Frozen rare earth sublattice and the rf magnetic properties of gadolinium orthoferrite

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The submillimeter spectra of $GdFeO_3$ crystals have been found to contain a quasiferromagnetic and a quasiantiferromagnetic mode of the antiferromagnetic resonance (AFMR). A study is made of the temperature dependence of their resonance frequencies $\nu_{1,2}(T)$, their linewidths $\Gamma^{(1,2)}(T)$, and the contributions $\Delta\mu_{1,2}(T)$ of these modes to the static magnetic permeability. For $T<40$ K an unusual increase of the AFMR frequencies is observed. It is shown theoretically that the observed behavior of $\nu_{1,2}(T)$ is due to the interaction of spin oscillations of the Fe^{3+} ions with the Gd sublattice, which is "frozen" at these frequencies.

I. INTRODUCTION

The magnetic properties of the rare earth orthoferrites $RFeO_3$, where R is a rare earth ion, are largely determined by the interaction between the antiferromagnetically ordered Fe subsystem and the paramagnetic (for $T>4$ K) rare earth subsystem.[1] In particular, this interaction is the cause of the various orientational phase transitions observed in these compounds.

It is accordingly of interest to elucidate the influence of the R subsystem on the rf magnetic properties of the orthoferrites, particularly on the behavior of the antiferromagnetic resonance (AFMR) frequencies. An important role should be played by the rare earth modes, whose frequencies, which are determined by the splitting of the ground multiplet of the rare earth ion in the crystalline and external fields and in the effective field due to the Fe subsystem, can lie above or below the AFMR frequencies. The behavior of the AFMR frequencies with decreasing temperature depends on which of these two possiblities is realized and can differ qualitatively in different orthoferrites, as the data of Refs. 2–5 attest.

In this paper we study the rf properties of gadolinium orthoferrite $GdFeO_3$. This orthoferrite has the feature that the splitting of the ground state of the Gd^{3+} ion at $H=0$ is extremely small (<0.1 cm^{-1}), i.e., the eigenfrequencies of the Gd sublattice are substantially lower than the AFMR frequencies in the Fe subsystem (~ 10 cm^{-1}). Our studies revealed that this circumstance leads to unusual behavior of the AFMR frequencies, viz., they show an increase with decreasing temperature rather than the expected decrease due to the decrease of the effective anisotropy constants on account of the Gd–Fe interaction.[6] This behavior of the AFMR frequencies is due to a peculiar effect in the dynamics of the Gd sublattice and its interaction with the Fe subsystem at frequencies $\nu \approx 10$ cm^{-1}. We show that at these frequencies the Gd sublattice can be considered "frozen" and the deviations of its magnetic moments from the equilibrium position can be neglected. In this case the oscillations of the spins of the Fe subsystem actually occur only in a static effective field due to the Gd subsystem. This dynamical behavior of the Fe and R subsystems is similar in many respects to the "magnetoelastic gap" or "frozen-lattice" effect.[7,8]

2. EXPERIMENT

Single crystals of $GdFeO_3$ were grown by float zoning with radiative heating.[9] The samples were cut from the original boules in the form of plane-parallel a-cut slabs with transverse dimensions ~ 1 cm and a thickness ~ 1 mm. The surfaces of the slabs were then optically processed.

Using the Epsilon submillimeter backward-wave-tube spectrometer,[10] we measured the transmission spectra of $GdFeO_3$ over the frequency range 150–1000 GHz at temperatures $T=4.2$–300 K in zero external magnetic field. We detected two AFMR modes: a quasiferromagnetic and a quasiantiferromagnetic mode, which are excited by rf magnetic fields $\mathbf{h}\perp c$ and $\mathbf{h}\parallel c$, respectively. By processing the transmission spectra by a technique described in Ref. 11, we determined the temperature dependence of the parameters of these modes: their resonance frequencies $\nu_{1,2}(T)$, their linewidths $\Gamma^{1,2}(T)$, and the contributions $\Delta\mu_{1,2}(T)$ of these modes to the static magnetic permeability. Figure 1 shows the temperature dependence of the resonance frequencies and linewidths of the two AFMR modes. The contribution to the static magnetic permeability was found to be practically independent of temperature at all the temperatures studied, and $\Delta\mu_y=\Delta\mu_1\approx\Delta\mu_z=\Delta\mu_2\approx 0.0008$.

Let us turn our attention to the considerable growth in the frequencies of the two AFMR modes for $T<40$ K (Fig. 1a). This behavior of the AFMR frequencies is somewhat unusual in view of the known static properties[6,12] of $GdFeO_3$. According to Ref. 12, at all temperatures $T<T_{N1}=650$ K the spin configuration of the iron ions in this orthoferrite is $\Gamma_4(G_xF_z)$, with the weak ferromagnetism vector **F** along the c axis and the antiferromagnetism vector **G** along the a axis. A study of the static magnetic properties of $GdFeO_3$ has revealed[6] that the anisotropy of the Ge–Fe interaction in this crystal leads to an appreciable decrease of the effective anisotropy constant K_{ac}^{eff} in the ac plane as the temperature is lowered. One would therefore expect a decrease in the frequency of the quasiferromagnetic mode of the AFMR at low temperatures, in accordance with the usual relation $\omega_1=\gamma(2H_EH_{ac}^{eff})^{1/2}$ (Fig. 1), where γ is the gyromagnetic ratio, H_E is the exchange field, and $H_{ac}^{eff}=K_{ac}^{eff}/M_0$ is the effective anisotropy field. (This relation holds very well in $TmFeO_3$, for example.)[13] We do not

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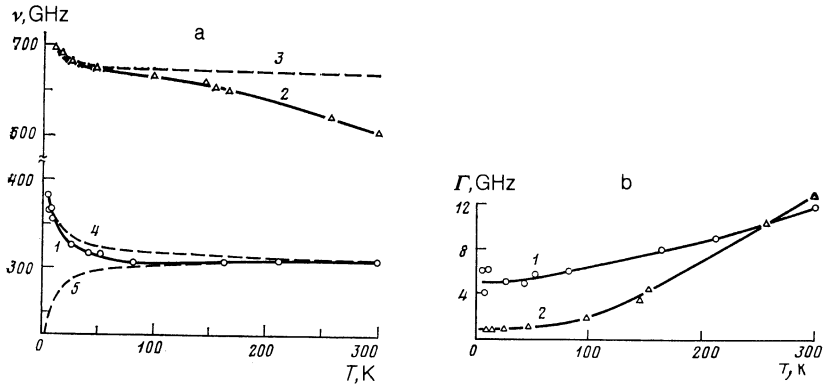

FIG. 1. Temperature dependence of the parameters of the AFMR modes in $GdFeO_3$: resonance frequencies $\nu_{1,2}$ (a) and linewidths $\Gamma^{(1,2)}$ (b) for the quasiferromagnetic (experimental curve 1) and quasiantiferromagnetic (experimental curve 2) modes of the AFMR. Curves 3 and 4 were calculated using Eq. (16'). Curve 5 was calculated by the formulas $\nu_1 = (\gamma/2\pi)[2H_E H_{ac}^{\text{eff}}(T)]^{1/2}$, where $H_{ac}^{\text{eff}}(T)$ was taken from Ref. 6.

see this in $GdFeO_3$, however. The reason for this diverse behavior, as we shall see, is that the dynamic susceptibility of the Gd subsystem in the frequency range investigated is much smaller than the static susceptibility, and the Gd subsystem is therefore in effect a frozen sublattice which creates only a static effective field acting on the Fe subsystem.

3. THEORY

To obtain a consistent description of the observed dynamical properties of $GdFeO_3$, let us consider the coupled oscillations of the antiferromagnetically ordered Fe subsystem and the paramagnetic Gd subsystem, which is polarized along the c axis on account of the Gd–Fe interaction. This interaction will be described in the effective (molecular) field approximation, which is widely used for describing the properties of orthoferrites.[1] The Hamiltonian of the Gd–Fe interaction is written[6]

$$\mathcal{H}_{\text{R-Fe}}^{\pm} = g_J \mu_B \mathbf{J}_R^{\pm}(a\mathbf{F} + \hat{P}^{\pm}\mathbf{G}) \equiv g_J \mu_B \mathbf{J}_R^{\pm}\mathbf{H}_{\text{eff}}. \quad (1)$$

This Hamiltonian has been shown[6] to give a good description of the static magnetic properties observed in $GdFeO_3$. Here $g_J = 2$, μ_B is the Bohr magneton, $\hbar \mathbf{J}_R^{\pm}$ is the operator for the total mechanical angular momentum of the Gd^{3+} ion,

$$\mathbf{F} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0, \quad \mathbf{G} = (\mathbf{M}_1 - \mathbf{M}_2)/2M_0$$

are the ferromagnetism and antiferromagnetism vectors of the Fe subsystem, which is treated in the two-sublattice ($\mathbf{M}_1$ and $\mathbf{M}_2$) approximation, $M_0 = |\mathbf{M}_1| = |\mathbf{M}_2|$ are the specific magnetic moments of the sublattices, a is the interaction constant for the isotropic Gd–Fe exchange,

$$\hat{P}^{\pm} = \begin{vmatrix} 0 & 0 & p_{xz} \\ 0 & 0 & \pm p_{yz} \\ p_{zx} & \pm p_{zy} & 0 \end{vmatrix} \quad (2)$$

is the matrix of the anisotropic Gd–Fe interaction, and the $\pm$ signs refer to the different crystallographically inequivalent positions of the Gd^{3+} ions.

To describe the dynamics of the magnetic moments of the Fe subsystem, we use the Landau-Lifshitz equations in which, with the aid of (1), we have introduced the effective fields acting on the Fe subsystem on account of the Gd subsystem:

$$\begin{aligned} \frac{1}{\gamma}\dot{\mathbf{F}} &= -[\mathbf{F}\mathbf{H}_F] - [\mathbf{G}\mathbf{H}_G] + \frac{1}{\gamma}(\alpha[\mathbf{F}\dot{\mathbf{F}}] + \beta[\mathbf{G}\dot{\mathbf{G}}]), \\ \frac{1}{\gamma}\dot{\mathbf{G}} &= -[\mathbf{G}\mathbf{H}_F] - [\mathbf{F}\mathbf{H}_G] + \frac{1}{\gamma}(\alpha[\mathbf{G}\dot{\mathbf{F}}] + \beta[\mathbf{F}\dot{\mathbf{G}}]), \end{aligned} \quad (3)$$

where the square brackets denote the vector (cross) product, α and β are damping coefficients,[14]

$$\begin{aligned} \mathbf{H}_F &= -M_0^{-1}(\partial\Phi_{\text{Fe}}/\partial\mathbf{F} + a\mathbf{F}), \\ \mathbf{H}_G &= -M_0^{-1}(\partial\Phi_{\text{Fe}}/\partial\mathbf{G} + \mathbf{F}_R\hat{P}_F + \mathbf{C}_R\hat{P}_C), \end{aligned} \quad (4)$$

$$\begin{aligned} \Phi_{\text{Fe}}(\mathbf{F}, \mathbf{G}) = {}& {}^1/_2 A\mathbf{F}^2 + {}^1/_2 K_{ac}^{\text{Fe}} G_z^2 + {}^1/_2 K_{ab}^{\text{Fe}} + {}^1/_4 K_2 G_z^4 + {}^1/_4 K_2' G_y^4 \\ & + {}^1/_2 K_2'' G_z^2 G_y^2 - d(F_x G_z - F_z G_x) - M_0 \mathbf{F}\mathbf{H} \end{aligned} \quad (5)$$

is the thermodynamic potential of the Fe subsystem (see, e.g., Ref. 1), $\mathbf{F}_R = (\mathbf{M}_R^+ + \mathbf{M}_R^-)/2$ and $\mathbf{C}_R = (\mathbf{M}_R^+ - \mathbf{M}_R^-)/2$ are the dynamical variables of the R subsystem, $\mathbf{M}_R^{\pm} = -Ng_J\mu_B\langle \mathbf{J}_R^{\pm} \rangle$ are the average specific magnetic moments of the two sublattices of the R subsystem, N is the number of rare earth ions per gram, $P_F = (P^+ + P^-)/2$, and $P_C = (P^+ - P^-)/2$.

The dynamics of the paramagnetic Gd subsystem is also described by equations of type (3), in which we have introduced a longitudinal relaxation in addition to the transverse relaxation (see, e.g., Ref. 15). We neglect the small splitting of the ground multiplet of the Ge^{3+} ion in the crystalline field ($\sim E < 0.1\ \text{cm}^{-1}$) and consider only the external field and the internal effective field, which is parallel to the c axis in a $\Gamma_4(G_xF_z)$ phase. Writing the dynamical variables in the form

$$\mathbf{F} = \mathbf{F}^0 + \Delta\mathbf{F}, \quad \mathbf{G} = \mathbf{G}^0 + \Delta\mathbf{G}, \quad \mathbf{F}_R = \mathbf{F}_R^0 + \Delta\mathbf{F}_R, \quad \mathbf{C}_R = \mathbf{C}_R^0 + \Delta\mathbf{C}_R,$$

where $\mathbf{F}^0$, $\mathbf{G}^0$, $\mathbf{F}_R^0$, and $\mathbf{C}_R^0$ are the average (statistical) values, and linearizing the equations of motion, we express the variables of the R subsystem in terms of $\Delta\mathbf{F}$ and $\Delta\mathbf{G}$ and the external magnetic field $\mathbf{h} = \mathbf{h}_0 \exp(i\omega t)$:

$$\Delta\mathbf{F}_R = \hat{\chi}_R(\omega)(\mathbf{h} + a\Delta\mathbf{F} + \hat{P}_F\Delta\mathbf{G}), \quad \Delta\mathbf{C}_R = \hat{\tilde{\chi}}_R(\omega)\hat{P}_C\Delta\mathbf{G}. \quad (6)$$

The dynamic susceptibilities $\hat{\chi}_R$ and $\hat{\tilde{\chi}}_R$ govern the response of the Gd subsystem to the variable effective fields, which cause a polarization of the ferromagnetic ($\Delta\mathbf{F}_R$) and antiferromagnetic ($\Delta\mathbf{C}_R$) types. In the Γ_4 phase they are of the form

$$\hat{\chi}_R = \begin{vmatrix} \chi_R^{xx} & \chi_R^{xy} & 0 \\ \chi_R^{yx} & \chi_R^{yy} & 0 \\ 0 & 0 & \chi_R^{zz} \end{vmatrix}, \quad (7)$$

where

$$\chi_R^{xx}=\chi_R^{yy}=\chi_R^0(1+i\delta)\,[(1+i\delta)^2-(\omega/\omega_R)^2]^{-1},$$
$$\chi_R^{xy}=-\chi_R^{yx}=[i\omega/\omega_R(1+i\delta)]\chi_R^{xx},\qquad \delta=\omega_r\omega/\omega_R^2, \tag{8}$$
$$\chi_R^{zz}=\chi_R^0(1+i\omega/\omega_{r1}')^{-1},\qquad \omega_{r1}'=\omega_{r1}(1+\Theta_F/T),$$

$\chi_R^0 = C/(T+\Theta_F)$ is the paramagnetic susceptibility of the Ge subsystem,

$$C=Ng_J^2\mu_B^2J_R(J_R+1)/3k_B,\qquad \omega_R=\gamma_R(H_z^0+aF_z^0+p_{zx}G_x^0)$$

are the resonance frequencies of the Ge^{3+} ion, and γ_R is the gyromagnetic ratio.

The longitudinal and transverse relaxation frequencies of the Ge subsystem are ω_{r1} and $\omega_r = \alpha_R\omega_R$. The susceptibility $\hat{\chi}_R(\omega)$ is given by formulas (7) and (8) with Θ_C in place of Θ_F. The paramagnetic Curie temperatures Θ_F and Θ_C here take into account the isotropic Gd–Gd interaction. The static values $\mathbf{F}_R^0$ and $\mathbf{C}_R^0$ are also determined by formulas (7) and (8), with $\omega = 0$ and with the corresponding static values H_z^0, F_z^0, and G_x^0.

By substituting the values of $\Delta\mathbf{F}_R$ and $\Delta\mathbf{C}_R$ into the linearized equations (3), we find the resonance frequencies and the total dynamic susceptibility of the system. In the $\Gamma_4(G_xF_z)$ phase that arises in $GdFeO_3$, there are two independent vibrational modes (see also Ref. 16). The first vibrational mode involves[1)] the variables $\Delta F_{x,y}$, ΔG_z, $\Delta F_R^{x,y}$, and $\Delta C_R^{x,y}$. The resonance frequencies are determined from the equation ($\alpha = 0$)

$$\omega^2[1+\chi_R^{xy}(\omega)\gamma aH_1/i\omega M_0]=\gamma^2[2H_E-a^2\chi_R^{yy}(\omega)/M_0]H_{ac}^{\text{eff}}(\omega), \tag{9}$$

where

$$H_1=aF_z^0-p_{xz}G_x^0,\qquad 2H_E=A/M_0,$$
$$H_{ac}^{Fe}=K_{ac}^{Fe}/M_0,\qquad H_{ac}^{\text{eff}}(\omega)=(H_{ac}^{Fe}+\chi_R^0p_{zx}^2/M_0)G_x^{02}$$
$$+2F_z^0G_x^0\chi_R^0ap_{zx}/M_0+H_z^0(F_z^0+\chi_R^0H_1/M_0)$$
$$-\chi_R^{xx}(\omega)H_1^2/M_0-\tilde{\chi}_R^{yy}(\omega)(p_{yz}G_x^0)^2/M_0. \tag{10}$$

In the absence of damping, Eq. (9) reduces to a cubic equation in ω^2 whose roots determine the three resonance frequencies. One of these frequencies is the analog of the frequency of the quasiferromagnetic mode of the Fe subsystem, and the other two are the quasirare-earth modes.

The second vibrational mode involves the variables ΔF_z, $\Delta G_{x,y}$, ΔF_R^z, and ΔC_R^z. In this case there is one resonance frequency, which is the analog of the quasiantiferromagnetic mode of the Fe subsystem, and there are also relaxational frequencies corresponding to the longitudinal vibrations of the Ge subsystem. The resonance frequency is determined from the equation

$$\omega^2=\gamma^2[2H_E-\chi_R^{zz}(\omega)H_2^2/M_0]H_{ab}^{\text{eff}}(\omega), \tag{11}$$

where

$$H_2=aG_x^0-p_{zx}F_z^0,$$
$$H_{ab}^{\text{eff}}(\omega)=(H_{ab}^{Fe}+\chi_R^0p_{zx}^2/M_0)G_x^{02}+G_x^0F_z^0(-H_D+\chi_R^0ap_{zx}/M_0)$$
$$+H_z^0(\chi_R^0p_{zx}/M_0)G_x^0-\tilde{\chi}_R^{zz}(\omega)(p_{zy}G_x^0)^2/M_0, \tag{12}$$
$$H_{ab}^{Fe}=K_{ab}^{Fe}/M_0,\qquad H_D=d/M_0.$$

The quantities $H_{ac}^{\text{eff}}(\omega)$ and $H_{ab}^{\text{eff}}(\omega)$ and (9) and (11) go over for $\omega\to 0$ to the effective anisotropy fields of $GdFeO_3$, which are related to the anisotropy constants and thermodynamic potential of the system by

$$K_{ac}^{\text{eff}}=(\partial^2\Phi/\partial\theta^2)_{\Gamma_4}=M_0H_{ac}^{\text{eff}}(0),$$
$$K_{ab}^{\text{eff}}=(\partial^2\Phi/\partial\varphi^2)_{\Gamma_4}=M_0H_{ab}^{\text{eff}}(0). \tag{13}$$

Here $\Phi(\theta,\varphi)$ is the thermodynamic potential of $GdFeO_3$ that was used in Ref. 6 (see also Ref. 17) for analysis of the static magnetic properties. The polar angle θ and azimuthal angle φ specify the orientation of the antiferromagnetism vector $\mathbf{G}$ of the Fe^{3+} ions. The derivatives in (13) are evaluated in the Γ_4 phase at $\theta = \pi/2$, $\varphi = 0,\pi$.

Depending on the relationship of the AFMR eigenfrequencies

$$\omega_1^{Fe}=\gamma(2H_EH_{ac}^{Fe})^{1/2},\qquad \omega_2^{Fe}=\gamma(2H_EH_{ab}^{Fe}+H_D^2)^{1/2}$$

of the Fe subsystem and ω_R of the R subsystem, formulas (9) and (11) lead to two different situations. Let us consider the case $\omega_R \ll \omega_{1,2}^{Fe}$, which obtains in $GdFeO_3$ in a relatively small external field. In particular, for $H = 0$

$$\nu_{Gd}=\gamma(aF_z^0+p_{zx}G_x^0)/2\pi\approx\gamma H_{\text{eff}}^c/2\pi\approx 1\text{GHz},$$

where, according to Refs. 6 and 12

$$|H_{\text{eff}}^c|=|p_{zx}-ad/A|=310\text{ Oe}.$$

Let us also assume that the relaxational frequencies ω_{r1} and ω_r are much lower than ω_1^F. Inthis case the dynamic susceptibility of the Gd subsystem at the AFMR frequencies will be much smaller than the static value χ_R^0. We can thus neglect the former in (9) and (11) and get the following expressions for the AFMR frequencies:

$$\omega_1^2=2\gamma^2H_EH_{ac}^{\text{eff}}(\infty),\qquad \omega_2^2=2\gamma^2H_EH_{ab}^{\text{eff}}(\infty). \tag{14}$$

This approximation is equivalent to neglecting all the oscillatory variables of the Gd subsystem ($\Delta\mathbf{F}_R$,$\Delta\mathbf{C}_R$) in the equations of motion (3) and taking only their static values into account. Thus, in this case the Gd sublattice will behave as a frozen system that produces only a static effective field at the Fe^{3+} ions.

We note that in the opposite case, when the frequency of the R subsystem is substantially higher than the AFMR frequency, formulas (14) should be written with $H_{ac,ab}^{\text{eff}}(\infty)\to H_{ac,ab}^{\text{eff}}(0)$ and $2H_e\to 2H_E-\chi_R^0a^2/M_0$. This case corresponds to a situation in which the variables of the R subsystem can instantaneously follow the variables of the Fe subsystem. Here one can use the Landau-Lifshitz equations with a thermodynamic potential renormalized by the R–Fe interaction (see, e.g., Ref. 11). This situation is typical for the quasiferromagnetic mode of the AFMR in $TmFeO_2$.[13]

Let us analyze in more detail the case of the frozen R sublattice in $GdFeO_3$. Calculating the equilibrium values for $H_z^0 = 0$.

$$F_z^0=G_x^0\left(-H_D+ap_{zx}\frac{\chi_R^0}{M_0}\right)\left(2H_E-\frac{a^2\chi_R^0}{M_0}\right)^{-1},\qquad G_x^0=\pm 1, \tag{15}$$

we find for the AFMR frequencies (14)

$$\omega_1^2=(\omega_1^{Fe})^2+2\gamma^2H_E\xi_1\chi_R^0(H_{\text{eff}}^c)^2/M_0,$$
$$\omega_2^2=(\omega_2^{Fe})^2+2\gamma^2H_E\xi_2\chi_R^0(H_{\text{eff}}^c)^2/M_0,$$
$$\varepsilon=\chi_R^0a^2/2H_EM_0,\qquad \xi_1=(1-\varepsilon)^{-2},\qquad \xi_2=(1-\varepsilon)^{-1}, \tag{16}$$

where $\omega_{1,2}^{Fe}$ are the resonance frequencies of the Fe subsystem in the absence of the Gd–Fe interaction, and $H_{eff}^{c} = p_{zx} - ad/A$ is the effective field at the Gd^{3+} ions in the Γ_4 phase. Formulas (16) give a good qualitative description of the observed temperature dependence of the AFMR frequencies $\nu_{1,2}(T)$. The quantity $\xi_{1,2}\chi_R^0 (H_{eff}^{c})^2/M_0$ is the static effective field acting on the Fe subsystem on account of the Gd^{3+} ions; this field stabilizes the Γ_4 phase and increases with decreasing temperature. However, to get a quantitative description of the observed temperature dependence $\nu_{1,2}(T)$ we would need a value $H_{eff}^{c} \approx 4$–5 kOe, which is substantially larger than the value $|H_{eff}^{c}| = 0.31$ kOe obtained from the static magnetic measurements.[12] This discrepancy may stem from the presence of addition effective fields at some of the Gd^{3+} ions due to the decompensation of the magnetic moments of the Fe^{3+} ions on account of defects or impurities in the crystal. The effect of these additional fields can be illustrated clearly for the example of magnetic vacancies in the Fe subsystem,[18] which arise when an Fe^{3+} ion is replaced by a nonmagnetic impurity ion. If the nearest-neighbor environment of a rare earth ion contains such a nonmagnetic impurity ion (a magnetic vacancy), the decompensation of the magnetic moments of the antiferromagnetically ordered Fe^{3+} ions will give rise to a random exchange field $H_{MB} \approx \pm \Lambda G$ at the rare earth ion, where Λ is the isotropic exchange constant of an R^{3+}-Fe^{3+} pair. A calculation analogous to the above shows that the AFMR frequencies in the frozen Gd sublattice approximation are given in this case by the formulas

$$\omega_{1,2}^2 = (\omega_{1,2}^{Fe})^2 + 2\gamma^2 H_E[\xi_{1,2}\chi_R^0 (H_{eff}^{c})^2 + xz\chi_R^0\Lambda^2]/M_0, \quad (16')$$

where x is the concentration of magnetic vacancies and $z = 8$ is the number of Fe^{3+} nearest neighbors of the rare earth ion. Assuming that $H_{eff}^{c} = 0.31$ kOe, $C = 0.03$ cm/g · deg, $\Theta_F = 4.3$ K, $a = -180$ kOe (Refs. 6 and 12), $\Lambda \approx a/z$, $\nu_1^{Fe} = 310$ GHz, $\nu_2^{Fe} = 670$ GHz, $H_E = 6.7 \cdot 10^6$ Oe, $M_0 = 106$ G · cm³/g, and $\gamma = 1.76 \cdot 10^7$ Hz/Oe, we find that formula (17) gives a rather good description of the observed temperature dependence $\nu_{1,2}(T)$ at magnetic-vacancy concentrations of only 0.4% (see Fig. 1a).[2)] Importantly, this magnetic-vacancy mechanism has practically no effect on the static magnetic properties of $GdFeO_3$ (since the susceptibility of the Gd^{3+} ions is isotropic) but can be manifested only in the dynamic properties, by virtue of the frozen Gd sublattice.

Let us conclude by considering the behavior of the total dynamic susceptibility tensor χ of $GdFeO_3$, which in the Γ_4 phase has the same form as in (7). In the approximation of a frozen Gd sublattice with small damping, the components of this tensor are ($H_z^0 = 0$):

$$\chi_{xx} = \frac{\chi_\perp^0}{(1-\varepsilon)^2}\frac{\omega_D^2}{\omega_1^2 - \omega^2 + i\omega\tilde{\Gamma}^{(1)}},$$
$$\chi_{yy} = \chi_\perp^0 \frac{\omega_1^2}{\omega_1^2 - \omega^2 + i\omega\tilde{\Gamma}^{(1)}}, \quad \chi_{xy} = -\chi_{yx} = \frac{i\omega}{\omega_1}(\chi_{xx}\chi_{yy})^{1/2}, \quad (17)$$
$$\chi_{zz} = \chi_\perp^0 \frac{\omega_2^2}{\omega_2^2 - \omega^2 + i\omega\tilde{\Gamma}^{(2)}},$$

where $\omega_D = \gamma H_D$, $\chi_\perp^0 = M_0/2H_E$, and $\tilde{\Gamma}^{(1)}$ and $\tilde{\Gamma}^{(2)}$ are coefficients which determine the linewidth of the quasiferromagnetic and quasiantiferromagnetic modes of the AFMR. The temperature dependence of $\Gamma^{(1,2)} = \tilde{\Gamma}^{(1,2)}/2\pi$ was determined experimentally (Fig. 1b). At low temperatures these coefficients are practically independent of T, indicating that the main contribution to the linewidths at low T is from defects and imperfections in the crystal.[19,20] With increasing T we see a growth of $\Gamma^{(1,2)}$ that is apparently due to an increase in the role of self-relaxation (four-magnon, six-magnon, etc.) processes.[21,22]

It follows from (17) that the contributions of the modes to the static magnetic permeability $\Delta\mu_1 = \Delta\mu_y = 4\pi\chi_\perp^0$ for $\mathbf{h} \| b$ and $\Delta\mu_2 = \Delta\mu_z = 4\pi\chi_\perp^0$ for $\mathbf{h} \| c$, should be identical and independent of temperature, as is well confirmed by the measurements. This allows us to find $\chi_\perp^0 = 0.8 \cdot 10^{-5}$ cm³/g and $H_E = 6.7 \cdot 10^6$ Oe, where we have assumed for $GdFeO_3$ a density of $\rho = 8$ g/cm³ and a sublattice spin magnetic moment $M_0 = 106$ G · cm³/g. Knowing $\chi_\perp^0$ and the AFMR frequencies, we can determine the anisotropy constants K_{ac}^{Fe} and K_{ab}^{Fe}. For $T = 300$ K we have $K_{ac}^{Fe} = 0.96 \cdot 10^5$ erg/g and $\tilde{K}_{ab}^{Fe} = K_{ab}^{Fe} + d^2/A = 3.7 \cdot 10^5$ erg/g.

4. CONCLUSION

Let us summarize the results of this study. We have detected two AFMR modes—one quasiferromagnetic and one quasiantiferromagnetic—in the submillimeter transmission spectra of the orthoferrite $GdFeO_3$. We have obtained the temperature dependence of the parameters of these modes, viz., the resonance frequencies $\nu_{1,2}(T)$, the linewidths $\Gamma^{(1,2)}(T)$, and the contributions $\Delta\mu_{1,2}(T)$ to the static magnetic permeability, in the temperature range 4.2–300 K.

We observed an increase in the AFMR frequencies of $GdFeO_3$ with decreasing temperature (for $T < 40$ K); this is unusual in view of the fact that the effective anisotropy constants, on the contrary, suffer a decrease due to the Gd–Fe interaction.

We have carried out a theoretical analysis of the coupled oscillations of the magnetic moment of the Fe and Gd subsystems. We have shown that the observed behavior of the AFMR frequencies can be explained by the interaction of the magnetic moments of the Fe^{3+} ions with the Gd sublattice, which is "frozen" at the frequencies in question because of its low natural or relaxation frequencies. Thus the spin oscillations of the Fe^{3+} ions in effect occur in a static effective field (which increases with decreasing temperature) due to the Gd subsystem. We have shown that the Gd–Fe interaction parameters obtained from the static magnetic measurements give only a qualitative description of the observed temperature dependence of $\nu_{1,2}$. To obtain a quantitative description we have invoked a mechanism involving the magnetic vacancies which arise in a real crystal because of defects and impurities. Better agreement between the experimental and theoretical $\nu_{1,2}(T)$ curves is found with allowance for the magnetic vacancy mechanism at a magnetic vacancy concentration of only 0.4%.

We have calculated the components of the dynamic magnetic susceptibility tensor and found that their behavior

is in good agreement with the measured contribution of the modes to the static magnetic permeability.

[1)]Since in the present description the wavelengths of the vectors of the Fe sublattice are conserved, i.e., $\mathbf{F}^2 + \mathbf{G}^2 = 1$, $\mathbf{F} \cdot \mathbf{G} = 0$, the variables $\Delta\mathbf{F}$ and $\Delta\mathbf{G}$ are coupled. For example, in the Γ_4 phase we have $F_z^0 \Delta G_z + G_x^0 \Delta F_x = 0$ and $F_z^0 \Delta F_z + G_x^0 \Delta G_x = 0$.

[2)]The deviation of the theoretical $\nu_2(T)$ curve from the experimental curve for mode 2 at $T > 100$ K is due to temperature dependence in $\nu_2^{Fe}(T)$, which we have ignored in the calculation.

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