Faraday effect in yttrium orthoferrite in the "low-frequency" antiferromagnetic resonance region

K. N. Kocharyan, R. M. Martirosyan, V. G. Prpryan, and É. L. Sarkisyan

Institute of Radiophysics and Electronics, Armenian Academy of Sciences
(Submitted 21 June 1983)
Zh. Eksp. Teor. Fiz. **86**, 639–643 (February 1984)

Magnetically ordered crystals are investigated at submillimeter wavelengths by a method based on quasi-optical backward-wave-oscillator (BWO) monochromatic spectroscopy. The temperature dependences of the low-frequency antiferromagnetic branch are obtained in the range from 77 to 600 K. The Faraday effect in $YFeO_3$ is investigated in the same spectral band, and it is shown that the specific rotation of the polarization plane increases sharply near the AFMR frequency. The results of the experiments are discussed within the framework of spin-wave theory.

1. INTRODUCTION

Yttrium orthoferrite $YFeO_3$ (space group $D_{2h}{}^{16}$) is at present the object of many investigations.[1–5] The increase interest is due primarily to the fact that its domain walls have high mobility and high velocity.[2] In addition, rare-earth orthoferites are low-symmetry antiferromagnets whose dynamic properties have been relatively little investigated and whose symmetry admits of a nonzero projection of the weak ferromagnetic moment on the direction of the crystal optical axis, a property that leads to a number of interesting magneto-optic effects.[5,6]

Antiferromagnetic resonance (AFMR) in the vicinity of an orientational phase transition induced by a strong magnetic field was first observed in $YFeO_3$ in Ref. 3. Theoretical calculations have shown that the AFMR spectrum of orthoferrites consists of two optical branches whose activation energies in the absence of a magnetic field depend on the equilibrium configurations of the magnetic moment that is realized in the specific orthoferrite; these branches are located as a rule in the submillimeter band. Fourier spectroscopy was used[4] to study the spin-wave spectrum of yttrium orthoferrite, and resonant absorptions due to both spin-wave branches were observed. Raman scattering of light was used[7,8] to investigate the spin-wave spectrum of a number of rare-earth orthoferrites at small wave numbers.

We report here an investigation of the temperature dependence of the high-frequency AFMR branch in a zero magnetic field, as well as a study of the distinguishing features of the Faraday effect at submillimeter wavelengths in $YFeO_3$.

2. EXPERIMENTAL PROCEDURE

To investigate the dynamics of magnetically ordered crystals in the submillimeter band we used, for the first time ever, a procedure based on quasioptic monochromatic backward-wave-oscillator (BWO) spectroscopy, which was developed and successfully used to study soft modes in ferroelectrics.[9] It seems to us that this procedure, which unlike the traditional one requires no external magnetic fields yet combines high resolution with a very broad investigation band, improves substantially the ability to study the dynamics and magneto-optics of magnetically ordered crystals in the submillimeter band. The procedure is based on obtaining the transmission spectra by comparing the intensities of the radiation passing through the quasi-optical channel with and without a plane-parallel-plate sample.

Submillimeter radiation generated by backward-wave tubes was shaped by a teflon lens, absorbing diaphragms, and polarizing one-dimensional reticular polarizing filters into a paraxial quasi-optical plane-parallel beam having a flat wavefront and with transverse dimensions $(15–20)\lambda$.

The sample was mounted in a frame whose displacement placed it in and out the path of the quasi-optical beam. The use of focusing lenses permitted investigation of samples with transverse dimensions $(3–5)\lambda$. The frame with the holder was placed inside a thermostat that permitted variation of the temperature in the interval 77–700 K. The radiation detector was an OAP-5M nonselective opto-acoustic receiver. In the Faraday-effect measurements the sample was placed in a solenoid that produced an axial magnetic field up to 500 Oe, and the rotation angle of the polarization plane was measured by rotating the analyzer at a fixed value of the magnetic field, thereby ensuring return to the initial intensity. The rotation-angle measurement accuracy was limited by the fluctuations of the radiation and amounted to $\pm 0.5°$. When anisotropic crystals were studied, the principal optical axes were aligned with the incident-radiation polarization plane by rotating the sample, placed between an analyzer crossed with a polarizer, around the axis of the optical beam until minimum transmission was reached.

3. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

Figure 1 shows the transmission spectrum of an yttrium orthoferrite sample of thickness $d = 1.210$ mm at room temperature in the 220–350 GHz band corresponding to the tuning range of one backward-wave tube. The presence of alternating maxima and minima in the transmission spectrum is a feature of low-absorption samples and is due to interference effects in the plane-parallel plate. The constancy of the oscillation period is evidence of absence of dispersion of the refractive index. From the known frequency dependence of the transmission coefficient (see, e.g., Ref. 9) one can find the refractive index and absorption coefficient. Our measurements have shown that the principal values of the refractive

 0038-5646/84/020373-03$04.00

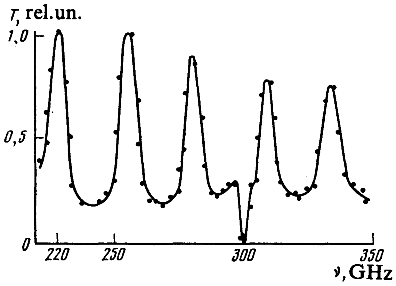

FIG. 1. Transmission spectrum of $YFeO_3$..

index for $YFeO_3$ are: $n_a = 4.807 \pm 0.005$, $n_b = 4.578 \pm 0.005$, $n_c = 5.050 \pm 0.005$. The absorption coefficients for the three principal crystallographic axes are practically the same at $k = 0.001 \pm 0.002$. The deep maximum at 330 GHz is due to the excitation of a low-frequency AFMR branch, as is evidenced by the temperature dependence of the AFMR frequency (Fig. 2), which tends to zero when the Néel temperature $T_N = 643$ K is approached. The AFMR absorption linewidth varied from sample to sample in the range 2.5–6 GHz at room temperature. The AFMR linewidth of some samples increased with decreasing temperature, and the absorption line intensity decreased and vanished completely at temperatures below 150 K. This behavior is typical of the "slow" relaxation mechanism[10] and is apparently due to the presence of monovalent iron ions in the samples.[11]

It is known that below the electric dipole transition frequencies a frequency-independent Faraday effect is observed[12] in antiferromagnets with uncomensated magnetic moment, with

$$\alpha_\Phi = (2\pi n/c)\gamma M,$$

leading to a rotation angle $\sim 1°$ for an $YFeO_3$ sample 1.210 mm thick magnetized to saturation and having the refractive indices cited above. However, the presence of low-frequency AFMR in $YFeO_3$ at submillimeter wave gave grounds for expecting a deviation from the known regularity in this spectral interval.

$YFeO_3$ is an optically biaxial crystal and, to exclude birefringence of the Faraday effect, was investigated by us with the light propagating along the optical axis. It is easy to deduce from the values of the principal refractive indices that in the submillimeter band the optical axes are located in the (100) plane and make an angle $\approx 45°$ with the [001] axis. It is remarkable that despite the more than doubling of the principal refractive indices on going from the IR to the microwave band, the directions of the optical axes change quite insignificantly. This is evidence of a very weak dispersion of the optical-axis direction from the nearest IR (Ref. 13) to the submillimeter band.

The plane-parallel sample perpendicular to the optical axis was cut from a $YFeO_3$ single crystal oriented beforehand in x-ray apparatus. The deviation of the optical axis from normal to the surface did not exceed 2°. The optical axis was aligned exactly with the radiation-propagation directions in the channel by superimposing the interference transmission maxima for two mutually perpendicular directions of the radiation polarization plane.

The frequency dependence of the Faraday effect at room temperature, for an yttrium orthoferrite sample magnetized to saturation, is shown in Fig. 3. The oscillations in this dependence are also due to interference effects inside the plane-parallel sample. The heights of the interference maxima, however, increase greatly as the AFMR frequency is approached, thus attesting to a strong increase of the specific rotation angle because of the influence of the antiferromagnetic resonance. This circumstance causes the specific rotation in $YFeO_3$ near the low-frequency AFMR branch to increase even above the specific rotation of iron-yttrium garnet, although the magnetization saturation of the latter is smaller by more than an order of magnitude.[14] We shall show that this behavior is typical of the Faraday effect near the low-frequency AFMR branch in orthorhombic antiferrmagnets with weak ferromagnetism, and can be explained by spin-wave theory.

The Hamiltonian for the iron sublattice of an orthoferrite can be written in the form

$$\mathscr{E} = 2M_0\mathscr{H},$$

$$\mathscr{H} = \frac{E}{2} m^2 - D(m_z l_x - m_x l_z) + \frac{a_1}{2} l_x^2 + \frac{c_1}{2} l_z^2 + \frac{a_2}{4} l_x^4 + \frac{c_2}{4} l_z^4 + \frac{f}{2} l_x^2 l_z^2 - \mathbf{m}\mathbf{H}_0.$$

Here

$$\mathbf{m} = \frac{\mathbf{M}_1 + \mathbf{M}_2}{2M_0}, \quad \mathbf{l} = \frac{\mathbf{M}_1 - \mathbf{M}_2}{2M_0},$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetic moments of the sublattices and M_0 their value at the given temperature T; E and D

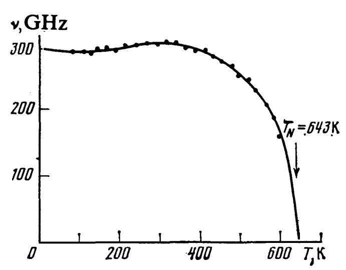

FIG. 2. Temperature dependence of the AFMR frequency of $YFeO_3$.

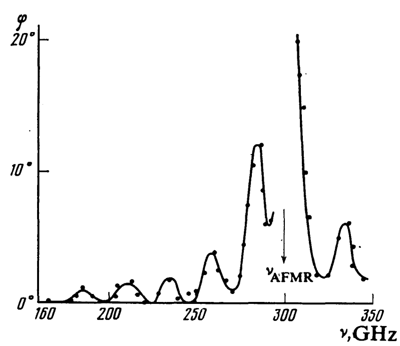

FIG. 3. Spectral dependence of the rotation angle of the polarization plane of an $YFeO_3$ sample 1.21 mm thick.

are respectively the effective fields of the symmetric and antisymmetric exchange; a_i, c_i, and f are the effective anisotropy fields.

Following Ref. 3, we shall assume that a_i, c_i, f, $D \ll E$. Analysis of the problem of forced oscillations of magnetic moments by starting from the Landau-Lifshitz equation of motion, without allowance for damping, under the condition that H_0 is directed along z, leads to the following systems of equations for small-oscillation amplitudes:

$$\begin{gathered} i\omega\mu_x = -(\omega_D+\omega_0)\mu_y + 2M_0\gamma m h_y, \\ i\omega\mu_y = -\omega_{A0}\lambda_z + \omega_0\mu_x - 2M_0\gamma m h_x, \\ i\omega\lambda_z = \omega_E\mu_y - 2M_0\gamma h_y; \end{gathered} \tag{1}$$

$$i\omega\mu_z = \frac{\omega_D(\omega_D+\omega_0) - \omega_{Ax}\omega_E}{\omega_E}\lambda_y, \tag{2}$$

$$i\omega\lambda_x = 0, \quad i\omega\lambda_y = -\omega_E\mu_z + \omega_D\lambda_x + 2M_0 h_z.$$

Here μ and λ are the amplitudes of the small oscillations of the vectors $\mathbf{m}$ and $\mathbf{l}$ about the equilibrium values $\mathbf{m}_0$ and $\mathbf{l}_0$, while h_i is the amplitude of the magnetic component of the electromagnetic field and $\omega_i = \gamma H_i$, where H_i are the values of the corresponding effective internal magnetic fields and the external one. The remaining notation is that of Ref. 3.

The system (1) has at

$$\omega_1^2 = \omega_{A0}\omega_E + \omega_0(\omega_0 + \omega_D)$$

a nontrivial solution that corresponds to the low-frequency AFMR branch. Analysis of the character of the magnetic-moment motion described by the system (1) shows that the total magnetization $\mathbf{m}$ processes about the z axis. The susceptibility tensor takes in this case the form

$$\{\chi\} = \begin{vmatrix} \chi_{xx} & i\chi_a & 0 \\ -i\chi_a & \chi_{yy} & 0 \\ 0 & 0 & 0 \end{vmatrix},$$

$$\chi_{xx} = 2M_0\gamma\frac{\omega_1^2/\omega_E}{\omega_1^2-\omega^2},$$

$$\chi_{yy} = 2M_0\gamma\frac{\omega_E m^2}{\omega_1^2-\omega^2},$$

$$\chi_a = 2M_0\gamma\frac{\omega m}{\omega_1^2-\omega^2}$$

It is easily seen that the off-diagonal component of the tensor χ_a, which is in fact responsible for the gyrotropic properties, increases when ω approaches the AFMR frequency, as is indeed observed in experiment.

In contrast to the case considered above, the system (2), to which corresponds the high-frequency AFMR branch $\omega_2^2 = \omega_D(\omega_D + \omega_0) - \omega_{Ax}\omega_E$, describes vibrational motion of the vector $\mathbf{m}$ along the z direction. It follows hence that no Faraday-effect anomalies should be observed near the resonance frequency ω_2. To confirm this statement experimentally the Faraday effect should be measured in the high-frequency AFMR region (Refs. 4, 7, 8).

In conclusion, the authors are sincerely grateful to G. V. Kozlov and S. P. Lebedev for help and consultation with the procedural problems and to A. M. Balbashov for supplying the samples.

[1] K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, Orientatsionnye perekhody v redokozemel'nykh magnetikakh (Orientational Transitions in Rare-Earth Magnets), Nauka, 1979.
[2] M. V. Chetkin, S. I. Gradetskii, and A. I. Akhutkina, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 373 (1982) [JETP Lett. **35**, 459 (1982)].
[3] V. I. Ozhogin, V. G. Shapiro, K. G. Gurtovoĭ, E. A. Galstyan, and A. Ya. Chervonenkis, Zh. Eksp. Teor. Fiz. **62**, 2221 (1972) [Sov. Phys. JETP **35**, 1162 (1972)].
[4] L. V. Velikov, E. A. Vitvinin, G. E. Ivannikova, F. F. Igoshin, A. P. Kir'yanov, and S. S. Markianov, Fiz. Tverd. Tela (Leningrad) **22**, 3612 (1980) [Sov. Phys. Solid State **22**, 2115 (1980)].
[5] B. B. Krichevtsov, K. M. Mukimov, R. V. Pisarev, and M. M. Ruvenshteĭn, Pis'ma Zh. Eksp. Teor. Fiz. **34**, 399 (1981) [JETP Lett. **34**, 379 (1981)].
[6] M. V. Chetkin and Yu. I. Shcherbakov, Fiz. Tverd. Tela (Leningrad) **11**, 1620 (1969) [Sov. Phys. Solid State **11**, 1314 (1969)].
[7] R. N. White, R. J. Nemanich, and C. Herring, Phys. Rev. **B25**, 1822 (1982).
[8] N. Koshizuka and K. Hayashi, J. Magn. and Magn. Mater. **31-34**, 569 (1983).
[9] A. A. Volkov, G. V. Kozlov, S. P. Lebedev, and A. M. Prokhorov, FIAN Preprint No. 2, 1980.
[10] A. G. Gurevich, Magnitnyĭ rezonans v ferritakh i antiferromagnetikakh (Magnetic Resonance in Ferrites and Antiferromagnets), Nauka, 1973.
[11] E. M. Gyorgy, J. P. Remeika, and P. L. Wood, J. Appl. Phys. **39**, 3499 (1968).
[12] G. S. Krinchik and M. V. Chetkin, Usp. Fiz. Nauk **98**, 3 (1969) [Sov. Phys. Usp. **12**, 307 (1969)].
[13] M. V. Chetkin, Yu. S. Didosyan, A. I. Akhutkina, and A. Ya. Chervonenkis, Pis'ma Zh. Eksp. Teor. Fiz. **12**, 519 (1970) [JETP Lett. **12**, 363 (1970)].
[14] S. N. Gradetskiĭ, V. P. Kalantaryan, K. N. Kocharyan, V. G. Prpryan, E. L. Sarkisyan, and M. V. Chetkin, Fiz. Tverd. Tela (Leningrad) **23**, 1250 (1981) [Sov. Phys. Solid State **23**, 736 (1981)].

Translated by J. G. Adashko