Spectrum of magnetic oscillations and interaction of antiferromagnetic and paramagnetic subsystems in $ErCrO_3$

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(Submitted April 18, 1975)
Zh. Eksp. Teor. Fiz. **69**, 1301–1311 (October 1975)

Homogeneous magnetic resonance spectra of a weak ferromagnet $ErCrO_3$ were investigated in the wavelength range 6–0.6 mm using pulse magnetic fields at temperatures 5–80°K. Paramagnetic resonance with the Er^{3+} ions was observed in addition to antiferromagnetic resonance in the chromium subsystem. An investigation was made of the interaction between oscillations in the antiferromagnetic and paramagnetic subsystems. The parameters of the Er-Cr interaction were determined. An additional branch was found in the magnetic oscillation spectrum and this branch was due to transitions in the excited Er^{3+} ions. A calculation was made of the spectrum of homogeneous magnetic oscillations of the system under investigation. A magnetic phase transition $\Gamma_4^- \to \Gamma_4^+$, associated with the interaction between the paramagnetic and antiferromagnetic subsystems, was detected.

PACS numbers: 76.50.+g, 75.30.Ce

Rare-earth orthoferrites and orthochromites contain two types of magnetic ions: 3d and 4f. The Néel temperature (T_N) of these compounds is governed by the strongest interaction, which is the exchange interaction within the 3d subsystem and which usually reaches several hundreds degrees Kelvin.

The exchange interaction between ions in the rare-earth subsystem is weak and usually it does not exceed several degrees Kelvin. At temperatures $10°K \leq T \leq T_N$ the interactions between the 4f ions alone cannot order magnetically the rare-earth subsystem, which remains in the paramagnetic state. However, the coupling with the magnetically ordered 3d subsystem induces in the rare-earth subsystem a magnetic order corresponding to one of the configurations which are possible in crystals of the space group in question. The 3d-4f interactions in the majority of orthoferrites and orthochromites are of the order of several tens degrees Kelvin.

Our aim was to investigate the spectrum of magnetic oscillations of the system composed of interacting antiferromagnetic (3d) and paramagnetic (4f) subsystems in erbium orthochromite $ErCrO_3$.

Low-lying excited states of rare-earth ions appear in the crystal field of some rare-earth orthoferrites and orthochromites because of the weakness of this field compared with the spin-orbit interaction. The gap between the ground and lowest excited states usually does not exceed 100 cm^{-1}. At temperatures of several hundreds degrees Kelvin such excited levels are populated and they contribute to the magnetic properties. In view of this, it would be interesting to detect and investigate the resonant absorption associated with the exchange and Zeeman splitting of the rare-earth ion levels in the ground and excited states. The presence of a strong "magnetization" influence exerted by the 3d subsystem establishes favorable conditions for the experimental detection of such transitions.

1. CRYSTAL AND MAGNETIC STRUCTURES

Erbium orthochromite $ErCrO_3$ (T_N = 133°K) has the orthorhombically distorted perovskite structure and the space group Pbnm.[1] Eight different magnetic configurations are possible in crystals with this space group[1,2] and three of them can occur in $ErCrO_3$ (Fig. 1).

The state Γ_1 appears at temperatures $T \leq 9.8°K$ in the absence of an external magnetic field. At T = 9.8°K in $H_0 = 0$ a transition takes place to the Γ_4 state[3] and this state is retained up to T_N. In the Γ_4 state the application of an external field H_0 along the c (z) crystal axis shifts the transition from Γ_1 to Γ_4 toward lower temperatures.[4] However, if the field is applied along the a (x) crystal axis, the magnetic system goes over to the state Γ_2 in a "flopping" field $H_0 = H_{fl}$.[4] In erbium orthochromite the ground state of the Er^{3+} ions is $^4I_{15/2}$. The two lowest excited states are split by the crystal field by 46 and 113 cm^{-1} from the ground state.[5-7] The exchange interaction and an external magnetic field split further these levels, which are Kramers doublets.

2. FREE ENERGY. MAGNETIC RESONANCE FREQUENCIES

The free energy in the state Γ_4 calculated including second-order invariants is

$$\begin{aligned} F={}&E\mathbf{J}_1\mathbf{J}_2+E_{xx}J_{1x}J_{2x}+E_{zz}J_{1z}J_{2z}-A_{xx}'(J_{1x}^2+J_{2x}^2)\\ &-A_{zz}'(J_{1z}^2+J_{2z}^2)-D(J_{1x}J_{2z}-J_{1z}J_{2x})-A(J_{1x}J_{1z}-J_{2x}J_{2z})\\ &-(\gamma_1\mathbf{J}_1+\gamma_2\mathbf{J}_2)\mathbf{H}_0+\mathscr{E}_{zz}^n(J_{1z}+J_{2z})I_z^n+\mathscr{E}_{xx}^n(J_{1x}+J_{2x})I_x^n\\ &+\mathscr{E}_{yy}^n(J_{y1}+J_{y2})I_y^n-d_1^n(J_{1z}-J_{2z})I_x^n-d_2^n(J_{1x}-J_{2x})I_z^n-\gamma^n\mathbf{l}^n\mathbf{H}_0. \end{aligned} \quad (1)$$

We shall be interested in the temperature range ($T \geq 10°K$) in which the interactions within the rare-earth subsystem can be ignored; $\mathbf{J}_{1,2}$ are the vectors of the mechanical moments of the chromium sublattices.

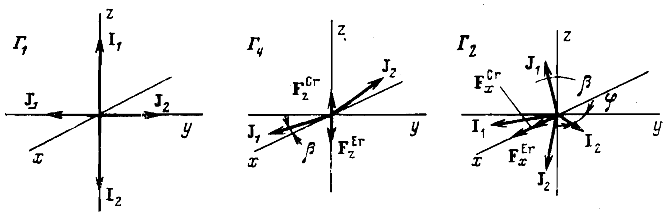

FIG. 1. Magnetic states of $ErCrO_3$ in the two-sublattice model of the chromium ($J_{1,2}$) and erbium ($I_{1,2}$) subsystems. The designations of the magnetic configurations and of the projections of the vectors of the irreducible representations $\Gamma_1(G_y^{Cr}, C_z^{Er})$, $\Gamma_4(G_x^{Cr}F_z^{Cr}, F_z^{Er})$, and $\Gamma_2(G_z^{Cr}F_x^{Cr}, C_y^{Er}F_x^{Er})$, are the same as in [2].

The sublattice concept will also be used for the induced moment of the erbium subsystem; $\mathbf{I}_n$ is the mechanical moment vector of the erbium sublattice. The index n will be used for a state in the erbium subsystem: this may be the ground state (n = 0) or one of the excited states (n = 1, 2, ...). Summation is carried out over the index n. Thus, the total induced moment in the erbium subsystem is the sum of the moment in the ground and excited states:

$$M_\alpha{}^{\mathrm{Er}}=\gamma_{\alpha\beta}{}^nI_\beta{}^n. \tag{2}$$

The parameters of the interaction between the erbium and chromium subsystems and the components of the g factor of the Er^{3+} ion in the ground state are different from the corresponding characteristics of each of the excited states. In Eq. (1) the magnetomechanical ratio tensors for the chromium sublattices $\boldsymbol{\gamma}_{1,2}$ are

$$\gamma_{1,2}=\left|\begin{array}{ccc}\gamma_{xx} & 0 & \pm\gamma_{xz}\\ 0 & \gamma_{yy} & 0\\ \pm\gamma_{zx} & 0 & \gamma_{zz}\end{array}\right|. \tag{3}$$

The indices 1 and 2 and the signs ± apply to the first and second chromium sublattices, respectively. The tensor $\boldsymbol{\gamma}^n$ for the erbium subsystem is

$$\gamma^n=\left|\begin{array}{ccc}\gamma_{xx}{}^n & 0 & 0\\ 0 & \gamma_{yy}{}^n & 0\\ 0 & 0 & \gamma_{zz}{}^n\end{array}\right|. \tag{4}$$

We shall find the normal frequencies of the system from the equations of motion for mechanical moments.[8] This faces us with a problem of mixed oscillations of the antiferromagnetic and paramagnetic subsystems. The solution of this problem gives

$$(\omega^2-\omega_{xy}{}^2)(\omega^2-\omega^{n2})-K_{xy}{}^2=0, \tag{5}$$

$$(\omega^2-\omega_z{}^2)=0. \tag{6}$$

In Eq. (6) the partial frequencies ω_{xy} and ω^n are

$$\begin{aligned}\omega_{xy}{}^2={}&2EJ_0\cdot A_{x-z}J_0+2EJ_0\cdot d_2{}^nI_0{}^n+(4AJ_0-\mathscr{E}_{zz}{}^nI_0{}^n)\\ &\times[(D+A)J_0-\mathscr{E}_{zz}{}^nI_0{}^n]+\gamma_{zz}H_0[(D+5A+2E\tau_2)J_0\\ &-2\mathscr{E}_{zz}{}^nI_0{}^n]+(\gamma_{zz}H_0)^2;\end{aligned} \tag{7}$$

$$\omega^n=2(d_2{}^n-\mathscr{E}_{zz}{}^n\sin\beta)J_0+\gamma_{zz}{}^nH_0; \tag{8}$$

here,

$$A_{x-z}J_0=(A_{xx}-A_{zz})J_0;\quad A_{xx}J_0=(E_{xx}+2A_{xx}')J_0,$$
$$A_{zz}J_0=(E_{zz}+2A_{zz}')J_0,\quad \tau_2=\gamma_{zx}/\gamma_{zz}.$$

The canting angle of the chromium sublattices is given by

$$\sin\beta=[(D+A)J_0-\mathscr{E}_{zz}{}^nI_0{}^n+\gamma_{zz}H_0]/2EJ_0. \tag{9}$$

The coupling between the oscillations ω_{xy} and ω^n is

$$\begin{aligned}K_{xy}{}^2={}&4(\mathscr{E}_{xx}{}^n\sin\beta+d_1{}^n)^2J_0{}^2(\mathscr{E}_{yy}{}^{n2}I_0{}^{n2}-\omega^nEI_0{}^n)\\ &-\omega_{xy}{}^2\omega^n\mathscr{E}_{yy}{}^{n2}I_0{}^n/E.\end{aligned} \tag{10}$$

In the weak coupling case ($K_{xy}^2 \ll \omega_{xy}^2\omega^{n2}$) and far from the intersection of the spectra ($\omega_{xy} = \omega^n$) this system should exhibit independent oscillations of the antiferromagnetic subsystem (ω_{xy}) and of the paramagnetic subsystem (ω^n). It is clear from Eq. (7) that in this case also the antiferromagnetic resonance (AFMR) frequency ω_{xy} depends on the parameters of the interaction between the antiferromagnetic and paramagnetic subsystems.

It is clear from Eq. (6) that oscillations in the rare-earth (erbium) subsystem ω^n do not interact with oscillations in the antiferromagnetic subsystem ω_z. Then, the AFMR frequency ω_z is given by

$$\begin{aligned}\omega_z{}^2={}&2EJ_0\cdot A_{xx}J_0+2EJ_0\cdot d_2{}^nI_0{}^n+(D+A)J_0[(D+A)J_0\\ &-\mathscr{E}_{zz}{}^nI_0{}^n]+\gamma_{zz}H_0(D+A+2E\tau_2)J_0.\end{aligned} \tag{11}$$

In the Γ_2 state ($\mathbf{H}_0 \parallel \mathrm{a}$ and $H_0 > H_{fl}$) the chromium and erbium subsystems have two sublattices each and this is true separately of the ground state and of each of the excited states. The expression for the energy then becomes

$$\begin{aligned}F={}&E\mathbf{J}_1\mathbf{J}_2+E_{xx}J_{1x}J_{2x}+E_{zz}J_{1z}J_{2z}-A_{xx}'(J_{1x}{}^2+J_{2x}{}^2)\\ &-A_{zz}'(J_{1z}{}^2+J_{2z}{}^2)-D(J_{1x}J_{2z}-J_{1z}J_{2x})-A(J_{1x}J_{1z}-J_{2x}J_{2z})\\ &-(\boldsymbol{\gamma}_1\mathbf{J}_1+\boldsymbol{\gamma}_2\mathbf{J}_2)\mathbf{H}_0+\mathscr{E}_{xx}{}^nJ_x{}^+(I_x{}^{n-}\cos\varphi+I_y{}^{n+}\sin\varphi)\\ &+\mathscr{E}_{yy}{}^nJ_y{}^+(I_y{}^{n-}\cos\varphi-I_x{}^{n+}\sin\varphi)+\mathscr{E}_{zz}{}^nJ_z{}^+I_z{}^{n+}\\ &+c_1{}^nJ_z{}^-(I_y{}^{n+}\cos\varphi-I_x{}^{n-}\sin\varphi)+c_2{}^nJ_y{}^-I_z{}^{n-}+e_1{}^nJ_z{}^-(I_x{}^{n-}\cos\varphi\\ &+I_y{}^{n+}\sin\varphi)+e_2{}^nJ_x{}^-I_z{}^{n+}+f_1{}^nJ_x{}^+(I_y{}^{n+}\cos\varphi\\ &-I_x{}^{n-}\sin\varphi)+f_2{}^nJ_y{}^+(I_x{}^{n+}\cos\varphi+I_y{}^{n-}\sin\varphi)\\ &-\gamma_{xx}{}^n(I_x{}^{n-}\cos\varphi+I_y{}^{n+}\sin\varphi)H_0.\end{aligned} \tag{12}$$

Here,

$$J_\alpha{}^\pm=J_{1\alpha}\pm J_{2\alpha},\quad I_\alpha{}^{n\pm}=I_{1\alpha}{}^n\pm I_{2\alpha}{}^n.$$

The noncollinearity of the erbium moments is represented by the angle φ described by

$$[\gamma_{xx}{}^nH_0-2J_0(\mathscr{E}_{xx}{}^n\sin\beta-e_1{}^n)]\cos\varphi-2c_1{}^nJ_0\sin\varphi=0. \tag{13}$$

The canting angle of the chromium sublattices in the Γ_2 state is given by

$$\sin\beta=[(D-A)J_0-(\mathscr{E}_{xx}{}^n\sin\varphi+f_1{}^n\cos\varphi)I_0{}^n+\gamma_{xx}H_0]/2EJ_0. \tag{14}$$

In the Γ_2 state the solution of the problem of mixed oscillations in the chromium and erbium subsystems gives

$$(\omega^2-\omega_{zy}^2)(\omega^2-\omega^{n2})-K_{zy}^2=0,\quad (\omega^2-\omega_x{}^2)(\omega^2-\omega^{n2})-K_x{}^2=0. \tag{15}$$

We shall give only the expressions for the frequencies of the independent oscillations which are excited in the weak coupling case far from the intersections of the spectra in question. The formulas for the AFMR frequencies in the chromium subsystem are

$$\begin{aligned}\omega_{zy}^2={}&-2EJ_0\cdot A_{x-z}J_0+2EJ_0\cdot a_1{}^nI_0{}^n+(4AJ_0\\ &+a_2{}^nI_0{}^n)[(D-A)J_0-a_2{}^nI_0{}^n]+\gamma_{xx}H_0[(D-5A\\ &-2E\tau_1)J_0-2a_2{}^nI_0{}^n]+(\gamma_{xx}H_0)^2;\end{aligned} \tag{16}$$

$$\begin{aligned}\omega_x{}^2={}&2EJ_0\cdot A_{zz}J_0+2EJ_0\cdot a_1{}^nI_0{}^n+(D-A)^2J_0{}^2\\ &-2\mathscr{E}_{xx}^n I_0{}^n\sin\varphi[(D-A)J_0-2a_2{}^nI_0{}^n]\\ &+\gamma_{xx}H_0[(D-A-2E\tau_1)J_0+2f_1{}^nI_0{}^n\cos\varphi].\end{aligned} \tag{17}$$

Here, $a_1^n = e_1^n \sin\varphi + c_1^n \cos\varphi$, $a_2^n = \mathscr{E}_{xx}^n \sin\varphi + f_1^n \times \cos\varphi$. The resonance frequencies in the erbium subsystem can be expressed in the form

$$\omega^n=2J_0[c_1{}^n\cos\varphi-(\mathscr{E}_{xx}^n\sin\beta-e_1{}^n)\sin\varphi]+\gamma_{yy}^n H_0\sin\varphi. \tag{18}$$

3. EXPERIMENTAL RESULTS

The magnetic resonance measurements were carried out employing a quasioptic monochromatic spectrometer in the wavelength range 6–0.6 mm and applying magnetic field pulses up to 80 kOe intensity at temperatures of 5–80°K. Backward-wave tubes were used as radiation sources. The resonance signals were detected with a cooled n-type InSb cell.

An optical cryostat with polyethylene terephthalate windows was placed inside the spectrometer. A sample was inserted into a corundum holder and placed inside a tube filled with gaseous helium which circulated through the helium bath of the cryostat. The temperature was varied by a heater wound on the holder and measured with a thermocouple (at T > 20°K) or with a carbon resistance thermometer (at T < 20°K).

The magnetic field was measured with a special system which produced a reference field. This system included additional coils with few turns, which were connected in series with the coils of the pulse electromagnet, and a centimeter-range resonator located between these coils. A standard in the form of an yttrium iron garnet sphere was placed inside this resonator. The reference field was provided by the ferromagnetic resonance signal of this sphere. The system was subjected to a preliminary calibration which gave the ratio of the magnetic field in the resonator with the standard to the magnetic field at the center of the measuring cell in the spectrometer. This ratio could be varied within a wide range by altering the working frequency of the reference system and moving the standard inside the resonator. The use of this method reduced the error in the field measurements below 2–3%.

Single crystals of erbium orthochromite were grown by spontaneous crystallization from the melt. The investigated samples were polished plates of ~ 5 mm^2 area and 0.1–0.2 mm thick.

The temperature dependences of the resonance fields were recorded at fixed frequencies in the range 50–500 GHz in an external magnetic field oriented along the principal crystal axes. These results were used to plot the dependences of the resonance frequency on the external field at fixed temperatures.

A. $H_0 \parallel c$ (State Γ_4)

The number and nature of the observed magnetic resonances depended strongly on the frequency and temperature. In the frequency range 150–300 GHz only one signal was observed at low temperatures. At higher frequencies there were two signals and in a narrow range near 370 GHz there were three signals.

Figure 2 shows the dependences $\omega_{\text{res}}(H)$ plotted for four different temperatures. We can see that at all temperatures there is a resonant absorption (ω^0) with a linear dependence of the resonance frequency on the field in a wide frequency range, and the slope of the dependence $\omega^0(H)$ as well as the position of $\omega^0(0)$ are practically independent of temperature. If we assume that the signal represents a paramagnetic resonance in the erbium subsystem in the ground state and if we apply Eq. (8), we find that the slope of the dependence $\omega^0(H)$ gives

$$g^0_{zz} = 5.8. \tag{19}$$

This value is identical with the g factor of the Er^{3+} ion in the ground state, deduced from measurements of the Zeeman shift of the optical lines of $ErFeO_3$.[10] Figure 3 shows the temperature dependence of the activation energy $\omega^0(0)$ for this magnetic oscillation branch. The quantity $\omega^0(0)$ represents the splitting of the ground level of the Er^{3+} ion due to the Er-Cr interaction. Figure 3 includes also the temperature dependences of this splitting obtained for $ErCrO_3$ from the Mössbauer effect[6] and from optical spectra.[7] We can see that the results agree at temperatures T > 10°K. It follows from[7] that at T = 10°K the splitting has a jump due to the phase transition from the state Γ_1 to the state Γ_4.[3] In our case this jump is not observed. This is due to the presence of an external field in our measurements (no field was used in[7]).

The signals denoted by ω_z in Fig. 2 are observed in a narrow frequency range near 370 GHz at all temperatures. The positions of these signals are practically independent of temperature. Similar signals were observed by us in a study of the longitudinal branch of AFMR in $YCrO_3$.[11] It follows from Eq. (11) that the slope of $\omega_z^2(H)$ for $ErCrO_3$ is independent of the parameters of the erbium subsystem and it is governed entirely by the chromium subsystem. A comparison of these slopes for $YCrO_3$[11] and $ErCrO_3$ shows that they are identical. Thus, the observed signals represent the longitudinal AFMR in the chromium subsystem.

It is clear from Fig. 2 that there are also signals whose resonance frequencies depend strongly on temperature (these are denoted by ω_{xy}). We shall attribute these signals to the transverse AFMR in the chromium subsystem. In fact, if we apply the formulas (7) and (11) for the AFMR frequencies for the chromium subsystem and if we bear in mind that $A_{xx}J_0 \gg A_{xz}J_0$ [11] we find that the temperature-dependent terms $\mathscr{E}^n_{zz}I^n_0$ and $d^n_2 I^n_0$ make a much greater contribution in the case of the transverse (ω_{xy}) oscillations then in the longitudinal (ω_z) case.

Applying Eqs. (7) and (11) to the experimental curves ω_{xy} and ω_z and using the parameters of the chromium subsystem deduced from the AFMR in $YCrO_3$,[11] we can calculate the values of $\mathscr{E}^n_{zz}I^n_z$ and $d^n_2 I^n_0$ which govern

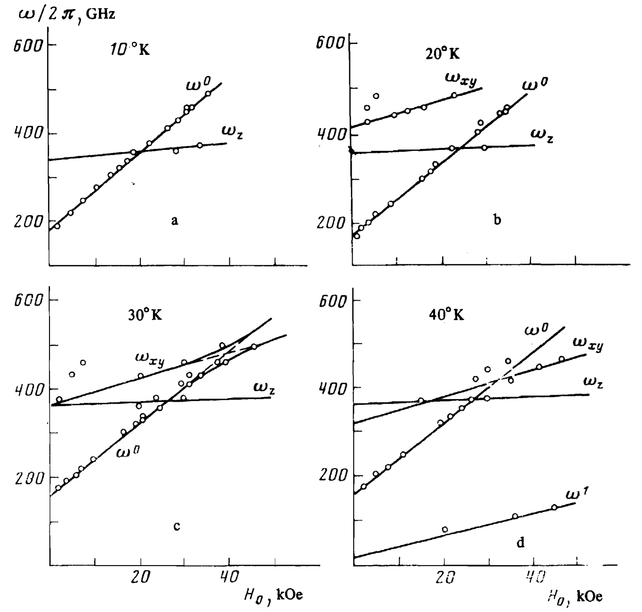

FIG. 2. Dependences of the resonance frequencies on the magnetic field obtained in the Γ_4 state at various temperatures. The ~450 GHz resonance signals in weak fields, observed at 20 and 30°K, are probably due to the inequivalence of the positions of the ions which is ignored in the two-sublattice model. The dependence $\omega(H)$ at T=77°K is given in [9].

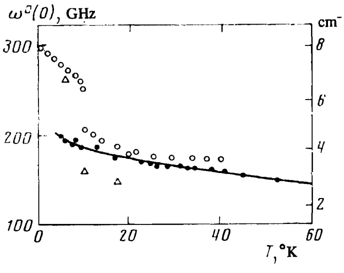

FIG. 3

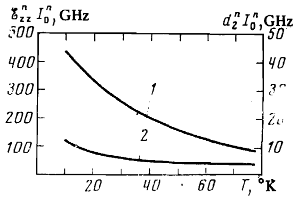

FIG. 4

FIG. 3. Temperature dependence of the initial splitting of Er^{3+} in the ground state: ●) our results; ○) optical data in [7]; △) from the Mossbauer effect [6].

FIG. 4. Temperature dependences of the Er-Cr interaction parameters: 1) $\mathscr{E}_{zz}^n I_0^n$; 2) $d_2^n I_0^n$.

the Er-Cr interaction energy in the Γ_4 state. The temperature dependences of these parameters are plotted in Fig. 4.

At temperatures above 35°K there are, in addition to the oscillation branches discussed above, absorption lines denoted by ω^1 in Fig. 2d. These lines are narrow and the slopes of the dependences are gentle. Since the absorption lines in question disappear at lower temperatures, we shall assume that they represent a resonance of the erbium subsystem in the first excited state. We can show that the g factor of the first excited state of Er^{3+} in $ErFeO_3$

$$g^1_{zz}=1.8, \qquad (20)$$

taken from[10], describes satisfactorily the slope of $\omega^1(H)$. Then, extrapolating $\omega^1(H)$ to zero we obtain the activation energy of these oscillations

$$\omega^1(0)\lesssim 20 \text{ GHz}. \qquad (21)$$

The temperature dependences of $\mathscr{E}^n_{zz}I^n_0$ and $d^n_2I^n_0$ and the temperature dependence of the magnetic moment in the Γ_4 state deduced from the magnetic measurements reported in[4] can be used, in combination with Eqs. (8) and (9), to calculate the initial splitting of Er^{3+} due to the Er-Cr interaction and to compare this splitting with the experimental value. At low temperatures ($T < 30°K$), when the population of the excited levels is weak ($N_1/N_0 < 0.1$), the calculated value effectively applies to the ground state. At higher temperatures there is generally some contribution from the excited states. However, according to Eq. (21) and Fig. 3, we have $\omega^1(0) \ll \omega^0(0)$. Thus, the calculated total splitting $\omega^n(0)$ should at all temperatures be equal to the activation energy $\omega^0(0)$ of the paramagnetic resonance of the erbium subsystem in the ground state. This is indeed found to be true. The lines in Figs. 2 and 3 are calculated from Eqs. (7)–(9) and (11) using the parameters found experimentally. The values of these parameters at 10 and 50°K are listed in Table I. We can see that the experimental points are described well by the calculated lines. Deviations in the line intersection regions are due to the interaction between the oscillations involved.

The equation for the frequencies of mixed oscillations (5) and the normal frequencies extrapolated from the experimental results to the intersection region can be used to find the coupling coefficient describing best the experimental dependence $\omega(H)$ in the interaction region. The results of such a calculation are plotted in Fig. 2c for the interaction between the oscillations ω_{xy} and ω^0 at $T = 30°K$. The optimal value of the coupling coefficient $\kappa^2_{xy} = K^2_{xy}/(\omega^2_{xy} + \omega^{n2})$ is found to be ~ 0.1. It follows from the curves in Fig. 2 that the dependences $\omega(H)$ for the oscillations ω_z and ω^0 also intersect. However, as expected, this intersection does not result in a strong interaction.

Measurements in the frequency range 200–250 GHz revealed also very strong absorption signals whose positions along the field axis depended weakly on temperature and frequency. A characteristic feature of these signals was a strong hysteresis: the magnetic fields corresponding to the absorption maximum were different for the rising and falling current (Fig. 5). As pointed out earlier, the Γ_4 state appears in $ErCrO_3$ in the temperature range $10°K < T < T_N$ and the ferromagnetic moments of the chromium F^{Cr}_z and erbium F^{Er}_z subsystems are antiparallel (Fig. 1). We shall denote

TABLE I. Free-energy parameters of $ErCrO_3$ (cm^{-1}).

T, °K	$2EJ_0$	DJ_0	AJ_0	$2E\tau_2J_0$	$A_{xx}J_0$	$A_{x-z}J_0$	$\mathscr{E}^n_{zz}I^n_0$	$d^n_2I^n_0$
10	300	7.14	1.0	3.90	$2.6\cdot10^{-1}$	$1.0\cdot10^{-2}$	1.43	0.39
50	290	6.9	0.97	3.77	$2.5\cdot10^{-1}$	$0.7\cdot10^{-2}$	$4\cdot10^{-2}$	$1.53\cdot10^{-2}$

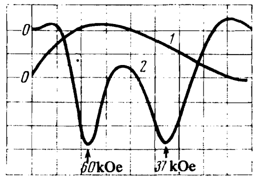

FIG. 5. Oscillograms of a magnetic field pulse (curve 1) and of an absorption signal for the Γ_4^-–Γ_4^+ transition (curve 2). T=38°K, ω=250 GHz.

this state by Γ_4^-. Clearly, if an external field oriented along the c axis (z axis) is sufficiently strong, a phase transition can take place from Γ_4^- to Γ_4^+ and in the latter case the two moments F^{Cr}_z and F^{Er}_z are canted with respect to the field. This transition may occur suddenly and be accompanied by hysteresis. The critical field of this transition $H_\mp$ can be found by equating the external field to the internal effective field exerted by the chromium on the erbium subsystem:

$$H_\mp=2J_0(d_2{}^n-\mathscr{E}^n_{zz}\sin\beta)/\gamma=\omega^n(0)/\gamma. \qquad (22)$$

An estimate based on the above formula gives $H_\mp \approx 60$ kOe, which depends weakly on temperature (Fig. 3). The signals mentioned above are wide (10–15 kOe) absorption bands and they are observed in fields not exceeding 60 kOe. We shall assume that these signals are associated with the absorption by a sample in some intermediate state which appears in the region of the Γ_4^-–Γ_4^+ phase transition.

B. Case $H_0 \parallel a$

In contrast to the $H_0 \parallel c$ case, which is characterized by a single magnetic state, Γ_4, in the $H_0 \parallel a$ configuration the magnetic system may be in one of the three states Γ_1, Γ_2, and Γ_4, depending on temperature and external field. Moreover, near the Γ_1–Γ_4 and Γ_4–Γ_2 transitions we may have intermediate states Γ_{14} and Γ_{42} which—like the pure states—have their own sets of normal oscillation frequencies. We shall not analyze in detail the magnetic oscillation spectra of all these states but we shall confine ourselves principally to a description of the experimental data obtained for the Γ_2 state.

Measurements at low frequencies (53 GHz) at $T > 10°K$ revealed a resonance in magnetic fields corresponding to the reorientation region (Γ_4–Γ_2 transition). Two pairs of signals, characterized by different temperature dependences of the absorption intensity were observed. The signals whose intensities fell rapidly with increasing temperature were attributed to the erbium subsystem and the signals with a weak temperature dependence of the intensity to the chromium subsystem.

At temperatures $T > 10°K$ and in fields $H_0 > H_{fl}$, when the state Γ_2 was obtained, there were three absorption lines denoted in Fig. 6 by ω_{zy}, ω^0, and ω^1. The signal corresponding to the ω_{zy} line was characterized by a weak temperature dependence. This signal was attributed to the transverse AFMR in the chromium subsystem (ω_{zy}). The dependence $\omega_{zy}(H)$ had a singularity in the Γ_4–Γ_2 transition region and the extrapolated value of the "flopping" (reorientation) field $H_{fl} = 7$ kOe

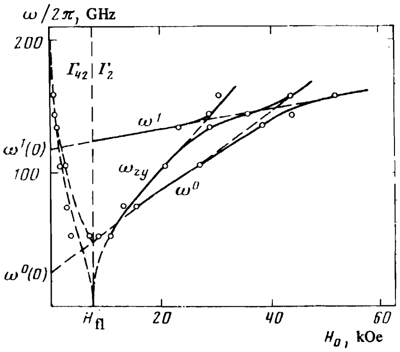

FIG. 6. Dependences of the resonance frequencies on the magnetic field; $H_0 \| a$, T=35°K.

agreed with the value obtained at this temperature from magnetic measurements in[4].

The signal denoted by ω_0 in Fig. 6 had a strong temperature dependence. The intensity of this signal fell with rising temperature. We attributed it to the paramagnetic resonance of the erbium subsystem in the ground state.

The signal denoted by ω^1 in Fig. 6 appeared only at higher temperatures ($T \sim 30-35°K$). In view of this feature, we attributed this signal to the absorption by the erbium subsystem in the first excited state (46 cm^{-1}).

It is clear from Fig. 6 that in the Γ_2 state the resonance frequencies of all three oscillations ω_{zy}, ω^0, and ω^1 are close and interact strongly. The interaction occurs not only between the oscillations of the chromium and erbium subsystems (ω_{zy} with ω^0 and ω_{zy} with ω^1) but there is also "pushing apart" of the branches at the intersection of ω^0 and ω^1 which is clearly due to the interaction within the erbium subsystem. At temperatures $T > 10°K$ this interaction is undoubtedly weak but when the spectra intersect it may also give rise to the interaction between oscillations.

It follows from Eqs. (13), (14), and (18) that the dependences $\omega^0(H)$ and $\omega^1(H)$ in the Γ_2 state are generally nonlinear since the equilibrium angles φ and β depend on the external field. However, these angles can be regarded as constant in weak fields. Then, applying Eq. (18), we find from the slopes of $\omega^n(H)$ that

$$\gamma_{yy}^0 \sin\varphi^0=3.0, \quad \gamma_{yy}^1 \sin\varphi^1=0.75. \tag{23}$$

Using the values of ω_{yy}^n from[10] (after ascertaining first that g_{zz}^n is the same for $ErCrO_3$ and $ErFeO_3$), we obtain

$$\varphi^0\cong 29°, \quad \varphi^1\cong 49°. \tag{24}$$

Thus, the canting angles of the erbium sublattices are different in the ground and excited states. Extrapolation of the dependences $\omega^0(H)$ and $\omega^1(H)$ to zero gives the activation energies

$$\omega^0(0)\cong 26\,\text{GHz}, \quad \omega^1(0)\cong 120\,\text{GHz}. \tag{25}$$

In contrast to the Γ_4 state, the initial splitting of the ground state of the Γ_2 configuration is less than in the excited state. This is also true of the components of the g factor ($g_{xx}^0 = 1.2$, $g_{xx}^1 = 4.1$[10]).

Thus, we detected and investigated a paramagnetic resonance of rare-earth ions in the ground and excited states in a magnetically ordered antiferromagnetic matrix. We investigated the influence of the interaction between the antiferromagnetic and paramagnetic subsystems on the spectra of magnetic oscillations. We found that a classical calculation of the frequencies of homogeneous oscillations and introduction of the sublattice concept for the antiferromagnetic and paramagnetic systems described well the experimental results.

We are deeply grateful to A. G. Gurevich and G. M. Nedlin for valuable discussions of our results. We are also grateful to T. A. Fomina for growing single crystals.

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Translated by A. Tybulewicz
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