

Coupled oscillations of iron rare-earth and elastic subsystems in orthoferrites with Kramers rare-earth ions

V. D. Buchel'nikov, I. V. Bychkov, and V. G. Shavrov

Institute of Radioengineering and Electronics, Russian Academy of Sciences

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A theory is derived for coupled oscillations of the iron, rare-earth, and elastic subsystems near spontaneous orientational phase transitions in rare-earth orthoferrites with Kramers rare-earth ions. The effect of the rare-earth subsystem on the spectrum of magnetoelastic oscillations is of fundamental importance. Expressions are derived for the activation of soft quasispin wave modes and for the velocities of transverse elastic waves near orientational phase transitions. The results are compared with the experimental results available. A theoretical explanation is offered for the anomalously large decrease in transverse sound velocity which was recently observed experimentally near a low-temperature spin reorientation in erbium orthoferrite.

1. INTRODUCTION

There is an extensive literature on the experimental and theoretical study of coupled oscillations in rare-earth orthoferrites (REOs) (see, for example, Refs. 1–7 and the bibliography cited there). The gapwidths of soft modes at points of different orientational phase transitions (OPTs) were determined and the coupled oscillations of the iron (d) and rare-earth (f) subsystems in REOs with non-Kramers^{4,5} and Kramers^{6,7} rare-earth ions (with even and odd number of $4f$ electrons, respectively) were investigated. In Refs. 8–14 changes in the sound velocity, attenuation, and Young's modulus were observed in many REOs near OPTs. Quantitative estimates of the frequencies of coupled oscillations and magnetoelastic constants were made on the basis of the theories developed in these works. A systematic theory of coupled oscillations of the iron and elastic subsystems (magnetoelastic waves) in REOs was proposed in Ref. 15. The theoretical results obtained there nonetheless cannot explain the experimental data completely. In particular, in experiments¹¹ near high-temperature OPTs the maximum change in the sound velocity is only $\sim 3\%$. According to theory, however, in the ideal case the sound velocity can change by 100%. The activation frequency of the soft modes at the points of high-temperature OPTs studied in Ref. 15 also does not agree with experiment. In addition, a new result was recently obtained experimentally in erbium REO near a low-temperature OPT which was not studied in Ref. 15: an anomalously large decrease (up to 25%) of the velocity of transverse sound was observed.^{13,14} All this indicates that the theory proposed in Ref. 15 requires further development.

It is well-known that the rare-earth subsystem plays an important role in static and dynamic properties of REOs.^{1,2} The effect of the f ions on the magnetic properties of REOs is significant even in the case when the f subsystem is in the paramagnetic state (in the effective field of the d subsystem). The spin-wave spectrum in REOs with non-Kramers f ions was investigated theoretically and experimentally in Refs. 4 and 5. It was shown that the spectrum consists of four branches, two of which describe oscillations of the d subsystem while the other two describe oscillations of the f subsystem. The interaction of the d and f modes is strongest near the points where the branches intersect. This interaction re-

sults in the fact that near an OPT the soft mode can become both a d and an f mode.

The problem of the effect of the f ions on the spectrum of magnetoelastic waves in REOs is of interest in connection with the importance of the f subsystem in the formation of the static and magnetic properties of these magnetic materials. In the present paper we investigate theoretically the spectrum of coupled oscillations of the elastic, dipole, and d and f subsystems near OPTs in REOs with Kramers f ions: ErFeO_3 and YbFeO_3 .

As an example we investigate $\Gamma_4-\Gamma_{24}$, $\Gamma_2-\Gamma_{24}$, and $\Gamma_2-\Gamma_{12}$ OPTs. All of these transitions occur in erbium REO at $T_1 \approx 100$ K, $T_2 \approx 90$ K, and $T_3 \approx 4$ K, respectively.^{9,13,14} In ytterbium REO the first two transitions occur at $T_1 \approx 8$ K and $T_2 \approx 7$ K, respectively.¹² The theoretical results obtained are compared with existing experimental data for these REOs.^{9,10,12-14}

2. ENERGY OF ORTHOFERRITE

We write the free energy density of the REO in the form^{1,2,4,5,15}

$$\mathcal{F} = \mathcal{F}_M + \mathcal{F}_{ME} + \mathcal{F}_E. \quad (1)$$

The energy density of the magnetic subsystem is given by

$$\mathcal{F}_M = \mathcal{F}_d + \mathcal{F}_{fd} + \mathcal{F}_f, \quad (2)$$

where

$$\begin{aligned} \mathcal{F}_d = & \frac{1}{2} A F^2 + \frac{1}{2} D (F G)^2 - d (F_x G_z - F_z G_x) - 2 M_0 F H \\ & + \frac{1}{2} \alpha \left(\frac{\partial G}{\partial x_i} \right)^2 + \frac{1}{2} K_{ac} G_z^2 + \frac{1}{2} K_{ab} G_y^2 + \frac{1}{4} K_{20} G_z^4 \\ & + \frac{1}{4} K_{20}' G_y^4 + \frac{1}{2} K_{20}'' G_y^2 G_z^2. \end{aligned}$$

$$\begin{aligned} \mathcal{F}_{fd} = & -N \{ f_x [\mu_x (H_x + a F_x) + B_z' G_z] \\ & + f_y \mu_y (H_y + a F_y) + f_z [\mu_z (H_z + a F_z) \\ & + B_x G_x] + c_x \mu_{xy} (H_y + a F_y) + c_y [\mu_{yx} (H_x + a F_x) + B_z'' G_z] + c_z B_y G_y \}, \\ \mathcal{F}_f = & -\frac{1}{2} N (\lambda_1 f_x^2 + \lambda_2 f_y^2 + \lambda_3 f_z^2 + \lambda_4 c_x^2 + \lambda_5 c_y^2 + \lambda_6 c_z^2 + 2 \lambda_7 f_x c_y \\ & + 2 \lambda_8 f_y c_x) - \frac{1}{2} N T [S(\sigma_1) + S(\sigma_2)], \end{aligned}$$

and A, D, α, d , and K are, respectively, the exchange, Dzyaloshinskii, and anisotropy constants in the d subsystem; \mathbf{H} is the magnetic field; N is the number of ions per cm^3 ; $\mu_\alpha = \mu_{\alpha\alpha}, \mu_{\alpha\beta} = \mu_B \mathbf{g}_{\alpha\beta}/2, \mu_B$ is the Bohr magneton, \hat{g} is the g tensor, a and B are the isotropic and anisotropic f - d exchange interaction constants; λ is the interaction constant in the f subsystem; and S is the entropy of the f subsystem

$$S(\sigma) = \ln 2 - \frac{1}{2}(1+\sigma) \ln(1+\sigma) - \frac{1}{2}(1-\sigma) \ln(1-\sigma), \quad \sigma_{1,2} = |\mathbf{f} \pm \mathbf{c}|.$$

The energy (2) is written in the approximation of two d and f sublattices. The vectors $\mathbf{F}, \mathbf{G}, \mathbf{f}$ and \mathbf{c} have the form

$$\mathbf{F} = (M_1 + M_2)/2M_0, \quad \mathbf{G} = (M_1 - M_2)/2M_0, \\ \mathbf{f} = (\sigma_1 + \sigma_2)/2, \quad \mathbf{c} = (\sigma_1 - \sigma_2)/2,$$

where M_i are the magnetizations d of the sublattices, $M_0 = |M_1| = |M_2| = \mu_d N, \mu_d = 5\mu_B$, and $\sigma_{1,2}$ are the average Pauli matrices of the sublattices of the f ions. We assume that the sublattices of the d subsystem are saturated ($D \rightarrow \infty$). In this case the vectors \mathbf{F} and \mathbf{G} satisfy the additional conditions

$$\mathbf{F}^2 + \mathbf{G}^2 = 1, \quad \mathbf{F}\mathbf{G} = 0. \quad (3)$$

In the expression for the magnetoelastic interaction energy we include only the interaction energy of the elastic and d subsystems

$$\mathcal{F}_{ME} = 2(B_{11}u_{xx} + B_{12}u_{yy} + B_{13}u_{zz})G_x^2 + 2(B_{21}u_{xx} + B_{22}u_{yy} + B_{23}u_{zz})G_y^2 + 2(B_{31}u_{xx} + B_{32}u_{yy} + B_{33}u_{zz})G_z^2 + 2B_{14}u_{yz}G_yG_z + 2B_{35}u_{xz}G_xG_z + 2B_{66}u_{xy}G_xG_y,$$

where \hat{u} is the strain tensor and \hat{B} is the tensor of magnetoelastic constants. We write the energy density of the elastic subsystem in the standard form:

$$\mathcal{F}_E = \frac{1}{2}\rho\hat{u}^2 + \frac{1}{2}(c_{11}u_{xx}^2 + c_{22}u_{yy}^2 + c_{33}u_{zz}^2) + c_{12}u_{xx}u_{yy} + c_{13}u_{xx}u_{zz} + c_{23}u_{yy}u_{zz} + 2c_{14}u_{yz}^2 + 2c_{55}u_{xz}^2 + 2c_{66}u_{xy}^2,$$

where \hat{c} is the tensor of elastic constants and ρ is the density of the material.

3. GROUND STATE

We now examine the case when the reorientation of the vectors \mathbf{G} and \mathbf{F} occurs in the xz plane and $H = 0$. We find the equilibrium values of the parameters of the subsystems in the phases Γ_2, Γ_4 , and Γ_{24} by minimizing the energy (1) with respect to $\mathbf{F}, \mathbf{G}, \mathbf{f}, \mathbf{c}$, and u_{ij} under the conditions (3). The results are as follows:

$$F_y = f_y = G_y = c_z = c_x = u_{xy} = u_{yz} = 0; \quad F_x = F_0 G_x, \quad F_z = -F_0 G_x; \quad (4) \\ f_z = (a\mu_z F_z + B_x G_x)/\lambda_3', \quad f_x = [(a\mu_x F_x + B_z' G_z)\lambda_7' + (a\mu_{yx} F_x + B_z'' G_z)\lambda_7'] / (\lambda_1' \lambda_5' - \lambda_7'^2), \quad c_y = [(a\mu_{yx} F_x + B_z'' G_z)\lambda_1' + (a\mu_x F_x + B_z' G_z)\lambda_7'] / (\lambda_1' \lambda_5' - \lambda_7'^2); \\ u_{\alpha\alpha} = \Delta_\alpha / \Delta, \quad u_{xz} = -B_{56} G_x G_z / 2c_{55}.$$

Here

$$F_0 \approx \frac{1}{A} [d + Na(\mu_x f_x + \mu_{yx} c_y) G_z - Na\mu_z f_z G_x], \quad (4a)$$

$$\lambda_i' = -\lambda_i + T \text{Arth } \sigma / \sigma,$$

$$\sigma = (f_x^2 + f_z^2 + c_y^2)^{1/2};$$

$$\Delta = c_{11} C_{2233}^2 + c_{12} C_{1323}^2 + c_{13} C_{1232}^2,$$

$$\Delta_\alpha = \frac{1}{2} E_\alpha C_{\beta\beta\gamma\gamma}^2 + E_\beta C_{\alpha\gamma\beta\gamma}^2 (\alpha \neq \beta, \beta \neq \gamma, \alpha \neq \gamma),$$

$$E_\alpha = -2B_{\mu\alpha} G_\mu^2, \quad C_{\alpha\beta\gamma\delta} = c_{\alpha\beta} c_{\gamma\delta} - c_{\alpha\gamma} c_{\beta\delta} \quad (4b)$$

$$(\alpha, \beta, \gamma, \delta, \mu = 1, 2, 3).$$

The relations (4), together with the relations (4a), are actually equations for determining σ, f_x, f_z , and c_y . For $\sigma \ll 1$ these equations become identities. The antiferromagnetism vector \mathbf{G} of the d subsystem in the collinear phase Γ_4 has the components $G_x = \pm 1$ and $G_z = 0$, while in the collinear phase Γ_2 the components are $G_x = 0$ and $G_z = \pm 1$. Here and below we employ the approximation $G^2 = 1 - F^2 \approx 1$. In the canted phase Γ_{24} the equilibrium values of the components of the vector \mathbf{G} are determined by the formulas

$$G_x^2 = 1 - G_z^2, \quad G_z^2 = -K_{ac}/K_2, \quad (5)$$

where K_{ac} and K_2 are the effective anisotropy constants

$$K_{ac} = K_{ac}^0 - \frac{8}{\Delta} [B_{11}(B_{11} - B_{31})C_{2233}^2 + B_{12}(B_{12} - B_{32})C_{1133}^2 + B_{13}(B_{13} - B_{33})C_{1122}^2 + (2B_{11}B_{13} - B_{11}B_{32} - B_{12}B_{31})C_{1323}^2 + (2B_{11}B_{23} - B_{11}B_{33} - B_{13}B_{31})C_{1232}^2 + (2B_{12}B_{13} - B_{12}B_{33} - B_{13}B_{32})C_{1213}^2 - B_{55}^2/c_{55} + NB_x^2/\lambda_3' - N(B_z''\lambda_5' + 2B_z' B_z''\lambda_7 + B_z''\lambda_7') / (\lambda_1' \lambda_5' - \lambda_7'^2)], \quad (6)$$

$$K_2 = K_{20} - \frac{8}{\Delta} [(B_{11} - B_{31})^2 C_{2233}^2 + (B_{11} - B_{32})^2 C_{1133}^2 + (B_{13} - B_{33})^2 C_{1122}^2 + 2(B_{11} - B_{31})(B_{12} - B_{32})C_{1323}^2 + 2(B_{12} - B_{32})(B_{13} - B_{33})C_{1232}^2 + 2(B_{11} - B_{31})(B_{13} - B_{33})C_{1213}^2] + 2B_{55}^2/c_{55} \quad (7)$$

We underscore the fact that the relations (5) are also actual equations for determining G_x and G_z , since the quantities λ_i' appearing in Eq. (6) depend on G_x and G_z . For $\sigma \ll 1$ the equations (5) become identities.

We now formulate the conditions under which these phases are stable.¹⁵ We examine the case $K_2 > 0$. For $T > T_1$, when $K_{ac} > 0$, the Γ_4 phase is stable. As the temperature decreases, at $T = T_1 (K_{ac}(T_1) = 0)$ the constant K_{ac} changes sign and the Γ_4 phase becomes unstable. A second-order OPT occurs: the system transforms into the canted phase Γ_{24} , in which the orientation of the vector \mathbf{G} is determined by Eq. (5). Reorientation terminates with a second-order OPT at $T = T_2 (K_{ac}(T_2) + K_2(T_2) = 0)$, when the system transforms into the phase Γ_2 .

In the REO ErFeO₃ another second-order $\Gamma_2-\Gamma_{12}$ OPT occurs as the temperature is further lowered.¹⁴ This transition occurs at $T = T_3$, when $K_{cb}(T_3) = 0$, where

$$K_{cb} = K_{ab}^0 + K_{20}'' - K_{ac}^0 + 4(B_{21} - B_{31})u_{xx}^0 + 4(B_{22} - B_{32})u_{yy}^0 + 4(B_{23} - B_{33})u_{zz}^0 - K_{20} - B_{44}^2/c_{44} - NB_y^2/\lambda_6' + N(B_z'^2\lambda_5' - 2B_z'B_z''\lambda_1' + B_z''^2\lambda_1')/(\lambda_1'\lambda_5' - \lambda_7'^2). \quad (8)$$

We note that the formulas (5)–(8) are presented in the approximations $K, B, d \ll A$, and $a = 0$.

4. EQUATIONS OF MOTION

In order to describe the dynamic properties of the f subsystem we start from the Landau–Lifshitz equations (the applicability of these equations for the f subsystem is justified in Refs. 4 and 5)

$$M_B \dot{\mathbf{f}}/g = -[\mathbf{f}, \vec{\mathcal{F}}_f] - [c, \vec{\mathcal{F}}_c] + M_B \mathbf{R}_f/g, \quad (9)$$

$$M_B \dot{\mathbf{c}}/g = -[\mathbf{f}, \vec{\mathcal{F}}_c] - [c, \vec{\mathcal{F}}_c] + M_B \mathbf{R}_c/g,$$

where $g = 2\mu_B$, $M_B = \mu_B N$, $\vec{\mathcal{F}}_f = \partial \mathcal{F} / \partial \mathbf{f}$, and \mathbf{R} are relaxational terms:⁵

$$\mathbf{R}_f = \Lambda_f \{[\mathbf{f}, \dot{\mathbf{f}}] + [c, \dot{c}]\}, \quad (10)$$

$$\mathbf{R}_c = \Lambda_f \{[\mathbf{f}, \dot{c}] + [c, \dot{\mathbf{f}}]\},$$

and Λ_f is the dissipation constant for the f subsystem. We note that the choice of the relaxational term in the form (10) permits imposing on the vectors \mathbf{f} and \mathbf{c} the additional conditions

$$\mathbf{f}^2 + \mathbf{c}^2 = 1, \quad \mathbf{f}c = 0, \quad (11)$$

which are analogous to the conditions (3) for the vectors \mathbf{F} and \mathbf{G} . The relations (3) and (11) automatically eliminate from analysis the longitudinal oscillations of the f and d subsystems.

The dynamics of the d subsystem is described by Eqs. (9)–(11), in which M_B , \mathbf{f} , \mathbf{c} , and Λ_f should be replaced by, respectively, M_0 , \mathbf{F} , \mathbf{G} , and Λ_d , where Λ_d is the dissipation parameter of the d subsystem. In addition, the sign of the terms on the right-hand sides of Eqs. (9) should be changed.

We now investigate the dynamical properties of the elastic subsystem in the standard manner with the help of the equations of motion for the displacements

$$\rho \ddot{u}_i = \partial \sigma_{ik} / \partial x_k, \quad (12)$$

where $\sigma_{ik} = \partial \mathcal{F} / \partial u_{ik}$ is the elastic-stress tensor. Dissipation in the elastic subsystem is small and for this reason we neglect it.

When spin and elastic waves are excited by electromagnetic waves Maxwell's equations must be added to the system of equations (9)–(12)

$$\text{rot } \mathbf{E} = -\frac{1}{v} \frac{\partial}{\partial t} (\mathbf{H} + 4\pi \mathbf{M}), \quad \text{rot } \mathbf{H} = \frac{1}{v} \varepsilon \frac{\partial \mathbf{E}}{\partial t}, \quad (13)$$

$$\text{div} (\mathbf{H} + 4\pi \mathbf{M}) = 0, \quad \text{div } \mathbf{E} = 0.$$

Here \mathbf{E} and \mathbf{H} are, respectively, the electric and magnetic field strengths, v is the velocity of light in free space, and ε is the permittivity of the REO (it is assumed that at the frequencies of interest the permittivity tensor satisfies

$\varepsilon_{ik} = \varepsilon \delta_{ik}$ and there is no electric conduction), \mathbf{M} is the total magnetization of the REO $\mathbf{M} = \mathbf{M}_d + \mathbf{M}_f$, $\mathbf{M}_d = 2M_0 \mathbf{F}$, $\mathbf{M}_f = N(\mu_x f_x + \mu_{yx} c_y + \mu_y f_y + \mu_{xy} \mu_z f_z)$.

The equations presented above completely describe coupled oscillations of the rare-earth, iron, and elastic subsystems of the REO.

5. DISPERSION EQUATIONS

In order to obtain the dispersion equations of coupled oscillations the equations of motion presented in Sec. 3 must be linearized near the equilibrium position (4). Depending on the magnetic phase under study, the dispersion equations for harmonic waves propagating along the z -axis have the following form:

1) Γ_4 phase. Modes of symmetry Γ_{23} :

$$\begin{aligned} &(\omega^2 - \omega_{5k}^2)(\omega^2 - \tilde{\omega}_{2k}^2)(\omega^2 - \tilde{\omega}_{1f}^2)(\omega^2 - \tilde{\omega}_{2f}^2) \\ &- \tilde{\omega}_E \omega_{me5} \omega_{5k}^2 (\omega^2 - \tilde{\omega}_{1f}^2)(\omega^2 - \tilde{\omega}_{2f}^2) \\ &- \omega^2 \tilde{\omega}_E \tilde{\omega}_{1fd}^3 (\omega^2 - \omega_{5k}^2) - \tilde{\omega}_E \tilde{\omega}_{2fd} \tilde{\omega}_{1f}^2 \tilde{\omega}_{2f}^2 (\omega^2 - \omega_{5k}^2) = 0. \end{aligned} \quad (14)$$

Here

$$\begin{aligned} \omega_{5k} &= s_5 k, \quad s_5 = (c_{55}/\rho)^{1/2}, \\ \tilde{\omega}_{2k}^2 &= \tilde{\omega}_E \tilde{\omega}_{ac}, \quad \tilde{\omega}_{E,ac} = \omega_{E,ac} - i\omega \Lambda_d, \\ \omega_{me5} &= g B_{55}^2 / M_0 c_{55}, \quad \omega_E = g A / M_0, \\ \omega_{ac} &= g K_{ac} / M_0 + \omega_{me5} + \omega_{2fd} + g \alpha k^2 / M_0 \\ &+ \omega_{d1p} / (1 - v^2 k^2 / \varepsilon \omega^2), \quad \omega_{d1p} = 16\pi g M_0 F_0^2, \\ \tilde{\omega}_{1f,2f}^2 &= \frac{g^2 N^2}{2M_B^2} f_z^2 (\tilde{\lambda}_1 \tilde{\lambda}_2 + \tilde{\lambda}_1 \tilde{\lambda}_5 \\ &- 2\tilde{\lambda}_7 \tilde{\lambda}_8 \pm [(\tilde{\lambda}_1 \tilde{\lambda}_2 + \tilde{\lambda}_1 \tilde{\lambda}_5 - 2\tilde{\lambda}_7 \tilde{\lambda}_8)^2 - 4(\tilde{\lambda}_1 \tilde{\lambda}_5 - \tilde{\lambda}_7'^2)(\tilde{\lambda}_2 \tilde{\lambda}_1 - \tilde{\lambda}_8'^2)]^{1/2}), \end{aligned} \quad (15)$$

$$\tilde{\omega}_{1fd}^3 = \frac{g^3 N^3}{M_0 M_B^2} f_z^2 (B_z'^2 \tilde{\lambda}_2 + 2B_z' B_z'' \tilde{\lambda}_8 + B_z''^2 \tilde{\lambda}_1),$$

$$\tilde{\omega}_{2fd} = \frac{gN}{M_0} (B_z'^2 \tilde{\lambda}_5 + 2B_z' B_z'' \tilde{\lambda}_7$$

$$+ B_z''^2 \tilde{\lambda}_1) / (\tilde{\lambda}_1 \tilde{\lambda}_5 - \tilde{\lambda}_7'^2),$$

$$\tilde{\lambda}_i = \lambda_i' - i\omega M_B \Lambda_f / gN + 4\pi N \mu_i^2 / (1 - v^2 k^2 / \varepsilon \omega^2),$$

$$i = 1, 2, 4, 5,$$

$$\tilde{\lambda}_{7,8} = \lambda_{7,8} - 4\pi N \mu_{1,2} \mu_{5,4} / (1 - v^2 k^2 / \varepsilon \omega^2),$$

$$\mu_1 = \mu_x, \quad \mu_2 = \mu_y, \quad \mu_4 = \mu_{xy}, \quad \mu_5 = \mu_{yx}.$$

We note that here and below we employ the approximation in which the constant A is larger than all other constants in Eq. (1), i.e., the frequency ω_E is much higher than all other frequencies appearing in the dispersion equations of the coupled oscillations. We also confine our attention to the case $a = 0$, since the terms containing a enter into the equations with the small factor F_0 .

We present the solution of the dispersion equation (14) for the quasispin and quasiaoustic branches for small wave numbers k (long-wavelength approximation) vk/ε , $\omega_{5k} \ll \omega_{20}$, $\omega_{1f,2f}$. For simplicity we assume that $\tilde{\lambda}_7 = \tilde{\lambda}_8 = B_z' = 0$. In this case one of the rare-earth modes (ω_{1f}) does not interact with the d and elastic modes:

$$\begin{aligned} \omega_f^2 &= \begin{cases} \omega_{2k}^2 + \omega_{2f}^2 \omega_E \omega_{2fd} / \omega_{2k}^2 + \omega_E \omega_{me5} \omega_{5k}^2 / \omega_{2k}^2, & \omega_{2k} > \omega_{2f}; \\ \omega_{2k}^2 - \omega_E \omega_{2fd} + \omega_E \omega_{me5} \omega_{5k}^2 / \omega_{2k}^2, & \omega_{2k} < \omega_{2f}; \end{cases} \\ \omega_{11}^2 &= \begin{cases} \omega_{2f}^2 - \omega_E \omega_{2fd} \omega_{2f}^2 / \omega_{2k}^2, & \omega_{2k} > \omega_{2f}; \\ \omega_{2f}^2 + \omega_E \omega_{2fd}, & \omega_{2k} < \omega_{2f}; \end{cases} \\ \omega_{111}^2 &= \omega_{5k}^2 [1 - \omega_E \omega_{me5} / (\omega_{2k}^2 - \omega_E \omega_{2fd})]. \end{aligned} \quad (16)$$

We also present the damping coefficient $\gamma = \text{Im } k(\omega)$ (Ref. 15) of the quasielastic wave ω_{III} :

$$\gamma_{\text{III}} = \frac{\omega^2 \omega_{\text{me5}} \omega_{\text{E}}^2 (\Lambda_d + \Lambda_f \omega_{2fd} / \omega_f')}{s_5 (\omega_{2k}^2 - \omega_{\text{E}} \omega_{2fd})^{1/2} (\omega_{2k}^2 - \omega_{\text{E}} \omega_{2fd} - \omega_{\text{E}} \omega_{\text{me5}})^{1/2}}, \quad (17)$$

where $\omega_f' = gN\lambda_5 / M_B$.

We note that when writing the frequencies of the quasispin branches $\omega_{1,\text{II}}$, here and below, we must set the cofactor $(1 - v^2 k^2 / \epsilon \omega^2)^{-1}$ in the formula (15) equal to unity, while in the expression for the frequency of the quasielastic branch ω_{III} and for its damping constant γ_{III} it must be set equal to zero.

2) Γ_2 -phase. Symmetry modes Γ_{12} (a) and Γ_{34} (b):

$$\begin{aligned} & (\omega^2 - \omega_{4k,5k}^2) (\omega^2 - \bar{\omega}_{1k,2k}^2) (\omega^2 - \bar{\omega}_{1f,2f}^2) \\ & - \bar{\omega}_{\text{E}} \omega_{\text{me4}, \text{me5}} \omega_{4k,5k}^2 (\omega^2 - \bar{\omega}_{1f,2f}^2) \\ & - \bar{\omega}_{\text{E}} \bar{\omega}_{1fd,2fd} \bar{\omega}_{1f,2f}^2 (\omega^2 - \omega_{4k,5k}^2) = 0. \end{aligned} \quad (18a,b)$$

Here

$$\begin{aligned} \omega_{4k} &= s_4 k, \quad s_4 = (c_{44} / \rho)^{1/2}, \quad \bar{\omega}_{1k,2k}^2 = \bar{\omega}_{\text{E}} \bar{\omega}_{cb,ca}, \\ \omega_{cb} &= gK_{cb} / M_0 + \omega_{\text{me4}} + \omega_{1fd} \\ & + g\alpha k^2 / M_0, \quad \omega_{\text{me4}} = gB_{44} / M_0 c_{44}, \\ \omega_{ca} &= -g(K_{ac} + K_2) / M_0 + \omega_{\text{me5}} + \omega_{2fd} + \omega_{dip} \\ & + g\alpha k^2 / M_0, \quad \bar{\omega}_{1fd,2fd} = gNB_{y,x}^2 / M_0 \bar{\lambda}_{6,3}, \\ \bar{\omega}_{1f,2f}^2 &= \frac{g^2 N^2}{M_B^2} \bar{\lambda}_{6,3} |\lambda'_{1,4} c_y|^2 \\ & + \bar{\lambda}_{5,2f,x}^2 + 2\lambda_{7,8} c_y f_x + 4\pi N (\mu_{1,4} c_y - \mu_{5,2f,x})^2 / (1 - v^2 k^2 / \epsilon \omega^2), \\ \bar{\lambda}_i &= \lambda_i' - i\omega M_B \Lambda_f / gN + 4\pi N \mu_z^2 \delta_{is}, \quad i=2, 3, 5, 6. \end{aligned} \quad (19)$$

The solution of the dispersion equations (18) for the quasispin and quasiaoustic branches in the long-wavelength approximation is determined by the formulas (16) and (17), in which for Eq. (18a) $\omega_f' = gN\lambda_6 / M_B$ and the indices 2 and 5 must be replaced by 1 and 4, respectively, while for Eq. (18b) $\omega_f' = gN(\lambda_3' + 4\pi N \mu_z^2) / M_B$.

6. DISCUSSION

We now analyze the behavior of the coupled oscillations near the OPTs of interest.

1) Γ_4 - Γ_{24} phase transition

It follows from Eq. (14) that in the Γ_4 phase the transverse elastic d - (quasiferromagnetic), and two f -branches interact with one another. The frequency ω_{III} (16), corresponding to the transverse elastic branch of the oscillations (polarized along the x -axis), at the phase transition point itself $K_{ac} = 0$ depends quadratically on k in the limit $k \rightarrow 0$:

$$\omega_{\text{III}} = s_5 (g\alpha / M_0 \omega_{\text{me5}})^{1/2} k^2, \quad (20)$$

while the velocity ω_{III} / k of this mode depends linearly on k and approaches zero as $k \rightarrow 0$. Thus the rare-earth subsystem does not affect the quadratic dispersion law of quasielastic waves near an OPT. The other branches at $k = 0$ have an activation frequency. In the case $\omega_{20} > \omega_{2f}$ the activation frequency of the quasi-iron branch ω_1 at the point of the OPT is determined by the magnetoelastic interaction (magnetoelas-

tic gap), the interaction of the d and f -subsystems, and the dipole interaction:

$$\omega_1^2(0) = \omega_{\text{E}} (\omega_{\text{me5}} + \omega_{2fd} + \omega_{dip}), \quad (21a)$$

while in the case $\omega_{20} < \omega_{2f}$ it is determined only by the magnetoelastic and dipole interactions:

$$\omega_1^2(0) = \omega_{\text{E}} (\omega_{\text{me5}} + \omega_{dip}). \quad (21b)$$

The activation frequency of the quasi-rare-earth mode at the point of the OPT is determined in the first case by the magnetoelastic and dipole interactions as well as by the interactions within the f -subsystem and between the d - and f -subsystems:

$$\omega_{\text{II}}^2(0) = (\omega_{2f} / \omega_{20})^2 \omega_{\text{E}} (\omega_{\text{me5}} + \omega_{dip}), \quad (21c)$$

and in the second case the activation frequency is determined by the interactions within the f -subsystem, the coupling of the d - and f -subsystems, and the dipole interaction

$$\omega_{\text{II}}^2(0) = \omega_{2f}^2 + \omega_{\text{E}} \omega_{2fd}. \quad (21d)$$

The damping coefficients Λ_d and Λ_f at high temperatures ($T \approx 100$ K) are equal, in order of magnitude, to $\Lambda_d \sim 10^{-4}$ and $\Lambda_f \sim 0.1-1$.^{4,5} As is well known,^{4,5} however, at low temperatures the damping coefficients of spin waves in the d - and f -subsystems can decrease by at least an order of magnitude. At high temperatures such a large value of the damping coefficient in the f -subsystem could imply that when the d and f -modes are excited by a high-frequency electromagnetic field it will be impossible to determine the activation frequency of these branches because the two absorption lines merge into one line. We note that the damping coefficient of sound ultimately is also determined by the damping coefficient of spin waves in the f sub-system (17). The strong sound absorption at high temperatures should result in the absence of echo signals near an OPT. Both effects (impossibility of determining the activation frequency of quasispin waves and the large damping of sound) have indeed been observed experimentally.^{11,18}

Using the data of Refs. 2, 9, 10, and 16-19 we now estimate the frequencies appearing in Eqs. (14)-(16) for erbium REO $M_0 \approx 830$ Oe, $\rho \approx 8$ g/cm³, $A \approx 9 \cdot 10^9$ erg/cm³, $B_x \approx 0.6$ K, $B_y \approx 1.3$ K, $B_z' \approx 2.4$ K (at $B_z'' = 0$), $|\lambda_i| \sim 3.5$ K, $B_{55} \approx 2 \cdot 10^6$ erg/cm³, $B_{44} \approx 4 \cdot 10^6$ erg/cm³, $c_{55} \approx 8.9 \cdot 10^{11}$ erg/cm³, $c_{44} \approx 1.2 \cdot 10^{12}$ erg/cm³, $d \approx 2 \cdot 10^8$ erg/cm³, since experimental estimates of the f - d and f - f interaction constants are available only for this crystal: $\omega_{\text{E}} \sim 2 \cdot 10^5$ GHz, $\omega_{2f} \sim 10^2$ GHz, $\omega_{20} \sim 10^3$ GHz, $\omega_{\text{me5}} \sim 10^{-4}$ GHz, $\omega_{2fd} \sim 1$ GHz, $\omega_{\text{dip}} \sim 5 \times 10^{-2}$ GHz. Hence it follows that in the region of the Γ_4 - Γ_{24} OPT the condition $\omega_{20} > \omega_{2f}$ is satisfied in the erbium REO and here the f -mode ω_{II} is the soft mode. The activation frequency of this mode at the point of the OPT is determined by the formula (21c): $\omega_{\text{II}}(0) \sim 10$ GHz. In order to obtain a more accurate estimate additional experiments must be performed in order to determine the f - d and f - f interaction parameters. In the experiment performed in Refs. 13 and 14 the gap of the soft mode in the erbium REO was not determined—a single absorption signal was observed at the point of the OPT.^{13,14} This fact can be explained, as already mentioned above, by the large damping in the f -subsystem at high temperatures (in ErFeO₃ at $T_1 \approx 100$ K).

2) Γ_2 - Γ_{24} phase transition

According to Eq. (18b), near this OPT the quasiferromagnetic branch of the d -subsystem, one branch of the f -subsystem, and transverse sound polarized along the x -axis interact with one another. Due to the effect of the dipole interaction¹⁵ the behavior of the quasielastic branch ω_{III} (16) at the point of the OPT where $K_{ac} + K_2 = 0$ holds differs from the behavior of this branch at the point of the Γ_4 - Γ_{24} transition

$$\omega_{III} = s_3 k [\omega_{dip} / (\omega_{me5} + \omega_{dip})]^{1/2}. \quad (22)$$

The velocity ω_{III}/k of this branch at the transition point, though it does decrease, remains finite.

In the experiment the velocity of transverse sound s_3 polarized along the x -axis is indeed observed to decrease near T_1 and T_2 . The change in the sound velocity ranges from 0.5% for YbFeO₃ to 1.5% for ErFeO₃.^{9,12} The magnitude of the change in YbFeO₃ is different at T_1 and T_2 : The decrease in the sound velocity at $T = T_2$ is larger than at $T = T_1$. In ErFeO₃ the sound velocity changes by approximately the same amount near T_1 and T_2 . Such a small change in the sound velocity and the fact that the size of the change is different at the points of the Γ_4 - Γ_{24} and Γ_2 - Γ_{24} OPTs in YbFeO₃ can be explained as follows.

The smallness of the change in the sound velocity at $T = T_1$ is probably due to the large damping of sound (17) occurring near the Γ_4 - Γ_{24} transition. In the region of the Γ_2 - Γ_{24} OPT the sound damping coefficient (17) decreases as a result of the dipole interaction (19). Therefore the change in the sound velocity near this transition can be larger than at $T = T_1$. This is indeed observed experimentally for YbFeO₃ (Ref. 12). The limit on the change in the velocity at $T = T_2$ also is explained by the effect of the dipole interaction directly on the sound speed. In the experiment an increase was also observed in the change in the sound velocity in the region of the Γ_4 - Γ_{24} OPT in YbFeO₃ when a constant magnetic field was applied parallel to the z -axis. In a field $H = 35$ kOe this change is almost an order of magnitude larger than the analogous change occurring in the absence of a field ($H = 0$). This effect can be explained by the decrease in the damping coefficient (17) of transverse sound both due to the decrease in the damping coefficient Λ_f accompanying ordering of the f -subsystem in a magnetic field and by an increase in the frequency $\omega'_f(H)$ in Eq. (17).

Once again using the data of Refs. 2, 9, 10, and 16–19, we obtain for the frequencies appearing in Eq. (18b) for ErFeO₃ at the point of the Γ_2 - Γ_{24} OPT ($T = T_2$, $K_{ac} + K_2 = 0$) the following estimates: $\omega_{2f} \sim 5 \cdot 10^2$ GHz, $\omega_{20} \sim 10^2$ GHz, $\omega_{2fd} \sim 0.1$ GHz, $\omega_{dip} \sim 0.1$ GHz, $\omega_{me5} \sim 10^{-4}$ GHz. One can see that the condition $\omega_{20} < \omega_{2f}$ is satisfied near the Γ_2 - Γ_{24} transition. Thus near the Γ_2 - Γ_{24} OPT the quasiferromagnetic d -mode ω_1 is the soft mode. The gapwidth of this mode at the point $T = T_2$ of the OPT is determined by the formula (21b), and for ErFeO₃ a numerical estimate gives $\omega_1(0) \sim 140$ GHz. This agrees in order of magnitude with the experimental value.¹⁴ Hence it follows that the activation frequency of the soft modes is different at the points T_2 and T_1 (in the latter case, as shown above, $\omega_{II}(0) \sim 10$ GHz). Such a difference is observed experimentally.¹²

3) Γ_2 - Γ_{12} phase transition

It follows from Eq. (18a) that near a phase transition the quasiantiferromagnetic branch of the d -subsystem, one f -mode, and the transverse elastic branch polarized along the y -axis interact with one another. At the point $K_{cb}(T_3) = 0$ of the OPT as $k \rightarrow 0$ the quasielastic branch ω_{III} (16) depends quadratically on k

$$\omega_{III} = s_4 (g\alpha / M_0 \omega_{me4})^{1/2} k^2. \quad (23)$$

Near the OPT the velocity ω_{III}/k of this branch depends linearly on k and approaches zero as $k \rightarrow 0$. We note that in contrast to the Γ_2 - Γ_{24} transition at $T = T_3$ the dipole interaction does not affect the behavior of the quasielastic branch ω_{III} [see Eqs. (16) and (19)]. Since the Γ_2 - Γ_{12} transition occurs at low temperatures, damping in the f -subsystem and together with it the damping γ_{III} of the quasielastic branch given by (17) should be smaller than in the region of the Γ_4 - Γ_{24} and Γ_2 - Γ_{24} transitions. These two factors (no effect of the dipole interaction at $T = T_3$ and decrease of the damping at low temperatures) should result in the fact that the change in the velocity of transverse sound s_4 near the Γ_2 - Γ_{12} OPT will be larger than at the Γ_2 - Γ_{24} and Γ_4 - Γ_{24} transition. A change of up to 25% in the sound velocity at $T = T_3$ has been observed experimentally.^{13–14} This is the first time that such a large change has been observed in the sound velocity in a REO.

In the region of the Γ_2 - Γ_{12} transition we obtain from Refs. 2, 9, 17, and 18 the following estimates of the frequencies in Eq. (18a) for the erbium REO (assuming that at low temperatures the magnetoelastic constant B_{44} increases by at least an order of magnitude (and this will be shown below) compared with its value at high temperatures): $\omega_{1f} \sim 500$ GHz, $\omega_{me4} \sim 2 \cdot 10^{-2}$ GHz, $\omega_{1fd} \sim 10$ GHz, $\omega_{10} \sim 10^{-3}$ GHz. Thus at $T = T_3$ we have $\omega_{10} > \omega_{1f}$ and near this transition the soft mode is the quasi-rare-earth mode, whose activation frequency is determined by a formula similar to Eq. (21c):

$$\omega_{II}^2(0) = (\omega_{1f}/\omega_{10})^2 \omega_B \omega_{me4}. \quad (24)$$

Hence we have an estimate of the gapwidth of the soft mode at $T = T_3$: $\omega_{II}(0) \sim 140$ GHz. This value agrees in order of magnitude with experiment.¹⁴

Another feature was observed experimentally in the region of the Γ_2 - Γ_{12} transition: sharp asymmetry in the behavior of the soft mode to the left and right of the transition point. This asymmetry follows from the different temperature dependence of the effective f - f interaction and anisotropy constants to the right and left of T_3 . We now show that the temperature dependence of the indicated constants is indeed different in different temperature intervals. In the region of the high-temperature transitions, when the f -subsystem is in the paramagnetic state (which means that $f, c, \sigma \ll 1$) $\tanh^{-1} \sigma \approx \sigma$ and hence, according to Eq. (4a), $\lambda'_\alpha \approx T - \lambda_\alpha \approx T$ [since at high temperatures $T \gg \lambda_\alpha$ (Refs. 4 and 5)]. From Eqs. (6) and (8) it follows that

$$\begin{aligned} K_{ac} &= \bar{K}_{ac}(T) - N(B_z'^2 + B_z''^2 - B_z^2)/T, \\ K_{cb} &= \bar{K}_{cb}(T) + N(B_z'^2 + B_z''^2 - B_z^2)/T, \end{aligned} \quad (25)$$

where the temperature dependence of the anisotropy constants $\bar{K}_{ac,cb}$ is determined by the temperature dependence of the van Vleck^{4,5,19} and magnetoelastic contributions to these

constants. As a result, taking into account the experimental data of Refs. 9 and 19 for the erbium REO in the temperature range where $\Gamma_4-\Gamma_{24}-\Gamma_2$ spin orientation occurs, the anisotropy constant K_{ac} depends linearly on T : $|K_{ac}| = |0.214 - 2.42 \cdot 10^{-3} T| \text{ K}$.

Near the $\Gamma_2-\Gamma_{12}$ OPT (at temperatures $T \gg T_3$) the f -subsystem is close to the ordered state (according to Ref. 16, in erbium REO spin reorientation in the d -subsystem at $T = T_3$ is accompanied by simultaneous antiferromagnetic ($c_z \neq 0$) ordering in the f -subsystem). In this case we have f , c , $\sigma \propto 1$. In order to determine the temperature dependence of the constants λ'_α and K_{cb} at temperatures $T \gg T_3$ we suppose, for simplicity, that $a = B'_z = \lambda_{7,8} = 0$ and $B'_z \gg \lambda_\alpha \sigma$. It then follows from Eq. (4) that in the Γ_2 phase we have $f_z = c_y = 0$ and $f_x = \sigma = B'_z / \lambda'_1$, and from Eq. (4a) it follows that $\tanh^{-1} \sigma \approx B'_z / T$, i.e., $f_x = \tanh(B'_z / T)$. Substituting these results into the formula for λ'_α (4a) and $K_{ac,cb}$, Eqs. (6) and (8), we find that for $T \gg T_3$

$$\lambda'_\alpha \approx B'_z / \text{th}(B'_z / T), \quad (26)$$

$$K_{ac,cb} \approx \tilde{K}_{ac,cb}(T) \mp N(B'_z{}^2 - B_{x,y}^2) \text{th}(B'_z / T) / B'_z.$$

Here the constants $\tilde{K}_{ac,cb}$ can depend on the temperature only through the magnetoelastic contribution, since at low temperatures the van Vleck contribution is practically constant.¹⁹ Using the experimental data of Refs. 2, 13, 14, and 19 we obtain finally from Eq. (27) the following expression for the constant K_{cb} in the erbium REO at temperatures $T \gg T_3$: $K_{cb} = [3.37 - 6.42 \tanh(2.4/T)] \text{ K}$. At temperatures $T < T_3$ the f -subsystem is ordered. In this case the temperature dependence of the f - f and f - d interaction parameters should change once again, and this likewise will result in a different temperature dependence of the anisotropy constants at temperatures $T < T_3$.

Thus the asymmetry in the behavior of the soft mode in the region of the $\Gamma_2-\Gamma_{12}$ OPT can indeed be explained by the different temperature dependences of the effective f - f and f - d interaction and anisotropy constants to the right and left of T_3 .

If the specific temperature dependence of the anisotropy constants is known, then the magnitude of the magnetoelastic constants B_{55} and B_{44} at different temperatures can be determined from the experimental dependence of the velocities $s(T) = \omega_{\text{III}}(T)/k$ of quasisound waves given by (16).^{11,13,14} In the erbium REO the following values were obtained in Ref. 11 for B_{55} and B_{44} near high-temperature transitions: $B_{55} \approx 2.2 \times 10^6 \text{ ergs/cm}^3$ and $B_{44} \approx 4 \times 10^6 \text{ ergs/cm}^3$. Using the experimental dependence of the velocity of transverse sound polarized along the y -axis^{13,14} and the T -dependence presented above for the anisotropy constant K_{cb} near the $\Gamma_2-\Gamma_{12}$ OPT at temperatures $T \gg T_3$, we find that the magnetoelastic constant B_{44} at low temperatures increases by almost two orders of magnitude: $B_{44} \approx 2.5 \cdot 10^8 \text{ ergs/cm}^3$. This significant increase of the magnetoelastic constant B_{44} in the region of the $\Gamma_2-\Gamma_{12}$ OPT can apparently be explained by the increase in the contribution of the f -subsystem at low temperatures to the magnetoelastic energy as a result of the closeness of the f -subsystem to the ordered state.

We note that the different degree of change in the sound velocity at these transitions can also be explained by the different value of the magnetoelastic constants B_{55} and B_{44} and the different temperature dependences of the anisotropy constants near the $\Gamma_4-\Gamma_{24}$, $\Gamma_2-\Gamma_{24}$, and $\Gamma_2-\Gamma_{12}$ transitions.

Indeed, assuming that the relation (25) holds in the region of the $\Gamma_4-\Gamma_{24}$ and $\Gamma_2-\Gamma_{24}$ transitions, we find that a factor of two decrease in the sound velocity should be observed as the OPT is approached to within $\Delta T \approx M_0 \omega_{me5} / (g \partial K_{ac} / \partial T) \sim 10^{-4} - 10^{-3} \text{ K}$, i.e., in order to observe large changes in the sound velocity near high-temperature transitions it is necessary to approach extremely close to the point of the OPT. In the region of the $\Gamma_2-\Gamma_{12}$ transition, however, such a decrease of the sound velocity will be observed when the OPT point is approached to within $\Delta T \approx M_0 \omega_{me4} / (g \partial K_{cb} / \partial T) \sim 10^{-2} - 10^{-1} \text{ K}$. In an experiment it is easily possible to approach the OPT point to within this temperature interval.

The OPT studied ($\Gamma_2-\Gamma_{12}$) in erbium REO is the only (and therefore unique) temperature transition for which such a significant change in the sound velocity as achieved.

7. CONCLUSIONS

The following conclusions can be drawn from the theoretical investigation, performed in this work, of the coupled oscillations of rare earth, iron, and elastic subsystems in REO with Kramers ions and from the comparison of the results with experiment.

Depending on the ratio of the oscillation frequency of the f -subsystem and the oscillation frequency of the d -subsystem, renormalized by the coupling with the f -subsystem, the elastic subsystem, and the dipole interaction, the soft mode near an OPT will be either one of the quasi-iron modes or one of the quasi-rare earth modes. For example, in erbium REO near the $\Gamma_4-\Gamma_{24}$ and $\Gamma_2-\Gamma_{12}$ transitions the soft mode is the quasi-rare-earth mode, while near the $\Gamma_2-\Gamma_{24}$ transition the soft mode is apparently the quasiferromagnetic mode.

Near the $\Gamma_4-\Gamma_{24}$ OPT the small change in the velocity of transverse sound polarized along the x -axis, is explained by the large damping constant in the paramagnetic f -subsystem (the linewidth depends on the temperature and at high temperatures it is of the order of the frequency itself^{4,5}), in terms of which the sound damping constant (17) is expressed. The damping constant can be so large that near a transition there will be no echo signal.¹¹ The small change in the sound velocity is also explained by the fact that the temperature interval near T_1 , in which significant decrease (by at least a factor of two) in the velocity occurs is extremely narrow ($\sim 10^{-4} \text{ K}$) and is not resolved experimentally.

Sound damping is weaker near the $\Gamma_2-\Gamma_{24}$ phase transition than near the $\Gamma_4-\Gamma_{24}$ transition. This is because of the effect of the dipole interaction, owing to the noncollinearity of the wave vector and the ferromagnetism vector, so that the change in the sound velocity in YbFeO_3 in the region of this transition is larger than in the case of the $\Gamma_4-\Gamma_{24}$ transition. Even here, however, the magnitude of the change in the sound velocity is small (0.5–3%) because the change itself is limited by the dipole interaction (22).

In the region of the low-temperature phase transition $\Gamma_2-\Gamma_{12}$ in erbium REO the experimentally observed 25% decrease of the velocity of sound, polarized along the y -axis, can be explained, first, by the fact that at low temperatures the damping in the f -subsystem and therefore the sound damping also decrease significantly. Second, near this transition the dipole interaction (23) does not limit the change in the sound velocity. The weak temperature dependence of the

frequency of the soft mode observed likewise near the Γ_2 - Γ_{12} transition above the transition is explained by the fact that at low temperatures the temperature dependence of the anisotropy constant (25) and (26) can change significantly. This in turn increases the temperature interval of "closeness" to the OPT point to tenths of a degree, which makes this transition unique, since such a large temperature interval of closeness to an OPT has still not been observed in any magnetic material.

Estimates of the gapwidths of the soft modes in the region of an OPT, which were obtained in the present work, agree in order of magnitude with the experimental values. In order to confirm the conclusions drawn in the present work and to compare theory and experiment more precisely, new experiments must be performed in order to determine the temperature dependence of the anisotropy constants as well as the magnetoelastic, f - f , and f - d interaction constants.

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