critical fluctuations. At present there are no known experimental studies of the details of the dependence of tanδ on E and ω in weak bias fields.

The author thanks O. G. Vendik and A. I. Sokolov for suggesting the topic and for interest in the work, and V. L. Gurevich and B. D. Laĭkhtman for reading the manuscript and for a number of important remarks.

[1)]We point out a misprint in Ref. 9; the columns in (35.15) should be interchanged.

[2)]It can be shown that since the longitudinal branch is separated for the considered directions, an admixture of longitudinal acoustic oscillations makes a contribution that is small compared with those considered below.

[3)]More accurately speaking, we have written out here the contribution of the absorption processes within the soft optical mode, since the contribution from such processes within the acoustic modes is less relative to the parameter T/Θ at $T < \Theta$. Here and below, when speaking of the increment to tanδ, we mean the increment to $\tan\delta(\mathbf{E}=0)$.

[4)]We estimate below $\Delta\varepsilon/\varepsilon$, which is equivalent to an estimate of E, but is more universal.

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Investigation of antiferromagnetic resonance and two-magnon absorption in the weak ferromagnet $CoCO_3$

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The high-frequency AFMR branch and two-magnon absorption are detected and investigated in $CoCO_3$. A frequency shift of the electric-dipole two-magnon absorption in a magnetic field is observed for the first time. The behavior of the low-frequency branch of the spectrum is investigated in magnetic fields comparable with the crystal exchange field. The experimental results are interpreted with the aid of a theory that takes into account the large value of the uniaxial anistropy and the appreciable Dzyaloshinskiĭ interaction. It is shown that the two-magnon absorption in $CoCO_3$ is due to simultaneous excitation of two magnons from the high-frequency branch of the spin-wave spectrum.

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1. INTRODUCTION

Spectroscopy in the longwave infrared (LIR) range of the spectrum is an extremely effective experimental method that has been used to investigate the high-frequency (HF) properties of many antiferromagnetic dielectrics (see, for example, the reviews of Foner[1] and of Richards[2]). The present paper reports the detection and investigation by this method of a HF branch of the spin-wave spectrum and of two-magnon absorption in the weak ferromagnet cobalt carbonate. It also reports an investigation of the low-frequency (LF) branch of the spectrum in magnetic fields comparable with the crystal exchange field. Preliminary results of these investigations were reported by us earlier.[3-5]

The magnetic properties of $CoCO_3$ (space group D_{3d}^6) have been studied in considerable detail. It was discovered by Borovik-Romanov and Ozhogin[6,7] that $CoCO_3$ changes to a magnetically ordered state at $T_N = 18.1$ K. The magnetic moments of the sublattices are canted, and the resultant weak ferromagnetic moment lies in the basal plane of the crystal. Subsequent neutron-diffraction data[8] indicate that the magnetic moments of the sublattices also lie in the (111) plane. In a phenomenological theory of magnetism, the appearance of a weak ferromagnetic moment is due to the existence of a Dzyaloshinskiĭ effective field H_D, whose value for $CoCO_3$, according to the data of Ref. 7, is $H_D = 28$ kOe. This value of H_D was later confirmed by investigations of the susceptibility of $CoCO_3$ in weak magnetic fields.[9] By "weak" fields are understood fields $H \ll H_E$, where H_E is the crystal exchange field. The magnetization of $CoCO_3$ in the basal plane has been investigated by use of pulsed magnetic fields of intensities up to 340 kOe.[10] Signs of an approach of the magnetization to saturation were detected; the saturation value was close to the pure-spin value $2g\mu_B SN = 16750$ cgs emu/mol, where $g = 2$, $S = \frac{3}{2}$, and $2N$ is Avogardro's number. The value determined for the exchange field was $2H_E = 300$ kOe.

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Approximately the same value of $2H_E$ is found from the results of Refs. 7 and 9, if one takes for the value of the saturation magnetization the pure-spin value.

The AFMR spectrum in antiferromagnets of the "easy plane" type consists of two branches—an acoustic, having no energy gap in the absence of an external magnetic field and of anisotropy in the basal plane, and an optical, with a gap[11,12]:

$$\omega_1^2=\gamma_\perp^2(HH_D+H^2), \tag{1a}$$

$$\omega_2^2=\gamma_\perp^2(2H_EH_A+H_D^2+HH_D) \tag{1b}$$

when $\mathbf{H}\perp C_3$, and

$$\omega_1^2=0, \tag{1c}$$

$$\omega_2^2=\gamma_\perp^2[2H_EH_A+H_D^2+(g_{zz}/g_\perp)^2H^2] \tag{1d}$$

when $\mathbf{H}\parallel C_3$. Here H_A is the effective uniaxial-anisotropy field, $g_\perp$ and g_{zz} are the components of the g factor, and $\gamma_\perp$ is the gyromagnetic ratio.

Earlier, only the LF branch of the spectrum, in magnetic fields $H\leq 23$ kOe, was investigated.[13,14] By use of the expression (1a), the values $g_\perp=3.3\pm0.2$ and $H_D=51.5\pm8$ kOe were determined. Investigation of EPR spectra of Co^{++} ions implanted in various crystal structures isomorphic to cobalt carbonate permitted determination of the components of the g factor for $CoCO_3$: $g_\perp=4.95\pm0.03$ and $g_\parallel=3.07\pm0.03$.[15] A paper of Borovik-Romanov *et al.*[16] reports a study of single-magnon Mandel'shtam-Brillouin light scattering by the acoustic branch of the spectrum. It was found that $H_D=27\pm16$ kOe and $g_\perp=4.1\pm1.1$. From the results enumerated, it is evident that values of H_D and $g_\perp$ determined in different experiments differ appreciably from one another.

In the present paper, a detailed investigation is made of both branches of the AFMR spectrum and of two-magnon absorption in $CoCO_3$. The experimental results are interpreted with the aid of a theory that takes into account the peculiarities of this crystal, and primarily the large value of the uniaxial anisotropy.

2. EXPERIMENT

Description of the experiment

The HF branch of the spin-wave spectrum and the two-magnon absorption were detected and investigated with the aid of a diffraction spectrometer for the LIR range of the spectrum, with an operating range, in wavelengths, from 20 μm to 1.3 mm. A description of the optical arrangement was given earlier.[17] The source of radiation in the spectrometer was a globar or mercury lamp. The receiver used was a bolometer of germanium alloyed with gallium, operating at $T=1.5$ K, or a photoreceiver of n-InSb, operating at $T=4.2$ K. The temperature of the specimen, which is located in a cryostat, can be varied over the range from 2 to 300 K. The specimen under investigation was located in an external stationary magnetic field of intensity up to 18 kOe. It was also possible to use a superconducting solenoid that permitted the obtaining of a magnetic field of intensity up to 50 kOe.

Investigation of the behavior of the LF branch of the spectrum in a magnetic field was carried out with the aid of a nonresonant spectrometer of the straight-through type with a straight-amplification receiver. The spectrometer permitted the carrying out of resonance investigations over the wavelength range 0.35–6 mm in a pulsed magnetic field of up to 300 kOe, at a specimen temperature determined by the cooling agent chosen. The sources of microwave radiation were generators of the backward-wave tube type; the transmission line was of quasioptic lens type; the receiver was an n-InSb crystal cooled to 4.2 K. The spectrum was recorded with a two-beam memory oscillograph C8-2. The scanning of the spectrum was done by the magnetic field, at fixed frequency of the microwave generator. Next to the specimen, which was located in the bore of the pulse solenoid, was placed a tablet of DPPH for calibration of the field value. The magnetic field was measured with a test coil located in the center of the solenoid.

The single crystals of $CoCO_3$ used in the experiment were grown by the hydrothermal method. The specimens contained as impurities iron (several tenths of a percent), manganese, and nickel (several hundredths of a percent). Orientation of the specimens was accomplished with x-rays and was of two types: the first with the plane of the specimen perpendicular to the third-order crystal axis, the second with the plane of the specimen parallel to the C_3 axis. The thickness of the specimens varied from 0.07 to 1 mm.

Results of the experiment

The absorption spectrum of $CoCO_3$ at $T=4.2$ K was investigated over the frequency range from 10 to 350 cm^{-1}. At frequencies above 170 cm^{-1} the specimens were opaque; this was because of excitation of phonon oscillations. Two absorption bands were found, at frequencies 34.6 ± 0.3 cm^{-1} and 54.7 ± 0.35 cm^{-1}; see Fig. 1. Lines with similar frequencies were simultaneously detected in the Raman light-scattering spectrum.[18] The parameters of the bands are given in Table I, with allowance for corrections for the instrumental function of the spectrometer. The absorption band at frequency 34.6 cm^{-1} has a symmetric form; in the absence of an external magnetic field, its polarization is $\mathbf{h}_\omega\perp C_3$ (the absorption is independent of the orientation of $\mathbf{h}_\omega$ in the basal plane of the crystal). Upon application of a magnetic field $\mathbf{H}\perp C_3$, $H\geq3$ kOe, absorption is observed only when $\mathbf{h}_\omega\parallel\mathbf{H}$, or equivalently when $\mathbf{h}_\omega\parallel\mathbf{m}$, where $\mathbf{m}$ is the weak ferromagnetic moment. This result is a consequence of the removal of the domain structure of $CoCO_3$ by the magnetic field. Upon application of a magnetic field, the specimen reconstructs itself into a single-domain state, and the reconstruction is completed at fields 3–3.6 kOe.[7,8]

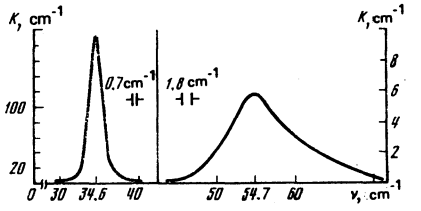

FIG. 1. Absorption spectrum of $CoCO_3$ at $T=4.2$ K.

TABLE I. Absorption-band parameters.

Absorption bands	ν, cm^{-1}	Polarization	K, cm^{-1}	I, cm^{-2}	Γ, cm^{-1}
First	34.6±0.3	$\mathbf{h}_\omega \| \mathbf{m}$	207	520	1.3
Second	54.7±0.35	$\mathbf{e}_\omega \perp C_3$	6.1	104	10.6

Note: here ν is the frequency of the absorption maximum, K is the absorption coefficient, I is the integrated intensity, and Γ is the width at level 0.5 K.

On heating of the specimen, the frequency of the absorption maximum shifts toward the LF region of the spectrum (see Fig. 2), the absorption band broadens, and observation of it becomes impossible when $T \gtrsim 14$ K.

Figure 3 shows the variation of the frequency of the absorption maximum with magnetic field intensity for two orientations: $\mathbf{H} \perp C_3$ and $\mathbf{H} \| C_3$. Except for the field range 18 kOe$< H \leqslant$50 Oe in the case $\mathbf{H} \perp C_3$, these relations were obtained with the experiment geometry shown in Fig. 4a. The magnetic field range indicated above can be obtained by use of the superconducting solenoid. Since the magnetic field vector must be perpendicular to the C_3 axis, the condition $\mathbf{H} \| \mathbf{m} \| \mathbf{k}$ is always satisfied, where $\mathbf{k}$ is the wave vector of the radiation incident on the crystal. The absorption line is observed when $\mathbf{h}_\omega \| \mathbf{m}$; that is, in this geometry observation of it is impossible.

The experimental data for fields 18 kOe$< H \leqslant$50 kOe with $\mathbf{H} \perp C_3$ were obtained with the geometry shown in Fig. 4b. In this case the specimen was placed so that the C_3 axis was horizontal, and was then rotated in a horizontal plane through an angle ψ. As a result, in the plane perpendicular to H and k there is a projection $\mathbf{m}_\perp$ of the magnetic moment. By making $\mathbf{h}_\omega$ coincident with the direction in which $\mathbf{m}_\perp$ lies, one can observe an absorption band with optical density $D \sin\psi$, where D is the optical density of the band for the given specimen under normal conditions of excitation, i.e., with $\mathbf{h}_\omega \| \mathbf{m}$.

The absorption band at frequency 34.6 cm^{-1} is very intense; its absorption coefficient is $K = 207$ cm^{-1}. Therefore for a specimen of thickness 0.8 mm with $\psi = 7°$, the intensity of the band should be approximately 15%, as was in fact observed experimentally. This intensity is sufficient for sure determination of the frequency position of the absorption maximum. The variation of frequency with magnetic field obtained with the geometry of Fig. 4b is given in Fig. 3 with allowance for corrections for perpendicularity of C_3 and H, which can be easily calculated.

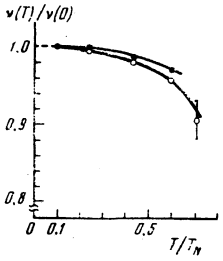

FIG. 2. Temperature variations of the frequencies of the absorption maxima: ○) $\nu(0) = 34.6$ cm^{-1}; ●) $\nu(0) = 54.7$ cm^{-1}. The sizes of the points correspond to the experimental error.

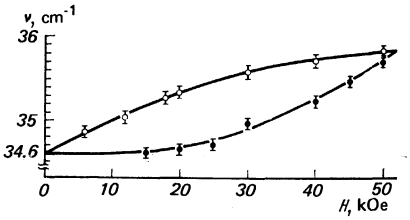

FIG. 3. Variation of the frequency of the HF branch of AFMR with magnetic field intensity: ○) $\mathbf{H} \perp C_3$; ●) $\mathbf{H} \| C_3$. The curves are calculated: the upper by formulas (3), (5), and (14); the lower by formulas (6), (7), and (14).

The second absorption band, at frequency 54.7 cm^{-1}, has an asymmetric form (see Fig. 1). Its polarization $\mathbf{e}_\omega \perp C_3$ does not change on application of the magnetic field. On heating of the specimen, the absorption maximum shifts toward the LF region of the spectrum somewhat less pronouncedly than is the case for the line with frequency 34.6 cm^{-1} (Fig. 2). At temperatures above 13 K, the absorption merges with the background. In a magnetic field $\mathbf{H} \perp C_3$, there is a nearly linear shift of the frequency toward the HF region of the spectrum; see Fig. 5. When $\mathbf{H} \| C_3$, the frequency position of the absorption maximum is unchanged in a field of intensity up to 50 kOe.

It was mentioned above that the LF branch of the spin-wave spectrum in $CoCO_3$ had been investigated earlier in magnetic fields $H \leqslant$23 kOe. We have investigated the behavior of the LF branch of the spectrum in fields of intensity up to 145 kOe. The results of investigation of the variation of the frequency of the LF branch of the spin-wave spectrum with magnetic field intensity at $T = 4.2$ K are shown in Fig. 6.

Discussion of the results of the experiment

We interpret the absorption line at frequency 34.6 cm^{-1} as excitation of the HF branch of the AFMR spectrum. The polarization, temperature, and field measurements support such an interpretation. In a paper of Turov and Guseĭnov,[19] conditions are determined under which oscillations of both branches of the spin-wave spectrum should be excited in rhombohedral weak ferromagnets. Oscillations of the HF branch should be excited when $\mathbf{h}_\omega \| \mathbf{H}$, oscillations of the LF branch when $\mathbf{h}_\omega \perp \mathbf{H}$. The experimentally established conditions for excitation of the HF branch of AFMR coincide with the deductions of the theory. The decrease of the AFMR frequency on heating of the specimen (Fig. 2) is characteristic of the majority of antiferromagnets. The

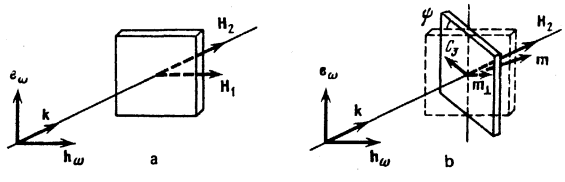

FIG. 4. Geometry of the experiment: $\mathbf{e}_\omega$, $\mathbf{h}_\omega$, and $\mathbf{k}$ are the electric, magnetic, and wave vectors of the LIR radiation; H_1 and H_2 are the vectors of the stationary magnetic field; $H_1 \leqslant 18$ kOe, $H_2 \leqslant 50$ kOe.

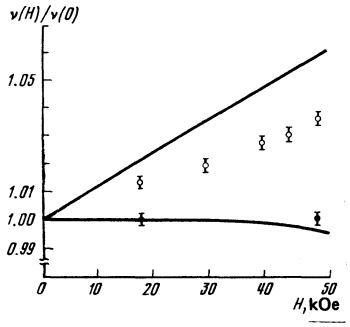

FIG. 5. Variation of two-magnon absorption frequency in $CoCO_3$ ($\nu(0) = 54.7$ cm^{-1}) with magnetic field intensity at $T = 4.2$ K: ○) $\mathbf{H} \perp C_3$; ●) $\mathbf{H} \parallel C_3$. Solid curves, relative changes of the energies of magnons with $\mathbf{k} = [\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2}]$: upper curve ($\mathbf{H} \perp C_3$), calculation by formulas (5), (19), and (14); lower curve ($\mathbf{H} \parallel C_3$), calculation by formulas (20) and (14).

large value of the absorption, which does not change when one goes over to specimens with a different impurity content, attests to the correctness of the interpretation. The variations of the frequency of the HF branch with magnetic field intensity are qualitatively described by the expressions (1b) and (1d). Quantitative agreement for $\mathbf{H} \perp C_3$ cannot be obtained for any values of the theoretical parameters that occur in the expressions (1). This is a consequence of the fact that the expressions (1) were derived on the assumption that $H_A \ll H_E$ and $H_D \ll H_E$. From the value of the frequency of the HF branch of AFMR in zero field, with values of the g factor determined by a number of authors,[13-15] it follows that the uniaxial-anisotropy field H_A is a quantity of the order of H_E; and from experimental data[7,9,14] it is evident that $H_D \approx (0.2-0.3)H_E$.

We attribute the absorption at frequency 54.7 cm^{-1} to simultaneous excitation of two magnons, with equal and oppositely directed wave vectors near the boundary of the Brillouin zone. The experimental data—the electric-dipole character of the absorption, a shift of the frequency toward the LF region of the spectrum that is less than for the HF branch of AFMR, the asymmetric form of the absorption line—support this interpretation.

Of special interest is the shift of frequency of two-magnon absorption in a magnetic field applied in the basal plane of the crystal (Fig. 5). To our knowledge, this is the first experimental observation of a shift of frequency of electric-dipole two-magnon absorption in a magnetic field. This result, together with the anomalous asymmetry of shape of the absorption (we have in mind the flatter HF absorption edge in Fig. 1), indicates important singularities in the magnon spectrum in weakly ferromagnetic $CoCO_3$.

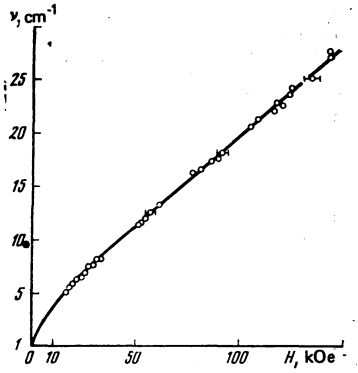

FIG. 6. Variation of the frequency of the LF branch of AFMR in $CoCO_3$ with magnetic field intensity: $\mathbf{H} \perp C_3$, $T = 4.2$ K. Solid curve: calculation by formulas (3), (5), and (14).

3. THEORY

For further discussion, it is necessary to obtain expressions for the spin wave spectrum that take into account the large value of the uniaxial anisotropy and the relatively large value of H_D in $CoCO_3$. We write the Hamiltonian of the crystal in the following form:

$$\mathcal{H} = \frac{1}{2}\sum_{a,b} J_{ab}(\mathbf{S}_a^{(1)}\mathbf{S}_b^{(1)} + \mathbf{S}_a^{(2)}\mathbf{S}_b^{(2)}) + \sum_{a,b} J_{ab}\mathbf{S}_a^{(1)}\mathbf{S}_b^{(2)}$$
$$+ \frac{a}{2}\sum_{a,b}[(S_{az}^{(1)})^2 + (S_{bz}^{(2)})^2] + \sum_{a,b} a_{ab}{}^1 S_{az}^{(1)} S_{bz}^{(2)}$$
$$+ \sum_{a,b} d_{ab}(S_{ax}^{(1)}S_{by}^{(2)} - S_{ay}^{(1)}S_{bx}^{(2)}) - \mu_B \mathbf{H}\sum_{a,b}\sum_{\alpha,\beta}(g_{\alpha\beta}^{(1)}\mathbf{S}_{a\beta}^{(1)} + g_{\alpha\beta}^{(2)}\mathbf{S}_{b\beta}^{(2)}). \quad (2)$$

Here the indices (1) and (2) refer to the sublattices, a and b to the sites; $\alpha, \beta = x, y, z$; μ_B is the Bohr magneton; and

$$g^{(1),(2)} = \begin{pmatrix} g_\perp & \pm g_{xy} & 0 \\ \mp g_{xy} & g_\perp & 0 \\ 0 & 0 & g_{zz} \end{pmatrix}.$$

The first and second terms in the Hamiltonian describe, respectively, the intrasublattice and intersublattice exchange interactions; the third and fourth, the single-ion and interionic anisotropies; the fifth, the antisymmetric exchange or Dzyaloshinskiĭ interaction; and the sixth, the energy of the magnetic material in the external magnetic field. The last term is written in the general form allowed by the symmetry of the crystal.

When no assumptions are made in advance about the values of the parameters in the Hamiltonian, it is appropriate to carry out the analysis by use of the formulas of second quantization. Diagonalization of the Hamiltonian by the method proposed in Ref. 20 leads to the following expression for the energies of the two branches of the spin-wave spectrum ($\mathbf{H} \perp C_3$, see Fig. 7a):

$$[\hbar\omega_{1,2}(\mathbf{k})]^2 = [S(\bar{J}_\mathbf{k} - \bar{J}_0) + S(J_0 \pm J_\mathbf{k})\cos 2\varphi + S(d_0 \pm d_\mathbf{k})\sin 2\varphi + \mu_B g_\perp H(\sin\varphi + \tau\cos\varphi)][S(\bar{J}_\mathbf{k} - \bar{J}_0) + S(a \mp a_\mathbf{k}{}^1) + S(J_0\cos 2\varphi \mp J_\mathbf{k}) + Sd_0\sin 2\varphi + \mu_B g_\perp H(\sin\varphi + \tau\cos\varphi)]. \quad (3)$$

Here the following notation has been introduced:

$$\bar{J}_\mathbf{k} = \sum_{a,b}\bar{J}_{ab}\exp[i\mathbf{k}(\mathbf{r}_a^{(1)} - \mathbf{r}_b^{(1)})],\quad J_\mathbf{k} = \sum_{a,b} J_{ab}\exp[i\mathbf{k}(\mathbf{r}_a^{(1)} - \mathbf{r}_b^{(2)})],$$
$$a_\mathbf{k}{}^1(d_\mathbf{k}) = \sum_{a,b} a_{ab}{}^1(d_{ab})\exp[i\mathbf{k}(\mathbf{r}_a^{(1)} - \mathbf{r}_b^{(2)})],\quad \tau = g_{xy}/g_\perp. \quad (4)$$

The upper sign in (3) refers to the HF branch of the spectrum, the lower to the LF branch. Minimization of the term of zeroth order in the magnon variables of the Hamiltonian leads to the following dependences of φ on H:

$$SJ_0\sin 2\varphi - Sd_0\cos 2\varphi - \mu_B g_\perp H(\cos\varphi - \tau\sin\varphi) = 0. \quad (5)$$

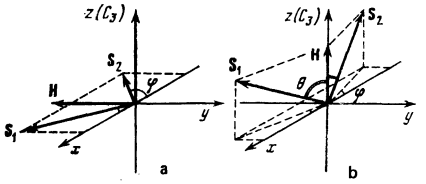

FIG. 7. Orientation of sublattice moments in a magnetic field: a) $\mathbf{H}\perp C_3$; b) $\mathbf{H}\|C_3$.

For the case $\mathbf{H}\|C_3$ (Fig. 7b), the orientation of the moments is determined by the following equations:

$$\operatorname{tg} 2\varphi=\frac{d_0}{J_0},\quad \cos\theta=\frac{\mu_B g_{zz} H}{S[J_0+(J_0^2+d_0^2)^{1/2}+(a+a_0^1)]}. \tag{6}$$

The expression for the energies of the LF and HF branches of the spectrum has the form

$$[\hbar\omega_{1,2}(\mathbf{k})]^2=S^2[(\bar{J}_{\mathbf{k}}-\bar{J}_0)+(J_0^2+d_0^2)^{1/2}\pm(J_{\mathbf{k}}^2+d_{\mathbf{k}}^2)^{1/2}]\times\{(\bar{J}_{\mathbf{k}}-\bar{J}_0)+(J_0^2+d_0^2)^{1/2}\mp J_{\mathbf{k}}+(a\mp a_{\mathbf{k}}^1)\sin^2\theta\pm\pm[(J_{\mathbf{k}}^2+d_{\mathbf{k}}^2)^{1/2}+J_{\mathbf{k}}]\cos^2\theta\}. \tag{7}$$

We shall obtain approximate expressions for the AFMR frequencies ($\mathbf{k}=0$). We introduce the effective fields

$$H_E=\frac{SJ_0}{\mu_B g_\perp},\quad H_A=\frac{S(a-a_0^1)}{\mu_B g_\perp},\quad H_a=\frac{S(a+a_0^1)}{\mu_B g_\perp},\quad H_D=\frac{Sd_0}{\mu_B g_\perp}. \tag{8}$$

Supposing that φ is a small quantity, we determine from equation (5) the trigonometric functions that occur in the expression (3) through terms in $(H/H_E)^2$, $(H_D/H_E)^2$, and τ; we retain this accuracy below. On substituting the results in (3), we obtain the approximate expressions for the AFMR frequencies ($\mathbf{H}\perp C_3$)

$$\left(\frac{\omega_1}{\gamma_\perp}\right)^2=H\left\{H_D\left[1+\frac{H_a}{2H_E}-\frac{1}{2}\left(\frac{H_D}{2H_E}\right)^2-\frac{3}{2}\frac{H_a}{2H_E}\left(\frac{H_D}{2H_E}\right)^2\right]+2H_E\tau+H_a\tau+H\left[1+\frac{H_a}{2H_E}+\left(\frac{H_D}{2H_E}\right)^2-3\frac{H_a}{2H_E}\left(\frac{H_D}{2H_E}\right)^2\right]\right\}, \tag{9a}$$

$$\left(\frac{\omega_2}{\gamma_\perp}\right)^2=2H_EH_A+H_D^2\left(1+\frac{H_A}{H_E}\right)+H\left\{H_D\left[1+\frac{H_A}{2H_E}-\frac{3}{2}\frac{H_A}{2H_E}\left(\frac{H_D}{2H_E}\right)^2+\frac{3}{2}\left(\frac{H_D}{2H_E}\right)^2\right]+2H_E\tau+H_A\tau+H\left[-\frac{H_A}{2H_E}+3\frac{H_A}{2H_E}\left(\frac{H_D}{2H_E}\right)^2+\tau\frac{H_D}{H_E}\right]\right\}. \tag{9b}$$

For $\mathbf{H}\|C_3$, we obtain from (6) and (7) exact expressions for the AFMR frequencies. Then on calculating $(H_E^2+H_D^2)^{1/2}$ with the accuracy indicated above, we get

$$(\omega_1/\gamma_\perp)^2=0, \tag{10a}$$

$$\left(\frac{\omega_2}{\gamma_\perp}\right)^2=2H_EH_A+H_D^2\left(1+\frac{H_A}{H_E}\right)+\left(\frac{g_{zz}}{g_\perp}\right)^2H^2\times\frac{4H_E^2-2H_EH_A+H_D^2(3-H_A/2H_E)}{4H_E^2+4H_EH_a+H_a^2+(H_D/2H_E)^2(1+H_a/2H_E)}. \tag{10b}$$

In accordance with formulas (9a) and (9b), we approximate the experimental results for $\mathbf{H}\perp C_3$, given in Figs. 3 and 6, by a polynomial of the second degree,

$$\nu^2=a_0+a_1H+a_2H^2. \tag{11}$$

It was found that for the LF branch, only the data at fields no larger than 50 kOe could be approximated by a second-degree polynomial. In accordance with the expression (10b), we approximate the results for $\mathbf{H}\|C_3$ by a contracted polynomial of the second degree,

$$\nu^2=a_0+a_2H^2. \tag{12}$$

The results of the approximations, made by the method of least squares, are given in Table II.

TABLE II. Results of approximations.

a_0, cm^{-2}	a_1, cm^{-2}/kOe	a_2, cm^{-1}/kOe^2	Note
0	1.246±0.039 [14]	0.0241±0.0031 [14]	LF AFMR, $\mathbf{H}\perp C_3$, $H\leq 23$ kOe[14]
0.02±0.02	1.221±0.03	0.0259±0.0035	LF AFMR, $\mathbf{H}\perp C_3$, $H\leq 50$ kOe
1196.5±5.5	3.1±0.35	−0.0275±0.0029	HF AFMR, $\mathbf{H}\perp C_3$, $H\leq 50$ kOe
1197±5.2	—	0.0282±0.0026	HF AFMR, $\mathbf{H}\|C_3$, $H\leq 50$ kOe

By comparing the coefficients of the various powers of H in the expressions (9a), (9b), and (10b) with the corresponding coefficients in the extrapolation polynomials, one can determine H_E, H_A, H_a, H_D, $g_\perp$, and g_{zz}. We neglect the nondiagonal components of the g factor. It should be noted that some terms in (9a), (9b), and (10b) exceed the accuracy of determination of the extrapolation coefficients. For estimates of them, we used the values of H_E and H_D given in the Introduction. Therefore for our experimental results, we write the following approximate expressions for the AFMR frequencies:

$$\left(\frac{\omega_1}{\gamma_\perp}\right)^2=HH_D\left(1+\frac{H_a}{2H_E}\right)+H^2\left(1+\frac{H_a}{2H_E}\right), \tag{13a}$$

$$\left(\frac{\omega_2}{\gamma_\perp}\right)^2=2H_EH_A+H_D^2\left(1+\frac{H_A}{H_E}\right)+HH_D\left(1+\frac{H_A}{2H_E}\right)+H^2\left(-\frac{H_A}{2H_E}\right) \tag{13b}$$

for $\mathbf{H}\perp C_3$, and

$$\left(\frac{\omega_2}{\gamma_\perp}\right)^2=2H_EH_A+H_D^2\left(1+\frac{H_A}{H_E}\right)+\left(\frac{g_{zz}}{g_\perp}\right)^2H^2\frac{4H_E^2-2H_EH_A}{(2H_E+H_a)^2} \tag{13c}$$

for $\mathbf{H}\|C_3$. These expressions together with the data of Table II permit determination of all the parameters of the Hamiltonian. At this stage of the calculation, the necessity for taking account of the interionic anisotropy in the Hamiltonian manifests itself. Without allowance for the interionic anisotropy, the system of equations obtained from the expressions (13) and the data of Table II has no solutions.

Thus by use of the approximate expressions for the AFMR frequencies, the following parameters are determined: $2H_E=190$ kOe, $H_A=130$ kOe, $H_a=-60$ kOe, $H_D=47$ kOe, $g_\perp=4.3$, $g_{zz}=3.9$. The errors in the determination of some of these parameters reach large values. Therefore these values were used as an initial approximation for a more accurate determination of them by use of the expressions (3) and (5)–(7) and of all the experimental data given in Figs. 3 and 6. With a computer, under the condition

$$|\nu_{\text{calc}}-\nu_{\text{exp}}|<\delta,$$

where δ is the experimental error, the following values were found:

$$2H_E=260\pm15\text{ kOe},\ H_A=116\pm5\text{ kOe},\ H_a=-61\pm6\text{ kOe},$$
$$H_D=47\pm4\text{ kOe},\ g_\perp=4.0\pm0.1,\ g_{zz}=3.65\pm0.35. \tag{14}$$

The relatively high accuracy of determination of these values is mainly due to the use of data obtained in fields of intensity up to 145 kOe (Fig. 6).

Figure 8 shows the variations of the frequencies of the HF and LF branches of the AFMR spectrum, over a wide range of magnetic fields, calculated by formu-

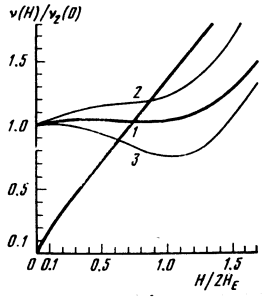

FIG. 8. Variation of AFMR frequencies with magnetic field intensity ($\mathbf{H}\perp C_3$): 1) calculation by formulas (3), (5), and (14); 2) $H_A/H_E = 0.33$, $H_D/H_E = 0.33$; 3) $H_A/H_E = 1$, $H_D/H_E = 0.17$.

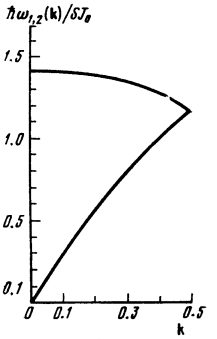

FIG. 9. Dispersion of spin waves in $CoCO_3$ for $\mathbf{k}\|[111]$. Calculation by formula (18).

las (3) and (5) with use of the values (14). Also shown here are the variations of the HF branch of the spectrum calculated with various relations among H_E, H_A, and H_D. Similar variations, but for other relations among these parameters, have been given in a paper of Ozhogin.[10]

The results obtained enable us to calculate the dispersion of spin waves in $CoCO_3$. The goal of the calculation is an explanation of the nature of the two-magnon absorption. Since electric-dipole two-magnon absorption may be due to singularities of the density of states at the points Z and A of the Brillouin zone,[21] we shall restrict ourselves to the case in which the wave vector does not depart from the $(1\bar{1}0)$ plane of the reciprocal lattice. In this case, the value of the reduced wave vector is (ζ, ζ, μ), and the expression (4) takes the form[22]

$$\bar{J}_{\mathbf{k}} = 2\bar{J}_1\{1+2[\cos 2\pi(\zeta-\mu)]\}. \tag{15a}$$

$$J_{\mathbf{k}} = 2J_1\{2\cos\pi\mu+\cos[\pi(2\zeta-\mu)]\} + 2J_2\{2\cos[\pi(2\zeta-\mu)]+\cos[\pi(2\zeta-3\mu)]\}, \tag{15b}$$

$$a^1_{\mathbf{k}}(d_{\mathbf{k}}) = 2a^1(d)\{2\cos\pi\mu+\cos[\pi(2\zeta-\mu)]\}. \tag{15c}$$

Here $\bar{J}_1$ is the exchange integral between nearest neighbors on a single sublattice; J_1 and J_2 are the exchange integrals between nearest and next-nearest neighbors on different sublattices; a^1 and d are the parameters of interionic anisotropy and of antisymmetric exchange for nearest neighbors on different sublattices.

The expressions (8) and (15b) enable us to determine the sum $J_1 + J_2$ of the intersublattice exchange integrals. Starting from the AFMR data (14) and setting $S = \frac{3}{2}$, we find

$$J_1 + J_2 = 81\cdot 10^9 \text{ sec}^{-1}. \tag{16}$$

It is impossible to determine from our experimental data the intrasublattice exchange integral $\bar{J}_1$; this prevents calculation of $\omega_{1,2}(\mathbf{k})$ for an arbitrary direction in the Brillouin zone. But such a calculation can be made for one of the directions of interest to us. If $\mathbf{k}\|[111]$, i.e., $\zeta = \mu$, then, as follows from the expression (15a), $\bar{J}_{\mathbf{k}} = \bar{J}_0$. In this case, by using (15) and setting $H = 0$, we get from formula (3) the following dependence of the spin-wave energies on the wave vector for $\mathbf{k}\|[111]$:

$$\left(\frac{\hbar\omega_{1,2}(\mathbf{k})}{SJ_0}\right)^2 = \left[(1\pm\cos\pi\mu)\cos 2\varphi + \frac{d_0}{J_0}(1\pm\cos\pi\mu)\sin 2\varphi\right] \times\left[\left(\frac{a}{J_0}\mp\frac{a_0^1}{J_0}\cos\pi\mu\right)+(\cos 2\varphi\mp\cos\pi\mu)+\frac{d_0}{J_0}\sin 2\varphi\right]. \tag{17}$$

From the data (14) for $CoCO_3$, we find a_0/J_0, a_0^1/J_0, and d_0/J_0; then by use of expressions (5) and (17), we get the following dependence of the spin-wave energies on the wave vector in $CoCO_3$:

$$\left(\frac{\hbar\omega_{1,2}(\mathbf{k})}{SJ_0}\right)^2 = 1.355(1\pm\cos\pi\mu)(1\mp 0.256\cos\pi\mu). \tag{18}$$

The dispersion relations calculated by formula (18) are shown in Fig. 9.

With the aid of expression (18) one can calculate the energy of magnons with wave vector $\mathbf{k} = [\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2}]$. This energy corresponds to frequency $\nu_{\text{calc}}(k_{z\,\max}) = 28.4$ cm^{-1}. This value agrees well with the two-magnon absorption frequency determined in the experiment: $2\nu_{\text{exp}}(\mathbf{k}_{\max}) = 54.7$ cm^{-1}.

From expressions (3), (5), and (15) we obtain the dependence of the energies of magnons with $\mathbf{k} = [\frac{1}{2}\,\frac{1}{2}\,\frac{1}{2}]$ on the magnetic field in the case $\mathbf{H}\perp C_3$:

$$\left(\frac{\hbar\omega_{1,2}(k_{z\,\max})}{SJ_0}\right)^2 = \left(\cos 2\varphi + \frac{d_0}{J_0}\sin 2\varphi + \frac{\mu_B g_\perp H}{J_0}\sin\varphi\right) \times\left(\frac{a}{J_0}+\cos 2\varphi+\frac{d_0}{J_0}\sin 2\varphi+\frac{\mu_B g_\perp H}{J_0}\sin\varphi\right). \tag{19}$$

By solving equation (5) numerically, we determine the orientation of the moments of the sublattices in $CoCO_3$ in a magnetic field $\mathbf{H}\perp C_3$; see Table III. By use of the value obtained for a/J_0, one can calculate the magnetic-field-dependent energy of a magnon with the wave vector belonging to point Z of the Brillouin zone. The results of the calculation are shown in Fig. 5. We get the analogous relation for $\mathbf{H}\|C_3$ from expressions (6) and (7):

$$\left(\frac{\hbar\omega_{1,2}(k_{z\,\max})}{SJ_0}\right)^2 = \left(1+\frac{d_0^2}{J_0^2}\right)^{1/2}\left[\left(1+\frac{d_0^2}{J_0^2}\right)^{1/2}+\frac{a}{J_0} - \frac{a}{J_0}\left(\frac{\mu_B g_{zz} H}{J_0+(J_0^2+d_0^2)^{1/2}+(a+a_0^1)}\right)^2\right]. \tag{20}$$

The calculated relation (20) for $CoCO_3$ is also shown in Fig. 5; it is evident that the calculated relations agree qualitatively with the experimental. The quantitative discrepancy in the case $\mathbf{H}\perp C_3$ may be a consequence of

TABLE III. Orientation of sublattice moments, $\mathbf{H}\perp C_3$.

φ, rad:	0.174	0.210	0.245	0.279	0.314	0.348
H, kOe:	0	10	20	30	40	50

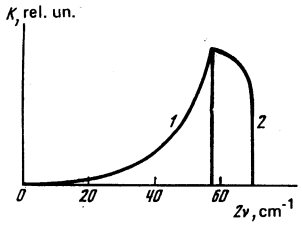

FIG. 10. Two-particle absorption coefficient (calculated by formula (21) with $W(\mathbf{k})=\text{const}$). The curves correspond to excitation of two magnons from the LF branch (1) and from the HF branch (2) of the spectrum.

the fact that in the calculation the magnon-magnon interaction, for example, was neglected; it may significantly change the spectrum near the edge of the zone.

By use of the dispersion relations (18), we calculate the two-particle absorption coefficient:

$$\alpha(2\nu)=Ck^2\left|\frac{dE(\mathbf{k})}{d\mathbf{k}}\right||W(\mathbf{k})|^2. \qquad (21)$$

Here $dE(\mathbf{k})/d\mathbf{k}$ is the density of states in the spin-wave band, and $W(\mathbf{k})$ is the probability of simultaneous excitation of two ions.[23] Figure 10 shows the results of calculations by formula (21). Curve 2 in Fig. 10 corresponds to simultaneous excitation of two magnons from the HF branch of the spectrum. Such a process is characterized by a flatter high-frequency absorption edge. The frequency of the absorption maximum of such a process [$2\nu(k_{z\,\max})=56.8$ cm^{-1}], and also the frequency interval in which observation of it is possible, are close to what was observed experimentally (Fig. 1). All of this enables us to conclude that the electric-dipole two-magnon absorption at frequency 54.7 cm^{-1} is due to simultaneous excitation of two magnons from the HF branch of the spin-wave spectrum.

4. CONCLUSION

We shall state briefly the basic results of the research. The HF branch of AFMR and two-magnon absorption have been experimentally detected and studied in detail in the weak ferromagnet $CoCO_3$; the behavior of the LF branch of AFMR has been investigated in magnetic fields comparable with the crystal exchange field; it has been shown that for quantitative description of the behavior of the resonance frequencies in a magnetic field, expressions obtained on the assumption that $H_A \ll H_E$ and $H_D \ll H_E$ are inapplicable; expressions for the AFMR frequencies have been obtained that take into account the large value of the uniaxial anisotropy and the appreciable Dzyaloshinskiĭ interaction; the basic parameters have been determined in the Hamiltonian that describes the magnetic properties of $CoCO_3$; by use of these parameter values, the dispersion of magnons along the direction [111] in the Brillouin zone has been calculated; it has been shown that the electric-dipole two-magnon absorption in $CoCO_3$ is due to simultaneous excitation of two magnons from the HF branch of the spectrum.

It must be noted that use of expressions for the AFMR frequencies that took into account the large value of the uniaxial anisotropy and the appreciable Dzyaloshinskii interaction did not remove the contradiction between the values of H_D determined from static and from resonance measurements. Also noteworthy is the large value of the interionic-anisotropy parameter obtained in the present paper. These two facts may be due to the fact that the sublattice moments depart from the basal plane of the crystal.

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