

# Visual observation of 180-degree antiferromagnetic domains

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(Submitted 26 February 1979)

*Pis'ma Zh. Eksp. Teor. Fiz.* **29**, No. 7, 432-435 (5 April 1979)

We report the first visual observation of 180-degree antiferromagnetic domains. Observations were conducted by means of a new linear magneto-optical effect that is proportional to the first power of magnetic field strength and the projection of the antiferromagnetic vector. The object of investigation was a collinear two-sublattice antiferromagnetic  $\text{CoF}_2$ .

PACS numbers: 75.60.Ch, 75.50.Ee, 78.20.Ls

Magnetically-ordered crystals in which an anti-inversion operation is absent as a symmetry operation, may sustain a linear magneto-optical effect (LMOE) for which there is no analog in the magneto-optics of non-magnetic crystals.<sup>1-3</sup> It consists of the occurrence in a magnetic field of birefringence of plane-polarized light whose value is proportional to the first power of the field strength in contrast to the quadratic Cotton-Mouton and Voigt effects. The resultant birefringence is connected to the induction by the magnetic field of a nonequivalence of the magnetic sublattices with respect to a 180° flip of their magnetic moments. The LMOE is described by components of the symmetric portion of the dielectric tensor which are linearly-dependent on field

intensity  $\Delta^s \epsilon_{ij} = E_{ijk} H_k$ . It follows from Onsager's relations for the magnetic-field-dependent kinetic coefficients<sup>4</sup> that the matrices of the symmetric kinetic coefficients and the corresponding coefficients of static properties share the same spatial and time symmetry in magnetically-ordered media. Consequently, the tensor  $E_{ijk}$  has the same symmetry as the tensor of the inverse piezomagnetic effect—axial  $c$ -tensor symmetry. Having shown interest in the dependence of  $\Delta^s \epsilon_{ij}$  on the internal characteristics of a two-sublattice antiferromagnetic and in considering the magnetic vectors  $\mathbf{m}$  and  $\mathbf{l}$  as sources of external effects on a crystal, it is evident that the sign of  $\Delta^s \epsilon_{ij}$  depends on the direction of the magnetic field and the directions of the sublattice moments. A flip in the direction of the latter leads to a change in the sign of the effect since the field direction remains unchanged. This circumstance provides a means of distinguishing the states of an antiferromagnetic crystal with inversely-oriented antiferromagnetic vectors. In this work we report on the first visual observation of the 180-degree antiferromagnetic domains.

We investigated the two-sublattice antiferromagnetic  $\text{CoF}_2$  which is characterized by  $\underline{4}/mmm$  crystallographic symmetry and  $4_z^+ 2_d^-$  magnetic ordering symmetry (point magnetic group  $\underline{4}/m\bar{m}\bar{m}$ ), with well-defined piezomagnetic properties.<sup>5,6</sup> Linear-in-the-field additions to the symmetrical part of the dielectric tensor may be expressed as follows:

$$\Delta^s \epsilon_{yz} = \text{sign} l_z E'_{yzk} H_x, \quad \Delta^s \epsilon_{xz} = \text{sign} l_z E'_{yzx} H_y,$$

$$\Delta^s \epsilon_{xy} = \text{sign} l_z E'_{xyz} H_z.$$

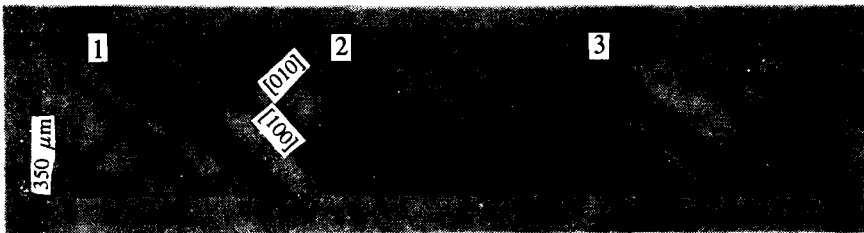


FIG. 1. 180-degree antiferromagnetic domain in  $\text{CoF}_2$ . Field intensity 49 kOe.



FIG. 2. Effect of magnetic field on antiferromagnetic domain structure. Photograph 1, 2 and 3 were obtained 20, 24 and 90 sec after the field intensity was increased from 29 to 44 kOe.

LMOE is simplest to achieve with longitudinal experimental geometry when  $\mathbf{H}$  and the light propagation vector  $\mathbf{k}$  are parallel to the tetragonal axis  $C_4$ . In this case birefringence occurs with zero background since both natural and spontaneous magnetic birefringence along the  $C_4$  axis are nonexistent. The optical axes of the  $\text{CoF}_2$  antiferromagnetic in a magnetic field  $\mathbf{H} \parallel C_4$  are positioned symmetrically with respect to  $C_4$  in the  $(110)$  or  $(\bar{1}\bar{1}0)$  planes depending on the sign of  $l_z$  for a fixed direction of  $\mathbf{H}$ . The angle between the axes, the magnitude of induced birefringence and the associated phase difference between the optical modes are, respectively<sup>3</sup>

$$\frac{n_g - n_m}{H} = 9,4 \cdot 10^{-7} \text{ kOe}^{-1}, \quad 2V/H^{-1/2} = 0,7 \text{ deg} \cdot \text{kOe}^{-1/2},$$

$$\delta / t H = 0,85 \text{ deg} \cdot \text{kOe}^{-1} \cdot \text{mm}^{-1}.$$

The above values correspond to a temperature of 11 K and an optical wavelength of 4000 Å. They indicate that the antiferromagnetic domains may be visually discernible in fields as low as 1 kOe.

Figures 1 and 2 show photographs of the antiferromagnetic structure of a  $\text{CoF}_2$  specimen with 1.7-mm thickness. The aperture of the illumination source is  $2^\circ$ . The specimen is in a vacuum and is attached with a maximum of freedom—it is pressed to the cold-duct with a strip of paper. Specimen temperature is approximately 24 K. Photograph #2 in Fig. 1 and all the photographs in Fig. 2 were taken in the absence of phase plates in the optics. In this case, antiferromagnetic domains  $l_z^+$  and  $l_z^-$  are visually indistinguishable since the polarization of the light transmitted through them differs only by the direction of rotation of the polarization ellipse for the same azimuth and axis ratio. However, their overlap regions are different since in the case of a slanting boundary between the domains polarization of the transmitted light varies smoothly from the left to the right ellipse, going through a linear polarization state. The direction of polarization of the incident light with respect to the planes of the optical axes in the domains was  $0, 90^\circ$ ,  $\mathbf{E} \parallel [110]$  (Fig. 1) and  $\pm 45^\circ$ ,  $\mathbf{E} \parallel [100]$  (Fig. 2). An analyzer was set to the maximum darkening of the overlap regions. We obtained photographs 1 and 3 (Fig. 1) with the aid of a  $\lambda/4$ -plate as compensator. Its position was set to achieve the same optical density in the transparent domains  $l_z^+$  and  $l_z^-$  for an analyzer orientation parallel to the plate. Subsequently, rotating the analyzer left or right resulted in the darkening of one or another antiferromagnetic domain. In the case of photographs 2 or 3 (Fig. 1) the rotation angles are approximately  $\pm 15^\circ$ . Contrast between the domains for such a placement of polarization elements was sufficient to observe the antiferromagnetic domains in a field of approximately 0.5 kOe.

The domain structure of a test specimen is movable. Upon heating and repeated cooling the domain structure assumed a different form, although certain of its common features were preserved (Figs. 1 and 2). The state of the domain structure is the following: predominant parallelism between the domain walls and type (100) planes, the wedge-shaped form of the domain edges—characteristic for twin crystals—affirms the significant effect magnetostrictive deformations and the occurring elastic stresses exert on the relative distribution of antiferromagnetic domains. The structure of the

latter is sensitive to the magnitude and direction of the magnetic field which may be associated not only with the presence of an inhomogeneous piezomagnetic moment constrained by residual elastic stresses but also with the occurrence and redistribution of elastic stresses of a multi-domain specimen in a magnetic field as a result of linear magnetostriction. Figure 2 also shows the rate at which a domain structure changes for a discrete field variation.

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