

ANTIFERROMAGNETISM OF  $\text{CoCO}_3$ 

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THE magnetic susceptibility of cobalt carbonate, which is isomorphous with the  $\text{MnCO}_3$  and  $\text{FeCO}_3$  crystals investigated in earlier work,<sup>1</sup> exhibits typically antiferromagnetic behavior from room temperature to 40°K, according to Bizette.<sup>2</sup> The decrease of the reciprocal susceptibility below this temperature range is especially rapid from 20° to 14°K, like that of a ferromagnet. Borovik-Romanov and Orlova<sup>3</sup> later obtained more detailed measurements of the susceptibility of powdered  $\text{CoCO}_3$ , finding less ferromagnetism than one would expect from ferromagnetic saturation. By analogy with the ideas of Matarrese and Stout<sup>4</sup> regarding the ferromagnetism of  $\text{NiF}_2$ , it was suggested that the observed weak ferromagnetism can result if the antiferromagnetic sublattices are not exactly antiparallel. According to Borovik-Romanov and Orlov, the ferromagnetism of  $\text{CoCO}_3$  is relatively large (~3% of the saturation moment) and can be detected by means of neutron diffraction.

We first began to study  $\text{CoCO}_3$  in 1958, when, since other sources were unavailable, we used the native  $\text{CoCO}_3$  mineral spherocobaltite from the vicinity of Riegelsdorf, Germany.\* However, the observed neutron-diffraction pattern could not be interpreted on the basis of the known  $\text{CoCO}_3$  parameters,<sup>5</sup> since neither the Bragg angles nor the reflection intensities agreed with the calculated values. Cooling to 4.2°K produced no change in

the pattern. It was evident that the spherocobaltite specimen did not possess the sought magnetic properties.

We have now investigated a finely crystalline hydrothermal specimen† of  $\text{CoCO}_3$ , the neutron-diffraction pattern of which at 4.2°K is shown in Fig. 1. The small-angle (111) and (100) reflections are of magnetic origin; they are absent at 20.4°K. Unlike the case of  $\text{MnCO}_3$ , the relative intensities  $I$  of these peaks show that magnetic scattering contributes mainly to the (100) reflection, whose intensity is more than twice that of the (111) reflection. This indicates that the magnetic moments of  $\text{Co}^{++}$  ions, unlike those of  $\text{Mn}^{++}$  ions in the analogous situation, depart from the (111) planes in such a way that the sum of the projections of moments on (100) planes considerably exceeds that on (111) planes. Calculation of the direction of antiferromagnetism relative to the rhombohedral axis, using the magnetic intensity ratio, indicates an angle of  $46 \pm 4^\circ$  between the moments and the [111] axis. Erickson's value for the form factor of  $\text{Co}^{++}$  was used.

In addition to the (111) and (100) magnetic reflections, as the temperature was reduced from 20.4° to 4.2°K some intensity increase was observed in the case of (110) and (211) reflections. In a number of successive runs the latter oscillated from 2 to 6.5% of the (211) nuclear intensity. The small magnetic contribution to reflections with even sums of indices for the given structure results from weak ferromagnetism. Dzyaloshin-skiĭ's thermodynamic theory<sup>7</sup> proposes three possible magnetic states of Mn, Fe, and Co carbonates at temperatures below  $T_N$ . For the present case the most suitable of these is state II, with the following conditions for the minimum of the thermodynamic potential:

$$\beta \approx \pi/2 - d/a, \quad \varphi = \pi/2, \quad m_x = q/B, \quad m_y = m_z = 0,$$

where  $\beta$  is the angle between the spins and (111),

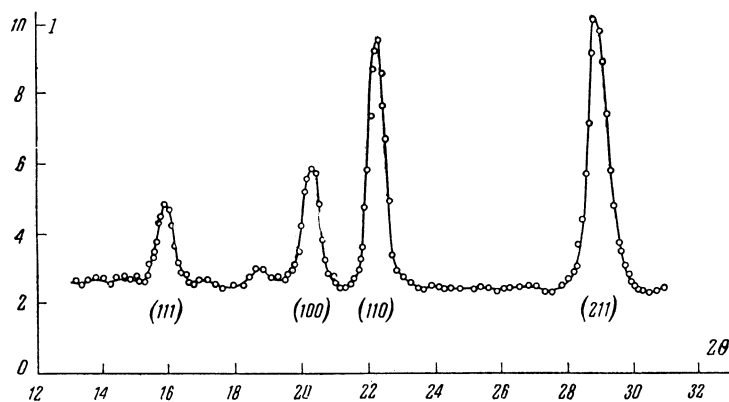


FIG. 1

$\varphi$  is the angle between the spin projections on (111) and the twofold symmetry axis,  $\mathbf{m}$  is the ferromagnetic moment, and  $d$ ,  $a$ ,  $g$ ,  $B$  are expansion coefficients [see Eq. (3a) in reference 7]. Figure 2 gives the experimentally observed arrangement of magnetic moments in  $\text{CoCO}_3$ , corresponding to state II. It should be noted that the angle  $\beta$  is very close to the direction of the shortest distance from the central ion to its nearest neighbor. This picture differs somewhat from the theoretical prediction for state II, since the coefficient ratio  $d/a$  is of the order of unity, whereas the theory predicts the very small ratio  $\sim v^2/c^2$ .

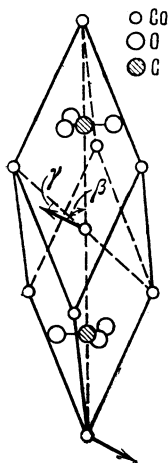


FIG. 2

In connection with this additional observed type of antiferromagnetic structure for isomorphous carbonates of the iron group, it should be remembered that in  $\text{FeCO}_3$  magnetic moments are directed along the [111] axis, while in  $\text{MnCO}_3$  they lie in the (111) plane.

The weak ferromagnetism of  $\text{CoCO}_3$ , which is indicated by the small magnetic contribution to the (211) reflections, shows that the magnetic moments of the ions form a small angle  $\gamma$  with the plane of symmetry, thus producing a total ferromagnetic moment along the twofold axis (Fig. 2). The ratio between ferromagnetic and antiferromagnetic contributions to the (211) and (100) reflections indicates  $15 \pm 5^\circ$  as the magnitude of  $\gamma$ . The existence of the ferromagnetic moment (8.6%) in the (111) plane was recently observed by Borovik-Romanov and Ozhogin,<sup>8</sup> who investigated the weak ferromagnetism of  $\text{CoCO}_3$  in crystals obtained from the same source as ours.† They calculated  $\gamma = 7^\circ$ , from their absolute data for the ferromagnetic moment and the calculated saturation moment of the ion. Aside from experimental errors, uncertainty regarding the

saturation moment of  $\text{Co}^{++}$  is the most likely source of a discrepancy regarding  $\gamma$ .

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†This specimen was prepared by I. Yu. Ikornikova at the Institute of Crystallography, Academy of Sciences, U.S.S.R.

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## DIPOLE MOMENT OF UNSTABLE ELEMENTARY PARTICLES

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PARITY nonconservation makes it possible for an elementary particle with spin to have a dipole moment.<sup>1</sup> Landau's theory of combined inversion<sup>2</sup> leads to time-reversal invariance, from which Landau concludes that elementary particles do not have an electric dipole moment.

However, this conclusion cannot be extended to