Observation of a gap in the spin-wave spectrum in YFeO$_3$ upon an orientational transition in a magnetic field

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(Submitted 1 April 1985)
Pis'ma Zh. Eksp. Teor. Fiz. **41**, No. 9, 391–393 (10 May 1985)

During spin flip in YFeO$_3$ induced by a magnetic field **H**||**a**, the antiferromagnetic resonant frequency does not vanish. It is predicted theoretically that the observed gap will have the behavior $\nu_0 \sim \sqrt{\chi_\parallel}$. This transition, continuous in H, does not have the properties of a second-order phase transition.

Previous studies of the antiferromagnetic resonance (AFMR) in yttrium orthoferrite (YFeO$_3$) in the region of the spin-flip transition induced by a magnetic field have been restricted to a few frequencies.[1] According to Refs. 1 and 2, the AFMR frequency corresponding to the softening mode of homogeneous magnetization oscillations vanishes in the field (H_t) at which the transition from the angular phase to the G_zF_x phase occurs.[3] However, there has been no detailed study of how the AFMR frequencies depend on the applied field in the vicinity of this transition.

The experiments which we are reporting here demonstrate for the first time that there is a significant energy gap in the field H_t. The experimental apparatus is described in Ref. 4. The electromagnetic radiation (100–200 GHz, $\lambda = 3$–1.5 mm) is produced by backward-wave tubes. The wavelength is measured within 0.5% by a Fabry-Perot interferometer.[5] The magnetic field is produced with the help of the Solenoid apparatus of the Laboratory of High Magnetic Fields, Institute of General Physics, Academy of Sciences of the USSR.[6] A field calibration within 1% is carried out on the basis of the positions of the ESR lines in diphenylpicrylhydrazyl on the sample.

In the experiments we measure the positions of the absorption lines for various values of the applied magnetic field at $T = 293$ K. The YFeO$_3$ sample was synthesized by crucible-free zone melting with radiative heating[7] in the form of a plate with dimensions of $6 \times 3 \times 0.4$ mm, oriented perpendicular ($\pm 1°$) to the **a** axis of the crystal. The sample was annealed in oxygen and then cooled slowly. The AFMR lines of the annealed sample are regular in shape and have a width of 600–700 Oe, while the lines of the unannealed sample have a width of 1.5–2 kOe. Precise positioning of the sample in the magnetic field was achieved by maximizing the distance between the AFMR peaks at $H > H_t$ and $H < H_t$. The error in the orientation of the field along the **a** axis in the **ac** plane is 5′. A $\pm 2°$ deviation of the field in the **ab** plane caused no changes of any sort in the positions of the AFMR lines.

The experimental results are shown in Fig. 1. Curve 1 corresponds to the field dependence of the square AFMR frequency at the highest attainable precision in the orientation of the sample. Curves 2 and 3 were measured during 5′ and 30′ deviations, respectively, of the magnetic field in the **ac** plane. The minimum energy gap found in

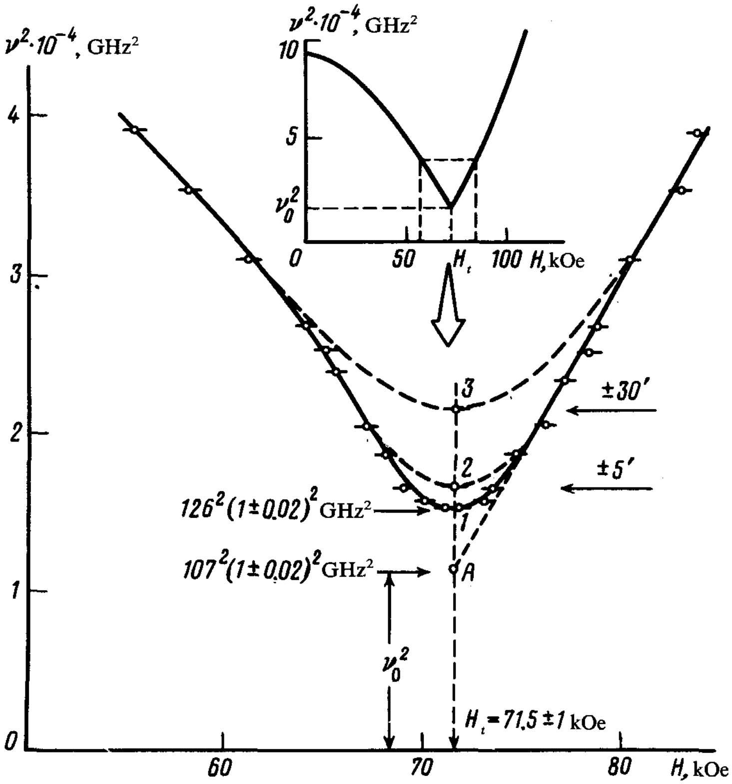

FIG. 1. Square of the AFMR frequency versus the external field. The inset shows the spectral region studied in the present experiments.

these experiments is $126\times(1\pm0.02)$ GHz. Extrapolation of $\nu(H)$ from fields $H>1.1H_t$ to the field of the spin-flip transition, H_t (point A in Fig. 1) yields $107\times(1\pm0.02)$ GHz; this value is stable in the face of a 2° field deviation in any direction.

These experimental results thus show that during a spin-flip transition induced by a magnetic field, with the magnetic field oriented highly precisely ($\pm 5'$) along the a axis of the crystal, the energy gap does not vanish in the spin-wave spectrum. At frequencies below ν_0, corresponding to the size of the energy gap, an absorption of electromagnetic radiation, which may be due to tails on the AFMR lines, is observed.

To interpret the experimental results, we use the nonequilibrium Landau thermodynamic potential written in accordance with the symmetry of an orthorhombic crystal[8,9,3]:

$$\begin{aligned}\Phi\,(\mathbf{M},\mathbf{L}) = {} & \Phi_0(\mathbf{L}^2) + \frac{1}{2}B\mathbf{M}^2 + \frac{1}{2}D(\mathbf{ML})^2 + d(M_xL_z - M_zL_x) - \mathbf{MH}\\ & + \frac{1}{2}a_1L_x^2 + \frac{1}{2}a_2L_y^2 + \frac{1}{2}a_3L_z^2 + \frac{1}{4}a_{11}L_x^4 + \frac{1}{4}a_{22}L_y^4 + \frac{1}{4}a_{33}L_z^4\\ & + \frac{1}{2}a_{12}L_x^2L_y^2 + \frac{1}{2}a_{13}L_x^2L_z^2 + \frac{1}{2}a_{23}L_y^2L_z^2\,,\end{aligned}$$

where $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2; \mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$; and $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations. A theoretical analysis shows that in the transition field H_t (at a finite temperature, $H_t = \chi_\perp(\chi_\perp - \chi_\parallel)^{-1}\times\{-H_D/2[H_D^2/4 + (\chi_\perp - \chi_\parallel)\chi_\perp^{-1}H_E H_{Aa}]^{1/2}\}$) the energy gap along the frequency scale is $\nu_0 = (2\pi)^{-1}\gamma(\chi_\parallel/\chi_\perp)^{1/2}H_t$, where $\chi_\perp^{-1} = B, \chi_\parallel^{-1} = B + DL_0^2, H_D = dL_0$, and $H_E H_{Aa} = \chi_\perp^{-1}L_0^2[a_3 - a_1 + (a_{33} - a_{13})L_0^2]$. To find this quantity, we use the simple thermodynamic equations of motion[10,11] under the condition $\mathbf{L}^2 = L_0^2$, where L_0 is the equilibrium value of the vector $\mathbf{L}$. In Fig. 1 and in the calculations, we used the value $\gamma_0 = ge/2mc$. for the kinetic coefficient γ.

Our estimates for the energy gap associated with the magnetoelastic interaction yield a value an order of magnitude smaller than that found experimentally.

In summary, this dynamic experiment has yielded a value for the ratio $\chi_\parallel/\chi_\perp$ for $YFeO_3$, an antiferromagnet with a slight ferromagnetism ($\chi_\parallel/\chi_\perp = 0{,}3 \pm 10\%$ at $T = 293$ K).

The theoretical analysis and the experimental results show that at $H = H_t$ the transition from the angular phase to the $G_z F_x$ phase does not exhibit the properties of a second-order phase transition: The AFMR frequency does not vanish on the line at which the angular solution loses its stability and on the coincident line at which the $G_z F_x$ phase becomes stable. The energy gap is thus nonzero at all temperatures to the extent that $(\chi_\parallel/\chi_\perp)^{1/2}$ is nonzero.

We are deeply indebted to Academician A. M. Prokhorov for constant interest in this study and for a discussion of the results. We sincerely thank V. G. Veselago and L. P. Maksimov for cooperation in the experiment using the Solenoid apparatus.

[1] V. I. Ozhogin, V. G. Shapiro, K. G. Gurtovoĭ, E. A. Galst'yan, and A. Ya. Chervonenkis, Zh. Eksp. Teor. Fiz. **62**, 2221 (1972). [Sov. Phys. JETP **35**, 1162 (1972)].
[2] A. A. Mukhin, Preprint No. 245, Institute of General Physics, Academy of Sciences of the USSR, 1984.
[3] K. P. Belov, A. K. Zvezdin, A. M. Kadomtseva, and R. Z. Levitin, Orientatsionnye perekhody v redkozemel'nykh magnetikakh (Orientational Transitions in Rare-Earth Magnets), Nauka, Moscow, 1979.
[4] E. G. Rudashevsky, A. S. Prochorov, and L. V. Velikov, IEEE Trans. Microwave Theory Tech. MTT-22, 1064 (1974).
[5] E. A. Vinogradov, E. M. Dianov, and N. A. Irisova, Pis'ma Zh. Eksp. Teor. Fiz. **2**, 320 (1965) [JETP Lett. **2**, 203 (1965)].
[6] V. G. Veselago, L. P. Maksimov, and A. M. Prokhorov, Prib. Tekh. Eksp. No. 4, 192 (1968).
[7] A. M. Balbashov, A. Ya. Chervonenkis, A. V. Antonov, and V. E. Bakhteuzov, Izv. Akad. Nauk SSSR, Ser. Fiz. **35**, 1243 (1971).
[8] I. E. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. **32**, 1547 (1957) [Sov. Phys. JETP **5**, 1259 (1957)].
[9] A. S. Borovik-Romanov, in: Antiferromagnetizm i ferrity. Ser. fix.-mat. nauk (Antiferromagnetism and Ferrites. Series of Physicomathemtical Sciences), Vol. 4, Izd. VINITI, Moscow, 1962.
[10] Yu. M. Gufan, Zh. Eksp. Teor. Fiz. **60**, 1537 (1971) [Sov. Phys. JETP **33**, 831 (1971)].
[11] E. G. Rudashevskiĭ, Tezisy dokladov na 16 Vsesoyuznoĭ konferentsii po fizike magnitnykh yavleniĭ (Proceedings of the Sixteenth All-Union Conference on the Physics of Magnetic Phenomena), Vol. 3, Tula, 1983, p. 150.

Translated by Dave Parsons