Light birefringence bilinear in the ferromagnetic and antiferromagnetic vectors in cobalt carbonate

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An increase was observed in the birefringence of plane-polarized light propagating parallel to the trigonal axis; this increase is due to the magnetic field directed along the light-propagation vector $\mathbf{H} \parallel \mathbf{k} \parallel C_3$. It is shown that the change of the birefringence can be phenomenologically described in terms of the contribution made to the dielectric susceptibility by terms proportional to the product of the projections of the ferromagnetism and antiferromagnetism vectors.

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In magnetically ordered multi-sublattice crystals, the changes that are produced in the refractive indices of the light in the course of ordering may be directly proportional to the changes in the projections of the magnetic vectors that characterize the structure of the magnetic subsystem. To our knowledge, however, no such relations have been observed. We report here the observation of magnetic birefringence (LB) bilinear in $\mathbf{m}$ and $\mathbf{l}$ in the two-sublattice antiferromagnet $CoCO_3$.

Cobalt carbonate is one of the most thoroughly studied weak ferromagnets with antiferromagnetic ordering symmetry of the type $3_z^+ 2_x^-$. Its magnetic properties were investigated in detail in[1], while the influence of the magnetic order on the optical properties of the crystal in the transparency region was investigated in[2]. In the present study we investigated the influence of the magnetic field on the birefringence of $CoCO_3$ in the case of light propagating along the axis $C_3 \| z$. The magnetic field was also directed along the C_3 axis. The errors in the $\mathbf{H} \| C_3$ and $\mathbf{k} \| C_3$ orientations did not exceed 1.5 and 0.1°, respectively. The birefringence was measured by two methods: with the aid of conoscopic figures, and by the method of a rotating analyzer with circularly polarized incident light.

Figure 1 shows photographs of the conoscopic figures for light with $\lambda \approx 4000$ Å. The centers of the dark circles are the points of emergence of the optical axes, and the distance between them is proportional to the angle between the optical axes $2V \sim (n_g - n_m)^{1/2} \approx (n_{xx} - n_{yy})^{1/2} = \Delta n_{xy}^{1/2}$. Plots of $\Delta n_{xy}(H)$ for light with $\lambda = 6328$ Å are shown in Fig. 2. In fields $H > 10$ kOe, where the sample is already homogeneous, the dependence of the LB on the magnetic-field intensity can be expressed by the empirical formula $\Delta n_{xy} = \Delta n_{xyo} + AH$, where at $T = 5$ K we have $\Delta n_{xyo} = (2.42 \pm 0.05) \times 10^{-4}$, $A = (3.76 \pm 0.08) \times 10^{-9}$ Oe^{-1}. The plane containing the optical axes is close to the $(C_3 C_2)$ plane.

It was shown in[2] that the LB produced in the course of magnetic ordering of $CoCO_3$ is well described by the dependence of ϵ_{ik} on the projections of the antiferromagnetic vector. Moreover, the only significant terms in the expression for Δn_{xy} are those that depend on the transverse components of $\mathbf{l}$. Taking this circumstance into

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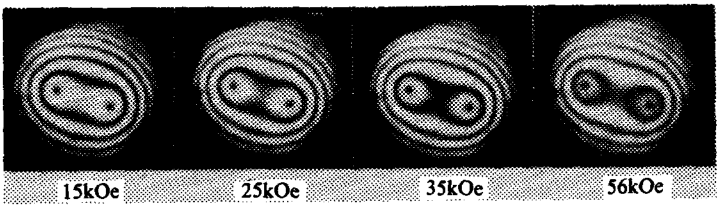

FIG. 1. Conoscopic figures of antiferromagnetic $CoCO_3$ as functions of the field intensity $H\|C_3(\lambda \approx 4000$ Å, $T=7$ K).

account and allowing for the ambiguity of the data on the orientation of the vector l in $CoCO_3$,[3] we could expect functions $\Delta n_{xy}(H)$ of two types: if $\theta_0=(\widehat{\mathbf{l}C_3})=\pi/2$,[3b] then Δn_{xy} should decrease with increasing field $\mathbf{H}\|C_3$, and if $\theta_0\neq\pi/2$,[3a] then Δn_{xy} can increase because of the rotation of l in the basal plane. It is interesting that in either case the changes are quadratic in H. Figure 2 shows plots of $\Delta n_{xy}(l_\perp)$ against H for $\theta_0=\pi/2$ (1) and $\theta_0=\pi/4$ (2). To calculate $l_\perp(H)$ we chose a potential that admits of an oblique orientation of the vector l on account of the opposite signs of the second- and fourth-order uniaxial anisotropy constants, and used the approximation in which $H \ll 1$ and $H_{a2}/H_{exc} \ll 1$. H_{ex} was assumed equal to 160 kOe, and according to[1] $H_{a_2}=50$ kOe. Higher values of H_{a2}[4] make the dependences of $\Delta n_{xy}(l_\perp)$ on H even weaker. It is seen from Fig. 2 that the deviation of the experimental results from the calculated curves greatly exceeds the measurement error.

The experimentally observed $\Delta n_{xy}(H)$ dependence can be explained only by assuming that an important contribution is made to the LB by the terms proportional to $m_i l_k$. The expression for the components of the tensor ϵ_{ik}^{-1} with allowance for the quadratic and bilinear terms in m and l can be written in the form[5]:

$$\epsilon_{ik}^{-1} = \Lambda_{ikrs}\, l_r l_s + \mathrm{M}_{ikrs}\, m_r m_s + \Delta_{ikrs}\, m_r l_s. \quad (1)$$

The second term in this expression can be neglected because of the small values of the transverse components of m. The corresponding increments to ϵ_{ik}^{-1} for a rhombohedral crystal take the form

$$\begin{aligned}
\delta\epsilon_{xx}^{-1} &= \Delta_5 m_z l_x + \Delta_3 m_x l_z + \Delta_2 m_y l_x - \Delta_1 m_x l_y, \\
\delta\epsilon_{yy}^{-1} &= -\Delta_5 m_z l_x - \Delta_3 m_x l_z + \Delta_1 m_y l_x - \Delta_2 m_x l_y, \\
\delta\epsilon_{zz}^{-1} &= \Delta_4 (m_x l_y - m_y l_x), \\
\delta\epsilon_{yz}^{-1} &= \Delta_6 (m_y l_x + m_x l_y) + \Delta_7 m_x l_z + \Delta_8 m_z l_x, \\
\delta\epsilon_{xz}^{-1} &= \Delta_6 (m_y l_y - m_x l_x) - \Delta_7 m_y l_z - \Delta_8 m_z l_y, \\
\delta\epsilon_{xy}^{-1} &= \frac{1}{2}(\Delta_2 - \Delta_1)(m_y l_y - m_x l_x) + 2\Delta_3 m_y l_z + 2\Delta_5 m_z l_y.
\end{aligned} \quad (2)$$

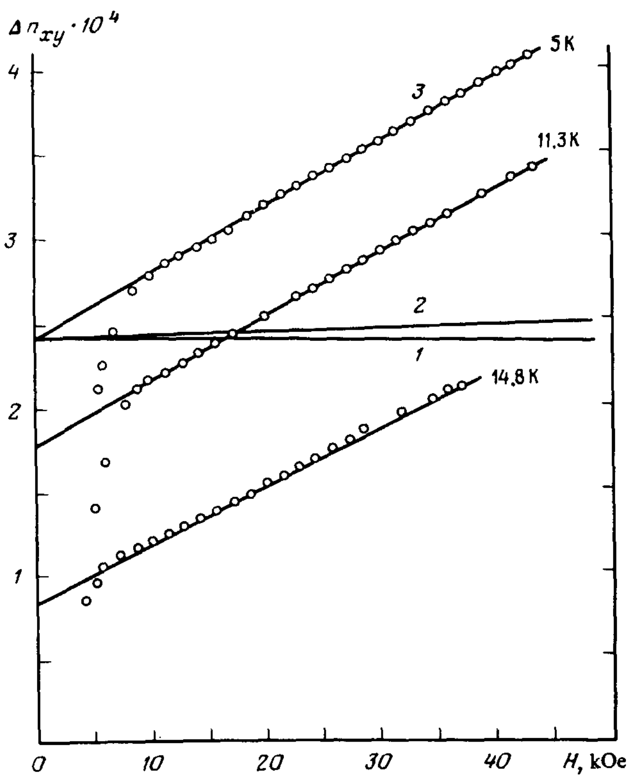

FIG. 2. Dependence of the LB on the intensity of the field $\mathbf{H}\|C_3$ for light propagating along the axis $C_3(\lambda=6328$ Å).

The projections m_r and m_s, which depend on $\mathbf{H}\|C_3$, must be obtained from the potential that includes the invariants responsible for the orientation of the vector $\mathbf{l}$ in the basal plane at $\mathbf{H}\|C_3$. In sufficiently strong fields $H>H_c$, the nonvanishing projections are l_x, m_z, and m_y, while m_x and l_y vanish.[6] The component l_z, even if different from zero, makes no noticeable contribution to $\Delta n_{xy}(l_i l_k)$[2a] and is insignificant for $\Delta n_{xy}(m_r l_s)$ at $\mathbf{H}\|C_3$. If no account is taken of the terms that depend on l_z, the following expression is valid:

$$\Delta n_{xy} = \frac{1}{2} n_o^3 \{ (\Lambda_{11}-\Lambda_{12}) l_x^2 + (\Delta_2 - \Delta_1) m_y l_x + 2\Delta_5 m_z l_x \}. \quad (3)$$

In fields $H_c < H < H_{exc}$, at which m_y, $l_x \approx$ const, $m_z = \chi_{zz}H$, the first two terms depend very little on H, while the last ensures a linear dependence of Δn_{xy} on the magnetic field intensity:

$$\Delta n_{xy}(H_c < H < H_{exc}) = \Delta n_{xyo} + \operatorname{sign} l_x \cdot 2 n_o^3 \Delta_5 M_o \chi_{zz} H. \quad (4)$$

The experimentally obtained relation (Fig. 2, line 3) yields for the magneto-optical coefficient $\Delta_5 = \Delta_{xxzx} = -\Delta_{yyzx}$ at $T=5$ K a value 1.03×10^{-12}(cgs emu/mole)^2, which exceeds the effective coefficient $[(\Lambda_{11}-\Lambda_{12})+q(\Delta_2-\Delta_1)]=2.73\times10^{-13}$ (cgs emu/mole)^2, that which gives rise to the spontaneous magnetic birefringence. The values $M_0 = 8.37 \times 10^3$ cgs emu/mole, $\chi_{zz}? = 3.45 \times 10^{-2}$ cgs emu/mole were taken from [1], and $q = m_0/l_\perp$, where m_0 is the spontaneous magnetic moment. The magneto-optical coefficients Δ_{ikrs}, which describe the changes linear in the field of the components of the symmetrical part of the tensor ϵ_{ik}, can be due to changes in the quantum mechanical states of the Co^{2+} ions, which are not equivalent in $CoCO_3$ with respect to right-hand and left-hand rotations of the spin in the direction towards the C_3 axis, as well as the elasto-optical effect, which is produced in the presence of linear magnetostriction. In cobalt compounds, both effects can be appreciable.[7] The independence of the induced birefringence of the sign of H_z is evidence of reversal of magnetization when the field directions of the both the ferromagnetic and antiferromagnetic vectors are reversed.

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