INFLUENCE OF UNIFORM COMPRESSION ON THE MAGNETIZATION OF DYSPROSIUM AND TERBIUM

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A study was made of the influence of pressure on the magnetization of polycrystalline samples of dysprosium, whose magnetic structure was antiferromagnetic in fields $H < H_{cr}$, and on the magnetization of terbium in its ferromagnetic state. The measurements were carried out in magnetic fields up to 17 kOe. Extrema were found in the curves of the field dependence of the pressure coefficient $(\Delta\psi/\psi)/p$ of dysprosium. The fields H'_{cr}, corresponding to the extremal values of $(\Delta\psi/\psi)/p$, increased with increasing temperature up to 146.5°K < T < 169.3°K, and then decreased. The transition through this range of temperatures was accompanied by a change in the sign of $(\Delta\psi/\psi)/p$ at $H \leq H'_{cr}$ (at $T \leq 145$°K, the effect was negative, while at $T \geq 169.3$°K, it was positive). The appearance of maxima and minima of the pressure effect at the transition from the antiferromagnetic to the ferromagnetic state and the change in the sign of $(\Delta\psi/\psi)/p$ near the Néel point at $H \leq H'_{cr}$ are explained by the influence of pressure on the values of the magnetic anisotropy constants. It was found that the magnetization of terbium decreased under uniform pressure. The pressure coefficient $(\Delta\psi/\psi)/p$ in weak fields, H < 10 kOe, was large ($\approx 10^{-4}$) and varied with pressure. At H = 17 kOe, the pressure coefficient was constant and independent of temperature over the whole investigated range of temperatures from 77 to 169°K. It is suggested that the observed reduction in the strong-field magnetization is connected with a change in the value of the exchange interaction integrals under uniform compression.

1. INFLUENCE OF UNIFORM COMPRESSION ON THE MAGNETIZATION OF DYSPROSIUM

NEUTRON-diffraction investigations of the magnetic structure of dysprosium[1] and measurements of its magnetic susceptibility, carried out at various temperatures,[2] have shown that below 85°K dysprosium is ferromagnetic, between 85 and 178.5°K it is antiferromagnetic, and above 178.5°K it is paramagnetic. In the antiferromagnetic state, dysprosium has a magnetic structure of the simple helix type, with the axis parallel to the c-axis of the crystal lattice. The angle of rotation q_0 of the magnetic moments in neighboring hexagonal layers depends on temperature and varies from 30° (at T = 85°K) to 43° (at T = 178.5°K). A slight modification of this helicoidal structure is observed below 140°K [a departure from a linear dependence $q_0(T)$], which is usually ascribed to an increase in the anisotropy in the basal plane below 110°K, as discovered in previous magnetic measurements.[2]

The application of a magnetic field H in a plane perpendicular to the c-axis alters the magnetic structure. At some critical value H_{cr} of the magnetic field, the antiferromagnetic state changes to ferromagnetic.

It has been shown[3-7] that the appearance of the helicoidal structure in rare-earth metals of the yttrium subgroup may be explained on the assumption of different signs of the exchange integrals for ions in the neighboring and more distant hexagonal layers. The existence of the ferromagnetic state at low temperatures is ascribed in[3-7] to an increase in the anisotropy constant in a plane perpendicular to the axis of the helix (in the basal plane). In this case, one would expect a considerable influence of the changes in the interatomic spacing on the processes of magnetization and on the magnetic structure.

A. Results of the Measurements

The method of measurement and the method of producing pressures were described earlier.[8] The measurements were carried out on polycrystalline samples in the temperature range from 77

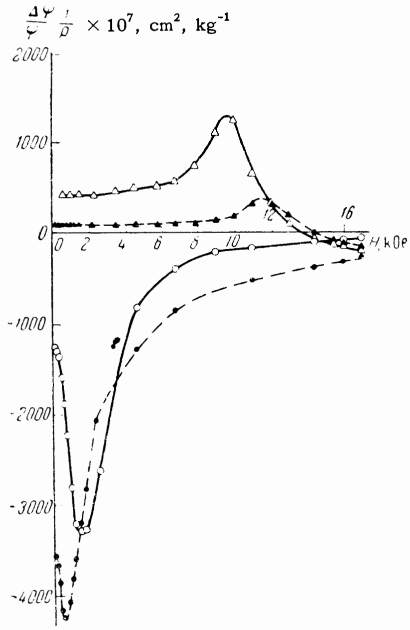

FIG. 1. Dependence of $(\Delta\psi/\psi)/p$ on the magnetic field. $T = 87.4°K$, $p = 1850$ kg/cm²: ○ – annealed samples, ● – samples subjected to a preliminary pressure of 3750 kg/cm². $T = 169.3°K$, $p = 2000$ kg/cm²: Δ – annealed samples, ▲ – samples subjected to a preliminary pressure of 3750 kg/cm².

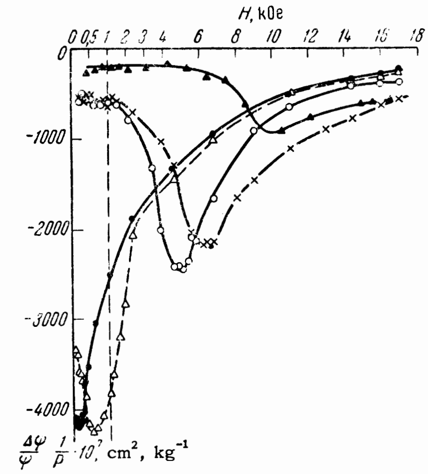

FIG. 2. Isotherms of $(\Delta\psi/\psi)/p$: ● – at $T = 77.3°K$, $p = 1820$ kg/cm²; Δ – 87.4°K, $p = 1850$ kg/cm²; ○ – 111.5°K, $p = 1900$ kg/cm²; × – 121.3°K, $p = 1900$ kg/cm²; ▲ – 146.5°K, $p = 1950$ kg/cm².

to 180°K. The samples of dysprosium were subjected to preliminary annealing in vacuum for 6 hours at 950°C, and then were slowly cooled in the furnace. The measurements showed that the effect of pressure on dysprosium (both annealed and unannealed) was irreversible at pressures higher than 2000 kg/cm²: under repeated uniform compression at 77 and 87°K, the absolute value of $\Delta\psi/\psi$ rose (here, $\Delta\psi/\psi$ is the relative change in the value of the magnetic flux of the sample under the action of pressure). Figure 1 gives the values of $\Delta\psi/\psi$ plotted against H for p = 1850 and 2000 kg/cm² at 87.2 and 169.3°K, respectively, for previously compressed and uncompressed annealed samples. After repeating the compression, a "quasi-stable" state was reached: $\Delta\psi/\psi$ ceased to rise. The value of $\Delta\psi/\psi$ was measured in such a "quasi-stable" state after preliminary compression at p = 3750 kg/cm², at 77.4°K. At all the temperatures, the measurements on dysprosium were carried out first at higher pressures (p = 3400–3700 kg/cm²), and then at lower pressures (p = 1800–2100 kg/cm²).

Figure 2 shows curves which give the dependence of $(\Delta\psi/\psi)/p$ on the magnetic field at constant temperatures and almost constant pressure (the pressure was varied from p = 1820 kg/cm² to p = 1950 kg/cm²). It follows from this figure that, in all fields in the temperature range from 77.3 to 146.5°K, the magnetization was reduced by uniform compression. At T = 111.5, 121.3, and 146.5°K in fields weaker than H_{cr}, the value of $\Delta\psi/\psi$ was independent of H. With further increase of the field intensity, negative maxima appeared in the curves, which were displaced in the direction of stronger fields by a rise in temperature. At T = 169.3°K, i.e., near the Néel point, the sign $(\Delta\psi/\psi)/p$ in weak fields ($H < H_{cr}$) changed to positive and then at H = 14.4 kOe it again became negative (Fig. 3). Above the Néel temperature (180.1°K), the sign of $(\Delta\psi/\psi)/p$ was negative in all fields.

The measurements carried out at various values of the pressure p showed that, both in the weak-field region $H < H_{cr}$ (where $\Delta\psi/\psi$ was independent of H) and in the region H > 15 kOe, the change $\Delta\psi/\psi$ was proportional to the pressure (Fig. 4). There was no proportionality to pressure in the

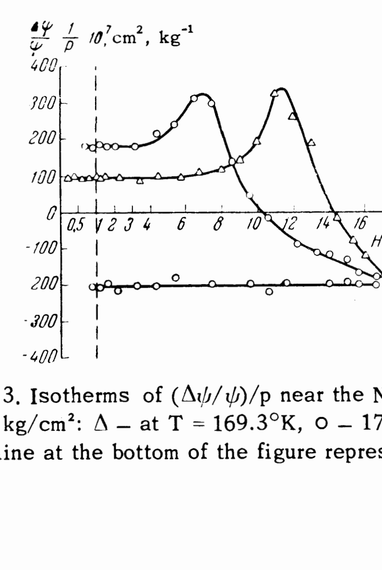

FIG. 3. Isotherms of $(\Delta\psi/\psi)/p$ near the Néel point at $p = 2000$ kg/cm²: Δ – at $T = 169.3°K$, ○ – 175.1°K. The straight line at the bottom of the figure represents 180.1°K.

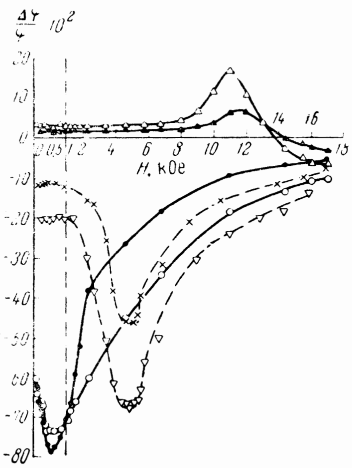

FIG. 4. Dependence of $\Delta\psi/\psi$ on the magnetic field at $T = 87.4°K$: ● – p = 1820 kg/cm², ○ – p = 3750 kg/cm²; at $T = 111.5°K$: × – p = 1900 kg/cm², ▽ – p = 3400 kg/cm²; at $T = 169.3°K$: ▲ – p = 2000 kg/cm²; Δ – p = 3400 kg/cm².

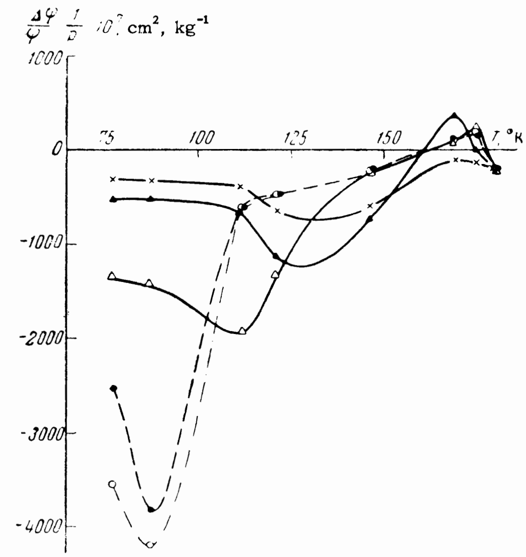

FIG. 5. Temperature dependence of $(\Delta\psi/\psi)/p$ in a constant magnetic field H: ○ – 0.4 kOe, ● – 1 kOe, Δ – 4.6 kOe, ▲ – 11 kOe, × – 16 kOe.

region of the maxima and minima referred to above.

Figure 5 shows the temperature dependence of $(\Delta\psi/\psi)/p$ in constant magnetic fields at a pressure of about 2000 kg/cm². All the curves, except the one for H = 16 kOe, exhibit two maxima corresponding to magnetic transitions: the first, the transition from the ferromagnetic to antiferromagnetic state in a given magnetic field; and the second, the transition from the antiferromagnetic state to paramagnetic. In H = 16 kOe, i.e., when the ferromagnetic structure exists almost throughout the investigated range of temperatures, the second maximum is absent.

B. Discussion of the Results

1. In contrast to gadolinium and ferromagnetic metals of the iron group, the magnetization of dysprosium in fields of 17 kOe does not reach saturation at low temperatures or at temperatures in which dysprosium is antiferromagnetic in weak fields. It is evident from Figs. 1–3 that in these fields the quantities of $(\Delta\psi/\psi)/p$ still do not reach their limiting values. It follows that over most of the investigated range of fields the changes in the magnetization under pressure may be associated with the corresponding changes in the magnetic anisotropy constant. To find how uniform compression affects the atomic magnetic moments of ferromagnetic dysprosium, it is necessary to carry out a similar study in fields of 20–30 kOe at low temperatures.

2. The values of $(\Delta\psi/\psi)/p$ of dysprosium below the Curie point Θ_C (Fig. 2) are negative, and have extremal values in relatively weak fields; moreover, their absolute values gradually decrease as the field increases, i.e., they behave as in the case of gadolinium and ferromagnetic metals of the iron group. Hence, we may conclude that at $T < \Theta_C$, the absolute values of the magnetic anisotropy constant of dysprosium increase under uniform compression (the application of pressure impedes the rotation of the magnetic moments of the domains in the direction of the field). The numerical values of the quantity $(\Delta K/K)/p$ may be determined by measurements on single crystals.

3. According to the existing theories [3-7], when the magnetic moments of dysprosium in the antiferromagnetic state rotate in the basal plane, the value of the magnetic susceptibility is governed mainly by the values of the exchange integrals J_i and, to a lesser degree, by the values of the magnetic anisotropy constants K_i. On the other hand, after the transition to the ferromagnetic state in fields $H > H_{cr}$, the susceptibility depends mainly on the values of K_i. Thus, if $(\Delta K_i/K_i)/p > (\Delta J_i/J_i)/p$, the transition from the antiferromagnetic state to ferromagnetic at $H \approx H_{cr}$ will be accompanied by an increase in the absolute value of $(\Delta\psi/\psi)/p$. It is evident from Figs. 2 and 3 that this increase is indeed observed, which proves the validity of the inequality given above. With further increase in the magnetic field intensity, $(\Delta\psi/\psi)/p$ varies in the same way as at temperatures $T < \Theta_C$, i.e., after passing through an extremal value its absolute value gradually decreases. Thus,

the maxima and minima observed in the curves of the field-dependence of $(\Delta\psi/\psi)/p$ (Figs. 2 and 3) may be explained by the influence of pressure on the values of K_i. As shown by Nagamiya, Nagata, and Kitano,[7] the value of the critical field H_{cr} depends on the anisotropy constant K′ in the basal plane and increases when the absolute value of this constant decreases. It is evident from Figs. 2 and 3 that the fields H'_{cr}, corresponding to the extremal values of $(\Delta\psi/\psi)/p$, increase as the temperature increases from the Curie point to the range $146.5 < T < 169.3$°K, and then the fields H'_{cr} decrease. The transition through this temperature range is accompanied by a change in the sign of $(\Delta\psi/\psi)/p$ (Fig. 3). The displacement of H'_{cr} toward weak fields indicates a rise in the absolute value of the magnetic anisotropy constant K′ in the basal plane, and a change of the sign of $(\Delta\psi/\psi)/p$ indicates, according to what has just been said, that the sign of $\partial|K'|/\partial p$ also changes. It is reasonable to assume that the sign of $\partial K'/\partial p$ does not change with the transition through the temperature range 146.5°K < T < 169.3°K, but that the sign of the value of K′ itself changes, and the absolute value of this quantity increases with further increase in the temperature. This immediately explains the observed variation of H'_{cr} (the displacement of H'_{cr} toward weak fields on increase of $|K'|$). The fact that in strong fields $(\Delta\psi/\psi)/p$ again becomes negative (cf. Fig. 3) obviously indicates the continued rise with pressure of the absolute values of the anisotropy constant K_0 in a plane perpendicular to the basal plane (we recall that at first the magnetic moments rotate in the basal plane under the action of the magnetic field--pressure aids this rotation at high temperatures by reducing the value of $|K'|$—and then they rotate from the basal plane toward the magnetic field direction—this rotation is impeded by pressure which increases $|K_0|$).

4. A change in the sign of $(\Delta\psi/\psi)/p$ in the temperature range 146.5°K < T < 169.3°K occurs also in fields $H < H_{cr}$, i.e., in the antiferromagnetic region. This obviously indicates that, although the value of the susceptibility χ at $H < H_{cr}$ depends primarily on the exchange integrals, the change in the susceptibility under uniform compression is due to a change in the value of K′, because $(\Delta K'/K')/p > (\Delta J_i/J_i)/p$ (K′ is the magnetic anisotropy constant in the basal plane).

5. The change in the helicoid angle q_0 under the influence of pressure [i.e., the quantity $(\Delta q_0/q_0)/p$] could be judged either from the sign of $(\Delta\psi/\psi)/p$ in sufficiently strong magnetic fields at high temperatures, or from the displacement of H_{cr} under the influence of pressure (according to[4,7], H_{cr} is proportional to q_0^4). However, since the limiting values of $(\Delta\psi/\psi)/p$ were not reached in the investigated range of fields, the conclusion that in strong fields $(\Delta\psi/\psi)/p$ becomes negative would be insufficiently justified [in very strong fields the influence of $(\Delta q_0/q_0)/p$ could be greater than the influence of $(\Delta K/K)/p$]. The accuracy of the measurement of the displacement of H_{cr} in our work was also in-

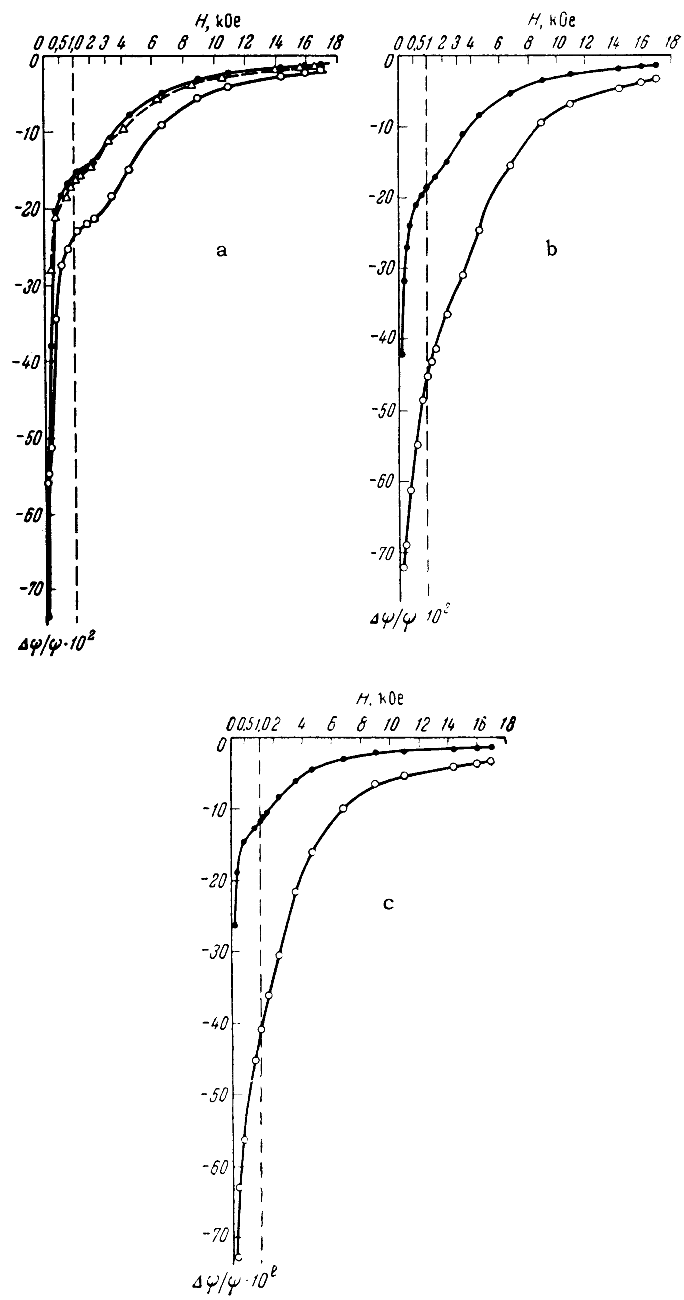

FIG. 6. Dependence of $\Delta\psi/\psi$ of terbium on the magnetic field: a) at T = 77.3°K: ● – p = 1820 kg/cm², ○ – p = 3000 kg/cm²; at T = 87.4°K: Δ – p = 1850 kg/cm²; b) at T = 111.5°K: ● – p = 1900 kg/cm², ○ – p = 5400 kg/cm²; c) at T = 169.3°K: ● – p = 2000 kg/cm², ○ – p = 5000 kg/cm².

sufficient to draw conclusions about the sign $(\Delta q_0/q_0)/p$.

2. INFLUENCE OF UNIFORM COMPRESSION ON THE MAGNETIZATION OF TERBIUM

The influence of pressure on the magnetization of terbium was investigated in the ferromagnetic state. The measurements of the change in the magnetization due to uniform compression of the samples were carried out in the temperature range from 77 to 169°K (according to[9], the Curie temperature of terbium is $\Theta = 218$°K).

Figures 6a, 6b, and 6c show curves representing the dependence of the relative change in the magnetic flux $(\Delta\psi/\psi)$, under uniform compression, as a function of the magnetic field H. It is clear from these curves that under uniform compression the magnetization decreases. In fields up to ≈ 11 kOe, the changes $\Delta\psi/\psi$ are large and not proportional to pressure.

We may assume that, as in the case of gadolinium,[8] in the rotation region the change in the magnetization under uniform compression is due to two factors: the change in the anisotropy constant and the change in the exchange interaction. An increase in the anisotropy constant under pressure decreases the magnetization in the technical-magnetization region.

At $H > 15$ kOe, the absolute value of $\Delta\psi/\psi$ decreases sharply; the pressure coefficient $\psi^{-1}(\Delta\psi/\Delta p)$ is independent of the value of p and changes only very slightly when the field intensity is increased further. It is reasonable to suppose that in strong fields ($H > 15$ kOe) the reduction in the magnetization under uniform compression is mainly due to a change in the exchange interaction due to the reduction of the interatomic spacings.

Using the well-known thermodynamic relationship

$$\sigma^{-1}(\Delta\sigma/\Delta p) = \psi^{-1}(\Delta\psi/\Delta p) - {}^{1}/_{3}\varkappa,$$

where σ is the magnetic moment per unit mass, κ is the compressibility of terbium, we may calculate the value of $(\Delta\sigma/\Delta p)/\sigma$.

The table lists the values of the pressure coefficients and $(\Delta\sigma/\Delta p)/\sigma$ at H = 17 kOe. The latter quantities were calculated using the compressibility of terbium, equal to 2.45×10^6 cm²/kg, obtained by measurements at room temperature.[2]

T, °K	p, kg/cm²	$-[(\Delta\psi/\Delta p)/\psi]\cdot 10^7$, cm²/kg	$-[(\Delta\sigma/\Delta p)/\sigma]\cdot 10^7$, cm²/kg
77.3	1820	65±7	73±7
	3000	63±6	71±6
87.4	1850	65±7	73±7
111.5	1900	63±6	71±6
	5400	61±6	69±6
169.3	2000	66±7	74±7
	5000	66±7	74±7

It is evident from the table that the pressure coefficient for H = 17 kOe is practically independent of temperature (within the experimental error). The value of $(\Delta\sigma/\Delta p)/\sigma$ is almost two orders of magnitude greater than for Gd,[8] and is close to the value observed for Invar alloys.[11]

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