Parametric excitation of magnons in antiferromagnetic $CoCO_3$

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The parametric excitation of magnons in the easy-plane antiferromagnet $CoCO_3$ is studied by the method of parallel pumping at frequencies $\omega_P/2\pi = 33$–40 GHz. The experiments were done in the temperature range 1.2–4.2 K. It is found that the excitation process is of a "hard" character. Measurements of the threshold field h_{c1} for parametric excitation of magnons in crystals with different Fe^{++} impurity contents have shown that at low temperatures the relaxation of the magnons is governed mainly by their interaction with these impurities. An anomaly is observed on the $h_{c1}(H)$ curve and is attributed to the intersection of the magnon and phonon spectra. A value $\alpha = (0.7 \pm 0.2)\cdot 10^{-2}$ Oe cm is found for the exchange constant in the magnon spectrum.

Cobalt carbonate is one of the best studied antiferromagnets with the Dzyaloshinskiĭ interaction. The symmetry of $CoCO_3$ is described by the space group D^6_{3d}. Neutron-diffraction[1] and magnetometric[2] studies of the magnetic structure have shown that below the Néel temperature $T_N = 18.1$ K $CoCO_3$ is an antiferromagnet exhibiting weak antiferromagnetism and easy-plane anisotropy. The magnon spectrum in antiferromagnets of this type has been calculated with allowance for the dipole-dipole interaction by Ozhogin[3] and Bar'yakhtar, Savchenko, and Tarasenko,[4] who proceeded from a phenomenological Hamiltonian. If the external field **H** lies in the easy, basal plane of the crystal ($z \| C_3, x \| \mathbf{H}$), then the lower, quasiferromagnetic branch of the spectrum is of the form

$$\left(\frac{\omega}{\gamma}\right)^2 = \left(\frac{\omega_0}{\gamma}\right)^2 + 4\pi\chi\left[\left(\frac{\omega_0}{\gamma}\right)^2\frac{k_z^2}{k^2} + (H+H_D)^2\frac{k_y^2}{k^2}\right], \qquad (1)$$

where

$$(\omega_0/\gamma)^2 = H(H+H_D) + \alpha_\perp^2(k_x^2+k_y^2) + \alpha_\|^2 k_z^2 + H_{\Delta 1}^2 + H_{\Delta 2}^2 \cos 6\varphi$$

is the magnon spectrum calculated without allowance for the dipole-dipole interaction.[5,6] Here H_D is the Dzyaloshinskiĭ field; γ is the magnetomechanical ratio; $\alpha_\|$ and $\alpha_\perp$ are the exchange constants for magnons propagating along the trigonal axis C_3 and in the basal plane, respectively; **k** is the magnon wave vector; $H_{\Delta 1}$ is a parameter determined by the interactions of the magnetic system with lattice vibrations, nuclear moments, and impurities; $H_{\Delta 2}$ is a parameter determined by the anisotropy in the basal plane; and χ is the magnetic susceptibility in the basal plane at $T = 0$ K.

Cobalt carbonate has been studied by the methods of static magnetometry,[2,7] calorimetry,[8] antiferromagnetic resonance,[9–13] electron paramagnetic resonance,[14] and Brillouin scattering.[15] The values of the magnetic parameters determined in these experiments are given in Table I. The magnon spectrum of $CoCO_3$ has also been calculated in the framework of a microscopic model by Ivanov, Loktev, and Pogorelov.[16] The calculated values of the parameters are also given in Table I. The substantial differences in the values of $\gamma^2 H_D$ obtained from antiferromagnetic-resonance measurements by different authors are probably due to the different Mn^{++} impurity contents in the investigated crystals; this impurity has been found[12] to have a substantial effect on the Dzyaloshinskiĭ field H_D. The introduction of one Mn^{++} ion per 100 Co^{++} ions in $CoCO_3$ decreases H_D by about 30%. We have measured the field dependence of the antiferromagnetic resonance frequency in the resistivity range $2\pi(16$–$53)$ GHz. The value of $\gamma^2 H_D$ for our samples is marked with an asterisk.

Zhotikov and Kreĭnes[17] also excited parametric magnons and phonons at a temperature below 2 K and detected them by Brillouin scattering. In the present study we investigate the parametric excitation of quasiparticles in $CoCO_3$ and study their relaxation.

TECHNIQUES AND SAMPLES

The $CoCO_3$ samples were grown by Ikornikova and Egorov at the Institute of Crystallography of the Academy of Sciences of the USSR.[12,14] In our measurements we used $CoCO_3$ samples grown in the form of thin slabs in order to decrease the influence of the demagnetizing fields. The ratio of the characteristic transverse dimension to the thickness (≈ 0.15 mm) for these samples was 10–12.

To study the parametric excitation of quasiparticles in $CoCO_3$ we used a microwave spectrometer that has been described in detail elsewhere.[18] The measurement cell was a high-Q cylindrical resonator ($Q \approx 10^4$) in which the TE_{012} mode was excited. The sample under study was mounted on the bottom of the resonator at an antinode of the microwave magnetic field h. The samples were prepared two ways. In the first case the sample was cut so that the crystallographic axis C_3 was perpendicular to the plane of the slab. In this case the sample was mounted with a small pocket of cigarette paper in order to prevent stresses from arising in the crystal on cooling. In the second case the sample was cut in such a way that the C_3 axis lay in the plane of the slab. In this case the sample was cemented by one end to the bottom of the resonator. The relative orientation of the microwave magnetic field **h**, the static magnetic field **H**, and the crystallo-

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TABLE I.

Method	$\chi \cdot 10^{-3}$, CGSM/cm^3	$H_{D\,stat}$, kOe	$H_{D\,rf}$, kOe	g	$\gamma^2 H_{D\,rf}$, GHz^2/kOe	$H^2_{\Delta 1}$ kOe^2	$H^2_{\Delta 2}$, kOe^2	$\alpha \cdot 10^{-2}$, Oe cm
Statics	1,84 [2]	27 [2,7]						
Specific heat [8]								0,68
EPR [14]				$g_\parallel = 3.07 \pm 0.03$				
				$g_\perp = 4.96 \pm 0.02$				
AFMR			51.5±8 [9]	3.3 [9]	1120±35 [9]	1 [13]	2–4 [13]	
			47±4 [11]	3.9 [11]	847 [10], 742 [12]			
					1473±130 [11] 890±80 *			
Brillouin scattering [15]					927±70			$\alpha_\perp = 1.24 \pm 0.06$; $\alpha_\parallel = 0.94 \pm 0.3$
Parametric excitation								0.7±0.2
Calculated values of the constants [16]		27,2	38,4	3,4	870			$\alpha_\perp = 0.19$; $\alpha_\parallel = 0.38$

graphic axis C_3 is shown in Figs. 1 and 3.

For the samples cut in the first way it was convenient to make angular measurements, since the values of the demagnetizing fields did not change as the magnetic field **H** was rotated in the plane of the slab. For the samples cut in the second way it was convenient to put the external static field **H** in the plane of the slab, since the antiferromagnetic resonance field depends strongly on the angle between **H** and the plane of the slab. The static magnetic field was measured by a Hall probe. The microwave sources were klystrons with frequencies $2\pi(16–53)$ GHz and powers of around 5 mW and a cw magnetron with a frequency $\omega_P = 2\pi \cdot 35.6$ GHz and a power ~ 10 W. The magnetron operated in the long-pulse regime with a repetition frequency of 50 Hz. The pulse length could be varied from 0.01 to 1 msec.

The resonator with the sample was placed in a Dewar containing liquid helium. The temperature was determined from the saturated vapor pressure of helium and could be varied from 1.2 to 4.2 K. The uncertainty in the measurement and maintenance of the temperature was less than 5%. The microwave pulse transmitted through the resonator with the sample inside was detected and observed on an oscilloscope. The power P, proportional to the square of the field h in the sample, was measured with a quadratic detector which was calibrated by thermistor measurements of the power. The field h in the sample was calculated from the value of P and the resonator parameters with an absolute error of 20%. Here it was assumed that the thin sample placed on the bottom of the resonator did not substantially change the field distribution in the resonator. The relative uncertainty in determining h in a single series of measurements was considerably less than 3%.

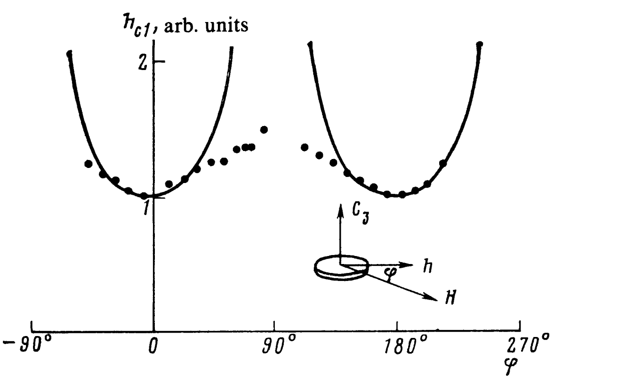

FIG. 1. Threshold field h_{c1} versus the angle φ between the static magnetic field **H** and the microwave magnetic field **h**; $H = 150$ Oe, $\omega_p/2\pi = 35.6$ GHz, $T = 1.2$ K.

PRESENTATION AND DISCUSSION OF EXPERIMENTAL RESULTS

The experiments showed that in low static fields, beginning at a certain level of the microwave field h_{c1} in the sample, the pulses transmitted by the resonator exhibit a characteristic distortion. The form of the distortion indicates that the absorption of microwave power by the sample is increasing in time. Thus we have observed the usual picture for a parametric process.

The threshold for parametric excitation can be expressed in terms of the coupling coefficient V of the excited quasiparticles with the microwave pump and their relaxation parameter $\Delta\omega$: $h_c = \Delta\omega / V$.[19] The coefficient V is calculated theoretically for each specific process and is determined by the characteristic magnetic properties of the material and the external conditions. In the experiments described here the threshold for excitation of parametric quasiparticles is minimum when the fields **H** and **h** are parallel and lie in the basal plane of the crystal, as can be seen from the angular dependence of the threshold field (Fig. 1). Judging from the shape of the angular dependence of the threshold field, one can assume that the observed parametric process depends substantially on the projection of the microwave field **h** onto the direction of **H** and we therefore measured all the temperature, frequency and field dependences for a parallel orientation of **h** and **H**.

At low temperatures the parametric excitation of quasiparticles is of a "hard" nature; i.e., is characterized by two threshold fields h_{c1} and h_{c2}, corresponding to the onset and cutoff of excitation. This type of process is typical for the parametric excitation of magnons[18,20] and can apparently be attributed to the "turning off" of a certain part of the relaxation as the number of excited quasiparticles grows. In $CoCO_3$ at 1.2 K the part of the relaxation that turns off reaches 50% of the total. Figure 2 shows the dependence of the threshold field h_{c1} on the static magnetic field. Also shown in Fig. 2 is a trace from an x-y chart recorder with the Hall-probe signal, proportional to the magnetic field H, fed to the x coordinate and the detector signal, proportional to the microwave power passing through the resonator, fed to the y coordinate. The arrow indicates the magnetic field H_0 at which the lowest-frequency magnons having k close to zero and propagating along the x axis have frequency $\omega_P/2$. The value of H_0 is calculated with formula (1) under the assumption that there are no gaps in the magnon spectrum. The sharp decrease in the absorbed power and the increase in the threshold field in fields close to H_0 suggests that the excited quasiparticles are magnons with frequency $\omega_P/2$. The coupling coefficient V of the microwave pump for the parametric excitation of magnons in easy-plane antiferromagnets has been calculated by Ozhogin.[19] The coefficient V has also been calculated with allowance for the dipole-dipole interaction by Lutovinov and Safonov.[21] According to Ref. 21, under the conditions of our experiment (pump frequency ω_P much lower than the homogeneous precession frequency ω_{20} of the quasi-antiferromagnetic branch, $H \ll H_D$, $\mathbf{H} \| \mathbf{h} \perp C_3$) the coupling coefficient V is independent (to $\sim 5\%$) of the direction of the wave vector of the excited magnons and is given by

$$V = \omega_P/\gamma^2 H_D. \qquad (2)$$

At a temperature of 1.2 K and in a field $H = 200$ Oe, sample

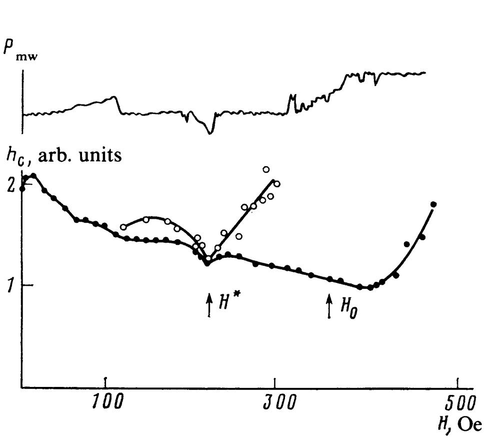

FIG. 2. Threshold field h_{c1} (●) and threshold field h_c for the second parametric instability (○) versus the field H. The upper part of the figure shows a chart-recorder trace of the microwave power P_{mw} transmitted by the resonator versus the field H; $\omega_P/2\pi = 35.6$ GHz; $T = 1.2$ K.

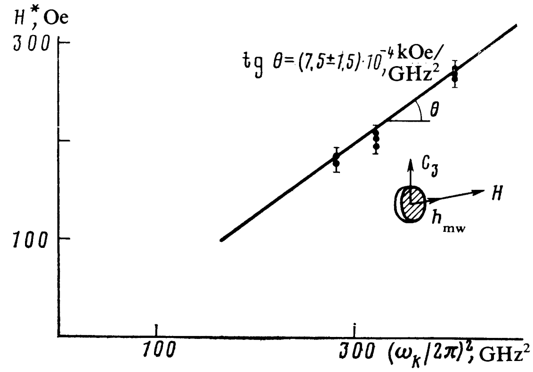

FIG. 3. The field H^* at which the anomaly is observed in the magnon relaxation versus the square of the magnon frequency $(\omega_k/2\pi)^2$, $T = 1.2$ K.

I, which was cut the first way, had a threshold field $h_{c1} = 0.2$ Oe. Having evaluated the coefficient V, we can estimate the relaxation frequency $\Delta\omega$, which is found to be 5 MHz. In small fields ($H \leqslant 40$ Oe) a peak appears on the $h_{c1}(H)$ curve. Such a peak of the threshold microwave field was observed in all of our samples and at all pump frequencies in the interval $2\pi(33$–$40)$ GHz. It is apparently due to a reorientation of the domains in the crystal.

In a field H^* all of our samples exhibited a resonant decrease in the threshold microwave field. Studies of the parametric excitation of magnons in $MnCO_3$ and $CsMnF_3$ crystals[22,23] have also revealed anomalies in the threshold field. These anomalies were attributed to the intersection of the magnon and phonon spectra. Figure 3 shows the dependence of the field H^* on the square of the magnon frequency $\omega_k = \omega_P/2$ in $CoCO_3$. Within the experimental accuracy this dependence is linear. Such a form of $H^*(\omega_k^2)$ suggests that the observed anomaly in $CoCO_3$ is of the same nature as the anomalies in $MnCO_3$ and $CsMnF_3$. The condition for intersection of the magnon and phonon spectra at a frequency $\omega_k = \omega_P/2$ in a field H^* is written

$$(\omega_k/\gamma)^2 = H^*(H^* + H_D) + \alpha_{\mathbf{k}}^2 k^2 + H_{\Delta\mathbf{k}}^2, \quad \omega_k = c_{\mathbf{k}} k, \qquad (3)$$

where $c_{\mathbf{k}}$ is the sound velocity in the $\mathbf{k}$ direction, $\alpha_{\mathbf{k}}$ is the exchange constant in the spectrum of magnons with wave vector $\mathbf{k}$, and $H_{\Delta\mathbf{k}}$ is the corresponding parameter in the spectrum of these magnons. Taking into account the fact that $H^* \ll H_D$, we obtain a solution of the system of equations (3) in the form

$$H^* = \frac{\omega_k^2(1 - \alpha_{\mathbf{k}}^2 \gamma^2 / c_{\mathbf{k}}^2) - \gamma^2 H_{\Delta\mathbf{k}}^2}{\gamma^2 H_D}. \qquad (4)$$

It is seen from Eq. (4) that the slope of the curve $H^*[(\omega_P/2)^2]$ is determined by the value of the exchange constant $\alpha_{\mathbf{k}}$ and sound velocity $c_{\mathbf{k}}$. More strongly coupled with magnons in an easy-plane antiferromagnet is the transverse sound, whose velocity in $CoCO_3$ varies, depending on the propagation direction, over the limits $(3.7$–$4.2) \cdot 10^5$ cm/sec.[15] From Fig. 3 one can estimate the value of the exchange constant $\alpha_{\mathbf{k}}$ of the excited group of spin waves. It is found to be $\alpha_{\mathbf{k}} = (0.7 \pm 0.2) \cdot 10^{-2}$ Oe cm.

Brillouin scattering by parametrically excited quasiparticles was studied in Refs. 17 and 24. According to the data from these studies no magnons with $\mathbf{k}\|x$ are excited at a pump frequency of $\omega_P/2\pi = 36$ GHz. This seems to imply that in the present case we are exciting magnons with $\mathbf{k}\|z$, which effectively interact with phonons propagating along the C_3 axis ($\mathbf{k}_{ph}\|z$) and polarized along $\mathbf{H}$.[25] Thus the value we have determined for $\alpha_\mathbf{k}$ corresponds to $\alpha_\|$. One might wonder why our results imply that the excited quasi-particles are magnons with $\omega_k = \omega_P/2$, while at the same pump frequency other authors[24] have observed scattering by phonons, and not magnons, at $\omega_{ph} = \omega_P/2$. This is possibly due to the different excitation conditions in the optical experiment: the fields $\mathbf{h}$ and $\mathbf{H}$ were applied at an angle of 45° to the basal plane. On the other hand, it is possible that the phonons observed in Ref. 24 are secondary quasiparticles arising as a result of a conversion of the parametrically excited magnons into phonons at impurity ions, without loss of energy.

When the field h appreciably exceeds the threshold value h_{c1} in the range of fields $H = 120$–300 Oe, yet another parametric process can occur. In this case the pulse transmitted by the resonator exhibits two chips, with different characteristic development times for the parametric instabilities. The threshold field h_c for the second instability is plotted in Fig. 2 as a function of $\mathbf{H}$. It is seen that in a field H^* the threshold fields for the two instabilities coincide. The second parametric process was observed in only one crystal—the one with the lowest threshold field h_{c1}. Judging from the coincidence of the threshold fields of the two processes at the point of intersection of the magnon and phonon spectra, one can assume that the second parametric process involves the creation of two phonons or a phonon and a magnon.

The threshold microwave field h_{c1} at a temperature of 1.2 K varied from sample to sample. For samples cut from different single crystals it varied by more than an order of magnitude (0.14–3 Oe). For samples cut from the same crystal, the threshold field was the same to within the absolute error of the experiment (20%). Since according to (2) the coupling coefficient V of the excited magnons with the microwave pump contains only the quantity $\gamma^2 H_D$, which can vary from sample to sample, while this coefficient itself does

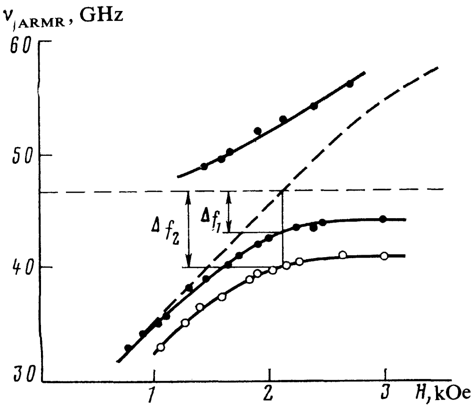

FIG. 4. Antiferromagnetic resonance spectrum for two crystals at temperature $T = 1.2$ K; (○) crystal I, (●) crystal II.

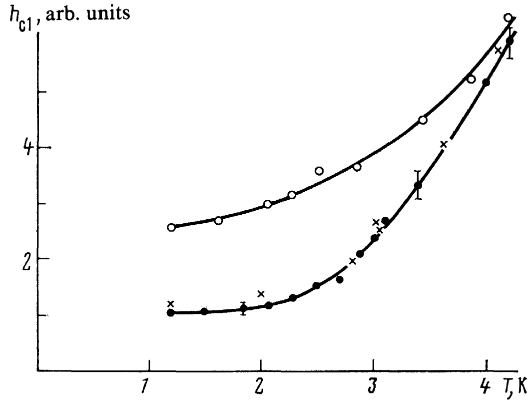

FIG. 5. Temperature dependence of h_{c1} for the same crystal in fields $H = 300$ Oe (●) and 150 Oe (×); ○ shows the temperature dependence of h_{c1} for another crystal in a field $H = 300$ Oe, $\omega_P/2\pi = 35.6$ GHz.

not change outside of an error of 10%, it is natural to assume that the relaxation of magnons at low temperatures in our $CoCO_3$ samples is governed by extrinsic processes—scattering by impurities.

Our $CoCO_3$ crystals could contain certain amounts of Fe^{++} and Mn^{++} impurities.[12] The frequency of the first excited level of Fe^{++} is $2\pi \cdot 45$ GHz,[12,13] i.e., rather close to the frequency of the excited quasi-particles: $2\pi \cdot 18$ GHz. It is natural to assume that the scattering of the quasi-particles by this impurity is the most active. The presence of Fe^{++} impurities also leads to splitting of the antiferromagnetic resonance spectrum. As was shown in Refs. 12 and 16, the splitting Δf is proportional to the square root of the impurity concentration C. We chose two $CoCO_3$ crystals (I and II) having very different threshold fields $h_{c1\text{I}}$ and $h_{c1\text{II}}$ at $T = 1.2$ K ($h_{c1\text{I}}/h_{c1\text{II}} = 6 \pm 1.5$) and measured the splitting of the antiferromagnetic resonance spectrum in these crystals (Fig. 4). The ratio of the squares of the splittings was found to be

$$\Delta f_\text{I}^2/\Delta f_\text{II}^2 = C_\text{I}/C_\text{II} = 4.5 \pm 1.$$

This result agrees qualitatively with the assumption that the main relaxation process of the parametrically excited magnons in our $CoCO_3$ samples at $T \sim 1.2$ K occurs at Fe^{++} ions. From the size of the splitting of the spectrum and the data of Ref. 12, one can estimate the content of impurity iron. Our estimate shows that in sample I there is one Fe^{++} ion for every 400 Co^{++} ions.

Figure 5 gives the temperature dependence of the threshold field h_{c1} for the excitation of magnons in the two crystals. At a temperature of 1.2 K the value of the threshold field h_{c1} was 2.25 times as large in one of the samples as in the other. At 4.2 K the values of h_{c1} for the two crystals differed by no more than 20%. This temperature dependences of h_{c1} could mean that at temperatures $\gtrsim 3$ K the intrinsic relaxation processes become more effective. Interestingly, the "hardness" $(h_{c1} - h_{c2})/h_{c1}$ of the parametric excitation of magnons decreases with increasing temperature, and at ~ 3 K the parametric process becomes "soft."

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