SPLITTING OF ANTIFERROMAGNETIC RESONANCE SPECTRUM IN $CoCO_3$

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In an investigation of the low-frequency branch of the spectrum of antiferromagnetic resonance (AFMR) in $CoCO_3$ in the frequency range 17 - 55 GHz, we observed a strong splitting of the AFMR spectrum. Cobalt carbonate below $T_N = 18.1°K$ is a rhombohedral antiferromagnet with weak ferromagnetism, which arises as a result of the tilting of the spins of the Co^+ magnetic ions, lying in the basal plane.

Single crystals of cobalt carbonate[1)] were prepared in the form of discs and spheres of

[1)] The authors are sincerely grateful to N. Yu. Ikornikova for supplying the $CoCO_3$ single crystals.

0.3 - 9.8 mm diameter. The plane of the disc always coincided with the basal plane of the crystal.

It was observed in [1], in an investigation of the low-frequency branch of AFMR in $CoCO_3$ in the frequency region 50 - 185 GHz and at T = 4.2°K, that the frequency dependence is given by the expression

$$f^2 = 1120\ \mathrm{GHz}^2/\mathrm{kOe}\ H + 21.7\ \mathrm{GHz}^2\mathrm{kOe}^2\ H^2.$$

Figure 1 shows a plot (curve 1) of this formula. Our results for sample 1 at 4.2°K are marked by circles.

These results were obtained in two runs. In the first (open circles) the sample was placed at the end of a short-circuited waveguide for the 8-mm band and the amplified signal corresponding to the change of the intensity of the reflected microwave power was recorded with an xy plotter. In these measurements, the high-frequency component of the magnetic field was parallel to the basal plane. In the second run (dark circles) the sample was placed in the center of a tunable resonator and the microwave component of the magnetic field was parallel to the C_3 axis. It is seen from Fig. 1 that the results of the two experiments are in good agreement. At frequencies higher than 46 GHz, a nonmonotonic dependence of the resonant field on the frequency was observed. It is also seen from the figure that our points approach curve 1 far from the 46 GHz frequency. In the vicinity of 46 GHz, however, a splitting of the AFMR spectrum is observed.

Sample 1 had the form of a cylinder and had the smallest line width of all the samples investigated by us (H ≈ 30 Oe). Control measurements were made on two other samples, in the form of a disc and a sphere, the minimum line widths of which were 100 and 65 Oe, respectively. They also revealed the same type of splitting near f = 46 GHz, but the magnitude of the splitting was 1.5 - 2 times larger than in the first sample. The data for the sphere are shown by the triangles in Fig. 1.

We investigated the angular dependence of the resonant field by placing the sample on the narrow wall of the waveguide at a distance 1.5 mm from the short-circuited end. We observed that the relation $H_\Theta = H_0/\sin\Theta$, where Θ is the angle between the direction of the external magnetic field and the C_3 axis and H_0 is the resonant field at $\Theta = 90°$, taken from curve 1, is satisfied far from the frequency at which the splitting takes place.

The angular dependence near the splitting frequency was more complicated. The results are shown in Fig. 2, where the abscissas represent the projection of the field on the basal plane ($H_\Theta \sin\Theta$). The frequency at which the splitting takes place apparently decreases with decreasing Θ, i.e., with increasing H_Θ. At present we still have no data for higher frequencies to be able to describe this dependence.

It was observed that the magnitude of the splitting depends strongly on the temperature. When the temperature is lowered to 1.1°K (Fig. 3), the splitting increases and vanishes at T = 10°K. Figure 4 shows the temperature dependence of the relative resonant field H/H_0. We see that the splitting decreases sharply between 5.5°K and 7.0°K. An increase of the resonant field, of equal magnitude for both frequencies, occurs above 8.5°K and is due to the

decrease of H_D.

We investigated the AFMR dependence for $MnCO_3$, which has the same crystallographic and magnetic structures as $CaCO_3$, in the frequency region from 24 to 52 GHz. No splitting was observed.

It is seen from the results that the elementary-excitation spectrum of the investigated $CoCO_3$ crystals has an additional branch (of frequency f = 46 GHz) at small values of the wave vector k), which interacts strongly with the spin-system oscillations. We are unable at present to propose a satisfactory explanation of the observed effects.

We note that this branch cannot be the high-frequency branch of the AFMR, which is shown by specific-heat measurements to lie much higher [3]. One might assume that the 46 GHz frequency corresponds to the oscillation frequency of the magnetic moments of some impurities (say Mn^{++}) in the effective field produced by the antiferromagnetic sublattices. However, our results on the temperature dependence do not agree fully with such an assumption.

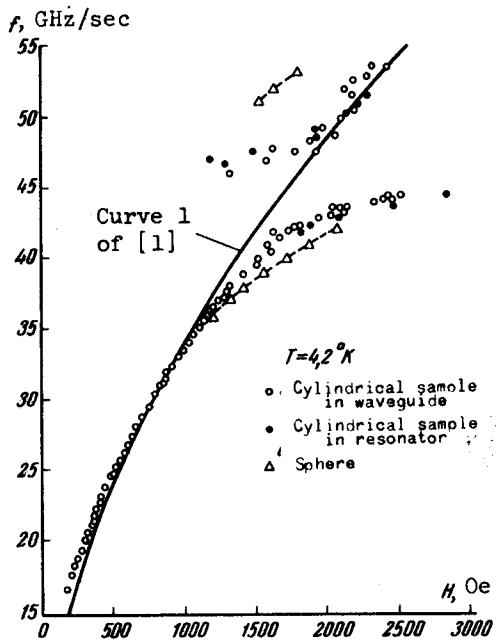

Fig. 1

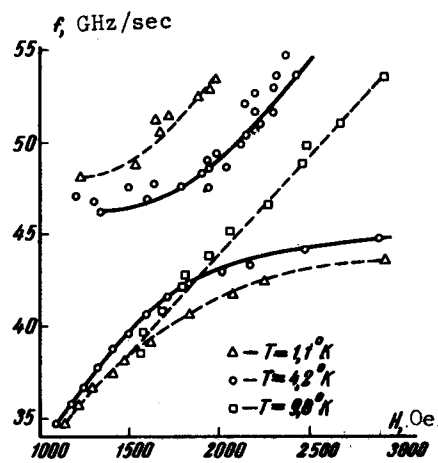

Fig. 3

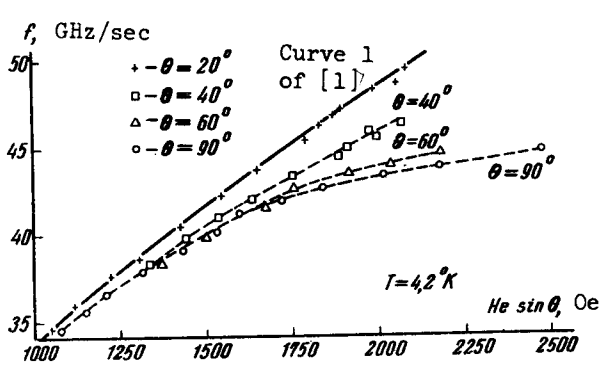

Fig. 2

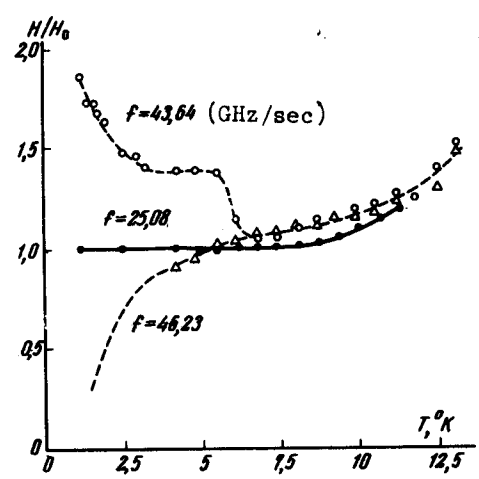

Fig. 4

The spin oscillations could change into acoustic phonons of definite frequency (∿46 GHz), if the crystal were to contain regions, with characteristic dimension ∿1000 Å, in which the antiferromagnetic structure is highly distorted. Such an assumption, however, has at present neither theoretical nor experimental confirmation.

In conclusion, we are deeply grateful to P. L. Kapitza for constant interest in the work, V. A. Tulin for useful discussions, and M. E. Mysin for help in preparing the experiments.

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[2] E. A. Turov and N. G. Guseinov, ibid. 38, 1326 (1960) [11, 955 (1960)].
[3] I. N. Kalinkina, ibid. 43, 2028 (1962) [16, 1422 (1963)].