Quadratic magnetic rotation of the polarization plane of light in the antiferromagnet CoF_2

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A magnetic-field-induced rotation of the polarization plane of light, proportional to the square of the magnetic field, has been observed experimentally in a two-sublattice antiferromagnetic crystal without an anti-inversion center. The rotation occurs in different directions for states of the crystal with opposite directions of the antiferromagnetic vector. The magnitude of the rotation is such that it is possible to visually observe 180° antiferromagnetic domains in moderate magnetic fields.

The ordering of a magnetic subsystem of a crystal allows new optical effects, which are forbidden by symmetry in magnetically disordered media. One such effect is a nonreciprocal rotation of the polarization plane of light, which is of even parity in the direction of the magnetic field. An even-parity magnetic rotation should not change sign when the magnetic field vector is reversed, but it should change sign upon the reversal of all the elementary magnetic moments associated with the magneto-optical properties of the crystal. That such an effect is possible in media which are not symmetric under the anti-inversion operation $\underline{\bar{I}} = \bar{I}\cdot\underline{I}$ was mentioned in Refs. 1 and 2 and stressed in Ref. 3. This possibility follows from the generalization to magnetically ordered media of the Onsager relations for the kinetic coefficients,[4] which lift the prohibition against terms of even parity in **H** in the expansion of the antisymmetric components of the dielectric tensor:

$$\epsilon_{ij}^{a} = f_{ij\alpha} H_\alpha + F_{ij\alpha\beta} H_\alpha H_\beta. \tag{1}$$

A quadratic magnetic rotation of the plane of polarization of light can be described by a polar fourth-rank c-tensor $F_{ij\alpha\beta}$ which is antisymmetric with respect to its first pair of indices and symmetric with respect to its second pair or by its dual, a third-rank axial c-tensor $G_{k\alpha\beta} = e_{kij}F_{ij\alpha\beta}$. A third-rank axial c-tensor $q_{ij\alpha\beta}$ also describes the linear magneto-optical effect,[2] but because of the different symmetries of the tensors $q_{ij\alpha\beta}$ and $G_{k\alpha\beta}$ with respect to the interchange of indices,the magnetic classes of crystals in which these effects are allowed are not completely identical. Both effects are forbidden in centrally symmetric crystals having a center of anti-inversion $\underline{\bar{I}}$ and in the classes 432, $\bar{4}3m$, and $m3m$. Furthermore, a quadratic magnetic rotation of the polarization plane of light is forbidden in the classes 422, $4mm$, $\bar{4}2m$, $4/mmm$, 622, $6mm$, $\bar{6}m2$, and $6/mmm$, where a linear magneto-optical effect is allowed. A quadratic magnetic rotation of the polarization plane of light is allowed in the same 31 pyromagnetic and 27 antiferromagnetic crystal classes, in which a magnetization quadratic in **H** is allowed.[5]

In the present letter we report the experimental observation of a rotation, quadratic in the field, of the polarization plane of light in an antiferromagnetic crystal. We studied the thoroughly investigated two-sublattice antiferromagnet CoF_2, whose structure is described by the magnetic point group $4/mm\underline{m}$. The components $G_{k\alpha\beta}$, which may be nonvanishing in this material, are G_{zxy} and $G_{xyz} = G_{yxz}$. The manifestation of a quadratic magnetic rotation of the polarization plane of light that might be detected most easily is the rotation of the polarization plane of light propagating along the C_4 axis in a field $\mathbf{H}\|[110]$. This rotation is described by the coefficient G_{zxy}. The experimental geometry $\mathbf{k}\|C_4\perp\mathbf{H}$ is favorable in that it is insensitive to a linear magnetic rotation of the polarization plane of light (the Faraday effect) and also insensitive with respect to a linear magneto-optical effect. Although the latter effect does lead to a splitting of the optic axis because of the appearance of components ϵ^s_{yz} and ϵ^s_{xz} in the dielectric tensor, the additional rotation of the indicatrix axis x_3 leaves the C_4 axis coinciding with one of the two optic axes.

In the experiments, two superconducting solenoids with mutually perpendicular axes are inserted into each other. The outer solenoid has the configuration of a Helmholtz coil. The solenoids make it possible to independently vary the longitudinal and transverse projections of the magnetic field, $\mathbf{H}_z\|C_4$ and $\mathbf{H}_{x'}\|[110]$, respectively. The ability to independently vary H_z and $H_{x'}$ makes it possible to vary the antiferromagnetic domain structure of the sample, since the field $H_{x'}$ induces a magnetic moment, whose z projection has a sign determined by the sign of the z projection of the antiferromagnetic vector $\mathbf{L}$. After the field H_z is removed,the antiferromagnetic domain structure remains essentially unaltered. By switching the direction of a sufficiently strong field $\mathbf{H}_z$ one can put the sample in a homogeneous state AFM^+ or AFM^-. Figure 1 shows the specific rotation angle φ versus H. Also shown here, by the solid lines, are the functions

$$\varphi/l = \pm BH^2 + cH, \qquad (2)$$

which give a good description of the positions of the experimental points for both states, AFM^+ and AFM^-, for the parameter values $B = 1.3\times 10^{-8}$ deg/(cm·Oe^2) and $c = 2\times 10^{-5}$ deg/(cm·Oe). Most probably, the increment which is linear in $\mathbf{H}$ is actually due to the presence of a projection of the magnetic field onto the light propagation direction; it is a Faraday rotation. The angle by which $\mathbf{H}$ would have to deviate from a perpendicular orientation is 1°, within the possible errors.

Let us compare the values of the constant (B) of the quadratic magnetic rotation of the polarization plane of light with the Verdet constant, V, noting that the Faraday rotation is proportional to the magnetization of the crystal. If we consider that we are applying the projections of the ferromagnetic and antiferromagnetic vectors, rather than the magnetic field, to the crystal, we can write the antisymmetric component ϵ^a_{xy} as the sum

$$\epsilon^a_{xy}(\mathbf{H}\perp C_4) = F^m_{xyz}M^{kb}_z + F^l_{xyxyz}L_xL_yL_z + F^{ml}_{xyxyz}M_xM_yL_z = F_{xyxy}H_xH_y, \qquad (3)$$

$$\epsilon^a_{xy}(\mathbf{H}'\| C_4) = f^m_{xyz}M_z = f_{xyz}H_z. \qquad (4)$$

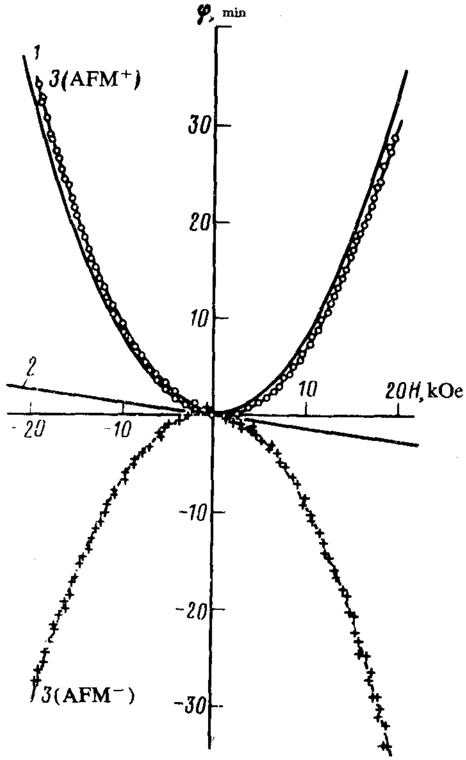

FIG. 1. The angle through which the polarization plane of light is rotated versus the strength of a magnetic field H‖[110] in the antiferromagnet CoF_2. ●, + —Experimental points corresponding to two antiferromagnetic states. Solid lines: 1—Quadratic magnetic rotation ; 2—Faraday effect due to a slight deviation of the angle $(\widehat{\mathbf{kH}})$ from 90°; 3—the dependence $\pm BH^2 + cH$ for $B = 1.3\times 10^{-8}$ deg/(cm·Oe^2) and $c = 2\times 10^{-5}$ deg/(cm·Oe). The sample thickness is $l = 1$ mm; $\mathbf{k}\|C_4$; $T = 15$ K; $\lambda = 6328$ Å.

Here $M_z = \chi_{zz} H_z$, and $M_z^{kb} = \beta h_x h_y h_z + \gamma M_x M_y h_z = C_{zxy} H_x H_y$ is the magnetization which is quadratic in **H**. The nature of the appearance of **M** is different for two different field orientations. Consequently, the coefficients F^m_{xyz} and f^m_{xyz} may not be equal. The presence of the two other terms in (3) means that a quadratic rotation may appear in the case $M_z^{kb} = 0$ also, but with nonvanishing $h_x h_y h_z$ or $M_x M_y h_z$. Since we do not know all the constants in (3), we set F^l_{xyxyz} and F^{ml}_{xyxyz} equal to zero and compare the experimental results found for $B_0^m = (\pi/\lambda n) F^m_{xyz}$ and $V_m = (\pi/\lambda n) f^m_{xyz}$.

Assuming $C_{xyz} = 6.8\times 10^{-9}$ emu/(cm^3·Oe^2), which follows from the value $M_z^{kb} = 360$ emu/mole, found in Ref. 6 at $H_{x'} = 50$ kOe, $X_{zz}(T = 15\text{ K}) = 6.0\times 10^{-4}$ emu/cm^3 (Ref. 7), and the measured value $V = 1.43$ deg/(cm·kOe), we find $B_0^m = \varphi / M_z^{kb} l = 2.0$ deg/(cm·G) and $V^m = V/\chi_{zz} = 2.4$ deg/(cm·G). The values of B_0^m and V^m are approximately the same. Since the systematic errors in the determination of χ_{zz} and (especially) C_{xyz} can be greater than 10%, we cannot rule out the possibility that B_0^m and V^m are equal. The equality of these magneto-optical coefficients would be evidence of the equality in CoF_2 of the specific contributions to the magnetic rotation of the polarization plane from quantum-mechanical states which are mixed with the ground state during the application of a field $\mathbf{H}\|C_4$ and **H**‖[110].

The quadratic magnetic rotation of the polarization plane of light can be used to visually distinguish 180° antiferromagnetic domains in a field **H**‖[110] of about 10 kOe.

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