New orientational transitions induced in orthoferrites by an external field

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The magnetization and magnetostriction of single crystals of dysprosium orthoferrite have been measured over the temperature range 4.2-60 °K, in magnetic fields up to kOe. Various types of reorientational transition, induced by an external magnetic field, have been observed at temperatures below the Morin point. On application of a field along the *c* axis of the crystal, there was observed a reorientation of the spins in the plane ab perpendicular to the field direction. A peculiarity of the transitions under the action of an external field along the *a* and *b* axes is that the reorientation occurs under the influence of the molecular field of the rare-earth ions, amplified by the external field. The experimental and theoretical phase diagrams H(T) agree qualitatively with each other.

INTRODUCTION

Despite the large amount of research devoted to the investigation of spin reorientation in rare-earth orthoferrites (RFeO₃), there has so far been little study of the reorientational transition from a weakly ferromagnetic to an antiferromagnetic state (a transition of the Morin type), which is observed for example in dysprosium orthoferrite. It is known that at high temperatures dysprosium orthoferrite is weakly ferromagnetic, the spins of the iron ions being oriented along the a axis of the rhombic crystal (the magnetic structure $G_X F_X$). On lowering of the temperature to ~ 40 K, the spins of the iron ions are reoriented to the b axis $(G_X F_Z \rightarrow G_y)$, and the crystal becomes antiferromagnetic^[1,2]. On further lowering of temperature, there occurs at about 4 K an antiferromagnetic ordering of the spins of the dysprosium ions, in the mode A_yG_x ; the spin configuration of the iron ions is unchanged^[3].

Although the exchange interaction of the rare-earth and iron ions does not exert appreciable influence on the ordering of the dysprosium ions, it determines to a significant degree the magnetic behavior of dysprosium orthoferrite at higher temperatures. The presence of exchange interaction can, in particular, lead to the occurrence in dysprosium orthoferrite of unusual reorientational transitions induced by an external magnetic field. It should be mentioned that the effect of a magnetic field on a reorientational transition of the Morin type in orthoferrites has hitherto received practically no attention.

THEORY

We shall consider the effect of a magnetic field on a phase transition of the Morin type in a rare-earth orthoferrite (dysprosium orthoferrite). It is convenient to present the results of an investigation of the effect of a magnetic field on phase transitions in the form of magnetic phase diagrams. We shall first investigate the problem of phase diagrams theoretically.

Phase diagrams can be calculated if one knows the free energy of the crystal, which we shall calculate on the basis of the following model. The magnetic properties of the crystal are described by two magnetic subsystems: the weakly ferromagnetic iron sublattice, characterized by the antiferromagnetism vector 1 and the magnetization m, and the paramagnetic system of rare-earth ions; these subsystems are coupled to each other by exchange interaction.

The free energy of the Fe³⁺ ions can be expressed in terms of 1 and m in the usual way, by using considerations of magnetic symmetry (see, for example,^[1,4]). To calculate the free energy of the paramagnetic subsystem it is necessary to know the spectrum and basic characteristics of the rare-earth ions (RI) in the crystalline field (the g tensor, the exchange parameters, etc.). The RI occupy four c sites in the crystal lattice and have two nonequivalent positions. For them, the local symmetry is described by point group C_S (a reflection plane parallel to the plane ab). In such a lowsymmetry environment, the original multiplet undergoes the maximum allowable splitting. Thus the multiplet ${}^{6}H_{15/2}$, corresponding to the ground state of the free Dy³⁺ ion, splits into doublets. According to optical measurements^[2], Dy³⁺ in DyFeO₃ has the following energy levels: $E_0 = 0$; $E_1 = 52.5$ cm⁻¹, $E_2 = 147 \pm 1$ cm⁻¹, $E_3 = 225 \pm 2 \text{ cm}^{-1}$, etc. Hereafter we shall limit ourselves to the case of low temperatures, where it is sufficient to consider only the lowest doublet (T < 77 K for $Dy FeO_3$).

In the presence of an external magnetic field H and of exchange interaction of the RI with the iron ions, the energy of the levels of this doublet can be described in the form

$$E_{o} = \frac{\beta}{2} |\tilde{g}(\mathbf{H} + \mathbf{h})|_{\sigma}, \qquad (1)$$

where $\sigma = \pm 1$, and where \tilde{g} is a tensor defined in the usual way in terms of the wave functions of the doublet^[3], in accordance with the local symmetry of the RI environment; it has the form

$$\tilde{g} = \begin{pmatrix} g_{zx} \pm g_{zy} & 0 \\ \pm g_{zy} & g_{yy} & 0 \\ 0 & 0 & g_{zz} \end{pmatrix}.$$
 (2)

The signs \pm correspond to the two nonequivalent positions of the RI.

We shall treat the exchange interaction in the molecular-field approximation (which in the present situation has a high degree of accuracy). In the same way as earlier^[4], using the magnetic symmetry of the orthoferrites, we shall define the anisotropic molecular field a that acts on the RI in the following form:

$$\mathbf{h}^{\pm} = \begin{pmatrix} \lambda_{1} & 0 & 0 \\ \pm \lambda_{4} & \lambda_{2} & 0 \\ 0 & 0 & \lambda_{3} \end{pmatrix} \mathbf{m} + \begin{pmatrix} 0 & 0 & \lambda_{3} \\ 0 & 0 & \pm \lambda_{6} \\ \lambda_{7} & \pm \lambda_{6} & 0 \end{pmatrix} \mathbf{l} = \hat{\lambda}_{1}^{\pm} \mathbf{m} + \hat{\lambda}_{2}^{\pm} \mathbf{l}.$$
(3)

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If these assumptions are made, the free energy of the crystal (per RFeO $_3$ molecule) can be described in the form

$$F = F_{Fe} + f_R^+ + f_R^-; \tag{4}$$

$$F_{\mathbf{F}_{\bullet}} = \frac{Am^{*}}{2} + a_{1}l_{x}^{2} + a_{2}l_{z}^{2} - d_{1}l_{x}m_{z} - d_{2}l_{z}m_{x} - mH, \qquad (5)$$

$$f_{R}^{\pm} = -\frac{T}{2} \ln 2 \operatorname{ch} \frac{\Delta^{\pm}}{T}.$$
 (6)

We shall neglect anisotropic terms of the type $R_1m_X^2/2$ and $b2m_Z^2/2$ in view of their smallness (m is comparable with $a_1l_X^2$ and $a_2l_Z^2$). The Zeeman splitting Δ^{\pm} of the doublet in the external and exchange field is

$$\Delta^{\pm} = \frac{\beta}{2} [g_{ij}g_{il}(H_j + h_j)(H_l + h_l)]^{\prime_l},$$
 (7)

where summation over repeated indices is understood, and where β is the Bohr magneton.

On minimizing the free energy (4) with respect to m at given 1, under the supplementary conditions (m1) = 0 and $l^2 = 1 - m^2 \approx 1$, we get

$$\mathbf{m} = A^{-1} [\mathbf{H}_{t} - (\mathbf{H}_{t}] \times \mathbf{I}], \qquad (8)$$

where $H_t = H + H_D + H_R$; H_D is the Dzyaloshinskiĭ field, with components $(d_1l_z, 0, d_2l_x)$;

$$\mathbf{H}_{R} = -\frac{\partial f^{+}}{\partial \mathbf{m}} - \frac{\partial f^{-}}{\partial \mathbf{m}} = \frac{1}{2} (\lambda_{1} + \mathbf{M}_{R} + \lambda_{1} - \mathbf{M}_{R})$$
(8a)

is the field exerted on the Fe³⁺ ions by the RI; and M_R^{\pm} is the magnetization of the RI. The values of M_R^{\pm} can be obtained by differentiation of the expression (6) with respect to H,

$$M_{j^{\pm}} = \left(\frac{\beta}{2}\right)^{2} \left(\operatorname{th}\frac{\Delta^{\pm}}{T}\right) \frac{g_{ij^{\pm}}g_{ik^{\pm}}(H+h^{\pm})_{k}}{\Delta^{\pm}},\tag{9}$$

the RI susceptibility is

$$\chi_{jk}^{\pm} = \left(\frac{\beta}{2}\right)^2 \frac{g_{ij}^{\pm}g_{ik}^{\pm}}{T}.$$
 (10)

In order not to complicate the presentation with details and with cumbersome formulas, and remembering that the orientational transitions under study occur in relatively quite strong magnetic fields, when the exchange field gives a smaller contribution than the external field to the Zeeman splitting of the doublet, we shall hereafter consider that M_R^{\pm} is a function only of the external field. On substituting (8) in (4) and taking into account terms of no higher order than the second in m, we get

$$F = -\frac{1}{2A} [\mathbf{H}_{i}^{2} - (\mathbf{H}_{i})^{2}] + \tilde{a}_{i} l_{z}^{2} + \tilde{a}_{z} l_{z}^{2}.$$
(11)

In this form, the free energy is a function of 1 (or of the angles θ and φ , which are defined in the usual way: $l_Z = l \cos \theta$, $l_X = l \sin \theta \cos \varphi$, $l_y = l \sin \theta \sin \varphi$). The expression (11) coincides in form with the free energy of a compensated, completely saturated antiferromagnet in a magnetic field, whose role in our case is played by H_t , the total effective field that acts on the Fe³⁺ ions. It is obvious that, depending on the orientation of the field H_t and on the character of the anisotropy, just as in the case of a compensated antiferromagnet, in the field H_t there can occur orientational transitions of the type of flipping of the magnetic sublattices (spin-flop). The external field here plays a double role. In addition to its direct influence on the iron sublattices, it induces RI magnetization and thus induces ("amplifies") the molecular field exerted on the Fe³⁺ ions by the RI. At

low temperatures, the "induced" molecular field may greatly exceed the external field, so that the flipping actually occurs in the internal molecular field. The induced molecular field depends strongly on temperature, and this leads to peculiar H-T phase diagrams for the system.

We shall consider concrete special cases.

1. $H_0 \parallel b$. According to (9) we have $(M_R^{\pm})_X = \pm M_X$, $(M_R^{\pm})_y = M_y$, $(M_R^{\pm})_z = 0$; substituting these values in (8a), we get $H_{RX} = 0$, $H_{Ry} = \lambda_2 M_y$, $H_{RZ} = 0$. Thus

$$\mathbf{H}_{\iota} = (dl_{\iota}, H_0 + H_R, dl_x). \tag{12}$$

The free energy (11) is

$$F = -\frac{(H_0 + H_R)^2}{2A} (l_x^2 + l_z^2) + K_{bc} l_x^2 + K_{bc} l_x^2 + \dots, \qquad (13)$$

where K_{bc} and K_{ba} are the anisotropy energies that "restrain" the vector 1 against rotation into the planes bc and ba of the crystal. We have omitted terms of higher order in 1 ($K_{bc} \equiv a_2 - d_2^2/2A$, $K_{ba} \equiv a_1 - d_1^2/2A$). Since at a temperature below the Morin point the stable state is $l_X = l_Z = 0$, obviously $K_{bc} > 0$, $K_{ba} > 0$.

The field $H_0 + H_R$ weakens the stability of the state $l_x = l_z = 0$. The critical field at which the mode $G_y(l_x = l_z = 0)$ loss its stability is obviously determined by the equation

$$(H_0 + H_R)_{\rm cr}^2 = 2A \min (K_{bc}, K_{ba}), \qquad (14)$$

or

$$(H_{0}+H_{R})_{\rm cr}^{2} = \begin{cases} 2AK_{bc}; & K_{bc} < K_{ba} \\ 2AK_{ba}; & K_{ba} < K_{bc} \end{cases}$$
(15)

On using the relations (8a), (9), and (10), we get

$$H_{o cr} = \frac{\left[24 \min(K_{bc}, K_{ba})\right]^{\nu_{t}}}{1 + \lambda_{2} \chi_{\nu \nu}^{R}(T)}.$$
 (16)

$$\chi_{\mu\nu}^{R} = \left(\frac{\beta}{2}\right)^{2} \frac{g_{\nu\nu}^{2} + g_{z\nu}^{2}}{T}.$$
 (17)

Possible variants of the phase diagrams for this case are shown in Fig. 1.

The physical mechanism of flipping of the iron sublattices is determined by competition of the anisotropy energy and the total magnetic energy $\chi_{\perp}(H + H_R)^2/2$.

2. $H_0 \parallel a$. According to (9), we have

$$M_{x}^{-}=M_{x}^{+}=\left(\frac{\beta}{2}\right)^{2}\operatorname{th}\frac{\Delta}{T}\frac{g_{xx}^{2}+g_{xy}^{2}}{\Delta}H, \quad \chi_{zx}^{\pm}=\left(\frac{\beta}{2}\right)^{2}\frac{g_{xx}^{2}+g_{xy}^{2}}{T}; \quad (18)$$
$$M_{y}^{\pm}=\left(\frac{\beta}{2}\right)^{2}\operatorname{th}\frac{\Delta}{T}\frac{\pm g_{xy}(g_{zx}+g_{yy})}{\Delta}H,$$
$$\chi_{yy}^{\pm}=\pm\left(\frac{\beta}{2}\right)^{2}\frac{g_{xy}(g_{xx}+g_{yy})}{T}; \quad (19)$$

$$M_{z}^{\pm}=0, \quad H_{Rz}=\lambda_{1}M_{z}+\lambda_{2}M_{y}, \quad H_{Ry}=H_{Rz}=0.$$
 (20)





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$$F = -\frac{1}{2A} [H_0 + (H_R)_x + dl_x]^2 (1 - l_x^2) - \frac{d^2}{2A} l_x^2 (1 - l_x^2) + \tilde{a}_1 l_x^2 + \tilde{a}_2 l_x^2.$$
(21)

We shall investigate the stability of the model G_y ($l_x = l_z = 0$). Formal analysis of (21), carried out under the condition that for $T < T_M$ and $H_0 = 0$ the mode G_y is stable, shows that in a magnetic field there occurs a smooth rotation of the antiferromagnetism vector 1 into the plane bc, with corresponding appearance of a magnetic moment along the x axis,

$$l_z = (H_0 + H_R) m_0 / 2K_{bc}, \quad l_x = 0.$$
 (22)

The fact that the reorientation of 1 occurs in the plane bc follows directly from the form of the free energy (21). In fact, on deflection of the vector 1 in the plane ab the magnetic energy increases by the amount $(H + H_R)^2 l_x^2/2A$, whereas deflection of 1 in the plane bc leads to decrease of the energy. The field at which the reorientation is completed is determined by the relation

$$(H_{\mathfrak{o}}+H_R)_{\mathrm{cr}}=2K_{bc}/m_{\mathfrak{o}}$$

$$(23)$$

or, by use of (18), (19), and (20),

$$H_{0 \text{ cr}} = \frac{2K_{bc}/m_0}{1 + \lambda_1 \chi_{xx} + \lambda_1 \chi_{xy}}.$$
 (24)

The phase diagram is shown in Fig. 2.

3. H₀ || c. According to (9) we have

$$M_z = M_y = 0, \quad M_z = \frac{\beta g_z}{2} \operatorname{th} \frac{\Delta}{T} H, \quad \chi_{zz} = \left(\frac{\beta}{2}\right)^2 \frac{g_{zz}^2}{T}, \quad (25)$$

 $H_{Rx} = H_{Ry} = 0$, $H_{Rz} = \lambda_3 M_z$

Substituting these values in (11), we get

$$F = -\frac{1}{2A} \left[(H_0 + H_R + dl_z)^2 (1 - l_z^2) \right] - \frac{d^2}{2A} l_z^2 (1 - l_z^2) + \tilde{a}_1 l_z^2 + \tilde{a}_2 l_z^2.$$
 (26)

It is easy to demonstrate that in this case the reorientation of the antiferromagnetism vector occurs in the plane ab,

$$l_z=0, \quad l_x=m_0(H_0+H_R)/2K_{ab}$$
 (27)

and that the critical field is determined by the relation

$$H_{0 \, cr} = 2K_{ab}/m_0 (1 + \lambda_s \chi_{zz}). \tag{28}$$

The phase diagram is shown in Fig. 3.

EXPERIMENTAL RESULTS

We have studied transitions from the antiferromagnetic to the weakly ferromagnetic state, in monocrystals of dysprosium orthoferrite, induced by an external magnetic field applied along various crystallographic directions. The investigations were made on monocrystals grown by the method of crucibleless zone fusion with optical heating^[5]. Figure 4 shows magnetization curves taken along the c axis of the rhombic crystal over the







FIG. 4. Isotherms of the magnetization of $DyFeO_3$ along the c axis, with external field H \parallel c, for various temperatures (°K): 1, 4.2; 2, 6.7; 3, 10.9; 4, 14.9; 5, 19.7; 6, 27.7; 7, 34.7; 8, 40.3; 9, 41.7.

FIG. 5. Temperature dependence of thermal expansion along the c axis.



wide temperature interval 4.2 to 100 K. As is evident from Fig. 4, at low temperatures, where the crystal is antiferromagnetic, application of a sufficiently large magnetic field along the c axis causes an abrupt transition to a weakly ferromagnetic state $(G_y \rightarrow G_x F_z)$. The value of the threshold field that produces this transition decreases with increase of temperature and vanishes at the Morin point, above which the crystal is a weak ferromagnet with magnetic moment along the c axis of the crystal $(G_x F_z)$.

Because of the strong influence of a magnetic field on the transition temperature, we determined the Morin point from the temperature dependence of the thermal expansion (Fig. 5), whose behavior exhibited a signifinificant anomaly at the instant of transition. The value thus obtained, $T_M \approx 41$ K, somewhat exceeds the value determined earlier^[7,8], when the Morin point was found from magnetic measurements.

From Fig. 5 it is seen that upon spontaneous transition from the antiferromagnetic to the weakly ferromagnetic state, the length of the crystal along the c axis decreases discontinuously $((\Delta l/l)_c = -1 \cdot 10^{-5})$. An analogous deformation of the crystal can obviously be produced also by application of a magnetic field below the Morin point along the c axis of the crystal, since in this case we also observed the transition $G_y \rightarrow G_X F_Z$. Figure 6a shows data on the change of longitudinal magnetostriction in a field applied along the c axis of the crystal at various temperatures. Above the Morin point, the magnetostriction occurs only because of the paramagnetism of the dysprosium ions, since 180° rotation of the spins of the iron ions in the field does not cause magnetostriction. At temperatures below the Morin point, in addition to the magnetostriction of the rare-earth ions there is magnetostriction due to rotation of the spins of the iron ions from the b axis to the a axis of the crystal under the influence of a magnetic field directed along the c axis of the crystal.

An unusual feature of this reorientational transition in dysprosium orthoferrite is that the rotation of the spins under the influence of a magnetic field parallel to



FIG. 6. Isotherms of the longitudinal magnetostriction along various crystallographic directions in DyFeO₃, for various temperatures (°K): a, $H \parallel c$ (1, 6.1; 2, 7.5; 3, 8.7; 4, 12.9; 5, 17; 6, 21.7; 7, 25.6; 8, 30.5; 9, 34; 10, 42). b, $H \parallel a$ (1, 6.1; 2, 8.2; 3, 10.9; 4, 15.6; 5, 19.7; 6, 23.7; 7, 29.7; 8, 36.1; 9, 42.4; 10, 48). c, $H \parallel b$ (1, 6.1; 2, 8.9; 3, 15.0; 4, 23.1; 5, 34.6; 6, 35.0; 7, 36.2; 8, 38; 9, 40.5; 10, 42).

the c axis occurs here in the plane ab perpendicular to the direction of the magnetic field. The temperature dependences of the threshold field that produces the transition from the antiferromagnetic to the weakly ferromagnetic state, as determined from the magnetization curves and from the magnetostriction curves, agree well with each other. The phase diagram obtained experimentally (Fig. 7) shows an increase of the threshold field with increasing distance from the Morin point. Near the Morin point, the dependence of H_{th} on T is practically linear; this is apparently due to the linear dependence of the anisotropy constant on temperature in this temperature range. According to^[6]

$$H_{\rm th} = -2K(T)/m(T).$$

Assuming that near T_M

$$K(T) = K(T - T_M)/T \tag{29}$$

we obtain from the phase diagram (Fig. 7) the value $K=3.3\cdot10^4 \text{ erg/cm}^3$.

The shift of T_M under the influence of a magnetic field applied along the c axis is of amount $\Delta T_M / \Delta H_{th}$



FIG. 7. Temperature dependence of the threshold field along the c axis that produces the transition $G_V \rightarrow G_X F_Z$.

FIG. 8. Experimental phase diagrams for DyFeO₃ in an external magnetic field (1, H || a; 2, H || b).

= 12 deg/kOe. Far from T_M , in the low-temperature region, $H_{th}(T)$ departs from a linear dependence, and there develops a rapid rise with lowering of temperature ($H_{th} = 7 \times 10^3$ Oe at 4.2 K).

As was pointed out above, the magnetic behavior of dysprosium orthoferrite is significantly influenced by exchange interaction of the rare-earth and iron ions. Above the Morin point, the presence of positive exchange interaction leads to a hyperbolic rise of the spontaneous magnetic moment along the c axis of the rhombic crystal with lowering of temperature.

The total spontaneous magnetic moment of the orthoferrite in this case can be described in the form [1]

$$\sigma_0 = \sigma_{Fe} + \chi_R H_0, \qquad (30)$$

where σ_{Fe} is the weakly ferromagnetic moment of the iron ions, and where H_0 is the exchange field that polarizes the paramagnetic rare-earth ions with susceptibility χR . For dysprosium orthoferrite, the value found by extrapolation to high temperatures is $\sigma_{Fe} = 1.5 \text{ G cm}^3/\text{g}$, which is close to the value for yttrium orthoferrite (the yttrium ions are nonmagnetic). The exchange field exerted by the iron ions on the dysprosium ions, above T_M , then had the value $H_0 = 5 \cdot 10^3 \text{ Oe}$; this somewhat exceeds that obtained in the paper of White^[9], because there a too low value of σ_{Fe} was taken.

Below the Morin point, the exchange field also acts along the c axis of the crystal; this promotes antiferromagnetic ordering of the dysprosium ions, of type C_Z , which however is not observed because of the small value of the g_Z factor. Exchange interaction between the rare-earth and iron ions can also lead, in turn, to an action of the rare-earth ions on the spins of the iron ions. The effect of this interaction should show up especially strongly along the a and b axes of the crystal, where the susceptibility of the Dy³⁺ ions is large.

As was considered in Sec. 1, the exchange field exerted by the rare-earth ions produces a potentiality for occurrence of a spin reorientation $(G_y \rightarrow G_x F_z)$ or $G_y \rightarrow G_z F_x)$ on application of a magnetic field along the a and b axes of the crystal. This type of reorientation, induced by an external magnetic field, is difficult to detect from magnetization curves, because the spontaneous magnetic moment is small and may appear along the c axis, perpendicular to the measurement direction. For exposure of a possible reorientational transition in a

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field, we carried out measurements of the longitudinal magnetostriction along the a and b axes of the crystal (Fig. 6b and c). It is evident that below T_M , in a sufficiently large field parallel to the a and b axes, there occurs an additional magnetostriction, apparently caused by reorientation of the spins of the iron ions, since the dysprosium ions at these temperatures are in a paramagnetic state. The value of the field at which a break in the magnetostriction curve is observed was taken as the threshold field that produces the transition at the given temperature.

The value of the threshold fields determined from the isotherms of longitudinal magnetostriction along the a axis decrease, with lowering of the temperature, in proportion to the distance from the Morin point. The experimental phase diagram for this case is shown in Fig. 8. In analogous fashion, an experimental phase diagram was constructed on the basis of values of the threshold fields determined from the isotherms of longitudinal magnetostriction along the b axis (Fig. 8). It is evident that in the case of a field $H \parallel b$ the dependence of the threshold field on temperature has a more complicated character. Comparison of the experimental phase diagrams with the theoretical curves shows good qualitative agreement. As is evident from Fig. 8, the threshold field along the a axis is larger than along the b axis; this is obviously due to the large value of the g-factor along the b axis, since, according to (16), the value of the threshold field is inversely proportional to the value of the susceptibility along the corresponding axes of the crystal.

It should be noted that reorientational transitions of this type, induced by an external magnetic field, have been observed here for the first time in orthoferrites. A specific peculiarity of the transitions in the case $H \parallel a$ and $H \parallel b$ is that the reorientation occurs under the influence of the molecular field of the rare-earth ions, amplified by the action of the external field. A distinctive trait of the transition in a field parallel to the c axis is that the reorientation of the spins occurs in a plane perpendicular to the direction of the external field; this also has not been observed previously in orthoferrites.

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