Effect of magnetic field on the crystal structure of terbium

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The effect of magnetic fields of intensity up to 16 kOe on the crystal structure of polycrystalline terbium was investigated in the temperature interval 77–300°K. Principal attention was paid to the study of structure effects in the ferromagnetic state, the region of existence of which in a magnetic field stretches to ~240°K. The ferromagnetic domains become reoriented, as a result of which the directions of the magnetic moments (the *b* axes) are disposed at the minimum possible angles to the magnetic-field intensity vector. The deformation of the crysal lattice in the magnetic field has a strongly anisotropic character. The contraction of the *b* axis is connected with rotation of the magnetic moment in the basal plane of the lattice, and the elongation of the *c* axis is connected with the enhancement of the indirect exchange interaction between the basal planes.

A characteristic feature of heavy rare-earth metals (REM) is the appearance of anomalously large strains both on going to the magnetically ordered state and when an external magnetic field is applied ("giant magnetostriction"[1]). The use of the method of x-ray structure analysis in a magnetic field[2] makes it possible to separate effects connected with phase transitions ("by temperature" and "by field") with magnetoelastic deformation of the crystal lattice, and with domain-reorientation processes.

We have previously investigated the influence of the magnetic field on the crystal structure of dysprosium[2-4] and gadolinium[5]. It was shown that in fields up to 16 kOe, a magnetic first-order "antiferromagnetism-ferromagnetism" transition takes place in dysprosium; the transition lowers the lattice symmetry from hexagonal to rhombic, and in the interval $T_C < T < T_N$ the critical field of the magnetic transition (H_{cr}) increases with increasing temperature, the jump in the elastic energy in H_{cr} decreases with increasing temperature, and the work of the magnetostriction increases. An x-ray diffraction procedure was developed for measuring the magnetoelastic deformation of the crystal lattice by determining the relative change in the interplanar distances (d_{hkl}) in a magnetic field[5]. This procedure, while less accurate than modern macroscopic methods, makes it possible to measure the change of the lattice parameters against the background of much stronger effects connected with the phase transitions and with the domain displacements. It was established that the magnetoelastic deformations of the lattice are maximal near the critical points.

What is considered in essence in[2-4] are the following two situations: weak magnetic anisotropy (Gd, $T \gtrless T_C$; Dy, $T > T_N$) and strong anisotropy of the easy-plane type (more accurately—an antiferromagnetic helicoid SS, Dy, $T_C < T < T_N$, $H < H_{cr}$). The case of uniaxial anisotropy was not considered in detail. The optimal object for the study of this situation can be terbium, which is a collinear ferromagnet (easy axis ⟨010⟩) at 0–222°K and a helicoidal antiferromagnet at 222–234°K [1)].

The magnetic[7] and crystalline[8,9] structures of Tb in magnetically ordered states were investigated earlier, and its various magnetic properties, such as magnetization[10,11], the magnetostriction of single-crystal[5,12,13] and polycrystalline samples[14], etc., have also been well investigated. The purpose of the present study was to determine the change of the crystalline structure of terbium at temperatures 77–300°K in magnetic fields up to 16 kOe.

RESULTS OF INVESTIGATION

The deformation of a hexagonal crystal lattice in a direction that forms angles β_i with the orthogonal axes a_i of the unit cell can be expressed in the form[2)][15]

$$\lambda=\lambda_1^{\alpha,0}(T)(\cos^2\beta_1+\cos^2\beta_2)+\lambda_2^{\alpha,0}(T)\cos^2\beta_3$$
$$+\lambda_1^{\alpha,2}(T,H)(\cos^2\beta_1+\cos^2\beta_2)(\cos^2\alpha_3-{}^1/_3)+\lambda_2^{\alpha,2}(T,H)\cos^2\beta_3(\cos^2\alpha_3-{}^1/_3)$$
$$+\lambda^{\gamma,2}(T,H)[{}^1/_2(\cos^2\beta_1-\cos^2\beta_2)(\cos^2\alpha_1-\cos^2\alpha_2)+2\cos\beta_1\cos\beta_2\cos\alpha_1\cos\alpha_2]$$
$$+2\lambda^{\varepsilon,2}(T,H)(\cos\beta_1\cos\alpha_1+\cos\beta_2\cos\alpha_2)\cos\beta_3\cos\alpha_3+\ldots \quad (1)$$

where α_i are the angles between the moment **M** and the axes a_i.

The magnetoelastic deformations $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ characterize the change in the dimensions of the hexagonal lattice in the basal plane and in the direction of the principal axis ⟨001⟩ in the magnetically-ordered state; they depend only on the values of the true magnetization. Strictly speaking, $\lambda_1^{\alpha,0}$ and $\lambda_2^{\alpha,0}$ should depend on the intensity of the magnetic field, but since the magnetization of terbium remains practically unchanged in fields up to 30 kOe[16], these deformations can be regarded as spontaneous and can be determined by extrapolating the curves a(T) and c(T) from the paramagnetic region[17]:

$$\lambda_1^{\alpha,0}=\frac{1}{2}\left(\frac{a-a_{\rm extr}}{a_{\rm extr}}+\frac{b-b_{\rm extr}}{b_{\rm extr}}\right)$$
$$\lambda_2^{\alpha,0}=\frac{c-c_{\rm extr}}{c_{\rm extr}}. \quad (2)$$

The normal magnetoelastic deformations $\lambda_1^{\alpha,2}$ and $\lambda_2^{\alpha,2}$ characterize the change of the dimensions of the lattice in the basal plane and along the principal axis when an external magnetic field is applied. The shear deformations $\lambda^{\gamma,2}$ and $\lambda^{\epsilon,2}$ describe the lowering of the symmetry of the basal plane and the deviation of the prinprincipal axis from the normal to the basis. To determine the induced deformations $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$, $\lambda^{\gamma,2}$, etc. it is necessary to incorporate in (1) information concerning the magnetic structure of the crystal, i.e., the values of cos α_i [4,5].

The study of the magnetic deformation of the crystal lattice reduces to a consideration of effects connected both with the appearance of magnetic ordering ($\lambda_1^{\alpha,0}$, $\lambda_2^{\alpha,0}$) and with the action of the magnetic field on the spin structure ($\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$, $\lambda^{\gamma,2}$, etc.). These two aspects of the magnetostriction of terbium will be considered below.

1. Spontaneous Magnetostriction

As already noted, to study the magnetic contribution to the dimensions of the crystal lattice it is necessary to perform precision measurements of the temperature dependences of the crystal-lattice parameters both in the magnetically ordered state and in the paramagnetic state. Measurements of the parameters of Tb at low temperatures were performed earlier[8,9] on metal of commercial purity. The present measurements were made on 99.8% pure Tb; we also succeeded in greatly increasing the accuracy of the lattice-parameter measurement ($\Delta a_i \sim 1 \times 10^{-4}$ Å) and increased somewhat the temperature-measurement range (77–300°K).

The experiments were performed with a low-temperature attachment to the URS-50I diffractometer[18]; we obtained x-ray diffraction patterns of the lines $(300)_h$ and $(006)_h$ of the hexagonal lattice. The reflection of $(300)_h$ at $T < 222$°K was "split" into two components:

$$(300)_h \rightarrow (330)_r + (060)_r, \quad 2\theta_{(330)_r} > 2\theta_{(060)_r},$$

where 2θ is the diffraction angle. This character of the splitting indicates that the hexagonal lattice is transformed into a rhombic lattice with an axis ratio $b/a > \sqrt{3}$.

The results of the measurements of the Tb lattice parameters are shown in Fig. 1. At 232 ± 1°K (T_N) inflections are observed on the a(T) and c(T) curves. At 222°K (T_C), the parameter a decreases jumpwise ($\Delta a = -8 \times 10^{-4}$ Å), while the parameters b and c increase ($\Delta b = 9 \times 10^{-4}$, $\Delta c = 13 \times 10^{-4}$ Å). The lowering of the symmetry and the jumplike change in the dimensions of the unit cell in the "helicoidal antiferromagnetism–collinear ferromagnetism" transition indicates that this is a first-order transition. Below T_C, the thermal expansion is positive along the a axis, and negative along the b axis, while the c(T) curve has an inflection at ~170°K. The axis ratio b/a increases monotonically with decreasing temperature after the discontinuity at 222°K ($\Delta b/a = 6 \times 10^{-4}$) (insert in Fig. 1).

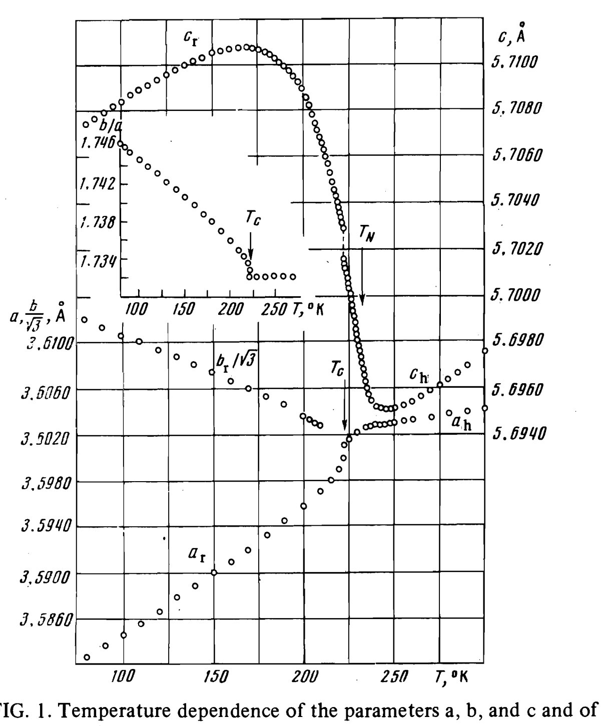

FIG. 1. Temperature dependence of the parameters a, b, and c and of the axial ratio b/a for Tb.

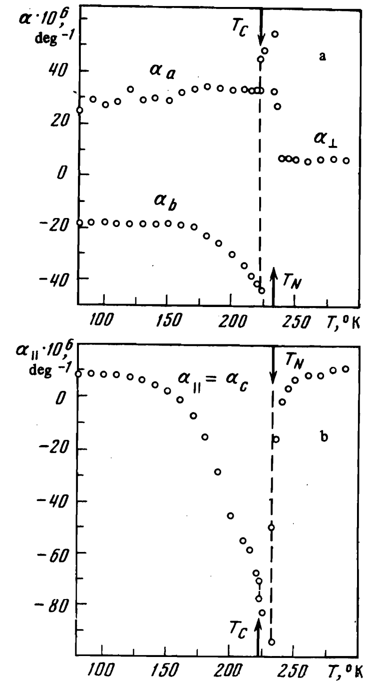

FIG. 2. Temperature dependence of the coefficient of linear expansion of Tb. a—α_a, α_b, $\alpha_\perp$; b—$\alpha_c = \alpha_\parallel$.

The values of the linear-expansion coefficient obtained by numerical differentiation of the Tb curves are shown in Fig. 2. In the paramagnetism-antiferromagnetism transition, clearly pronounced λ anomalies are seen on the $\alpha_i(T)$ curves; these anomalies are typical[3)] of second-order phase transitions[19]. Approximating these anomalies by Landau discontinuities ($\Delta\alpha_\perp > 0$, $\Delta\alpha_\parallel < 0$) and starting from the Ehrenfest equations for hexagonal crystals[20]

$$\Delta\alpha_{11} = \Delta\alpha_\perp = \frac{\Delta C_p}{T_N V}\frac{dT_N}{d\sigma_{11}}, \qquad \Delta\alpha_{33} = \Delta\alpha_\parallel = \frac{\Delta C_p}{T_N V}\frac{dT_N}{d\sigma_{33}} \tag{3}$$

(ΔC_p is the discontinuity of the specific heat in T_N; $V = a^2 c\sqrt{3}/4$ is the atomic volume; σ_{11} and σ_{33} are clamping stresses applied along the axes a and c), we find that the Neel temperature should increase when the single crystal is compressed along the a axis and should decrease upon compression along c. This agrees with the results of direct measurements of $dT_N/d\sigma_{11}$ and $dT_N/d\sigma_{33}$[21].

With the aid of (2) we calculated the spontaneous magnetic deformations of the Tb crystal lattice (Fig. 3). The magnetostriction is maximal along the c axis ($\lambda_2^{\alpha,0}$), and the character of the $\lambda_2^{\alpha,0}(T)$ dependence correlates well with the temperature dependence of the spontaneous magnetization[4)][16]. The deformation of the basal plane ($\lambda_1^{\alpha,0}$) is smaller by one order of magnitude, although the effects along the a and b axes ($\lambda_a < 0$, $\lambda_b > 0$) are quite large.

2. Stimulated Magnetostriction

The procedure used for the structural investigation of terbium in a magnetic field did not differ significantly from that described earlier[2]. Using radiation from a chromium anticathode, we recorded the reflections from the planes $(114)_h$, $(203)_h$, $(210)_h$, $(211)_h$, $(212)_h$, and $(105)_h$, and on going into the ferromagnetic state the lines of the $(hkl)_h$ type split into two or three rhombic components $(h, h + 2k, l)_r$, $(k, 2h + k, l)_r$,

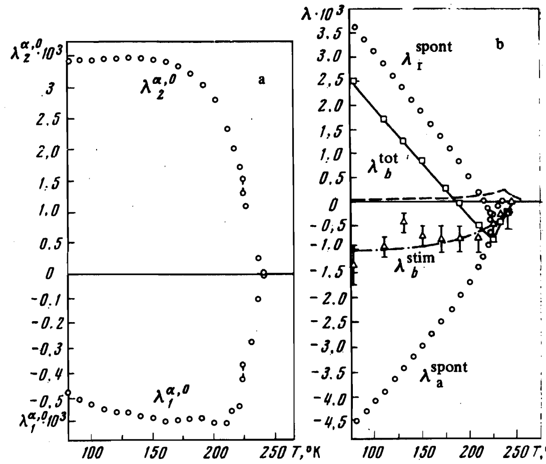

FIG. 3. Magnetic deformation of Tb crystal lattice: a—$\lambda_1^{a,0}(T)$, $\lambda_2^{a,0}(T)$; b—$\lambda_a^{spont}(T)$, $\lambda_b^{spont}(T)$ (○), $\lambda_b^{stim}(T)$ at H = 16 kOe (△), $\lambda_b^{tot}(T)$ (□), dashed—$\lambda_b^{stim}(T)$ from [12] at H = 16 kOe.

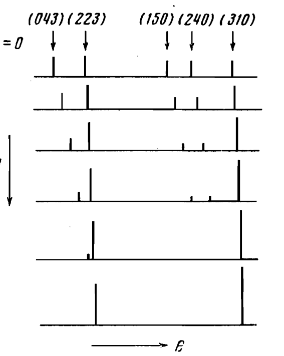

FIG. 4. Change of the diffraction pattern of Tb at $T < T_C$ in a magnetic field (scheme).

$(h + k, h - k, l)_r$. The relative error in the measurement of the interplanar distances d_{hkl} did not exceed 1×10^{-4} Å.

Owing to the small value of the critical field of the antiferromagnetism-ferromagnetism transition at $T < T_N$, we observed only the diffraction lines of rhombic Tb in the magnetic field. The character of the change of the diffraction picture with increasing magnetic field intensity is shown schematically in Fig. 4. When the magnetic field intensity is increased, the x-ray diffraction patterns reveal a decrease in the distance between reflections of the type $(h, h + 2k, l)_r$, $(k, 2h + k, l)_r$, and $(h + k, h - k, l)_r$, and a decrease of the intensity of the interferences $(h, h + 2k, l)_r$ and $(k, 2h + k, l)_r$ with a simultaneous increase of the intensity of the lines $(h + k, h - k, l)_r$. The coming together of the rhombic lines indicates unequivocally a lowering of the degree of rhombic distortion of the crystal lattice ($b/a \to \sqrt{3}$).

The redistribution of the intensity of the diffraction lines means that a rearrangement of the domain structure takes place wherein all the easy-magnetization axes $\langle 010 \rangle_r$ start to orient themselves at the minimal possible angle to the vector **H** ("dynamic magnetic texture" [2,3]). The complete extinction of the reflections of the type $(h, h + 2k, l)_r$ and $(k, 2h + k, l)_r$ is evidence of completion of the domain-displacement process; with increasing temperature, the domain reorientation occurs in fields of lower intensity.

Figure 5 shows the isotherms of the dependence of the interplanar distance $d_{(115)_r}$ on the magnetic field intensity. (The isotherms of the field dependences of

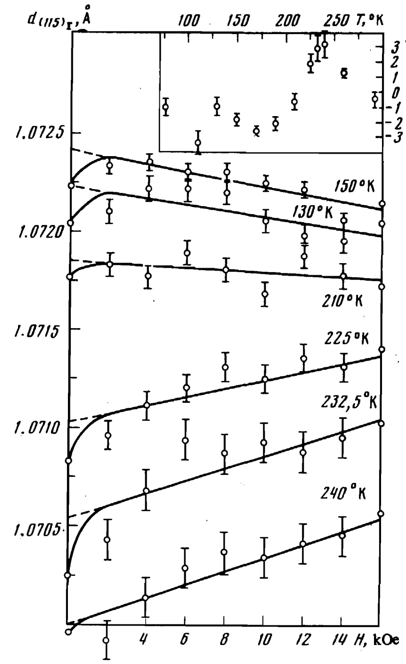

FIG. 5. Field dependences of the interplanar distance $d_{(115)_r}$ of Tb.

d_{hkl} for other crystallographic planes are in principle of the same character). In the interval $2 < H < 16$ kOe, the $d_{hkl}(H)$ plots were approximated by straight lines; the temperature dependence of the angular coefficients $\xi_{hkl} = d_{hkl}^{-1}\delta d_{hkl}/\delta H$ is given in the insert. As a rule, the values of d_{hkl} at H = 0 deviate noticeably from the course of the $d_{hkl}(H)$ isotherms.

The plots of $\xi_{hkl}(T)$ reveal at ~240°K singularities similar to the λ anomalies of the second derivatives of the thermodynamic potentials at second-order phase-transition points. The shift of the "apparent Curie points"[5)] of Tb was noted in a number of papers [11-13].

From the isotherms $d_{(310)_r}(H)$, $d_{(223)_r}(H)$ and $d_{(115)_r}(H)$ we calculated the magnetostriction deformations $\lambda_1^{\alpha,2}$, $\lambda_2^{\alpha,2}$, and $\lambda^{\gamma,2}$ (or their derivatives with respect to the magnetic field $\partial\lambda_i/\partial H$).

For ferromagnetic Tb ($\cos\alpha_1 = 0$, $\cos\alpha_2 = 1$, $\cos\alpha_3 = 0$ [7]), the field-dependent terms of Eq. (1) take the form

$$\lambda(H) = -\tfrac{1}{3}(\cos^2\beta_1 + \cos^2\beta_2)\lambda_1^{\alpha,2}(H) - \tfrac{1}{3}\cos^2\beta_3\lambda_2^{\alpha,2}(H) - \tfrac{1}{2}(\cos^2\beta_1 - \cos^2\beta_2)\lambda^{\gamma,2}(H). \qquad (4)$$

The values of $\cos\beta_i$ are obtained from pure crystallographic relations of the type [4]:

$$\cos^2\beta_i = d_{hkl}^2 h_i^2 / a_i^2 \qquad (5)$$

for an orthogonal (rhombic) lattice. Since the initial equation (1) is valid for the case M ∥ H, the magnetoelastic deformations measured in polycrystals

$$\lambda(H) = \frac{d_{hkl}(H) - d_{hkl}(H=0)}{d_{hkl}(H=0)} \qquad (6)$$

were renormalized for calculation by means of formula (4):

$$\lambda'(H) = \lambda(H)/\langle\cos\delta\rangle, \qquad (7)$$

where $\langle\delta\rangle$ is the averaged angle between the vectors **M** and **H** [4].

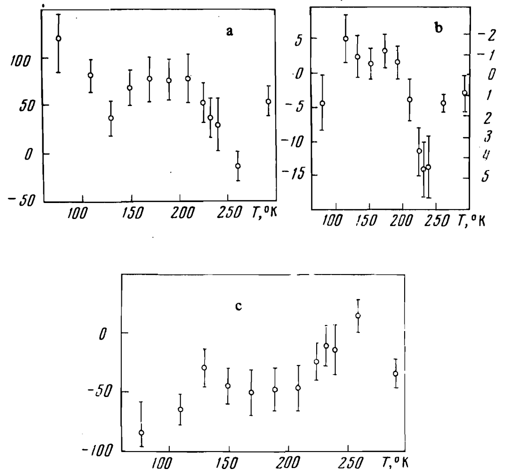

FIG. 6. Temperature dependence of the magnetostriction coefficients of Tb:

$$\text{a) } \frac{\partial\lambda_1^{\alpha,2}}{\partial H}\cdot 10^8,\ \mathrm{Oe}^{-1},\ \text{b) } \frac{\partial\lambda_2^{\alpha,2}}{\partial H}\cdot 10^8\ \mathrm{Oe}^{-1} = -3\frac{\partial\lambda_c^{\mathrm{stim}}}{\partial H}\cdot 10^8\ \mathrm{Oe}^{-1},$$

$$\text{c) } \frac{\partial\lambda^{\gamma,2}}{\partial H}\cdot 10^8\ \mathrm{Oe}^{-1} = \frac{\partial\lambda_{b/a}^{\mathrm{stim}}}{\partial H}\cdot 10^8\ \mathrm{Oe}^{-1}$$

Results of the calculation of $\partial\lambda_1^{\alpha,2}/\partial H$, $\partial\lambda_2^{\alpha,2}/\partial H$ and $\partial\lambda^{\gamma,2}/\partial H$ are shown in Fig. 6. For all the stimulated-magnetostriction coefficients one observes a weak temperature dependence below 170°K, a "peak" near ~240°K, and a rapid decrease of $\partial\lambda_i/\partial H$ in the paramagnetic region.

DISCUSSION OF RESULTS

The results obtained in this study indicate that the effect of a magnetic field on the crystal structure of Tb reduces, first, to reorientation of the ferromagnetic domains, and, second, to a change in the dimensions and shape of the unit cell.

The domain reorientation results in the appearance of giant macroscopic deformations in the basal plane, since the spontaneous deformations of the lattice along the three possible easy magnetization axes (b) are oriented parallel to the external magnetic field. It appears that the displacement processes are also accompanied by a certain change in the lattice dimensions (see Fig. 5, in which the points at H = 0 do not fit the general plots of $d_{hkl}(H)$).

The cause of the Tb crystal lattice deformation in a magnetic field should be taken to be the change in the character of the indirect exchange interaction. According to present-day notions [24], the s-f indirect exchange integral of rare-earth metals is formed by conduction electrons, which are very sensitive to the external field; this can lead to modification of the exchange integral.

It is profitable in this connection to compare the effects of the spontaneous and stimulated deformations of the orthogonal crystal axes. It follows from (4) that

$$\lambda_{\langle 100\rangle}^{\mathrm{stim}}(H)=\lambda_a^{\mathrm{stim}}(H)=-{}^1/_3\lambda_1^{\alpha,2}(H)-{}^1/_2\lambda^{\gamma,2}(H),$$
$$\lambda_{\langle 010\rangle}^{\mathrm{stim}}(H)=\lambda_b^{\mathrm{stim}}(H)=-{}^1/_3\lambda_1^{\alpha,2}(H)+{}^1/_2\lambda^{\gamma,2}(H), \quad (8)$$
$$\lambda_{\langle 001\rangle}^{\mathrm{stim}}(H)=\lambda_c^{\mathrm{stim}}(H)=-{}^1/_3\lambda_2^{\alpha,2}(H).$$

It is also obvious that

$$\lambda_{b/a}^{\mathrm{stim}}(H)=\lambda^{\gamma,2}(H). \quad (9)$$

In fields of intensity up to 16 kOe, the stimulated deformation of the lattice along the easy magnetization

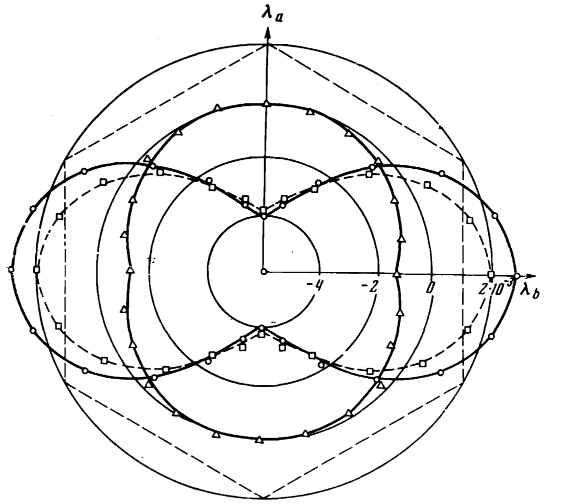

FIG. 7. Magnetic deformation in the (001) plane for Tb at T = 110°K. ○–λ^{spont} at H = 0; △–λ^{stim} at H = 16 kOe; □–λ^{tot} at H = 16 kOe.

axis b is of the same order as the spontaneous deformation (see Fig. 3b). The deformation along the a axis is zero within the limits of measurement accuracy. The c-axis deformation due to the application of the external magnetic field, while small in absolute magnitude, greatly exceeds the possible measurement errors (see Fig. 6b). The "lengthening" of the c axis both when the magnetic field is increased and when the temperature is lowered indicates that the spontaneous and stimulated magnetostrictions have a common mechanism, namely, that the indirect exchange interaction between the basal planes of the crystal lattice becomes stronger.

A more complicated picture of the magnetic deformation of the Tb lattice is observed in the basal plane (Fig. 7), namely, application of a magnetic field causes flattening of the spontaneous magnetostriction ellipsoid in the (001) plane, since $\lambda_b^{\mathrm{stim}}$, unlike λ^{spont}, has a negative character. We note that when a magnetic field is applied along the b axis [12], i.e., under conditions when only the paraprocess magnetostriction is possible, the magnetic deformation in this direction is positive (see Fig. 2b). The negative stimulated magnetostriction in the basal plane can apparently be attributed to rotation of the magnetic moment **M** in the (001) plane in the direction of the projection of the magnetic-field intensity vector **H**. Rather crude guesses, based on the change of the axis ratio b/a in the magnetic field, show that the angle between the vectors **M** and **H** does not exceed 10° at H ~ 16 kOe. We note that this enables us to use formula (4) for the calculations in the entire range of values of H without introducing noticeable errors into the results.

The violation of the coherence relation between the moment **M** and the "easy axis" b should lead to a decrease in the spontaneous magnetostriction in the direction of this axis. In sufficiently strong magnetic fields (H > 50 kOe) one should expect a further enhancement of this effect for polycrystals and even a lifting of the rhombic distortions of the crystal lattice of ferromagnetic Tb. The tendency of the axis ratio b/a to decrease with increasing magnetic field intensity (see Fig. 4) points to the possibility of formation of a ferromagnetic structure of Tb with arbitrary orientation of the moment **M** in the basal plane, i.e., to a transition from anisotropy of the easy-axis type (in weak fields) to anisotropy of the easy-plane type (in strong fields). This assumption agrees with the results of the measurement of the magnetization of Tb single crystals in strong magnetic fields[25], namely, at $H > 50$ kOe the difference between the properties along the axes a and b vanishes.

Thus, we have obtained the following results:

1. In the temperature interval 77—300°K and in magnetic fields of intensity up to 16 kOe, we investigated the crystal structure of polycrystalline terbium of 99.8% purity.

2. We have shown that in the ferromagnetic state the spontaneous magnetostriction is negative in the direction of the a axis of a rhombic lattice, and is positive in the direction of the b and c axes.

3. In the ferromagnetic state, a reorientation of the domains is observed in a magnetic field, as a result of which the easy magnetization axes b align themselves at the minimal possible angle to the magnetic field intensity vector **H**.

4. Rotation of the magnetic moment in the basal plane in the direction of the vector **H** causes the rhombic distortions of the crystal lattice to decrease.

5. All the coefficients of the stimulated magnetostriction reveal anomalies in the vicinity of the Neel point.

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1)The helicoidal SS terbium magnetic structure is destroyed in weak magnetic fields ($H_{cr}^{max} = 442$ Oe [6]).

2)It is assumed that the field H is applied parallel to the magnetic moment M.

3)In the absence of jumps in the lattice parameters at T_N (see Fig. 1).

4)According to the presently held opinions [22], the spontaneous magnetostriction should be proportional to the square of the magnetization of the ferromagnet.

5)We recall that the true Curie points are isolated points on the (T, H) planes, located at $T = T_C$ and $H = 0$ [23].

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