THE ONE-ION MODEL AND THE MAGNETOSTRICTION OF TERBIUM AND ITS GADOLINIUM ALLOY

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We measure the temperature dependence of the magnetostriction, the magnetostrictive contribution to the thermal expansion, and that of the magnetization of monocrystalline terbium and the alloy $Tb_{0.5}Gd_{0.5}$. We find that the experimental data agree with the predictions of the one-ion theory as regards the temperature dependence of the magnetostrictive contribution to the thermal expansion. We calculate from experimental data and thermodynamic relations the dependence of the transition temperature and the magnetic-crystalline interaction on the lattice parameters. We demonstrate the agreement of these relationships with estimates made according to the theory of the crystalline field using the one-ion model.

There are two models currently in use for rare-earth metals (REM) which explain differently the occurrence of the magnetic anisotropy and, hence, the large magnetostriction of the REM's. The theory developed in [1-3] is based on the one-ion model. According to this theory, the principal cause of the large magnetic anisotropy and magnetostriction is the interaction of the anisotropic cloud of the $4f$ electrons with the crystalline field of the lattice.

The anisotropy and, hence, the magnetostriction of the REM's can be explained [4] by the fact that their exchange interaction is strongly anisotropic, i.e., dependent on the direction of the magnetization vector relative to the crystallographic axes, due to the anisotropic distribution in the crystal lattice of the conduction electrons, which are magnetized by the indirect exchange interaction with the $4f$ electrons.

This paper is devoted to the investigation of the possibility of using the one-ion model for describing the temperature dependence of the magnetostrictive contribution to the thermal expansion of terbium and its allow with gadolinium $Tb_{0.5}Gd_{0.5}$. Additionally, it is the goal of this paper to compare the predictions of the one-ion model with the values calculated from the magnetostriction data for the shifts of the transition temperature when the interatomic distances are changed.

The method of growing the single crystals and the specification of their quality control were given earlier [5]. The methods of measuring the magnetostrictive strain and the thermal expansion were described in [6].

Figures 1 and 2 shows the temperature dependence of the magnetostrictive contributions to the thermal expansion along the crystallographic axes a, b, and c for monocrystalline terbium and the alloy $Tb_{0.5}Gd_{0.5}$. These contributions were determined according to the method used earlier [6], from measurements of the temperature dependence of the thermal expansion in a magnetic field applied along the easy axis of magnetization (the b axis).

The magnetostrictive contribution to the thermal expansion is governed, according to the theory [1,2], by the two-ion isotropic exchange contributions (magnetostriction constants $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$), which are proportional to the square of the specific magnetization m, and the one-ion aniostropic contributions (magnetostriction constants $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$ and $\lambda^{\gamma,2}$), the temperature dependence of which is described by the hyperbolic Bessel function $\hat{I}_{5/2}(m)$ as shown in the figures.

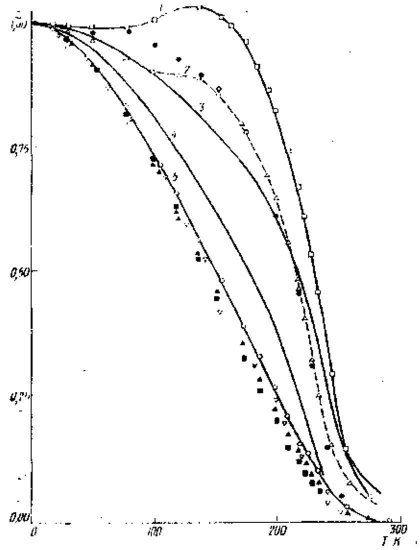

Fig. 1

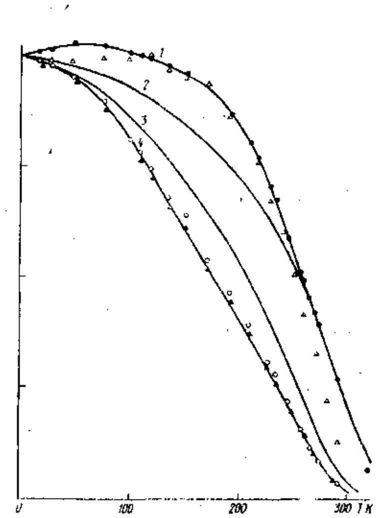

Fig. 2

Fig. 1. Temperature dependence of the magnetization and the magnetostrictive contribution to the thermal expansion along the a, b, and c axes for terbium in a magnetic field of 15 kOe applied along the b axis: curve 1 is λ_c/λ_{0c} from the data of [11]; 2 is λ_c/λ_{0c} from our data; 3 is the specific magnetization m; 4 is m^2; 5 is $\hat{I}_{5/2}(m)$; ○ – λ_a/λ_{0a} and ■ – λ_b/λ_{0b} from our data; ▲ – λ_b/λ_{0b} from the data of [11]; ∇ – λ_b/λ_{0b} calculated according to Eq. (2); ● – λ_c/λ_{0c} calculated from Eq. (3).

Fig. 2. Temperature dependence of the magnetization and the magnetostrictive contribution to the thermal expansion along the a and c axes for the alloy $Tb_{0.5}Gd_{0.5}$ in a magnetic field of 50 kOe applied along the b axis: curve 1 is λ_c/λ_{0c} from the experimental data; 2 is the specific magnetization m; 3 is m^2; 4 is $\hat{I}_{5/2}(m)$, ○ – λ_a/λ_{0a} from the experimental data; Δ – λ_c/λ_{0c} calculated according to Eq. (3); ▲ – λ_a/λ_{0a} calculated from Eq. (1).

Thus, for the magnetostrictive contribution to the thermal expansion along the crystal axes a, b, and c, we will have [2]:

$$\lambda_a(T) = \lambda_1^{\alpha,0} m^2 - \left(\frac{1}{3}\lambda_1^{\alpha,2} + \frac{1}{2}\lambda^{\gamma,2}\right)\hat{I}_{5/2}(m), \qquad (1)$$

$$\lambda_b(T) = \lambda_1^{\alpha,0} m^2 - \left(\frac{1}{3}\lambda_1^{\alpha,2} - \frac{1}{2}\lambda^{\gamma,2}\right)\hat{I}_{5/2}(m), \qquad (2)$$

$$\lambda_c(T) = \lambda_2^{\alpha,0} m^2 - \frac{1}{3}\lambda_2^{\alpha,2}\hat{I}_{5/2}(m). \qquad (3)$$

The experimental results shown in Figs. 1 and 2 for the magnetostrictive contributions to the thermal expansions are well described by Eqs. (1), (2), and (3). The contributions $\lambda_a(T)$ and $\lambda_b(T)$ are determined mainly by the terms proportional to $\hat{I}_{5/2}(m)$; this indicates the predominant role of the one-ion contributions. In the temperature dependence of the magnetostrictive contribution to the thermal expansion along the c axis there is a substantial contribution from the two-ion iostropic term $\lambda_2^{\alpha,0} m^2$, as well as from the one-ion term $\frac{1}{3}\lambda_2^{\alpha,2}\hat{I}_{5/2}(m)$.

It has earlier been established experimentally [5] that for terbium and its alloy $Tb_{0.5}Gd_{0.5}$ the magnetostriction constant $\lambda^{\gamma,2}$ has a temperature dependence to $\hat{I}_{5/2}(m)$, in agreement with the formulas which follow from a one-ion mechanism. The values of the magnetostriction constants found by adjusting the theoretical curves given in Eqs. (1)-(3) to the experimental data for λ_a, λ_b, and λ_c are close to the values determined by other methods [7].

Thus, the theory [1,2], according to which the magnetostriction and the magnetostrictive contributions to the thermal expansion are caused by the two-ion isotropic exchange and the one-ion anisotropic magnetic-crystalline interaction, describes well the temperature dependence of the magnetostriction constants. For explanation of magnetoelastic phenomena it is not necessary to take into account the dependence of the exchange energy on the direction of the magnetization in the crystal lattice.

Some of the discrepancy between the theoretical and experimental curves for $\lambda_c(T)$ in the low-temperature region can be explained by the fact that Eqs. (1)-(3) do not take into account the magnetostriction constants of higher orders, which evidently play an appreciable role at low temperatures.

The results given above, however, do not permit the quantitative comparison of the experimental data on the magnetoelastic properties with the predictions of the theory of magnetostriction based on the one-ion model and the theory of the crystalline field, which do permit calculation of the dependence of the transition temperature on the lattice parameters of the crystal. We will show below that this dependence can also be found from the measurements of the magnetostriction and the magnetization near the Curie point.

In a ferromagnet located in a magnetic field directed along the easy axis which is strong enough to break down the domain structure, the magnetization I is a function of the temperature relative to the Curie temperature as well as a function of the strength of the magnetic field. Hence, using the well-known thermodynamic relation [9]

$$\left(\frac{\partial I}{\partial p_i}\right)_H = -\left(\frac{\partial \lambda_i}{\partial H}\right)_{p_i}, \qquad (4)$$

where p_i is a uniaxial stress acting along the i axis and λ_i is the magnetostriction along the i axis (i = a, b, c), one obtains

$$\frac{\partial \theta}{\partial p_i} = \frac{\theta}{T\left(\frac{\partial I}{\partial T}\right)_{p_i,H}} \left(\frac{\partial \lambda_i}{\partial H}\right)_{p_i,H}. \qquad (5)$$

This formula was obtained for the isotropic case by K. P. Belov [9], who used it to determine the shift of the Curie point of Invar alloys with pressure.

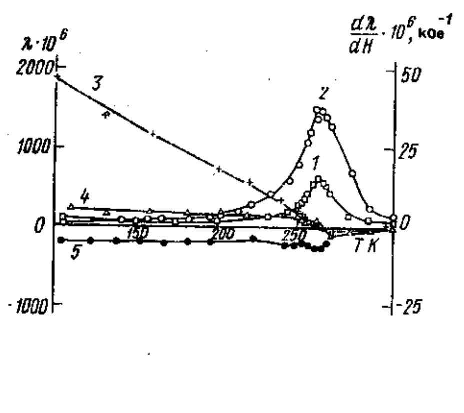

Fig. 3. Temperature dependence of magnetostriction λ and slope of magnetostriction isotherms, $\partial\lambda/\partial H$, in 14-kOe field along a, b, and c axes of $Tb_{0.5}Gd_{0.5}$ single crystal; 1) λ_c, 2) $\partial\lambda_c/\partial H$, 3) λ_b, 4) $\partial\lambda_b/\partial H$, 5) λ_a, 6) $\partial\lambda_a/\partial H$.

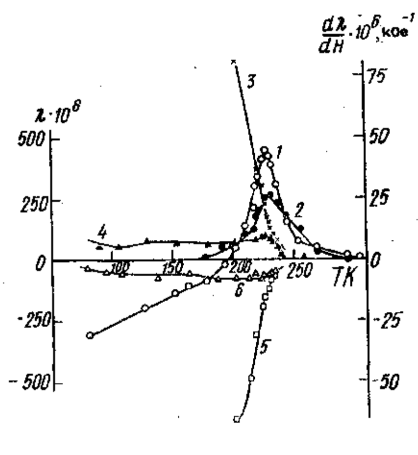

Fig. 4. Temperature dependence of magnetostriction λ and slope of magetostriction isotherms, $\partial\lambda/\partial H$, in 14 kOe field along a, b, and c axes for Tb single crystal; 1) λ_c, 2) $\partial\lambda_c/\partial H$, 3) λ_b, 4) $\partial\lambda_b/\partial H$, 5) λ_a, 6) $\partial\lambda_a/\partial H$.

We measured the magnetostriction and magnetization of terbium and its alloy with gadolinium $Tb_{0.5}Gd_{0.5}$ in samples in the form of disks 7 mm in diameter and 1.5 mm thick, with the aim of obtaining data on the shift of the Curie point under the action of uniaxial stresses (see Figs. 3 and 4). According to our data, the alloy $Tb_{0.5}Gd_{0.5}$ is ferromagnetic at temperatures from 4.2°K to the Curie point (Θ = 262°K).

Terbium is also ferromagnetic at Θ = 230°K in magnetic fields H > 500 Oe, which are sufficient for breaking down the helicoidal magnetic structure which is observed in terbium in the temperature interval 219-230°K. Measurements of the magnetostriction and magnetization of Tb and $Tb_{0.5}Gd_{0.5}$ were carried out in a magnetic field along the b axis, which is the easy axis of magnetization, located in the basal plane of the hexagonal lattice.

For terbium at T = 230°K and H = 14 kOe we found:

$$\frac{\partial I}{\partial T} = 18{,}1 \text{ G/deg}, \quad \frac{\partial \lambda_c}{\partial H} = 2{,}2 \cdot 10^{-8}\,\text{Oe}^{-1},$$
$$\frac{\partial \lambda_b}{\partial H} = 0{,}9 \cdot 10^{-8}\,\text{Oe}^{-1}, \quad \frac{\partial \lambda_a}{\partial H} = -0{,}6 \cdot 10^{-7}\,\text{Oe}^{-1}.$$

For the alloy $Tb_{0.5}Gd_{0.5}$ at T = 262°K and H = 14 kOe we obtained:

$$\frac{\partial I}{\partial H} = -20{,}5 \text{ G/deg}, \quad \frac{\partial \lambda_c}{\partial H} = 3{,}7 \cdot 10^{-8}\,\text{Oe}^{-1},$$
$$\frac{\partial \lambda_b}{\partial H} = 0{,}12 \cdot 10^{-8}\,\text{Oe}^{-1}, \quad \frac{\partial \lambda_a}{\partial H} = 0{,}7 \cdot 10^{-8}\,\text{Oe}^{-1}.$$

Substituting these values into Eq. (5), one can obtain the values of the shift of the Curie temperature under the action of uniaxial stresses (Table 1).

Table 1

Shift of the Curie Temperature under the Action of Uniaxial Stresses for Tb and $Tb_{0.5}Gd_{0.5}$

Substance	$\frac{\partial\theta}{\partial p_a}, \frac{K}{kbar}$	$\frac{\partial\theta}{\partial p_b}, \frac{K}{kbar}$	$\frac{\partial\theta}{\partial p_c}, \frac{K}{kbar}$
Tb	0,33	—0,50	—1,22
$Tb_{0,5}Gd_{0,5}$	0,34	—0,06	—1,80

The shift of the Curie point with changes in the parameters of the crystal lattice a, b, and c can be determined from the system of equations:

$$-\frac{\partial\theta}{\partial p_a} = \frac{\partial\theta}{\partial \log a} S_{11} + \frac{\partial\theta}{\partial \log b} S_{12} + \frac{\partial\theta}{\partial \log c} S_{13}, \tag{6}$$

$$-\frac{\partial\theta}{\partial p_b} = \frac{\partial\theta}{\partial \log a} S_{12} + \frac{\partial\theta}{\partial \log b} S_{11} + \frac{\partial\theta}{\partial \log c} S_{13}, \tag{7}$$

$$-\frac{\partial\theta}{\partial p_c} = \frac{\partial\theta}{\partial \log a} S_{13} + \frac{\partial\theta}{\partial \log b} S_{13} + \frac{\partial\theta}{\partial \log c} S_{33}, \tag{8}$$

where S_{ij} are the elastic compliance constants, which differ little for Tb and Gd [7,10].

The values of $\frac{\partial\theta}{\partial \log a_i}$ calculated from Eqs. (5)-(8) are given in Table 2.

Table 2

Shift of the Curie Temperature for Changes in the Parameters of the Crystal Lattice

Substance	$\frac{\partial\theta}{\partial \log a}$, K	$\frac{\partial\theta}{\partial \log b}$, K	$\frac{\partial\theta}{\partial \log c}$, K
Tb	172	544	965
$Tb_{0,5}Gd_{0,5}$	188	370	1300

According to the calculations of [11], the Curie point of REM's depends on both the parameter I_0 and on the magnetic-crystalline energy (the parameter θ_A):

$$\theta = \frac{2}{3}(g-1)^2 J(J+1)\frac{I_0}{k} + \theta_A. \tag{9}$$

It follows from this that the shift of the Curie temperature under changes of the interatomic distance occurs as a result of the change of both the exchange and the magnetic-crystalline energy:

$$\frac{\partial\theta}{\partial \log a_i} = \frac{2}{3}(g-1)^2 J(J+1)\frac{\partial I_0/k}{\partial \log a_i} + \frac{\partial\theta_A}{\partial \log a_i}, \tag{10}$$

where $a_i = a, b, c$.

Using the values for Tb obtained earlier [12]

$$\frac{\partial I_0/k}{\partial \log a} = \frac{\partial I_0/k}{\partial \log b} = -23 \quad \text{and} \quad \frac{\partial I_0/k}{\partial \log c} = 200,$$

and also our values for $\frac{\partial\theta}{\partial \log a_i}$, which are given in Table 2, one can calculate from Eq. (7) the quantities $\frac{\partial\theta_A}{\partial \log a_i}$, which characterize the dependence of the magnetic-crystalline energy on the parameters of the crystal lattice (Table 3).

Table 3

Dependence of the Magnetic-Crystalline Energy on the Parameters of the Crystal Lattice

	$\frac{\partial\theta_A}{\partial \log a}$, K	$\frac{\partial\theta_A}{\partial \log b}$, K	$\frac{\partial\theta_A}{\partial \log c}$, K
Tb experiment	−330	705	−435
Tb theory	−338	817	−531

Comparison of the data for $\frac{\partial\theta_A}{\partial \log a_i}$ obtained by us with the values calculated earlier [12] according to the theory of the crystalline field using the one-ion model shows that this theory permits the correct calculation of the dependence of the magnetic-crystalline interaction on the interatomic distance and, consequently, the value of the anisotropic magnetostriction, which arises because of this dependence.

Thus, one can deduce that the one-ion theory of magnetostriction explains the temperature dependence of the magnetostriction of terbium and its alloys with gadolinium and permits evaluation of the dependence of the magnetic-crystalline energy on the interatomic distance in good agreement with the experimental data.

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