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HAL Id: jpa-00209808
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Submitted on 1 Jan 1984

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Birefringence and Frederiks effect in a cubic liquid crystal

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(Reçu le 24 octobre 1983, accepté le 12 décembre 1983)

Résumé. — Les expériences montrent que les cristaux liquides nématisques existants sont uniaxial avec une symétrie de groupe continu. L'existence de cristaux liquides présentant une symétrie de groupe discret, par exemple la symétrie du groupe du cube, n'est pas interdite. Les candidats les plus probables pour cette classe de composés sont les smectiques D qui ne présentent pas d'anisotropie optique. La question se pose de savoir comment distinguer expérimentalement un cristal liquide cubique d'un liquide isotrope. Dans ce travail, on propose d'utiliser à cette fin l'effet Kerr anisotrope.

Abstract. — Experiments show that the existing nematic liquid crystals are uniaxial with the continuous group symmetry. The existence of liquid crystals possessing a discrete group symmetry — for instance, the cubic group symmetry — is not forbidden. The most probable candidates for this class of compounds are so-called smectics D which do not show optic anisotropy. The question arises of how to distinguish experimentally a cubic liquid crystal from an isotropic liquid. In this work the anisotropic Kerr effect is proposed for this purpose.

Nelson and Toner [1] proposed that there exist smectic liquid crystals with the cubic group symmetry. The most probable candidates for this class of compounds are so-called smectics D, which do not show optic anisotropy [2, 3]. The question of how to distinguish experimentally a cubic liquid crystal from an isotropic liquid arises. In this work the anisotropic Kerr effect is proposed for this purpose.

According to Nelson and Toner [1] (see also Patsashinskii and Mitus [4]), the order parameter in a cubic liquid crystal is defined by a fourth order tensor \( \lambda_{\alpha\beta\gamma\delta} \) where \( \mathbf{n'} \) are the unit vectors directed along the fourth-order axes of a cube.

\[
\lambda_{\alpha\beta\gamma\delta} = \lambda \left( \sum_{i=1}^{3} n_i' n_i' n_i' n_i' - \frac{1}{3} (\delta_{\alpha\beta} \delta_{\gamma\delta} + \delta_{\alpha\gamma} \delta_{\beta\delta} + \delta_{\alpha\delta} \delta_{\beta\gamma}) \right)
\]

where \( \mathbf{n'} \) are the unit vectors directed along the fourth-order axes of a cube.

Let us consider a cubic liquid crystal placed in the external electric field \( \mathbf{E} \). The energy of interaction of such a crystal with the external field can be written using rotational invariance considerations:

\[
H_1 = \frac{1}{4} g \lambda a x y z E_x E_y E_z E_a = \frac{1}{4} g \lambda \sum_{i=1}^{3} (\mathbf{n'} \cdot \mathbf{E})^4.
\] (2)

One can easily find the anisotropic part of the dielectric tensor from equation (2):

\[
e^{ab}_{\theta} = \frac{\partial^2 H_1}{\partial E_a \partial E_b} - \frac{1}{3} \delta_{ab} \frac{\partial^2 H_1}{\partial E_i^2} = g \lambda \sum_{i=1}^{3} n_i' n_i' (\mathbf{n'} \cdot \mathbf{E})^2 - \frac{1}{3} g \lambda \delta_{ab} E^2.
\] (3)

Generally the eigenvalues of the tensor \( e^{ab}_{\theta} \) are

\[
e^{a}_{\theta} = g \lambda [(\mathbf{n'} \cdot \mathbf{E})^2 - \frac{1}{3} E^2]
\] (4)

where the principal axes are directed along the vectors \( \mathbf{n'} \). When the field \( \mathbf{E} \) is directed along a diagonal of the cube or along one of the fourth-order axis, the anisotropy is uniaxial. This situation is difficult to distinguish from the Kerr effect in an isotropic liquid. In order to distinguish a cubic liquid crystal from an isotropic liquid it is necessary to measure the Kerr effect while varying the direction of the electric field \( \mathbf{E} \). However, the variation of the direction of the electric field cannot always be followed by the change of the Kerr effect. Indeed, so far we consider...
the triad $n'$ to be oriented rigidly, while the field $E$ rotates freely with respect to it. Provided the effect of boundaries and other orienting factors is negligible, the field $E$ will orient the cubic liquid crystal itself. The resulting orientation depends on the sign of $g$. If $g > 0$, the crystal is oriented in such a way as to make the direction of $E$ coincide with a diagonal of the cube. If $g < 0$, $E$ is directed along one of the fourth-order axes. So the cubic liquid crystal is oriented by the electric field in such a way as to make it impossible to distinguish from an isotropic liquid. Hence the experiments should be done in layers sufficiently thin to orient the liquid crystal with the walls.

Though we do not know anything about the orientational action of walls, the effect of wales is probably to orient a crystal in such a way as to direct the normal to a wall along a diagonal or along the edge of the cube.

In the vicinity of a wall the direction of a diagonal or an edge of the cube is fixed; however, a free rotation remains possible in the plane perpendicular to the indicated direction. The electric field $E$, whose direction does not coincide with that of the normal to the surface, fixes this azimuthal angle. If the diagonal of the cube is perpendicular to the surface and $g > 0$, the projection of an edge and the projection of the electric field point in opposite directions, while at $g < 0$ their directions coincide (see Figs. 1 and 2). If one of the cube edges is normal to the surface and $g > 0$, the projection of the electric field coincides with the diagonal of the cube face, lying on the surface. If $g < 0$, one of the cube edges is directed along the projection of the electric field (see Figs. 3, 4). We shall designate these different geometric situations by $A^\pm$ and $B^\pm$. The letters $A, B$ indicate whether the diagonal or the edge of the cube coincides with the normal to the surface, while indices $\pm$ refer to the sign of the constant $g$.

The texture of a cubic liquid crystal can be characterized by one angle $\theta$, which can be chosen to be the angle between the normal and the diagonal of the cube in the cases $A^\pm$ and to be the angle between the normal and the edge of the cube in the cases $B^\pm$.

The coordinates of the triad in the case $A^+$ are written explicitly below:

$$
n^1\left[ \frac{1}{\sqrt{2}} \cos(\theta_0 - \theta), -\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}} \sin(\theta_0 - \theta) \right]
n^2\left[ \frac{1}{\sqrt{2}} \cos(\theta_0 - \theta), \frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}} \sin(\theta_0 - \theta) \right]
n^3\left[ -\sin(\theta_0 - \theta), 0, \cos(\theta_0 - \theta) \right]
$$

where $\theta_0 = \arctan\sqrt{2}$. We will not write out the other cases, as the generalization is obvious.

The angle between the diagonal of the cube and the field $E$ can be fixed arbitrarily until the Frederiks transition.

\begin{align*}
\text{Fig. 1.} & \quad \text{The arrangement of the cube and the electric field vector for } g > 0. \text{ L is the unit vector directed along the diagonal of the cube. } n^1, n^2, n^3 \text{ are the unit vectors directed along the edges of the cube. The normal to the surface coincides with the } z\text{-axis.}
\end{align*}

\begin{align*}
\text{Fig. 2.} & \quad \text{The arrangement of the main directions for } g < 0. \text{ The normal to the surface coincides with the diagonal } L \text{ of the cube.}
\end{align*}

\begin{align*}
\text{Fig. 3.} & \quad \text{The arrangement of the cube and the electric field vector for } g > 0. \text{ The normal to the surface coincides with the edge of the cube.}
\end{align*}

\begin{align*}
\text{Fig. 4.} & \quad \text{The arrangement of the cube and the electric field vector for } g < 0.
\end{align*}
We now consider this transition. First of all, the general form of the elastic distortion energy in a cubic liquid crystal must be found. Restricting ourselves to the effects which are quadratic over the tensor $\lambda_g e_{ij}$ as well as over the derivatives of this tensor, the cubic symmetry defines unambiguously the form of the elastic energy:

$$H_e = \frac{1}{2} K_1 \sum_{i=1}^{3} (\text{div } n_i)^2 + \frac{1}{2} K_2 \sum_{i=1}^{3} (n_i \cdot \text{rot } n_i)^2 + \frac{1}{2} K_3 \sum_{i=1}^{3} [n_i \times \text{rot } n_i]^2. \quad (6)$$

This equation differs from the usual expression for a nematic liquid crystal only in the summation over $i$. The values $K_i$ can be naturally called the Frank constants. The triad distribution corresponds to a minimum of the energy $H = H_1 + H_e$ at given boundary conditions.

The total energy in an electric field for the case $A^+$ has the form

$$H = \frac{1}{2} \int \left[ K_+ + K_- \cos 2(\theta - \theta_0) \right] (d\theta)^2 dz + \frac{1}{8} \lambda_g E^4 \int \left[ \frac{9}{8} + \frac{9}{8} \cos 2(\theta_0 + \theta_E - \theta) + \frac{3}{8} \cos 4(\theta_0 + \theta_E - \theta) \right] dz \quad (7)$$

where $\theta_E$ is the angle between the normal to the surface and the direction of the electric field,

$$K_+ = K_1 + \frac{1}{4}(K_2 + 3 K_3), \quad K_- = \frac{1}{4}(K_3 - K_2). \quad (8)$$

Assuming $\theta$ to be small everywhere we obtain approximately from (7)

$$H = \int \left[ \frac{1}{2} \left( K_+ - \frac{1}{3} K_- \right) \frac{d\theta^2}{dz} + \frac{1}{8} \lambda_g E^4 \left( \frac{9}{8} - \frac{9}{8} \cos 4(\theta_0 + \theta_E - \theta) + \frac{1}{4} b + ab \right) \right] dz \quad (9)$$

where

$$a = \sin 2(\theta_0 + \theta_E) + \frac{3}{2} \sin 4(\theta_0 + \theta_E)$$

$$b = 2[\cos 2(\theta_0 + \theta_E) + 3 \cos 4(\theta_0 + \theta_E)]. \quad (10)$$

The Frederiks transition takes place at $a = 0$, $b > 0$ only. Using equation (10) we find these conditions to be satisfied at $\theta_E = -\theta_0$ or $\theta_E = \pi/2 - \theta_0$. The critical value $E_c$ can be found by solving the linear Euler-Lagrange equation for the functional (9) and requiring $\theta$ to be zero at $z = 0$ and $z = d$:

$$E_c = \left( \frac{\pi}{d} \right)^{1/2} \left[ \frac{8(3 K_1 + K_2 + 2 K_3)}{3 \lambda_g b} \right]^{1/4}. \quad (11)$$

The constant $b$ has the value $b = 4$ at $\theta_E = -\theta_0$ and $b = 8$ at $\theta_E = \pi/2 - \theta_0$.

If $E < E_c$ and $\theta_E$ has the values indicated above, the solution $\theta = 0$ is valid. If $\theta_E$ deviates from the distinguished directions $-\theta_0$ and $\pi/2 - \theta_0$, the angle $\theta$ is nonzero at any value of $E$. The solution of the linear equation is

$$\theta = 2 \frac{a}{b} \frac{\sin (pz/2) \sin [(d - z)/2]}{\cos (pd/2)} \quad (12)$$

where

$$p^2 = 3 \lambda_g E^4 b [8(3 K_1 + K_2 + 2 K_3)]$$

and $d$ is the thickness of the cubic liquid crystal layer. The equation (12) is valid provided $0 < 1$ and $b > 0$. In particular, at small $E/E_c$ we have $\theta \sim (E/E_c)^4$; at $\theta$ close to $\theta_0$ we have $\theta \sim |\theta_0 - \theta_E| E_c/|E - E_c|$.

Omitting the calculations, we write down the values $E_c$ for another geometrical situation $A^-, B^-$:

The case $A^-$:

$$E_c = \left( \frac{\pi}{d} \right)^{1/2} \left[ \frac{8(3 K_1 + K_2 + 2 K_3)}{3 \lambda_g b} \right]^{1/4} \quad \theta_E = 2 \theta_0 \text{ or } \theta_E = 0, \quad b = -16/3.$$  

The case $B^+$:

$$E_c = \left( \frac{\pi}{d} \right)^{1/2} \left[ \frac{4(K_1 + K_3)}{g \lambda b} \right]^{1/4} \quad \theta_E = 0 \text{ or } \theta_E = \pi/2, \quad b = 4.$$  

So at $E \ll E_c$ generally, and at $E < E_c$ in the vicinity of some distinguished directions, the deviation from the orientation fixed by the walls is everywhere small. This yields the possibility of observing the Kerr effect at a given value $\theta_E$ and $\theta = 0$. The interaction-induced anisotropy of the dielectric tensor is uniaxial. However, in contrast to the case of an isotropic liquid, the anisotropy axis coincides with the electric field direction. We would like to note also that the difference of the refraction coefficients $n_e - n_o$ depends on the electric field direction.

The situation differs slightly if the usual Kerr effect $\delta e_{\theta \theta} \sim E_c E_p$ appears to contribute significantly. In this case one of the principal axes of the dielectric tensor is perpendicular to the plane containing the
diagonal of the cube, its edge and the electric field vector. The eigenvalue associated with this axis of the dielectric tensor $\varepsilon_1$ is a linear function of $\cos 2(\theta_0 + \theta_2)$.

Let us draw attention to a peculiarity of the Frederiks effect when the angle $\theta_E = \arctg \sqrt{2}$.

It is possible to define the constants $\lambda, g, K'$ ($K' = 3 K_1 + K_2 + 2 K_3$) which measure the critical field and the angular dependence of the dielectric anisotropy.

Here some estimates for $E_c$ are presented. Assuming the Franck constants to be $10^{-6}$ dyne as in a nematic liquid crystal and $\lambda \sim 1, g \sim E_{at}^{-2} \sim 4 \times 10^{-10}$ CGSE ($E_{at}$ is a characteristic atomic electric field) we obtain $E_c \sim 10^4$ V/cm for layer thickness $d \sim 10^{-1}$ cm.

Acknowledgments.

In conclusion, I express my gratitude to V. L. Pokrovsky for the statement of the problem and to E. I. Kats for useful discussions.

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