Scattering of light in weakly ferromagnetic $CoCO_3$ following excitation of the spin system by large microwave power

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Intense peaks corresponding to magnons and phonons parametrically excited by microwave pumping have been observed in the spectrum of light scattered in weakly ferromagnetic $CoCO_3$. The frequency of these peaks is equal to half the pump frequency.

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We have investigated the spectral composition of light scattered at 90° to the incident beam in a $CoCO_3$ crystal at $T \lesssim 2$ K in a magnetic field, following application of relatively large microwave power.

The spectral instrument was the high-contrast ($\sim 10^7$–10^8) three-pass scanning Fabry-Perot interferometer made by the Burleigh firm (USA). All the investigations were made with a setup that differed from that described in[1] in that (a) the light incident on the interferometer was scattered by the sample at 90° to the incident beam; (b) the recording instrument (past the photon counter) was a multichannel storage unit DAS-I, also made by Burleigh. This instrument served also to adjust the interferometer automatically; (c) the microwave radiation power had a frequency $\nu = 35.4$ GHz and an output power ~ 2 W; (d) the power of the light source ($\lambda = 6328$ Å)—LG–36A laser—was ~ 50 mW. The experiments were performed on the same $CoCO_3$ sample[1)] as in[2]. The employed experimental geometry made it possible to investigate the scattering of light by elementary excitations (spin waves or phonons) with wave vector $\mathbf{k} = 2.5 \times 10^5$ cm^{-1}.

Below $T_N = 18.1$ K, the crystal $CoCO_3$ is an antiferromagnet with a weak ferromagnetism. Its magnetic, resonant, and optical properties have by now been sufficiently well investigated.[3–6]

In the previous study,[2] we investigated the scattering of light in this crystal by spin waves that were either thermal or excited by AFMR at low microwave powers (~ 5 mW). The increase of the microwave power led to observation of a number of new experimental facts, which are reported here. We investigated scattering by elementary excitations propagating either along the C_3 axis (z axis) of the crystal ($\mathbf{k} = \mathbf{k}_z$) or in the basal plane of the x axis[2)] ($\mathbf{k} = \mathbf{k}_x$).

We report first the results obtained for $\mathbf{k} = \mathbf{k}_z$.

I. At a definite level of the applied microwave power (pump) $P_0 \approx 50$ mW, under conditions of excitation of the low-frequency AFMR branch ($\nu = 35.4$ GHz, $H = 1280$ Oe) there appear in the

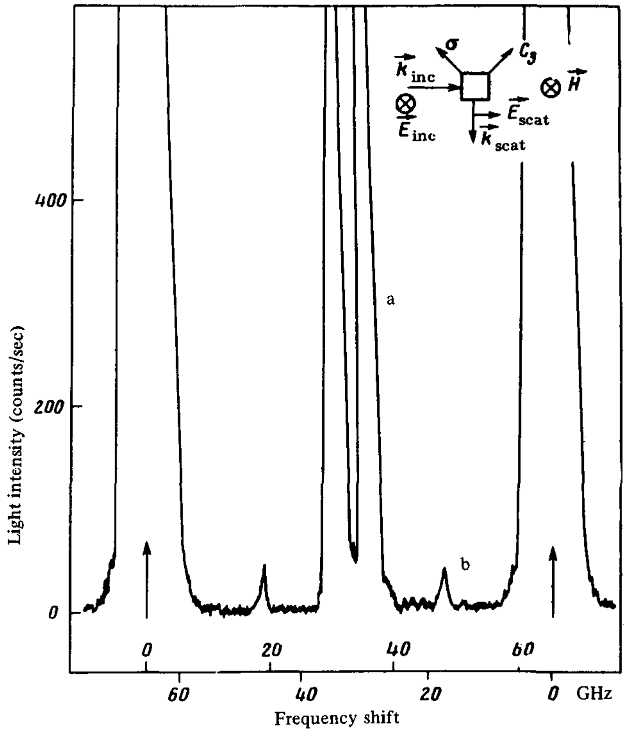

FIG. 1. Spectrum of light scattered at 90° in $CoCO_3$ ($T \lesssim 2$ K) by excitations propagating along z, following application of microwave power ($P \sim 400$ mW) under AFMR conditions ($H = 1280$ Oe, $\nu = 35.4$ GHz): a—enhanced magnons, b—excitations at half the frequency.

scattered light-spectrum, besides the peaks corresponding to the enhanced magnons (compared with the thermal ones at $H = 1280$ Oe[2]) at the pump frequency (Fig. 1a), additional satellites of lower intensity, with frequency equal to half the pump frequency (Fig. 1b). These satellites exist in a narrow interval of magnetic field ~ 40 Oe, which coincides in width with the region of the existence of the amplification of the magnon peaks at the fundamental frequency. However, the maximum of the intensity of these lines is shifted towards larger fields by ~ 20 Oe in comparison with the maximum of the intensity of the enhanced magnons. The pump power P_0 corresponds to the start of the deviation from linearity in the dependence of the intensity of the enhanced magnon peaks on the microwave power. The polarization conditions in these experiments were the same as in the observation of thermal magnons, i.e., $\mathbf{E}_{scat} \perp \mathbf{E}_{inc}$.

II. By investigating the spectrum of the light scattered in a weak magnetic field ($H = 230$ Oe, the same experimental geometry as in Sec. I) with microwave pumping at 35.4 GHz, we were able to observe the enhancement of thermal magnons at a frequency 17.8 ± 0.4 GHz (Fig. 2). This enhancement likewise exists in a very narrow interval of magnetic fields, ± 10 Oe. The dependence of the enhancement on the microwave power seems to have no threshold. At powers ~ 300 mW, saturation of the enhancement takes place. The intensity of the enhanced peaks is then ~ 20 times larger than that of the thermal peak at a microwave power ~ 500 mW. In these experiments, too, the condition $\mathbf{E}_{scat} \perp \mathbf{E}_{inc}$ is satisfied.

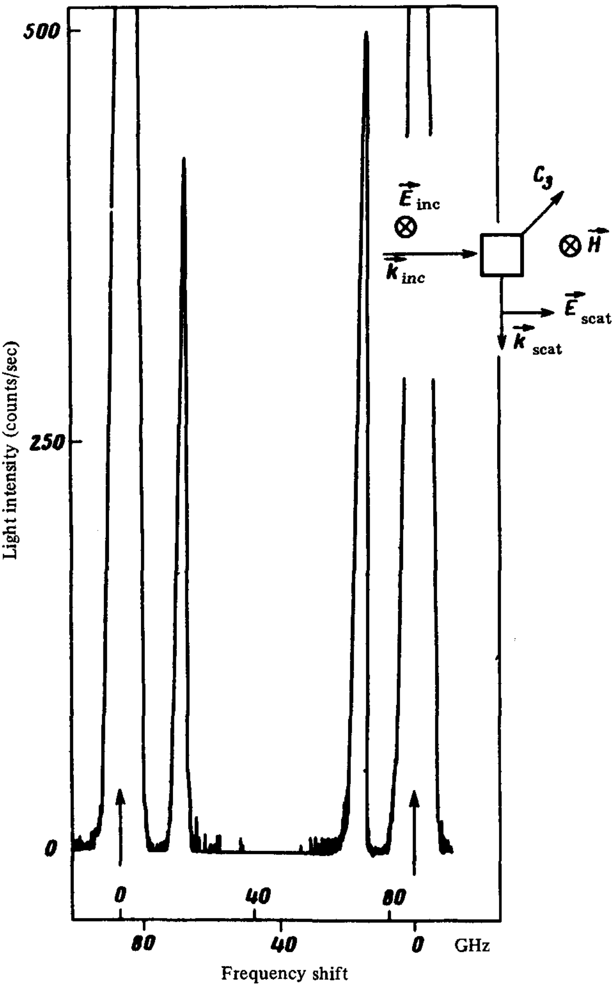

FIG. 2. Spectrum of light scattered in $CoCO_3$ by magnons propagating along z, following application of microwave power ($P \sim 500$ mW) with $\nu = 35.4$ GHz and in a magnetic field $H = 230$ Oe.

III. We turn now to the results obtained at $\mathbf{k} = \mathbf{k}_x$. The geometry of the experiment for this case is shown in Fig. 3.

In this configuration, in weak magnetic fields, when microwave power at $\nu = 35.4$ GHz is applied, a satellite at half the frequency likewise appears in the spectrum of the scattered light. However, in contrast to the preceding cases, it exists in a wide interval of magnetic fields, ~ 500 Oe. Additional absorption of the microwave power is observed in the same field interval (Fig. 3, curve 2). The frequency of the satellite does not depend on the magnetic field (points on Fig. 3). The polarization conditions of the observations also differ from the preceding case, namely, the scattered light contains both the $\mathbf{E}_{\parallel}$ component and the $\mathbf{E}_{\perp}$ component (relative to the incident light).

We are presently able to explain qualitatively the nature of the effect described in Secs. II and III. As to the effect described in Sec. I, the spectra of the thermal magnons and phonons obtained by us[3)] for $CoCO_3$ cannot explain this effect as being due to the simple processes of decay of a

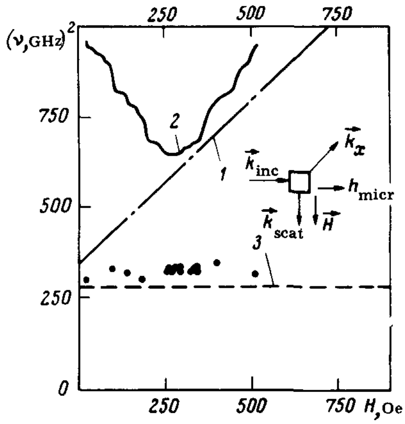

FIG. 3. Dependence of the frequency squared on the magnetic field for thermal and pumped elementary excitations propagating along x, for $CoCO_3$ at $T \lesssim 2$ K. The points correspond to pumped excitation. Curves: 1—spectrum of thermal magnons with $\mathbf{k}=\mathbf{k}_x$, 2—microwave power absorption lines, 3—frequency of transverse phonons in the basal plane at $T \sim 100$ K.

magnon with $\nu = 35.4$ GHz into magnons or phonons at half the frequency. This question calls for further investigation.

The results described in Sec. II can be attributed to parametric excitation of magnons at half the frequency for perpendicular pumping ($\mathbf{H} \perp \mathbf{h}_{micro}$) in a magnetic field whose magnitude is determined by the spectrum of the low-frequency magnons. The microwave photons with energy $\hbar\nu$ produces in this case two magnons with energies $\hbar\nu/2$ and with wave vectors $\mathbf{k}$ and $-\mathbf{k}$. We emphasize that in this experiment both the magnitude and the direction of the observed magnons are fixed. No microwave-power absorption comparable with that observed at resonance was observed by us in this field.

In the third case it follows from the experiment first (Fig. 3) that the frequency of the satellite does not depend on the magnetic field and, second, that the polarization conditions correspond to the conditions for observation of transverse phonons in the basal plane. In addition, the frequencies of these phonons at given $\mathbf{k}$ (determined at $T \sim 100$ K) are close to the frequency of the satellite. It is natural to assume that in this experiment we observe the scattering of light by microwave-pumped phonons or by mixed magnetoelastic excitations. In the absence of microwave pumping, the number of phonons at $T \gtrsim 2$ K is small and is not observed in the experiment.

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[1]The authors thank N.Yu. Ikornikova and V.M. Egorov for supplying the $CoCO_3$ crystals.
[2]The x axis in the basal plane is chosen to be parallel to the external magnetic field.
[3]These spectra will be published by us in a detailed article.

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