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Nature of the Magnetic Anisotropy of Dysprosium: Investigation of the Anisotropy of Dysprosium-Gadolinium Alloys

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The magnetization of monocrystals of Dy_xGd_{1-x} alloys ($0.046 \leq x \leq 0.49$) was measured in pulsed magnetic fields up to 280 kOe, along various crystallographic directions. From the data obtained, the coefficient of uniaxial anisotropy, k_2^0, was determined. It is shown that the experimental dependence of the anisotropy coefficient k_2^0 on temperature and on alloy composition agrees, within the limits of experimental error, with the theoretical dependence for the single-ion anisotropy model.

INTRODUCTION

ONE of the characteristic properties of heavy rare-earth metals (HREM) is a huge uniaxial magnetic anisotropy. In recent years it has been shown experimentally[1-7] that in HREM (except gadolinium) the uniaxial magnetic anisotropy energy exceeds 10^8 erg/cm^3, which is about two orders of magnitude larger than the anisotropy of ferromagnets of the iron group. This large anisotropy of HREM is explained qualitatively by the fact that in these metals, because of strong localization of the electrons of the unfilled 4f-shell, the spin-orbit interaction is significantly larger than the interaction of the orbital moment of the 4f-electrons with the crystalline field of the lattice; the total angular momentum $J = L + S$ is a "good" quantum number, and upon rotation of the magnetic moment in the field there occurs a change of orientation not only of the spin moment S, but also of the orbital moment L.

At present two mechanisms for the uniaxial magnetic anisotropy of HREM have been treated in detail theoretically: 1) electrostatic interaction of the anisotropic cloud of charges of the 4f-electrons with the crystalline field of the lattice—single-ion anisotropy (SIA)[8-11]; 2) anisotropy of the indirect exchange interaction of the 4f-electrons with nonvanishing orbital moment via the conduction electrons—exchange-interaction anisotropy (EIA)[12-16]. Both of these mechanisms give for the magnitude of the uniaxial anisotropy of HREM an estimate consistent with the experimental data, and at present the problem of the contribution of each of these mechanisms to the anisotropy of HREM remains unsolved. This problem, as was shown in a paper of Irkhin[17], can in principle be clarified by an analysis of the dependence of the magnitude of the uniaxial anisotropy constant of HREM on the number of the rare-earth element (in other words, on the quantum numbers J, L, S), since this dependence is different for the SIA and EIA models. But this difference, for the majority of TREM (except thulium, for which the anisotropy in the magnetically ordered state has not been measured), is small and lies within the limits of experimental error[1]. Measurements of the temperature dependence of the anisotropy of HREM give evidence in favor of the SIA model. For terbium, dysprosium[1], and holmium[6] the temperature dependence of the first anisotropy coefficient k_2^0 (in an expansion of the anisotropy energy in harmonic polynomials) agrees better with the temperature dependence given by the SIA model than with that given by the EIA model. At the same time, recent investigations of the magnetic structure of rare-earth alloys[20], and also experiments on inelastic scattering of neutrons on terbium[21], indicate that exchange-interaction anisotropy has an important effect on a number of properties of HREM.

A direct answer to the question of the nature of the magnetic anisotropy of HREM can be obtained by investigating the anisotropy of alloys of HREM with each other. If the magnetic anisotropy is single-ion, then it is the sum of additive contributions from the anisotropy of the separate atoms, and consequently it should be, in the alloy, a linear function of the concentrations of the components. But if the anisotropy is due to anisotropic exchange interaction, then its magnitude is proportional to the number of pair interactions in the alloy; that is, it should be a quadratic function of the concentration of the components. It is most convenient to use, as one of the components of the alloy, gadolinium. Because the trivalent gadolinium ion is in an S-state and has zero orbital moment, the anisotropy of gadolinium is appreciably smaller than the anisotropy of the other HREM[7], and it may be treated as a magnetically isotropic matrix. Consequently, the anisotropy of alloys of gadolinium in the SIA case will be proportional to the concentration x of the anisotropic component (the anisotropy per atom of the anisotropic component is independent of x), whereas in the EIA case it will vary quadratically with x (the anisotropy per atom of the anisotropic component is proportional to x)[2]. In investigation of the anisotropy of alloys, it is necessary to pay attention to the change of the crystalline-field parameters in the alloy as compared with these parameters in the pure metals.

In the present research, in order to clarify the nature of the magnetic anisotropy of HREM, an investigation was made of the uniaxial magnetic anisotropy of

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[1] Recent data on the anisotropy of the paramagnetic susceptibility of thulium[18] agree better with the SIA model. But Irkhin and Karpenko[19] have shown that just as good an agreement can be obtained also with the EIA model if one takes account of exchange via conduction electrons with a nonvanishing orbital moment.

[2] Very recently, in papers of Japanese authors[22], the anisotropy of dilute (1–2%) alloys of gadolinium with other rare-earth metals was investigated, and it was shown that the anisotropy per atom of dysprosium and terbium in the alloy was smaller by a factor of about two than in the pure metal.

alloys of gadolinium with a varying content of dysprosium. Dysprosium was chosen because, first, data exist on the anisotropy of this metal[1,2,5]; second, in gadolinium-dysprosium alloys a helicoidal structure either is absent or is broken down in comparatively weak fields[23] and can be disregarded in calculation of the anisotropy; third, dysprosium has a comparatively high vapor pressure and is easily cleansed by the method of distillation.

SPECIMENS; EXPERIMENTAL METHOD

Monocrystals of Dy_xGd_{1-x} alloys were grown by the method of crucibleless zone fusion, with induction heating, in an atmosphere of helium. The original materials were first cleansed of impurities by vacuum distillation. The composition of the alloys and their homogeneity were determined by means of atomic absorption analysis. Monocrystals of Dy_xGd_{1-x} alloys were grown with x = 0.046, 0.103, 0.183, 0.296, 0.378, and 0.492. Specimens for magnetization measurements, in the form of bars with dimensions 7 × 1.4 × 1.4 mm, oriented parallel to the hexagonal axis of the crystal and parallel to the basal plane, were cut from the monocrystals by the electric-spark method. The orientation of the specimens was accomplished by an x-ray method, by the method of inverse mapping. The accuracy of the orientation was 2–3°. The magnetic anisotropy of the alloys was determined by measurement of the magnetization along various crystallographic directions. The measurements of magnetization were made in pulsed magnetic fields up to 280 kOe in the temperature interval 4.3–300°K (on alloys with x = 0.378 and 0.492, along the hexagonal axis, in the temperature interval 78–300°K), by the induction method, with use of differential coils[24]. The accuracy of measurement of the absolute value of the magnetization was 10–12%; the relative values of the magnetization (as a function of the field, the temperature, and the composition) were determined with accuracy 6–8%.

EXPERIMENTAL RESULTS

Figure 1 shows the field dependence of the magnetization of alloys with various contents of dysprosium, in the basal plane and along the hexagonal axis, at temperature 78°K. Similar dependences were obtained at other temperatures. It is seen that for all the alloys, the basal plane is a plane of easy magnetization: saturation in this plane is attained in weak fields. The hexagonal axis of the crystal is an axis of difficult magnetization: the magnetization in this direction increases, with increase of field, considerably more slowly than in the basal plane, and it becomes saturated upon attainment of a saturation field H_S whose magnitude increases with increase of the dysprosium content in the alloy. From the measurements of magnetization in the basal plane, the magnetic saturation moments of the alloys were determined. Within the limits of experimental error, the dependence of the saturation moment (per atom) on the composition of the alloy can be represented in the form

$$\mu_{\text{alloy}} = x\mu_{0\text{Dy}} + (1 - x)\mu_{0\text{Gd}}, \qquad (1)$$

where $\mu_{0\text{Dy}} = 10.2\ \mu_B$, $\mu_{0\text{Gd}} = 7.5\ \mu_B$ are the saturation moments obtained from measurements on the pure metals[11].

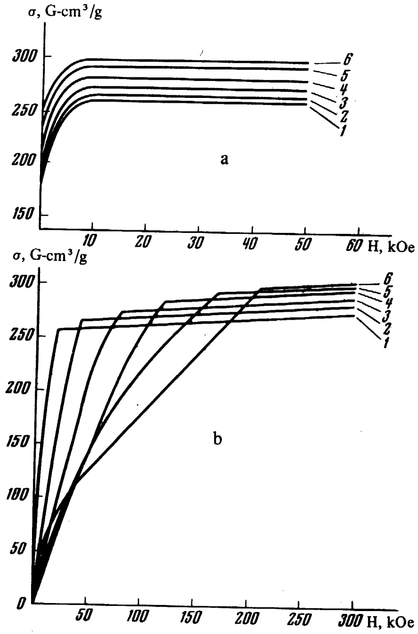

FIG. 1. Field dependence of the magnetization of Dy_xGd_{1-x} alloys 78°K: a, field H parallel to the basel plane; b, field H parallel to the hexagonal axis. 1, x = 0.046; 2, x = 0.103; 3, x = 0.183; 4, x = 0.296; 5, x = 0.378; 6, x = 0.492.

DISCUSSION OF EXPERIMENTAL RESULTS

A. Determination of Anisotropy Coefficients; Peculiarities of Magnetization Processes in Rare-Earth Alloys

The experimentally obtained dependence of the magnetization on the field enables us to calculate the uniaxial-anisotropy constants of the alloys. Usually[25] in the calculation of magnetization curves along various directions, attention is paid only to the uniaxial-anisotropy energy and to the Zeeman energy. But as was shown earlier[6], such a calculation is correct only when the anisotropy energy is much smaller than the exchange-interaction energy. In rare-earth metals and alloys, the magnetic anisotropy energy is comparable with the exchange-interaction energy[26], and this leads to a number of peculiarities in the interpretation of magnetization curves of these materials.

First[6], in this case it may not be supposed, as is usually done, that the magnetization in fields smaller than the saturation field is independent of the field. This effect shows up most strongly on magnetization curves near the magnetic transition temperature; and estimates show that in the alloys investigated, it may be neglected, within our accuracy of measurement, at temperatures 20–30°K below the Curie temperature. Therefore we shall ignore this region near the Curie temperature.

Second, as was first noted in[22], upon magnetization in a direction of difficult magnetization in an alloy consisting of two components with different anisotropy, a noncollinear magnetic structure will occur. This phenomenon is due to the fact that the magnetic moments of the components of the alloy are acted upon by differ-

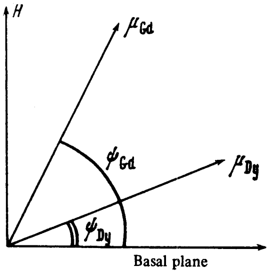

FIG. 2. Orientation of the magnetic moments of the dysprosium and of the gadolinium in the alloy, in a field parallel to the hexagonal axis.

ent anisotropy fields; and upon rotation of the magnetic moments in an external field, as a result of "competition" of the anisotropy fields and the exchange interaction, an angle between the magnetic moments of the different components will be produced. In this case, in order to describe the magnetization processes in a field parallel to the hexagonal axis, it is necessary to take into account, in the expression for the free energy, not only the Zeeman energy and the anisotropy energy, but also the energy of exchange interaction between the components of the alloy:

$$\begin{aligned}E = &-I_{\text{Dy-Gd}}x(1-x)\mu_{\text{Gd}}\mu_{\text{Dy}}\cos(\psi_{\text{Gd}}-\psi_{\text{Dy}})\\ &+[({}^{3}/{}_{2}k_2{}^0-{}^{15}/{}_{4}k_4{}^0)\sin^2\psi_{\text{Dy}}+{}^{35}/{}_{8}k_4{}^0\sin^4\psi_{\text{Dy}}]\\ &-[Hx\mu_{\text{Dy}}\sin\psi_{\text{Dy}}+H(1-x)\mu_{\text{Gd}}\sin\psi_{\text{Gd}}].\end{aligned} \tag{2}$$

Here the first term describes the exchange interaction between the dysprosium and gadolinium atoms in the alloy; $I_{\text{Dy-Gd}}$ is the molecular-field coefficient, ψ_{Dy} and ψ_{Gd} are the angles between the directions of the magnetic moments of the dysprosium and of the gadolinium in the basal plane (Fig. 2), and μ_{Dy} and μ_{Gd} are the magnetic moments per atom. The second term is the anisotropy energy of the alloy (we assume that the anisotropy is entirely due to the presence of dysprosium in the alloy); k_2^0 and k_4^0 are the anisotropy coefficients in the expansion of the anisotropy energy in harmonic polynomials. The third term is the Zeeman energy of interaction of the magnetic moments with the field.

From the equilibrium conditions $\partial E/\partial\psi_{\text{Dy}} = \partial E/\partial\psi_{\text{Gd}} = 0$ it is easy to obtain the following equations for the equilibrium directions of the magnetic moments in a field:

$$I_{\text{Dy-Gd}}\mu_{\text{Dy}}\sin(\psi_{\text{Gd}}-\psi_{\text{Dy}}) = H\cos\psi_{\text{Gd}}, \tag{3}$$

$$\begin{aligned}&(3k_2{}^0-{}^{15}/{}_{2}k_4{}^0)\sin\psi_{\text{Dy}}\cos\psi_{\text{Dy}}+{}^{35}/{}_{2}k_4{}^0\sin^3\psi_{\text{Dy}}\cos\psi_{\text{Dy}}\\ &= H[\mu_{\text{Dy}}x\cos\psi_{\text{Dy}}+\mu_{\text{Gd}}(1-x)\cos\psi_{\text{Gd}}].\end{aligned} \tag{4}$$

The quantity measured in our experiments—the magnetization of the alloy along the hexagonal axis—is (Fig. 2)

$$\mu_c = \mu_{\text{Dy}}x\sin\psi_{\text{Dy}}+\mu_{\text{Gd}}(1-x)\sin\psi_{\text{Gd}}. \tag{5}$$

If the values of $I_{\text{Dy-Gd}}$ and of μ_{Dy} and μ_{Gd} are known, equations (3) and (5) enable us to find the angles ψ_{Dy} and ψ_{Gd}; and knowing these, we can find from equation (4) the uniaxial-anisotropy coefficients k_2^0 and k_4^0 of the alloy. Thus in order to calculate the anisotropy coefficients of the alloys, it is necessary to find $I_{\text{Dy-Gd}}$, μ_{Dy}, and μ_{Gd}. In the molecular-field approximation, the magnetic moments of the components of the alloy can be described in the form

$$\begin{aligned}\mu_{\text{Dy}} &= \mu_{0\text{Dy}}B_{15/2}\left[\frac{g_{\text{Dy}}J_{\text{Dy}}}{kT}H_{\text{eff Dy}}\right],\\ \mu_{\text{Gd}} &= \mu_{0\text{Gd}}B_{7/2}\left[\frac{g_{\text{Gd}}J_{\text{Gd}}}{kT}H_{\text{eff Gd}}\right],\end{aligned} \tag{6}$$

where the effective molecular fields acting on the magnetic moments of the components of the disordered alloy are[27][3]

$$\begin{aligned}H_{\text{eff Dy}} &= x\,\mu_{\text{Dy}}I_{\text{Dy-Dy}}+(1-x)\,\mu_{\text{Gd}}I_{\text{Gd-Dy}},\\ H_{\text{eff Gd}} &= (1-x)\,\mu_{\text{Gd}}I_{\text{Gd-Gd}}+x\,\mu_{\text{Dy}}I_{\text{Gd-Dy}}.\end{aligned} \tag{7}$$

As has been shown by numerous theoretical and experimental researches (see, for example,[29]), in a number of HREM the coefficient of exchange interaction between the spins S may be considered constant. Hence it is easy to obtain the relation between the molecular-field coefficients $I_{\text{Dy-Dy}}$, $I_{\text{Gd-Gd}}$, and $I_{\text{Gd-Dy}}$:

$$I_{\text{Dy-Dy}} = \frac{g_{\text{Gd}}{}^2(g_{\text{Dy}}-1)^2}{g_{\text{Dy}}{}^2(g_{\text{Gd}}-1)^2}I_{\text{Gd-Gd}},\quad I_{\text{Dy-Gd}} = \frac{g_{\text{Gd}}(g_{\text{Dy}}-1)}{g_{\text{Dy}}(g_{\text{Gd}}-1)}I_{\text{Gd-Gd}}. \tag{8}$$

The value of $I_{\text{Gd-Gd}}$ can be found from the Curie temperature of pure gadolinium (Θ_{Gd} = 293°K):

$$I_{\text{Gd-Gd}} = \frac{3k\Theta_{\text{Gd}}}{\mu_{0\text{Gd}}g_{\text{Gd}}(J_{\text{Gd}}+1)} = 17k\ \text{deg}/\mu_{\text{B}} \tag{9}$$

and further to determine from formulas (6)—(8) the values of $I_{\text{Dy-Gd}}$, μ_{Gd}, and μ_{Dy}. We remark that the relations (8) are approximate and, as is shown by comparison with experiment, are fulfilled with an accuracy not exceeding 10%. This leads to additional errors in the determination of the anisotropy coefficients k_2^0 and k_4^0 from experimental data. Estimates show that the error in determination of k_2^0 due to this cause increases only slightly the total experimental error (the error of determination of the absolute value of k_2^0 amounts to 20—22%; the relative measurement of k_2^0 is determined with accuracy 10—12%). But this additional error is decisive in the calculation of k_4^0: the change in the value of $I_{\text{Dy-Gd}}$ (within the limits of accuracy) leads to a change not only of the value but also of the sign of k_4^0. Therefore it is impossible to determine k_4^0 from our data.

B. Nature of the Uniaxial Magnetic Anisotropy of Gadolinium-Dysprosium Alloys

Figure 3 shows the dependence on temperature of the anisotropy coefficient of gadolinium-dysprosium alloys with a larger dysprosium content; and a sharp increase of k_2^0 is observed upon lowering of the temperature. In order to explain the nature of the uniaxial anisotropy, the experimental temperature dependence of the anisotropy coefficient of the alloys was compared with the theoretical for various models. H. Callen and E. Callen[30] have shown that both SIA and EIA depend on temperature via the temperature dependence of the relative magnetization (in the present case, $m = \mu_{\text{Dy}}/\mu_{0\text{Dy}}$). For SIA, this dependence can be described in the form

$$k_2{}^0(T) = k_2{}^c(0)\hat{I}_{5/2}[\mathscr{L}^{-1}(m)] \tag{10}$$

(here $k_2^0(0)$ is the anisotropy coefficient at 0°K, $\hat{I}_{5/2}$ is the reduced hyperbolic Bessel function, $\mathscr{L}^{-1}(m)$ is the inverse Langevin function); for EIA,

$$k_2{}^0(T) = k_2{}^0(0)m^2. \tag{11}$$

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[3] Since the gadolinium and dysprosium atoms have nearly equal atomic radii, atomic ordering in gadolinium-dysprosium alloys is improbable[28].

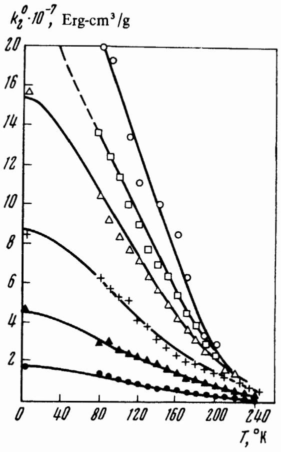

FIG. 3

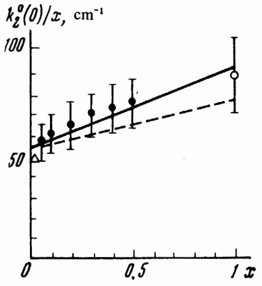

FIG. 4

FIG. 3. Temperature dependence of the coefficient of uniaxial anisotropy k_2^0 of Dy_xGd_{1-x} alloys. Points are experimental data: ●, x = 0.046; ▲, x = 0.103; +, x = 0.183; △, x = 0.296; □, x = 0.378; ○, x = 0.492. Curves are the theoretical dependence for the single-ion anisotropy model.

FIG. 4. Dependence of the coefficient of anisotropy per atom of dysprosium, $k_2^0(0)/x$, on the dysprosium concentration x. ●, our data; △, data of paper [22]; ○, data of papers [1,5]; ———, experimental dependence; - - - - - - - - -, theoretical dependence for single-ion anisotropy with allowance for the dependence of the crystalline-field parameters on the composition of the alloy.

The temperature dependence of the relative magnetization $m = \mu_{Dy}/\mu_{0\,Dy}$ was determined, in the molecular-field approximation, by formulas (6)–(9). As is seen from Fig. 3, the theoretical temperature dependence for the SIA model agrees well with the experimental curves $k_2^0(T)$ of the alloys studied. The analysis shows also that formula (11) for the EIA model does not agree with the observed dependence $k_2^0(T)$. Thus the temperature dependence of the coefficient k_2^0 of the alloys points to the single-ion character of the uniaxial anisotropy of these materials.

As has already been indicated above, one can attempt to solve the problem of the nature of the magnetic anisotropy by studying the dependence of the anisotropy on the dysprosium content. It is convenient to consider the anisotropy per atom of dysprosium. This quantity, as has already been mentioned, is independent of the concentration x in the SIA case and is proportional to x in the EIA case. Figure 4 shows the dependence of $k_2^0(0)/x$ (that is, of the coefficient of anisotropy per atom of dysprosium) on the composition of the alloy. In the same figure are plotted data of other authors for pure dysprosium[1,5] and for an alloy of gadolinium with 1.3 atomic percent of dysprosium[22]. As is seen from the figure, the coefficient $k_2^0(0)/x$ increases on increase of the dysprosium content in the alloy, and its dependence on the composition of the alloy can be represented in the form

$$k_2^0(0)/x = [(54 \pm 10) + (38 \pm 10)x]\,\mathrm{cm}^{-1}, \qquad (12)$$

the data of other authors are also included on this straight line. The first component in the expression (12) is independent of composition and describes single-ion anisotropy. As for the second component, proportional to x, it could be interpreted as anisotropy caused by exchange interaction. In our opinion, however, it is more probable that the increase of $k_2^0(0)/x$ with increase of x is caused by a change of the crystalline-field parameters on change of the concentration of the alloy. The crystalline field that acts on this dysprosium in the alloy, and consequently also the value of the single-ion anisotropy, depend on the interatomic distances. Kasuya[11] showed that for an ideal hexagonal lattice, the coefficient k_2^0 is zero, and that its value is proportional to the deviation of the lattice-parameter ratio c/a from its value for the ideal lattice ($(c/a)_{id} = 1.633$):

$$k_2^0 \sim 1{,}633 - c/a. \qquad (13)$$

The ratio c/a is 1.59 for pure gadolinium and decreases approximately linearly with increase of the dysprosium concentration in the alloy, to 1.573 for pure dysprosium[11,31]; this causes some increase of the single-ion anisotropy on increase of x. The theoretical dependence of the single-ion contribution to the anisotropy coefficient $k_2^0(0)/x$ on the dysprosium concentration, with allowance for the change of the crystalline-field parameters in the alloy, is plotted as the dotted line in Fig. 4; as is seen from the figure, it agrees with the experimental dependence within the limits of error.

We note that the theoretical treatment is approximate. The values of c/a were determined at room temperature. At low temperatures, because of spontaneous isotropic and anisotropic magnetostriction, the ratios c/a change somewhat; there also occur rhombic distortions of the crystal lattice, which are especially large in pure dysprosium[32]. Furthermore, at low temperatures there is an appreciable contribution to the uniaxial anisotropy of HREM from magnetoelastic interaction[33]. Estimates show that allowance for these facts can improve somewhat further the agreement of the experimental dependence of $k_2^0(0)/x$ on the composition of the alloy with the theoretical dependence obtained in the SIA model.

Thus the uniaxial magnetic anisotropy of dysprosium and of dysprosium-gadolinium alloys is basically single-ion, and is due to electrostatic interaction of the multiplet moments of the 4f-electrons of the dysprosium with the crystalline field of the lattice.

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215