Antiferromagnetic resonance in $FeBO_3$

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We investigated antiferromagnetic resonance (AFMR) in the weakly ferromagnetic single crystal $FeBO_3$ in the wavelength range 14–0.5 mm in magnetic fields up to 100 kOe and in the temperature interval from 4.2 to 350 °K. We studied the temperature dependence of the energy gap of magnetoelastic origin in the low-frequency branch of the AFMR. The high-frequency branch of the AFMR was obtained experimentally, and its study has made it possible to determine the value of its energy gap H_c ($T \to 0$ °K) = 165.7 ± 0.2 kOe (g factor equal to 2), and also the temperature dependence of $H_{AE}(T)$, which turned out to agree well with the temperature dependence of the spontaneous magnetization. The obtained temperature dependences of the effective Dzyaloshinskiĭ fields from the low-frequency and high-frequency branches of the AFMR differ from each other. An appreciable difference was observed in the temperature dependences of the resonance-absorption linewidths of the high-frequency and the low-frequency AFMR.

INTRODUCTION

We have investigated antiferromagnetic resonance (AFMR) experimentally in weakly ferromagnetic iron borate. This investigation is of interest both for explanation of the influence of various types of interactions on AFMR in weakly ferromagnetic structures, and for study of the dependence of the physical parameters characterizing $FeBO_3$ as a magnetic crystal on the external conditions (for example, on the magnetic fields, temperatures, pressures, etc.) and for comparison of these parameters with the theoretical ones.

Iron borate is a rhombohedral antiferromagnet with weak ferromagnetism[1]. $FeBO_3$ was synthesized recently[2] and has been investigated less thoroughly than other antiferromagnets of this group. The great interest presently evinced in this compound is due to the fact that $FeBO_3$ is transparent in the visible region of the spectrum and remains at the same time weakly ferromagnetic up to the temperature T_N = 348°K[3]. There are only two presently known compounds, $FeBO_3$ and FeF_3, which have a spontaneous moment at room temperature and are at the same time transparent in the visible region of the spectrum.

From the point of view of experimental study, $FeBO_3$ offers a number of advantages over the previously investigated (including by the AFMR method) antiferromagnets[4] that are isomorphic to $FeBO_3$. Indeed, the Neel temperature of $FeBO_3$ (T_N = 348°K) lies in a temperature range that is convenient for the investigation of critical phenomena, since it greatly exceeds the Neel temperatures of the known weakly ferromagnetic carbonates $MnCO_3$ (T_N = 32°K), $CoCO_3$ (T_N = 18°K), $NiCO_3$ (T_N = 22°K). In addition, $FeBO_3$ is an ideal object for correct quantitative comparison of the conclusions of the spin-wave theory with the experimental results, since the temperature region in which the conclusions of the spin-wave theory are usually assumed to be valid ($T \lesssim 0.1\ T_M$) is quite wide for $FeBO_3$ (Θ = 35°K).

An important feature of $FeBO_3$ is the large value of the Dzialoshinskiĭ interaction, which is just as large as in $CoCO_3$ and 20 times larger than in hematite; this greatly favors the study of the manifestations of this interaction. It should be noted that in $CoCO_3$ the features of the splitting of the orbital levels in the crystal field (incompletely quenched orbital angular momentum) greatly complicate the theoretical interpretation of the experimental results.

Before this investigation was started, there were very few studies of the magnetic properties of $FeBO_3$. Following the reported synthesis of $FeBO_3$, reports were published of static measurements of the magnetization of polycrystalline samples[5], neutron-diffraction investigations[6], and also studies of the temperature dependence of the position of the resonance-absorption line in $FeBO_3$ at 34.5 GHz[7]. This resonance absorption was treated by the authors as ferromagnetic resonance in a ferromagnet with an easy-plane anisotropy. On the other hand, the data of[5], and also the neutron-diffraction results[6], favored the assumption that $FeBO_3$ is an antiferromagnet with weak ferromagnetism. In view of the foregoing, it was of interest to study the resonance in a wide range of frequencies and magnetic fields. It was also of interest to study the frequency-field dependences of the resonance absorption in a wide temperature interval, so as to determine the limits of applicability of the theoretical dependences of the resonance-absorption frequencies on the external magnetic field, which are usually obtained under the assumptions of spin-wave theory, which are valid at $T \ll T_N$.

Simultaneously with the publication of the first results of our research[8, 9], a number of reports appeared on the properties of $FeBO_3$. Investigations were made of the magnetoelastic interaction[10], a nuclear spin echo was observed and studied[11], and the magnetization of single-crystal samples by static methods was studied[10, 12]. Thus, the research performed has made it possible to compare results obtained with different samples as well as to interpret simultaneously experimental results obtained by different methods.

For the case when the external magnetic field is perpendicular to the ternary axis of the crystal ($\mathbf{H} \perp C_3$), the AFMR modes in rhombohedral structures with weak ferromagnetism take the form[4]

$$\omega_1 = \gamma\left[H(H+H_D)\frac{\chi_\perp}{\chi_\parallel} + H_\Delta^2\right]^{1/2}, \quad (1)$$

$$\omega_2 = \gamma[2H_A H_E + H_D(H+H_D)]^{1/2}, \quad (2)$$

$\gamma = ge/2mc$ is the gyromagnetic ratio, H_D is the effective Dzyaloshinskiĭ field causing the weak ferromagnetism, H_A and H_E are respectively the effective

anisotropy and exchange fields, and H_Δ^2 is the isotropic energy gap resulting either from the interaction of the nuclear and electron spins[13] or from the magnetoelastic interaction[14, 15].

As follows from (1) and (2), one AFMR mode is characterized by a small energy gap. The second mode, owing to the large exchange interaction, can have an appreciable energy gap, just as in the case of easy-axis antiferromagnets. However, unlike the easy-axis antiferromagnets, where the energy gap at $T \to 0°K$ can be determined from the sublattice flipping field, in antiferromagnets with easy-plane anisotropy the energy gap in the high-frequency mode is not known beforehand and cannot even be estimated approximately. Thus, for example, the discrepancy between the energy gap determined from measurements of the specific heat[16] and the value obtained by direct resonance measurements on $MnCO_3$[17, 18] reaches 100%. Therefore the observation of antiferromagnetic resonance corresponding to the second branch entails considerable experimental difficulties. Foremost among these difficulties is the need to cover a large frequency interval in order to "find" the resonance absorption, the frequency of which, besides being dependent on the magnetic field, is also strongly temperature-dependent. The energy gap in the high-frequency AFMR mode of rhombohedral weak ferromagnets corresponds to electromagnetic-radiation quanta of wavelength shorter than 1–2 mm. This range was not very accessible until recently, owing to the absence of radiation sources. However, following the development of backward-wave tubes (BWT) for this band,[19] it has become possible to investigate AFMR in the short-wave part of the millimeter band and in the submillimeter band.

EXPERIMENTAL PROCEDURE AND SAMPLES

A block diagram of the spectrometer, in which 500–4000 μ electromagnetic radiation was generated by backward-wave tubes, is shown in Fig. 1.

A magnetic field up to 100 kOe was produced by water-cooled solenoids of the Physics Institute of the Academy of Sciences.[20] The magnetic field was measured by the method proposed in[21] at wavelengths up to $\lambda = 930$ μ (the EPR field in DPPH is $H_{res} = 100$ kOe). The accuracy with which the magnetic field was measured was not worse than 0.5%. To improve the signal/noise ratio, the useful signal was synchronously detected by modulating the radiation at 30 Hz with a mechanical chopper. The radiation passed through the sample to a receiver based on n-type indium antimonide cooled to liquid-helium temperatures[22]. The signal from the receiver was amplified with a narrow-band low-frequency amplifier, from which it was fed to the synchronous detector. The reference signal to the synchronous detector was picked off a photoresistor illuminated with a small lamp. The light flux from the lamp was also periodically interrupted by the aforementioned mechanical chopper.

The signal from the synchronous detector was fed to the y coordinate of an x-y recorder, whose x coordinate received a voltage proportional to the magnetic field from a shunt of the solenoid. The electromagnetic radiation generated by the backward-wave tube was transmitted from the generator to the sample through standard copper waveguides of cross section 7.2 × 3.4 mm, which were thus used as multimode beam guides. The wavelength of the electromagnetic

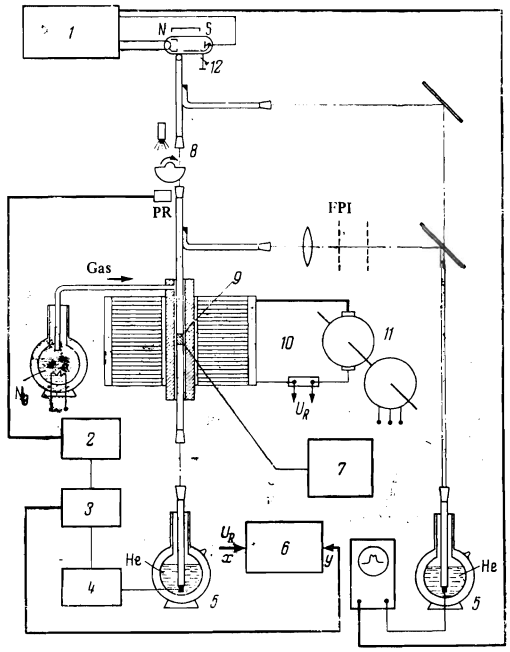

FIG. 1. Block diagram of spectrometer for the millimeter and submillimeter bands: 1–high-voltage stabilized BWT power supply, 2, 4–narrow-band low-frequency amplifiers, 3–synchronous detector, 5–receivers based on n-InSb, 6–automatic recorder, 7–system for the stabilization and measurement of the temperature at the investigated sample, 8–modulator of electromagnetic radiation, 9–section of waveguide with investigated sample, 10–water-cooled 100 kOe solenoid, 11–dc generator ($I_{max} = 12$ kA), 12–backward-wave tube; FPI–plane-parallel Fabry-Perot interferometer; PR–photoresistor.

radiation was measured with a plane-parallel Fabry-Perot interferometer with metallic grids[23]. The measurement accuracy was no worse than 0.5%.

At $T = 4.2°K$, the measurements in the strong magnetic field were made in a special cryostat, in which the indium-antimonide receiver was placed. A distinguishing feature of the cryostat design was its large length ($l = 1600$ mm) at small transverse dimensions ($d = 48$ mm). The large length of the cryostat was dictated by the need to place the indium-antimonide receiver outside the region of influence of the magnetic field. In the temperature interval $T = 77$–$350°K$, the system was cooled by blowing cold nitrogen gas over the waveguide section with the sample. The temperature stabilization and measurements were effected with a system analogous to that described in[24]. The accuracy with which the temperature was measured in the indicated range was no worse than 0.5°K, and the temperature was kept constant within not more than 0.2°K.

A typical sample mounting is shown in Fig. 2, which shows the part of the waveguide with the sample and the electromagnetic radiation concentrator. The sample was placed on the short wall of the 7.2 × 3.4 mm waveguide. The electromagnetic-radiation concentrator was a spring of hardened brass fitted tightly to the inside dimensions of the waveguide. With the aid of this device it was possible to improve the signal/noise ratio by approximately two orders of magnitude. The concentrators were used for measurements at wavelengths shorter than 1000 μ.

Some of the measurements (at wavelengths exceeding 3500 μ) were performed with an ordinary reflex direct-amplification microwave spectrometer. The generators in this case were standard klystron oscil-

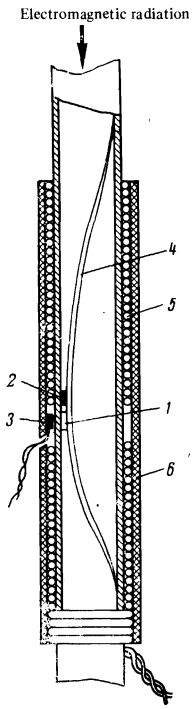

FIG. 2. Section of beam guide with investigated sample: 1–sample, 2–DPPH, 3–thermocouple junction, 4–electromagnetic-radiation concentrator, 5–heater, 6–thermal insulation.

lators. The wavelength of the electromagnetic radiation wave measured in the range from 3.8–14 mm with wavemeters of accuracy from 0.05 to 0.5%, equipped with a rectangular H_{10} resonator. The sample was mounted in this case on the tuning piston of the resonator.

The $FeBO_3$ single crystals were grown by the spontaneous-crystallization method from a solution in the melt, using a technology analogous to that in[7]. The reagents were specially pure (OSCh grade) Fe_2O_3 (136.0 g), B_2O_3 (313.6 g), PbF_2 (192.0 g), and chemically pure (grade KhCh) PbO (40.0 g). All the reagents were dried at 120°C for 16 hours, and then ground through a sieve, weighed in a glass bulb, and thoroughly mixed. The mixed reagents were melted in a platinum crucible of 275 cm^3 volume at 830°C. The crucible was covered with a platinum cover and placed in an electric resistance oven whose temperature control was programmed. Crystal growth was in accordance with the following temperature sequence: heating to 1135°C at 200°C/hr, soaking at this temperature for 10 hours, and then cooling at 2°C/hr to 600°C. The oven was turned off at 600°C. During the time of soaking and cooling, the crucible was rotated at 30 rpm.

The crystals from the cooled melt were separated by leaching in 20% hot aqueous solution of nitric acid. The grown crystals were perfect light-green hexagonal flat prisms 10 to 500 μ thick. The basal planes of the crystals always coincided with the planes of the plates. Some of the crystals were colored light-brown. A qualitative spectral analysis revealed the presence of traces of Pb, Ni, Mn, and Si in the crystals. An x-ray analysis of the grown crystals yielded lattice parameters that agreed with the data of[5].

EXPERIMENTAL RESULTS AND DISCUSSION

1. Low-Frequency AFMR Mode at $H \perp C_3$

The resonant absorption corresponding to the low-frequency mode of the spin waves in $FeBO_3$ was investigated in the wavelength range λ = 3.5–14 mm in a constant magnetic field up to 10 kOe in the temperature range 4.2–400°K. In this section we present the results of the measurements in the temperature interval 4.2–300°K. The results of the measurements at higher temperatures were published earlier[9,22,25].

With the sample so oriented that the external magnetic field was in the basal plane of the crystal, resonance absorption lines were observed. Typical plots of the derivatives of the absorption lines are shown in Fig. 3. It was observed that the shape and position of the resonance-absorption lines are strongly influenced by the method of mounting the samples. Figure 3a shows the derivatives of the resonance-absorption line at the frequency 36.160 GHz (T = 77.3°K), obtained with the sample mounted without adhesive on the tuning piston of the resonator. Figures 3b and 3c show the derivatives of the resonance-absorption lines of the same sample at the same frequency at 77.3°K in the case when the sample was secured to the piston with vacuum grease (b) and with BF-6 (c). We see that the gluing of the sample or securing it with vacuum grease that solidifies at liquid-nitrogen temperature leads to a general broadening of the resonance-absorption line, to a shift of this line relative to the position of the line of the sample mounted without adhesive, and to the appearance of a clearly pronounced fine structure. After the adhesive or grease was removed and the sample was mounted in the resonator without adhesive, the observed absorption lines are just as narrow as prior to gluing. The widths of the fine-structure peaks are close to the resonance-absorption line width obtained with the sample mounted without adhesive.

The observed complicated fine structure of the lines could be due, in principle, for example, to the following factors: the appearance of magnetostatic types of precession, excitation of spin waves by macroscopic inhomogeneities of the surface, etc.

There is also another possible way of explaining the observed phenomenon. Indeed, as observed in[15],

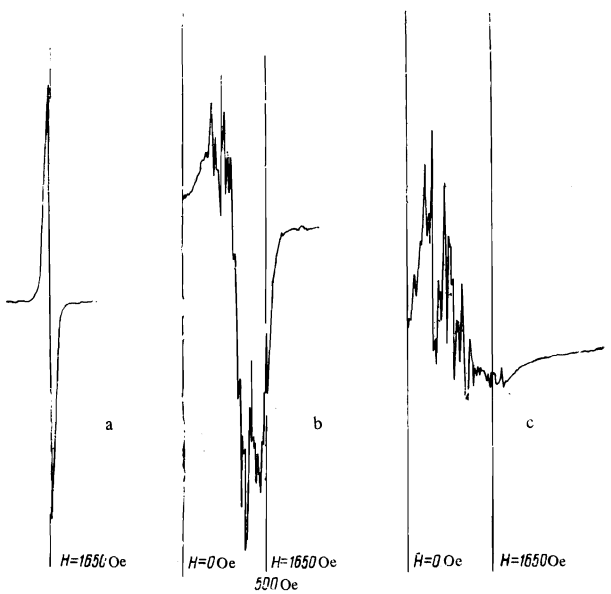

FIG. 3. Typical plots of the derivatives of the resonance-absorption lines of $FeBO_3$, corresponding to low-frequency AFMR, at ω = 36.160 GHz and T = 77.4°K: a–sample mounted without adhesive, b–sample secured with vacuum grease, c–sample glued to resonator piston with BF-6 adhesive.

the position of the AFMR line in rhombohedral antiferromagnets with weak ferromagnetism is extremely sensitive to deformations in the basal plane of the crystal. It was also shown in that reference that the low-frequency mode of the spin-wave spectrum of an easy-plane antiferromagnet can contain an energy gap due to the magnetoelastic interaction (see formula (1)):

$$H_\Delta^2=H_{\Delta 1}^2+H_\Delta^2(\mathbf{p})=2H_{me}(0)H_E+2H_{me}(\mathbf{p})H_E, \quad (3)$$

where

$$H_{\Delta 1}^2=2H_EH_{me}(0)=\frac{B}{2M_0}\frac{4\lambda_3^2\mu_5+\lambda_4^2\mu_3-\lambda_3\lambda_4\mu_6}{4(\mu_3\mu_6-\mu_6^2)}(l_x^2+l_y^2)^2, \quad (4)$$

$$H_\Delta^2(\mathbf{p})=2H_{me}(\mathbf{p})H_E=\begin{cases}-2pAH_E & \text{with } \mathbf{p}\parallel\mathbf{H}\\ 2pAH_E & \text{with } \mathbf{p}\perp\mathbf{H}\end{cases}, \quad (5)$$

λ_i and μ_i are respectively the elastic and magnetoelastic constants, A is the spontaneous-striction amplitude and is expressed in terms of λ_i and μ_i[15], and $\mathbf{p}$ is the external force acting on the crystal. If the crystal is compressed or distended in the basal plane, then, as follows from (1) and (5), the resonance-absorption line should shift.

To ascertain the origin of the fine structure, experiments were performed to study the influence exerted on the shape and position of the resonance-absorption line by the dimensions and shapes of the samples, by the various methods of mounting the samples, and also by the degree of homogeneity of the high-frequency field of the resonator. The experiments have shown that the line structure does not depend on the location of the sample in the resonator, i.e., the degree of homogeneity of the high-frequency field does not affect the shape and structure of the observed resonance-absorption lines. The phenomenon was observed in samples of various shapes and dimensions. Study of different methods of mounting and gluing the crystals led to the conclusion that the shift of the position of the center of gravity of the absorption lines and the appearance of the fine structure are due to homogeneous and inhomogeneous mechanical stresses that are produced in the sample when it is glued. It appears that these stresses exert different effects on different blocks of the crystal, and this causes a shift of the resonance-absorption lines in individual blocks relative to the unshifted AFMR line.

This phenomenon was most strongly pronounced in the study of AFMR in pure light-green $FeBO_3$ crystals with absorption line widths $\Delta H \approx 10$–100 Oe at a frequency $\omega \approx 30$ GHz and below room temperature. The effect was strongly dependent on the temperature and was stronger the lower the temperature at which the measurement was performed. At room temperature and above, gluing of the sample had practically no effect on the width, shape, and position of the absorption line, and there was no fine structure. Such high sensitivity of the width, shape, and position of the resonance-absorption line to deformations of the investigated sample indicates that the quality of the crystal should be considerably affected by the rate of cooling during the course of crystallization and annealing of the crystals.

All the experimental data cited below were obtained with single crystals of light-green tint, since the results of measurements made on brownish single crystals were not reproducible from sample to sample. The brown tint seems to be due to uncontrollable impurities, particularly Fe^{2+} and Fe^{4+} [7].

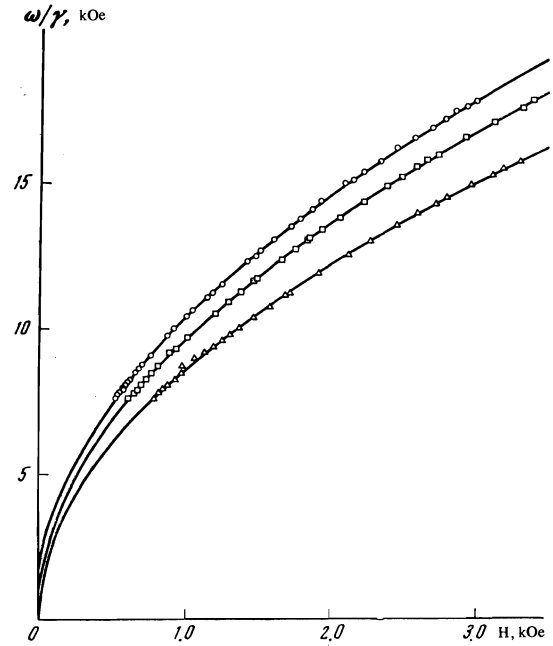

FIG. 4. Dependence of the frequency of low-frequency AFMR on the external magnetic field at temperatures 4.2 (○), 194.7 (□), and 273 (△) °K.

Figure 4 shows three series of experimental points corresponding to plots of the AFMR frequency against the external magnetic field at three temperatures, 4.2, 194.7, and 273°K. The measurement results were reduced with a computer by least squares using formula (1) and γ corresponding to $g = 2.00$ as obtained from our earlier measurements[9]. As seen from Fig. 4, the experimental results agree well with the theoretical curves (solid lines) corresponding to relation (1) with the computer-calculated coefficients. The agreement is observed in the temperature range from 4.2 to 300°K.

As follows from (1), the resonance experiments make it possible to reveal, in principle, the anisotropy of the susceptibility. This was done earlier for hematite[22], where the susceptibility anisotropy could be observed only in magnetic fields up to 100 kOe, owing to the relations between the parameters that are contained in (1). In $FeBO_3$, the low-frequency AFMR mode was investigated in fields up to 10 kOe. In this range of fields, as follows from (1), the term $H^2\chi_\perp/\chi_\parallel$ has a much smaller effect than the term $H_DH\chi_\perp/\chi_\parallel$, so that it was impossible to state that the ratio $\chi_\perp/\chi_\parallel$ differs from unity. As follows from the form of relation (1) and from the analysis of the experimental data on α-Fe_2O_3[22], to detect susceptibility anisotropy in $FeBO_3$ it is necessary to perform the measurements in magnetic fields up to ~500 kOe, inasmuch as the Dzyaloshinskiĭ field is exceedingly strong, $H_D \approx 100$ kOe, and is approximately 5 times larger than the Dzyaloshinskiĭ field in hematite. Exact resonance measurements in such strong magnetic fields are difficult, and there are no other experiments pointing to the presence of susceptibility anisotropy (for example, static measurements). On the other hand, the iron ion Fe^{3+} in $FeBO_3$, like the magnetic ions in $MnCO_3$ and α-Fe_2O_3, is in the s state. This gives grounds for assuming that the anisotropy of the susceptibility is small. Our experiments have made it possible to determine the values of $H_{DD_1} = H_D\chi_\perp\chi_\parallel$ for a number of temperatures; these

are listed in the table. The temperature dependence of this quantity is shown in Fig. 5a. The solid curve is the Brillouin function for the spin s = 5/2. We did not calculate the values of H_{DD_1} by formula (1) at temperatures above 300°K. At these temperatures, the magnetic order induced by the external magnetic field and investigated in detail in earlier papers[9, 25] comes into play, and the question of the possibility of determining the Dzyaloshinskiĭ fields from relation (1) near the Neel temperature[1)] still remains open[9].

As seen from Fig. 4, an energy gap H_Δ^2 exists in the low-frequency AFMR mode. We have investigated the temperature dependence of $H_\Delta^2(T)$ (Fig. 6), calculating H_Δ^2 from (1) and using a series of measurements at different temperatures. However, comparison of the experimental results on the temperature dependence of H^2 (T) with the experimental temperature dependence of $H^4_{DD_1}(T)$ ($H^4_{DD_1} \sim l^4$) obtained in the same experiments shows that the discrepancy seems to exceed the possible measurement errors (see Fig. 6). The solid line in Fig. 6 also shows the fourth power of the Brillouin function for the magnetization (spin s = 5/2).

It follows from Fig. 6 that $H_\Delta^2|_{T=300°K} \approx 0.1\, H_\Delta^2|_{T=4.2°K}$. Therefore, owing to the smallness of the magnetoelastic interaction, the effect exerted by the stresses due to gluing of the sample on the shape and position of the AFMR line should be much smaller than at low temperatures. This experimentally observed phenomenon confirms the conclusion that the gap in the low-frequency mode of the spin waves in $FeBO_3$ is of magnetoelastic origin.

We have already noted that a study of the temperature dependence of the position of the resonance-absorption line at 34.5 GHz was made in[7]. This resonance absorption was treated in that reference as ferromagnetic resonance in a ferromagnet with anisotropy of the "easy plane" type. The authors of[7] used the following formula for the dependence of the resonance-absorption frequency on the external magnetic field:

$$(\omega/\gamma)^2 = H(H + H_A + 4\pi M_s),$$

where M_S is the saturation magnetic moment of the sample. This formula, as noted by the authors earlier[8], coincides in form with formula (1) if H_Δ^2 is neglected. In this case the quantity $H_A + 4\pi M_S$ should be taken to mean the Dzyaloshinskiĭ field.

The positions of the resonance-absorption lines observed by us at 34.5 GHz coincided to within no more than 3% with the line positions observed in[7], and at all temperatures. The agreement between the results obtained with crystals of different origin indicates that the single crystals used in both investigations were of sufficient purity.

The values of the Dzyaloshinskiĭ field H_D obtained in[10] with the aid of static measurements of the magnetization of samples from the same source as those investigated coincide with the values of H_{DD_1} obtained by us at T = 77.4°K, and differ somewhat from them at room temperature (see the table).

The Dzyaloshinskiĭ fields obtained from static measurements[12] differ by 10–12% from our data and from the data of[10], both at 77.4°K and at room temperature. Without stopping to discuss the correspondence of the Dzyaloshinskiĭ field obtained from static measurements with the Dzyaloshinskiĭ fields obtained from AFMR, we note the following apropos the indicated difference. We studied samples with different tints. It turned out that the Dzyaloshinskiĭ fields H_{DD_1} determined from experiments on samples with brownish tint were not reproducible from sample to sample and were systematically 10–40% lower than the Dzyaloshinskiĭ fields determined from experiments on light-green samples (the difference depends on the tint). This circumstance indicates that the Dzyaloshinskiĭ field H_{DD_1} decreases in the presence of impurities that color the crystal brown.

2. High-Frequency AFMR Mode at H ⊥ C_3

The high-frequency AFMR mode in $FeBO_3$ was investigated at wavelengths λ = 500–1700 μ in constant magnetic fields up to 100 kOe.[2)] When the sample was so mounted that the external magnetic field was in the basal plane, absorption lines corresponding to the high-frequency mode of the spin-wave spectrum were observed.

The dependence of the frequency of the AFMR high-frequency mode on the external magnetic field was investigated in the temperature interval from 4.2 to 350°K. Formula (2) leads to a linear dependence of the square of the frequency of the high-frequency resonance on the external magnetic field applied in the basal plane. Indeed, all the experimental results, which were reduced with the computer by least squares, agree well with the formula

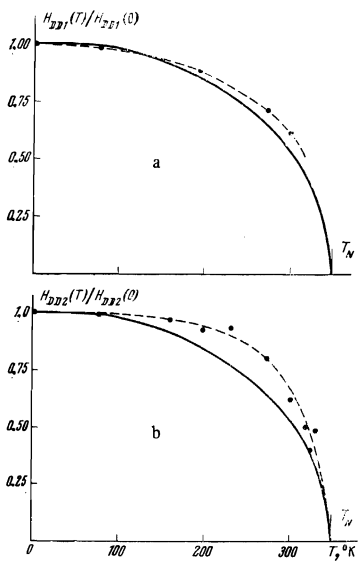

FIG. 5

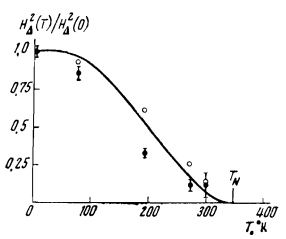

FIG. 6

FIG. 5. Temperature dependences of the Dzyaloshinskiĭ fields, determined from measurements of the low-frequency (a) and high-frequency (b) AFMR.

FIG. 6. Temp erature dependence of the energy gap in the low-frequency mode of the spin-wave spectrum in $FeBO_3$: ○–experimental values of $H_\Delta^2(T)/H_\Delta^2(0)$, ●–experimental values of $H^4_{DD1}(T)/H^4_{DD1}(0)$.

Certain magnetic parameters of $FeBO_3$, determined from resonance and static [10,12] measurements

T, °K	H_Δ^2, kOe²	H_{DD_1}, kOe	H_{DD_2}, kOe	H_c, kOe	H_D, kOe [10]	H_D, kOe [12]
4.2	4.9 ±0.2	101.2±0.1	92.7±0.2	165.7±0.2	—	—
77.3	4.25±0.25	99.3±0.2	91.5±0.9	164.5±0.5	99	87±7
198,5	1.6 ±0.2	89.7±0.1	85.1±0.9	152±1	—	—
231	—	—	84.7±1.3	143 ±1	—	—
273	0.6±0.2	72.0±0.1	82.0±1.0	127 ±1	—	—
300	0.6 ±0.5	62.0±0.5	57.4±0.2	111±1	66	48

$$(\omega_2/\gamma)^2 = H_{DD2}H + H_c^2, \quad (6)$$

where the parameters H_{DD_2} and H_c^2 depend on the temperature. Figure 7 shows, by way of example, the experimental results for five temperatures. The solid lines were obtained by reducing the experimental data in accord with formula (5) by least squares. This reduction, carried out at all measurement temperatures, has made it possible, first, to determine the temperature region in which relation (2) is valid and second, to study the temperature dependences of the Dzyaloshinskiĭ field $H_{DD_2}^{(T)}$ and the value of the energy gap $H_c^{(T)}$.

As seen from Fig. 7, the experimental points are fitted well by straight lines of the type (6) up to T = 330°K. It was impossible to verify (6) in the immediate vicinity of the Neel temperature because of the strong broadening of the resonance-absorption lines and of the inevitably resulting errors in the determination of their positions.

The temperature dependence of H_{DD_2} is shown in Fig. 5b. The dashed line is drawn through the experimental points and the solid line is the Brillouin function for the spin s = 5/2. The values of H_{DD_2} for a number of temperatures are listed in the table. As can be seen from the table, the Dzyaloshinskiĭ fields H_{DD_1} and H_{DD_2} differ by almost 10%, which is more than the possible experimental errors. We are unable to state at present whether this difference is a consequence of the presence of susceptibility anisotropy, or whether there is a real difference between the Dzyaloshinskiĭ fields determined from measurements of the low-frequency and high-frequency AFMR, as in the case of hematite[22]. Attention is also called to the different character of the temperature dependences of H_{DD_1} and H_{DD_2} (see Figs. 5a and 5b).

The temperature dependence of $H_{AE}(T)$ shown in Fig. 8 was calculated using the experimentally obtained values of H_{DD_2}: $H_{AE}^2(T) = H_c^2(T) - H_{DD_2}^2$. As seen from the figure, the experimental points agree well with the temperature dependence of the weak ferromagnetic moment $\sigma = \sigma(T)$ taken from[10].

We also investigated the temperature dependence of the energy gap $H_c(T)$ in the high-frequency AFMR mode. Extrapolation of the function $H_c(T)$ to T = 0 yields the energy gap in the low-frequency mode of the spin-wave spectrum in $FeBO_3$, with a value 165.7 kOe (the high-frequency resonance in the absence of an external magnetic field corresponds at T = 0°K to a wavelength $\lambda = 640\ \mu$).

The size of the gap corresponds to $T_{AE} = 22.1$°K ($T_{AE} = \mu_{eff} H_c/k$)[3], i.e., it is at this temperature that one should observe kinks on the temperature dependences of the sublattice magnetization, doubling of the specific heat, etc. It should be noted that it is difficult to obtain this result with other experiments, since large volumes of the substance are required. It is difficult to separate a large number of pure crystals. The use of a mixture of crystals with different impurity contents can lead to incorrect results, since the size of the energy gap also depends on the value of the Dzyaloshinskiĭ field, and the latter, according to our data, can vary from 60 to 100 kOe, depending on the impurity content. The size of the gap in $FeBO_3$ greatly exceeds the energy gap in manganese carbonate $MnCO_3$ (which amounts to $H_c|_{T\approx 0°K} \approx 44$ kOe according to the data of Richards[17] and of Borovik-Romanov and Prozorova[18])

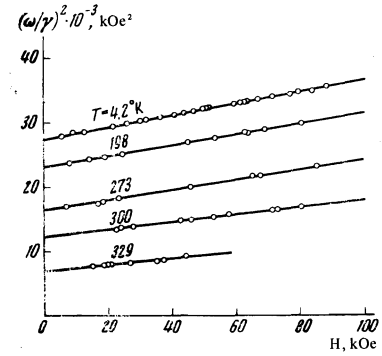

FIG. 7. Dependence of the frequency of the high-frequency AFMR on the external magnetic field at various temperatures.

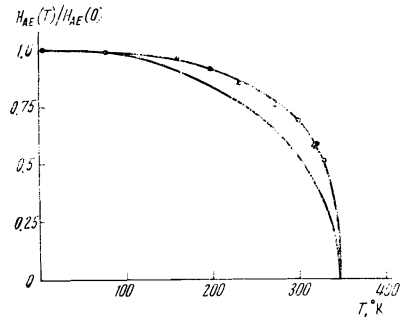

FIG. 8. Temperature dependence of the quantity $H_{AE} = H_{AE}(T)$; the solid curve passing through the points is the temperature dependence of the weak ferromagnetic moment $\sigma = \sigma(T)$ [10]. The lower curve is the Brillouin function for the spin s = 5/2.

and that in the weakly-ferromagnetic hematite α-Fe_2O_3[27].

So large an energy gap in the high-frequency mode of the $FeBO_3$ spin waves is due, first, to the appreciable value of the Dzyaloshinskiĭ field, and second, to the large value of the exchange field.

3. Temperature Dependence of the Line Width of the High-Frequency and Low-Frequency AFMR

We investigated the temperature dependences of the AFMR linewidths. It was observed that the absorption linewidths of the low-frequency and high-frequency resonances behaved differently with rising temperature (see Fig. 9). The line widths are expressed in terms of the frequency ($\Delta\omega$). The temperature dependences of the resonance-absorption line widths shown in the figure demonstrate the variation of the high-frequency and low-frequency AFMR line widths in a constant magnetic field with changing temperature. It is seen from Fig. 9 that the increase of the line widths of the high-frequency resonance occurs at much lower temperatures than that of the low-frequency resonance. The dark circles on the diagram correspond to the high-frequency AFMR line widths in the magnetic field H ≈ 50 kOe, and the triangles correspond to the low-frequency line widths in a field H ≈ 5 kOe. In fields stronger than 5 kOe, the line width of the absorption corresponding to the low-frequency AFMR begins to broaden at temperatures closer to T_N, owing to the fact that the external magnetic field suppresses the fluctuations that destroy the magnetic order near T_N[25]. Such a strong difference between the temperature dependence of the line widths of the low- and high-frequency AFMR indicates that the relaxation mechanisms for the oscillations of the low- and high-frequency modes are different.

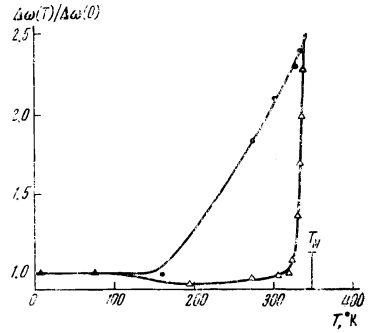

FIG. 9. Temperature dependences of the line widths of the low-frequency (Δ) and high-frequency (●) AFMR.

CONCLUSION

The main results of our investigations are the following:

1. We observed a fine structure in the absorption lines corresponding to the low-frequency AFMR, and have shown that it results from elastic stresses that appear when the sample is glued. It is shown that gluing shifts the position of the AFMR line significantly.

2. We investigated the low-frequency AFMR in $FeBO_3$. We have shown that the dependence of the AFMR frequency on the external magnetic field coincides with the theoretical one in the temperature range 4.2–300°K.

3. An energy gap of magnetoelastic origin was observed in the low-frequency AFMR mode, and its temperature dependence was investigated.

4. High-frequency AFMR in $FeBO_3$ was observed and investigated. It is shown that the dependence of the AFMR frequency on the external magnetic field agrees with the theoretical dependence in the temperature range 4.2–330°K.

5. We determined the value of the energy gap H_c in the high-frequency mode of the spin-wave spectrum, namely $H_{c}|_{T=0°K} = 165.7$ kOe ($\lambda = 640\ \mu$), $T_{AE} = 22.1$°K.

6. We determined the $H_{AE}(T)$ dependence and have shown that it agrees with the temperature dependence of the spontaneous magnetization $\sigma(T)$.

7. The Dzyaloshinskiĭ fields were determined independently from the low-frequency and high-frequency AFMR modes, and their temperature dependences were investigated. It was observed that these dependences differ.

8. A significant difference was observed in the temperature dependences of the linewidths of the high- and low-frequency resonances, which seemingly points to a difference between the mechanisms that govern these widths.

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[1)]The Neel temperature of the investigated crystals was determined by the nuclear gamma resonance method without an external magnetic field, and turned out to be 348.1°K [9].

[2)]A preliminary report of observation of AFMR corresponding to the high-frequency mode of the spin-wave spectrum in $FeBO_3$ was published in [27].

[3)]After this paper was sent to press, the authors learned of the work of M. Eibschutz and M. E. Lines (Phys. Rev. B7, 4907 (1973), in which theoretical estimates were made of T_{AE} and H_D for $FeBO_3$. Their data agree well with the results of our measurements.

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