THE ANTIFERROMAGNETS $CoCO_3$, CoF_2, *AND* $FeCO_3$ *IN STRONG FIELDS*

V. I. OZHOGIN

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THE considerable interest shown recently in studies of the magnetization of antiferromagnets in strong fields [1-3] is due to the importance of the information on the magnitude of interactions in antiferromagnets which is obtained from the nonlinearity of the dependence of the magnetic moment m on the field H. Below we give the results of an experimental study of the m(H) curve for some antiferromagnets (selected for the reasons given below) at the temperature T = 1.9°K (i.e., $T \ll T_N$) in fields up to 180 kOe.

The measurements were carried out by a pulse method which, apart from a few details, was similar to that described in [4]. The dependences H(t) and m(t) were recorded with an OK-24 two-beam oscillograph, one input of which received an integrated signal from a Rogowski loop, and the other an integrated signal from a system of coils for measuring the magnetic moment of the sample (the effective number of turns × 1 cm^2 was of the order of 100; the integrator was electronic). Figure 1 gives the oscillograms obtained.

Figure 2 shows the dependence of the magnetic moment of single-crystal $CoCO_3$ (T_N = 18.1°K) on the field applied at right angles to the sample three-fold symmetry axis. From Dzyaloshinskiĭ's theory [5] it follows that the weak ferromagnetism of $CoCO_3$ leads to the dependence $m_\perp = \sigma + \chi_\perp H$, where $\sigma = \beta l$, and l is the projection of the antiferromagnetic vector along the direction perpendicular to the field and the three-fold axis. With increase of the field intensity the angle by which the magnetic moments of the sublattices are rotated toward alignment along the field increases and this projection decreases. Because of this the curve m(H) should gradually assume the shape of the $\chi_\perp H$ curve on increase of the field. Figure 2 shows that this effect had not yet appeared in the available fields. The values of σ and $\chi_\perp$, found from Fig. 2, are, respectively, 1500 cgs emu/mole (±20%) and 0.047 cgs emu/mole (±10%), in agreement with the results of measurements in weak fields. [6]

Measurements of the dependence m(H) for CoF_2 with the field in the basal plane of a single crystal were of interest because of the peak of the transverse susceptibility of this substance near T_N = 37.7 K. [7] The origin of the peak is obviously related to the mechanism proposed for the explanation of the similar peak in the case of $CoCO_3$: [6] antiferromagnetic ordering is induced by the magnetic field above T_N because of the Dzyaloshinskiĭ interaction. When $T < T_N$ the weak ferromagnetism of CoF_2 does not appear because the anisotropy is of type different from that in $CoCO_3$. However, in a sufficiently strong field directed at right angles to the axis of the crystal, the antiferromagnetic vector may alter its position markedly, becoming oriented at right angles to this axis and the field. Such a rotation is accompanied by an increase of the anisotropy energy and a reduction of the energy given by the term $\beta(m_x l_y + m_y l_x)$ in the expansion of the thermodynamic potential [8] (the possibility of such an effect was noted in [9]). A qualitative calculation of this effect, actually observed in CoF_2 (Fig. 3), may be carried out as follows.

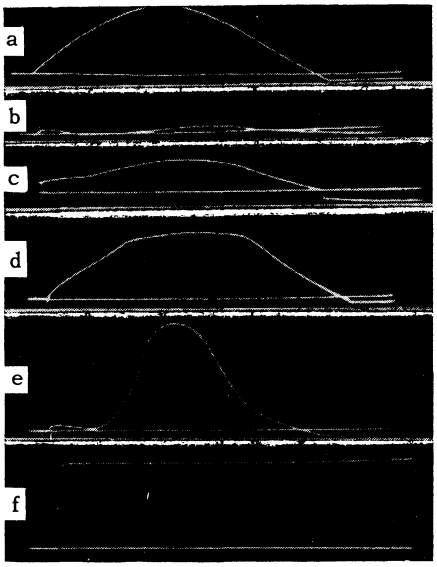
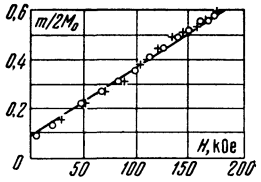


FIG. 1. Oscillograms from which the curves of Figs. 2-4 were obtained: a) field as a function of time, the time from zero to maximum being τ_{max} = 1.2 msec; b) integrated signal from the measuring coils in the absence of a sample, test temperature T = 1.9°K; c) magnetic moment of $CoCO_3$ as a function of time, T = 1.9°K; d) m(t) for CoF_2, T = 1.9°K; e) m(t) for $FeCO_3$, T = 1.9°K; f) calibration of the second beam: m(t) for a nickel sample (3.9 mm diameter, h = 7 mm), T = 300°K, H_{max} = 60 kOe.

FIG. 2. Magnetization curve of $CoCO_3$ in the basal plane; T = 1.9°K; $2M_0$ = 16 750 cgs emu/mole (calculated on the assumption of orbital moment "freezing").



Writing the Hamiltonian of the system in the form

$$\mathcal{H} = (2M_0)^2 [\tfrac{1}{2} A\lambda^2 + \tfrac{1}{2} B\mu^2 - \beta(\lambda_x\mu_y + \lambda_y\mu_x) - \tfrac{1}{2} a\lambda_z^2 - \mu h] \quad (1)$$

and eliminating λ by use of $\lambda^2 + \mu^2 = 1$ (which is a consequence of $|\mathbf{M}_1|^2 = |\mathbf{M}_2|^2 = \mathbf{M}_0^2$, where $\mathbf{M}_1$ and $\mathbf{M}_2$ are the magnetic moments of the sublattices), we obtain the following expression which must be minimized for μ and θ:

$$U(\mu, \theta) = \tfrac{1}{2}(\gamma + a)\mu^2 - \beta\mu\sqrt{1-\mu^2}\sin\theta + \tfrac{1}{2} a(1-\mu^2)\sin^2\theta - \mu h \quad (2)$$

(here $\mathbf{M}_1 - \mathbf{M}_2 \equiv \mathbf{l} \equiv 2\mathbf{M}_0\boldsymbol{\lambda}$, $\mathbf{M}_1 + \mathbf{M}_2 \equiv \mathbf{m} \equiv 2\mathbf{M}_0\boldsymbol{\mu}$, $\mathbf{H} \equiv 2\mathbf{M}_0\mathbf{h}$, $B - A = \gamma$, $\mathbf{h} = (h, 0, 0)$, $\boldsymbol{\mu} = (\mu, 0, 0)$, $\boldsymbol{\lambda} = (0, \lambda\sin\theta, \lambda\cos\theta)$, θ is the angle between $\mathbf{l}$ and the axis $z \parallel C_4$). Not restricting ourselves by the condition $4\beta \ll \gamma$, but assuming (quite roughly) that $h \ll (\gamma^2/8\beta + \beta)$, we can obtain:

$$H < H_c: \quad m = \chi_1 H, \quad \chi_1 = (\gamma + a - \beta^2/a)^{-1}; \quad (3)$$

$$H > H_c: \quad m = \sigma + \chi_2 H, \quad \sigma = 2M_0[\sqrt{1/2 + (\gamma/4\beta)^2} - \gamma/4\beta],$$
$$\chi_2 = [4\beta\sqrt{1/2 + (\gamma/4\beta)^2}]^{-1}. \quad (4)$$

Here

$$H_c = 2M_0(\beta h_1^2 - a h_1 h_2)/(\beta^2 - a^2), \qquad h_1^2 \equiv a\gamma + a^2 - \beta^2, \quad h_2^2 \equiv a\gamma. \quad (5)$$

Having found from Fig. 3 that $\chi_1 \sim 0.055$ cgs emu/mole, $\sigma/2M_0 \sim 0.25$, $\chi_2 \sim 0.027$ cgs emu/mole, and using Eqs. (3) and (4), we can estimate the interaction constants: $\gamma \sim 29$, $a \sim 4.4$, $\beta \sim 8.2$. Substituting these values into Eq. (5) we obtain $H_c \sim 80$ kOe, which is close to the experimental value $H_c \sim 110$ kOe (see also Fig. 3). The absence of a sharp transition may be accounted for by the field inhomogeneity in the interior of the sample ($\approx 5\%$), the influence of terms not allowed for in the Hamiltonian of Eq. (1), the inaccuracy of the orientation of the sample with respect to the field ($\pm 10°$), etc.

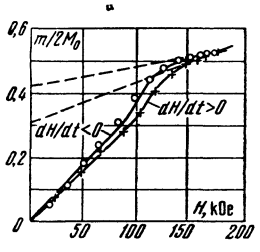


FIG. 3

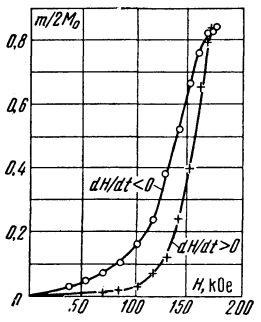


FIG. 4

FIG. 3. Magnetization curve of CoF_2 along a two-fold axis; T = 1.9°K; $2M_0$ = 16 750 cgs emu/mole.

FIG. 4. Magnetization curve of $FeCO_3$ along a three-fold axis; T = 1.9°K; $2M_0$ = 22 300 cgs emu/mole.

It was also of definite interest to determine the magnetization curve of single-crystal $FeCO_3$ (siderite[1)]) in strong fields directed along the C_3-axis, i.e., the axis along which an anomalous susceptibility peak occurs near T_N. Experiments actually showed a strong nonlinearity of m(H) which appeared in fields H > 100 kOe (Fig. 4), and, in contrast to the reversal effect in normal antiferromagnets (for example, in MnF_2), the value of m obviously increased to its nominal value. This behavior of m(H), for $FeCO_3$, as well as the form of the curves $\chi_{\parallel}(T)$ and $\chi_{\perp}(T)$ near T_N, resembles very much the behavior of metamagnets (of $FeCl_2$ type). However, final conclusions on the type of interactions in $FeCO_3$ can be drawn only after measuring the dependence m(H) in still stronger fields, which the author proposes to do in the immediate future (together with a study of the magnetization curves of other antiferromagnets having a peak of the transverse susceptibility or an anomalous peak of the longitudinal susceptibility near T_N). The considerable hysteresis between the m(H) curves in increasing and decreasing fields (as in the case of CoF_2) may be due to trivial causes (for example, rotation of the sample under the action of ponderomotive forces because of inaccurate orientation with respect to the field) or due to processes of establishing equilibrium in the case of sublattice moment reversal, if the times taken by these processes are not too short. However, this problem requires special study.

After writing this letter the author became acquainted with the work of Jacobs[11] giving the results of an experimental study of m(H) for siderite, which in the main are in agreement with those given above.

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[1)]Natural siderite single crystals (from the Neudorf deposit in Germany) were obtained from the Mineralogical Museum of the USSR Academy of Sciences, through the courtesy of its director Barsanov.

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