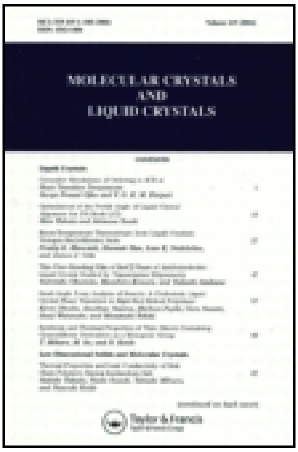

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# The Orientational Instability of Nematic Homeostropic Layers under Osciliatory Shear

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# THE ORIENTATIONAL INSTABILITY OF NEMATIC HOMEOTROPIC LAYERS UNDER OSCILLATORY SHEAR

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**ABSTRACT** Using an acoustooptical method we show that under plane oscillatory shear the director acquires a stationary inclination $\theta_c$. This phenomena have a threshold character. The shear amplitude threshold $a_1$ of the emergence of $\theta_c$ does not depend on the excitation frequency in the range $50<f<500$ Hz. We found the second threshold $a_2>a_1$, when the director motion is elliptical. In this case the director is out of in the third dimension. It has been shown, that the threshold $a_2$ is also independent on the shear frequency.

## INTRODUCTION

It is well known[1] that for the NLC with $\alpha_3/\alpha_2 \geq 0$ and a linear flow with large gradients $\partial v/\partial z$, in homeotropically aligned nematic layers the director tends to align in the flow plane at angle $\theta = \pm\tan^{-1}(\alpha_3/\alpha_2)^{1/2}$. For oscillatory shear flow the that becomes more complicated due to the amergence of a stationary component of the director's inclination along the oscillatory shear plane. The origin of this phenomenon may be related with a slight ellipticity of the shear plate oscillation[2]. However, other explanations of such behavior may be suggested[3], including the instability in the in shear plane motion and the transition to the out-of-plane director motion[4].

The aim of this paper is a detailed experimental investigation of the formation of the stationary angle in director's inclination; we also intendent to study the effect of the out-of-plane director motion at large shear amplitudes.

## EXPERIMENTAL

The case has been studied where a linear polarized monochromatic light wave is incident perpendicularily on the layer of a uniaxial nematic liquid crystal (NLC), and the wave vector of incident light **k** forms angle θ with the optical axis of NLC.

Birefringence Δn may be determined with an accuracy to of the order of $\sin^4\theta$ as[5]:

$$\Delta n = n_e - n_o = \frac{a}{2}\sin^2\theta + \frac{3a^2}{4}\sin^4\theta,$$

where $a = \dfrac{n_o(n_e^2-n_o^2)}{n_e^2}$, $n_o$ - is the refractive index of an ordinary wave, $n_e$ - extraordinary light wave. Accordingly, intensity I of the transmitted light through the layer is equal to[6]:

$$I = I_o \sin^2 2\phi \sin^2(\delta/2), \qquad (1)$$

where $I_o$ is the intensity of light incident on the polarizer, φ is the angle between the polarization plane of a polarizer and an optical axis of NLC. The phase difference between ordinary and extraordinary waves with an inhomogeneous distribution of the director over the sample thickness will be equal to:

$$\delta = \frac{2\pi}{\lambda}\int_{-h/2}^{h/2} \Delta n(z)\, dz\ , \qquad (2)$$

where λ is the length of the incident light monochromatic wave, h is the layer thickness of LC.

Now, let us make an assumption about angle θ. It is supposed that $\theta = \theta_c + \theta_t$, where $\theta_c$ is the permanent inclination of the director relative to the normal of the NLC cell and $\theta_t$ is a time-dependent part of director inclination.

Then, let us assume that $\theta_c$ and $\theta_t$ are small. Having decomposed $\langle \sin^2\theta \rangle$ in the vicinity of $\theta_c$, we get the following value for $\langle \Delta n \rangle$:

$$\Delta n = \frac{b}{2} \left[\langle \sin^2\theta_c \rangle + \langle \sin 2\theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle\right],$$

where $\langle\ \rangle$ is an averaging along coordinate z, $b = \frac{a}{2} + \frac{3}{4}a^2$. The latter expression retains its character at $\theta_c = 0$.

Expression (1) in this case will have the form (at $\phi=45^\circ$):

$$I = I_o \sin^2\left[\frac{\pi d b}{\lambda}\left(\langle \sin^2\theta_c \rangle + 2\langle \sin\theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle\right)\right]. \quad (3)$$

Let us denote $\langle \sin^2\theta_c \rangle = A$, $2\langle \sin\theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle = B$, $\frac{\pi d b}{\lambda} = C$.

Further transformation will bring about the following result:

$$I = I_o\Big\{\sin^2 C\langle \theta_c^2 \rangle + C(2\langle \theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle) \sin 2C\langle \theta_c^2 \rangle +$$
$$+ C^2(2\langle \theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle)^2 \cos^2 C\langle \theta_c^2 \rangle\Big\}. \quad (4)$$

From (4) it follows that besides constant component $I_c$, the optical signal contains a whole spectrum of $I_{if}$ harmonics which reflect the dynamics of the director motion in plane ZV formed by the cell normal and the shear wave vector.

Next, we will analyze the acoustooptical effects observed in NLC layer. With reference to[2], it may be tentatively assumed that the director's time-dependent component $\delta n(t)$ is

$$\delta n(t) = \frac{\rho v_o}{\eta_1 q} \exp[2^{3/2} q(z-\delta)] \cos(2^{3/2} q(z-\delta)) \sin\omega t$$

$$\text{or} \quad \delta n(t) = \theta_t(t) = \theta_o \exp[-\lambda z] \cos\lambda z \sin\omega t. \quad (5)$$

Introducting expression (5) into (3) we obtain:

$$I = I_o\{\sin^2 C\langle \sin^2\theta_C \rangle + C[2\langle \sin\theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle] *$$
$$* \sin 2C\langle \sin^2\theta_c \rangle + C^2[2\langle \sin\theta_c * \theta_t \rangle + \langle \theta_t^2 \rangle]^2 *$$
$$* \cos^2 C\langle \sin^2\theta_c \rangle\}. \quad (6)$$

And assuming that $\theta_t = \theta_o \sin\omega t$, then $[\theta_o = \theta_o(a, v)]$, let us single out constant and variable components of the optical signal, restricting our interest to $I_c'$, $I_{1f}'$, $I_{2f}'$. On the whole, superior harmonics are of little interest to our study.

Thus, using expression (5) and taking into consideration that at this frequency of disturbance $qd<1$, let us average $\theta_1$ and $\theta_1^2$ along cell thickness h, the ensuing result will be:

$$I'_c = I_c/I_0 = \{\sin^2 C\langle\sin^2\theta_c\rangle + \frac{C\theta_0^2}{2}\sin 2C\langle\sin^2\theta_c\rangle + 2C^2\langle\sin\theta_c * \theta_0\rangle^2 \cos^2 C\langle\sin^2\theta_c\rangle + \frac{3}{8}C^2(\theta_0^2)^2 * \cos^2 C\langle\sin^2\theta_c\rangle\}, \quad (7)$$

$$I'_{1f} = I_{1f}/I_0 = \{2C\langle\sin\theta_c * \theta_0\rangle \sin 2C\langle\sin^2\theta_c\rangle + 3C^2\langle\sin\theta_c * \theta_0\rangle * (\theta_0^2)\cos^2 C\langle\sin^2\theta_c\rangle\}\sin\omega t, \quad (8)$$

$$I'_{2f} = I_{2f}/I_0 = \{-\frac{C\theta_0^2}{2}\sin 2C\langle\sin^2\theta_c\rangle - 2C^2\langle\sin\theta_c * \theta_0\rangle^2 * \cos^2 C\langle\sin^2\theta_c\rangle - \frac{C^2\theta_0^4}{2}\cos^2 C\langle\sin^2\theta_c\rangle\}\cos 2\omega t, \quad (9)$$

Now, let us consider another experimental situation, which makes it possible to define the character of director motion under the shear excitation, i.e. we are interested to know whether the director moves in the oscillation plane, formed by the normal of the cell and the oscillating direction of the movable plate or whether it moves into a third dimension. Figure 1 illustrates a situation where the director is moving in the ZY plane, where Y coincides with the direction of the plate, and Z coincides with the normal to the cell plane. Such motion is characterized by angle $\theta$ (the angle of the director deviation from the state of equilibrium), the deviation of the ZX-plane is characterized by angle $\alpha$.

Therefore, in contrast to the previous case, the director has three components. In view of the latter circumstance, let us analyse the optical response in this state.

Let us examine expression (1). In general, it should look like

$$I = I_o \sin^2 2(\phi - \phi') \sin^2 \delta/2, \quad (10)$$

where $\phi'$ is the director exit angle from oscillation plane

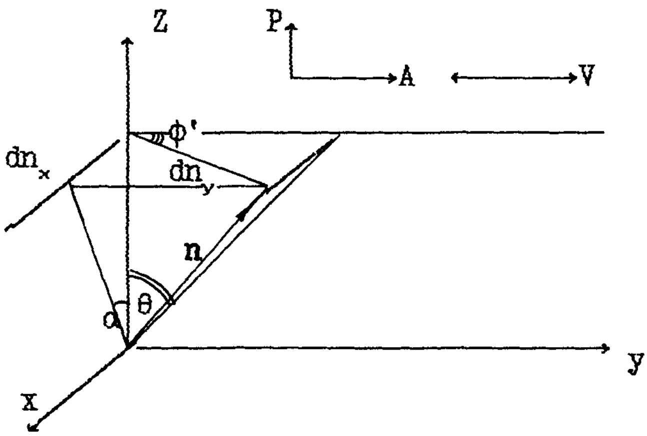


FIGURE I. Possible director motion in the oscillation plane.

ZY.

Since $\phi$ in this case (experimental situation, figure 1, $\phi$=90°, expression 10) assumes the following features:

$$I = I_o \sin^2 2\phi' \sin^2 \delta/2 \quad (11)$$

Let us denote components $\delta n_x$ and $\delta n_y$ in the forms of:

$$\begin{cases} \delta n_x = \delta n_x^o \sin\omega t, \\ \delta n_y = \delta n_y^o \sin(\omega t + \Phi), \end{cases} \quad (12)$$

where $\delta n_x^o = \sin \alpha$, $\delta n_y^o = \sin \theta$, whereas $\Phi$ is the phase difference between $\delta n_x$ and $\delta n_y$. Let us use formula $tg\ \phi' = \frac{\sin\alpha * \sin\omega t}{\sin\theta * \sin(\omega t + \Phi)}$ in (II). Then, taking into account (I2), we obtain:

$$I = \frac{4\ I_o \sin^2\alpha/\sin^2\theta * \sin^2\omega t/\sin^2(\omega t+\Phi)}{(1+\sin^2\alpha/\sin^2\theta * \sin^2\omega t/\sin^2(\omega t+\Phi))^2} \sin^2\delta/2. \quad (I3)$$

The case, where $\Phi$=0° according to[4] does not realize. So, let us consider the case where the phase difference is $\Phi=90^\circ$. After some transformations, expression (13) may be reduced to:

$$I = 1/2\ I_0\beta^2[I_1 - I_2\cos 2\omega t + I_3\cos 4\omega t]\ \sin^2\delta/2, \qquad (14)$$

here $\beta^2=\sin^2\alpha/\sin^2\theta$, $\gamma^2 = (\sin^2\theta-\sin^2\alpha)/\sin^2\theta$,
where

$$I_1/I_0 = 1/2\ \beta^2[1 + \gamma^2 + 5/16\gamma^4] = A, \qquad (15)$$

$$A = 1/2\ \beta^2[1 + \gamma^2 + 5/16\gamma^4], \qquad (16)$$

$$I_2/I_0 = -1/2\ \beta^2[1/2\gamma^2 + 1/4\ \gamma^4]\ \cos 2\omega t = B\ \cos 2\omega t, \qquad (17)$$

$$B = 1/4\ \beta^2[\gamma^2 + 1/2\gamma^4], \qquad (18)$$

$$I_3/I_0 = -1/2\ \beta^2[1 + \gamma^2 + 1/4\gamma^4]\ \cos 4\omega t = M\ \cos 4\omega t, \qquad (19)$$

$$M = 1/2\ \beta^2[1 + \gamma^2 + 1/4\gamma^4] \qquad (20)$$

From (3) we have $\sin^2\delta/2 = D - F\cos 2\omega t + L\cos 4\omega t$. Then we get the following components of the optical signal:

$$I''_c = I_c/I_0 = 1/2\ (2AD + BF - ML), \qquad (21)$$

$$I''_{2f} = I_{2f}/I_0 = 1/2\ (MF - MD - BL - 2BD - 2AF)\cos 2\omega t, \qquad (22)$$

$$I''_{4f} = I_{4f}/I_0 = 1/2\ (BF - BL - MD + 2AL)\ \cos 4\omega t. \qquad (23)$$

Thus, studying amplitude dependencies of values (21-23), it is possible to define the character of the director motion under the impact of a shear wave. In this case it may be supposed that having reached a certain threshold $a_2$ and still oscillating in plane ZV, the director leaves the oscillation plane and begins elliptical motion.

The system used for the acoustooptical measurements was constructed on the basis of a polarized optical microscope and shown in figure 2. Measurements were taken in the sound-frequency range of 20...20000 Hz. The nematic layers of a liquid crystal with homeothropic or quasi-homeothropic orientations were studied in our experiments. Metallic chromium was deposited on the glass

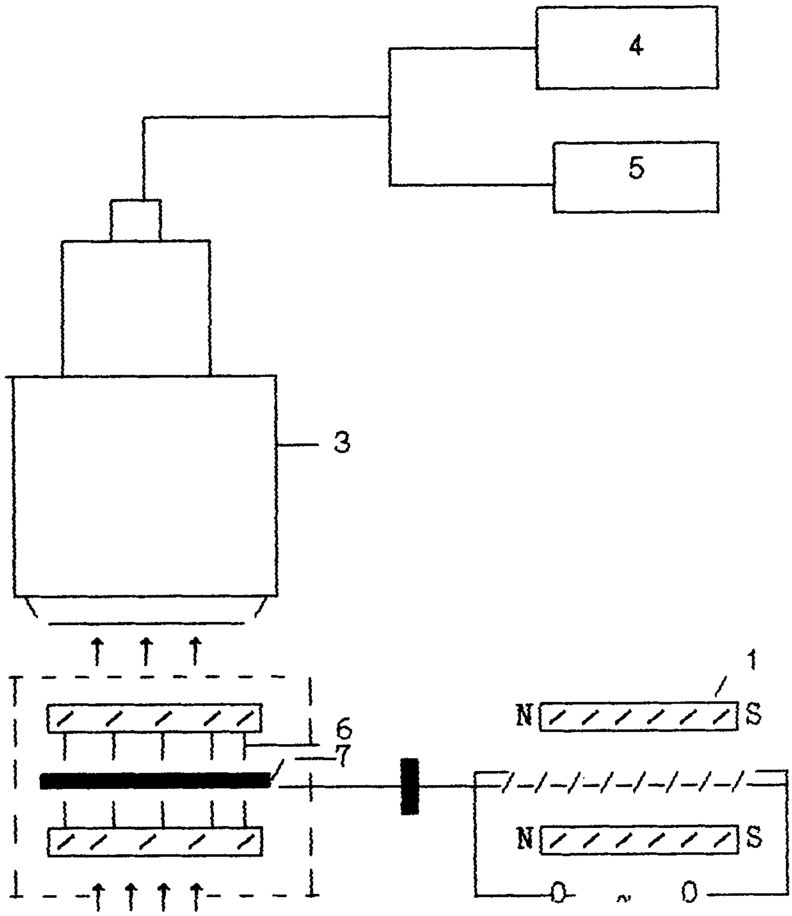


FIGURE 2. The set up of a experimental unit for study acoustooptical effects. 1 - source of sound oscillations, 2 - microscopic stage, 3 - polarizing microscope with a photometric adapter, 4 - DC microvoltmeter, 5 - selectiv amplifier, 6 - LC-sample, 7 - movable slide.

substrates to provide a required orientation. We used the liquid crystal MMBA. A NLC cell consisted of two semitransparent glass plates. Between them was placed a thin glass slide 150 μm thick with a similar coating deposited on both sides; the slide was oscillating in its own plane.

Shear oscillations in a NLC sample where stimulated by an electronic vibrator, its membrane was connected to glass slide by means of glass waveguide; the amplitude was measured optically with a microscope. For this purpose, a line (about 10 μm thick) was drawn on the movable slide. The amplitude was measured by the degree of the line smearing. Simultaneously, oscillation amplitude of the movabel thin slide were measured by an inductive method, its results were used to calibrate a scale a = a(U) where U stands for voltage in the induction coil.

## RESULTS AND DISCUSSION

Let us consider the action of a low frequency shearing wave (f=100 Hz) on a NLC layer (h ≃ 20 μm). Periodic change in the local properties of an oriented homeotropic layer induced change in the optical response of the LC cell.

Figure 3 shows experimental dependencies of $I'_c(a)$, $I'_{1f}(a)$, $I'_{2f}(a)$. $I'_c$ displays a non-linear growth and then acquires an oscillating character. These oscillations emerge as a result of the phase difference according to equation (1). The emergence of the minimum is also linked to such phase difference: $\delta = 2\pi n$, whereas the maxumum is $\delta = (2n + I)\ \pi$. In fact, oscillations in optical signal $I'_c(a)$ indecates a constant inclination angle ($\theta_c$).

Solving a system of equations connecting two experimental relationships $I'_c(a)$ and $I'_{2f}(a)$ (7-9), we have obtained functions $\langle \sin^2\theta_c \rangle(a)$ and $\langle \theta_o^2 \rangle(a)$ of shearing amplitude a. Curves $\langle \sin^2\theta_c \rangle(a)$ and $\langle \theta_o^2 \rangle(a)$ are plotted in Figure 4 They show that the emergence of $\langle \sin^2\theta_c \rangle(a)$ has a threshold character, whereas $\langle \theta_o^2 \rangle(a)$ reaches its maximum at certain values of amplitude a, then it begins to decrease.

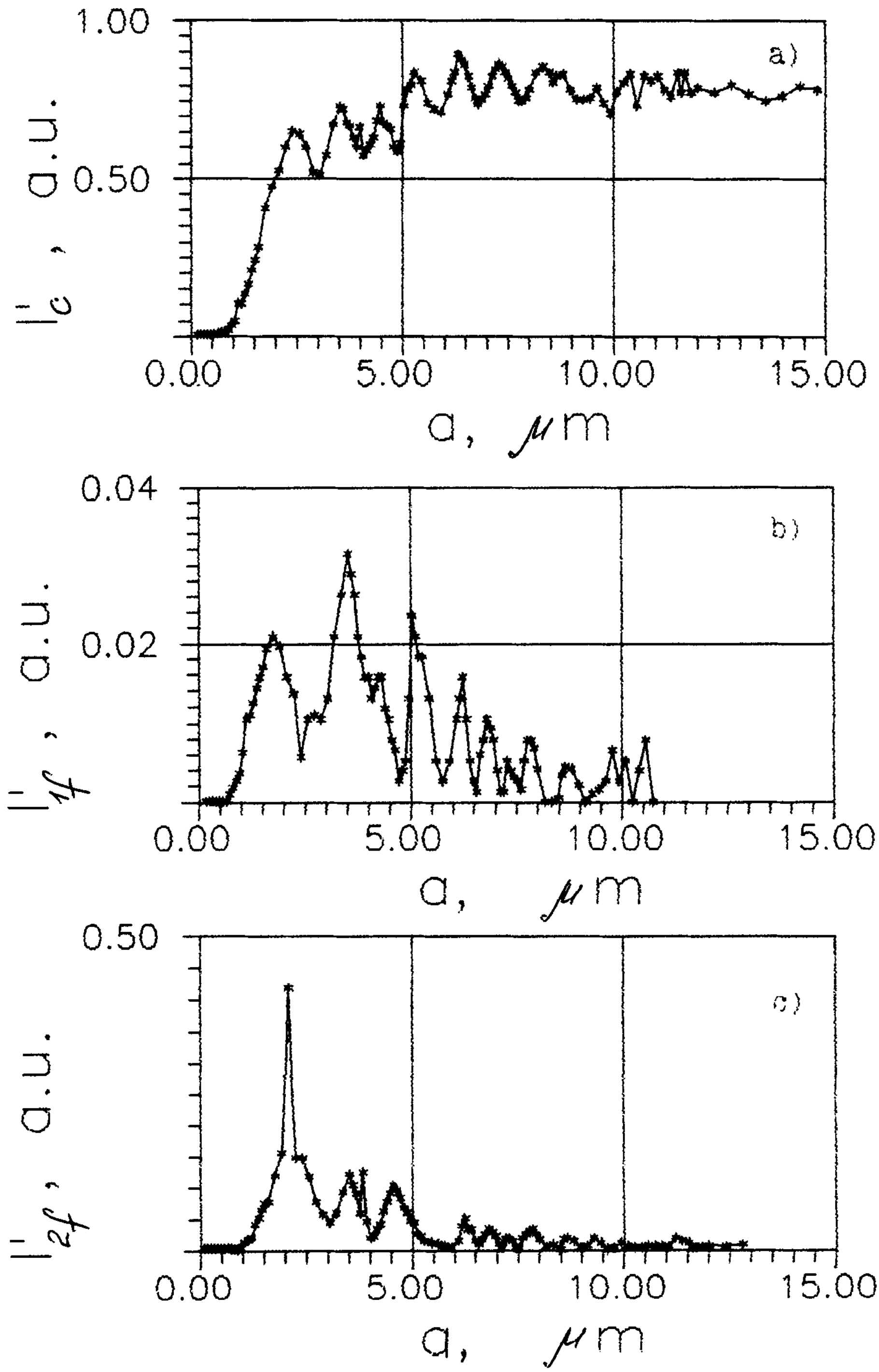


FIGURE 3. Dependencies of a) $I'_c$, b) $I'_{1f}$, c) $I'_{2f}$ on the shearing amplitude a.

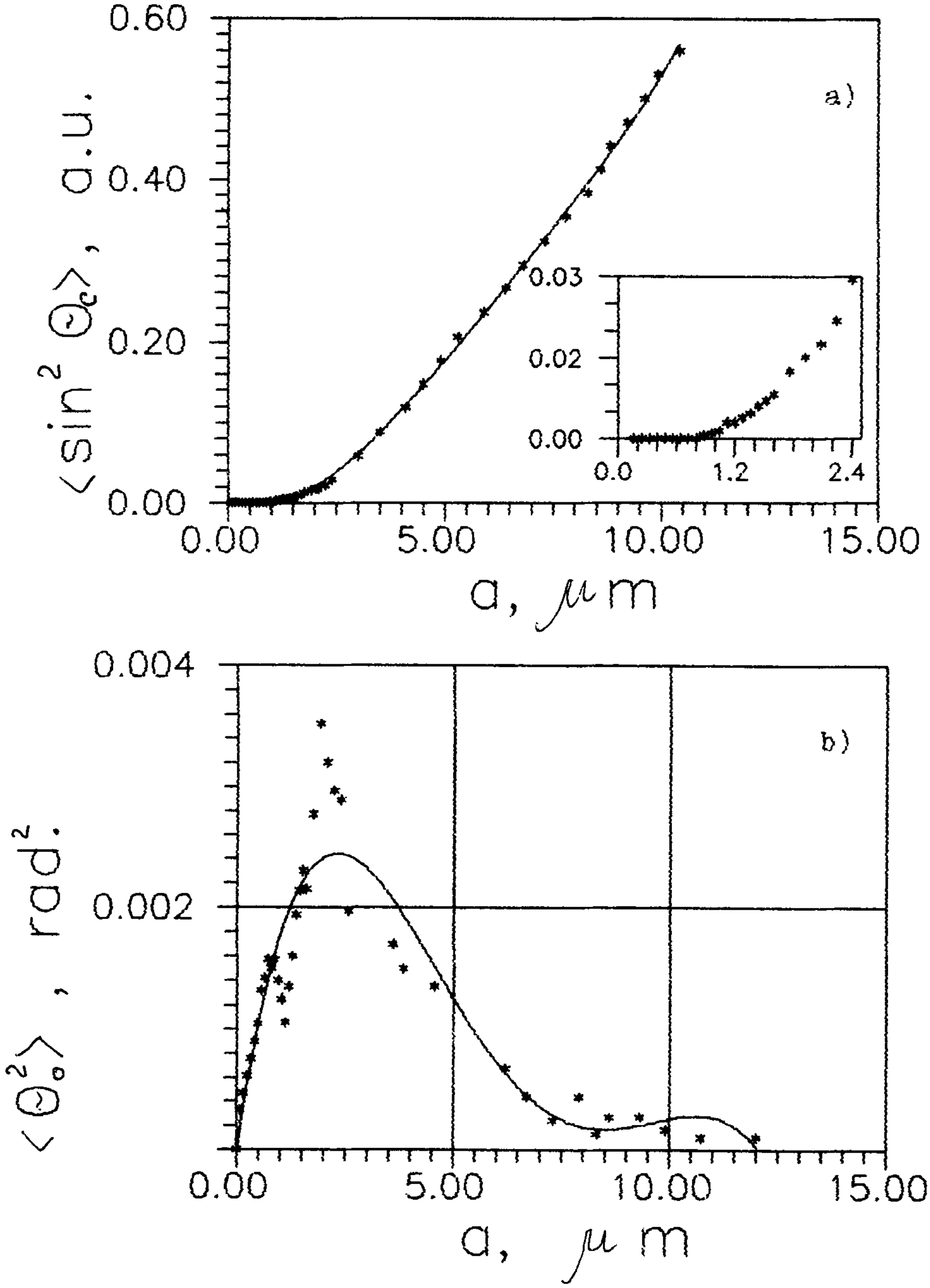


FIGURE 4. Dependences a) $\langle \sin^2 \theta_c \rangle$, b) $\langle \theta_0^2 \rangle$ on the shearing amplitude a.

It is apparently due to the fact that the director's stationary inclination angle tends to decrease dynamic susceptibility of the the LC layer. Further growth in the shearing amplitude leads to the formation of periodic structures of the "rolls" type.

It should be noted that the oscillating part of the director tends to decrease to zero. Further growth of the shearing amplitude leads to the destruction of periodic structures, so that macroscopic streams are formed and the system passes over to a mode of turbulization and dynamic chaos.

Thereby, we have been able to find that in condition of a periodic shear, when a certain threashold is reached, the director acquires a stationary inclination. In fact, determining the type of such an orientational transition in the field of a shear wave may be reduced to the study of a frequency dependence of threshold value $a_1$ and the emergence of stationary inclination of the director. The threshold amplitude of the stationary inclination was measured at the emergence of $I'_{1f}$ because $I'_{1f}$ $\langle \sin\theta_c * \theta_D \rangle \sin \omega t$. If $a_1(f)$=const, then, in this case it is possible to consider the effect in terms of an acoustic analogue of the Fredereecks transition. Figure 5 presents a dependence of threshold $a_1(f)$ on shear frequency f of the oscillating plates. This dependence demonstrates that in the frequency range under study i.e. $50<f<500$ Hz, $a_1$ does not depend on the frequency.

To study the effect of azimutal director instability, let us coincide the polarizer with the direction of shear V. In this configuration the sample can acquire bleaching if the director leaves plane ZV. Indeed, in this case, a weak but sufficiently stable signal is registered at the double frequency of excitation. It means that the director, while oscillating in the ZV-plane, reaches another threshold - $a_2$ and leaves the above mentioned plane. According to expression (21-23), the registered optical signal should also display a constant ($I_c''$) and the fourth harmonic ($I_{4f}''$), which have actually been registered. Figure 6 shows dependencies $I_c''(a)$, $I_{2f}''(a)$, $I_{4f}''(a)$ demonstrating that the components of the optical signal grow in a non-linear manner and possess a threshold character.

From experimental data on $I_{2f}''(a)$ it is possible to calculate the elliptical parameter - $\beta^2 = \frac{\sin^2\alpha}{\sin^2\theta}$ as a function of shear amplitude. Figure 7a at shows that function $\beta^2(a)$ develops in a threshold manner, at first it increases to a certain maximum, then decreases. These data make it possible to estimate angle $\alpha$ at which the director leaves the oscillation plane $\sin^2\alpha = \beta^2 \langle\theta^2\rangle$, since $\theta^2$ is small, then $\sin^2\theta \sim \theta^2$.

Dependence $\alpha(a)$ is shown in figure 7b it also has a threshold character and actually demonstrates, that under the impact of periodic shear excitation, the director oscillations reaches a certain threshold value of the amplitude, leaves its original plane of motion and starts moving in space on an elliptical trajectory. Measurements of frequency dependency of the director's threshold exit from plane ZV (Figure 5) show that threshold $a_2$ does not depend on the excitation frequency. The latter conclusion is confirmed by other studies.[4]

## CONCLUSION

Thus, it has been established that there exist two thresholds of orientational instability of homeotropically oriented nematic liquid crystal in shear wave field. The first $a_1$ is connected with the formation of a stationary angle of director inclination $\theta_c$. The second $a_2 > a_1$ is related to the director exit out of the plane of shear oscillation into the third dimension.

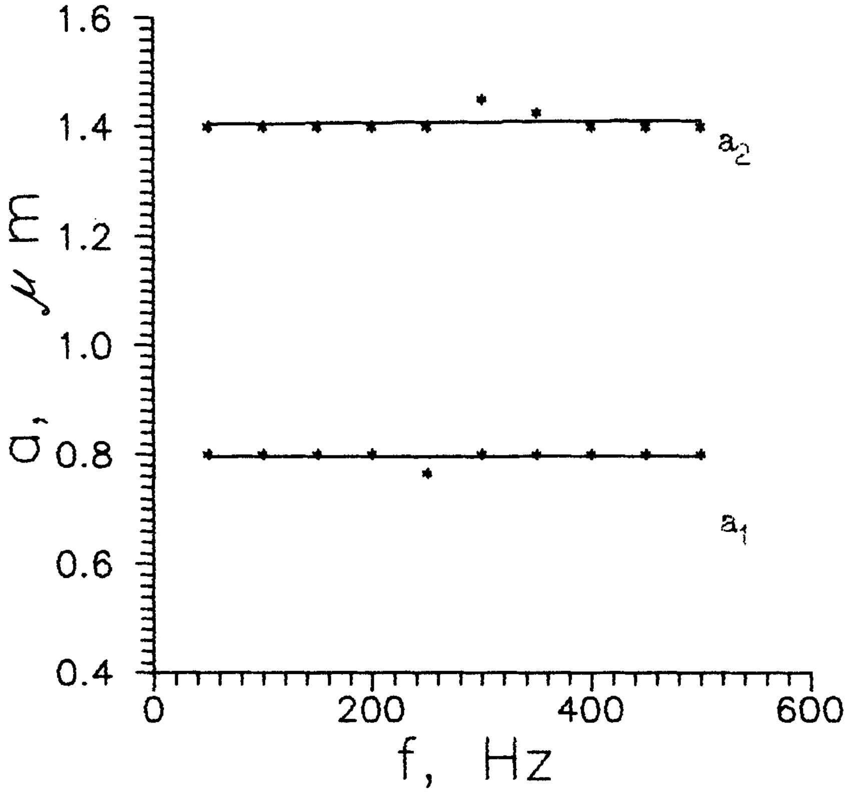


FIGURE 5. Dependencies of thresholds $a_1$ and $a_2$ on the frequency of the oscillating plates.

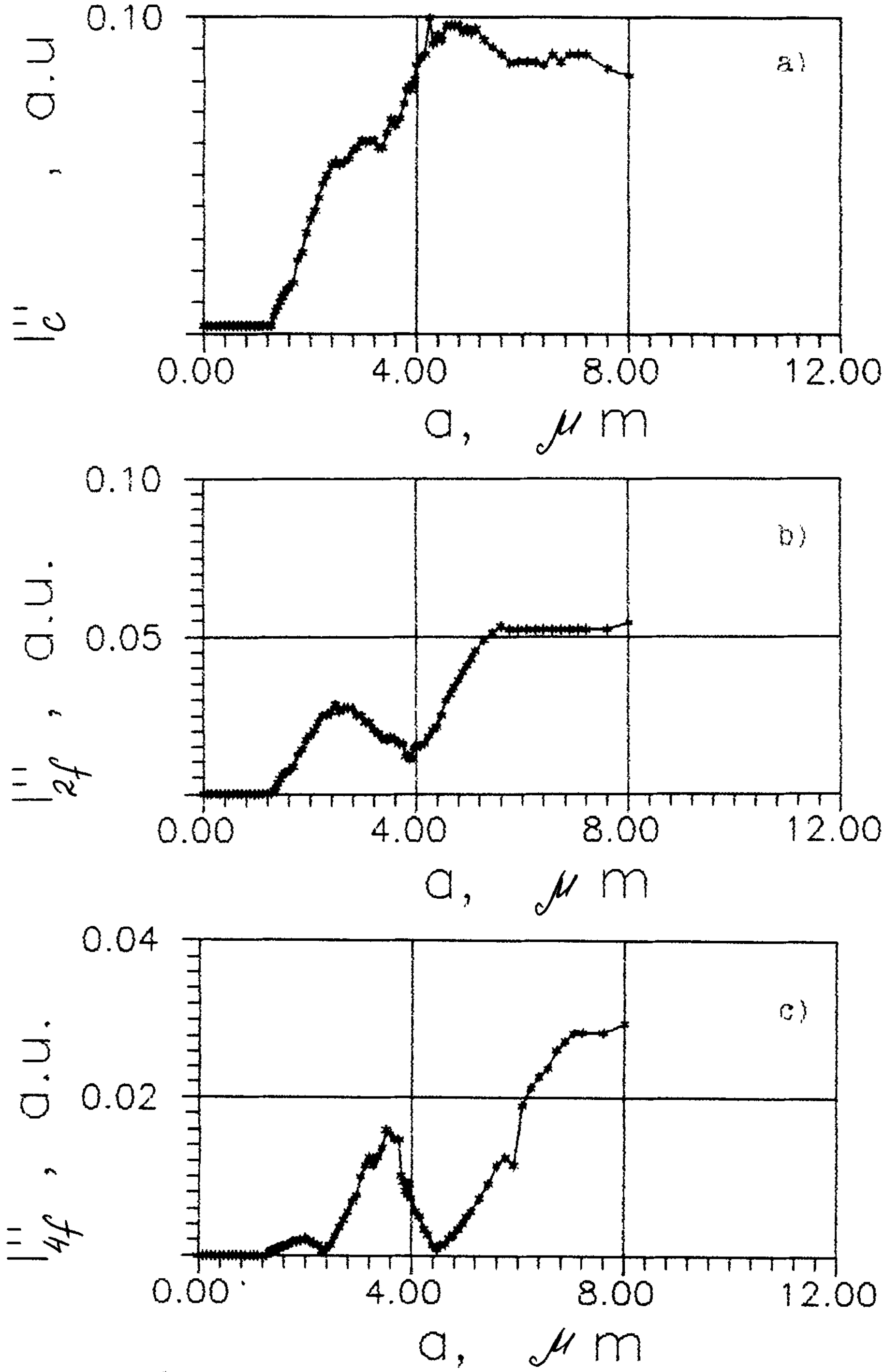


FIGURE 6. Dependencies of a) $I''_c$, b) $I''_{2f}$, c) $I''_{4f}$ on the shearing amplitude a.

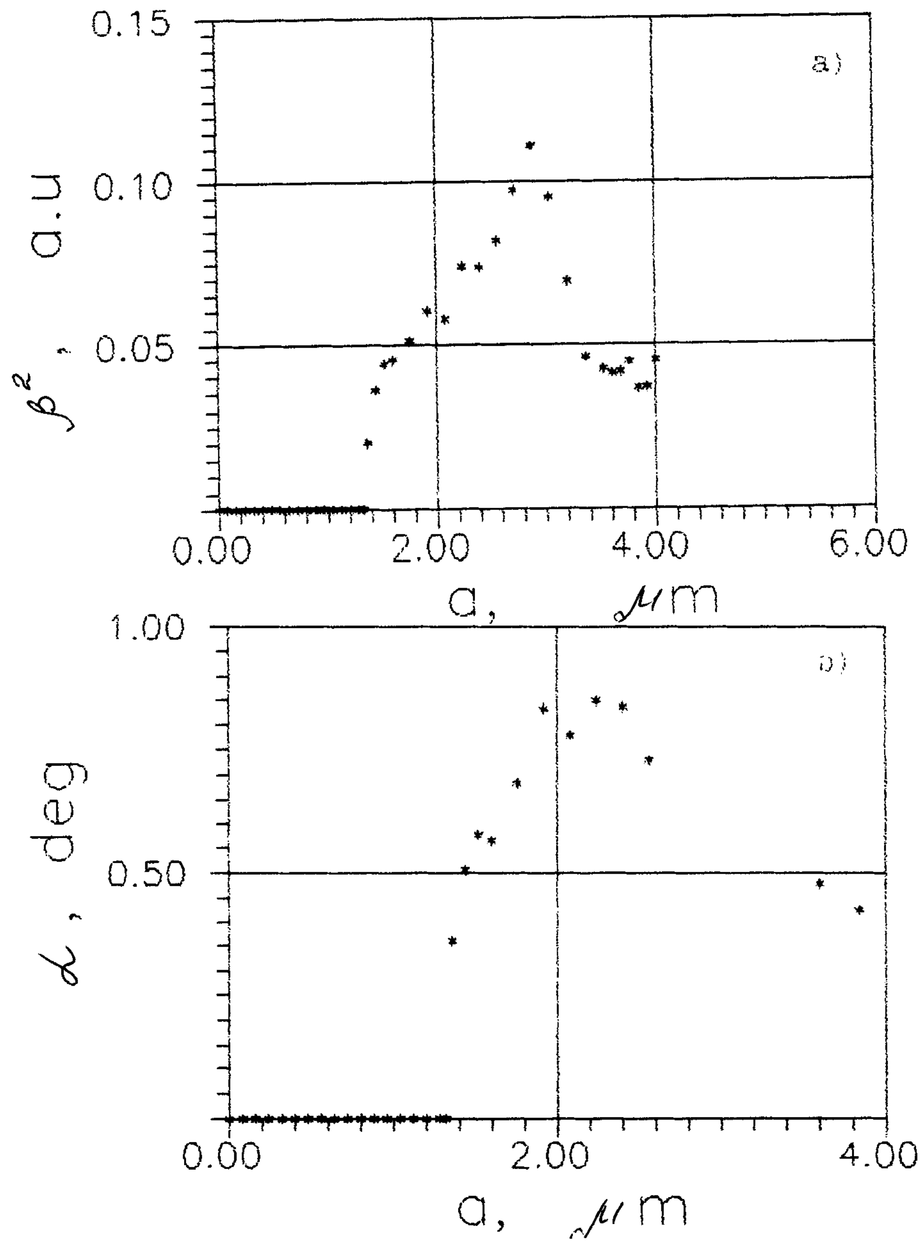


FIGURE 7. Dependencies of a) elliptical parameter $\beta^2$, b) angle $\alpha$ on the shearing amplitude a.

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