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Translated by J. G. Adashko

Nature of magnetic anisotropy of dysprosium. Paramagnetic susceptibility of dysprosium-yttrium alloys

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(Submitted June 24, 1976)
Zh. Eksp. Teor. Fiz. 71, 2381–2388 (December 1976)

The magnetic susceptibility of single-crystal dysprosium-yttrium alloys was measured in the basal plane and along the hexagonal axis. It is shown that the susceptibility of these alloys obeys the Curie-Weiss law, the effective magnetic moments along the different directions are equal, and the paramagnetic Curie temperatures are different. The difference between the paramagnetic Curie temperatures in the basal plane and along the hexagonal axis does not depend on the dysprosium concentration in the alloys. This, as revealed by comparison with the theoretical models of magnetic anisotropy demonstrates that the magnetic anisotropy of dysprosium-yttrium alloys is of the one-ion type.

PACS numbers: 75.30.Cr, 75.30.Gw

INTRODUCTION

Important information on the contribution of the two principal mechanisms (one-ion anisotropy and exchange anisotropy) to the magnetic anisotropy of heavy rare earth metals (HREM) can be obtained by investigating alloys of HREM with one another. If the magnetic anisotropy is of one-ion origin, then it is made up additively of the anisotropies of the individual atoms and is a linear function of the alloy-component concentrations. But if the magnetic anisotropy is due to anisotropic exchange, then its magnitude is proportional to the number of pair interactions, i.e., it depends quadratically on the concentration of the alloy components.

The anisotropy of HREM depends strongly on the deviations of the real crystal structure from the ideal hcp structure: in the point-charge model, the coefficient of uniaxial anisotropy (when the anisotropy energy is expanded in harmonic polynomials) is for both anisotropy mechanisms[1,2]

$$k_2^0 \propto \eta = a^{-3}(1.633 - c/a). \tag{1}$$

To discuss the experimental result it is therefore necessary to take into account the change of the crystal lattices parameters a and c with changing alloy composition.

Starting with the premises set forth above, we have reduced in[3,4] the experimental data on the magnetic anisotropy of dysprosium-gadolinium alloys in the magnetically ordered state and in the paramagnetic region, and have shown that the concentration dependence of k_2^0 of these alloys can be described within the framework of the single-ion model. It was therefore concluded in those papers that the magnetic anisotropy of dysprosium is single-ion and is due to the interaction of the anisotropic cloud of $4f$ electrons with the crystal lattice field. Recently, however, Boutron,[2,5] using essentially the same concept, analyzed the experimental data on the anisotropy of dilute HREM alloys in gadolinium[6] and of pure HREM,[7–9] and arrived at the opposite conclusion, that a substantial contribution to the magnetic anisotropy of HREM (and, in particular, dysprosium) is made by the anisotropy due to exchange interaction: according to his calculations, it amounts to more than 50% of the one-ion anisotropy.

To determine more accurately the roles played by the various mechanisms in the magnetic anisotropy of dysprosium, and to ascertain the cause of the fundamental difference between our conclusions concerning the nature of the anisotropy of this metal and those of Boutron, we have investigated the magnetic anisotropy of dysprosium-yttrium alloys in the paramagnetic state. Yttrium has a zero magnetic moment and is a Pauli paramagnet. Therefore its susceptibility in the alloy can be regarded as an additive increment to the susceptibility due to the dysprosium, and this facilitates the calculation of the magnetic anisotropy of the alloy from the experimental anisotropy of the paramagnetic susceptibility. In addition, in dysprosium-yttrium alloys the coefficient η, which characterizes the deviation of the crystal structure of the alloy from the ideal hcp lat-

TABLE I.

Parameter	x					
	0	0.10	0.30	0.50	0.70	1.0
a, Å	3.646	3.645	3.633	3,621	3.609	3.592
c, Å	5,741	5.732	5.714	5.696	5,678	5.650
c/a	1.573	1.573	1.573	1.573	1.573	1.573
$10^3\eta$, Å^{-3}	−1.25	−1.25	−1.26	−1.27	−1.28	−1.29
$\alpha_{bp}\cdot 10^6$	5.9	5.9	5.5	5.6	5.8	5.0
$\alpha_c\cdot 10^6$	20.5	20.0	20.5	20.3	20.0	20.0
$\beta\cdot 10^6$	−398	−385	−408	−402	−390	−410

tice, is practically independent of the composition. Therefore, in contrast to dysprosium-gadolinium alloys, there is no need for introducing a correction for the change in the crystal structure in the theoretical analysis of the concentration dependence of the magnetic anisotropy of dysprosium-yttrium alloys.

SAMPLES AND MEASUREMENT PROCEDURE

Single crystals of the alloys Dy_xY_{1-x} (x = 0.10; 0.30; 0.50; 0.70; 1.0) were grown by the crucibleless zone melting method with induction heating in an atmosphere of helium in the laboratory for rare metals and alloys of the Metallurgy Institute of the USSR Academy of Sciences. The composition of the alloys and their homogeneity were monitored with the aid of an atomic-absorption analysis and by the electric probe method.

The coefficients of the thermal expansion of the alloys in the basal plane (α_{bp}) and along the hexagonal axis (α_c) were measured in the temperature range 300–900 °K with the aid of the quartz dilatometer of the "Metallurgy combine" installation developed at the Metallurgy Institute.[10] Measurements of the paramagnetic susceptibility of the single crystals of the alloys in the basal plane (χ_{bp}) and along the hexagonal axis (χ_c) were made with a vibration magnetometer in fields up to 15 kOe in the temperature interval 78–400 °K, on samples in the form of spheres of 2.5–3 mm diameter. The orientation of the samples was effected by a magnetic method and was monitored by an x-ray method. The experimental susceptibility was represented in the form

$$\chi_{\text{alloy}}=\chi_Y+\chi_{Dy}, \tag{2}$$

where χ_Y is the susceptibility of the yttrium and χ_{Dy} is the susceptibility due to the presence of dysprosium in the alloy. By using the values of χ_Y from[11] we obtained χ_{Dy}.$^{1)}$

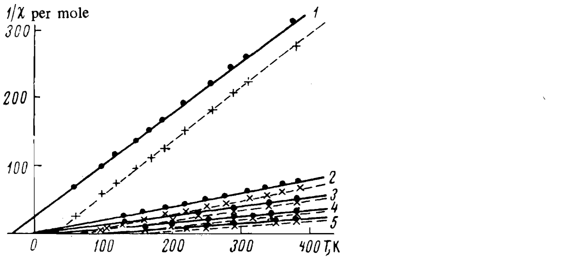

FIG. 1. Dependence of the reciprocal paramagnetic susceptibility of single-crystal Dy_xY_{1-x} alloys on the temperature: dashed lines—in the basal plane, solid lines—along the hexagonal axis. 1) x = 0.10, 2) x = 0.30, 3) x = 0.50, 4) x = 0.70, 5) x = 1.0.

EXPERIMENTAL RESULTS

Table I lists the parameters of the crystal lattice of the dysprosium-yttrium alloys in accordance with the data of Markova and Finkel',[12,13] and also the coefficients of the thermal expansion of these alloys.[10] The table shows also the ratio c/a and the parameter η that characterizes the deviation of the real crystal structure of the alloy from the ideal hexagonal structure. Although the lattice constants a and c vary somewhat with changing alloy concentration, the ratio c/a is almost independent of x, so that the parameter η remains unchanged when the alloy composition changes. This parameter does depend, however, on the temperature. This dependence can be characterized by a temperature coefficient β equal to

$$\beta=\frac{1}{\eta}\frac{d\eta}{dT}=\frac{(\alpha_c-\alpha_{bp})\,c/a}{1.633-c/a}-3\alpha_{bp}\,. \tag{3}$$

The values of β for dysprosium-yttrium alloys, calculated from the experimental values of the lattice parameters and the thermal-expansion coefficients, are given in Table I, and likewise depend little on the alloy concentration.

Figure 1 shows the temperature dependences of the paramagnetic susceptibility χ_{Dy}. It is seen that the susceptibility obeys the Curie-Weiss law and can be represented in the form (per dysprosium atom)

$$\chi_{Dy}=\mu^2_{\text{eff alloy}}/3k(T-\Theta_p), \tag{4}$$

where $\mu_{\text{eff alloy}}$ is the effective magnetic moment of the alloy due to the dysprosium, and Θ_p is the paramagnetic Curie temperature. Table II gives the values of the paramagnetic Curie temperatures and the effective magnetic moments of the investigated alloys in the basal plane and along the hexagonal axis. Plots of $\Theta_{p\,bp}$ and Θ_{pc} on the alloy concentration are also shown in Fig. 2.

As follows from our data, the paramagnetic Curie temperatures in the basal plane and along the hexagonal axis increase with increasing dysprosium content, and for all the alloys we have $\Theta_{p\,bp}>\Theta_{pc}$, while the difference $\Delta\Theta_p=\Theta_{p\,bp}-\Theta_{pc}$ is independent of the concentration within the limits of experimental error. The effective magnetic moment of the alloy in the basal plane and along

TABLE II.

Parameter	x				
	0.1	0.3	0.5	0.7	1.0
$\Theta_{p_{bp}}$, K	33±2	65±2	82±2	116±2	164±2
Θ_{pc}, K	−30±2	0±2	20±2	51±2	107±2
$\Delta\Theta_p=\Theta_{p_{bp}}-\Theta_{pc}$, K	63±4	65±4	62±4	65±4	57±4
$\mu_{\text{eff bp}}/\mu_B$	10.6±0.3	11.2±0.3	10.6±0.3	10.8±0.3	10.6±0.3
$\mu_{\text{eff }c}/\mu_B$	10.8±0.3	11.5±0.3	11.1±0.3	11.3±0.3	10.9±0.3
$\Delta\Theta_p=\frac{\mu^2_{\text{eff}}}{3k}\left(\frac{1}{\chi_c}-\frac{1}{\chi_{bp}}\right)$, K (at 300 K)	49±5	52±5	44±5	47±5	46±5

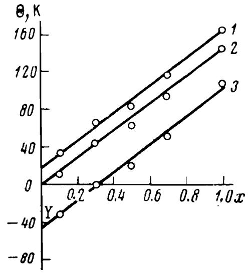

FIG. 2. Dependence of the p paramagnetic Curie temperatures of single crystal Dy_xY_{1-x} alloys on the dysprosium concentration x: 1) $\Theta_{p\,bp}$, 2) Θ_{p0}, 3) Θ_{pc}.

the hexagonal axis is approximately the same and does not depend on the dysprosium content.

DISCUSSION OF RESULTS

In the molecular-field approximation, the paramagnetic susceptibility of a two-compolnent rare-earth alloy, with account taken of the magnetic anisotropy, was calculated by us earlier.[4] It was shown that the susceptibility of such an alloy obeys the Curie-Weiss law, and the effect of the anisotropy reduces to making the paramagnetic Curie temperature dependent on the orientation of the field relative to the hexagonal axis. Thus, the formula for the magnetic susceptibility of a two-component anisotropy alloy can be represented in the form (4), where Θ_p, and consequently also χ, are functions of the angle φ between the hexagonal axis of the crystal and the field direction.

The effective magnetic moment and the magnetic Curie temperature depend on the properties of the alloy components. In the case of dysprosium-yttrium alloys, when one of the alloy components (yttrium) has a zero magnetic moment, the expressions given in[4] for $\mu_{\text{eff alloy}}$ and Θ_p become much simpler and can be represented in the form

$$\mu_{\text{eff alloy}}=\mu_{\text{eff Dy}},\quad \Theta_p(\varphi)=\Theta_{p0}-{}^1/_3(3\cos^2\varphi-1)\Delta\Theta_p,$$
$$\Theta_{p0}=\frac{I_0}{3k}\mu^2_{\text{eff Dy}}\left(\frac{g_{\text{Dy}}-1}{g_{\text{Dy}}}\right)^2 x,\quad \Delta\Theta_p=\frac{1}{k}(a_{\text{Dy}}x+b_{\text{Dy}})\mu^2_{\text{eff Dy}}, \tag{5}$$
$$\mu_{\text{eff Dy}}=g_{\text{Dy}}[J_{\text{Dy}}(J_{\text{Dy}}+1)]^{1/2}.$$

Here I_0 is the dysprosium-dysprosium exchange-interaction coefficient, while the coefficients a_{Dy} and b_{Dy} characterize respectively the exchange magnetic anisotropy and the one-ion anisotropy, and are connected with the coefficient of the magnetic anisotropy of the alloy k_2^0 by the relation[14]

$$k_2^0=a_{\text{Dy}}\mu^2_{0\text{Dy}}x^2+{}^5/_3\, b_{\text{Dy}}\mu^2_{0\text{Dy}}x, \tag{6}$$

where $\mu_{0\text{Dy}}=g_{\text{Dy}}J_{\text{Dy}}$ is the saturation moment of the dysprosium in the alloy. It follows from formulas (5) that

$$\Theta_{p0}={}^1/_3(2\Theta_{p\,pb}+\Theta_{pc}),\quad \Delta\Theta_p=\Theta_{p\,pb}-\Theta_{pc}. \tag{7}$$

We note that relations (7) were obtained under the assumption that the constants a_{Dy} and b_{Dy} do not depend on the temperature. This is not the actual case, inasmuch as the parameter η varies with temperature as a result of thermal expansion. This leads to a temperature dependence of $\Delta\Theta_p$, so that the values of Θ_{p0} and $\Delta\Theta_p$ obtained from the experimental data differ somewhat from the theoretical ones determined from formulas (5). Estimates show, however, that this difference is small: the thermal expansion increases the experimental values of $\Delta\Theta_p$ by not more than 3–4 °K, and the experimental values of Θ_{p0} by less than 2 °K in comparison with the theoretical values determined by relations (5) and (7). Since these systematic errors are less than the random measurement errors, we introduced no corrections for the thermal expansion.

In addition, we have calculated (see Table II) $\Delta\Theta_p$ from the data on anisotropy of the paramagnetic susceptibility of the alloys at 300 °K by the method proposed by Boutron[5]

$$\Delta\Theta_p=\frac{\mu^2_{\text{eff alloy}}}{3k}\left(\frac{1}{\chi_c}-\frac{1}{\chi_{bp}}\right). \tag{8}$$

The quantities $\Delta\Theta_p$ obtained by this method are determined by the values of a_{Dy} and b_{Dy} at room temperature and are somewhat smaller than the values $\Delta\Theta_p$ calculated from the difference of the paramagnetic Curie temperatures and characterizing these parameters at 0 °K as a result of the temperature dependence of a_{Dy} and b_{Dy}.

The experimental data on the paramagnetic susceptibility of the dysprosium-yttrium alloys agree well with the theoretical relations (5) and (7): the $\mu_{\text{eff alloy}}$ are independent, within the limits of errors, of the composition of the alloy and are close to the theoretical value of the effective magnetic moment of dysprosium (10.65 μ_B), while Θ_{p0} increases linearly with increasing concentration of the dysprosium in the alloy (see Table II and Figure 2).

Of greatest interest is the concentration dependence of the difference of the paramagnetic Curie temperatures $\Delta\Theta_p$, inasmuch as from this dependence we can draw conclusions concerning the nature of the magnetic anisotropy of the dysprosium in dysprosium-yttrium alloys. It can be assumed that in dysprosium-yttrium alloys the parameters a_{Dy} and b_{Dy} do not depend on the dysprosium concentration, since the coefficient η hardly changes with the concentration (see Table I). Consequently, as seen from formula (5), if $\Delta\Theta_p$ is constant in the alloys, then the magnetic anisotropy is determined by the parameter b_{Dy} and is of the one-ion type, and if $\Delta\Theta_p\sim x$, then the magnetic anisotropy depends on the parameter a_{Dy} and is due to the anisotropic exchange interaction.

It is seen from Table II that, with an error not exceeding 8–10%, the quantity $\Delta\Theta_p$ (whether determined from (7) or from (8)) does not depend on the concentration of the alloy. Consequently, it can be concluded from our data that in dysprosium-yttrium alloys the magnetic anisotropy is of the one-ion type. Thus, measurements of the anisotropy of the paramagnetic susceptibility of dysprosium-yttrium alloys confirm the conclusion that the anisotropy of the dysprosium is of the one-ion type; this conclusion was drawn by us earlier[3,4] in a study of dysprosium-gadolinium alloys.

Knowing $\Delta\Theta_p$, we calculated from formulas (5) and (6) the value of the coefficient of uniaxial anisotropy of the

TABLE III.

Element	k_2^0 at 0 °K (10^8 erg/cm^3)				
	From measurements in the magnetically ordered state*			From measurements of the anisotropy of the paramagnetic susceptibility	
Dy	4.1±1.6	[15–17] **	a)	3.3±0.6	[17]
	5.5±1.2	[18]	b)	3.2±0.3	[26]
	5.5±1.1	[19, 20]	a)	3.5	[8]
	4.9	[21]	a)	3.7±0.3	our data
Tb	3.3±1.3	[15–17] **	a)	2.9±0.3	[26]
	5.5±1.2	[18]	b)	3.17	[8]
	5.56±1.1	[19, 20]	a)		
	5.3±0.5	[22]	c)		
	3.1±0.5	[23]	c)		
	4.5	[24]	a)		
Ho	1.5±0.2	[25] **	a)	0.95±0.18	[26]
	2	[24]	a)		
	4.16±1.1	[19, 20]	a)		
Dy in the alloy $Dy_{0.046}Gd_{0.954}$	3.6±0.5	[3]	a)	2.2±0.4	[4]

*Obtained from magnetization measurements (a), from torque measurements (b), and from ferromagnetic-resonance measurements (c).
**Extrapolation from the region of nitrogen temperatures.

dysprosium k_2^0 at 0 °K. It is given in Table III. The same table gives the values of the coefficient of uniaxial anisotropy of dysprosium, terbium, holmium, and the alloy $Dy_{0.046}Gd_{0.954}$, calculated from the published data on the anisotropy of the paramagnetic susceptibility, and also measured directly in the magnetically ordered state by various methods. We call attention to the fact that, as already noted earlier,[4] the values of k_2^0 at 0 °K, determined from the paramagnetic susceptibility, are smaller by an approximate factor 1.5 than those measured directly at this temperature. The reason for this large difference is still not clear. It cannot be attributed to a change in the parameter b_{HREM} with changing temperature as a result of thermal expansion, since estimates show that in the temperature interval 0–300 °K the parameter η decreases for pure HREM by only 15–20%, while for the alloy of dysprosium with gadolinium it even increases (the coefficient β for this alloy is positive). It is possible that this difference is due to the approximate character of the theory of the temperature dependence of magnetic anisotropy,[14] from which formula (6) was deduced, and also to the fact that formulas (1) and (3), which describe in the point-charge approximation the influence exerted on the magnetic anisotropy of HREM by the thermal expansion, are not exact.

Be it as it may, the difference between the coefficients of single-ion anisotropy (or, equivalently, the crystal-field parameters) of HREM, obtained from measurements in the paramagnetic state and in the magnetically-ordered region, must be taken into account when reducing the experimental data, inasmuch as neglect of this difference can lead to incorrect conclusions. This was exactly the case in Boutron's comparison[2,5] of the anisotropy of dilute alloys of HREM in gadolinium in the magnetically ordered state (at 0 °K) with the anisotropy of pure HREM in the paramagnetic region, namely, he neglected this difference and assumed that the one-ion anisotropy calculated from these measurements is the same in alloys as in pure HREM (with allowance for the change of the crystal-lattice parameters), and the difference between the anisotropy of pure HREM and the anisotropy of the alloys is due to the contribution made to the anisotropy by the anisotropic exchange interaction. In fact, this difference, as shown by the foregoing data, is due to the difference between the coefficients of the one-ion anisotropy calculated from measurements in the magnetically ordered and in the paramagnetic states.

We thank K. P. Belov, E. M. Savitskiĭ, and V. F. Terekhov for interest and attention, and S. A. Nikitin for valuable discussions.

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Translated by J. G. Adashko