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INVESTIGATION OF ANTIFERROMAGNETIC RESONANCE IN COBALT CARBONATE OVER A BROAD FREQUENCY RANGE

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The dependence of the antiferromagnetic resonance frequency on magnetic field is studied in $CoCO_3$ at T = 4.2°K in the frequency range from 50 to 180 GHz. The results obtained do not agree with the existing phenomenological theory of resonance.

1. INTRODUCTION

$CoCO_3$ is a rhombohedral antiferromagnetic material with weak ferromagnetism. In substances isomorphic to it, α-Fe_2O_3 and $MnCO_3$, antiferromagnetic resonance (AFMR) has been studied in detail, and good agreement has been obtained between experiment and theory. $CoCO_3$ differs from the antiferromagnets mentioned by a strong anisotropy of the g-factor. Resonance in $CoCO_3$ was observed by Rudashevskiĭ[1] at frequencies up to 36 GHz. There are not enough of these data, however, to clear up the whole spectral picture. In the present research, AFMR in $CoCO_3$ has been observed in the frequency range from 50 to 180 GHz. This range was covered continuously.

2. MEASUREMENT METHOD AND SPECIMENS

The investigation of AFMR in $CoCO_3$ was made on a millimeter-range radiospectrometer, a block diagram of which is shown in Fig. 1.

The sources of microwave power were a backward-wave tube OV-612 (120 to 180 GHz)[2] and an orotron (50 to 125 GHz). The orotron is a new type of generator in the millimeter range, developed by Rusin and Bogomolov[3]. Its broad tuning range, high frequency stability, and large power (5 watts in the pulse) make it convenient for application in magnetic radiospectrometers. The frequency of the microwave signal was measured in the range 50 to 80 GHz with standard wavemeters (accuracy 0.02%), and in the range 80 to 180 GHz by means of open resonators with micrometer reading of the distance between mirrors (accuracy 0.25 to 1%).

The magnetic field was produced with a superconducting solenoid, made from wires of 65-niobium-titanium, with maximum field value 50 kOe. The magnetic field was calibrated on the basis of paramagnetic resonance in $MnCl_2 \cdot 4H_2O$, for which g = 2.00[4]. The accuracy of the field measurement was basically determined by the errors of the recording potentiometer and was ±100 Oe over the whole range.

The $CoCO_3$ specimens were placed in a shorted section of a copper wave guide, with cross section 3.6 × 1.8; this made it possible to make measurements over a wide frequency interval with a single mounting of the specimen being studied. The reflected signal was registered by a silicon detector and, after amplification, was fed to the Y-coordinate of a recording potentiometer PDS-021. To the X-coordinate of the poteniometer was fed a voltage proportional to the current in the superconducting solenoid. A record of the resonance-absorption line was made at fixed frequencies by smooth variation of the magnetic field.

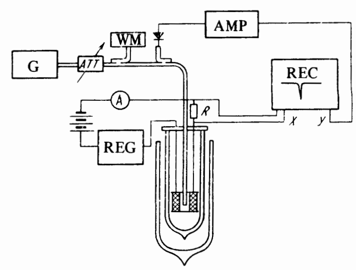

FIG. 1. Block diagram of the millimeter-range spectrometer: G, generator; WM, wavemeter; AMP, amplifier; REC, recorder; REG, regulator for the current in the superconducting solenoid.

The $CoCO_3$ crystals were obtained by the hydrothermal method, in the Institute of Crystallography of the USSR Academy of Sciences by N. Yu. Ikornikova[1)][5]. They are thin plates of violet color, whose plane coincides with the basal plane. From these crystals cylindrical and spherical specimens, of diameter 0.3 to 0.2 mm, were made by a mechanical method.

3. RESULTS AND DISCUSSION

Figure 2 shows the results of the investigation of the dependence of the resonance absorption frequency ν for $CoCO_3$ on the applied static magnetic field H. The specimens were always so placed that the field H lay in the basal plane of the crystal. The measurements were made at temperature 4.2°K.

The width ΔH of the resonance line, for the specimens investigated, varied from 200 to 300 Oe (at H = 12 kOe). The method of attachment and the shape of the specimens had no effect on the position of the resonance line, within the limits of accuracy of our experiment. Likewise, no anisotropy of the value of the resonance field in the basal plane was noticed.

The results of the experiment were analyzed by the method of least squares by means of an M-20 elec-

1)The authors thank N. Yu. Ikornikova for providing the $CoCO_3$ monocrystals.

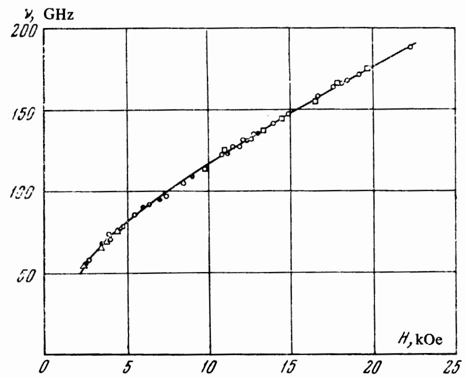

FIG. 2. Dependence of antiferromagnetic resonance frequency for $CoCO_3$ on size of the static magnetic field, at T = 4.2°K.

tronic computer[2)]. It was found that the experimental data were well approximated by the parabola

$$\nu^2 = a_1 H + a_2 H^2 \quad (1)$$

with the following coefficients:

$$a_1 = 1120 \pm 35\ \mathrm{GHz^2/kOe}, \quad a_2 = 21.7 \pm 2.76\ \mathrm{GHz^2/kOe^2}.$$

Rudashevskiĭ[1] found other values of the coefficients a_1 and a_2. Within the limits of the indicated error, however, the experimental points from his work agree with formula (1).

The theoretical dependence of the AFMR frequency on magnetic field for rhombohedral crystals (group D_{3d}^6), in the case of an isotropic g-factor, was derived by Borovik-Romanov[6] and by Turov[7]. For the low-frequency branch of the AFMR spectrum, which was investigated in our work, this dependence has the form

$$(\nu/\gamma)^2 = H(H + H_D), \quad (2)$$

where γ is the gyromagnetic ratio and H_D is the Dzyaloshinskiĭ field, which can be determined independently from static measurements. Turov and Guseĭnov[8] showed that allowance for anisotropy in the basal plane leads to an additional term of the form $H_\Delta^2 \cos 6\varphi$ in formula (2). Borovik-Romanov and Meshcheryakov[9] detected anisotropy of the resonance in $CoCO_3$ at low frequencies. The amplitude of the hexagonal anisotropy of the resonance field in the region of the spectrum investigated by us, on the basis of the data of paper[9], is $\delta H < 50$ Oe. This quantity is less than the accuracy of our measurements.

As has already been said, the Co^{++} ion is distinguished by a large anisotropy of the g-factor. A systematic theory of AFMR, taking into account that the magnetic properties are determined not only by the spin but also by the orbital moment of the electron, does not exist at present. Turov[10], however, carried out a calculation of the AFMR spectrum, with allowance for anisotropy of the g-factor, within the framework of a phenomenological model. He considered in particular antiferromagnets of tetragonal structure (of the type of NiF_2) and of rhombohedral structure (of the type of $CoCO_3$). In the case of NiF_2, allowance for anisotropy of the g-tensor significantly changes the form of the AFMR spectrum, and the results of the calculation agree well with the experimental data of Richards[11]. In the case of rhombohedral crystals, allowance for the anisotropy of the g-factor does not change the form of formula (2).

By comparing the experimental dependence (1) with the theoretical formula (2), one can calculate the g-factor and the Dzyaloshinskiĭ field:

$$g_\perp = 3.3 \pm 0.2, \quad H_D = 51.5 \pm 8\ \mathrm{kOe}.$$

In observation of paramagnetic resonance on Co^{++} ions in the rhombohedral crystals $CdCO_3$[12] and $CaCO_3$[13], a value of g between 4.8 and 5.0 was observed. The discrepancy between the values of the g-factors in EPR and in AFMR does not contradict the phenomenological theory of AFMR, since in the latter the values of the g-factors are arbitrary.

The value obtained for the Dzyaloshinskiĭ field H_D, however, is almost twice as large as that calculated from static measurements, $H_D = 27 \pm 1.5$ kOe[14]. Such a large discrepancy lies clearly outside the limits of error of the experiment and indicates that the phenomenological theory of AFMR is apparently not in a state to explain the resonance picture in such strongly anisotropic crystals as $CoCO_3$.

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[2)]The calculations on the M-20 machine were made by R. A. Latypova. The authors extend to her their cordial thanks.

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