# Influence of optical excitation of holmium impurity ions on magnetic resonance in yttrium iron garnet

G. A. Petrakovskiĭ and G. S. Patrin

*L. V. Kirenski Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR, Krasnoyarsk*

(Submitted 27 September 1985)
Zh. Eksp. Teor. Fiz. **90**, 1769–1780 (May 1986)

The changes in the parameters of the magnetic resonance in a $Y_3Fe_5O_{12}$:$Ho^{3+}$ single crystal subjected to laser radiation of the $\lambda = 1.064\,\mu$m wavelength have been investigated experimentally and theoretically. A model in which the crystal matrix is considered in the continuum limit and the impurity ions are regarded as quasi-Ising particles with a moment $\tilde{J} = 1$ is used to show that the main experimental results can be explained by an effective reduction in the concentration of the impurity ions because of a transition to an optically excited state and by the departure of the populations of the levels of the ground-state quasidoublet from the Gibbs distribution in the course of a cycle comprising optical excitation and relaxation. A theoretical explanation is provided of the characteristics of the angular and temperature dependences of the resonance field and of the width of the magnetic resonance line of $Y_3Fe_5O_{12}$:$Ho^{3+}$ in the ground state.

## 1. INTRODUCTION

The development of modern physical experimental methods has aroused the interest of researchers in physical systems which are in nonequilibrium states. These systems may be "frozen" states such as amorphous materials,[1] metastable states of the type created by magnetic fields, pressure,[2] or optical excitation,[3] and also dynamic excited states typical of, for example, laser systems. We shall consider the influence of optical excitation on the magnetic state of matter. We shall confine our attention to the effects that result from optical excitation of impurity ions in magnetic insulators. Such excitation appreciably alters the electron states of the ions affected,[4] which—under certain conditions—may alter the magnetic properties of matter. These effects differ fundamentally from the optical effects of photomagnetic annealing, since in the latter case the essential feature is not the presence of optically excited states but redistribution of some of the active magnetic centers (for example, $Fe^{2+}$ ions in $Y_3Fe_5O_{12}$:$Si^{4+}$ crystals[5,6]) under the influence of illumination. It should also be mentioned that this mechanism differs greatly from the photomagnetism of magnetic semiconductors.[7,8]

Selective transfer of paramagnetic ions to optically excited states generally alters all the interactions in a magnetic material. However, depending on the predominant mechanism, it is convenient to divide the optical excitation effects into exchange and relativistic effects. Since optical excitation may alter fundamentally the magnetic state of ions and the exchange interactions, it is clear that the ground state of the whole crystal may change drastically in a great variety of ways. The new magnetic state may be observed when the spin system is in dynamic equilibrium subjected to optical pumping or may be metastable. In both cases the new state may be collective (in phase transitions) or it may be a single-ion state. In the former case the effect should be manifested as a phase transition of the percolation type with an intensity threshold. An example of such an effect is an optically induced change in the magnetic state reported for $EuCrO_3$ (Ref. 3).

The second class of effects associated with a change in the relativistic interactions is characterized by modifications of the magnetic properties of magnetic materials. This class of effects stems from the basic dependence of the magnetic properties of matter (for example, the single-ion anisotropy) on the electron state of a magnetic ion. An effect of this type has been observed[9] as an optically induced orientational transition in $ErCrO_3$. In this case the optically excited $Er^{3+}$ ions act as impurities.

It was found previously[10] that pulsed laser radiation of the $\lambda = 1\,\mu$m wavelength alters the parameters of a magnetic resonance in a $Y_3Fe_5O_{12}$:$Ho^{3+}$ single crystal. It has also been shown that these changes are mainly due to a change in the magnetic state of the holmium ions when they undergo transition to an optically excited state.

We shall report a detailed investigation of the influence of laser excitation of the $Ho^{3+}$ ions in the $Y_3Fe_5O_{12}$ matrix on magnetic resonance in steady state, carried out with the aim of identifying the mechanisms of optically induced changes.

## 2. PROPERTIES OF A $Y_3Fe_5O_{12}$ CRYSTAL DOPED WITH $Ho^{3+}$ IONS

Yttrium iron garnet $Y_3Fe_5O_{12}$ is one of the most thoroughly investigated ferrites. This is due to the fact that this insulator possesses unique properties, particularly resonant, acoustic, and optical. An important feature is also the highly advanced technology available for the preparation of high-quality $Y_3Fe_5O_{12}$ single crystals.[11]

For optical excitation to have an effect on the properties of matter there must be a long-lived (metastable) excited level and the ability to transform the system efficiently to this level. The situation is fully analogous to that encoun-

0038-5646/86/051038-07$04.00

tered in the search for laser-active materials. If the electronic properties of ions in the ground and excited states are different, then the transformation of the system to an excited state will alter greatly some characteristics of a magnetic material. Therefore, effective controlled modification of material properties by optical radiation requires the knowledge of details of its electron structure and of the mechanisms that determine its properties.

It is known[12] that the resonance properties of $Y_3Fe_5O_{12}$ depend sensitively on the presence of rare earth impurities. For example, the width $\Delta H$ of the FMR line representing homogeneous precession of the magnetization of "pure" $Y_3Fe_5O_{12}$ is approximately proportional to temperature in the range 0–3000 K and at $T = 300$ K it represents a few tenths of an oersted (in the $\lambda = 3$ cm range). The addition of 1 at.% holmium ions alters the FMR line width by about 750 Oe at the maximum on the temperature scale when the magnetizing field $\mathbf{H}_0$ is oriented along the [111] axis. The temperature dependence of $\Delta H$ has a characteristic maximum at a temperature that depends on the orientation of a sample. The behavior of the resonance properties of $Y_3Fe_5O_{12}$ crystals doped with rare earths is normally described using a theory of "slow" relaxation,[12] but there are some points that still remain unclear.

On the other hand, a detailed study of the optical properties of "pure" and $Ho^{3+}$-doped iron garnets[13] has made it possible to reconstruct and describe the structure of the ground $^5I_8$ and first excited $^5I_7$ multiplets of the $Ho^{3+}$ ion, and also to identify the channels for transfer of the electron excitation energy by the holmium ions and the efficiency of these channels. In a $Y_3Fe_5O_{12}$:$Ho^{3+}$ crystal the $^5I_7$ multiplet is metastable and its lifetime at $T = 78$ K is about 4 ms and excitation of this crystal with radiation of the 0.4–2 $\mu$m wavelengths generates $^5I_7 \to {}^5I_8$ luminescence in the region of $\lambda = 2\,\mu$m. Stafford *et al.*[14] reported emission of stimulated radiation as a result of pulsed laser radiation pumping ($\lambda = 1.06\,\mu$m) via the absorption band of the $Fe^{3+}$ ions at the octahedral positions. In this case the levels of the $^5I_7$ multiplet were filled as a result of the $^4T_{1g}(Fe^{3+}) \rightsquigarrow {}^5I_7(Ho^{3+})$ nonradiative transition (Fig. 1) when the threshold pump density was 3 J/cm$^2$ at $T = 78$ K.

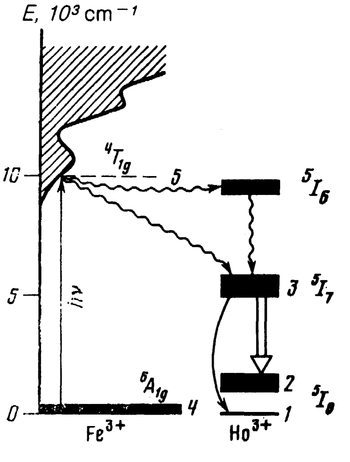

FIG. 1. Diagram of the energy levels in a $Y_3Fe_5O_{12}$:$Ho^{3+}$ crystal and the transition scheme.

## 3. EXPERIMENTAL METHOD

All the measurements were carried out using the spectrometer described in Ref. 10. The radiation source was a cw laser with the following parameters: $\lambda = 1.06\,\mu$m, beam diameter 3 mm, radiation power 0.5–3 W. The measurements were made on $Y_3Fe_5O_{12}$:$Ho^{3+}$ (0.1 at.%) samples in the form of disks 2 mm in diameter and 0.2 mm thick, cut parallel to the (110) plane. These disks were polished on both sides to give an optical surface finish. The direction of propagation of the exciting radiation was perpendicular to the plane of the sample and to the direction of an external magnetic field.

The experimental results were obtained by the following procedure: first a microwave absorption line was recorded for an unilluminated sample, then optical radiation of $P_1$ power was applied to the sample and after 2–3 min the absorption line was recorded during illumination. In this time a thermal equilibrium was established in the working chamber of a cryostat and the temperature of the sample remained subsequently constant. The radiation was then switched off. The time taken to recover the initial temperature was 1–2 min. This procedure was repeated for the next value of the power $P_2 > P_1$.

We carried out measurements at values of the optical radiation power such that the increase in the temperature of a sample $\Delta T$ was less than the error in the temperature determination shown in the plots (for example, when the optical radiation power density was 10 W/cm$^2$, the temperature rise was $\Delta T = 2$ K).[15]

Figures 2 and 3 show the experimental results which

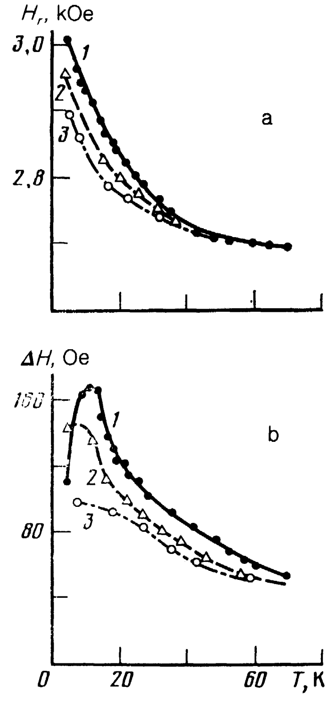

FIG. 2. Experimental temperature dependences of the resonance field (a) and of the line width (b) obtained for a $Y_3Fe_5O_{12}$:$Ho^{3+}$ (0.1 at.%) crystal when the magnetic field was directed along the [001] axis: 1) unilluminated sample; 2) radiation power density 7 W/cm$^2$; 3) 12 W/cm$^2$.

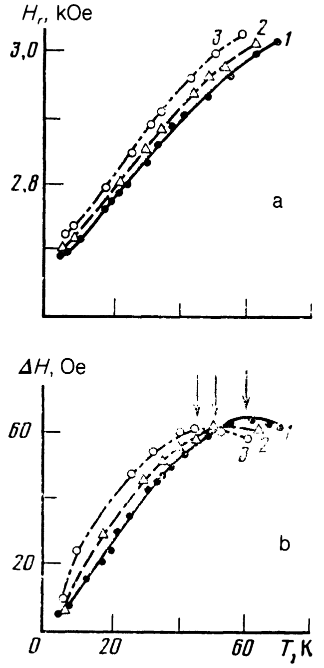

FIG. 3. Experimental temperature dependences of the resonance field (a) and of the line width (b) obtained for a $Y_3Fe_5O_{12}$:$Ho^{3+}$ (0.1 at.%) crystal when the magnetic field was directed along the [111] axis: 1) unilluminated sample; 2) radiation power density 7.35 W/cm$^2$; 3) 10 W/cm$^2$. The arrows identify the positions of the maxima.

were obtained for a $Y_3Fe_5O_{12}$ sample in which the $Ho^{3+}$ ion concentration was $x = 0.1$ at.% and the measurements were carried out at different optical pump powers when the magnetic field direction was parallel to the [001] and [111] axes. Clearly, the influence of optical radiation on the magnetic resonance depended on the direction of the external magnetic field and, in the range of pump powers investigated, it disappeared in the region of $T = 50$ K. The temperature dependence of the line width had a common feature for both directions: illumination reduced the line widths at its maximum and shifted the maximum toward lower temperatures.

We also carried out experiments on an yttrium ferrite garnet single crystal free of dopants and characterized by a microwave absorption line width of about 1 Oe. Within the limits of the experimental error, there were no changes in the line width or in the resonance field.

## 4. THEORY

These experimental results can be understood on the basis of the following model.

In many-level systems one frequently uses in practice the fact that adjacent levels form groups, which makes it possible to describe some low-temperature magnetic properties by introducing an effective moment. As a rule, the $g$ factor and the exchange interactions are strongly anisotropic.[16] For example, in the case of a $Y_3Fe_5O_{12}$ crystal doped with holmium ions the low-temperature behavior is described in Ref. 17 allowing only for the lowest quasidoublet of the $Ho^{3+}$ ion and assuming quasi-Ising behavior. The latter assumption is justified because rare-earth ions with an even number of electrons have two levels which are quite distant from the other levels and we then have $g_\perp = 0$ (Ref. 18).

In a qualitative analysis of the influence of optically excited states we can limit ourselves to introduction of an additional level separated from the ground-state quasidoublet by an energy equal to the photon energy $h\nu$. This situation can be described mathematically by a three-level model of a center using an effective moment $\tilde{J} = 1$. Figure 1 shows the diagram of the energy levels and the transition scheme for a $Y_3Fe_5O_{12}$:$Ho^{3+}$ crystal.

We shall assume that the impurity ions are distributed with equal probability among six inequivalent positions in a crystal. Each ion is then characterized by a site index $i = 1,...,N_k$ and by a position index $k = 1,...,6$, where $N_k = N_0/6$ and $N_0$ is the total number of impurity ions.

In the case of the $i$th ion located in the $k$th position we can apply the molecular field approximation and include the first nonvanishing terms of a crystal field with local orthorhombic symmetry. The Hamiltonian of this ion can be written as follows:

$$H_{ik}=(\lambda \mathbf{M}_0-g\mathbf{H}_0)\xi\tilde{\mathbf{J}}_{z_{ik}}-D(\tilde{J}^2_{z_{ik}}-1)+\Delta(\tilde{J}^2_{x_{ik}}-\tilde{J}^2_{y_{ik}})/2. \quad (1)$$

Here, $\lambda$ $\mathbf{M}_0$ is the molecular field exerted on the $Ho^{3+}$ ions by the "iron" subsystem; $\mathbf{H}_0$ is the external magnetic field; $\Delta$ and $D$ are the crystal field parameters; $\xi$ is a parameter in the theory (in the present case it represents the maximum magnetic moment of an ion); $\mathbf{M}_0$ is the total magnetization of the investigated crystal; $\tilde{\mathbf{J}}_{z_{ik}}$, $\tilde{\mathbf{J}}_{x_{ik}}$, and $\tilde{\mathbf{J}}_{y_{ik}}$, are the corresponding components of the effective moment in the local coordinate system.

Diagonalization of expression (1) subject to the condition $D > 0$ yields the eigenvalues

$$\varepsilon_{ik}^{1,2} = \mp\{[(\lambda M_0-gH_0)\xi\gamma_{ik}]^2+\Delta^2/4\}^{1/2}, \quad \varepsilon_{ik}^3=D, \quad (2)$$

where $\gamma_{ik}$ is the cosine of the angle between the total field acting on the moment of the $i$th and the local $z$ quantization axis in the $k$th position in the garnet structure (the local $z$ axes of a dodecahedral position coincide with the [100], [010], and [001] crystallographic directions).

If we ignore the details of the populations the levels of the impurity ion, we find that its behavior in a laser radiation field can be described by means of rate equations.

In the case of the $i$th ion in the $k$th position the system of equations is

$$dn_{ik}^l/dt=\sum_{l\neq m} d_{lm}n_{ik}^m, \quad l, m=\{1, 2, 3\}, \quad (3)$$

and the whole crystal satisfied the condition

$$\sum_{l,ik} N_k n_{ik}^l = N_0.$$

Here, $n^l_{ik}$ is the population of the $l$th level; $d_{lm}$ is the probability of the transition from the $l$th to the $m$th level; $d_{13}$ (or $d_{23}$) is the probability of the transition in accordance with the 4 → 5 → 3 scheme (Fig. 1).

In general, an analysis of the behavior of the ions interacting with optical radiation should be carried out bearing in mind that impurity ions are in nonequilibrium states and that the effective temperature may differ from the temperature of the matrix.

In an analysis of heat transfer in solids the thermal equilibrium is established in a wide range of temperatures (with the exception of the ultralow region) by thermal diffusion. The time taken to establish such an equilibrium after a heat pulse is applied to one of the faces of a sample of thickness $L$ is[19]

$$\tau^*=\rho c_v L^2/\varkappa,$$

where $\rho$ is the density, $c_v$ is the specific heat at constant volume, and $\varkappa$ is the thermal conductivity. For example, in the case of quartz[20] at $T=5$ K we have $\tau^*=1.7\times10^{-5}$ s for $L=0.2$ mm. A comparison of the lifetime $\tau$ of the $Ho^{3+}$ ions in the $^5I_7$ state with the time $\tau^*$ shows that $\tau\gg\tau^*$. This allows us to assume that a holmium ion in an optically excited state is at every moment in thermal equilibrium with the crystal matrix.

Optical radiation does not induce direct transitions between the levels of the ground-state quasidoublet of an impurity ion so that the probabilities of relaxational transitions between these levels obey the following condition because of detailed balance[21]

$$d_{12}/d_{21}=\exp(-\Delta/k_BT),\quad \Delta=\varepsilon_2-\varepsilon_1, \tag{4}$$

where $k_B$ is the Boltzmann constant and $T$ is the lattice temperaure.

When the magnetization vector precesses about a specific axis at a frequency $\omega$, the projections of the molecular field on the local $z$ axes also vary at a frequency $\omega$. This alters the splitting $\Delta$ and causes a redistribution of the populations of the sublevels 1 and 2. In the case of small variations in the direction of the magnetization Eq. (4) becomes

$$d_{12}/d_{21}=\exp(-\Delta_{ik}^0/k_BT)\,[1-(\delta_{ik}/k_BT)\,\exp i\omega t], \tag{4a}$$

where $\Delta_{ik}^0$ is the steady-state splitting of the levels of the $i$th ion in the $k$th position,

$$\delta_{ik}=(\partial\Delta_{ik}/\partial\theta)\Delta\theta+(\partial\Delta_{ik}/\partial\varphi)\Delta\varphi, \tag{5}$$

and $\theta$ and $\varphi$ are the polar and azimuthal angles of the magnetization vector. Bearing in mind the above discussion and solving the system (3) by the method of successive approximations, we obtain the first-order solution for $n_{ik}^l$ (see the Appendix 1).

The behavior of the system can be described as a function of the changes in the parameters if we know the form of the free energy. We shall consider the $Y_3Fe_5O_{12}$ crystal matrix in the continuum limit. The parameters which describe this matrix are the magnetization $\mathbf{M}(T)$ and the magnetocrystalline anisotropy constant $K_1(\mathbf{M},T)$. The free energy of a crystal with impurities subjected to a laser radiation field is

$$U=F_Z+F_A+f+W, \tag{6}$$

where

$$F_Z=-\mathbf{MH}_0,\quad F_A=-{}^1/_2|K_1|\sum_{j=x,y,z}M_j^4+\mathbf{M}\overleftrightarrow{N}\mathbf{M}/2,$$

$$f=\sum_{l,i,k}n_{ik}{}^l\varepsilon_{ik}{}^l-ST,$$

$S$ is the entropy of the impurity subsystem, $\overleftrightarrow{N}$ is the tensor of the demagnetization coefficients, and $W$ is the energy of the optical radiation field.

Solution of the Landau-Lifshitz equations yields expressions for the resonance field $H_r$ and the magnetric resonance relaxation frequency $\omega_r$. We shall assume, as usual, that the oscillation frequency is $\omega=\omega_1+j\omega_r$. We then have

$$H_r=-(H_{\theta\theta}{}^A+H_{\varphi\varphi}{}^A+f_{\theta\theta}+f_{\varphi\varphi})/2$$
$$+[(H_{\theta\theta}{}^A-H_{\varphi\varphi}{}^A+f_{\theta\theta}-f_{\varphi\varphi})^2/4+(\omega_1/\Gamma)^2]^{1/2}, \tag{7}$$

$$\Delta H=\omega_r/\Gamma=(\Gamma/\omega_1)[H_r(S_{\theta\theta}+S_{\varphi\varphi})+H_{\theta\theta}{}^AS_{\varphi\varphi}+H_{\varphi\varphi}{}^AS_{\theta\theta}]/2. \tag{8}$$

Here, $\Gamma$ is the gyromagnetic ratio and the expressions for $H_{\theta\theta}^A$, $H_{\varphi\varphi}^A$, $f_{\theta\theta}$, $f_{\varphi\varphi}$, $S_{\theta\theta}$, and $S_{\varphi\varphi}$ are given in Appendix 1.

The equilibrium values of the angles $\theta_0$ and $\varphi_0$ are determined from the minimum of the free energy (6) on the assumption that the Zeeman energy is much greater than the other terms. In the experiments a sample was usually oriented so that the magnetic field $\mathbf{H}_0$ was in the (110) plane. This corresponds to an equilibrium azimuthal angle of $\theta_0=\pi/4$.

The equilibrium value of $\theta$ is determined from the expression

$$\theta_0=\theta_H-[U'(\theta_H)/MH][1-U''(\theta_H)/MH]^{-1}, \tag{9}$$

where $\theta_H$ is the angle between the external magnetic field and the $z$ axis in the crystallographic coordinate system (the [001] direction in the present case). A prime represents differentiation with respect to the variable $\theta$. All stages of this calculation were carried out in the linear approximation.

The quantities defined by Eqs. (7) and (8) were calculated for the following values of the parameters[22]:

$$\lambda M_0=125\,\text{koe},\quad \xi=10\mu_B,\quad \omega_1=9{,}3\,\text{GHz},$$
$$M_0=218\ \text{G},\quad \Delta^0=8\,\text{cm}^{-1},\quad K_1=24{,}5$$
$$\times10^3\,\text{erg/cm}^3,\quad N_0=1{,}267\cdot10^{19}\ \text{particles/cm}^3,$$
$$\tau_0=1{,}08\cdot10^{-10}\,\text{sec},\quad D=5000\ \text{cm}^{-1}.$$

## 5. RESULTS AND DISCUSSION

### a. System in the absence of optical pumping

A resonance in the absence of laser radiation can be described by substituting in Eqs. (A1.2) and (A1.3) the values $A_1=A_2=B=C=0$, which corresponds to zero population of the level 3. Figure 4 shows the angular dependence of the line width calculated from Eq. (8) (curve 1) and the experimental dependence taken from Ref. 23 (curve 2). We can see that the angular dependence of the resonance field is practically identical with that obtained in Ref. 24 without allowance for the delay of the transitions between the levels of the $Ho^{3+}$ ions.

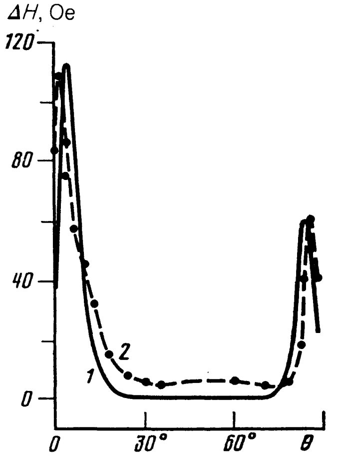

FIG. 4. Angular dependence of the line width at helium temperatures: 1) calculated for $T = 5$ K; 2) experimental curve taken from Ref. 23 and obtained at $T = 4.2$ K.

Figure 5 shows the temperature dependence of the theoretical line width for the magnetic field directions close to [111] (curve 1) and [110] (curve 4), as well as the experimental curves obtained for the same directions and taken from Ref. 23 (curves 2 and 3, respectively). In all these calculations it was assumed that $\tau_0$ was independent of temperature and that the condition $\omega_1\tau_0 = 1$ was obeyed. Clearly, the angular dependence exhibited good agreement between the theory and experiment.

The experimental and calculated temperature dependences of the line width were in general agreement. When the magnetic field was directed close to the [110] axis, there were two maxima which had not been explained earlier, whereas in the case of the field directed along the [001] and [111] axes, the positions and amplitudes of the maxima were close to those found experimentally. The discrepancy between the curves may be attributed to the imperfections of our model because only two levels are considered in dealing with the ground state. Nevertheless, this approximation may be justified and the results obtained can be explained.

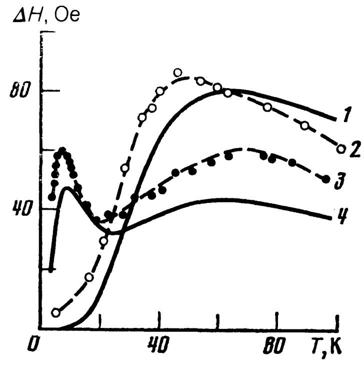

FIG. 5. Temperature dependences of the line width: 1) calculation for $\theta_H = 55°$; 2) experimental dependence for a magnetic field along the [111] axis; 3) experimental dependence for a magnetic field along the [011] axis; 4) calculation for $\theta_H = 86.5°$. The experimental points were taken from Ref. 23.

It is known that rare-earth ions with a moment $J$ have wave functions $\Phi_i$ of the form

$$\Phi_i = \sum_m C_i^m |J, m\rangle$$

($m$ is the magnetic quantum number and $C_i^m$ is the coefficient in the above expansion).

All the theoretical[25,26] and experimental[22] results provide grounds for assuming that the lowest quasidoublet of the holmium ion consists mainly of the states $|8,8\rangle$ and $|8,-8\rangle$, i.e., the wave functions of the states $|1\rangle$ and $|2\rangle$ are

$$|1\rangle = \alpha|8, 8\rangle + \beta|8, -8\rangle + (\ldots),$$

$$|2\rangle = \delta|8, 8\rangle + \eta|8, -8\rangle + (\ldots),$$

where the brackets represent a small contribution of the higher levels and there is a large difference between the coefficients $\alpha$, $\beta$ and $\delta$, $\eta$.

A transition of an ion from the $|1\rangle$ to the $|2\rangle$ state maximizes the change in its magnetic moment. At low temperatures and for the directions characterized by the minimum value of $\Delta$ the change dominates the state of the whole system. This is indicated by the good agreement between the calculated and experimental curves obtained at low temperatures. When the field $\mathbf{H}_0$ is directed along the [111] axis the splitting $\Delta$ between the $|1\rangle$ and $|2\rangle$ states of an ion is approximately 60 $\text{cm}^{-1}$, but the levels with different values of the magnetic moments are below this value. Clearly, these levels are "activated" earlier when temperature is increased, which shifts the temperature dependences toward lower temperatures.

The amplitudes of the peaks and their positions at low temperatures depend very strongly on the values of $\Delta$ and $\xi$. We made no attempt to improve the agreement between the calculations and the experimental results by fitting the parameters because the model is too crude.

### b. Influence of an optically excited state

An allowance for the existence of an optically excited state of an impurity ion has the effect that the coefficients $A_1$, $A_2$, $B$, and $C$ in Eqs. (A1.2)–(A1.4) become functions of the pump power. The population of the level 3 is then nonvanishing. Clearly, the dynamics of the transitions is governed by the ratio of the probabilities of the transitions between the levels. However, there is at present no detailed information on the probabilities of transitions in $Y_3Fe_5O_{12}$:$Ho^{3+}$, so that their selection is to some extent arbitrary.

We shall try to understand what happens in our system when an impurity ion is raised to an excited level by assuming, for the sake of simplicity, that $d_{13} = d_{23} = \rho_0$. This approximation is permissible because the $^5I_7$ level is filled as a result of a nonradiative transition which clearly involves the transfer of excitation by phonons[27] and this process is independent of the polarization of the incident radiation. Moreover, since the final level in the $^5I_7 \rightarrow {}^5I_8$ radiative transition is separated from the ground state by about 400 $\text{cm}^{-1}$, we shall assume that $d_{31} = 0$. The fact that the lifetime $\tau$ of the excited level is $4\times10^{-3}$ s gives $d_{32}=250$. We shall also use the relationships in Eq. (A2.4) and the fact that $\omega_1\tau_0=1$.

Figures 6 and 7 show the theoretically calculated temperature dependences of the resonance field and line width for the external magnetic field directions parallel to the [001] and [111] axes in the absence of optical excitation (curve 1) and in the case of pumping of level 3 (curves 2 and 3). Curves 2 and 3 represent populations of level 3 amounting to 10 and 30% of the total number of the impurity centers at $T=\Delta/k_B$.

A comparison of the theoretical and experimental dependences shows that the resonance field behavior is in qualitative agreement with the theory when the external field $\mathbf{H}_0$ is oriented along the [001] axis or along the [111] axis. It should be noted that the calculations were carried out for a spherical sample. In the case of the line width of a sample subjected to a magnetic field oriented along the [001] axis there is again a qualitative agreement between the theory and experiment, whereas in the case of the [111] direction the reduction in the amplitude of the maximum is considerably greater than that found experimentally.

Optical radiation reduces effectively the concentration of impurity ions because of transitions from the states $|1\rangle$ and $|2\rangle$ to the state $|3\rangle$ in which the magnetic moment is zero. Moreover, in the process of optical relaxation from the level 3 the Gibbs distribution of the populations of the $|1\rangle$ and $|2\rangle$ states is disturbed, which alters the equilibrium direction of the magnetization described by Eq. (9).

Since the resonance field is governed primarily by the static magnetic characteristics of a material, the optically induced changes can be explained by a change in the magnitude of the magnetic moments and in the energy of the magnetic anisotropy of the impurity subsystem. Relaxation processes in such systems are governed by the structure and dynamics of transitions between the sublevels of the ground-state multiplet. Therefore, when the external field is oriented along the [001] axis and the main role is played by the quasi-doublet of the $|1\rangle$ and $|2\rangle$ states, the model is clearly close to reality. In the case when $\mathbf{H}_0\|$ [111] we can assume that the processes of the magnetic and optical relaxation from the level 3 (or the process of absorption of light) involves the states lying below level 2 separated from level 1 by the energy $\Delta$ ($\theta_0=55°$). It should be noted that in this model there is a tendency for the temperature maximum of the line width to shift toward lower temperatures, although the reduction in the amplitude of the maximum predominates because of a reduction in the number of ions in the $|1\rangle$ and $|2\rangle$ states.

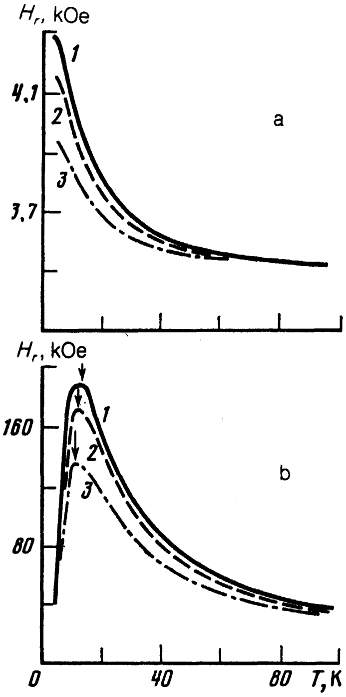

FIG. 6. Theoretical temperature dependences of the resonance field (a) and line width (b) for a magnetic field directed along the [001] axis: 1) population $n_3=0$; 2) $n_3=0.1$; 3) $n_3=0.3$; $T=\Delta/k_B$. The arrows identify the maxima of the two quantities.

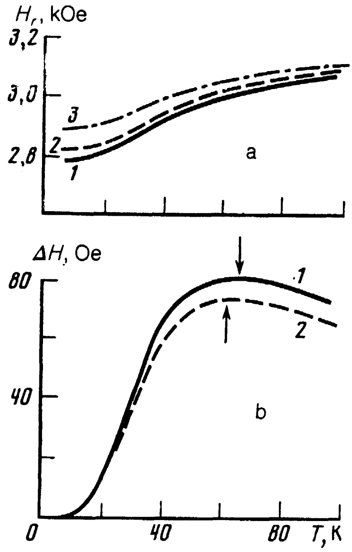

FIG. 7. Theoretical temperature dependences of the resonance field (a) and line width (b) for a magnetic field directed along the [111] axis: 1) population $n_3=0$; 2) $n_3=0.1$; 3) $n_3=0.3$; $T=\Delta/k_B$. The arrows identify the maxima of the two quantities.

Other combinations of the transition probabilities $d_{lm}$ yield qualitatively similar results. The situation when $d_{31}$ and $d_{32}$ are comparable with $d_{21}$ is more interesting, but it does not reproduce the conditions in our experiments and we shall not deal with it.

## 6. CONCLUSIONS

It follows from the above theoretical analysis that the resonance properties of an yttrium iron garnet crystal doped with $Ho^{3+}$, including the effects of optical excitation, can be described qualitatively by regarding an $Ho^{3+}$ impurity ion as a three-level center with zero magnetic moment in the excited state. The results obtained are in full agreement with those obtained in static magnetic investigations, which indicate quasi-Ising nature of the behavior of the holmium ions in the $Y_3Fe_5O_{12}$ matrix.[17] The theoretical model used in the present study can be refined by a more detailed consideration of the electron structure of the $Ho^{3+}$ ions and of the dynamics of electron transitions between the levels. It may also be important to go beyond equilibrium thermodynamics in a more rigorous analysis of the static and resonance properties of an optically excited system.

We shall conclude by noting that our results give grounds for assuming that the resonance properties of yttrium-holmium iron garnet can be described allowing for the features of the energy structure of the impurity centers and for the distributionof the impurities between the local positions in a crystal, and that the mechanism of "slow" relaxation is not the dominant one.

The authors are deeply grateful to K. A. Sablina for growing single crystals of yttrium-holmium iron garnet.

## APPENDIX 1

The expressions for the populations of the levels of an $i$th impurity ion in an optical radiation field are

$$n_l{}^i=n_l{}^{0i}+\tilde{n}_l{}^i, \qquad l=\{1,\ 2,\ 3\}.$$

Here,

$$n_1{}^{0i}=(1+A_1)/Q, \tag{A1.1}$$

$$n_2{}^{0i}=[\exp(-\Delta_i^0/k_\mathrm{B}T)+A_2]/Q, \tag{A1.2}$$

$$n_3{}^{0i}=[\exp(-\Delta_i^0/k_\mathrm{B}T)+1)B+A_3]/Q, \tag{A1.3}$$

$$\tilde{n}_{1,2}^{\,i}=\mp\frac{\delta_i n_1{}^{0i}}{k_\mathrm{B}\mathrm{T}}\exp(-\Delta_i{}^0/k_\mathrm{B}T)\exp(i\omega t)$$

$$\times\{1+i\omega/d_{21}[\exp(-\Delta_i{}^0/k_\mathrm{B}T)+1]\}^{-1},$$

$$\tilde{n}_3{}^i=0, \tag{A1.4}$$

where

$$Q=[\exp(-\Delta_i^0/k_\mathrm{B}T)+1](1+B)+C,$$

$$A_1=(d_{23}d_{31})/q, \quad A_2=d_{13}d_{32}/q, \quad A_3=d_{13}d_{23}/q, \tag{A1.5}$$

$$B=(d_{23}+d_{13})d_{21}/q, \quad C=A_1+A_2+A_3, \quad q=(d_{32}+d_{31})d_{21}.$$

## APPENDIX 2

The notation used in the text is as follows:

$$H_{\psi\psi}{}^A=\partial^2F_A/\partial\psi^2, \tag{A2.1}$$

$$f_{\psi\psi}=\Big\{\sum_{k=1}^{6}N_k\Big[\frac{\exp(-\Delta_h^0/k_\mathrm{B}T)+A_2-1-A_i}{[\exp(\mathrm{p}\Delta_k^0/k_\mathrm{B}T)+1](1+B)+C}\Big]$$

$$\times\frac{\partial^2\Delta_k{}^0}{\partial\psi^2}\Big\}-S_{\psi\psi}/\omega_1\tau_0, \tag{A2.2}$$

$$S_{\psi\psi}=\sum_{k=1}^{6}N_k\frac{2}{k_\mathrm{B}T}\frac{\omega_1\tau_0}{1+(\omega_1\tau_0)^2}\Big(\frac{\partial\Delta_k{}^0}{\partial\psi}\Big)^2(1+A_1)$$

$$\times\Big\{\Big[\exp\frac{\Delta_k}{k_\mathrm{B}T}+\exp\Big(-\frac{\Delta_k^0}{k_\mathrm{B}T}\Big)\Big]^2(1+B)$$

$$+C\Big(1+\exp\frac{\Delta_k^0}{k_\mathrm{B}T}\Big)\Big\}^{-1}, \tag{A2.3}$$

$$\psi=\theta,\ \varphi, \quad 1/\tau_0=d_{21}+d_{12}. \tag{A2.4}$$

[1] G. A. Petrakovskiĭ, Usp. Fiz. Nauk **134**, 305 (1981) [Sov. Phys. Usp. **24**, 511 (1981)].

[2] É. A. Zavadskiĭ and V. I. Val'kov, Magnitnye fazovye perekhody (Magnetic Phase Transitions), Naukova Dumka, Kiev, 1980.

[3] E. I. Golovenchits, V. A. Sanina, and T. A. Shaplygina, Zh. Eksp. Teor. Fiz. **80**, 1911 (1981) [Sov. Phys. JETP **53**, 992 (1981)].

[4] G. F. Imbusch, S. R. Chinn, and S. Geschwind, Phys. Rev. **161**, 295 (1967).

[5] B. T. Sharp and R. W. Teale, J. Phys. C **7**, 965 (1974).

[6] V. F. Kovalenko and S. N. Lyakhimets, Fiz. Tverd. Tela (Leningrad) **26**, 3014 (1984) [Sov. Phys. Solid State **26**, 1817 (1984)].

[7] A. A. Berdyshev and B. M. Letfulov, Fiz. Tverd. Tela (Leningrad) **14**, 2119 (1972) [Sov. Phys. Solid State **14**, 1824 (1973)].

[8] Magnitnye poluprovodniki/Pod red. V. G. Veselago (magnetic Semiconductors, ed. by V. G. Veselago), Tr. Fiz. Inst. Akad. Nauk SSSR **139** (1982).

[9] S. Kurita, K. Toyokawa, K. Tsushima, and S. Sugano, Solid State Commun. **38**, 235 (1981).

[10] G. A. Petrakovskiĭ and G. S. Patrin, Solid State Commun. **48**, 25 (1983).

[11] Yu. M. Yakovlev and S. Sh. Gendelev, Monokristally ferritov v radioélektronike (Ferrite Single Crystals in Radioelectronics), Sovetskoe Radio, Moscow, 1975.

[12] A. G. Gurevich, magnitnyĭ rezonans v ferritakh i antiferromagnetikakh (Magnetic Resonance in Ferrites and Antiferromagnets), Nauka, Moscow, 1973.

[13] L. F. Johnson, J. F. Dillon Jr., and J. P. Remeika, Phys. Rev. B **1**, 1935 (1970).

[14] R. G. Staford, H. Masui, R. L. Farrow, R. K. Chang, and L. G. Van Uitert, J. Appl. Phys. **47**, 2483 (1976).

[15] G. S. patrin, V. V. Ovchinnikov, and S. N. Kuliev, V kn.: Magnitnye, élektrischeskie i rezonansnye svoĭstva magnitodiélektrikov (in: Magnetic, Electrical, and Resonance Properties of Magnetic Insulators), Institute of Physics, Krasnoyarsk, 1982, p. 169.

[16] A. R. Kessel' and G. O. Berim, magnitnyĭ rezonans izingovskikh magnetikov (Magnetic Resonance in Ising Magnetic Materials), nauka, Moscow, 1982.

[17] V. I. Silant'ev, A. I. Popov, R. Z. Levitin, and A. K. Zvezdin, Zh. Eksp. Teor. Fiz. **78**, 640 (1980) [Sov. Phys. JETP **51**, 323 (1980)].

[18] S. A. Al'tshuler and B. M. Kozyrev, Élektronnyĭ paramagnitnyĭ rezonans, Nauka, M., 1972 (Electron Paramgnetic Resonance, Academic Press, New York, 1964).

[19] H. Jeffreys and B. S. Swirles, Methods of Mathematical Physics, 3rd ed., Cambridge University Press, 1956. (Russ. Transl., Vol. 3, Mir, Moscow, 1970).

[20] A. A. Novitskiĭ and I. G. Kozhevnikov, Teplofizicheskie svoĭstva materialov pri nizkikh temperaturakh (Thermophysical Properties of Materials at Low Temperatures), Mashinostroenie, M., 1975.

[21] A. Isihara, Statistical Physics, Academic Press, New York, 1971 (Russ. Transl., Mir, Moscow, 1973).

[22] S. Krupička, Physik der Ferrite und der verwandten magnetischen oxide, Vieweg, Brunswick, 1973 (Russ. Transl., Mir, Moscow, 1976).

[23] A. N. Ageev, A. G. Gurevich, Fiz. Tverd. Tela (Leningrad) **11**, 2122 (1969) [Sov. Phys. Solid State **11**, 1712 (1970)].

[24] R. Z. Levitin, A. I. Popov, and V. V. Snegirev, Fiz. Tverd. Tela (Leningrad) **24**, 3138 (1982) [Sov. Phys. Solid State **24**, 1777 (1982)].

[25] H. Kamimura and T. Yamaguchi, Phys. Rev. B **1**, 2902 (1970).

[26] V. V. Druzhinin, V. V. Shkarubskiĭ, and N. M. Chulkov, Fiz. Tverd. Tela (Leningrad) **25**, 2942 (1983) [Sov. Phys. Solid State **25**, 1696 (1983)].

[27] G. M. Zverev, I. I. Kuratev, and A. M. Onishchenko, V kn.: Spektroskopiya kristallov (in: Spectroscopy of Crystals), Nauka, Moscow, 1975, p. 184.

Translated by A. Tybulewicz