Structural transformations in planar nematic crystals in an ultrasonic field

D. I. Anikeev, O. A. Kapustina, and V. N. Lupanov

N. N. Andreev Acoustics Institute of the Academy of Sciences of the USSR
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An unconventional description of ultrasonically induced changes of the orientational state of planar nematic liquid crystals (NLCs) is discussed. The NLC is described on the basis of nonequilibrium hydrodynamics approach, which takes into account the relaxational character of these changes and the anisotropy of the elastic properties of the mesophase, whereas the approach based on classical Leslie–Ericksen hydrodynamics considers only the anisotropy of the viscous properties of the NLC. The agreement between the theory and the obtained experimental data on the threshold characteristics of the observed spatially modulated structures confirms that the new approach is well-grounded.

INTRODUCTION AND FORMULATION OF THE PROBLEM

The effect of acoustic oscillations on an oriented sample of a nematic liquid crystal (NLC) under certain conditions leads to a change inthe ordering of the molecules. This change is manifested in the formation of stationary spatial structures having different scales.[1] Thus at acoustic frequencies structures with a spatial period $\Lambda \sim d$ or $\Lambda \sim \lambda^{1/3}$ are formed in planar samples of an NLC.[2-5] (Here d is the thickness of the layer of NLC and λ is the wavelength of the sound wave.) Such structures also arise in the samples in the ultrasonic range.[6] However they form by a different mechanism from those of Refs. 2–4, which are valid for acoustic frequencies. According to Ref. 2, at acoustic frequencies, where the wavelength of the viscous wave satisfies $\lambda_{\text{vis}} \gg d$, spatial structures form via orientational instability of acoustically driven oscillatory hydrodynamic flow of a nematic liquid.

In this paper we present the results of experimental investigations of the conditions under which spatial structures appear at ultrasonic frequencies in samples with planar orientation of the molecules. The results obtained are interpreted on the basis of the theoretical model of Ref. 7, where a new approach is proposed for describing structural transformations induced in NLC by ultrasound. This model presupposes the following physical interpretation of the phenomenon in an NLC layer: Random and nonuniform (parallel to the layer) deformation of the planar structure, when the molecules protrude out of the plane of the layer by an angle $\theta = \theta_0 f(x,z)$, and compression of the medium in the ultrasonic wave generates anisotropic shear stresses in the NLC layer of the form

$$\sigma_{xz}=\theta(\mu_3 v_{zz}+\Delta E u_{zz})$$

(see Fig. 1). Here u_{zz} and v_{zz} are, respectively, the compression and the rate of compression in the ultrasonic wave; θ_0 is the amplitude of the angle of deflection; $\mu_3 = f(\omega, \tau_1, \tau_2)$ and $\Delta E = f(\omega, \tau_1, \tau_2)$ are the real and imaginary parts of the modulus of elasticity; τ_1 and τ_2 are the relaxation times of the order parameter and the orientation of the terminal groups of the molecules, respectively; and $\omega = 2\pi f$, where f is the ultrasonic frequency. These stresses generate oscillatory vortex flows (which are periodic along the layer) with the velocity $v_z' \sim \theta_0 \cos qz$. The interaction of these flows with the starting ultrasonic field results in the appearance of stationary shear stresses $\langle \sigma_{xz} \rangle$. (Here q is the wave number, describing the periodicity of the distortion along the x axis, and the symbol $\langle \cdot \rangle$ signifies averaging over the period.)

The model postulates two physical mechanisms which are responsible for this effect: a) convective interaction of these flows and of the oscillatory motion of the nematic liquid in the ultrasonic field and b) periodic variation of the viscosity accompanying the compression $u_{zz} \sim e^{-i\omega t}$ of the medium, which destroys the temporal symmetry of the oscillatory flow, in the ultrasonic wave. Stationary stresses generate stationary flows in the layer whose velocity varies periodically along the layer, just as does the velocity v' of the oscillatory flows. The viscous stationary moments, which are proportional to the gradients of the velocities of the stationary flows, amplify the initial distortion of the structure and hence the spatial harmonic of the form

$$\theta \sim \theta_0 \sin qx \cdot \sin pz,$$

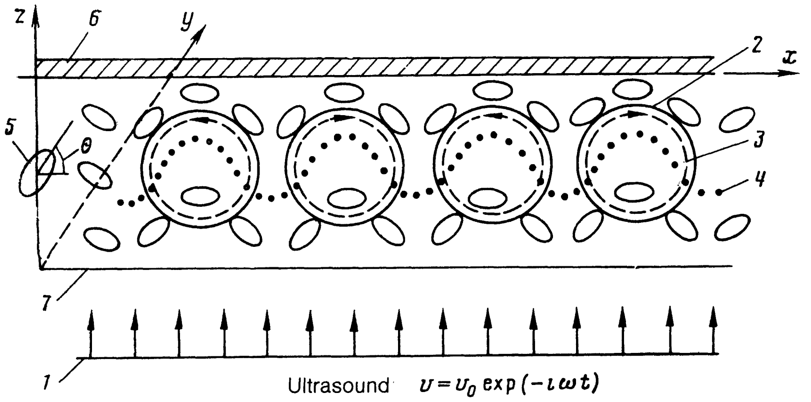

FIG. 1. Illustration of the mechanism of orientational distortion of a planar sample of NLC in an ultrasonic field according to the theoretical model of Ref. 7: 1) ultrasound, 2) oscillating flow, 3) stationary flow, 4) profile of distortions of the orientation of the NLC, 5) NLC molecule, 6) acoustically rigid boundary, 7) acoustically nonrigid boundary.

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where $p = \pi/d$ and $q = \pi/\Lambda$, is amplified most. At the threshold of the effect the action of the viscous stationary moments on the molecules is compensated by the Franck elastic moments.

This interpretation of the phenomena, based on nonequilibrium hydrodynamics and taking into account the anisotropy of the dynamic modulus of elasticity and the coefficient of bulk viscosity of the NLC, differs from the usual approach based on Leslie-Ericksen hydrodynamics, which considers the anisotropy only of the viscous properties of the mesophase.

In Ref. 7 the calculation was performed for the case of normal incidence of ultrasound on a layer of NLC with an acoustically rigid boundary ($z = 0$) that completely reflects the ultrasound. This results in the formation of a standing wave of the form

$$V_z = 2V_0 \sin kz \cdot \cos \omega t,$$

where $k = \omega/c$, c is the velocity of ultrasound in the NLC, and V_0 and V_z are, respectively, the amplitude of the particle velocity and its component along the z axis. The region of applicability of the theoretical model is determined by the following inequalities:

$$\frac{2\pi\eta}{\rho d^2} \ll f \ll \frac{2\pi c}{d}.$$

(Here η and ρ are, respectively, the viscosity and density of the NLC.)

EXPERIMENT

The NLC studied—a eutectic mixture of MBBA and EBBA—filled a flat capillary, consisting of a 2 mm thick glass plate and a thin polymer film with a light-reflecting aluminum coating. In order to create a planar orientation the surfaces bounding the layer were coated with polyvinyl alcohol and then polished in a definite direction. In the experiments the thickness of the layer was varied from 10 to 360 μm.

The orientational state of such samples was observed in polarized reflected light using the standard schemes, which are described in Refs. 1 and 6, for a wave field with a different degree of nonuniformity. For this, in one series of experiments the distance l between the radiator and the sample, which were immersed in water, was varied (it reached $0.75R^2/\lambda$, where R is the radius of the ultrasound radiator), while in another series a Straubel radiator,[8] which produced a quasiuniform wave field in the NLC layer, was employed.[1)] In the latter case acoustic contact between the radiator and the NLC was made through a thin adhesion layer, which was deposited on the outer surface of the polymer film and had a thickness l_{ad}. The driving frequency was varied from 0.3 to 3 MHz. The magnitude of the perturbation was checked by measuring the voltage supplied to the ultrasonic radiator as well as by measuring the intensity of ultrasound incident on the NLC layer by the acoustic-radiometer method.[8] The thermal-stabilization system made it possible to regulate the temperature of the NLC over the range 22–45 °C and to maintain a constant temperature to within 0.5 °C.

EXPERIMENTAL RESULTS AND DISCUSSION

The observations showed that an ultrasonic field, whose frequency satisfies the condition presented above, at a certain threshold in planar NLC layers generates a special type of nonuniform distribution of the director orientation. This distribution is manifested in the form of a system of alternating light and dark bands which are orthogonal to the director $\mathbf{m}^0$ in the undisturbed state. In order to describe this effect quantitatively, we studied experimentally the dynamics of the development of structures and determined their period Λ and threshold particle velocity $V_{0\,th}$. In the experiments we varied one of the wave field parameters, the thickness of the sample, and the temperature of the NLC.

Figure 2a shows a plot of the spatial period Λ of the structures versus the degree of excitation. Here U and U_{th} are, respectively, the running and threshold values of the voltage supplied to the radiator. It is easy to see that near threshold the period of the structures is somewhat greater than its value above threshold and at $U \approx 1.5\,U_{th}$ it reaches a constant value of $\sim 0.5d$, which, according to visual observations, corresponds to stabilization of the pattern of distortion in the NLC. These data pertain to a 40 μm thick sample, placed at a distance $l = 2$ cm from the radiator. The ultrasonic frequency is equal to 3.2 MHz and the temperature of the NLC is equal to 30.2°C. As the observations showed, such changes in the period of the structure accompanying a transition into an above-threshold regime occur for all NLC samples studied in the experiments.

The plot in Fig. 2b summarizes the results of these observations. It shows the values of the spatial period Λ, corresponding to a stable pattern of distortion ($U = 2U_{th}$), for 10–360 μm thick samples in wave fields with different degree of nonuniformity. The labels 1 and 2 show the values of Λ for NLC samples placed at distances $l = 2$ and 20 cm, respectively, from the radiator. The ultrasonic frequency was equal to 3.2 MHz and the temperature was equal to 30.2 °C. The data 3 were obtained in a quasiuniform wave field ($l = l_{ad}$), for which the ultrasonic frequency was equal to 1 MHz and the temperature was equal to 21 °C. It is easy to see that the above threshold, for $U \geqslant 1.5\,U_{th}$, the relation between the spatial period of the structures and the thickness of the film is linear, and in the range of values of d studied for wave fields with different degree of nonuniformity is of the form $\Lambda \approx 0.5d$.

Data on the effect of the ultrasonic frequency on the spatial period of the structures above threshold at $U = 2U_{th}$ are presented in Fig. 2c. The data labelled 1 and 2 show the values of Λ for 40 μm thick samples 2 and 20 cm from the radiator; the temperature was equal to 30.2 °C. As one can see, for ultrasonic frequencies in the range 2.8–10 MHz the spatial period of the structures remains practically constant and is determined by the thickness of the NLC layer.

The experimental results on the effect of the temperature of the NLC on the spatial period of structures above threshold at $U = 2U_{th}$ are presented in Fig. 2d. They pertain to a 60 μm thick sample, a frequency of 3 MHz, and $l = 2$ cm.

It is easy to see that in the temperature interval 20–40 °C the period of the structures is practically constant and is determined only by the thickness of the NLC layer. We note that the distortion becomes irregular at 45 °C and these structures vanish at 49 °C.

We now discuss the basic relationships characterizing the magnitude of the effect at threshold. Table I summarizes the experimental data, which were obtained in a series of

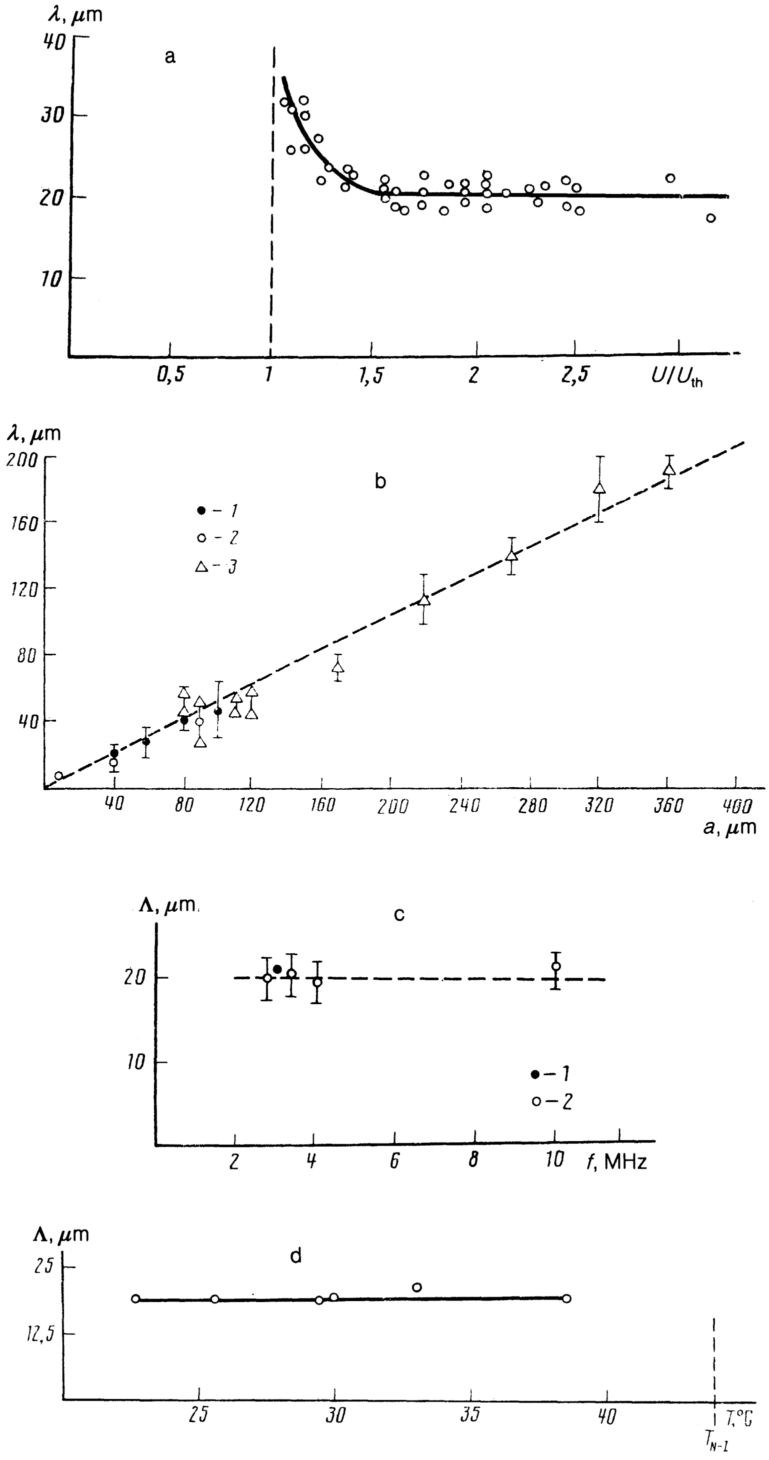

FIG. 2. Effect of acoustic parameters, the dimensions of the sample, and the temperature of the NLC on the spatial period of the structures formed in an ultrasonic field: (a) change in the spatial period of the structures above threshold; (b) effect of the thickness of the NLC layer on the period of the structures; (c) spatial period of the structures in the frequency band 2.8–10 MHz; (d) effect of the temperature on the period of the structures.

independent experiments for wave fields with different degree of nonuniformity, on the effect of the thickness of the NLC layer and the ultrasonic frequency on the threshold particle velocity. These data show that as the thickness of the samples increases in the interval of values of d studied the threshold particle velocity $V_{0\text{th}}$ decreases within comparatively narrow limits. Decreasing the ultrasonic frequency decreases $V_{0\text{th}}$. Thus, for samples 90–100 μm thick, $V_{0\text{th}}$ from 2.5 to 7.8 cm/s, i.e., approximately by a factor of three as the frequency decreases from 3 MHz to 1 MHz.

The experimental data on the temperature dependence of the threshold voltage U_{th} at which structures form are presented in Fig. 3. These voltages are normalized to the threshold voltage U^*_{th} at 22.8 °C;

$$\Delta T = T_{N-I} - T$$

is the difference between the temperature T_{N-I} of the nematic-phase—isotropic-liquid phase transition[10] and the running value of the temperature. The thickness of the sample was equal to 40 μm and the frequency was equal to 3.2 MHz.

TABLE I. The threshold amplitude as a function of the thickness of the NLC layer and the ultrasound frequency.

d, μm	f, MHz	$\Delta T°$, C	V_{0th}, cm/s		Type of apparatus
			Experiment	Calculation	
10	3,2	23	3,6*	5	
40	3,2		2,89	4,5	
90	3,2		2,2	3,7	$l = 20$ cm
10	1	23	—**	5,8	
100	1		7,8	4,5	
110	1		7,5	4,2	
120	1		8	4	
150	1		7,5	3	$l = l_{ad}$
170	1		7,72	1,7	
220	1		5,91	—	
270	1		4,24	—	
320	1		4,8	—	
100	0,1		—	5,8	
100	1		7,8 **	4,45	
100	3,5		2,5 *	3,5	
100	10		—	2,8	$l = 2$ cm

*The threshold amplitude was determined from measurements of the ultrasonic radiation pressure.[8]

**The threshold amplitude was determined from measurements of the voltage on the radiator and its conversion to V_{0th} by the method described in Ref. 9.

One can see that as the phase transition is approached the threshold decreases somewhat.

We now compare the experimental data on the behavior of the threshold characteristics with the theoretical premises which follow from the model of Ref. 7. According to Ref. 7, the amplitude of the threshold particle velocity is described in the most general form by the following expression:

$$V_{0th} = {}^1/_2 \pi c \left[\frac{\mathcal{K}_3 \eta F(s_{min})}{\Delta E \gamma_1}\right]^{1/2} d^{-1}. \quad (1)$$

Here K_3 and γ_1 are the Franck elastic constant and the rotational viscosity of the NLC, $s = q^2/p^2$, and s_{min} is the value of the parameter s that minimizes the function $F(s)$.[2] This implies a relation between the spatial period Λ of the structures and the thickness of the NLC layer:

$$\Lambda = \frac{d}{s_{min}^{1/2}}\,. \quad (2)$$

Estimates show (see Ref. 7) that at low and high frequencies, which are determined by the limiting values of the parameter

$$B = \frac{\pi^2 \mu_3 \eta}{\rho d^2 \Delta E}$$

($B \gg 1$ and $B \ll 1$, respectively), the minimum of the function $F(s)$ corresponds to the following values of s_{min}: 3.7 and 4.8. This gives for the spatial period Λ the values $0.52d$ and $0.48d$, respectively. Thus the changes produced in the spatial period by a change in the ultrasonic frequency are very insignificant; this agrees with the experimentally observed fact that the period of the structures does not depend on the ultrasonic frequency. The theoretical values of Λ presented above are equal to the period of the structures observed above threshold $U \gg 1.5 U_{th}$. The difference between the theoretical and experimental values of Λ at threshold could be due to the development of hydrodynamic flows,[3)] which also give rise to deflection of the director. Near threshold the main effect is still small, and because of the influence of this factor a spatial distribution of the director with wave vector different from $q = \pi/\Lambda$ can form in the NLC layer.

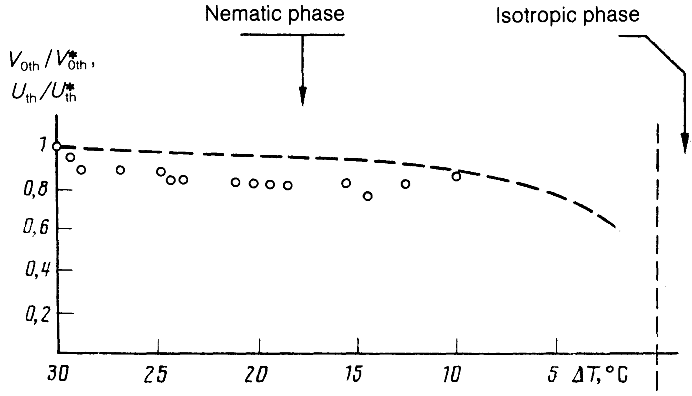

FIG. 3. Effect of the temperature of the NLC on the parameters characterizing the wave field in the NLC layer: the running values of U_{th} and V_{0th} are normalized to the values of U^*_{th} and V^*_{th} corresponding to the lower limit of the temperature interval studied ($T = 22.8$ °C).

Table I gives the values of the threshold particle velocity which were calculated from Eq. (1) for 10–170 μm thick layers of MBBA in the frequency range 1–10 MHz at the temperature $\Delta T = T_{N-I} - 10\,^\circ\mathrm{C}$. As one can see, $V_{0\mathrm{th}}$ is virtually independent of the thickness of the layer. This agrees with the experimental data. As the ultrasonic frequency increases $V_{0\mathrm{th}}$ decreases. This also agrees qualitatively with the experimental results. The small difference between the absolute experimental and computed values of the threshold particle velocities could be associated with the errors of measurement of the intensity of ultrasound by the acoustic-radiometer method, which gives the average intensities over the cross section of the beam, as well as with the approximate character of the amplitudes of the particle velocities estimated by the well-known bridge method of calculating the voltage supplied to the radiator.[9] In addition, the value used in the calculations for the energy of adhesion of molecules to the surface of the supporting plates $w = 2.2\cdot 10^3$ cal/mole can differ somewhat from the true value of w, corresponding to the conditions of the experiments, in which this quantity was not checked.

According to Ref. 7, in the band of frequencies satisfying the condition $\omega\tau_1 \gg 1$ (for MBBA the relaxation time of the order parameter is equal to $2.2\cdot 10^{-7}$ s, so that the frequencies at which the corresponding experimental data were obtained fall into this band), the amplitude of the threshold particle velocity is virtually independent of the temperature and satisfies the following law:

$$V_{0\mathrm{th}} \sim \Delta T^{-1/3}.$$

The plot in Fig. 3 shows this dependence, calculated from the relation (1) with $s_{\min} = 4$ for a 50 μm thick MBBA sample at an ultrasonic frequency of 3.5 MHz. It was constructed from the numerical value of the amplitude of the particle velocity at a temperature corresponding to the condition $\Delta T = 10\,^\circ\mathrm{C}$. The value of $s_{\min}$ chosen in these calculations is the average of the values corresponding to low and high frequencies. One can see that this plot fits the experimental data obtained under conditions close to the design conditions ($d = 60\,\mu$m and $f = 3.2$ MHz).

The foregoing analysis provides that the new mechanism, proposed in Ref. 7, of stationary distortion of planar samples of NLC in a standing ultrasonic wave is valid. The new ideas take into account the relaxational character of the change induced in the orientational state of an NLC by ultrasound and they are important for correctly interpreting previous data,[6,11–13] obtained independently in different laboratories at ultrasonic frequencies while studying different types of mesophases.

We take this opportunity to thank E. N. Kozhevnikov for participating in a discussion of the results.

[1)] Here we mean a quartz radiator in the form of a plate, whose shape follows the curve of the square root of the modulus of elasticity (according to Straubel), thanks to which the oscillations of the surface of the plate accompanying thickness perturbations are more uniform.

[2)] The expression for the function $F(s)$ is not given here because it is too complicated. It can be found in Ref. 7.

[3)] The mechanism responsible for hydrodynamic flows in an NLC layer under conditions similar to those described in the experiments discussed in Ref. 1. These include: the presence of nonuniformities in the NLC layer, nonuniform distribution of the wave parameters over the cross section of the ultrasonic wave, oblique incidence of ultrasound on the layer, etc.

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