Scattering of light by spin waves in antiferromagnetic $CoCO_3$

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We investigate Mandel'shtam–Brillouin scattering of light ($\lambda = 0.63\ \mu$) at 90° in antiferromagnetic $CoCO_3$ ($T_N = 18.1$°K) at $T \sim 2$°K. Scattering by thermal spin waves and by the spin waves produced in the sample when the low-frequency AFMR branch is excited in it are observed. The study of the spectral composition and intensity of the light scattered by the excited spin waves shows that the dominant process of relaxation is elastic two-magnon scattering.

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We have attempted to investigate Mandel'shtam Brillouin scattering of light by spin waves in the crystal $CoCO_3$ at $T \sim 2$ °K.

$CoCO_3$ goes over below $T_N = 18.1$ °K, into an antiferromagnetic state with weak ferromagnetism and with anisotropy of the easy-plane type. [1] The spin-wave spectrum of such a magnetic structure consists of two branches. [2,3] The low-frequency branch, which will be dealt with from now on, is of the form

$$\left(\frac{\nu}{\gamma}\right)^2 = H(H + H_D) + \alpha_z^2 k_z^2 + \alpha_\perp^2 k_\perp^2 , \tag{1}$$

where H is the external magnetic field, H_D is the Dzyaloshinskiĭ field, γ is the gyromagnetic ratio, α_z and $\alpha_\perp$ are exchange constants, while the values of γ and H_D determined from AFMR experiments on $CoCO_3$ are $\gamma = 5.6$ and $H_D = 27$ kOe according to [5] and $\gamma = 4.6$ and $H_D = 51.5$ kOe according to [6]. The absence of a gap in this branch of the spectrum enables us to investigate the low-frequency spectrum of this structure with the aid of Mandel'shtam-Brillouin scattering of light by spin waves with $k \neq 0$. [4]

Investigation of the optical properties of $CoCO_3$ [7] has shown that this crystal becomes optically biaxial in the antiferromagnetic state, and the resultant anisotropic magnetic birefringence is $n_x - n_y = 27 \cdot 10^{-5}$ (or ~ 1500 deg/cm at $\lambda = 0.63\ \mu$. The refractive-index difference is $n_z - n_x = 16 \cdot 10^{-5}$. No noticeable Faraday rotation (less than 100 deg/cm) was observed by us in this compound.

To investigate the scattering of light we used the same installation as in [8], except that the light fed into the Fabry-Perot inteferometer was scattered by the sample at 90° to the incident beam.

The $CoCO_3$ sample was prepared in the form of a rectangular parallelepiped with base $\sim 1 \times 1$ mm and height ~ 1.5 mm. The z (C_3) axis was directed along the diagonal of the parallelepiped base. The geometry of the scattering experiment is illustrated in the insert of Fig. 1. At this geometry, we observed scattering by spin waves with $k_z = 2.5 \times 10^5$ cm^{-1}.

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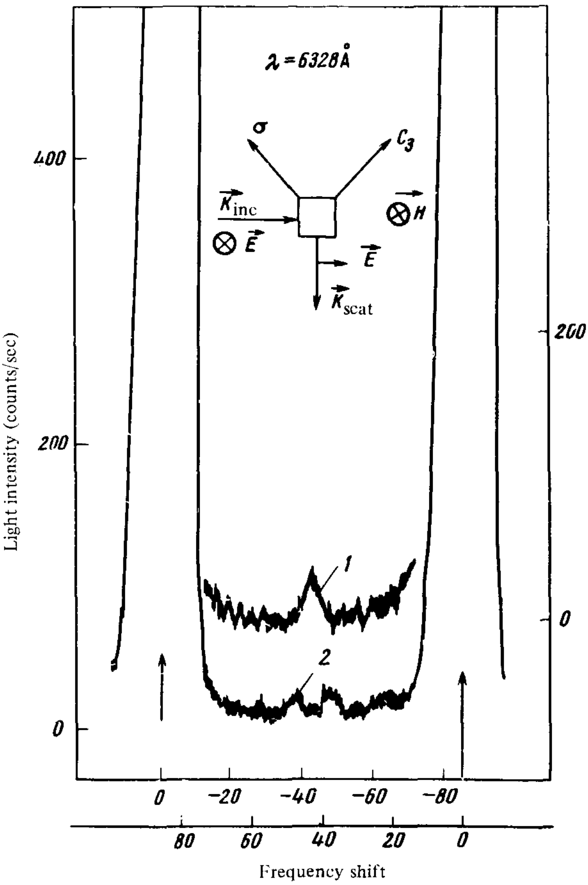

FIG. 1. Spectrum of 90° scattering by thermal magnons for $H = 1408$ Oe (1) and $H = 1014$ Oe (2) in $CoCO_3$ ($T \sim 2$ °K).

All the experiments were performed at temperatures 1.5 – 2 °K in magnetic fields up to 2 kOe. The power of the employed light source (LG-36A laser) was ~ 50 mW.

We investigated in the experiment the scattering of light by thermal spin waves in the absence of microwave power, and by spin waves excited when microwave power was applied to the sample under AFMR conditions.

a) *Thermal spin waves*. A typical spectrum of the light scattered by thermal spin waves is shown in Fig. 1. It is seen that the registered spectrum contains a component shifted by $\pm\Omega$ due to scattering by thermal magnons besides the undisplaced component corresponding to the incident-light frequency. The frequency of the displaced component depends on the applied magnetic field.[1)] Curve 1 corresponds to $H = 1408$ Oe and curve 2 to $H = 1014$ Oe.

The spectrum shown in Fig. 1 was obtained under the following polarization conditions: the light incident on the crystal was polarized along the external

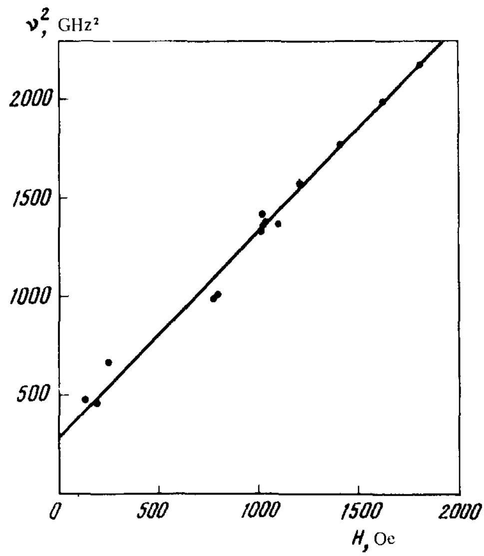

FIG. 2. Dependence of the frequency on the magnetic field for thermal magnons in $CoCO_3$ ($T \sim 2$ °K).

magnetic field, the interferometer received the scattered-light component polarized at 90° to the magnetic field. In the case when the incident and scattered light were polarized along the magnetic field, the signal at the displaced frequencies was completely missing from the registered spectrum. The results obtained for the spectra of scattering by thermal magnons allow us to state that, accurate to ~30%, the intensities of the Stokes and anti-Stokes components are equal.

Figure 2 shows the dependence of the square of the magnon frequency on the external magnetic field in the interval 0 – 2 kOe, deduced from the measured spectra. As seen from this figure, this dependence is sufficiently well approximated by a straight line. According to formula (1), the slope of this line yields for $\gamma^2 H_D$ (under the condition $H \ll H_D$) a value 1050 GHz²/kOe. This value is 5% smaller than that obtained from the data of [6], and exceeds by 20% the value obtained from the data of [5]. Extrapolating this straight line to $H = 0$ we obtain the value $\nu^2 = a_z^2 k_z^2$, from which we get $a_z = 1.5 \times 10^{-5}$ kOe-cm (or 6.9×10^{-5} GHz-cm). The value of α_{av} determined from measurements of the heat capacity of $CoCO_3$ [10] is 0.68×10^{-5} kOe-cm.

b) *Excited spin waves*. We have demonstrated experimentally in [8,9] that excitation of antiferromagnetic resonance in a $CoCO_3$ sample produces in it both spin waves with $\mathbf{k} = 0$ and with $\mathbf{k} \neq 0$. The number of the latter is $\sim 10^4$ times larger than that of the former. To ascertain into which of the spin waves with $\mathbf{k} \neq 0$ the homogeneous precession decays under AFMR conditions, we have investigated the spectral composition of the light scattered at 90° (i.e., $k_z = 2.5 \times 10^5$) when the low-frequency AFMR branch is excited in it. In the present experiment we satisfied the same polarization conditions as in the investigation of the thermal magnons. Figure 3 shows a typical spectrum of the scattered light when microwave power (~5 mW) is applied in a field $H = 1014$ Oe. From a comparison of Figs. 1 and 3 we see that when AFMR is excited the number of the spin waves with given k increases by at least 20 times. Such an in-

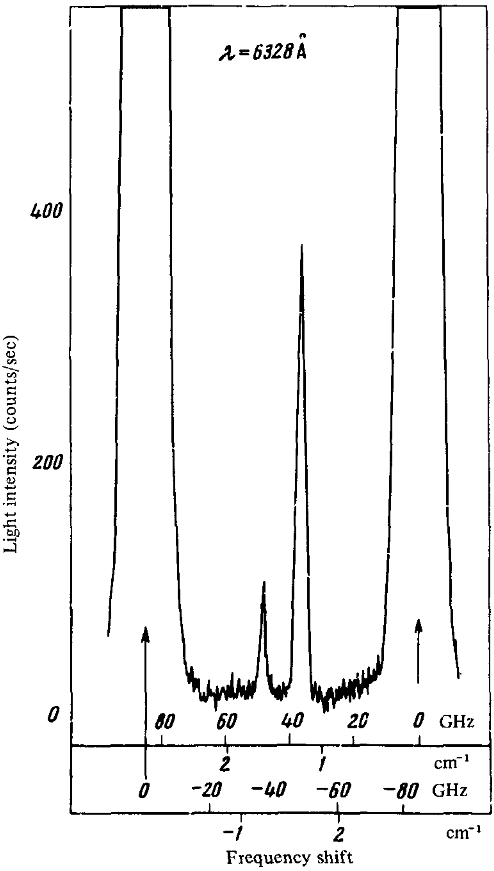

FIG. 3. Spectrum of scattering at 90° by magnons excited in AFMR in $CoCO_3$ ($T \sim 2$ °K).

crease of the magnon peaks is observed only in a narrow interval of magnetic fields, $H = 1014 \pm 25$ Oe, although absorption of microwave power in the sample investigated in the same experiment was observed in the range of fields from $H = 620$ to 1200 Oe. The large width of the resonance line is connected with excitation of various magnetostatic modes in the sample and is in qualitative agreement with the calculations of Bar'yakhtar *et al.* for easy-plane antiferromagnets.[11]

The frequency of the AFMR-enhanced magnons was always equal to the microwave frequency $\nu = 36$ GHz. This result indicates that the dominant relaxation process of the homogeneous precession is a two-magnon process in which the spin-wave energy is rigorously conserved, but the wave vector is not (see, e.g., [12]). Attention is called to the strong difference, in Fig. 3, between the intensities of the Stokes and anti-Stokes components of the light scattered by the magnons. The intensity of the anti-Stokes component always turned out to be larger than that of the Stokes component, even though the ratio of their values varied when the sample was reset. This fact can be interpreted as the existence of a directed flux of magnons with $k_z = 2.5 \times 10^5$ through the region from which the scattering is observed.

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[1)]All the magnetic field cited in this article take into account the demagnetizing factor of the sample.

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