p \Delta\Theta_p$ against x , which is particularly noticeable at small x , can be attributed to the change of the parameter of the crystal field acting on the dysprosium when the alloy composition is changed (for more details, see^[12]). The theoretical plot, constructed with allowance for this circumstance, gives better agreement with the experimental data (Fig. 4). Thus, an investigation of the dependence of the anisotropy of the paramagnetic Curie temperature on the composition of dysprosium-gadolinium alloys confirms the previously drawn conclusion^[12] that the anisotropy of these alloys is of the single-ion type.

From the data on the paramagnetic susceptibility we can estimate the coefficient of uniaxial anisotropy $J_2^0(0)$ at 0°K . For single-ion anisotropy^[22] we have

$$K_2^0(0)_i = \frac{5}{3} b_i \mu_{0i}^2. \quad (24)$$

It follows from our measurements of the anisotropy of the paramagnetic Curie temperature that $b = 0.35 \text{ cm}^{-1}/\mu_B^2$ for dysprosium, from which we obtain $K_2^0(0) = 59 \text{ cm}^{-1}$, in good agreement with estimates obtained by others^[18,23] from measurements in the paramagnetic region. This value, however, is smaller by an approximate factor of 1.5 than the value determined from measurements of the anisotropy of dysprosium in the magnetically ordered state^[12]. The reasons for this

⁴⁾We call attention to the fact that in the discussion of the properties of dysprosium-gadolinium alloys in the paramagnetic region one must not neglect the anisotropy of the gadolinium (as was done in the interpretation of the results of the measurement of these alloys in the magnetically-ordered state^[12]). The reason is that the uniaxial anisotropy of gadolinium has an anomalous temperature dependence and goes through a maximum near the temperature Θ_2 ^[23]. We note also that, since $\Delta\Theta_p \text{Dy} \gg \Delta\Theta_p \text{Gd}$, the theoretical plot constructed under the assumption that the gadolinium has single-ion anisotropy is close to the plot obtained under the assumption that the anisotropy of gadolinium is due to exchange interaction.

⁵⁾The anisotropy of $\mu_{\text{alloy,eff}}$ of dysprosium-gadolinium alloys is small (theoretical estimates show that it does not exceed 3–4%), and is therefore not observed at our measurement accuracy.

difference are not yet clear. It is possible that this difference is due in part to the influence of magnetoelastic effects, the contribution of which was not taken into account in the derivation of (24).

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