2) We note that "induced diffusion of the second kind," due to indirect nuclear-nuclear interaction,[16, 17] will be *a fortiori* weaker than the direct relaxation for any r_{ij}; therefore its role may be neglected.

3) The crystals were obtained from the Institute of Semiconductors of the Ukrainian Academy of Sciences.

4) Here and later on, the magnetic quantum numbers correspond to the states that are significantly predominant in the given energy level (no pure states are realized in the spectrum of the given material).

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Nature of the spontaneous and field-induced low-temperature orientational transitions in erbium orthoferrite

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A theory is constructed and an experimental investigation is carried out on the field-induced spin-flip transitions in $ErFeO_3$ at low temperatures, for two orientations of the external magnetic field: $\mathbf{H}\|a$ and $\mathbf{H}\|c$, where a and c are the crystal axes. The threshold fields of the phase transitions are determined from the isotherms of the magnetostriction $\lambda(H)$ along the c axis. The dependence of the threshold fields on the ratio of the Er-Er and Er-Fe interactions is explained; from a comparison of the theoretical and experimental results, estimates are obtained for the values of these interactions in $ErFeO_3$. It is shown that interaction between rare-earth ions plays a dominant role in low-temperature spin-flip phenomena in erbium orthoferrite.

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$ErFeO_3$ is the only orthoferrite in which ordering of the rare-earth ions at low temperatures is accompanied by a simultaneous spin-flip (SF) transition in the subsystem Fe^{3+} ions, such that the rare-earth and iron spin configurations resulting from the phase transition belong to a single representation of the symmetry group of the orthoferrites, i.e., are compatible spin configurations. It has been established experimentally[1,2] that the low-temperature SF transition in the subsystem of Fe^{3+} spins at $T = T_R \approx 4$ K is a second-order phase transition $\Gamma_2 - \Gamma_{12}$.1) We recall that in the configuration Γ_2, the antiferromagnetism vector $\mathbf{G}\|c$, while in the configuration Γ_1, $\mathbf{G}\|b$; here c and b are the crystal axes, and Γ_i denotes the appropriate irreducible representation of the symmetry group of orthoferrites. Γ_{12} is a reducible representation, consisting of Γ_1 and Γ_2; in the configuration Γ_{12}, the vector $\mathbf{G}$ lies in the plane bc. On lowering of temperature, the angle of deviation of the iron spins from the c axis increases continuously; extrapolation to $T = 0$ K gives $\varphi_0 = 49°$. The Er^{3+} ions are, for $T < T_R$, ordered antiferromagnetically along the c axis (according to the mode C_z

with a small admixture of modes F_x and C_y).

Since the configurations of the rare-earth and iron spins that occur at $T < T_R$ are compatible, the presence of one of them necessarily induces the appearance of the other. This means that the observed spontaneous phase transition may be determined in various degrees both by the Er–Er and by the Er–Fe interactions. Investigation of the low-temperature magnetic properties of $ErFeO_3$ has been the subject of a large number of papers (we mention, in particular, Ref. 3, in which a systematic theory of spontaneous reorientation was developed); but so far it remains an open question whether the SF transition $\Gamma_2-\Gamma_{12}$ is primarily a consequence of ordering of the Er^{3+} ions (the Er-Fe interaction may then be very small), or whether the reorientation of the iron spins occurs because of the strong anisotropy of the Er-Fe exchange, while the ordering of the Er^{3+} ions is actually a polarization of the rare-earth subsystem by the Er–Fe exchange field. It should be noted that zero-field experiments on the temperature behavior of the mode C_z of the Er^{3+} spins or of the angle of rotation of the Fe^{3+} spins in the *bc* plane do not in principle enable us to separate these interactions (see below). The assumption made in Ref. 3 regarding the closeness of the values of the Er–Er interactions in $ErFeO_3$ and in the isomorphic compound $ErAlO_3$ (the temperature of ordering of the Er^{3+} spins is $T_{Er} \approx 0.6$ K), and the conclusion that follows from it regarding the dominant influence of the Er–Er interaction on the low-temperature magnetic properties of erbium orthoferrite, are evidently inadmissible. One can involve as a counterexample the value of the parameter $T_{Er} \approx 3.8$ K in $ErCrO_3$[4] (closer to $ErFeO_3$ with respect to lattice parameters than is $ErAlO_3$).

For the purpose of explaining the mechanisms of low-temperature phase transitions and of estimating the values of the corresponding interactions, SF transitions induced in $ErFeO_3$ by an external magnetic field were investigated in the present work. The threshold fields of these transitions depend primarily on the ratio of the Er–Er and Er–Fe interactions. Independent interest attaches to an investigation, carried out in the paper, of phase transitions with $\mathbf{H} \parallel c$, when the behavior of erbium orthoferrite approximates the behavior of an Ising antiferromagnet under the conditions for manifestation of the magnetic analog of a cooperative Jahn-Teller effect. In fact, the spontaneous phase transition at $T = T_R$ is, as it were, a synthesis of an "order-order" transition (a magnetic Jahn-Teller transition, with breakdown of the symmetry in the subsystem of Fe^{3+} spins and additional splitting of the ground state of the Er^{3+} ions[5]) and a "disorder-order" transition (antiferromagnetic ordering of the Er^{3+} spins because of their own interaction). The H, T phase diagram of such a magnet, when $\mathbf{H} \parallel c$ (in the idealized model, when $\mathbf{H}$ is parallel to the Ising axis of the rare-earth ion) will have the specific form characteristic of systems described by a many-component order parameter (see below).

1. THEORY

The low-temperature spontaneous spin flip in $ErFeO_3$ has been treated with sufficient completeness by Vitebskii and Yablonskii.[5] We shall be interested primarily in the dependence of the values of the threshold fields of the field-induced SF transitions on the ratio of the Er–Er and Er–Fe interactions; for $H = 0$, the formulas of the present section are analogous to those obtained in Ref. 3.

We write the total free energy of $ErFeO_3$ in the form

$$\mathscr{F} = \mathscr{F}_{Fe}(\mathbf{G}, \mathbf{F}; \mathbf{H}) + \mathscr{F}_{Er}(\mathbf{G}, \mathbf{H}_R^+, \mathbf{H}_R^-; \mathbf{H}, T), \tag{1}$$

where the quantities $\mathbf{G}$, $\mathbf{F}$, $\mathbf{H}_R^+$, and $\mathbf{H}_R^-$ are regarded as variational parameters. Here $\mathscr{F}_{Fe}$ is the free energy of the subsystem of Fe^{3+} spins, for which we have, in the two-sublattice approximation, the standard expression[6]

$$\mathscr{F}_{Fe} = {}^1/_2 A\mathbf{F}^2 - {}^1/_2 b_1 G_x^2 - {}^1/_2 b_3 G_z^2 - d(F_x G_z - F_z G_x) - \mathbf{FH}, \tag{2}$$

whereas $\mathscr{F}_{Er}$ contains the energy of Er–Er and of Er–Fe interactions. It is assumed that the Er–Fe interaction is determined primarily by antisymmetric terms; therefore $\mathscr{F}_{Er}$ in (1) is independent of $\mathbf{F}$. The quantities $\mathbf{H}_R^+$ and $\mathbf{H}_R^-$ have the meaning of molecular fields produced by the Er–Er interaction. At low temperatures the subsystem of Fe^{3+} spins is completely saturated, and the dependence of $\mathscr{F}_{Fe}$ on T may be neglected.

We introduce angles θ and φ that determine the orientation of the vectors $\mathbf{G}$ and $\mathbf{F}$ (see Fig. 1): θ is the angle between $\mathbf{F}$ and the c axis in the *ac* plane[2]; φ is the angle between $\mathbf{G}$ and the *ac* plane in the plane perpendicular to the vector $\mathbf{F}$. The angle values $\theta = 0$, $\varphi = 0$ correspond to the configuration Γ_4; $\theta = \pi/2$, $\varphi = 0$ to the configuration Γ_2. The configuration Γ_1 corresponds to the value $\varphi = \pi/2$. After substitution in (2) of $G_x = -G\cos\theta\cos\varphi$, $G_z = G\sin\theta\cos\varphi$, $F_x = F\sin\theta$, $F_z = F\cos\theta$ and the minimization with respect to F, we get

$$\mathscr{F}_{Fe}(\theta, \varphi; H) = K_{ac}\cos^2\theta\cos^2\varphi - K_{bc}\cos^2\varphi - \mathbf{F}_0\mathbf{H}, \tag{3}$$

where $K_{ac} = \frac{1}{2}G^2(b_3 - b_1)$ and $K_{bc} = \frac{1}{2}G^2(d^2/A + b_3)$ are the anisotropy constants of the iron sublattices in the *ac* and the *bc* planes, respectively, of the crystal; $\mathbf{F}_0 = (F_{0x}, 0, F_{0z})$, where

$$F_{0x} = (dG/A)\sin\theta\cos\varphi,$$
$$F_{0z} = (dG/A)\cos\theta\cos\varphi$$

(terms $\propto H^2$ in (3) have been discarded).

An expression for the free energy $\mathscr{F}_{Er}$ is most simply

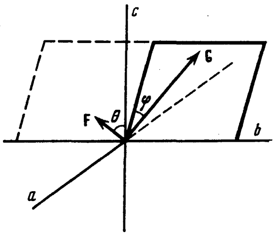

FIG. 1. Orientation of the vectors G and F.

obtained by means of a variational principle. We take into account that in $ErFeO_3$ at low temperatures, only the ground doublet of the Er^{3+} ion is occupied, and that it can be described by an effective spin $S=\frac{1}{2}$ with an anisotropic g factor. The model free energy of the system of Er^{3+} ions, located in the exchange field $\mathbf{H}_{Fe}^{\pm}$ exerted by the Fe^{3+} ions and also in an external magnetic field $\mathbf{H}$, and interacting with each other, can be written, by use of a variational theorem of Bogolyubov,[7] in the form (per $ErFeO_3$ molecule)

$$\begin{aligned}\mathscr{F}_{Er}(\theta,\varphi,\mathbf{H}_R^+,\mathbf{H}_R^-;\mathbf{H},T)=&-{}^1/_2T\ln\left(\mathrm{ch}\frac{\Delta^+}{T}\mathrm{ch}\frac{\Delta^-}{T}\right)\\&+{}^1/_2\mathbf{M}^+\hat{\lambda}\mathbf{M}^-+{}^1/_2\mathbf{M}^+\mathbf{H}_R^++{}^1/_2\mathbf{M}^-\mathbf{H}_R^-,\end{aligned}\tag{4}$$

where the signs ± distinguish two nonequivalent positions of the rare-earth ions in the orthoferrite structure. The corresponding splittings (more accurately, half-splittings) of the ground doublet of the Er^{3+} ions,

$$\Delta^{\pm}=|\hat{g}^{\pm}(\mathbf{H}_{Fe}^{\pm}+\mathbf{H}_R^{\pm}+\mathbf{H})|\tag{5}$$

and the quantities

$$\mathbf{M}^{\pm}=\frac{\partial}{\partial\mathbf{H}_R^{\pm}}\frac{T}{2}\ln\left(\mathrm{ch}\frac{\Delta^+}{T}\mathrm{ch}\frac{\Delta^-}{T}\right),\tag{6}$$

which have the meaning of magnetic moments of the Er^{3+} ions in the nonequivalent positions, are functions of the variational parameters θ, φ, $\mathbf{H}_R^+$, and $\mathbf{H}_R^-$. The form of the tensors $g^{\pm}$ and of the dependences of the fields $\mathbf{H}_{Fe}^{\pm}$ on the orientation of the vector $\mathbf{G}$ is given, for example, in the paper of Zvezdin, Mateveev, and Popov[8]; $\hat{\lambda}$ is the Er–Er interaction tensor. The splittings (5) are measured in degrees Kelvin (K), the components of the g tensor in [K/kOe] (g[K/kOe] $=\frac{1}{15}g$).

By minimizing the sum of the expressions (3) and (4) (the total free energy of $ErFeO_3$) with respect to θ, φ, $\mathbf{H}_R^+$, and $\mathbf{H}_R^-$, one can find the equilibrium values of the variational parameters and the corresponding equilibrium states of system. As has already been mentioned in Ref. 3, in erbium orthoferrite it is apparently possible, in first approximation, to take into account only the interactions between the S_z^+ and S_z^- components of the effective spin. In any case, experiment[9] indicates absence of any temperature variation of the splitting of the ground doublet of the Er^{3+} ion in the configuration Γ_2 (spin in the *ab* plane) all the way to $T=T_R$. Below, we shall take this fact into account and shall use as variational parameters not the vectors $\mathbf{H}_R^{\pm}$ but the signal-component quantities $H_{Rz}^{\pm}\equiv H_R^{\pm}$. In this approximation, the tensor $\hat{\lambda}$ obviously has only one nonvanishing component $\lambda_{zz}\equiv\lambda$.

We turn to the investigation of concrete cases.

A. External field H parallel to the *a* axis of the crystal, and H = 0

Since at low temperatures the easiest plane of anisotropy in $ErFeO_3$ is the *bc* plane ($\mathbf{G}$ in the *bc* plane, $\mathbf{F}\parallel a$), and since a field $\mathbf{H}\parallel a$ does not divert the vector $\mathbf{G}$ from it, we may set $\cos\theta=0$ in (3) and (4). Then the molecular field $\mathbf{H}_{Fe}^{\pm}$ has the form[8] $\mathbf{H}_{Fe}^{\pm}=(a_1\cos\varphi,\pm a_2\cos\varphi,\pm a_3\sin\varphi)$, where a_i are certain constants. Since $H_{Fe z}^+=-H_{Fe x}^-$, it follows from symmetry considerations that[3)] $H_R^+=-H_R^-\equiv H_R$, and in accordance with (5)

$$\Delta^+=\Delta^-\equiv\Delta=[(\Delta_1\sin\varphi+g_{zz}H_R)^2+\Delta_2^2\cos^2\varphi+2aH\cos\varphi+bH^2]^{1/2},\tag{7}$$

where

$$\Delta_1=g_{zz}H_{Fe\,z}\text{ and }\Delta_2=[(g_{xx}H_{Fe\,x}+g_{xy}H_{Fe\,y})^2+(g_{yx}H_{Fe\,x}+g_{yy}H_{Fe\,y})^2]^{1/2}$$

are the splittings of the ground doublet of the Er^{3+} ion, in the configurations Γ_1 and Γ_2 respectively, because of the Er–Er interaction; $b=g_{xx}^2+g_{yx}^2$; $a=(g_{xx}^2+g_{yx}^2)H_{Fe\,x}+(g_{xx}g_{xy}+g_{yy}g_{yx})H_{Fe\,y}$; $H_{Fe\,i}\equiv H_{Fe\,i}^+$; and $g_{ij}\equiv g_{ij}^+$. The expression for the free energy simplifies to

$$\mathscr{F}(\varphi,H_R;H,T)=-K_{bc}\cos^2\varphi-F_0H\cos\varphi-T\ln\mathrm{ch}(\Delta/T)-{}^1/_2\lambda M^2+MH_R.\tag{8}$$

The quantity

$$M\equiv M^+=-M^-=\frac{\partial}{\partial H_R}\left(T\ln\mathrm{ch}\frac{\Delta}{T}\right)$$

in (8) is a function only of the two variational parameters φ and H_R; $F_0=|\mathbf{F}_0|=dG/A$.

From the extremization conditions $\mathscr{F}'_{\varphi}=0$ and $\mathscr{F}'_{H_R}=0$, two solutions are found: 1) $\sin\varphi=H_R=0$ (Γ_2); 2) $\sin\varphi\neq0$, $H_R\neq0$ (Γ_{12})[4)]; in the second case, the equilibrium values of $\sin\varphi$ and H_R for given H and T are determined by the system of equations

$$H_R=\lambda M=\frac{\Delta_1\lambda gf}{1-\lambda g^2f}\sin\varphi,\tag{9a}$$

$$2K_{bc}-f\left(\frac{\Delta_1^2}{1-\lambda g^2f}-\Delta_2^2\right)=(F_0-af)H,\tag{9b}$$

where

$$f=f(\varphi,H_R;H,T)=\frac{1}{\Delta}\mathrm{th}\frac{\Delta}{T}\tag{10}$$

($\Delta=\Delta(\varphi,H_R;H)$ from (7), $g_{zz}\equiv g$).

The regions of stability of the configurations Γ_2 and Γ_{12} in the H,T phase diagram are separated by a line of second-order phase transition. On it, the temperature dependence of the threshold field is given by the equation

$$2K_{bc}-f'\left(\frac{\Delta_1^2}{1-\lambda g^2f'}-\Delta_2^2\right)=(F_0-af')H,\tag{11}$$

where $f'=f(0,0;H,T)$. It is simpler, however, to obtain in explicit form the inverse relation $T=T(H)$. In fact, equation (11) reduces to a quadratic equation in f', with coefficients independent of T. On expressing f' from (11) as a function $f'=f'(H)$, we get $f'(H)=f(0,0;H,T)$, or

$$T=\Delta_2(H)/\mathrm{arcth}[\Delta_2(H)f'(H)],\tag{12}$$

where $\Delta_2(H)=(\Delta_2^2+2aH+bH^2)^{1/2}$. We shall not give the cumbersome formulas, containing radicals, for $f'=f'(H)$; for concrete values of the parameters of the theory (see below), the second-order phase-transition lines $\Gamma_2-\Gamma_{12}$ are plotted by use of (12) in Fig. 4.

When $H=0$, (11) reduces to an equation that determines the temperature T of spontaneous phase transition $\Gamma_2-\Gamma_{12}$:

$$\begin{gathered}2K_{bc}-f''\left(\frac{\Delta_1^2}{1-\lambda g^2f''}-\Delta_2^2\right)=0,\\f''=\frac{1}{\Delta_2}\mathrm{th}\frac{\Delta_2}{T}.\end{gathered}\tag{13}$$

When $T>T_R$, the left member of this equation (the effective anisotropy) is positive, and the configuration Γ_2 is stable. On decrease of temperature, the value of f'' increases, and with it the second (negative) term in the effective anisotropy. Upon satisfaction of the condition (13) at the point $T=T_R$, a phase transition occurs. It is obvious that $T_R=\Delta_2/\operatorname{arctanh}\Delta_2 f_0$ [see (12)]; f_0 is a constant, dependent only on the parameters Δ_1, Δ_2, K_{bc}, and λ of the theory, which is determined by equation (13).

As is seen from (9b), in the configuration Γ_{12} at $H=0$ the quantity $f=f(\varphi, H_R; 0, T)=f_0=\text{const}$. This fact means the following:

1) $f_0=f(\varphi, H_R; 0, 0)=\Delta_0^{-1}$ (Δ_0 is the splitting of the ground doublet of the Er^{3+} ion in the configuration Γ_{12} at $T=0$ and $H=0$);

2) in accordance with (9a), $H_R \propto M \propto \sin\varphi$;

3) $\Delta=\Delta(\varphi, H_R; 0)=[(\Delta_1 \sin\varphi+gH)^2+\Delta_2^2\cos^2\varphi]^{1/2} =(\tilde{\Delta}_1^2\sin^2\varphi+\Delta_2^2\cos^2\varphi)^{1/2}=\Delta(\varphi)$, where $\tilde{\Delta}_1=\Delta_1/(1-\lambda g^2 f_0) =\text{const}$;

4) one can find the relation $T=T(\varphi)$ [and the corresponding relations $\sin\varphi(T)$, $M(T)$, and $H_R(T)$] by means of the formula $T=\Delta(\varphi)/\operatorname{arctanh}[\Delta(\varphi)/\Delta_0]$.

It follows from the formulas given about that experiments on investigation of the temperature behavior of the splitting of the ground doublet and of the angle of rotation of the Fe^{3+} spins in the *bc* plane (or of the mode $C_z=2M$ of the rare-earth spins) can give values of the parameters T_R, Δ_2, Δ_0, and $\sin\varphi_0$ ($\varphi_0=\varphi(T=0)$), and of the quantity $\tilde{\Delta}_1$. These data are insufficient for separation of the Er–Er and Er–Fe interactions. For estimation of these interactions, it is necessary to know the anisotropy of the Fe^{3+} ion in the *bc* plane. Then from equation (13), which can be written in the form

$$2K_{bc}-f_0(\tilde{\Delta}_1\Delta_1-\Delta_2^2)=2K_{bc}-\frac{\tilde{\Delta}_1\Delta_1-\Delta_2^2}{\Delta_0}=0, \tag{14}$$

the parameter Δ_1 would be determined. But the relation between λg^2 and Δ_1 is determined by the value of the parameter $\tilde{\Delta}_1$. Unfortunately the constant K_{bc} is unknown; furthermore, it can be estimated only in an indirect way for orthoferrites in which spontaneous spin-orientation transitions are observed in the *bc* (or *ab*) plane. Thus it is necessary to determine the constant K_{bc} from known values of Δ_1 and λg^2, rather than the other way around. At the same time, the values of the threshold fields of the field-induced phase transitions depend on the ratio of the values of the parameters Δ_1 and λg^2 [as is illustrated (see below) by curves 1 and 2 in Fig. 4]. Measurement of the threshold fields enables us to estimate the Er–Er and Er–Fe interactions in $ErFeO_3$.

B. External field H parallel to the *c* axis of the crystal

In this case it is necessary to use the general expression for the free energy [see (3) and (4)] and to perform a minimization with respect to the four parameters θ, φ, H_R^+, and H_R^-. From the extremum conditions we get

$$H_R^{\pm}=-\lambda M=-\lambda g\Delta_z^{\mp}f^{\mp}, \tag{15a,b}$$

$$\sin\varphi=-\frac{\Delta_1(H_R^+-H_R^-)}{2\lambda g[2K_{bc}+{}^1/_2\Delta_2^2(f^++f^-)]}=\frac{\Delta_1 C_z}{2g2K_{bc}^{\text{eff}}}, \tag{15c}$$

$$\cos\theta\cos\varphi=-\frac{\Delta_4(H_R^++H_R^-)}{2\lambda g[2K_{ac}-{}^1/_2\Delta_2^2(f^++f^-)]}=\frac{\Delta_4 F_z}{2g2K_{ac}^{\text{eff}}}, \tag{15d}$$

where $f^{\pm}=1/\Delta^{\pm}\tanh\Delta^{\pm}/T$, $\Delta^{\pm}=[(\Delta_z^{\pm})^2+\Delta_2^2\sin^2\theta\cos^2\varphi]^{1/2}$, $\Delta_z^{\pm}=\pm\Delta_1\sin\varphi+gH_R^{\pm}+\Delta_4\cos\theta\cos\varphi+gH$; Δ_4 is the splitting of the ground doublet in the configuration Γ_4 by the Er–Fe exchange, $C_z=M^+-M^-$, $F_z=M^++M^-$. Even without detailed analysis of the complicated equations (15) it is evident that a field $\mathbf{H}\|c$ leads to the appearance and growth of the mode F_z and diminution of the mode C_z in the rare-earth subsystem, thereby inducing, through the Er–Fe interaction, a spatial SF transition $\Gamma_{12}-\Gamma_{124}-\Gamma_{24}$ in the subsystem of Fe^{3+} spins (when $T<T_R$).[5)] Here is realized a special case of the phenomenon of enhancement of the external field.[10] when SF transitions occur actually not in the external field but in the Er–Fe molecular field.[6)] In particular, far from T_R (for $T\to 0$) substantial changes in the state of the system should take place for $H\sim\Delta_0/g$, when the effect of the field on the values of the F_z and C_z modes of the antiferromagnetically ordered rare-earth subsystem is greatest.

In $ErFeO_3$ the value of Δ_4 is very small.[9] According to (15c, d), this means that $\cos\theta\approx 0$ and that for $T<T_R$, the SR transition $\Gamma_{12}-\Gamma_{124}-\Gamma_{24}$ is close to the transition $\Gamma_{12}-\Gamma_2$ (that is, a field $\mathbf{H}\|c$ induces a SF transition inverse to the spontaneous transition). Nevertheless, even in the approximation $\cos\theta=0$ exact solutions (φ, H_R^+, H_R^-) of equations (15) in their stability region on the *H*, *T* phase diagram can be found only by numerical methods. It is interesting that simplification of the model by the assumption $\Delta_2=0$ makes it possible to carry out the investigation in analytical form. Having our experimental results in mind, we shall give a theoretical *H*, *T* phase diagram when $\mathbf{H}\|c$ for the case $\lambda g^2>\Delta_1$. It is shown in Fig. 2. We note the following features of this diagram, 1) Near T_R ($T_1<T<T_R$), the field-induced SF transition $\Gamma_{124}-\Gamma_{24}$ is a second-order phase transition; 2) At low temperatures ($T<T_2$), it is a first-order phase transition[7)]; 3) In the intermediate range $T_2<T<T_1$, the reorientation $\Gamma_{124}-\Gamma_{24}$ occurs by two phase transitions of first and second order ($\Gamma_{124}-\Gamma_{124}-\Gamma_{24}$). On the diagram there is a critical point *O* of termination of the line of phase transitions of the first order. In a real crystal, in the region bounded by the lines of loss of stability *OA* and *OB* the existence of an intermediate state is possible.

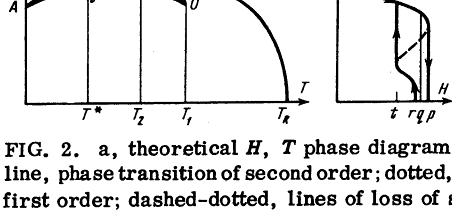

FIG. 2. a, theoretical *H*, *T* phase diagram when $\mathbf{H}\parallel c$. Solid line, phase transition of second order; dotted, phase transition of first order; dashed-dotted, lines of loss of stability. b, schematic dependence of the angle φ on the magnetic field when $T=T^*$.

2. EXPERIMENT AND DISCUSSION OF RESULTS

On a single crystal of erbium orthoferrite, grown by the method of zone melting, isotherms of the magnetostriction along the c axis of the crystal were measured for orientations of the external magnetic field along the a and c axes, over the temperature range 2.17–4.2 K, in fields up to 60 kOe. The measurements were made by the strain-gauge method, by means of special strain gauges that had a small galvanomagnetic effect at low temperatures.

A. $\mathbf{H}\|a$. Isotherms of the magnetostriction $\lambda_{ca}(H)$ for this case are shown in Fig. 3. When $T<4$ K, there is a break on them that we attribute to the field-induced SF transition $\Gamma_{12}-\Gamma_2$. The break field H_{th} (the treshold field of the SF transition) increases with lowering of temperature and reaches 20 kOe at $T=2.17$ K. The additional strain of the crystal caused by rotation of the Fe^{3+} spins in the bc plane is $-3\cdot10^{-5}$ at $T=2.17$ K. The temperature dependence of the threshold field, obtained from the $\lambda_{ca}(H)$ curves, is shown in Fig. 4 (experimental points). The error in the determination of the threshold field was ±0.5 kOe. Plotted in the same figure are theoretical curves, from formula (12), of the threshold field for the two extreme cases $\lambda g^2=0$ (Curve 1) and $\Delta_1\to0$ (Curve 2), for the parameter values $\Delta_2=2.26$ K, $\Delta_0=4.42$ K,[9] $a=0.15$ K/kOe, $b=0.014$ K/kOe². The first case corresponds in (12) to $f'(H)=\Delta_0^{-1}(\Delta_1^2-\Delta_2^2-F_0\Delta_0H)/(\Delta_1^2-\Delta_2^2-aH)$, the second to $-f'(H)=f_0=\Delta_0^{-1}=\text{const}$. The parameters a and b (together with Δ_2) determine the magnetic moment and susceptibility of the Er^{3+} ion along the a axis of the crystal resulting from splitting of the ground doublet [in particular, $M_a^{\mathrm{Er}}(H,T)=af$] and can be found from the experimental values of $M_a^{\mathrm{Er}}(H,T)$ and $\chi_a^{\mathrm{Er}}(H,T)$. The values of a and b given above were obtained by processing of the experimental data of Refs. 11 and 12; incidentally an estimate was obtained for the Van Vleck susceptibility of Er^{3+} along the a axis: $\chi_{VV}=5\cdot10^{-4}$ G/gOe.

As seen from Fig. 4, the dependence of the threshold fields of the SF transition $\Gamma_{12}-\Gamma_2$ on the ratio of the values of the Er–Er and Er–Fe interactions shows up most clearly at the lowest temperatures. It is characteristic that when $\Delta_1\geqslant\lambda g^2$, the values of these fields vary little with the ratio of the parameters Δ_1 and λg^2; in particular, the curve corresponding to $\Delta_1=\lambda g^2$ (Curve 3 in Fig. 4) is very close to Curve 1. The theoretical threshold-field curve that agrees best with the experimental points corresponds to $\lambda g^2=3.4$ K; that is, the Er–Er interaction dominates. By using (14), we can now obtain the anisotropy constant of the Fe^{3+} ions in the bc plane; $K_{bc}=0.2$ K (for comparison, the constant K_{ac} in yttrium orthogerrite as 0.18 K[13]).

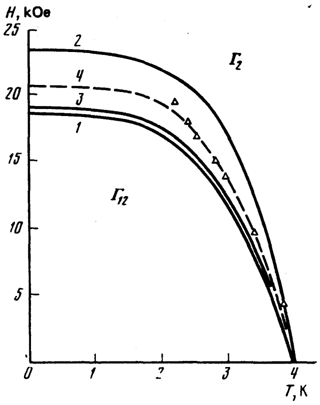

FIG. 4. H, T phase diagram of $\mathrm{ErFeO_3}$ when $\mathbf{H}\|a$. The numerals mark the theoretical curves: 1, $\lambda g^2=0$; Δ_1 0; 3, $\Delta_1=\lambda g^2$; 4, $\Delta_1=1.3$ K and $\lambda g^2=3.4$ K. The symbols Δ are the experimental points.

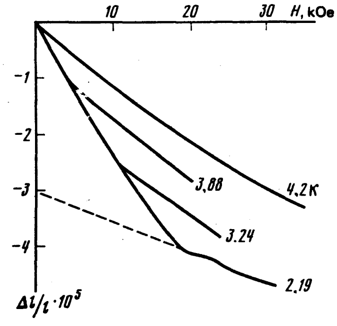

FIG. 3. Isotherms of the magnetostriction of an $\mathrm{ErFeO_3}$ crystal for $\mathbf{H}\|a$ and $\lambda\|c$.

B. $\mathbf{H}\|c$. Characteristic isotherms of the magnetostriction $\lambda_{cc}(H)$ for this case are shown in Fig. 5. In the temperature range $2.17<T<3.4$ K, application of an external magnetic field causes an abrupt rise of the strain, with a split maximum at $H\sim4$ kOe (Curve 1). When $T>3.4$ K, the split maximum of the strain disappears, and the value of the maximum decreases with rise of temperature (Curve 2). For $T>4$ K, there is no anomaly on the isotherms $\lambda_{cc}(H)$ (Curve 3). We note also that a positive blip on the magnetostriction isotherms is observed at precisely those fields where the magnetization of $\mathrm{ErFeO_3}$ along the c axis is changing most rapidly.[8)]

It is obvious that at low temperatures, a substantial contribution to the magnetostriction should be made by the strongly magnetized rare-earth ions.[9)] In our case, the contribution of the Er^{3+} ions to $\lambda_{cc}(H)$ can be written in the form

$$\lambda_{cc}^{\mathrm{Er}}=\lambda[(M^+(H,T))^2+(M^-(H,T))^2]=\frac{\lambda}{4}[C_z^2(H,T)+F_z^2(H,T)], \quad (16)$$

if we suppose that the rare-earth magnetostriction is

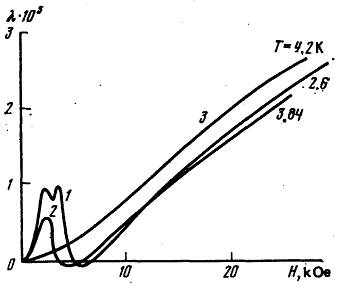

FIG. 5. Isotherms of the longitudinal magnetostriction of an $\mathrm{ErFeO_3}$ crystal for $\mathbf{H}\|c$, $\lambda\|c$.

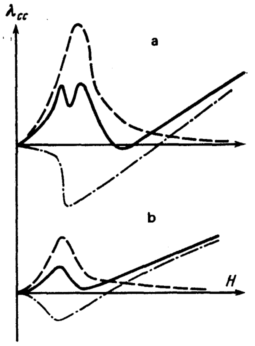

FIG. 6. Contributions to the magnetostriction from the Er^{3+} and from the Fe^{3+} ions when $\mathbf{H} \parallel c$: a, low temperatures; b, near T_R. Dotted, λ_{cc} (H); dashed-dotted, $\lambda_{cc}{}^{Fe}$ (H); solid line, $\lambda_{cc}{}^{Fe}$ $(H)+\lambda_{cc}{}^{Er}$ (H).

of single-ion character. From the point of view of microscopic theory, the symmetric formula (16) is seemingly not completely rigorous (we shall not discuss this question); but qualitatively it correctly reflects the fact that during the metamagnetic transition $C_z \rightarrow F_z$ in the subsystem of Er^{3+} spins, at the instant when one of the sublattices (M^-) is completely demagnetized (the external field equal to the molecular), the strain of the crystal must differ significantly from the strains that are nearest to each other in the saturated states ($F_z = 0$ and $C_z \sim 0$). If the parameter $\lambda < 0$, then at $M^- = 0$ there will be observed on the $\lambda_{cc}^{Er}(H)$ curve a positive blip $\Delta\lambda_{cc}^{Er} \sim \lambda(M^+(0, T))$. The Fe^{3+} ions are responsible for a negative jump of the strain during a SF transition of first order ($T < 3.4$ K) or a break on the $\lambda_{cc}^{Fe}(H)$ curve during a SF transition of second order ($T > 3.4$ K). Figure 6 shows schematically how the unusual character of the magnestriction isotherms $\lambda_{cc}(H)$ is due to a difference of the contributions from the Er^{3+} and from the Fe^{3+} ions. This interpretation of the experimental results for $\mathbf{H} \parallel c$ agrees qualitatively with the theoretical H, T phase diagram (Fig. 2). More detailed information about the H, T phase diagram (the presence of critical points, of an intermediate state, etc.) can be provided by experimental methods that permit direct tracing of the orientation of the iron spins (for example, the NMR method[2]).

In conclusion, we give still another estimate, which shows that in fields $\mathbf{H} \parallel c$ of the order of magnitude of the threshold fields, it is correct in formulas (15) to disregard the direct interaction of the external field with the Fe^{3+} spins. We have $F_0 H_{th}/\Delta_1/gC_z \sim F_0/M_c^{Er} \sim 10^{-2}$, since $H_{th} \sim \Delta_0/g \sim \Delta_1/g$.

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1) In $ErFeO_3$ at higher temperatures ($T \sim 100$ K) there is still another SR transition $\Gamma_2 - \Gamma_{24} - \Gamma_4$, which is not considered in this paper.

2) This paper investigates the behavior of $ErFeO_3$ for orientations of the external magnetic field along the crystal axes a and c. In these cases, the weakly ferromagnetic **F** lies in the ac plane.

3) This fact can of course be established directly by minimization of the general expression for the free energy of $ErFeO_3$.

4) The solution $\cos\varphi = 0$, $H_r \neq 0$ that is possible when $H = 0$ is not realized in $ErFeO_3$.

5) Depending on the parameters of the theory (when $\Delta_4 \gg \Delta_1$), the sequence $\Gamma_{12} - \Gamma_{124} - \Gamma_{14}$ is also possible. In both cases, the final configuration in very large fields will be the configuration Γ_4.

6) Direct interaction of H with the iron sublattice is negligibly small and is disregarded in (15). Appropriate estimates for $ErFeO_3$ are given in the next section.

7) This does not contradict the presence on the H, T phase diagram, for $T < T_2$, of a line of phase transtion of second order, which is a line of direct transition from the metastable phase Γ_{124}, and which can manifest itself only on hysteresis curves during decrease of the external field (Fig. 2b).

8) Our experimental curves $M_c = M_c(H)$ for $T < T_R$ have the form that is usual for the metamagnetic transition $C_z \rightarrow F_z$ (see, for example, Ref. 1) and are not given here.

9) For example, the Tb^{3+} ions in $TbAlO_3$ give noticeable magnetostriction.[14]

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