Lowering of the optical class of an antiferromagnetic crystal, induced by a longitudinal magnetic field

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It was observed in experiment that a longitudinal magnetic field induces birefringence, linear in the field intensity, in a two-sublattice collinear tetragonal antiferromagnet in which linearly polarized light propagates along the tetragonal axis. The magnitude of the birefringence makes it possible to distinguish visually, in a magnetic field, antiferromagnetic states that have different directions of the antiferromagnetic vector. It is shown that the distortions in the tetragonal-structure field are not the decisive factor in the observed magneto-optical effect.

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The presence of a magnetic subsystem in a magnetically ordered crystal can lead, under certain conditions, to somewhat unusual optical properties of crystals with higher crystallographic and magnetic syngonies. For example, tetragonal crystals in whose magnetic point group the operation of rotation through an angle $\pi/2$ is a symmetry operation only when combined with the time-reversal operation R, must inevitably become of a lower optical class in a magnetic field **H** that is directed along the C_4 axis and does not upset the collinear arrangement of the magnetic moments of the ions.[1] The considered magneto-optical effect is linear in H_z and is related to the inverse piezomagnetic effect in uniaxial crystals.

We report here observation of a birefringence, linear in the magnetic-field intensity, of linearly polarized light propagating along the crystallographic axis of the tetragonal collinear antifferromagnet CoF_2. Measurements of the birefringence (LB) was carried out in pulsed magnetic fields by a conoscopic method with circularly polarized light. The stresses due to the mounting of the sample led to a small deformation of the optical indicatrix, such that the plane of the optical axes was close to the (100) plane

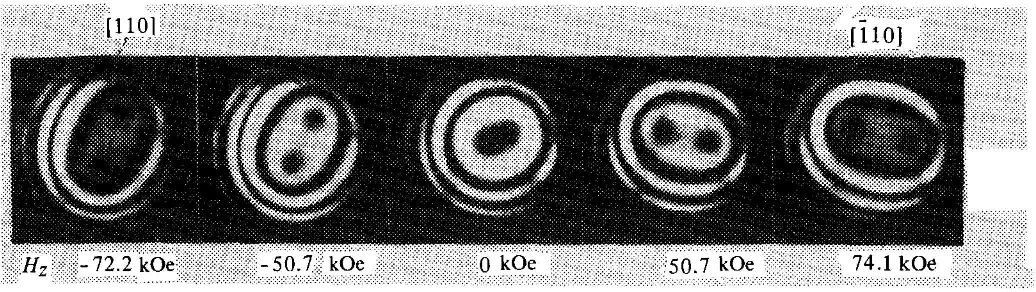

FIG. 1. Change of conoscopic figures of the collinear antiferromagnet CoF_2 as a function of the direction and magnitude of the field $\mathbf{H} \parallel C_4 \parallel \mathbf{Z}$. The sample is in a monodomain antiferromagnetic state λ_z^+, $\lambda \approx 4000$ Å, and $T \approx 30$ K.

(Fig. 1). These stresses contributed to the formation of a definite monodomain antiferromagnetic structure. Turning on a magnetic field $\mathbf{H} \| C_4$ gives rise to a distinctly observable increase of the angle between the optical axes, and a rotation of the plane in which the axes are located. The direction of the rotation depends on the direction of **H** and on the prior history of the sample, which determines the direction of the antiferromagnetic vector in the crystal. Figure 1 demonstrates the change of the indicatrix of a monodomain antiferromagnetic (AFM) crystal as a function of the direction and magnitude of the magnetic field. The dependence of the quantity $(\tilde{n}_g - \tilde{n}_m)$, determined from the conoscopic figures, on the field intensity is nearly linear in a field exceeding 15 kOe, and the difference between the azimuths of the planes in which the axes are located, at opposite field directions, tends to 90° with increasing field.

The *DB* induced by the field is most conveniently described by the dependence of the components on the dielectric tensor on the orientation and magnitude of the magnetic vectors that describe the magnetic structure of the crystal. For the CoF_2 crystal with the magnetic ions subdivided into sublattices of the type $4_z^- 2_d^+$, we can write

$$\begin{aligned}
\delta \epsilon_{xx}^{-1} &= \Delta_{xxxy} m_x l_y + \Delta_{xxyx} m_y l_x , \\
\delta \epsilon_{yy}^{-1} &= \Delta_{xxxy} m_y l_x + \Delta_{xxyx} m_x l_y , \\
\delta \epsilon_{zz}^{-1} &= \Delta_{zzxy} (m_x l_y + m_y l_x), \\
\epsilon_{yz}^{-1} &= \Delta_{yzxz} m_x l_z + \Delta_{yzzx} m_z l_x , \\
\epsilon_{xz}^{-1} &= \Delta_{yzxz} m_y l_z + \Delta_{yzzx} m_z l_y , \\
\epsilon_{xy}^{-1} &= \Delta_{xyxx} (m_x l_x + m_y l_y) + \Delta_{xyzz} m_z l_z .
\end{aligned} \tag{1}$$

In longitudinal fields $H < 10^5$ Oe, the magnetic structure of CoF_2 remains collinear, the transverse components m_x, m_y, λ_x, and λ_y are equal to zero, and the difference between the principal refractive indices in a crystal free of mechanical stresses is

$$n_g - n_m = 2n_0^3 \Delta_{xyzz} M_0 \chi_{zz} H . \qquad (2)$$

The aximuth of the optical axes relative to the X axis is $\phi = \pi/4\,\mathrm{sign}H_z \mathrm{sign}\, l_z$. Taking into account the observed initial deformation of the indicatrix, this azimuth differs from 45°, and the difference between the refractive indices is

$$n_{x'x'} - n_{y'y'} = (\tilde{n}_g - \tilde{n}_m)\sin 2\tilde{\phi} = 2n_0^3 \Delta_{xyzz} M_0 \chi_{zz} H\, \mathrm{sign}\, l_z\, \mathrm{sign} H_z , \qquad (3)$$

where $(\tilde{n}_g - \tilde{n}_m)$ and $\tilde{\phi}$ are measured quantities, the axes X' and Y' are directed parallel to the [110] and [$\bar{1}$10] axes, in contrast to $X \parallel [100]$ and $Y \parallel [010]$.

Figure 2 shows the obtained dependence of the *LB* $\Delta n_{x'y'} = n_{x'x'} - n_{y'y'}$ on the field intensity. The two straight lines correspond to two monodomain states of the sample with opposite directions of the AFM vector **1**. The stable direction of this vector depends on the magnitude and direction of the field. The sample remains monodomain also after the field is turned off. The reversal of magnetization of the sublattices, wherein the direction of **m** remains the same and the energywise unprofitable direction of **1** is reversed, proceeds via formation of a domain structure of two AFM states λ_z^+ and λ_z^-. The magnetization-reversal process is illustrated in Fig. 3 with conoscopic figures. Figure 3 corresponds to an inhomogeneous state of the sample with several large AFM domains. From the plot of $\Delta n_{x'y'}(H)$ (Fig. 2), we can determine the coefficient Δ_{xyzz}, which turned out to be 1.3×10^{-12} (cgs emu/mole)^2 at $T = 11$ K and $\lambda = 4000$ Å. The required values $\chi_{zz} = 1.3 \times 10^{-2}$ cgs emu/mole and 2 $M_0 = 16\,750$ cgs emu/mole were taken from Refs. 2 and 3, and the refractive index was assumed to be 1.5.[4]

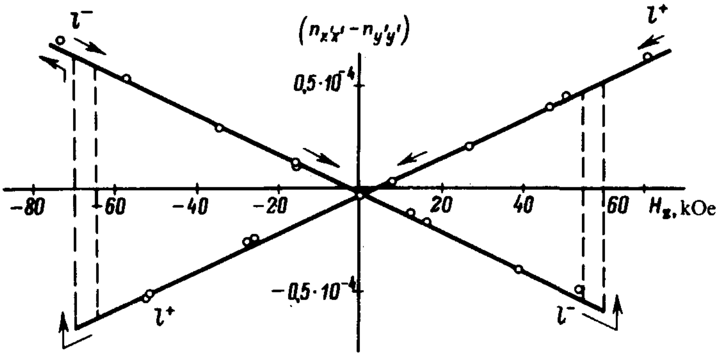

FIG. 2. Birefringence, induced by a magnetic field $\mathbf{H} \parallel C_4 \parallel \mathbf{Z}$ for light propagating along the C_4 axis as a function of the direction and magnitude of the field. The two straight lines correspond to two antiferromagnetic states of the sample; $\lambda \approx 4000$ Å, $T \approx 11$ K.

The magneto-optical coefficient responsible for the *DB* which is linear in the field can receive contributions from different mechanisms. The apperance of $\epsilon_{xy} \neq 0$ is due to the different polarizability of the non-equivalent Co^{++} ions, jointly with a ligand environment, in the magnetic field as a result of the following: 1) the non-equivalent

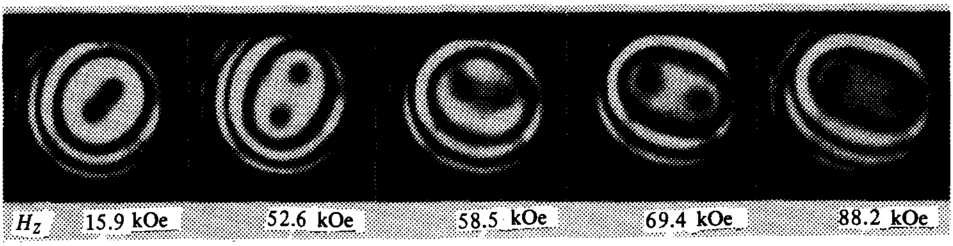

FIG. 3. Change of the conoscopic figures in the course of the reversal of magnetization of the antiferromagnetic state of the sample. The initial antiferromagnetic state λ_z is energywise not favored in a field $H_z > 60$ kOe.

quantum-mechanical mixing of the states in the magnetic field under the joint influence of the low-symmetry crystal field and the spin-orbit interaction; 2) the difference between the populations of the low-symmetry excited states in the non-equivalent ions; 3) the different shifts of the energy levels of the ions of the first and second sublattices. In addition, an appreciable contribution to ϵ_{xy} can be made by a secondary photoelastic effect due to the magnetostriction in the case of the inverse piezomagnetic effect. To determine the secondary *DB*, we measured the piezo-optical effect in CoF_2. The sample was compressed in the [110] direction and the temperature was 20.4 K. The photoelastic coefficient π_{66} was close to 0.6×10^{-13} cm²/dyn. Knowing the magnetostriction deformations along [110] $|\Delta l/l|_{xy} = 4.9\times10^{-10} H$ (Ref. 5) and the elastic constant $C_{66} = 8.5\times10^{11}$ dyn/cm² (Ref. 4), we obtain the contribution made to the *DB* by the linear magnetostriction in a field **H** || **Z**:

$$|\Delta n_{x'y'}|_{pe} = 4.9\times10^{-10} \quad n_0^3 \pi_{66} C_{66} H,$$

where H is in oersteds. At $H = 5\times10^4$ Oe we have $\Delta n_{x'y'}|_{pe} = 0.4\times10^{-5}$, which is larger by one order of magnitude than the observed 0.5×10^{-4}. Consequently, distortions of the tetragonal structure are not decisive for the observation of the magneto-optical effect.

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