Interaction of antiferromagnetic resonance modes in $DyFeO_3$ under spin reorientation conditions in a magnetic field

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An investigation of the antiferromagnetic resonance (AFMR) spectra of dysprosium orthoferrite was made with spin reorientation induced by a magnetic field up to 130 kOe applied along the antiferromagnetic axis at temperatures 78–150 K. Anomalous behavior of the AFMR modes was observed: below ~ 145 K a mode which was quasiantiferromagnetic in the absence of a field became soft. This effect was demonstrated to be due to the interaction of AFMR modes exhibiting the same symmetry of oscillations under spin reorientation conditions and the degree of interaction was governed by the applied magnetic field.

INTRODUCTION

Rare-earth orthoferrites $RFeO_3$ (where R is a rare-earth ion, Lu, or Y) have a distorted perovskite structure (with the space group D_{2h}^{16}) and antiferromagnets with a weak ferromagnetism at temperatures $T < 605$ K. In accordance with the magnetic symmetry, orthoferrites can exhibit three spin configurations: Γ_4 (when the vector $\mathbf{L}$ is directed along the **b** axis of an orthorhombic crystal and $\mathbf{M}$ vanishes), $\Gamma_2(L_z,M_x)$, and $\Gamma_4(L_x,M_z)$, where $\mathbf{M}=\mathbf{M}_1+\mathbf{M}_2$ and $\mathbf{L}=\mathbf{M}_1-\mathbf{M}_2$; $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations. At room temperature most orthoferrites exhibit the $\Gamma_4(L_x,M_z)$ configuration. Cooling results in various types of spin reorientations because of the magnetic interaction between the R^{3+} and Fe^{3+} ions: two second-order phase transitions via an intermediate phase $\Gamma_4 \rightarrow \Gamma_{42} \rightarrow \Gamma_2$ (or $\Gamma_2 \rightarrow \Gamma_{24} \rightarrow \Gamma_4$, depending on the initial state) or a first-order phase transition $\Gamma_4 \rightarrow \Gamma_1$, as found, for example, in the case of $DyFeO_3$ (detailed information on spin reorientations and a good selection of the published literature can be found in Ref. 1). Moreover, the $\Gamma_4 \rightarrow \Gamma_2$ spin reorientation can also be induced by a magnetic field applied along the **a** axis.

Investigations of the response of orthoferrites to hf radiation have revealed two antiferromagnetic resonance (AFMR) modes: quasiferromagnetic ν_1 (σ mode) and quasiantiferromagnetic ν_2 (γ mode).[2-4] According to Ref. 4, the σ mode is excited by a transverse hf field $\mathbf{h} \perp \mathbf{M}$, and the γ mode by a longitudinal hf field $\mathbf{h} \| \mathbf{M}$.

The frequencies of these modes, ν_{10} and ν_{20}, are determined in the absence of an external magnetic field in the $\Gamma_4(L_x,M_x)$ configuration by the effective anisotropy constants in the **ac** and **ab** planes, respectively. Usually the frequency of the σ mode is 1.5–2 times less than the frequency of the γ mode (at room temperature ν_{10} corresponds to radiation with wavelength ~ 1 mm, whereas ν_{20} corresponds to the wavelength ~ 0.5 mm—see Refs. 2 and 3). In the case of the $\Gamma_4 \rightarrow \Gamma_{42} \rightarrow \Gamma_2$, spin reorientation induced by a magnetic field applied along the antiferromagnetic axis (**a** axis of an orthorhombic crystal) both modes have the same symmetry and, therefore, we can observe in principle their interaction and repulsion at the crossing points. However, this spin reorientation softens $\nu_1(H)$ and increases the already large difference between the frequencies of the σ and γ modes, so that there is no need to consider the interaction of oscillations in AFMR experiments. However, in the case of $DyFeO_3$, cooling reduces ν_{20} and at $T^* \sim 150$ K the two frequencies ν_{10} and ν_{20} become comparable[3] [the reduction in ν_{20} is due to a reduction in the effective anisotropy constant in the **ab** plane which leads at $T = T_M \sim 50$ K to the Morin transition $\Gamma_4(L_x,M_2) \rightarrow \Gamma_1(L_y)$].

Therefore, in the case of dysprosium orthoferrite at temperatures $T_M < T < T^*$ we have a unique opportunity for experimental observation of the interaction of the σ and γ modes in the case of a spin reorientation induced by a magnetic field applied along the **a** axis ($\Gamma_4 \rightarrow \Gamma_{12} \rightarrow \Gamma_2$ transition). The interaction of AFMR modes was first investigated by Borovik-Romanov and Prozorova[5,6] applying a magnetic field in the plane of an antiferromagnet $MnCO_3$ exhibiting the easy-plane anisotropy. As shown in Ref. 7, in this case the degeneracy of AFMR the branches occurs in a field $(2H_E H_A)^{1/2}$, where H_E and H_A are the fields of the exchange and anisotropic interactions, respectively. In the investigations of Borovik-Romanov and Prozorova[5,6] this degeneracy was lifted by tilting the magnetic field by a small angle out of the plane; this reduced the symmetry of the oscillations and made it possible to see the AFMR modes interact and push apart, in contrast to the case when the field is exactly oriented in the plane of a crystal.

The fundamental difference between the process of spin reorientation investigated in the present study and the somewhat artificially "induced" interaction described above is that in a magnetic field $\mathbf{H} \| \mathbf{a}$ the oscillations of both modes have the same symmetry in the course of all spin orientations and it is not possible to achieve branch degeneracy (with the exception of the $H = 0$ case) by any method. A specific case of the interaction of an impurity mode with one of the AFMR modes was also detected and investigated by Kocharyan and Rudashevskiĭ.[8]

We investigated the AFMR spectra of $DyFeO_3$ in the case of a spin reorientation in a field $\mathbf{H} \| \mathbf{a}$ applied at temperatures 78–150 K. We observed an interaction between the σ and γ modes and found that, depending on the ratio of the frequencies ν_{10} and ν_{20}, one could study different cases of the interaction and the mode repulsion was determined by the external field.

THEORY

Calculations were carried out using the thermodynamic potential of orthoferrites in which we included the correc-

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tions for the interaction of the magnetic subsystems of the R^{3+} and Fe^{3+} ions.[1,9] The procedure of calculation of the equilibriun state and AFMR frequencies for this case became extremely time-consuming and tedious. However, the calculations could be simplified greatly using the results of Ref. 10, where estimates were obtained of some parameters of the R–Fe interaction in $DyFeO_3$ in the investigated range of temperatures. For example, it follows from Ref. 10 that above liquid nitrogen temperature it is not necessary to allow for the magnetic field "gain" η_x and the anisotropic part of the R–Fe interaction $\tau H_x L_z$ has to be included up to 150 K (the notation is the same as in Refs. 1, 9, and 10). It follows from an estimate of τ obtained in Ref. 10 and from the expressions in Ref. 9 that the correction to the Dzyaloshinskii interaction due to the magnetic R ions can be ignored.

One of the important results of Ref. 10 is the conclusion that the thermodynamic potential must include the term responsible for the susceptibility along the antiferromagnetic axis all the way up to liquid nitrogen temperature. Consequently, during spin reorientation in a magnetic field the vectors **M** and **L** are not perpendicular to one another: $(\mathbf{M}\cdot\mathbf{L})\neq 0$ and an energy gap appears in the spectrum of a quasiferromagnetic AFMR mode in the field in which the spin reorientation is completed.[11]

Subject to all these comments, the thermodynamic potential is found to be renormalized by the R–Fe interaction in a field $\mathbf{H}\|\mathbf{a}$ and at temperatures $T\gtrsim 78$ K this potential is described by

$$\Phi(\mathbf{M},\mathbf{L})=\Phi_0(\mathbf{L}^2)+\frac{1}{2}B\mathbf{M}^2+\frac{1}{2}D(\mathbf{ML})^2+d(M_xL_z-M_zL_x)$$
$$-M_xH_x-\tau H_xL_z+\frac{1}{2}\sum_{i=1}^{3}a_iL_i^2$$
$$+\frac{1}{4}\sum_{i,j=1}^{3}a_{ij}L_i^2L_j^2, \quad x,\ y,\ z=1,2,3. \tag{1}$$

The only model assumption used in the calculations is the constancy of the antiferromagnetic vector: $\mathbf{L}^2\neq L_0^2$.

In calculations of the AFMR frequencies it is necessary to solve the equations of motion. The Landau–Lifshitz equations are unsuitable for this case because it is necessary to assume the condition $(\mathbf{M}\cdot\mathbf{L})=0$, which is in conflict with the available experimental results.[10] Therefore, we shall use more general thermodynamic equations of motion in the exchange approximation[12,13]:

$$\Delta\dot{\mathbf{M}}=\gamma_1[\mathbf{M}_0\mathbf{H}_M]+\gamma_2[\mathbf{L}_0\mathbf{H}_L],$$
$$\Delta\dot{\mathbf{L}}=\gamma_2[\mathbf{L}_0\mathbf{H}_M]+\gamma_3[\mathbf{M}_0\mathbf{H}_L], \tag{2}$$

where $\Delta\mathbf{M}=\mathbf{M}-\mathbf{M}_0$, $\Delta\mathbf{L}=\mathbf{L}-\mathbf{L}_0$; $\mathbf{M}_0$ and $\mathbf{L}_0$ are the equilibrium values of the ferromagnetic and antiferromagnetic vectors;

$$\mathbf{H}_M=\frac{\partial\Delta\Phi}{\partial\Delta\mathbf{M}}, \quad \mathbf{H}_L=\frac{\partial\Delta\Phi}{\partial\Delta\mathbf{L}}, \quad \Delta\Phi=\frac{1}{2}\sum_{i,j=1}^{6}\alpha_{ij}\Delta X_i\Delta X_j;$$

$\alpha_{ij}=\partial^2\Phi/\partial X_i\partial X_j$ is the stability matrix of the thermodynamic potential; $\mathbf{X}=\{\mathbf{M},\mathbf{L}\}$ is a six-dimensional vector; γ_i are kinetic coefficients. It is shown in Ref. 11 that γ_1 and γ_2 for $YFeO_3$ differ from one another and from $\gamma_0=ge/2mc$ by no more than 1–2%. This is due to the fact that the Fe^{3+} ion is in the S state and the anisotropy of the γ factors cannot be significant. It is also shown there that the condition $\mathbf{L}^2=L_0^2$ necessarily yields $\gamma_3=0$. Therefore, in our calculations we shall use the approximation $\gamma_1\sim\gamma_2\sim\gamma_0$, $\gamma_3=0$. Substituting the solution in the form $\Delta\mathbf{X}=\Delta\mathbf{X}_0e^{i\omega t}$ into Eq. (2), we obtain the following secular equation for AFMR frequencies:

$$\det\|\hat{\gamma}\hat{\alpha}-i\omega\hat{E}\|=0, \tag{3}$$

where $\hat{\gamma}$ is the dynamic matrix[12]; $\hat{\alpha}$ is the stability matrix; $\hat{E}$ is a unit matrix.

Expanding Eq. (3) in the case when $H\leqslant H_r$, where H_r is the field in which the $\Gamma_4\to\Gamma_{42}\to\Gamma_2$, spin reorientation is completed, we obtain an expression for the frequencies of both AFMR modes. We shall write down this expression in a form known from the classical theory of oscillations of interacting branches[14]:

$$\nu_{1,2}^2=\frac{1}{2}\{\nu_\sigma^2+\nu_\gamma^2\pm[(\nu_\gamma^2-\nu_\sigma^2)^2+\rho^2]^{1/2}\}, \tag{4}$$

where

$$(2\pi\nu_\sigma/\gamma_0)^2=(\Delta_1+3A_1\sin^2\varphi)\cos^2\varphi+H^2(\sin^2\varphi-\beta),$$
$$(2\pi\nu_\gamma/\gamma_0)^2=\Delta_2+A_2\sin^2\varphi+H^2(1+\beta)^2\cos^2\varphi-\beta H^2,$$
$$\rho=\pi^{-1}\nu_\sigma\gamma_0H(1+\beta)\cos\varphi;$$

Δ_1 and Δ_2 are the energy gaps in the AFMR spectrum in the absence of a magnetic field, corresponding to the frequencies ν_{10} and ν_{20}:

$$\Delta_1\equiv(2\pi\nu_{10}/\gamma_0)^2=\chi_\perp^{-1}L_0^2[a_3-a_1+(a_{13}-a_{11})L_0^2],$$
$$\Delta_2\equiv(2\pi\nu_{20}/\gamma_0)^2=\chi_\perp^{-1}L_0^2[a_2-a_1+(a_{12}-a_{11})L_0^2]+H_D^2;$$

A_1 and A_2 are governed by the biquadratic anisotropy constants in the **ac** and **ab** planes, respectively:

$$A_1=\chi_\perp^{-1}L_0^4(a_{11}+a_{33}-2a_{13}), \quad A_2=\chi_\perp^{-1}L_0^4(a_{11}-a_{12}+a_{23}-a_{13}),$$
$$H_D=dL_0, \quad \chi_\perp^{-1}=B, \quad \chi_\parallel^{-1}=B+DL_0^2, \quad \beta=1-\chi_\parallel/\chi_\perp,$$

φ is the angle between the vector **L** and the **a** axis in the case of a spin reorientation in the **ac** plane, found from the equations of state

$$\frac{\partial\Phi(\mathbf{M},\varphi)}{\partial\mathbf{M}}=0, \quad \frac{\partial\Phi(\mathbf{M},\varphi)}{\partial\varphi}=0.$$

Direct substitution readily shows that in the absence of a magnetic field ($\varphi=0$) or when $H=H_r$ ($\varphi=\pi/2$), i.e., in those cases when ρ responsible for the interaction of modes in Eq. (4) vanishes, the values of ν_σ and ν_γ govern the frequencies of the σ and γ modes.

We shall find the symmetry of the oscillations using the results of Ref. 15, where small oscillations of a spin system are classified in accordance with the irreducible representations of the magnetic symmetry group of a crystal. For example, according to Ref. 15, in the Γ_4 phase the symmetry of oscillations of the σ mode corresponds to the Γ_{23} representation, the symmetry of the γ mode corresponds to the Γ_{14} representation, and in the Γ_2 phase we have the Γ_{34} and Γ_{12} representations. A table of irreducible representations of the group D_{2h}^{16} (see Refs. 1 and 15) readily demonstrates that in the Γ_4 phase the σ mode is governed by small oscillations of

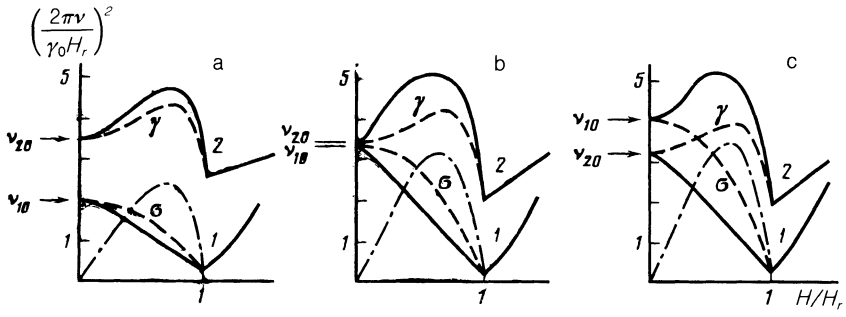

FIG. 1. Squares of the frequencies of the quasiferromagnetic (1) and quasiantiferromagnetic (2) AFMR modes versus the magnetic field **H**||**a**, plotted using Eq. (4) and different ratios of ν_{10} to ν_{20}. Here, σ denotes $\nu_\sigma^2(H)$ and γ denotes $\nu_\gamma^2(H)$; the chain curve represents $\rho(H)$. For clarity, the curves are plotted using dimensionless coordinates normalized to the reorientation transition field H_r (explanations in text).

the components M_x, M_y, and L_z and the γ mode is determined by the components L_x, L_y, and M_z. The same result can be obtained in solving the secular equation of Eq. (3), which in simple cases splits into products of two blocks each containing "admixtures" of oscillations of the components of the vectors **M** and **L** corresponding to one or the other AFMR mode.

We thus find that in the case of a spin reorientation in a field **H**||**a** the symmetry of the oscillations of both modes in the Γ_{42} phase is governed by the Γ_{1234} representation and two components of the vectors **M** and **L** participate in each AFMR branch; the separation of variables (decoupling of the modes) occurs only for $H=0$ and $H=H_r$.

We shall consider separately three cases.

1. The frequency of the γ mode is considerably higher than the frequency of the σ mode in the absence of a magnetic field, i.e., $\nu_\gamma(0) \gg \nu_\sigma(0)$ (or $\nu_{20} \gg \nu_{10}$). This situation is encountered in practically all orthoferrites at room temperature. Figure 1a shows plots of the dependences $\nu_1^2(H)$ and $\nu_2^2(H)$ for a spin reorientation in a field **H**||**a**, deduced from Eq. (4). This figure includes also $\nu_\sigma^2(H)$ and $\nu_\gamma^2(H)$, and also $\rho(H)$. In spite of the fact that the σ and γ modes are separated by a considerable frequency interval, the high strength of the interaction $\rho(H)$ (chain curve; all the curves in Fig. 1 are plotted on the same scale) in the $0<H<H_r$ case the modes are repelled so that $\nu_2(H)$ lies somewhat above $\nu_\gamma(H)$, and $\nu_1(H)$ is slightly below $\nu_\sigma(H)$. (All the parameters needed to plot Fig. 1 were deduced from the experimental data of Ref. 10.)

2. The frequencies ν_{20} and ν_{10} are practically equal, but ν_{20} still exceeds ν_{10}. This case is observed for $DyFeO_3$ at ~ 150 K (Refs. 3 and 10). The results of frequency calculations are given in Fig. 1b, which demonstrates how a strong interaction $\rho(H)$ results in considerable repulsion of the modes. In both cases under discussion after completion of spin reorientation ($H \geqslant H_r$) the oscillation mode of each of the branches of the AFMR spectrum is the same as the corresponding mode in the absence of a magnetic field, whereas in the course of reorientation ($0<H<H_r$) the oscillations are mixed: the σ mode includes a contribution of oscillations of the γ mode and vice versa, the magnitude of the "admixture" being governed by the interaction $\rho(H)$. Therefore, changes in the nature of the oscillation modes in the course of spin reorientation can be represented by $\sigma \to \sigma\gamma \to \sigma$, $\gamma \to \gamma\sigma \to \gamma$. We shall write down these changes using the above-mentioned classification of oscillations in accordance with the symmetry group representations:

$$\nu_1\colon\ \Gamma_{23} \to \Gamma_{1234} \to \Gamma_{34},$$
$$\nu_2\colon\ \Gamma_{14} \to \Gamma_{1234} \to \Gamma_{12}.$$

3. The most interesting situation should be observed in the case when $\nu_{20}<\nu_{10}$ (Fig. 1c), which applies to dysprosium orthoferrite at temperatures $T_M<T<T^*$. Now $\nu_\sigma(H)$ and $\nu_\gamma(H)$ intersect at some value of the field and this, together with the strong interaction $\rho(H)$ results in qualitative changes in the behavior of the branches of the AFMR spectrum. As the magnetic field **H** is increased, an oscillation of different symmetry is mixed into each of the modes so that in the field corresponding to completion of the spin reorientation process the admixture becomes the main oscillation, the modes are apparently transposed, and the changes in the nature of oscillations occur in accordance with the scheme $\sigma \to \sigma\gamma \to \gamma$, $\gamma \to \gamma\sigma \to \sigma$, or

$$\nu_1\colon\ \Gamma_{23} \to \Gamma_{1234} \to \Gamma_{12},$$
$$\nu_2\colon\ \Gamma_{14} \to \Gamma_{1234} \to \Gamma_{34}.$$

Therefore, in the case of $DyFeO_3$ in the temperature range 50–140 K the result of a spin reorientation in a field **H**||**a** is softening of the quasiantiferromagnetic mode $\nu_2(H)$, whereas the quasiferromagnetic mode $\nu_1(H)$ becomes hard. This paradoxical situation appears mainly because of inexact terminology as both modes are so identified in zero external field. In the presence of a magnetic field the oscillation modes have the same symmetry and which of them becomes softer (and which does not) can be determined only if we know the values of ν_{10}, ν_{20} and $\nu_1(H_r)$, $\nu_2(H_r)$. As already mentioned, in the case of orthoferrites the value of ν_{20} is usually much higher than ν_{10} in a wide range of magnetic fields and temperatures, which corresponds to the first case discussed above so that the somewhat inaccurate names for the modes in the case of a spin reorientation in a magnetic field, adopted for the zero-field (temperature) spin reorientation, cause no misunderstanding.

EXPERIMENTS

We investigated the AFMR spectra of $DyFeO_3$ using a direct-amplification spectrometer in the frequency range 75–400 GHz ($\lambda = 4$–0.75 mm) using magnetic fields 3–130 kOe at temperatures from 78 to 150 K. The radiation sources were backward-wave tubes. The magnetic field was generated in the "Solenoid" device at the Division of High Magnetic Fields of the Institute of General Physics, USSR Academy of Sciences.[16]

In these experiments we determined the positions of the absorption lines of the incident electromagnetic radiation as a function of a magnetic field applied to a sample in the **H**||**a** orientation at various temperatures. A stabilization system made it possible to keep the temperature constant to within ± 0.5 K in each experiment. The experimental method and

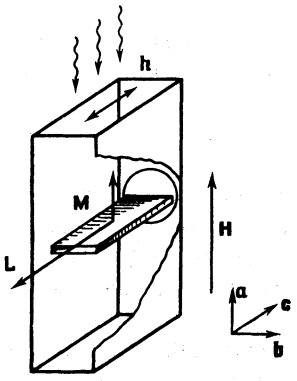

FIG. 2. Section of a waveguide containing a sample and relative orientations of **M**, **L**, **H**, and **h** when $H \geqslant H_r$. The directions of the crystallographic axes of a single crystal are also shown.

the exact setting of the orientation of a crystal in a magnetic field were described in detail in Ref. 11.

A $DyFeO_3$ single crystal was cut from a boule grown by the floating zone method using radiation heating[17]; it was a plate of dimensions $6\times3\times0.5$ mm oriented (by an x-ray diffraction method) at right-angles (to within $\pm$ 1°) to the **a** axis of the orthorhombic crystal.

The relative orientations and positions of the magnetic field **H**, of the sample in the waveguide, and of the incident completion electromagnetic radiation with the field **h** are shown in Fig. 2. The experimental geometry was such that after completion of the spin reorientation process the weak ferromagnetic moment **M** was parallel to the magnetic field **H** and to the direction of propagation of the incident radiation. Therefore, the γ mode was not excited in fields $H \geqslant H_r$, because this would require a component of **h** parallel to **M**, whereas in our case we had **h**$\perp$**M**. This condition allowed us to determine directly the symmetry of oscillations of each of the two experimentally observed branches of the AFMR spectrum after completion of the spin reorientation process (in fields $H > H_r$ only the σ mode was excited).

The experimental results are presented in Fig. 3. At $T = 146$ K (Fig. 3a) the inequality $\nu_{20} > \nu_{10}$ was obeyed and the nature of oscillations of each of the AFMR modes was

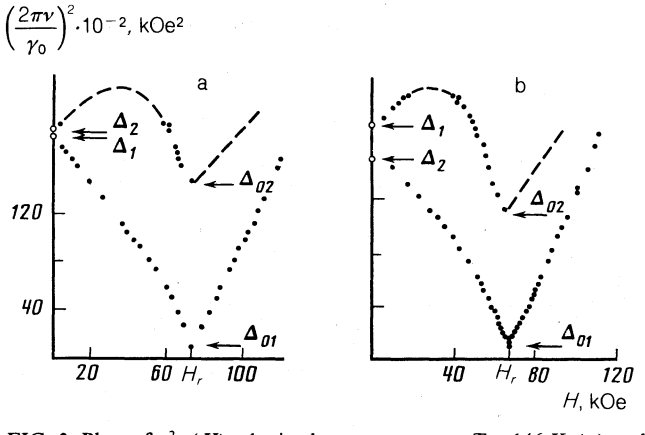

FIG. 3. Plots of $\nu_{1,2}^2(H)$, obtained at temperatures $T = 146$ K (a) and $T = 128$ K (b). Here, Δ_1 corresponds to ν_{10}^2 and Δ_2 corresponds to ν_{20}^2; Δ_{01} and Δ_{02} are the energy gaps in the field $H = H_r$. In the range $H > H_r$ only the σ mode is excited. The results taken from Ref. 3 are represented by open circles (○).

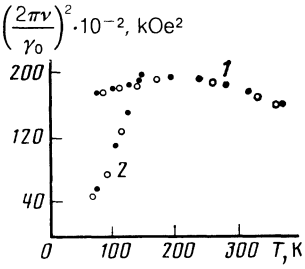

FIG. 4. Plots of $\nu_{10}^2(T)$ (1) and $\nu_{20}^2(T)$ (2). The results of the present study are represented by black dots (●) and the results taken from Ref. 3 are denoted by open circles (○).

preserved, in full agreement with the second case discussed above, as demonstrated by the "continuity" of the σ mode. Figure 3b illustrates the last and most interesting case of changes in the mode symmetry. We can clearly see that the branch $\nu_1(H)$ (representing the σ mode in the absence of the field) was not excited in the range $H > H_r$ (i.e., it reduced to the γ mode) and, conversely, the quasiantiferromagnetic (in $H = 0$) mode $\nu_2(H)$ because quasiferromagnetic on completion of spin reorientation. An indirect confirmation of the theory was the identity, at different temperatures, of the values of ν_{10} and ν_{20} found by extrapolation of $\nu_1(H)$ and $\nu_2(H)$ to zero magnetic field and the corresponding values given in Ref. 3, where the temperature dependences $\nu_{10}(T)$ and $\nu_{20}(T)$ were determined (Fig. 4) (open circles are used in Figs. 3 and 4 for the results reported in Ref. 3). The symmetry of oscillations of each of the AFMR modes was determined uniquely in Ref. 3 by the **h** polarization of the incident hf radiation. Therefore, there was no doubt that below $\sim$140 K in the course of a spin reorientation in the field **H**$\|$**a** it was the $\nu_2(H)$ mode that became softer.

It is quite clear from Fig. 3 that at the point of completion of the spin reorientation process the spectra of both modes exhibited large energy gaps Δ_{01} and Δ_{02} in full agreement with the theoretical calculations (Fig. 1).

Quasiferromagnetic mode whose frequencies were nonvanishing in the course of a spin reorientation transition in a magnetic field was first reported in Ref. 18. It was shown in Refs. 11 and 18 that Δ_{01} was mainly due to nonconservation of the magnetic moments of the sublattices in the antiferromagnet, i.e., it was represented by the parallel susceptibility $\chi_\parallel$. A detailed analysis of this effect is outside the scope of the present paper. However, it should be mentioned that Δ_{02} decreased much more rapidly with temperature than did Δ_{01} (see also Ref. 10). This was due to "activation" of the R–Fe interaction as a result of cooling and reduction in the anisotropy constant in the **ab** plane, which in the final analysis resulted in the Morin transition in $DyFeO_3$. Extrapolation of the dependences $\Delta_{01}(T)$ and $\Delta_{02}(T)$ to low temperatures showed that at $T \sim 60$–70 K the value of Δ_{02} became less than Δ_{01}. Then, $\nu_\gamma(H)$ in the range of fields $0 < H < H_r$ was always less than $\nu_\sigma(H)$ and we should go back to the second case, when the nature of oscillations of each of the AFMR modes on completion of spin reorientation was the same as the nature of oscillations in $H = 0$, except that now $\nu_2(H)$ should lie below $\nu_1(H)$ and the mode $\nu_1(H)$ which became softer as a result of this phase transition was much higher ($\Delta_{01} > \Delta_{02}$), than the quasiferromagnetic mode. It follows

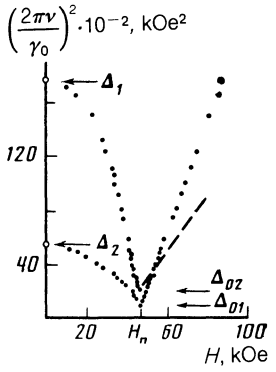

FIG. 5. Plots of $\nu_{1,2}^2(H)$ at $T = 78$ K.

from the calculations of Ref. 19 that a reduction in the frequency $\nu_2(H_r)$ demonstrates that $DyFeO_3$ may exhibit a new unusual spin reorientation phase transition which is described by a phase diagram with a tetracritical point in the (T,H) plane.

The effects described above ($\Delta_{02} < \Delta_{01}$ and the unusual spin reorientation) should be observed also at $T < 78$ K, but our apparatus was not suitable for such measurements. By way of illustration, we plotted in Fig. 5 the spectra of both AFMR modes at $T = 78$ K. It is quite clear that Δ_{01} and Δ_{02} have similar values, but Δ_{02} is again slightly greater than Δ_{01}, and we are still within the framework of the last of the cases discussed above.

The R–Fe interaction results in coupling of oscillations of the magnetic moments of the dysprosium and iron ion subsystems. In the calculations such a coupling is allowed only by the thermodynamic potential renormalized by the R–Fe interaction.

The model used in the present investigation is essentially a quasistatic approximation, when the magnetic moments of Dy^{3+} ions follow instantaneously the moments of the Fe^{3+} ions. This model is justified if $\nu_i^R \gg \nu_{1,2}$, where ν_i^R are the resonance frequencies of the R subsystem, corresponding to different transitions between energy levels of the ground state multiplet of R^{3+} split by the crystal and exchange fields. The spectrum of the Dy^{3+} ion consists of doublets with the energies $E_1 = 0$, $E_2 = 52$ cm^{-1}, $E_3 = 147$ cm^{-1}, etc.[20] The exchange and dipole R–Fe interactions result in splitting of the doublets of Dy^{3+} by an amount ~ 1 cm^{-1} (which corresponds to $\nu = 30$ GHz). Therefore, the frequencies ν_i^R associated with the $E_1 \leftrightarrow E_2$, $E_1 \leftrightarrow E_3$, exceed considerably $\nu_{1,2}$, which partly justifies the approximation we have adopted. It is shown in Ref. 3 that at temperatures $T \gtrsim 50$ K we can again ignore the contribution due to the splitting of the doublets. However, this result is obtained in the absence of an external magnetic field. In a field $H = 50$ kOe (we recall that in the case of $DyFeO_3$ at $T = 78$ K the reorientation-completing field is $H_r = 46$ kOe) the splitting of the lower doublet reaches ~ 15 cm^{-1} ($\nu = 440$ GHz).

Since an increase in the field from zero to H_r reduces the value of $\nu_{1,2}$ from 380 to 50 GHZ, whereas ν_i^R conversely increases from 30 to 440 GHz, it follows that in principle we should be able to observe the interaction between the ν^R and $\nu_{1,2}$ modes. However, it has not been possible to detect this in experiments at ν^R. Clearly, this is due to the much lower intensity of the rare-earth modes compared with intensities of the $\nu_{1,2}$ modes. The result obtained supports the above model which does not allow explicitly for the dynamics of the R subsystem.

CONCLUSIONS

The AFMR spectra of $DyFeO_3$ have been studied during the spin reorientation transition induced by a magnetic field $\mathbf{H}\|\mathbf{a}$ at temperatures in the range 78–150 K at frequencies 75–400 GHz in fields up to 130 kOe. Anomalous behavior of the AFMR modes was observed and it was manifested by the fact that below ~ 145 K the transition softened the mode which was quasiantiferromagnetic in the absence of the field. The observed effect was attributed to a strong interaction and repulsion between the AFMR modes characterized by the same symmetry of the oscillations in the course of the spin reorientation and the degree of this interaction was governed by the magnetic field. It was established that, depending on the ratio of the frequencies of the two modes, the situations were fundamentally different in the fields $H = 0$ and $H = H_r$: in the former case the type of oscillations of each of the AFMR modes was identical, after completion of the spin orientation, with the type of the corresponding mode in $H = 0$, whereas in the other case there was an exchange of symmetries: the mode which was quasiferromagnetic in the absence of the field, became quasiantiferromagnetic on completion of the spin reorientation transition and, conversely, the γ mode became the σ mode, which resulted in its softening in the field corresponding to completion of the reorientation process.

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