Acousto-optical effect induced by an incident ultrasonic beam in a normally oriented nematic liquid crystal layer

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The change of the orientation of a nematic liquid crystal layer following normal incidence of an ultrasonic beam, and the ensuing optical effect when the layer is placed between crossed polaroids and illuminated, are investigated theoretically. The crystal molecules are rotated as a result of the stationary liquid streams produced when ultrasound acts on the liquid layer. An expression is obtained for the transparency (which determines the part of the light flux passing through the layer). It is shown that the layer section spanned by the ultrasonic beam should be made transparent by the ultrasonic field; the results of the theory are compared with the experimental data.

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The effect of ultrasound on the structure of ordered layers of a nematic liquid crystal (NLC) was investigated in many papers (see the review¹). It was shown experimentally that when ultrasound is incident on an oriented NLC layer, the crystal optical axis, which coincides with the direction of alignment of the elongated molecules, is tilted away from the initial orientation, so that the optical properties of the layer as a whole are changed. In the absence of external action, a normally oriented NLC layer placed between crossed polaroids is opaque to the light wave that illuminates the layer along the crystal axis; tilting of the crystal molecules in an acoustic field makes the layer transparent. When the layer is illuminated at an angle to the axis, the action of the sound on the crystal alters the intensity of the transmitted light flux. In high-power ultrasound fields, the structure of the NLC becomes inhomogeous, and this leads to strong scattering of the light.

Clearing and the change of transparency of a normally oriented NLC layer in a sound field were considered theoretically in Refs. 2-8. An attempt to treat the acousto-optical effects as the acoustic analog of the Freedericksz transition^{2,3} was unsuccessful, since the obtained sound-intensity thresholds were higher by 2-3orders of magnitude than those used in experiment; similar results were obtained in Ref. 4, where the vibrohydrodynamic instability of the orientation of NLC was considered. Sripaipan⁵ proposed a mechanism for clearing an NLC layer with free ends, in which the interaction between the longitudinal oscillations produced as a result of the motion of the free ends of the layer when it is compressed and the transverse oscillations produces stationary liquid streams and, as a consequence, to rotation of the crystal molecules. The corresponding clearing theory⁸ agrees satisfactorily with the experimental data, but does not explain the bleaching of a layer with closed ends, which is investigated in most experiments. In Ref. 6 was considered the role of the streams that are produced in a layer because of radiation forces in an ultrasonic beam; the theoretical picture of the layer clearing agrees with that observed in experiment when the beam is focused on a region with dimensions of the order of the layer thickness. It is proposed in Ref. 7 that the molecule tilting is due to the

anisotropy of the sound absorption in the oriented liquid crystal: the molecules tend to tilt in such a way that the losses in the sound wave were decreased. This mechanism predicts the effect at large angles of incidence of the sound wave on the crystal (~ 1 rad).

In this paper we develop a theory of the acoustic-optical effect in the case of normal incidence of an ultrasound beam of restricted width on an NLC layer. The clearing mechanism consists in the following. When the sound wave passes through the NLC layer it is partially reflected from the solid boundary; a standing compression wave is established in the layer and has a width that coincides with the dimensions of the ultrasound beam. The compression gradients along the layer lead to longitudinal vibrations of the liquid particles. If the edges of the ultrasound beam are sharp enough, the compression gradients on the edges produce longitudinal waves that propagate both inside the beam and to the outside. If the intensity is in addition inhomogeneous over the beam cross section, longitudinal waves are excited also inside the beam. The time-averaged forces, which are proportional to the product of the velocity of the longitudinal oscillations and the compression, produce in the liquid a stationary flow in which the velocity gradients over the thickness of the layer tilt the crystal molecules. In the absence of a sound field, an NLC layer placed between crossed polaroids is opaque to a light wave normally incident on the layer. When the optical axis of the crystal is tilted, the rotation of the molecules in the layer results in an ordinary and extraordinary light waves. The superposition of the waves at the layer boundary determines the depolarization of the incident light wave passing through the second polaroid.

Let us consider thus a normally oriented layer of a nematic liquid crystal of thickness *h*. Assuming that the width of the ultrasound beam is so much smaller than the layer width that we can neglect in the calculation of the wave field in the layer the longitudinal waves reflected from the ends of the layers (since they are damped) we calculate the effect for an NLC layer of infinite width. We consider a two-dimensional problem in which all the particle vibrations, the propagation of the longitudinal waves, the streams, and the tilting of the molecules take place in the (x, z) plane; the z axis is directed along the normal to the layer and the x axis along the layer.

We choose the origin at the midpoint of the layer, so that the upper and lower boundaries of the layer have coordinates $z=\pm h/2$, while the boundaries of the ultrasound beam of width 2a have coordinates $x=\pm a$.

We confine ourselves to ultrasound frequencies ω such that the length of the sound wave in the nematic liquid crystal is much larger, and the length of the viscous wave much shorter, than the layer half-thickness, assuming the following inequalities to be satisfied

$$kh/2 \ll 1, \quad qh/2 \gg 1,$$
 (1)

where $k = \omega/c$ and $q = (\rho \omega/2 \eta)^{1/2}$ are the wave numbers in the acoustic and viscous layers, while ρ and c are the density and speed of sound in the nematic crystal. We consider only small molecule tilt angles $\varphi < 1$ in the sound field; this allows us to linearize the equations with respect to angle.

We calculate the effect by a perturbation method. From the linearized hydrodynamic equations we obtain the velocity of the liquid when the layer is compressed; retaining in the equations the terms quadratic in the velocities, we obtain the velocity of the stationary stream. Using the gradients of the stationary velocities we can determine the optical effects from the molecule angle rotation.

Bearing in mind the satisfaction of the obvious inequality $\omega \eta / \rho c^2 \ll 1$ in the considered frequency range $\omega \leq 10^8 \sec^{-1}$, we discard in the description of the longitudinal waves the viscous stresses compared with the elastic stresses; viscous effects are taken into account when the acoustic wave field is determined only by the presence of viscous waves that propagate from the boundaries of the layer along the z axis. Recognizing also that the considered compression amplitudes in the layer are much less than 5×10^{-3} , at which the interaction of the velocity field of the director lead to threshold effects,⁴ we regard the nematic crystal as an isotropic liquid with dynamic shear viscosity η corresponding to the indicated viscous waves in the nematic

$$\eta = \eta_1 + \frac{\alpha_2}{2} \left(1 - \frac{\gamma_2}{\gamma_1} \right),$$

where

$$\eta_2 = \frac{1}{2}(-\alpha_2 + \alpha_4 + \alpha_5), \quad \gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_3 + \alpha_2,$$

and α_i are the Leslie viscosity coefficients.⁹

We specify the compression in the layer in terms of the motion of its boundaries in the form

where v_0 is the amplitude of motion of the upper boundary, the function $\chi(x)$ describes the distribution of the amplitudes of the oscillations along the layer boundary, and consequently the distribution of the intensity of the ultrasound beam over its cross section. We choose for the calculation a unit-step distribution of the ultrasound intensity:

$$\chi(x) = \begin{cases} 1, & |x| \leq a \\ 0, & |x| > a \end{cases}.$$

The solution of the linearized equation of motion of a viscous liquid

$$\rho \frac{\partial^2 \mathbf{v}}{\partial t^2} + \eta \operatorname{rot} \operatorname{rot} \frac{\partial \mathbf{v}}{\partial t} - \rho c^2 \operatorname{grad} \operatorname{div} \mathbf{v} = 0$$

with boundary conditions (2) and with allowance for inequalities (1) is of the form

$$v_{z} = v_{\delta} \chi(x) \frac{2z+h}{2h} \cos \omega t,$$
$$v_{z} = \operatorname{Re} \left\{ i \frac{v_{\delta}}{2kh} [1-T(z)] [\exp\{(ik-\delta) (x+a)\} - \exp\{-(ik-\delta) (x-a)\}] e^{-iat} \right\}, \quad |x| \le a.$$

The function T(z) corresponds to viscous waves generated by a longitudinal wave on the boundary of the layer:

$$T(z) = \exp\{(1-i)q(z-h/2)\} + \exp\{-(1-i)q(z+h/2)\}$$

and causing damping of the longitudinal waves in a layer with a coefficient $\delta = k/2qh$. The velocity v_x at |x|>a is also represented as a sum of longitudinal waves propagating from the ends of the beam.

The equations for the velocity of the stationary flow v_{x_2} of the nematic and for the stationary rotation angle of the molecules φ_2 , under the condition

$$\partial/\partial x \sim k \ll \partial/\partial z \sim \pi/h$$

are obtained in the form

$$\frac{\partial^2 v_{zz}}{\partial z^3} = f(x, z), \quad K_3 \frac{\partial^2 \varphi_2}{\partial z^2} = \alpha_2 \frac{\partial v_{zz}}{\partial z} + (*), \tag{3}$$

where

η

$$f(x,z) = \rho \frac{\partial^2}{\partial x \partial z} \left(\langle v_x^2 \rangle - \langle v_z^2 \rangle \right) + \rho \left(\frac{\partial^2}{\partial z^2} - \frac{\partial^2}{\partial x^2} \right) \langle v_x v_z \rangle + (**).$$

The angle brackets $\langle \rangle$ denote time averaging, (*) denote terms of the form $\alpha \langle v \nabla \varphi \rangle$, while (**) denote terms of the form

$$\propto \frac{\partial^2}{\partial z^2} \langle \varphi \nabla v \rangle$$
 and $\propto \frac{\partial^2}{\partial z^2} \langle v \nabla \varphi \rangle$,

which make small contributions to v_{sz} and φ_2 (here φ is the molecule tilt angle in viscous waves); K_s is the Frank elastic constant.

From the system (3), under the conditions

$$v_{\alpha 2}|_{z=\pm h/2}=0, \quad \phi_2|_{z=\pm h/2}=0$$

and from the condition, in the form

$$\int_{-h/2}^{h/2} v_{x2} dz = 0$$

that the flow be closed, we obtain φ_2 :

$$\varphi_{z} = \frac{\rho v_{0}^{z} \alpha_{z}}{2 \cdot 2^{n} k q \eta_{z} K_{3}} \left\{ F_{z}(x) \left[\left(\frac{z}{h} \right)^{3} + \frac{1}{2} \left(\frac{z}{h} \right)^{2} - \frac{z}{4h} - \frac{1}{8} \right]$$
(4)
+ $F_{z}(x) (kh)^{z, s} \left(\frac{\rho c h}{2\eta} \right)^{0, s} \left[\frac{1}{60} \left(\frac{z}{h} \right)^{s} + \frac{1}{24} \left(\frac{z}{h} \right)^{4} - \frac{1}{120} \left(\frac{z}{h} \right)^{3} - \frac{1}{48} \left(\frac{z}{h} \right)^{2} + \frac{1}{960} \frac{z}{h} + \frac{1}{384} \right] \right\},$

where

$$F_{i}(x) = \left\{ \cos \left[k(a-x) - \frac{\pi}{4} \right] e^{\delta(x-a)} - \cos \left[k(a+x) - \frac{\pi}{4} \right] e^{-\delta(x+a)} \right\} \chi(x),$$

$$F_{2}(x) = \left\{ \sin \left[k(x+a) \right] e^{-\delta(x+a)} + \sin \left[k(x-a) \right] e^{\delta(x-a)} \right\} \chi(x).$$

The clearing of the layer following the tilt of the molecules is characterized by a transparency m, which is taken to mean the ratio of the light flux passing through the second polaroid to the light flux incident on the layer after going through the first polaroid. If the molecule tilt angle varies smoothly over the thickness of the layer, and if the slope is small, the transparency is given by a known equation¹⁰ in which $\hbar \varphi_2^2$ is replaced by

$$m = \sin^2 \left\{ \frac{\Delta n}{2} k_0 \int_{-h/2}^{h/2} \varphi_2^2 dz \right\} \sin^2 2\theta.$$

Here $\Delta n = n_{\parallel} - n_{\perp}$, while n_{\parallel} and n_{\perp} are the refractive indices of the light along and across the optical axis of the crystal, k_0 is the wave number of the light in vacuum; θ is the angle between the tilt plane of the molecules and the orientation of one of the polaroids. Integrating φ_2^2 with respect to z, we obtain the following expression for the transparency:

 $m = \sin^2 \{BN(x)\} \sin^2 2\theta, \tag{5}$

where

$$B = \frac{\Delta n}{840} k_0 h \frac{\rho v_0^4 \alpha_2^2 \eta}{k^2 \omega n_2^2 K_2^2}.$$

The function N(x) describes the distribution of the transparency along the NLC layer:

$$N(x) = F_1^2(x) - 3.15 \cdot 10^{-2} (kh)^{2,5} \left(\frac{\rho ch}{2\eta}\right)^{-6,5} F_1(x) F_2(x)$$

+2.9 \cdot 10^{-4} \frac{(kh)^5 \rho ch}{2\eta} F_2^2(x).

At |x|>a, the function $N(x) \equiv 0$, meaning that the clearing region is limited to the size of the ultrasound beam. A system of alternating dark and light fringes should be observed on the layer, and the distance between the centers of the light fringes is of the order of the sound wavelength. The distribution of the transparency of an NLC layer of thickness $h=10^{-2}$ cm at a frequency 1 MHz and a beam width 2a=2 cm is shown in Fig. 1.

In the experiments, the NLC layer is usually placed in water. Recognizing that the density and the sound velocity in the nematic liquid crystals and in water are close in value, we represent the amplitude of the vibrational velocity in the ultrasound beam incident on the layer in the form $V=v_0/2kh$; the parameter *B* is then given by

$$B = \frac{2}{105} \frac{\Delta n k_0 h^5 \omega \eta \alpha_2^2}{\rho c^4 \eta_2^2 K_3^2} I^2,$$



FIG. 1. Plot of the function N(x) at a = 1 cm, $h = 10^{-2}$ cm, and $\omega = 10^7$ sec⁻¹.

where $I = \rho V^2 c$ is the intensity of the ultrasound incident on the layer.

The theoretically predicted dependence of the transparency on the intensity of the ultrasound, namely m $\sim \sin^2(\text{const} \cdot I^2)$, or $m \sim I^4$ in the case of low intensities, agrees with the experimental data.^{5,11} The dependence of the ultrasound intensity corresponding to the threshold of observation of the effect, I_{thr} , on the layer parameters, have been investigated in a number of studies. To determine the analogous dependence in the theory, we take the threshold intensity to be the value of Icorresponding to a transparency $m = 10^{-2} \sin^2 2\theta$. At kh $\ll 1$ we have $N(x) \approx F_1^2(x)$. At large layer thicknesses, when $ka/2qh \leq 1$, the theoretically predicted dependence of I_{tbr} on the layer thickness, $I_{ttr} \sim h^{-2.5}$, is close to the experimental $I_{\rm thr} \sim h^{-2}$; the dependence of $I_{\rm thr}$ on the sound frequency ω is determined by the relation $I_{
m thr}$ ~ ω^{-2} . At small layer thickness, when $ka/2qh \gg 1$, the main effect should be observed near the edges of the ultrasonic beam and vanishes with increasing parameter ka/2qh.

We estimate now the effect numerically at a frequency 1 MHz with a=1 cm for an MBBA liquid-crystal having a thickness $h=10^{-2}$ cm and the following properties: η_2 = 1.03 P, $\alpha_2=0.78$ P, $\eta=0.25$ P, $\rho\approx1$ g·cm⁻³, c=1.5×10⁵ cm · sec⁻¹, $K_3=10^{-6}$ dyn, and $\Delta n=0.21$.⁹ The wave number k_0 is assumed equal to 10^5 cm⁻¹, and the angle θ is assumed equal to $\pi/4$. The value of I_{thr} in the central relation of the light fringes, where $N(x) \approx 4$, is I = 1.7 mW ° cm⁻², and the first maximum of the transparency is reached at $I_1=6.5$ mW · cm⁻². These values of I_{t-1} and I_1 are of the same order as the experimental data.^{6.11}

The presented calculations are valid for sound intensities I at which the molecule rotation is small, $\varphi_2 < 1$. From Eq. (4) for φ_2 the condition for the validity of the results in the form

$$I \leq 3 \frac{q\eta_2 K_3 c}{kh^2 |\alpha_2| \max|F_1(x)|}.$$

For the numerical example considered above, this inequality takes the form $I \leq 20 \text{ mW} \cdot \text{cm}^{-2}$.

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