Acousto-optical effect in oblique incidence of ultrasound on a nematic-liquid-crystal layer

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We consider the change of orientation of a nematic-liquid-crystal layer and the ensuing optical effect following oblique incidence of an ultrasound wave and ultrasound beam on the layer. The molecule rotation is due to velocity gradients of the stationary flow produced by the action of nonlinear boundary forces in a real liquid-crystal cell in a sound field. Incidence of a sound waveon the layer causes a homogeneous deformation of the crystal structure. The effect produced by incidence of an ultrasound beam is calculated in the region outside the beam. The calculation results are compared with experiment.

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§1. INTRODUCTION

The action of ultrasound on a nematic-liquid-crystal (NLC) layer can lead to a change of the molecule orientation, and to destruction of the crystal structure at high ultrasound powers. These structure changes lead in the first case to depolarization of the linearly polarized light wave incident on the NLC layer, or to strong scattering of the light in both the first and second case.[1]

The experimental investigations indicate that the extent of the action of the ultrasound on the crystal structure depends strongly on the ultrasound-wave incidence angle.[2] The action of ultrasound on an NLC layer at normal incidence of the sound wave was explained in Refs. 3–5 as being due to the onset in the layer of stationary streams proportional to the product of the rates of layer compression in the sound wave by the displacement velocity in the longitudinal wave propagating along the layer when the latter is inhomogeneously compressed or in the presence of free boundaries. The velocity gradients of the stationary stream along the layer thickness cause rotation of the molecules: this effect is optically observed at incident-ultrasound intensities 1 mW$\cdot$cm^{-2}. The action of ultrasound obliquely incident on an NLC layer was considered in Refs. 6–9. Dion[6-8] proposed that the rotation of the liquid-crystal molecules in a traveling wave is due to anisotropy of the absorption: the molecules tend to rotate so as to decrease the losses in the sound waves. Assuming in error that the sound is absorbed within distances shorter than the layer thickness, Dion obtained too high a result; a correct calculation predicts for this mechanism too small an effect that might be observed at a sound intensity larger by three orders of magnitude than those used in experiment, and which is masked in a real case by stronger effects. Candau *et al.*[9] considered the action of the radiation forces in an ultrasound beam obliquely incident on an NLC layer. The radiation forces produce in the liquid layer a stream that turns around outside the region subject to the action of the ultrasound; the stream velocity gradients cause rotation of the molecules. Far from the "sounded" region, the theory agrees qualitatively with experiment but, as will be shown below, the quantitative disparity is very large. At large beam dimensions, comparable with the dimensions of the liquid-crystal cell, and when an ultrasound wave is incident on the NLC layer, radiation forces that are homogeneous along the layer likewise do not lead to the onset of streams and to deformation of the structure.

In this paper we propose for the acoustooptical effect in oblique incidence of ultrasound a mechanism based on the analysis of the boundary forces concentrated in thin layers of thickness of the order of the length of the viscous wave near the boundaries of the NLC layer. These forces arise when sound acts on a liquid-crystal cell in which a nematic crystal is placed between solid plates that flank the NLC layer and set the orientation of the crystal molecules. The acoustic properties of the liquid crystal and of the plates are different, so that the incident sound wave produces in the NLC, beside the longitudinal waves, also two viscous wave propagating into the interior of the layer from its boundaries. The boundary forces, which are proportional to the time-averaged product of the vibrational velocities in the longitudinal and viscous waves, produce a stationary stream; the velocity gradients of the stationary stream deform the structure of the nematic crystal. The optical effect due to the molecule rotation is observed by passing a beam of light through the liquid crystal cell placed between crossed polaroids. At the normal molecule orientation and in the absence of a sound wave the system is opaque. When the optical axis of the crystal is inclined as a result of the molecule rotation, ordinary and extraordinary light waves are produced in the crystal, and their superposition at the exit from the layer determines the depolarization of the incident light wave and the fraction of the light flux passing through the second polaroid. The light scattering is increased at the same time, making it possible to observe the effect directly.

The calculation of the acoustooptical effect on the basis of the indicated mechanism was carried out for oblique incidence, on the liquid crystal cell, of an ultrasound wave and of an ultrasound beam whose dimensions are smaller than those of the cell. In both cases it is assumed that the initial orientation of the crystal is normal.

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§ 2. OBLIQUE INCIDENCE OF ULTRASOUND WAVE

The effect is calculated in the following manner. Assuming that the motions of the NLC-layer boundaries are given, we obtain the wave field in the layer; retaining in the hydrodynamic equations the terms quadratic in the velocity, we obtain the velocities of the stationary stream; from the molecule rotation angle in the stream we determine the optical effect. The motion of the NLC-layer boundaries following the incidence of an ultrasound wave on the cell is determined numerically with a computer.

We confine ourselves in the calculation to sound frequencies such that the length of the viscous wave in the layer is much less than the layer thickness h, assuming the following inequality

$$qh \gg 1, \tag{1}$$

where $q = (\rho\omega/2\eta)^{1/2}$ is the wave number in the viscous wave, and ρ and η are the density and dynamic viscosity of the nematic crystal. We consider only small molecule-rotation angles $\varphi < 1$; this allows us to linearize the equations in angle. In the description of the longitudinal waves we discard the viscous stresses compared with the elastic ones ($\eta\omega \ll \rho c^2$, where the c is the speed of sound). When the wave field in the layer is determined, the only viscous effects involved are only the viscous waves propagating from the layer boundaries in the normal direction. Recognizing also that the considered compression amplitudes in the layer are much smaller than the value 5×10^{-3} at which the crystal structure becomes unstable,[10] we regard the nematic crystal as an isotropic liquid with dynamic shear viscosity η corresponding to the indicated viscous waves in the nematic:

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

where $\gamma_1 = \alpha_3 - \alpha_2, \gamma_2 = \alpha_3 + \alpha_2$, and α_i are the Leslie viscosity coefficients.[11] The static shear viscosity in the stream along the layer is then equal to η_2.

We direct the z axis along the normal to the layer, choose the origin $z = 0$ on the lower boundary of the layer, and direct the x axis along the layer in the ultrasound-wave incidence plane. In this case the acoustic vibrations of the particles, the stationary flow, and the rotation of the molecules take place in the (xz) plane.

We specify the velocities v_x and v_z of the NLC-layer boundary vibrations in the form

$$v_\varkappa|_{z=\beta} = v_{\varkappa\beta}\exp(-i\omega t + ik_x x). \tag{2}$$

The subscript $\varkappa = x, z$ labels here the velocity components, and the index $\beta = 0$, h labels the layer boundary; $k_x = k\sin\theta$, where k is the wave number in the ultrasound wave incident on the layer, and θ is the incidence angle. The velocities $v_{\varkappa\beta}$ are compiled:

$$v_{\varkappa\beta} = v_{\varkappa\beta 1} + iv_{\varkappa\beta 2}. \tag{3}$$

The solution of the wave equation of the liquid motion in the layer

$$\rho\frac{\partial^2\mathbf{v}}{\partial t^2} + \eta\,\mathrm{rot}\,\mathrm{rot}\frac{\partial\mathbf{v}}{\partial t} - c^2\Delta\mathbf{v} = 0$$

with boundary conditions (2) is

$$v_x = \mathrm{Re}\left\{\frac{\exp\{i(k_x x - \omega t)\}}{\sin k_z h}\{i\,\mathrm{tg}\,\theta[v_{z0}\cos(k_z(h-z)) - v_{zh}\cos k_z z] + e^{(i-1)qz}[v_{x0}\sin k_z h - i\,\mathrm{tg}\,\theta(v_{z0}\cos k_z h - v_{zh})] + e^{(i-1)q(h-z)}[v_{xh}\sin k_z h - i\,\mathrm{tg}\,\theta(v_{z0} - v_{zh}\cos k_z h)]\}\right\},$$

$$v_z = \mathrm{Re}\left\{\frac{\exp\{i(k_x x - \omega t)\}}{\sin k_z h}[v_{zh}\sin k_z h + v_{z0}\sin(k_z(h-z))]\right\},$$

where $k_z = (\omega/c)\cos\theta$.

Recognizing that the time-averaged quantities are independent of x in this case, we obtain, following Ref. 12, an equation for the stationary-stream velocity v_2:

$$\eta_2\frac{d^3 v_{2x}}{dz^3} = \rho\frac{d^2}{dz^2}\langle v_x v_z\rangle + (*), \quad v_{2z} = 0. \tag{4}$$

the angle brackets denote here time averaging, and (*) stands for terms of the form $\alpha(d^2/dz^2)\langle\varphi\nabla v\rangle$ and $\alpha(d^2/dz^2)\langle v\nabla\varphi\rangle$, which make a small contribution to the solution of the equation.

Under the same conditions, the equation for the stationary molecule rotation angle φ_2 (Ref. 11) takes the form

$$K_3 d^2\varphi/dz^2 \approx \alpha_2 dv_{2x}/dz, \tag{5}$$

where k_3 is the Frank elastic constant.

Solving the system (4) and (5) with the conditions of the sticking of the liquid and preservation of the molecule orientation on the NLC-layer boundaries

$$v_{2x}|_{z=0,h} = 0, \quad \varphi_2|_{z=0,h} = 0,$$

as well as under the condition that the flow be closed, in the form

$$\int_0^h v_{2x}\,dz = 0,$$

we obtain φ_2:

$$\varphi_2 = \frac{\rho v_0^2\alpha_2 h}{2q\eta_2 K_3}\left\{Q\frac{z}{h} + (Q-2S)\left(\frac{z}{h}\right)^2 + (Q-S)\left(\frac{z}{h}\right)^3\right\},$$

where v_0 is the vibrational velocity in the incident sound wave; Q and S are parameters independent of v_0:

$$Q = \frac{1}{v_0^2}\left\{v_{z01}v_{x01} + v_{z02}v_{x02} + v_{z02}v_{x01} - v_{z01}v_{x02} + (v_{z02}v_{zh1} - v_{z01}v_{zh2} - v_{z01}v_{zh1} - v_{z02}v_{zh2})\frac{\mathrm{tg}\,\theta}{\sin k_z h} + (v_{z02}^2 + v_{z01}^2)\,\mathrm{tg}\,\theta\,\mathrm{ctg}\,k_z h\right\},$$

$$S = \frac{1}{v_0^2}\left\{v_{zh1}v_{xh1} + v_{zh2}v_{xh2} + v_{zh2}v_{xh1} - v_{zh1}v_{xh2} + (v_{zh1}v_{z02} - v_{zh2}v_{z01} - v_{z01}v_{zh1} - v_{z02}v_{zh2})\frac{\mathrm{tg}\,\theta}{\sin k_z h} + (v_{zh1}^2 + v_{zh2}^2)\,\mathrm{tg}\,\theta\,\mathrm{cth}\,k_z h\right\}.$$

The optical effect is characterized by the transparency m, taken to mean the ratio of the light flux passing through the polaroid to the flux incident on the NLC layer. If the molecule inclination angle varies smoothly, the transparency can be estimated using a known equation,[13] in which $\varphi^2 h$ is replaced by $\int_0^h \varphi^2 dz$:

$$m = \sin^2\left[\frac{\Delta n}{2}k_0\int_0^h \varphi^2\,dz\right]\sin^2 2\psi, \tag{6}$$

Here $\Delta n = n_\parallel - n_\perp$; $n_\parallel$ and $n_\perp$ are the refractive indices along and across the crystal axis, k_0 is the wave number of the light in vacuum, and ψ is the angle between the plane of polarization of the incident light and the (x, z) plane.

Integrating φ_2^2 with respect to z, we obtain the following expression for the transparency:

$$m=\sin^2\{B\Lambda\}\sin^2 2\psi,$$

where

$$B=\frac{\Delta n}{32}k_0h\left(\frac{\rho v_0^2\alpha_2 h}{K_3 q\eta_2}\right)^2,$$

Λ is a parameter determined by the cell data, the angle θ, and the frequency ω:

$$\Lambda=\frac{1}{105}Q^2+\frac{1}{70}QS+\frac{1}{105}S^2.$$

In experiment, the acoustooptical cell is usually placed in water. Since the sound speeds in water and in the liquid crystal are close, we express the parameter B in terms of the intensity $I=\rho v_0^2 c$ of the incident sound wave:

$$B=\frac{\Delta n}{32}k_0h\left(\frac{I\alpha_2 h}{K_3 q\eta_2 c}\right)^2.$$

We obtain Λ by computer calculation of the amplitude of the NLC-layer boundary motion due to incidence of a sound wave on a system consisting of a solid plate of thickness H_1, the liquid-crystal layer, and a solid plate of thickness H_2 (see, e.g., Ref. 14). The acoustooptical cell is assumed placed in water, and the plates are assumed to be of the same material. The boundaries of the plates are regarded as free to move in their own plane, meaning the inequality $\omega\eta \ll \mu$, where μ is the shear modulus of the plate. The values of Λ and the sound transmission coefficient in terms of the intensity D, at a frequency 1 MHz and at various NLC-layer thicknesses, are plotted vs. the angle θ in Fig. 1. We chose for the calculations the following values of H_1, H_2, Young's modulus in the longitudinal wave, the shear modulus μ, and the plate densities ρ_n, viz., $H_1=0.02$ cm, $H_2=0.3$ cm, $E=0.425\times 10^{12}$ g·cm^{-2}, $\mu=0.225\times 10^{12}$ g·cm^{-1}sec^{-2}, and $\rho_n=2.5$ g·cm^{-3}. The density and the sound velocity in the water and in the liquid crystal were assumed equal: $\rho=1$ g·cm^{-3} and $c=1.5\times 10^5$ cm·sec^{-1}.

The maximum values of Λ, and hence of the optical transparency of the cell, correspond to maximum acoustic transparency; this correlation was observed experimentally in Ref. 2. The small shift of the peak values of Λ on the θ scale with changing layer thickness h shows that the maximum-transparency angles θ are determined by the parameters of the boundary plates.

We now estimate numerically the effect at the frequency $\omega=1$ MHz, after determining the sound intensity $I_{\rm thr}$ corresponding to the arbitrary threshold transparency $m_{\rm thr}=\sin^2 2\psi$. For an MBBA liquid crystal with $\alpha_2=-0.78$ P, $K_3=0.7\times10^{-6}$ dyn, $c=1.5\times10^5$ cm·sec^{-1}, $\rho=1$ g·cm^{-3}, $\Delta n=0.12$, $\eta=0.25$ P, $\eta_2=1.03$ P (Ref. 11) and layer thickness $h=10^{-2}$ cm we obtain at $\Lambda\sim1$ the value $I_{\rm thr}\sim0.1$ mW·cm^{-2}. At peak values $\Lambda\sim10^2$ ($\theta\approx17°$) we obtain $I_{\rm thr}\sim10^{-2}$ mW·cm^{-2}.

The dependence of the transparency on the NLC layer

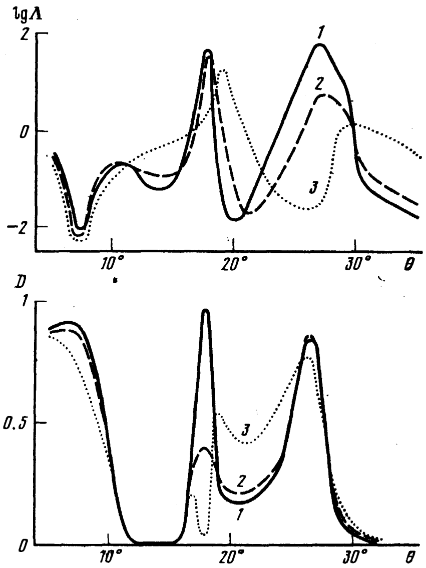

FIG. 1. Dependence of three optical (Λ) and acoustic (D) transparencies of an acoustooptical cell on the ultrasound incidence angle at $\omega/2\pi=1$ MHz. Curves 1, 2, and 3 correspond to h equal to 10^{-3}, 3×10^{-2}, and 10^{-2} cm, respectively.

thickness is different at different sound-incidence angles. At the first transparency peak we have $\Lambda\sim h^{-0.5}$ and $I_{\rm thr}\sim h^{-1.25}$, and at the second $\Lambda\sim h^{-1.6}$ and $I_{\rm thr}\sim h^{-0.7}$. These relations are closed to the experimental $I_{\rm thr}\sim h^{-1}$ obtained for oblique incidence of the sound.

§3. OBLIQUE ULTRASOUND BEAM INCIDENCE ON NLC LAYER

Neglecting multiple reflections of the ultrasound from the layer boundaries and the broadening of the angle spectrum of the beam on passing through the cell, we consider the action of ultrasound on an NLC layer in a cylindrical region of radius $r=a$. We assume the cylinder generatrices to be perpendicular to the layer, which is permissible at $a\gg h$. The equations for the velocity of the stationary stream and for the pressures take the form

$$\begin{aligned}&\eta_2\Delta \mathbf{v}_2=\nabla P_2+\rho\{\langle(\mathbf{v}\nabla)\mathbf{v}\rangle+\langle\mathbf{v}(\nabla\mathbf{v})\rangle\},\quad r<a,\\&\eta_2\Delta\mathbf{v}_2=\nabla P_2,\quad r>a,\quad \operatorname{div}\mathbf{v}_2=0,\end{aligned}\tag{7}$$

where $v(v_x, 0, v_z)$ are the vibrational velocity in the "sounded" region.

The radial derivative $\partial P_2/\partial r$ of the pressure (r is the distance to the region of the sounded region) changes jumplike at $r=a$. From (7) we obtain

$$\left.\frac{\partial P_2}{\partial r}\right|_{r=a+0}-\left.\frac{\partial P_2}{\partial r}\right|_{r=a-0}=-\rho\left.\left(\frac{\partial}{\partial x}\langle v_x^2\rangle+\frac{\partial}{\partial z}\langle v_xv_z\rangle\right)\right|_{r=a}\cos\chi,\tag{8}$$

where χ is the angle between the radius r and the x axis.

We consider the acoustooptical effect in a liquid-crystal region outside the ultrasound beam, at $r-a>h$. In this region the motion of the liquid is due to the gradients of a pressure that is constant over the layer thickness; the z-dependent harmonics of P_2 vanish when the distance from the sounded region increases to a distance $\sim h$. Neglecting small effects proportional to the square of the sound absorption coefficient δ, we obtain from (7) an equation for the pressure P_2 averaged over the layer thickness:

$$\Delta P_2 = 0, \quad r \neq a. \tag{9}$$

Averaging the condition (8) over the layer thickness and taking the sound absorption in the crystal into account, we obtain the connection between the external and internal pressure gradients:

$$\left.\frac{\partial P_2}{\partial r}\right|_{r=a+0} - \left.\frac{\partial P_2}{\partial r}\right|_{r=a-0} = (F_1+F_2)\cos\chi, \tag{10}$$

where $F_1 = \rho v_0^2 \delta D \sin\theta$ is the x component of the radiation forces (the action of these forces was considered earlier qualitatively in Ref. 9), and F_2 is a term connected with the action of the boundary forces, defined at $\delta = 0$ by

$$F_2 = -\frac{\rho}{2h}(v_{xh1}v_{zh1} + v_{xh2}v_{zh2} - v_{x01}v_{z01} - v_{x02}v_{z02}).$$

Equations (9) and (10) have a solution that is bounded at $r=0$ and $r \to \infty$ and is continuous at $r=a$, in the form

$$P_2 = \begin{cases} -{}^1/_2(F_1+F_2)\, r\cos\chi, & r<a \\ -{}^1/_2(F_1+F_2)\dfrac{a^2}{r}\cos\chi, & r>a \end{cases}. \tag{11}$$

In the outer zone, where $\partial^2/\partial r^2 \sim 1/r^2 \ll \partial^2/\partial z^2 \sim 1/h^2$, we have $\Delta \approx \partial^2/\partial z^2$. Substituting P_2 from (11) in the second equation of (7) and solving it under the condition that the liquid sticks at the layer boundaries, we obtain the flow-velocity components v_{2x} and v_{2y}:

$$v_{2x} = \frac{(F_1+F_2)a^2}{\eta_2}\frac{\cos 2\chi}{4r^2}z(h-z),$$

$$v_{2y} = \frac{(F_1+F_2)a^2}{\eta_2}\frac{\sin 2\chi}{4r^2}z(h-z).$$

Solving the equation for the molecule rotation angle

$$K_3\frac{\partial^2\varphi_2}{\partial z^2} = \alpha_2\frac{\partial|\mathbf{v}_2|}{\partial z}$$

at a constant orientation of the molecules at the layer boundaries, we obtain

$$\varphi_2 = \frac{(F_1+F_2)a^2\alpha_2}{24\eta_2K_3r^2}(3z^2-2z^3-z).$$

The optical effect produced when linearly polarized light is passed through the NLC layer is determined from the phase difference d between the ordinary and extraordinary light waves at the exit from the crystal. According to (6), d is equal to

$$d = \frac{\Delta n}{2}k_0\int_0^h \varphi^2\,dz \approx \frac{\Delta n\cdot k_0\alpha_2^2}{2{,}4\cdot 10^5\eta_2^2K_3^2}\frac{(F_1+F_2)^2h^7a^4}{r^4}. \tag{12}$$

Since d is independent of χ, at a sufficiently high incident-sound intensity there should be observed outside the sounded region a system of light and dark circular fringes corresponding to the values $d_p = \pi p/2$ ($p = 1, 2, 3, \ldots$), and a dark cross determined by the orientation of the crossed polaroids. This picture was observed experimentally in Ref. 9, where the acoustooptical effect was investigated for oblique incidence of an ultrasound beam at an angle corresponding to the maximum acoustic transparency of the layer ($D \approx 1$).

For a quantitative comparison of the results with the experimental data, we estimate the ratio of the radius R_1 of the first dark ring to the radius a of the sounded region. From (12) we obtain

$$\frac{R_1}{a} = \left\{\frac{\Delta n\cdot k_0h^7\alpha_2^2(F_1+F_2)^2}{2{,}4\cdot 10^5\pi\eta_2^2K_3^2}\right\}^{1/4}. \tag{13}$$

At the frequency $\omega/2\pi = 2.8$ MHz and at a sound intensity $I = 40$ mW $\cdot$ cm^{-2} experiment yielded for a PCB liquid-crystal layer $h = 0.02$ cm thick, at the maximum of the acoustic transparency, the ratio $R_1/a = 3$ to 5 (Ref. 9). To estimate R_1/a from Eq. (13) we put $\rho = 1$ g $\cdot$ cm^{-3}, $\delta = 0.1$ cm^{-1} (Ref 15), $c = 1.5\times 10^5$ cm $\cdot$ sec^{-1}, $\alpha_2 \approx \eta_2$, $K_3 = 10^{-6}$ dyn, $k_0 = 10^5$ cm^{-1}, and $\Delta n = 0.15$. In this case F_1 equals 0.13 g $\cdot$ cm^{-3} $\cdot$ sec^{-2}. At the maximum acoustic transparency of the layer we can estimate the order of magnitude of $|F_2|$ from the relation $|F_2| \approx \rho v_0^2/2h$. This estimate agrees well with the numerical calculation of F_2 for the cell parameters considered in §2. At $\theta \approx 25°$ and $I = 40$ mW $\cdot$ cm^{-2} the value obtained was $F_2 = 128$ g $\cdot$ cm^{-2}. The theoretical value $F_2 = 128$ $R_1/a = 4.5$ agrees with the experimental data.

A comparison of the values of F_1 and F_2 shows that the acoustoelectric effect for an ultrasound beam incident on an NLC layer is determined by the action of the boundary forces in the sounded region. The radiation forces considered in Ref. 9, while leading to qualitative agreement between the picture of the experimentally observed effect, cannot explain it quantitatively: the ratio $R_1/a = 0.14 < 1$ obtained for the radiation forces in the example considered is meaningless.

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