SOVIET PHYSICS JETP VOLUME 30, NUMBER 3 MARCH, 1970

STUDY OF THE INTERACTION BETWEEN TWO SPIN OSCILLATION MODES DURING ANTI-FERROMAGNETIC RESONANCE IN THE WEAKLY FERROMAGNETIC PHASE OF HEMATITE

L. V. VELIKOV, S. V. MIRONOV, and E. C. RUDASHEVSKIĬ

P. N. Lebedev Physics Institute, USSR Academy of Sciences

Submitted April 18, 1969

Zh. Eksp. Teor. Fiz. 57, 781–790 (September, 1969)

The interaction between two spin oscillation modes during antiferromagnetic resonance in the weakly ferromagnetic phase of hematite ($\alpha-Fe_2O_3$) near the intersection point of two magnetic moment oscillation branches is investigated in detail for wavelengths between 1.40 and 1.70 mm, magnetic field strengths up to 100 kOe, and temperatures between 290 and 310°K. The dependences of the resonance field on temperature, frequency, and orientation of the external magnetic field relative to the crystallographic axes of the sample are obtained. The experiments show that, just as in the case of $MnCO_3$, the branches corresponding to two different oscillation modes of the magnetic moments strongly interact upon deviation of the external magnetic field from the crystal basal plane, and repel each other. A dependence of the antiferromagnetic resonance frequencies on the magnitude of the applied magnetic field and its direction relative to the crystallographic axes is derived theoretically under the assumption that the Dzyaloshinskiĭ field responsible for the weak ferromagnetism is not zero. The calculation is in good agreement with the experimental results.

INTRODUCTION

A characteristic feature of rhombohedral antiferromagnets (space group D_{3d}^6), which include the hematite (α-Fe_2O_3) investigated in the present work, is, as shown by Borovik-Romanov[1] and Turov[2], the absence of an energy gap in one of the branches of the spin-wave spectrum, under the condition that the magnetic moments of the sublattices are directed perpendicular to the threefold axis of the crystal. In this case, a deviation of the magnetic moments of sublattices from rigorous antiparallelism is also observed and is explained by the Dzyaloshinskiĭ theory[3], together with an uncompensated magnetic moment directed perpendicular to the threefold axis. This weak ferromagnetic moment can be described by an effective Dzyaloshinskiĭ field H_D and a perpendicular magnetic susceptibility $\chi_\perp$:

$$\sigma = \chi_\perp H_D. \tag{1}$$

For the antiferromagnetic-resonance frequencies as functions of the applied external field H and on the angle φ between the basal plane perpendicular to the threefold axis and the direction of the magnetic field, one obtains from the Hamiltonian proposed by Dzyaloshinskiĭ[3] the expressions[1,2,4]

$$\omega_1/\gamma = \sqrt{H\cos\varphi(H\cos\varphi + H_D)}, \tag{2}$$

$$\omega_2/\gamma = \sqrt{2H_AH_E + H_D(H\cos\varphi + H_D) + H^2\sin^2\varphi}, \tag{3}$$

where H_A and H_E are the effective anisotropy and exchange magnetic fields, and γ is the gyromagnetic ratio.

The theoretical relation (2) was verified experimentally by a number of workers. In[5], where synthetic single crystals of sufficiently high grade were investigated (the width of the absorption line corresponding to the low-frequency antiferromagnetic resonance did not exceed 200 Oe at a wavelength 8 mm and 80 Oe at 2.5 cm), it was shown that the experimental results are well described by the relation

$$\omega_1/\gamma = \sqrt{H\cos\varphi(H\cos\varphi + H_D) + H_\Delta^2} \tag{4}$$

with values $H_D = 22.0 \pm 0.1$ kOe and H_Δ^2 independent of the direction of the external magnetic field in the basal plane. The origin of H_Δ^2 was explained in[6] on the basis of the Hamiltonian proposed by Borovik-Romanov and one of the authors. It was shown that the gap $H_\Delta^2 = 2H_{MC}H_B$, where H_{MC} is the effective magnetic field resulting from the magnetoelastic interaction. It was also shown that the magnetoelastic interaction should contribute also to the high-frequency branch, changing slightly the value of the effective anisotropy field H_A (without changing the form of relation (3)). The high-frequency resonance described by relation (3) was observed and investigated in detail for the case $\varphi = 0°$ in[7]. It was shown, in particular, that the quantity $2H_AH_E$ depends very strongly on the temperature when $T > 260°$K (a temperature change of 10° is equivalent to application of an external magnetic field ~30 kOe), with $2H_AH_E = \alpha T + \beta$, where $\alpha = 67 \pm 5$ kOe^2/deg, and $\beta = 16{,}900 \pm 800$ kOe^2.

Thus, using the foregoing theoretical relations and the experimental results, one can make assumptions concerning the magnitude of the external magnetic field and of the temperature at which the antiferromagnetic resonances corresponding to relations (3) and (4) will be observed at the same frequency, i.e., the branches of the oscillations intersect.

The intersection of the branches in antiferromagnetic resonance was observed by Borovik-Romanov and Prozorova in $MnCO_3$ ($T_N = 32.5°$K)[8,9]. They have shown that if the magnetic field is accurately oriented in the basal plane, the two modes of spin oscillations do not interact. But if the angle between the magnetic field and the basal plane differs from zero, then the oscillations do interact, and the following relation holds for the frequencies of the coupled oscillations:

$$\left(\frac{\omega}{\gamma}\right)^4-\left(\frac{\omega}{\gamma}\right)^2\left[\left(\frac{\omega_1}{\gamma}\right)^2+\left(\frac{\omega_2}{\gamma}\right)^2+H^2\sin^2\varphi\right]+\left(\frac{\omega_1}{\gamma}\right)^2\left(\frac{\omega_2}{\gamma}\right)^2=0, \quad (5)$$

where $\omega_1/\gamma = H$ and $\omega_2/\gamma = \sqrt{2H_AH_E}$, i.e., the values given by (2) and (3) under the assumption that $H_D = 0$ and $\varphi = 0°$.

The investigated antiferromagnet $MnCO_3$ has a very small Dzyaloshinskiĭ field H_D ($H_D = 4.4$ kOe)[1,10]. Therefore, bearing in mind the large magnitude of the external magnetic field at which antiferromagnetic resonance is observed ($H \approx 45$ kOe) in the region of the oscillation-branch intersection, it was possible to carry out the calculation and to compare it with experiment under the assumption that $H_D = 0$, In hematite, however, H_D is much larger[5] ($H_D = 22$ kOe), and one could therefore expect the appreciable value of H_D to change the relations characteristic of the interacting branches of the magnetic oscillations.

In addition, hematite is a very convenient object for a detailed study of the interaction of two oscillation modes, in view of the foregoing features: first, antiferromagnetic resonance in hematite is observed at room temperature; second, the frequency of the high-frequency resonance can be readily varied by varying the temperature, making it possible to choose convenient ranges of the frequencies and of the fields; third, the sufficiently narrow absorption lines[7] make it possible to determine accurately their position, and fourth, the entire temperature interval in which the interaction can be observed is quite small compared with T_N ($T_N = 974°K$ for α-Fe_2O_3), by virtue of the large rate of change of the frequency of the upper branch with changing temperature.

Thus, it seemed advisable to investigate in detail the antiferromagnetic resonance in α-Fe_2O_3 in order to determine the qualitative and quantitative regularities that occur when the two types of magnetic oscillations interact.

SAMPLES AND EXPERIMENTAL PROCEDURE

We used hematite single crystals grown by the Institute of Solid State Physics of the Czechoslovak Academy of Sciences; these were previously used in[5,6,7] 1). The samples were in the form of thin plates measuring 2 × 1 × 0.2 mm.

The experiments were performed with a direct-amplification spectrometer 2), similar to that used in[7], in the wavelength range 1.40–1.70 mm and in a constant magnetic field up to 100 kOe, obtained with the water-cooled FIAN solenoid[11]. The wavelength of the radiation was measured with a Fabry-Perot interferometer with metallic grids[12].

The sample was placed on a foamed-polystyrene holder, which could be rotated in the waveguide in such a way as to vary the orientation of the external constant magnetic field relative to the basal plane. The rotating device made it possible to set the sample with accuracy ±10′. This device had no metallic elements protruding into the interior of the waveguide, since the sample was set in rotation by a small rotating part of the narrow wall of the waveguide, which was mounted flush with the interior surface. To decrease the influence of standing waves, decoupling attenuators were placed in the entire submillimeter channel, including the sections near the sample; these decreased quite strongly the power passing through the channel. To ensure reliable operation with small useful signals, very sensitive and low-noise receivers of indium antimonide (InSb), cooled to liquid-helium temperature, were used. The absorption signals, as usual, were recorded on an x-y plotter, with the voltage corresponding to the magnitude of the magnetic field being applied to the plotter directly from a shunt through which the total solenoid current ($I_{max} = 10$ kA) flowed. This method of scanning the magnetic field, as established in[13], ensured a linearity within 1%. The external magnetic field was calibrated by EPR in DPPH at all the operating frequencies during each measurement. The sample temperature was measured with a thermocouple and stabilized in the 290–310°K interval accurate to ±0.2° with the aid of the system described in[14].

EXPERIMENTAL RESULTS

As indicated above, the main purpose of the investigation was to study in detail the connection between the two modes of the magnetic moments near the intersection points of two branches of antiferromagnetic resonance. Bearing in mind the strong temperature dependence of the energy gap for the upper branch of the AFMR branch, two series of measurements were performed.

In the first series of experiments, the region of intersection of the magnetic-moment oscillation branches was scanned by varying the temperature of the sample at a fixed frequency of the electromagnetic radiation. A typical record of the absorption lines in this case is shown in Fig. 1, which shows also clearly the DPPH marker on the right side. It was observed that the values of the resonant fields depend strongly on the angle φ between the basal plane of the crystal and the direction of the magnetic field. The shown absorption lines correspond to $\varphi = 2°30'$. The corresponding temperature dependences of the resonant fields are shown in Fig. 2. As seen from the figure, far from the point of intersection of the oscillation branches, one branch ("low-frequency" in the previously employed terminology) is practically independent of the temperature. It is represented in the figure by the dashed line drawn parallel to the abscissa axis. The "high-frequency" strongly-temperature-dependent branch corresponds to the other dashed line. Since an interaction takes place between the oscillations when the magnetic field orientation is inclined to the basal plane, the "low-frequency" absorption line goes over into the "high-frequency" one with increasing temperature, and vice versa, whereas a broadening of one of the absorption lines is observed in the immediate

1) The authors are grateful to M. Vihr of the institute of Solid State Physics of the Czechoslovak Academy of Sciences for supplying the single crystals.

2) The authors are grateful to A. P. Bazhulin and E. A. Vinogradov of the Lebedev Physics Institute for help in constructing the spectrometer.

vicinity of the intersection point (see the plot corresponding to T = 294.2°K in Fig. 1).

The other series of experiments was performed at a constant temperature (T = 300.1°K) and at several fixed frequencies. At each frequency we investigated the dependence of the magnetic field, in which the resonance was observed, on the angle φ between the basal plane and the direction of the magnetic field. The experimental points obtained at wavelengths λ equal to 1.45, 150, 1.53, 1.59 and 1.69 mm are shown in Fig. 3. The strongest interaction corresponds to Fig. 3c.

Figure 4 shows plots of the antiferromagnetic resonance frequencies against the magnetic field (γ corresponds to g = 2.0) for the angles 0°, 6°, and 12°. As seen from the figure, at φ = 0° the lines pass one through the other without interacting. At $\varphi \neq 0°$, the "low-frequency" branch goes over into the "high-frequency" branch, and vice versa, in analogy with the foregoing, but this now is a result of the change of frequency and not of temperature.

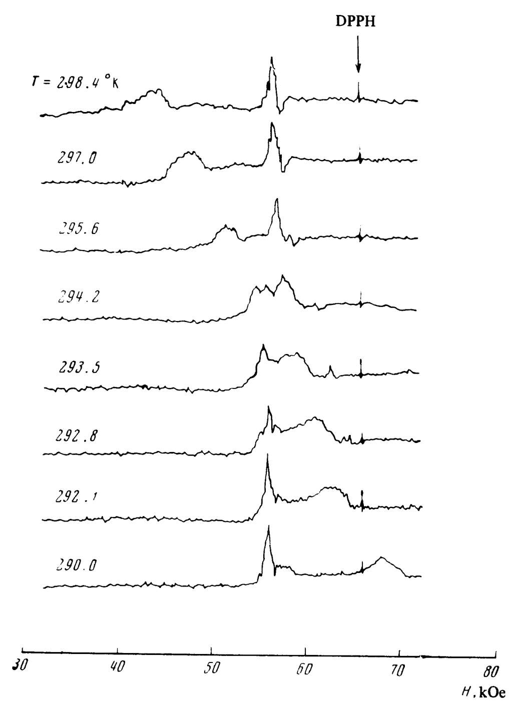

FIG. 1. Absorption lines for two branches of antiferromagnetic resonance at different temperatures. The angle between the direction of the magnetic field and the basal plane is φ = 2°30′, the wavelength of the electromagnetic radiation is λ = 1.60 mm.

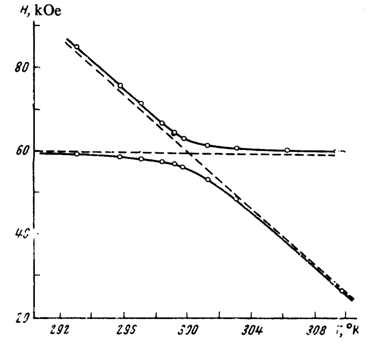

FIG. 2. Dependence of the resonance-absorption field on the temperature; φ = 2°30′, λ = 1.52 mm.

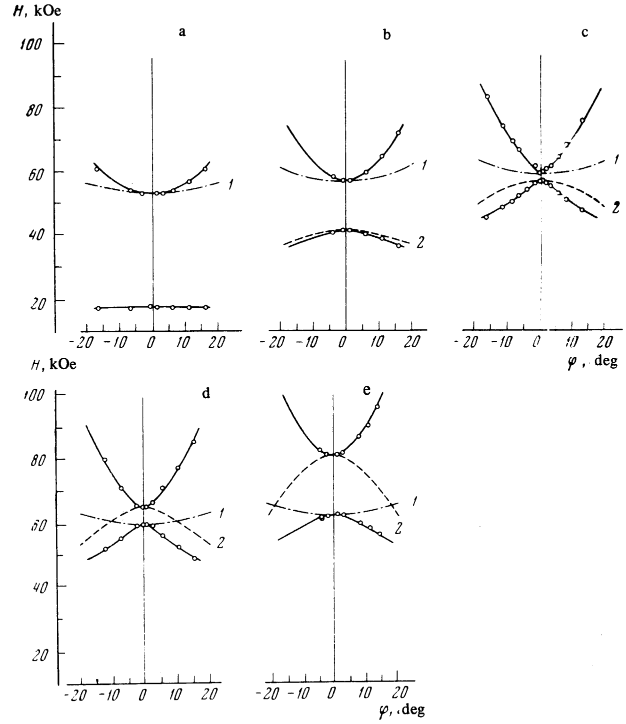

FIG. 3. Dependence of the resonant field on the angle φ. Wavelength of the electromagnetic radiation: a – 1.69 ± 0.01 mm; b – 1.59 ± 0.01 mm; c – 1.53 ± 0.01 mm; d – 1.50 ± 0.01 mm; e – 1.45 ± 0.01 mm; T = 300.1° K. The dashed lines are based on formulas (3) and (4), and the solid lines are the result of calculation by means of formulas (11).

To illustrate the appearance of the interaction following the deviation from φ = 0°, special absorption line curves were obtained as shown in Fig. 5. When the magnetic field lies exactly in the basal plane (φ = 0°), the absorption lines coincide, and the more intense line has a smaller width. But even a very small deflection of the magnetic field away from the basal plane (±1°30′) greatly broadens the narrower of the absorption lines. This is evidence of a strong mutual influence of the oscillations when the interaction turned off, realized by deflecting the magnetic field away from the direction φ = 0°.

DISCUSSION

To explain the results it is necessary to perform a calculation by using the Landau-Lifshitz equations and the well known phenomenological Hamiltonian[3] for rhombohedral antiferromagnets with weak ferromagnetism

$$\mathcal{H} = {}^1\!/_2 B\mathbf{m}^2 + {}^1\!/_2 a l_z^2 + q(l_x m_y - l_y m_x) - \mathbf{mh}, \qquad (6)$$

where $\mathbf{m} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$ is the magnetic moment of the antiferromagnet ($\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations, with $\mathbf{M}_1^2 = \mathbf{M}_2^2 = \mathbf{M}_0^2$); $\mathbf{l} = (\mathbf{M}_1 + \mathbf{M}_2)/2M_0$ is a vector defining the direction of the magnetic moments of the sublattices in the antiferromagnetic stage; $\mathbf{m} \cdot \mathbf{h} = 2M_0\mathbf{m} \cdot \mathbf{H}$ is the energy of the antiferromagnet in the external magnetic field. The OZ axis coincides with the threefold axis of the crystal. The previously introduced effective fields are expressed in terms of the Hamiltonian constants as follow:

$$2H_A H_E = aB / 4M_0^2, \quad H_D = q / 2M_0. \tag{7}$$

Calculation shows that in the case when $H_D \neq 0$ ($q \neq 0$) the following equation is obtained for the natural frequencies of the antiferromagnet

$$\left(\frac{\omega}{\gamma}\right)^4 - \left(\frac{\omega}{\gamma}\right)^2\left[\left(\frac{\omega_1}{\gamma}\right)^2 + \left(\frac{\omega_2}{\gamma}\right)^2 + H_z^2\right] + \left(\frac{\omega_1}{\gamma}\right)^2\left(\frac{\omega_2}{\gamma}\right)^2 + H_D H_\perp H_z^2 = 0, \tag{8}$$

where ω_1 and ω_2 are determined by the formulas for the frequencies of the antiferromagnetic resonance with an external magnetic field lying in the basal plane:

$$\omega_1 / \gamma = \sqrt{H_\perp (H_\perp + H_D)}, \tag{9}$$

$$\omega_2 / \gamma = \sqrt{2H_A H_E + H_D (H_\perp + H_D)}. \tag{10}$$

In the foregoing equations, $H_Z = H \sin\varphi$ and $H_\perp = H \cos\varphi$ are the projections of the applied magnetic field on the basal plane and the threefold axis.

By using formulas (9) and (10) we can calculate the natural frequencies of the oscillations of the antiferromagnet at any value of the angle between the basal plane and the direction of the magnetic field; these values hold true for any mutual arrangement of the branches ω_1 and ω_2:

$$\left(\frac{\omega}{\nu}\right)^2 = \frac{1}{2}\left[\left(\frac{\omega_1}{\gamma}\right)^2 + \left(\frac{\omega_2}{\gamma}\right)^2 + H_z^2\right] \pm \left\{\frac{1}{4}\left[\left(\frac{\omega_2}{\gamma}\right)^2 - \left(\frac{\omega_1}{\gamma}\right)^2 + H_z^2\right]^2 + H_\perp^2 H_z^2\right\}^{1/2}. \tag{11}$$

When the natural frequencies ω_1 and ω_2 are close or equal, the first term in the radicand is small and the formulas for the frequencies of the coupled oscillations have a form different from (2) and (3). But if it is assumed that ω_1 and ω_2 differ greatly, then the term $H_\perp^2 H_Z^2$ in the radicand can be neglected, and we obtain the previously known expressions (2) and (3). It should also be noted that when $\varphi = 0°$ and $\varphi = 90°$, the obtained relations coincide exactly with (2) and (3).

In expression (11) for the natural frequencies of the antiferromagnetic, the Dzyaloshinskiĭ field H_D, which causes the weak ferromagnetism, enters only via ω_1 and ω_2. This allows us to conclude that in the absence of weak ferromagnetism (i.e., $H_D = 0$) the form of the formulas for the coupled oscillations remains unchanged (it is assumed that anisotropy of the "easy plane" type obtains). The results of the calculation with $H_D \neq 0$ also confirm the conclusions of[9], namely that there is no interaction when the external magnetic field lies exactly in the basal plane. In view of the fact that expression (11) contains the frequencies ω_1 and ω_2, it is necessary to use the relations (3) and (4) obtained in[5,7] when the aforementioned relation for these frequencies is experimentally verified. The solid curves of Fig. 3 were calculated from formulas (11)[3)] using (3) and (4), and the values of the effective fields H_D, $2H_A H_E$, and H_Δ^2 were determined from our experiments at $\varphi = 0°$ and $T = 300.1°K$.

As can be seen from Fig. 3, the agreement with the calculation is good. The dashed and dash-dot curves in the same figure describe the known relations (3) and (4). The numbers 1 and 2 denote the non-interacting

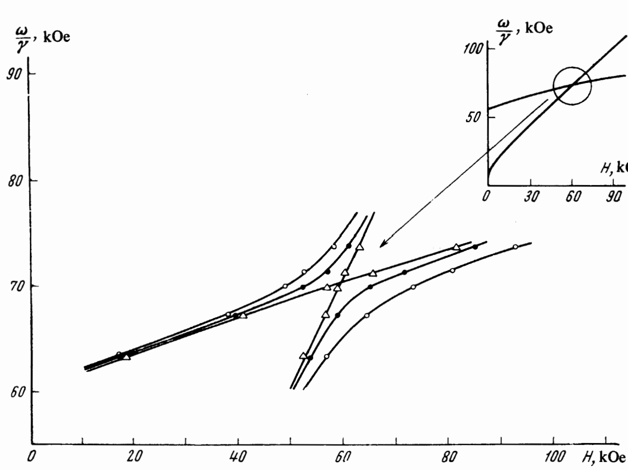

FIG. 4. Spectrum of antiferromagnetic resonance near the intersection point: Δ – $\varphi = 0°$; ● – $\varphi = 60°$; ○ – $\varphi = 12°$. Solid lines – result of calculation of formulas (11).

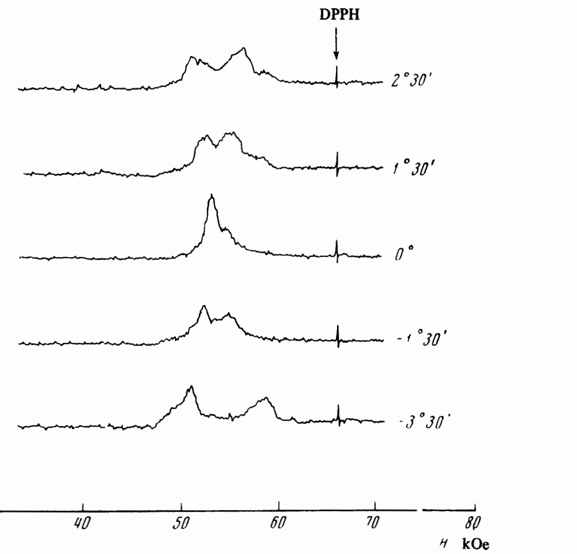

FIG. 5. Absorption lines for two branches of antiferromagnetic resonance at different values of the interaction, which is determined by the angle of inclination of the magnetic field to the basal plane of the crystal, near the intersection point.

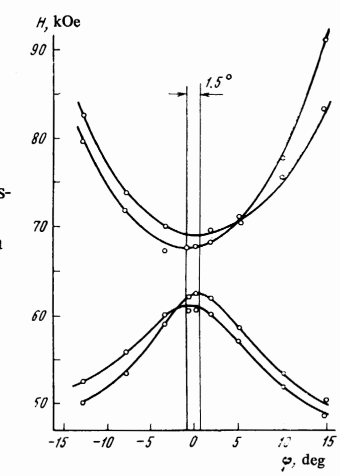

FIG. 6. Dependence of the resonant fields on the angle φ for a crystal having a fine structure as a result of the presence of blocks.

---

3) The authors are grateful to A. T. Matachun for help with the computer calculations.

branches of the oscillations ω_1 and ω_2. Owing to the interaction, the splitting at $\varphi \neq 0°$ increases as the exact values of the fields and frequencies corresponding to the intersection of the oscillation branches are approached. In addition, the interaction explains the deviation from the $1/\cos\varphi$ dependence, which is characteristic of the low-frequency branch[5], and which was observed by Elliston and Troup[15]. This deviation, owing to the temperature dependence of the low-frequency branch, also depends on the temperature, although in the previously obtained relations (2) and (3) the effective fields H_D and H_Δ^2 do not depend on the temperature.

Above the intersection point, when $\varphi \neq 0°$, the branches of the oscillations go over one into the other, and to explain the experimental results it is therefore necessary to use already not the relations (3) and (4), but expression (11). This is seen most clearly in Fig. 3e, where the solid and dashed lines vary quite differently with increasing angle φ. It should be noted that in the investigation of antiferromagnetic resonance at $\varphi = 0°$, in the presence of another closely-lying level (ω_2), deviations from the known expressions (3) and (4) are possible, owing to the inaccurate angle setting of the sample relative to the magnetic field, and a temperature dependence of the lower branch (ω_1) may appear. This phenomenon should be borne in mind not only in the study of hematite, but also in the investigation of other antiferromagnets. Indeed, to explain antiferromagnetic resonance one customarily uses formulas that do not take the mutual placement of the spin-wave branches into account.

The dependence of the antiferromagnetic resonance frequencies on the external magnetic field for three values of the angle φ are shown in Fig. 4.

We have investigated in passing the influence of the high-frequency branch of the antiferromagnetic resonance on the width of the absorption lines of the low-frequency branch. The experiments have shown that the second branch does not exert any noticeable influence, with the exception of a certain vicinity of the intersection point, where a broadening of the antiferromagnetic resonance line, corresponding to 1, is observed.

Absorption lines were observed having a fine structure in certain samples. For two corresponding fine-structure lines we plotted the angular dependence shown in Fig. 6. As can be seen from the figure, the extrema for the two pairs of curves differ from each other in direction by approximately 1–2°, thus indicating that the individual fine-structure lines pertain to two blocks, the threefold axes of which make an angle of several degrees.

In conclusion, the authors are deeply grateful to A. M. Prokhorov for continuous interest and discussions, and are sincerely indebted to A. S. Borovik-Romanov for interest in the work and fruitful discussions. The authors are grateful to N. A. Irisova for help in mastering the submillimeter-band techniques, to V. G. Veselago and L. P. Maksimov for collaboration in the work with strong magnetic fields, and also to N. P. Lopukhov and A. G. Khar'kovtsev for preparing the apparatus and help with the experiments.

[1] A. S. Borovik-Romanov, Zh. Eksp. Teor. Fiz. **36**, 766 (1959) [Sov. Phys.-JETP **9**, 539 (1959)].

[2] E. A. Turov, ibid **36**, 1254 (1969) [**9**, 890 (1959)].

[3] I. E. Dzyaloshinskiĭ, Journal Phys. Chem. Solids **4**, 241 (1958).

[4] E. A. Turov and N. Guseĭnov, Zh. Eksp. Teor. Fiz. **38**, 1326 (1960) [Sov. Phys.-JETP **11**, 953 (1960)].

[5] E. G. Rudashevskiĭ and T. A. Shal'nikova, ibid **47**, 886 (1964) [**20**, 593 (1965)].

[6] A. S. Borovik-Romanov and E. G. Rudashevskiĭ, ibid **47**, 2095 (1964) [**20**, 1407 (1965)].

[7] L. V. Velikov and E. G. Rudashevskiĭ, ibid. **56**, 1557 (1969) [**29**, 836 (1969)].

[8] A. S. Borovik-Romanov and L. A. Prozorova, ZhETF Pis. Red. **4**, 57 (1966) [JETP Lett. **4**, 39 (1966)].

[9] L. A. Prozorova and A. S. Borovik-Romanov, Zh. Eksp. Teor. Fiz. **55**, 1727 (1968) [Sov. Phys.-JETP **28**, 910 (1969)].

[10] A. S. Borovik-Romanov, N. M. Kreĭnes, and L. A. Prozorova, Zh. Eksp. Teor. Fiz. **45**, 64 (1963) [Sov. Phys.-JETP **18**, 46 (1964)].

[11] V. G. Veselago, L. P. Maksimov, and A. M. Prokhorov, PTÉ No. 4, 192 (1968).

[12] E. A. Vinogradov, E. M. Dianov, and N. A. Irisova, ZhETF Pis. Red. **2**, 323 (1965) [JETP Lett. **2**, 205 (1965)].

[13] L. V. Velikov, V. G. Veselago, M. V. Glushkov, V. M. Ivanov, Yu. V. Kosichkin, A. M. Prokhorov, E. G. Rudashevskiĭ, and V. I. Chernykh, PTÉ No. 3, 187 (1969).

[14] S. V. Mironov, E. G. Rudashevskiĭ, and V. I. Chernykh, ibid. No. 5, 1969.

[15] P. R. Elliston and G. J. Troup, J. Phys., C (Proc. Phys. Soc.) **12**, 169 (1968).

Translated by J. G. Adashko
89