HEXAGONAL ANISOTROPY OF THE POSITION OF THE ANTIFERROMAGNETIC RESONANCE LINE IN $CoCO_3$

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Hexagonal anisotropy is observed in the position of the antiferromagnetic resonance line on rotation of the external magnetic field in a plane perpendicular to the third-order axis of a $CoCO_3$ crystal. The magnitude of the observed anisotropy amplitude is smaller by almost an order of magnitude than that which was to be expected on the basis of results obtained from static measurements[5,2]. Possible reasons for the observed discrepancy are discussed.

1. INTRODUCTION

THE crystallographic structure of the rhombohedral antiferromagnet $CoCO_3$ belongs to the space group D_{3d}^6; and the Hamiltonian, as was shown by Dzyaloshinskiĭ[1], has the form, through terms of the sixth order,

$$\mathcal{H} = {}^1/_2Bm^2 + {}^1/_2bm_z^2 + {}^1/_2al^2\cos^2\theta + ql\sin\theta(m_y\cos\varphi - m_x\sin\varphi) + dl^4\sin^3\theta\cos\theta\sin 3\varphi + el^6\sin^6\theta\cos 6\varphi - \mathbf{mh}.$$

The z axis is directed along the ternary axis of the crystal; θ is the angle between the z axis and the direction of l; φ is the angle between the direction of easy magnetization and the direction of the external field H ($\mathbf{m} \parallel \mathbf{H}$); and

$$\mathbf{m} = \frac{\mathbf{M}_1 + \mathbf{M}_2}{2M_0}, \quad \mathbf{l} = \frac{\mathbf{M}_1 - \mathbf{M}_2}{2M_0}, \quad \mathbf{h} = 2M_0\mathbf{H},$$

where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the sublattice magnetizations, with $\mathbf{M}_1^2 = \mathbf{M}_2^2 = M_0^2$.

The crystallographic anisotropy energy in the basal plane is defined by the expression $e' \cos 6\varphi$, where $e' = e + d^2/4a$. Kaczér and Kreĭnes[2,3], by means of a torsion magnetometer, observed hexagonal anisotropy in the rhombohedral antiferromagnet $CoCO_3$ and determined the value of $K_3 = 2e' = 656$ erg/cm^3. This hexagonal anisotropy constant, as was shown by Turov and Guseĭnov[4], should enter into the expression for the antiferromagnetic resonance spectrum. For the low-frequency branch of the spectrum, this expression has the form

$$4M_0^2(\omega/\gamma)^2 = h(q + h) + 36e'B\cos 6\varphi.$$

On substituting the values of $B/4M_0^2$, $q/2M_0$, and e' from[5,2], we find that on rotation of the external magnetic field in the basal plane, the resonance field for $CoCO_3$ should change with an amplitude of 350 Oe for fields of order 1 kOe.

The frequency dependence of the antiferromagnetic resonance spectrum in $CoCO_3$ in the frequency interval 10 to 34 GHz was studied by Rudashevskiĭ[6]. No sixty-degree anisotropy was detected. P. Maksimenko (thesis, Moscow Chemico-technological Institute) observed in several specimens, at frequency 136 GHz, an irregular change of the position of the resonance line in the basal plane, with amplitude ~ 1.5 kOe. The availability of more perfect monocrystals of the right form has made it possible to obtain a line width $\Delta H = 30$ Oe for f = 34 GHz (whereas in[6] $\Delta H = 400$ Oe at the same frequency) and to detect the hexagonal anisotropy satisfactorily.

2. SPECIMENS AND METHOD OF MEASUREMENT

The monocrystals of cobalt carbonate[1)] had the form of plates of irregular form, with transverse dimension 0.5 to 1.5 mm and thickness 0.1 to 1 mm. The plane of the plates always coincided with the basal plane, as was checked by x-ray studies[2)]. The measurements were carried out in a straight-amplification spectrometer at frequencies f equal to 21.94 and 33.87 GHz, at T = 4.2° K. The specimens, fastened on the end of a quartz stick, were placed in the middle of a resonator, in which H_{011} oscillations were excited. The external magnetic field always lay in the basal plane and was directed perpendicularly to the microwave field.

In order to eliminate the effect of the demagnetizing factor when the external field was rotated in the basal plane, specimens were prepared in the form of disks. The crystals were fastened at their surface, with shellac, to the end of a brass rod, of diameter 0.4 to 0.8 mm, which was clamped in the chuck of a lathe. An abrasive stone, supported by hand, was brought into contact with the lateral surface of the plate. By changing the position at which the brass rod was clamped in the chuck, it was possible to regulate the pressure exerted by the abrasive on the specimen. This made it possible to obtain circular disks without shearing. It was found that gluing affected the position and width of the line, and therefore in the investigation of hexagonal anisotropy all specimens were fastened by the method shown in Fig. 1.

3. RESULTS

The initial measurements of resonance line position were made on unmachined specimens. On rotation of the magnetic field in the basal plane, large irregular changes of position of the resonance line were observed,

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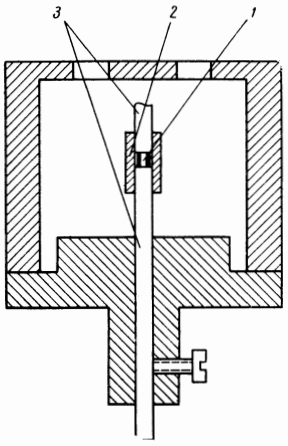

FIG. 1. Fastening of the specimen in the resonator: 1, specimen; 2, paper cup; 3, quartz sticks.

different for different specimens. A typical curve for one of them is shown in Fig. 2, a. After the specimens were made into the form of a disk, the change of position of the line, as is shown in Fig, 2, b, had a regular character for all specimens; that is, clear sixty-degree anisotropy was observed. An unmachined specimen had a broad and complex line, which became smoother and narrower after machining. When the specimen lay freely on its support, and when its orientation corresponded to a direction of easy magnetization, the value of the resonance field corresponded to a minimum in Fig. 2, b. X-ray investigations showed that the direction of easy magnetization lies along a second-order axis; this agrees with the results of[3].

It has been mentioned that gluing of the specimen affects the shape, width, and position of the line. For all glued specimens, the line had the right shape and a minimal width when the external magnetic field H was parallel to the direction of easy magnetization, and it became wider by a factor two or three when H was parallel to a direction of hard magnetization; on the sixty-degree anisotropy there were superposed irregularities of the kind shown in Fig. 2, b. For this reason, in the measurement of the hexagonal anisotropy all specimens were fastened by the method shown in Fig. 1. After this, the linewidth remained practically constant on change of orientation of the external field and corresponded to the minimal value for a glued specimen. Figure 3 shows the observed dependence of the value of the resonance field on the orientation of the magnet for an unglued cylindrical specimen with line-width ΔH = 30 Oe.

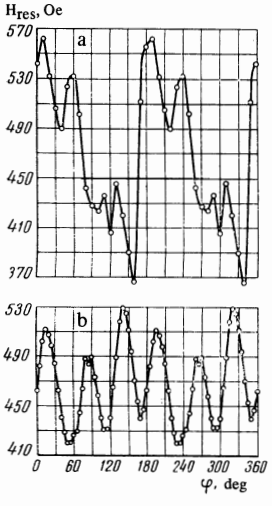

FIG. 2. Anisotropy of resonance-line position on rotation of the field in the basal plane for the same specimen: a, unmachined; b, cylindrical; f = 21.940 GHz.

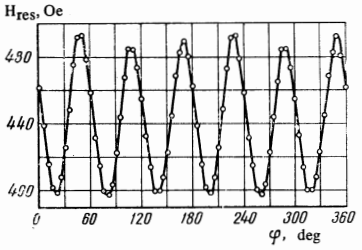

FIG. 3. Hexagonal anisotropy of resonance-line position for a specimen with line-width ΔH = 25 to 32 Oe; f = 21.940 GHz.

Measurements of the magnitude of the hexagonal anisotropy were carried out on a large number of specimens. The linewidth of these specimens fluctuated between 30 and 150 Oe. It was established that the anisotropy amplitude $H_A = \frac{1}{2}(H_{max} - H_{min})$ and the field $H_0 = \frac{1}{2}(H_{max} + H_{min})$ change markedly from specimen to specimen. Figure 4 presents the results obtained; from them it is clear that the anisotropy amplitude varied from 40 to 80 Oe and the resonance field from 400 to 500 Oe for f = 21.94 GHz. A definite correlation is observed between the value of the field H_0 and the anisotropy amplitude H_A, whereas there was no correlation with line-width. A similar picture was observed in measurements at frequency f = 33.87 GHz, where the resonance field was of order 1 kOe. Measurements made at a frequency at which the resonance field was greater than 2 kOe showed that the anisotropy remains also in fields in which the specimen is known to be saturated.

One of the cylindrical specimens, of thickness 0.4 mm, with H_0 = 420 Oe and H_A = 50 Oe, was ground to thickness 0.04 mm. After this the following results were obtained: H_0 = 480 Oe, H_A = 76 Oe.

4. DISCUSSION OF RESULTS

A. Effect of shape. Since our measurements indicated a strong dependence of the resonance-line position on the shape of the specimen, we repeated the calculation of Turov and Guseĭnov[4] on the dependence of antiferromagnetic resonance frequency on field with allowance for the energy of the demagnetizing field. On introducing into the Hamiltonian a term determining the energy of the demagnetizing field, in the form

$$\frac{1}{2}N_i(M_{1i}^2 + M_{2i}^2) = \frac{1}{2}n_i(m_i^2 + l_i^2),$$

(where $N_i = 2M_0^2 N_i$, and N_i is a demagnetizing factor

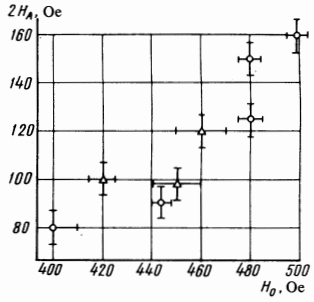

FIG. 4. Resonance field $H_0 = \frac{1}{2}(H_{max} + H_{min})$ and doubled anisotropy amplitude $2H_A = H_{max} - H_{min}$ for different cylindrical specimens; Δ, specimens with the same ratio of height to diameter.

taking values from 0 to 4π), we get for an external field $H \parallel x$:

$$4M_0^2\left(\frac{\omega}{\gamma}\right)^2 = \left(1+\frac{b+\Delta n_{zx}}{B+n_x}\right)(h+\Delta n_{yx}m_{0x})(h+q) - (B+b+\Delta n_{zy})\Delta n_{yx} + 36e'(B+b+\Delta n_{zy}), \quad (2)$$

where $\Delta n_{ik} = n_i - n_k$. An estimate shows that the change of resonance-line position δH is determined by the second term in the right side of Eq. (2), and in our case it gives the value $\delta H = 10^3\delta(\Delta N_{yx})$ [Oe]. This estimate agrees with the variations of resonance field observed in an unmachined specimen.

From (2) it is clear that a difference in the value of H_0 cannot be explained by different values of N_i. In our measurements there is also observed no correlation between N_i and H_0.

B. Anisotropy. We shall compare the data obtained on anisotropy amplitude H_A with the results of[5,2]. From Eq. (2), on setting $\Delta n_{yx} = 0$ and taking into account that n_x, Δn_{zx}, $\Delta n_{zy} \ll b$, we get in the usual notation:

$$\delta H = H_A\frac{36e'(B+b)/4M_0^2}{2H+H_D}. \quad (3)$$

On substituting in (3) the values $e' = 328$ erg/cm^3 from[2] and $\chi_{\parallel} = 4M_0^2/(B+b) = 1.21\times10^{-3}$, $q/2M_0 = H_D = 27$ kOe from[5], we get $H_A = 350$ Oe. The observed amount of anisotropy, $H_A = 40$ to 80 Oe, is much smaller than 350 Oe. At present we see no explanation either of this discrepancy or of the large change of H_A from specimen to specimen.

C. Resonance-line position. We shall discuss the possible reasons for the nonuniqueness of the value of H_0. One of these may be a difference in H_D for different specimens. As was shown by Morrish and Searle[7], addition of 0.38 mole of Ti^{+4} ions decreases H_D in α-Fe_2O_3 from 22 Oe to 15.6 Oe. Since the Co^{++} ion is characterized by a much smaller spin-lattice relaxation time, which indicates strong coupling with the lattice, the sensitivity of H_D to impurities may be significantly larger in $CoCO_3$. On expressing the field H_D in explicit form in (3) and substituting in the formula

$$(\omega/\gamma)^2 = H(H+H_D) + H_\Delta^2\cos 6\varphi, \quad (4)$$

we get for $\varphi = \pi/12$:

$$H_A \approx H_\Delta^2\left(\frac{\omega}{\gamma}\right)^{-2}H_0.$$

This expression, however, does not agree with the result presented in Fig. 4, where the dependence between H_0 and H_A has the form

$$H_A = 0{,}4(H_0 - 300)\ \text{[Oe]}.$$

Presence of magnetostatic modes may also be a reason for the observed discrepancy. Nonuniformity of the static magnetic field at the surface of the cylinder may lead to the occurrence of boundary conditions under which a nonuniform type of precession can arise. All the observed values of H_0 lie in the region of the spectrum that was calculated for plates by Bar'yakhtar, Savchenko, and Tarasenko[8] with allowance for the presence of magnetostatic modes.

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