It should be noted that in accord with the susceptibility data the gap begins to decrease when the temperature rises above $T \approx 116$ K, and it is precisely in this region that the logarithmic derivative of the resistance has a maximum. It was noted above that the activation energy of the conductivity is $E_a \approx 5.90$ K, in good agreement with the susceptibility data. Thus, for the obtained values of the energy gap ($2E_a \approx 1180$ K) and of Peierls transition temperature ($T_P \approx 330$ K) we get $2\Delta(0)/T_P \approx 3.5$, which agrees with the exact calculation within the framework of the molecular field approximation.

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Metamagnetic phase transitions and instability of magnetic structure in rare-earth orthoferrites

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We have investigated the metamagnetic and spin-reorientation transitions that occur in $TbFeO_3$ in an external magnetic field when **H** is parallel to the *a* axis of the crystal and when it is parallel to the *b* axis. We detected anomalies, corresponding to these transitions, on the magnetostriction field-dependence curves (one anomaly for $\mathbf{H} \| a$ and two for $\mathbf{H} \| b$). We estimated the R–R and R–Fe interaction constants in $TbFeO_3$. It is shown that these phase transitions may be regarded as transitions of the Jahn-Teller type.

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1. INTRODUCTION

Certain rare-earth ions (RI) behave, in crystals at low temperatures, like Ising ions; that is, their magnetic moment is directed along a definite crystal axis for arbitrary directions of the field acting on them. Such, for example, are the Dy^{3+}, HO^{3+}, and Tb^{3+} ions in orthoaluminates and orthoferrites.

Metamagnetic transitions in Ising anti-ferromagnets have been investigated in detail[1,2] only in those with a single rare-earth subsystem ($TbA1O_3$, $DyA1O_3$, etc.).

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In such compounds, the metamagnetic transition is a magnetic reversal of those ions whose magnetic moments are directed opposite to the external field. There then occur discontinuities on the magnetization curves along the corresponding directions. The critical field at which the metamagnetic transition occurs is equal, in order of magnitude, to the effective field exerted on the given ion by the ions of the other sublattice.

When the crystal contains still another d-ion subsystem (for example, in TbFeO and $DyFeO_3$), the magnetic behavior of the system becomes more complicated and more interesting; simultaneously with the metamagnetic transitions, there may occur spin-re-orientation (SR) transitions in the d-ion subsystem. Such transitions have been detected in $TbFeO_3$[3,4] in an investigation of magnetization curves on which discontinuities, corresponding to metamagnetic transitions, were observed (one discontinuity when H was parallel to the a axis of the crystal, two when H was parallel to the b axis); in the interpretation of these discontinuties, attention was paid to possible reorientation of the spins of the iron ions ($\Gamma_4 - \Gamma_2$), and this made it possible to explain the experimental results quantatively for $\mathbf{H}\|a$ but only qualitatively for $\mathbf{H}\|b$.

It should be mentioned that on the magnetization curves of $TbFeO_3$, because of the smallness of the weakly ferromagnetic moment of the iron ions, the change of state of the Fe subsystem in the SR transitions is practically undetectable. Therefore in experimental investigation of the instability of magnetic structure that occurs in metamagnetic transitions, it is important to measure, along with the magnetization, such effects as would react directly to the state of the Fe subsystem, for example the Faraday effect, the magnetostriction, and NMR. In the present work, magnetostriction measurements were used for this purpose.

A theoretical investigation of the equilibrium magnetic configurations in $TbFeO_3$, carried out with allowance for the peculiarities of the magnetic properties of the Tb^{3+} ion and for its interaction with the Fe^{3+} ions in orthoferrites, made it possible to determine the sequence of magnetic transitions in a field and the values of the fields corresponding to the phase transitions for $\mathbf{H}\|a$ and for $\mathbf{H}\|b$. It was shown that the phase transitions investigated may be regarded as a manifestation of an instability of magnetic structure of the crystal that occurs when the lowest energy levels of the Tb^{3+} ions in the field intersect (approach each other) (a magnetic analog of the Jahn-Teller effect[5]). By comparison of the results of the theory with experimental data, the R–R and R–Fe interaction constants in $TbFeO_3$ were estimated.

2. THEORY

An elementary cell of an orthoferrite contains four RI, located at crystallographic positions with local environmental symmetry C_s. The ground state of the Tb^{3+} ions in the crystalline field of the orthoferrite is an accidental doublet, quite clearly separated from the excited levels ($E_1 \gtrsim 10^2$ cm^{-1}).[6] An important property of the ground quasidoublet of the Tb^{3+} ion is the strong anisotropy of its g factor. In fact the Tb^{3+} ion in orthoferrites may be regarded as an Ising ion, with an axis of quantization lying in the ab plane of the crystal at angle $\alpha = \pm 36°$ to the a axis; the magnetic moment of the ion (at $T=0$) is close to the maximum possible value $m_0 \approx 9\ \mu_B$.[3]

The R–Fe interaction (dipole and exchange) leads to splitting of the ground quasidoublet of the Tb^{3+} ion only when the vector G leaves the ab plane, and H reaches a maximum when the spins of the iron ions are in the phase $\Gamma_2(G_zF_x)$.[7-9] This is the reason for the spin reorientation $\Gamma_4 - \Gamma_2$ in $TbFeO_3$ at $T \approx 6.5$ K. In addition to the splitting of the ground quasidoublet of the Tb^{3+} ion, the R–Fe interaction leads to downward shift of its center of gravity through interaction with the excited states; allowance for this is important in the explanation of a number of magnetic properties of $TbFeO_3$ (χ_c, m_c) and leads, in particular, to an additional anisotropy of the Fe^{3+} ions, which stabilizes the phase $\Gamma_4(G_xF_z)$.[9] At low temperatures, the interaction of the RI themselves with each other leads to an antiferromagnetic ordering of them, of the type $A_x'G_y'$, which produces the reverse reorientation of the spins of the Fe^{3+} ions, $\Gamma_2 - \Gamma_4$, at $T \approx 3.1$ K.[7,9]

In investigating magnetic transitions in an external magnetic field in the low-temperature range, we shall start from the free energy of $TbFeO_3$ at $T=0$, which, in the 4-sublattice approximation for the RI, can be represented in the form[3] (per RI)

$$E = E_{Fe} - \frac{1}{4}\Big[\sum_{\alpha=1}^{4} m_\alpha h_\alpha - \frac{1}{2}\sum_{\alpha,\beta=1}^{4} m_\alpha T_{\alpha\beta} m_\beta\Big], \tag{1}$$

where the first term in (1) is the energy of the Fe^{3+} ions:[1)]

$$E_{Fe} = \frac{A\mathbf{F}^2}{2} + \frac{1}{2}\sum_{i=x,y,z} b_i G_i^2 - \mathbf{F}\mathbf{H}\mu_{Fe} + d(F_zG_x - F_xG_z) + \text{terms of fourth order in } \mathbf{G}. \tag{2}$$

The second term in (1) is the energy of the RI in the effective field, where h_α is its projection on the axis of quantization of the RI. It includes the external field H, the exchange and dipole fields exerted by the Fe^{3+} ions, and the effective field exerted by the other RI:

$$h_\alpha = (H_x + aF_x + p'G_z)\cos\alpha \pm (H_y + aF_y)\sin\alpha + \sum_\beta T_{\alpha\beta} m_\beta, \tag{3}$$

where the signs + and – refer to RI in positions 1, 3 and 2, 4 respectively; a is the constant of isotropic R–Fe exchange, p' is the constant of antisymmetric R–Fe dipole interaction ($p' = 2959$ Oe, $a \sim 10^5$ Oe according to Ref. 9); $T_{\alpha\beta}$ are parameters of the R–R interaction of the rare-earth sublattices, with the following symmetry properties[8]:

$$T_{\alpha\beta} = T_{\beta\alpha},\quad T_{\alpha\alpha} \equiv T_1,\quad T_{12} = T_{34} \equiv T_2,\quad T_{13} = T_{24} \equiv T_3,\quad T_{14} = T_{23} \equiv T_4.$$

The magnetic moments m_α of the sublattices are defined in the local RI axes (that is, they are along the corresponding Ising axes) and are determined by $m_\alpha = m_0$ sign (h_α).[2)] The second term in brackets in (1) appears because the R–R interaction has been included twice in the term $\Sigma m_\alpha h_\alpha$.

On minimizing the energy (1) with respect to F under the conditions $\mathbf{F}\cdot\mathbf{G} = 0$ and $\mathbf{G}^2 = 1 - \mathbf{F}^2 \approx 1$, we get[3)]

$$\mathbf{F}=\{\mathbf{H}_i-(\mathbf{H}_i\mathbf{G})\mathbf{G}\}/A, \tag{4}$$

$$E=-\frac{\mathbf{H}_i^2-(\mathbf{H}_i\mathbf{G})^2}{2A}+\frac{1}{2}\sum b_iG_i^2-p'M_x^RG_z$$
$$-H_xM_x^R-H_yM_y^R-\frac{1}{8}\sum_{\alpha,\beta}m_\alpha T_{\alpha\beta}m_\beta$$
$$+\text{terms of fourth order in }\mathbf{G}, \tag{5}$$

where

$$\mathbf{H}_i=(dG_z+\mu_{Fe}H_x+aM_x^R,\ \mu_{Fe}H_y+aM_y^R,\ -dG_x+\mu_{Fe}H_z),$$
$$M_x^R={}^1/_4(m_1+m_2+m_3+m_4)\cos\alpha,\quad M_y^R={}^1/_4(m_1-m_2+m_3-m_4)\sin\alpha,$$
$$\mu_{Fe}=5\mu_B.$$

On the basis of the system energy (5), which depends only on the orientation of G, we shall now consider magnetic transitions in $TbFeO_3$ when $\mathbf{H}\|a$ and when $\mathbf{H}\|b$.

A. $\mathbf{H}\|a$. In this case (5) can be expressed in the form[4)]

$$E={}^1/_2[K_1^0-k'(M_x^R)^2]\cos^2\theta+{}^1/_4K_2^{Fe}\cos^4\theta$$
$$-m_x^{Fe}H_x\cos\theta-(\lambda'\cos\theta+H_x)M_x^R-{}^1/_8\sum_{\alpha,\beta}m_\alpha T_{\alpha\beta}m_\beta, \tag{6}$$

where $K_1^0=K_1^{Fe}+K_1^{VV}>0$ is the anisotropy constant of the vector G in the ac plane of the crystal, θ is the angle of inclination of G to the c axis, and

$$m_x^{Fe}=\mu_{Fe}d/A,\quad \lambda'=p'+ad/A,\quad k'=a^2/A.$$

For the effective fields on the RI we have the following expression:

$$h_\alpha=(H_x+\lambda'\cos\theta+k'M_x^R\cos^2\theta)\cos\alpha+\sum_\beta T_{\alpha\beta}m_\beta. \tag{7}$$

Minimization of the energy (6) with respect to θ leads to the following picture of the behavior of the system in a field $\mathbf{H}\|a$. Two phases are possible. Phase A, in which the magnetic structures of the iron and rare-earth sublattices are described by the same irreducible representation of the group D_{2h}^{16}, Γ_2 (corresponding to the magnetic group mm). In this phase

$$\cos\theta=1,\quad m_1=m_2=m_3=m_4=m_0.$$

Phase B, in which the rare-earth sublattice is described by the reducible representation Γ_{842} and the iron by the reducible representation Γ_{42}. In this phase

$$\cos\theta\approx m_x^{Fe}H_x/K_1^0,\quad m_1=-m_2=-m_3=m_4=m_0.$$

Table I gives the magnetic symmetry elements and the nomenclature of the phases[5)] that occur when $\mathbf{H}\|a$; their stability limits are determined by the values of

TABLE I. Magnetic symmetry and nomenclature of phases for $\mathbf{H}\|a$.

Phase	Fe^{3+} ions: Representation of group D_{2h}^{16}	Fe^{3+} ions: Magnetic configuration	R^{3+} ions: Representation of group D_{2h}^{16}	R^{3+} ions: Magnetic configuration	Magnetic symmetry elements*	Region of stability of phases
A	Γ_2	G_zF_x	Γ_2	$F_x'C_y'$	$\begin{cases}E,\ I,\ \tilde{2}_x,\ R\tilde{2}_y,\\ R\tilde{2}_z,\ \tilde{\sigma}_x,\ R\tilde{\sigma}_y,\ R\tilde{\sigma}_z\end{cases}$	$H>H_1^a$
B	Γ_{42}	$G_{xz}F_{zx}$	Γ_{842}	$A_x'G_y'$	$E,\ R\tilde{2}_y$	$H<H_2^a$

*R is the time-inversion operator; definitions of the other symmetry elements are given, for example, in Ref. 10. In the magnetic configurations of the R subsystem, we here and hereafter omit small components of the RI Ising vector, of Van Vleck origin.

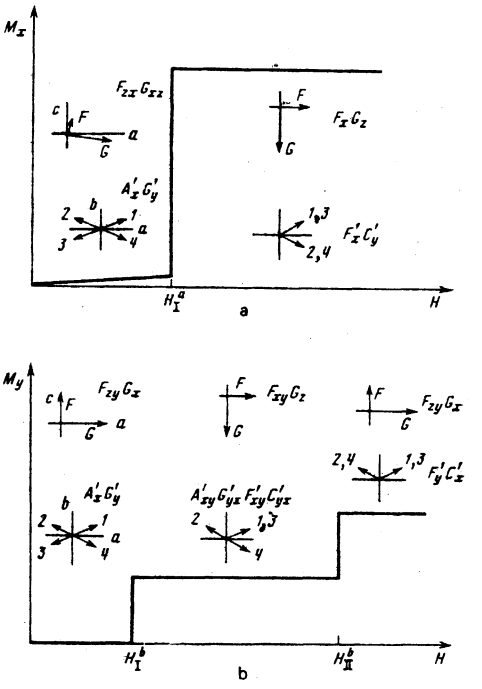

FIG. 1. Magnetization curves of $TbFeO_3$ for $\mathbf{H}\|a$ (a) and for $\mathbf{H}\|b$ (b) (schematic).

H_1^α and H_2^α:

$$H_1^0=-(T_1+T_2+T_3+T_4)m_0/\cos\alpha-(\lambda'+k'm_0\cos\alpha),$$
$$H_2^0\approx(T_1-T_2-T_3+T_4)m_0/\cos\alpha[1+m_x^{Fe}\lambda'/K_1^0]. \tag{8}$$

The critical field at which a transition of the first kind occurs between phases A and B is

$$H_1^0\approx[-(T_2+T_3)m_0^2-(\lambda'+{}^1/_2k'm_0\cos\alpha)m_0\cos\alpha$$
$$+{}^1/_2K_1^0+{}^1/_4K_2^{Fe}]/(m_0\cos\alpha+m_x^{Fe}). \tag{9}$$

This is a metamagnetic transition $A_x'G_y'\to F_x'C_y'$ in the rare-earth subsystem and an orientational transition $G_{xz}F_{zx}\to G_zF_x$ in the subsystem of Fe^{3+} ions (Fig. 1a).

In this transition there is a striking behavior of the weakly ferromagnetic moment of the Fe^{3+} ions. In the phase G_xF_z, the weakly ferromagnetic moment along the c axis is determined by the cant of the magnetic moments of the sublattices of Fe^{3+} ions solely in the Dzyaloshinskiĭ field $F_z=-d/A$. But in the phase G_zF_x, the Dzyaloshinskiĭ field is supplemented by a strong isotropic field exerted by the RI and by the comparatively weak external field, which produce an additional canting of the spins of the Fe^{3+} ions:

$$F_x=(d+am_0\cos\alpha+\mu_{Fe}H_x)/A.$$

We shall estimate the value of this additional canting of the magnetic moments of the iron sublattices. On setting $H_D\sim10^5$ Oe ($d=\mu_{Fe}H_D$) and a $\sim10^5$ Oe, we get $\Delta F_x/F_z\gtrsim100\%$.

B. $\mathbf{H}\|b$. We express the energy of the system, with rotation of the vector G in the ac plane, in the form

$$E={}^1/_2[K_1^0-k'(M_x^R)^2]\cos^2\theta+{}^1/_4K_2^{Fe}\cos^4\theta$$
$$-\lambda'M_x^R\cos\theta-M_y^RH_y-{}^1/_2k'(M_y^R)^2-{}^1/_8\sum_{\alpha,\beta}m_\alpha T_{\alpha\beta}m_\beta. \tag{10}$$

For the effective h_α along the corresponding RI axes, we have

$$h_\alpha=(\lambda'\cos\theta+k'M_x^R\cos^2\theta)\cos\alpha\pm(H_y+k'M_y^R)\sin\alpha+\sum_\beta T_{\alpha\beta}m_\beta. \quad (11)$$

Minimization of (10) with respect to θ leads to the following picture. Here three phases are possible. Phase A, in which the magnetic structure of the rare-earth and iron sublattices is described by the reducible representation Γ_{43} of the group D_{2h}^{16}. In this phase

$$\cos\theta=0, \quad m_1=-m_2=m_3=-m_4=m_0.$$

Phase B, in which the magnetic structure of the iron sublattice is the same as in phase A, but that of the rare-earth sublattice is determined by the irreducible representation Γ_{843}. In this phase

$$\cos\theta=0, \quad m_1=-m_2=-m_3=m_4=m_0.$$

And phase C, in which the magnetic structure of the iron sublattice is described by the reducible representation Γ_{32}, of the rare-earth by the reducible representation Γ_{8532} of the group D_{2h}^{16}. Phase C is doubly degenerate; in it

$$\cos\theta=1, \quad m_1=-m_2=m_3=m_4=m_0$$

or

$$\cos\theta=-1, \quad m_1=-m_2=-m_3=-m_4=m_0.$$

Table II gives the magnetic symmetry elements and the nomenclature of the phases when $\mathbf{H}\|b$; their stability limits are determined by the values of H_1^b, H_2^b, H_3^b, and H_4^b:

$$\begin{aligned}
H_1^b&=-(T_1-T_2+T_3-T_4)m_0/\sin\alpha-k'm_0\sin\alpha,\\
H_2^b&=(T_1-T_2-T_3+T_4)m_0/\sin\alpha,\\
H_3^b&=(-T_1-T_2-T_3+T_4)m_0/\sin\alpha-(\lambda'+{}^1/_2k'm_0\cos\alpha)\operatorname{ctg}\alpha-{}^1/_2k'm_0\sin\alpha,\\
H_4^b&=(T_1+T_2-T_3+T_4)m_0/\sin\alpha+(\lambda'+{}^1/_2k'm_0\cos\alpha)\operatorname{ctg}\alpha-{}^1/_2k'm_0\sin\alpha.
\end{aligned} \quad (12)$$

The critical fields at which first-order transitions occur between the corresponding phases are:

$$H_I^b=\frac{(-T_2-T_3+T_4)m_0}{\sin\alpha}-\left(\lambda'+\frac{1}{4}k'm_0\cos\alpha\right)\operatorname{ctg}\alpha-\frac{1}{4}k'm_0\sin\alpha+\frac{K_1^0+{}^1/_2K_2^{Fe}}{m_0\sin\alpha} \quad (13)$$

in the case of a transition between phase B and phase C;

$$H_{II}^b=\frac{(T_2-T_3+T_4)m_0}{\sin\alpha}+\left(\lambda'+\frac{1}{4}k'm_0\cos\alpha\right)\operatorname{ctg}\alpha-\frac{3}{4}k'm_0\sin\alpha-\frac{K_1^0+{}^1/_2K_2^{Fe}}{m_0\sin\alpha} \quad (14)$$

for a transition between phase C and phase A; and

$$H_{III}^b=(-T_3+T_4)m_0/\sin\alpha-{}^1/_2k'm_0\sin\alpha \quad (15)$$

for a transition between phase B and phase A.

When $H_I^b<H_{III}^b$, a transition from the antiferromagnetic phase B to the ferromagnetic phase A occurs through the intermediate phase C, and this leads to the occurrence of two discontinuities[3] on the magnetization curve for $\mathbf{H}\|b$ (see Fig. 1b). In this case, as in the preceding (for $\mathbf{H}\|a$), there occurs an additional canting of the magnetic sublattices of the iron ions because of the strong isotropic field produced by the RI at the Fe^{3+} ions, both in the intermediate phase C ($F_y=\frac{1}{2}am_0(\sin\alpha)/A, F_x=(d+\frac{1}{2}am_0\cos\alpha)/A$ and in the final ferromagnetic phase A ($F_y=am_0(\sin\alpha)/A$).

TABLE II. Magnetic symmetry and nomenclature of phases for $\mathbf{H}\|b$.

Phase	Fe^{3+} ions: Representation of group D_{2h}^{16}	Fe^{3+} ions: Magnetic configuration	R^{3+} ions: Representation of group D_{2h}^{16}	R^{3+} ions: Magnetic configuration	Magnetic symmetry elements*	Region of stability of phases
A	Γ_{43}	G_xF_{zy}	Γ_{43}	$F_y'C_x'$	$E, I, R\tilde{2}_x, R\tilde{\sigma}_x$	$H>H_1^b$
B	Γ_{43}	G_xF_{zy}	Γ_{843}	$A_x'G_y'$	$E, R\tilde{2}_x$	$H<H_3^b$
C	Γ_{32}	$F_{yx}G_z$	Γ_{8532}	$\begin{cases}A_{xy}'G_{yx}'\\F_{xy}'C_{yx}'\end{cases}$	E	$H_3^b<H<H_4^b$

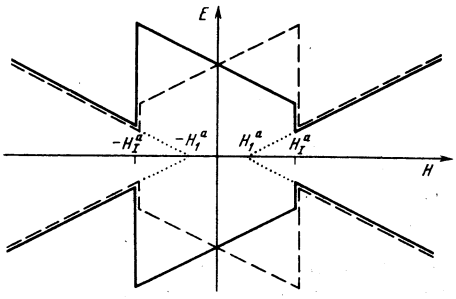

FIG. 2. Behavior of the lowest energy levels of the Tb^{3+} ions for $\mathbf{H}\|a$ (Solid line, positions 1, 4; dashed line, positions 2, 3). The dotted lines show how the lowest levels would behave if there were no phase transitions.

We shall consider the behavior of the lowest energy levels of the Tb^{3+} ions during these phase transitions. Figure 2 shows the field dependence, for $\mathbf{H}\|a$, of the lowest levels of the Tb^{3+} ions, determined as $E_\alpha=\pm m_0|h_\alpha|$ in the various phases. At sufficiently high fields, all the lower levels of the RI, magnetized in an effective field

$$(H_a+\lambda'+k'm_0\cos\alpha)\cos\alpha+\sum_\beta T_{\alpha\beta}m_\beta,$$

are equally split. It is evident from Fig. 2 that decrease of the absolute value of the external field would inevitably lead to intersection of the lower levels of the Tb^{3+} ions if there were no phase transitions in the system (the dotted curves in Fig. 2). Such an intersection (approach) of the lower levels of the ions present in a magnetic crystal is energetically disadvantageous and may lead to instability of the magnetic structure (a magnetic analog of the Jahn-Teller effect[5]). The resulting development of instability of the magnetic structure causes repulsion of the lower levels. In the present case, this instability manifests itself at $H=H_I^a$ as a SR transition $\Gamma_{42}\to\Gamma_2$ in the subsystem of Fe^{3+} ions and a metamagnetic transition $\Gamma_{842}\to\Gamma_2$ in the subsystem of Tb^{3+} ions. Figure 2 illustrates the repulsion of levels that then occurs.

A similar picture of the behavior of the lowest RI levels during phase transitions holds also when $\mathbf{H}\|b$. From this point of view, the phase transitions being considered are very similar to structural phase transitions of the Jahn-Teller type.[11,12]

3. EXPERIMENTAL RESULTS AND DISCUSSION OF THEM

In order to investigate the instability of magnetic structure that occurs during metamagnetic transitions, we undertook to measure the magnetic properties and magnetostriction of terbium orthoferrite monocrystals along various crystallographic directions. The mea-

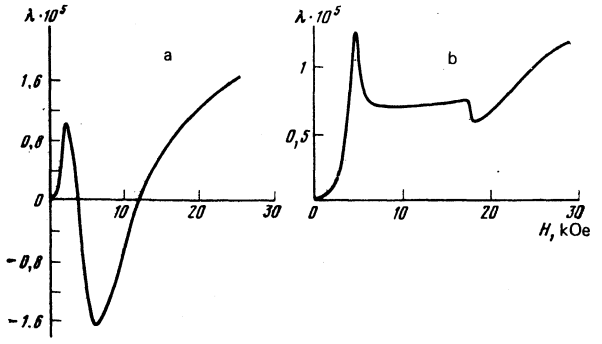

FIG. 3. Field dependence of the magnetostriction of $TbFeO_3$: a, for H||a (T = 2.7 K, λ||a); b, for H||b (T = 2.88 K, λ||b).

surements were made over the temperature interval from 2.18 to 4.2 K, in a magnetic field up to 60 kOe. The terbium orthoferrite monocrystals were grown by the method of zone melting, with optical heating.

Figure 3a shows the field variation of the longitudinal magnetostriction along the a axis of the rhombic crystal. It is evident that an anomaly of the magnetostriction occurs in the field range from 2 to 10 kOe, where, according to Ref. 3, a metamagnetic transition occurs.

Occurrence of an anomaly of the magnetostrictive strains on reorientation of the spins of the iron ions was observed earlier[13] in orthoferrites. Thus in the reorientational transition $\Gamma_4 \rightarrow \Gamma_2$, there occurs along the a axis of the crystal an additional deformation $\Delta l/l \approx 10^{-5}$,[13] due to the Fe^{3+} ions, while the magnetostriction produced by the RI, as a rule, is small and varies quadratically with the field.

In our case, there is observed in the low-temperature range a more complicated type of field variation of the magnetostriction, apparently due to an increase of the rare-earth contribution to the magnetostriction. Besides the positive magnetostriction (~10^{-5}), which can be attributed to reorientation of the spins of the Fe^{3+} ions ($\Gamma_4 \rightarrow \Gamma_2$), there is observed an abrupt change of sign of the magnetostriction (the value of the strain is $\Delta l/l \approx -3 \cdot 10^{-5}$), which is apparently due to the metamagnetic transition $A_x'G_y' \rightarrow F_x'C_y'$ in the rare-earth subsystem. The subsequent increase of the magnetostriction in fields above 10 kOe may be explained on the basis of the contribution to the magnetrostriction of the R–Fe interaction due to additional canting of the sublattices of iron ions in an external magnetic field (see above).

The correctness of these ideas is confirmed by the results of measurements of the longitudinal magnetostriction in $TbAlO_3$ monocrystals with H||a, where a qualitatively different type of field dependence of the magnetostriction was observed: at large fields the magnetostriction, produced in this case solely by the rare-earth ions, reached saturation and remained unchanged with increase of the field. The sign of the magnetostriction accompanying the metamagnetic transition was, just as in $TbFeO_3$, negative.

When the field is applied along the b axis, two anomalies of the magnetostriction occur (Fig. 3b), at fields $H_I^b \sim 5$ kOe and $H_{II}^b \sim 18$kOe, which correspond to the two-stage metamagnetic transition observed in Ref. 3 with H||b. In this case, magnetic inversion of the RI occurs through the intermediate phase Γ_{8532} and is accompanied by reorientation of the spins of the Fe^{3+} ions in the phase Γ_2. This, in particular, is corroborated by the positive spike of the magnetostriction at $H_I^b \sim 5$kOe and the negative at $H_{II}^b \sim 18$kOe.

TABLE III. R–R interaction constants in $TbFeO_3$.

$T_\alpha \cdot 10^{-21}$, см$^{-3}$	Dipole contribution	Exchange contribution	Total value	$T_\alpha \cdot 10^{-21}$, см$^{-3}$	Dipole contribution	Exchange contribution	Total value
T_1	28.5	−29.5	−1.0	T_3	−12.4	−51.2	−63.6
T_2	3.4	−3.5	−0.1	T_4	10.2	12.4	22.6

Thus our investigations of magnetostriction in terbium orthoferrite show that during the metamagnetic transitions in $TbFeO_3$ there occurs an instability of the magnetic structure of the Fe^{3+} ions.

By use of the experimental values of the critical fields for H||a and for H||b and of the expressions (9), (13), and (14) for them obtained above, it is possible to estimate the exchange contribution T_α^{ex} to the R–R interaction constant, and the constant a of isotropic R–Fe exchange. As supplementary relations we shall use the expression for the temperature of the first SR transition in $TbFeO_3$ obtained in Ref. 9 ($kT_{R1} = (\lambda' m_0 \cos\alpha)^2/K_1^0 + (T_1 + T_2 + T_3 + T_4)m_0^2$) and the value $T_N^0 = (T_1 - T_2 - T_3 + T_4)m_0^2/k_B = 4.3$ K, determined in Ref. 9 by matching with experimental data on the temperatures of the spontaneous SR transitions in $TbFeO_3$. On setting $H_I^a = 2$ kOe, $H_I^b = 5$ kOe, $H_{II}^b = 18$ kOe, $H_E = A/\mu_{Fe} \approx 10^7$ Oe, $H_D \approx 10^5$ Oe, $T_{R1} \approx 6.5$ K, K_1^{Fe} and K_2^{Fe} the same as in $YFeO_3$ ($K_1^{Fe} = 0.556 \cdot 10^{-16}$ erg/ion, $K_2^{Fe} \approx 0.4K_1^{Fe}$,[14] $K_1^{VV} = 0.1 \cdot 10^{-16}$ erg/ion[9]), we get a ≈ 118 kOe and the values of the R–R interaction constants given in Table III. There also is given the dipole component of T_α (for an infinite specimen), which makes it possible to separate out the exchange contribution to the R–R interaction.

[1)] Here the coefficients b_i contain a contribution from Van Vleck (VV) interaction of the ground quasidoublet of the Tb^{3+} ion with the excited states. In the expression (2) we have omitted several small terms due to this Van Vleck interaction and important only when H|| c (for details, see Ref. 9).

[2)] Here we neglect the splitting Δ_0 of the ground quasidoublet of Tb^{3+} in the crystalline field, since it is small[3] ($\Delta_0 \lesssim 1$ K) in comparison with the splitting in the effective field h_α.

[3)] The contribution of the external field to H_i can be neglected, since in the field range under consideration $\mu_{Fe}H_{x,y} \ll d$, $aM_{x,y}^R$.

[4)] The constants λ', p', k' are connected with the corresponding parameters λ, p, k used in Ref. 9 by the relations $\lambda = \lambda' \cos\alpha$, $p = p' \cos\alpha$, $k = k' m_0^2 \cos^2\alpha$.

[5)] In the system under consideration still another phase is possible, in which the magnetic structure of the rare-earth sublattice is described by the reducible representation Γ_{42} ($F_x' C_y'$) of group D_{2h}^{16}, and that of the iron sublattice by the reducible representation Γ_{42} (G_{xz} F_{zx}). In this phase, the iron subsystem is in an angular configuration, in which the angle θ is determined by the equation $K_2^{Fe}\cos^3\theta + (K_1^0 - k'm_0^2\cos^2\alpha)\cos\theta - (\lambda' m_0 \cos\alpha + H_x m_x^{Fe}) = 0$. But in $TbFeO_3$ this phase does not occur.

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Dependence of the NMR relaxation time on the magnetic field in quasi-one-dimensional and quasi-two-dimensional crystals

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The effect of electron transitions between chains on the dependence of the NMR relaxation rate T_1^{-1} on the magnetic field H in highly conducting quasi-one-dimensional compounds is considered. It is shown that the study of this dependence is an effective method for determining the transverse resonance integrals in quasi-one-dimensional compounds. The dependence of T_1^{-1} on H in donor-acceptor crystals of the TTF-TCNQ and HMTTF-TCNQ type is found. The transverse resonance integrals in TTF-TCNQ are determined from data for the NMR relaxation. The experimentally observed logarithmic dependence of the NMR relaxation rate on the magnetic field in the compound HMTSF-TCNQ is explained. The dependence of the relaxation rate on the magnetic field in quasi-two-dimensional compounds is found.

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1. INTRODUCTION

Experimental studies of the NMR relaxation in highly conducting quasi-one-dimensional crystals of the TTF-TCNQ type[1-4] show that, at room temperatures and in sufficiently strong magnetic fields (10–40 kOe), the dependence of the NMR relaxation rate T_1^{-1} on the magnetic field H is observed to be of the form $T_1^{-1} \sim H^{-1/2}$ characteristic for one-dimensional systems. The increase of the quantity T_1^{-1} with decrease of H breaks down in fields below about 10 kOe, and T_1^{-1} tends to a constant limit as $H \to 0$. At the same time, the theory of NMR relaxation in metals shows that, when scattering of electrons by impurities or phonons is disregarded, the quantity T_1^{-1} in systems of any dimensionality is independent of the magnetic field H so long as the Zeeman energy $\hbar\omega_e$ of the electrons remains small compared with their Fermi energy ε_F. The latter condition is fulfilled by a large margin in the quasi-one-dimensional crystals investigated in Refs. 1–4. In connection with this it was noted in Ref. 1 that the dependence $T_1^{-1} \sim H^{-1/2}$ in quasi-one-dimensional crystals may be associated with nuclear-spin relaxation via the electron system in conditions of strong scattering of electrons moving along the chain. In this situation the spectrum of the long-wavelength electron spin excitations responsible for the NMR relaxation via the electron system acquires a diffusive character: $\omega(k) = Dk^2$, where D is the coefficient of diffusion of the electrons along the chain. When the motion of the spin excitations has a diffusive character the spin density at a given site decreases with time t like $t^{-1/2}$, t^{-1}, and $t^{-3/2}$ as $t \to \infty$ in systems with dimensionaltiy 1, 2 and 3, respectively. This decay gives dependences of the rate of diffusion of the excitations on the frequency ω_e of the form $\omega_e^{-1/2}$, $|\ln\omega_e|$, and const as $\omega_e \to 0$ for systems with dimensionality 1, 2, and 3.[1)]

It has also been noted[1] that the limitation of the relaxation rate as $H \to 0$ in quasi-one-dimensional metals is connected with the effects of the three-dimensional motion of the electron spin excitations, i.e., with transitions of electrons from filament to filament (in layer crystals the limitation of the increase as $H \to 0$ is connected with transitions of electrons between layers).[2)]

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