

Domain structure of cholesteric liquid crystal acted upon by ultrasound

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(Submitted 18 September 1986)

Zh. Eksp. Teor. Fiz. **92**, 1306–1315 (April 1987)

A theory of the action of ultrasound on the structure of a planar-oriented cholesteric liquid crystal layer is developed. It is shown that at sound intensities above threshold, cholesteric layers become distorted, forming a domain structure of the square-grid type. The threshold amplitude of the velocity in the ultrasound wave and the size of the domains are determined for an equilibrium layer and for one stretched along the crystal axis. The calculation results are compared with the experimental data.

The action of sound on the structure of liquid crystals (LC) is of interest from the standpoint of visualizing a sound field and transforming acoustic information into optic. Most experimental and theoretical studies made to date were on nematic LC. The action of sound on the structure of nematics, with homeotropic molecule orientation, reduces to rotation of the molecules in acoustic streams at high frequencies,¹ to a linear-in-amplitude deformation of the sound oscillations at low frequencies,² and to the onset of domains following an elliptic³ and a one-dimensional shear.⁴ The action of both periodic shear⁵ and periodic compression on the structure of a nematic with planar orientation of the molecules also leads to domain formation.

The action of sound on the structure of a cholesteric LC (CLC) has been less investigated. The dependence of the pitch of the CLC helix on the temperature, which changes as the crystal is heated in the acoustic field, was used in Ref. 7 to visualize strong acoustic fields. Ultrasound action, not connected with thermal effects, on the structure of CLC was investigated experimentally in Refs. 8 and 9, where it was shown that when an ultrasound wave is incident on a CLC layer with planar orientation of the molecules, periodic distortions of the CLC structure are produced and have the form of domains; the effect has a threshold. If the initial orientation is homeotropic, domains of the fingerprint type or bubble domains are produced in various situations.

We construct here a theory for the action of ultrasound on a CLC layer with planar initial orientation of the molecules and with a helix pitch much smaller than the layer thickness. The effect is analyzed on the basis of the CLC hydrodynamic equations, in which are retained the quadratic terms proportional to the product of the molecule-rotation angles by the velocity of the liquid. Allowance for quadratic terms leads to the following picture of the effect. Random distortions of cholesteric layers lead, owing to molecule rotation in the plane of the layer or to departure of molecules from the cholesteric plane, to the appearance of oscillating eddies whose dimension along the layer determines the domain dimensions, and the dimension along the crystal is of the order of the layer thickness. The interaction of interacting streams and the molecule rotation with compression they produce in the ultrasound wave leads to time-averaged torques that add to the distortion of the cholesteric structure. At the threshold of the effect these torques are stabilized by elastic Frank torques. At sound-velocity amplitudes above threshold, stationary distortion, periodic along the

layer, is produced in the cholesteric layers and is observed as a domain structure. Two cases are analyzed here; an unstretched CLC layer with an equilibrium helix pitch, and a stretched one with the helix pitch larger than the equilibrium value.

When determining the viscous torques and stresses in the hydrodynamics of CLC with weakly distorted structure, we shall treat the CLC as a twisted nematic and disregard the inhomogeneity of the molecule orientation. The molecule-rotation and motion equation stake in this case has the form

$$\left[\mathbf{n}, \left\{ \gamma (\mathbf{N} - \hat{\mathbf{v}} \cdot \mathbf{n}) - \nabla_i \frac{\partial g}{\partial \nabla_i \mathbf{n}} + \frac{\partial g}{\partial \mathbf{n}} \right\} \right] = 0, \quad (1)$$

$$p\mathbf{v} = -\nabla P + \nabla \cdot \hat{\sigma} + \mathbf{F}.$$

Here \mathbf{n} is the director that determines the molecule-alignment direction ($\mathbf{n}^2 = 1$); \mathbf{v} is the velocity, $\hat{\mathbf{v}}$ the strain-rate tensor, $\mathbf{N} = \dot{\mathbf{n}} - (1/2)\text{curl}[\mathbf{v} \times \mathbf{n}]$ specifies the rotation of the molecules relative to the surrounding liquid; ρ is the density, γ the coefficient of rotational viscosity, g the density of the Frank elastic energy, $G = \int g dV$, and P the pressure. We write the viscous-stress tensor $\hat{\sigma}$ in the form

$$\sigma_{ij} = (\mu_1 v_{hh} + \mu_2 v_{km} n_k n_m) \delta_{ij} + \mu_3 v_{hh} n_i n_j + \alpha_4 v_{ij} + \alpha_2 N_i n_j + \alpha_5 v_{ik} n_k n_j + \alpha_6 v_{jk} n_k n_i,$$

where μ_i and α_i are the Leslie viscosity coefficients. Assuming, in analogy with nematic crystals, that the viscosity coefficients α_1 and α_3 are small, we set them equal to zero, so that $\gamma = \alpha_2 = \alpha_5 - \alpha_6$.

Note that allowance for the inhomogeneity of the director orientation in the expressions for the viscous torques and stresses leads to a more complete hydrodynamic description of the CLC; in the expressions for the torques and the stresses there appear additional terms that are linear in \mathbf{N} and $\hat{\mathbf{v}}$ and contain in the coefficients the component of the unit vector \mathbf{l} which is directed along the helix axis.¹¹ The description of the effect in this case becomes unjustifiably cumbersome, and the result will differ from those obtained below on the basis of Eqs. (1) only by a dimensionless factor in the expression for the threshold velocity, a factor consisting of the viscosity coefficients that appear in the complete hydrodynamic description of CLC. The values of these viscosities are unknown, as are the factors made up of them. It is therefore impossible to establish the difference between the results of the complete and incomplete descriptions of the ef-

fect, and in the present paper the analysis of the effect is based on the simplified equations (1).

The force \mathbf{F} arises in a deformed cholesteric structure and is determined by varying the density g of the Frank elastic energy with respect to the displacement \mathbf{u} :

$$\mathbf{F} = -\frac{\partial g}{\partial \mathbf{u}} + \nabla_i \frac{\partial g}{\partial \nabla_i \mathbf{u}} - \Delta \frac{\partial g}{\partial \Delta \mathbf{u}} \dots \quad (2)$$

The Frank elastic energy G takes in the two-constant approximation the form¹²

$$G = \frac{1}{2} K \int_V \{ (\operatorname{div} \mathbf{n})^2 + \lambda (\mathbf{n} \operatorname{rot} \mathbf{n} + q)^2 + [\mathbf{n} \operatorname{rot} \mathbf{n}]^2 \} dV,$$

where $K = K_1 = K_3$, $\lambda = K_2/K$, K_i are the Frank elastic constants, $q = 2\pi/P_0$, and P_0 is the pitch of the helix. In the unperturbed state, the director lies in cholesteric planes and rotates uniformly on going from one plane to another, with a period P_0 . We define the structure distortions by the angles θ and φ that determine the deviation of the molecules, respectively, from the unperturbed cholesteric plane and (in the plane) from the initial orientation. Uniform rotation through an angle φ displaces the cholesteric layers along the crystal axis by a distance φ/q ; a nonuniform rotation of the molecules along the layer distorts the layer. We regard hereafter the angles θ and φ as spatial variables. The Frank elastic energy takes in the approximation quadratic in θ and φ the form

$$G = \frac{1}{2} K \int_V \{ (\nabla \varphi)^2 + (\nabla \theta)^2 + q^2 \theta^2 + 4q (\mathbf{n} \nabla) \varphi + (\lambda - 1) (n_1 \nabla_2 \theta - n_2 \nabla_1 \theta - \nabla_{\parallel} \varphi)^2 \} dV,$$

where \mathbf{n} is the director in the unperturbed CLC structure, with components n_1 and n_2 in the cholesteric plane; ∇_{\parallel} and ∇_{\perp} (∇_1, ∇_2) are the gradients along and perpendicular to the cholesteric helix, respectively.

The cholesteric-crystal structure is least stable to perturbations whose energy is a minimum—such perturbations are the angles φ that vary slowly along the crystal axis and the rapidly varying angles θ , which can be represented in the form

$$\theta = n_{\alpha} \tau_{\alpha}, \quad \alpha = 1, 2,$$

where τ_{α} is a function that varies slowly along the crystal axis. These are precisely the perturbations considered in the present article. The integrals of the terms $n_1 \nabla_2 \theta \nabla_{\parallel} \varphi$, $n_2 \nabla_1 \theta \nabla_{\parallel} \varphi$ and $n_1 n_2 \nabla_1 \theta \nabla_2 \theta$ over the volume of the CLC layer vanish if the molecule orientation does not change on the layer boundaries; the elastic energy assumes the following final form:

$$G = \frac{1}{2} K \int_V \{ (\nabla_{\perp} \varphi)^2 + \lambda (\nabla_{\parallel} \varphi)^2 + (\nabla \theta)^2 + q^2 \theta^2 + 4q \theta (\mathbf{n} \nabla) \varphi + (\lambda - 1) [n_1^2 (\nabla_2 \theta)^2 + n_2^2 (\nabla_1 \theta)^2] \} dV. \quad (3)$$

In the particular case when two-dimensional structure distortions are considered in a plane perpendicular to the cholesteric layers, assuming the elastic moment conjugate to the angle θ to be zero, expressing the angle φ in terms of the displacements of the liquid along the crystal axis \mathbf{u} ($\varphi = -q\mathbf{u}$), assuming that $\lambda \ll 1$, and averaging over the

pitch of the helix in Eq. (3), we arrive at a known formula for the elastic energy¹³:

$$G = \int_V \left[\frac{1}{2} K_2 q^2 (\nabla_{\parallel} \mathbf{u})^2 + \frac{3}{16} K_3 (\Delta_{\perp} \mathbf{u})^2 + \frac{1}{2} K_3 (\nabla_{\parallel} \nabla_{\perp} \mathbf{u})^2 \right] dV.$$

Consider an ultrasonic wave of frequency ω and vibrational velocity v_0 normally incident on a CLC layer. We confine ourselves to frequencies at which the following inequalities hold:

$$\eta/\rho h^2 \ll \omega < c/h,$$

where h is the CLC layer thickness, η the viscosity, and c the speed of sound in the liquid crystal. These inequalities mean that the length of the viscous wave is assumed shorter and that of the sound wave longer than the CLC-layer thickness. The latter inequality allows us to determine the threshold of the effect by replacing the action of the sound by its average over the layer.

We describe the effect in the following geometry. The z axis is normal to the layer, with $z = 0$ the lower face of the layer; the axes x and y lie in the plane of the lower face of the layer; the unperturbed cholesteric layers are parallel to the (xy) plane. We assume furthermore that

$$n_1 = n_x, \quad n_2 = n_y, \quad \nabla_1 = \partial_x, \quad \nabla_2 = \partial_y.$$

Action of the sound on the distorted CLC structure ($\varphi \neq 0$, $\theta \neq 0$) produces both acoustic waves and eddies, in which the velocities differ by a factor $\eta\omega/\rho c^2$. Confining ourselves in addition to the frequencies ω , at which this ratio is small ($\omega < 10^{10} \text{ sec}^{-1}$ in a real case), we exclude acoustic modes from consideration applying the curl operation to the equation of motion (1). Retaining in the equations the quadratic terms proportional to the product of molecule rotation angles θ and φ by the velocity, and carrying the necessary transformations, we arrive at the following systems of equations.

The rotation equations:

$$\begin{aligned} \gamma \{ \varphi, \iota + qv_z - 1/2 (\operatorname{rot} \mathbf{v})_z + n_x n_y (v_{xx} - v_{yy}) + (n_x^2 - n_y^2) v_{xy} \} - \Gamma_{\varphi} &= 0, \\ \gamma (\dot{\theta} - n_{\alpha} v_{z, \alpha} - \theta v_{zz}) - \Gamma_{\theta} &= 0. \end{aligned} \quad (4)$$

Here Γ_{θ} and Γ_{φ} are the elastic torques conjugate to the angles θ and φ and expressed in terms of the free energy (3) by the Euler equations:

$$\begin{aligned} \Gamma_{\varphi} &= \nabla \frac{\partial g}{\partial \nabla \varphi} - \frac{\partial g}{\partial \varphi} = K (\Delta_{\perp} \varphi + \lambda \varphi_{,zz} + 2q n_{\alpha} \theta_{, \alpha}), \\ \Gamma_{\theta} &= \nabla \frac{\partial g}{\partial \nabla \theta} - \frac{\partial g}{\partial \theta} \\ &= K [\Delta \theta - q^2 \theta - 2q n_{\alpha} \varphi_{, \alpha} + (\lambda - 1) (n_x^2 \theta_{,yy} + n_y^2 \theta_{,xx})], \end{aligned}$$

Here and elsewhere the subscript α runs through the values $\alpha = x$ and y , and $\Delta_{\perp} = \partial_x^2 + \partial_y^2$.

The equations of motion:

$$\begin{aligned} (\rho \partial_t \Delta - \bar{\Delta}^2) v_z &= -\gamma \Delta_{\perp} n_{\alpha} \theta_{, \alpha} + (\mu_3 + \alpha_5) v_{zz} \Delta_{\perp} n_{\alpha} \theta_{, \alpha} \\ &\quad - \alpha_6 \partial_z^2 (v_{zz} n_{\alpha} \theta_{, \alpha}) + (\operatorname{rot} \operatorname{rot} \mathbf{f})_z + \Delta_{\perp} F_z - \partial_z (F_{\alpha, \alpha}), \end{aligned}$$

$$[\rho\partial_t - (\eta + \gamma)\partial_z^2 - (\eta - \alpha_i/2)\Delta_\perp](\text{rot } \mathbf{v})_z = -\gamma\Delta_\perp(\varphi, \dot{v}_z + qv_z) + \alpha_6\partial_z[v_{zz}(n_y\theta_{y,x} - n_x\theta_{x,y})] + \partial_z(\text{rot } \mathbf{f})_z + F_{y,x} - F_{x,y},$$

$$\text{div } \mathbf{v} = 0.$$

We have introduced here the notation

$$\bar{\Delta}^2 = \eta\partial_z^2\Delta_\perp + \eta_2\partial_z^4 + (\eta + \gamma/2)\Delta_\perp^2,$$

$$\eta = 1/2(\alpha_4 + \alpha_5/2), \quad \eta_2 = \alpha_4 + \alpha_6/4$$

\mathbf{f} is the force, nonlinear in velocity, due to convection and to compression of the medium:

$$\mathbf{f} = \rho[(\nabla\mathbf{v})\mathbf{v} + \mathbf{v}(\nabla\mathbf{v})].$$

To derive the force \mathbf{F} from Eq. (2) we must transform in the expression for the Frank elastic energy to the material angle variables θ_m and φ_m and exclude molecule rotations connected with rotation of the medium as a whole. This transition is implemented by substituting in (3) the angles θ and φ in the form

$$\theta = \theta_m - n_\alpha u_{z,\alpha}, \quad \varphi = \varphi_m - qu_z - 1/2(u_{x,y} - u_{y,x}).$$

Fixing the values of θ_m and φ_m in the elastic energy and substituting the latter in (2), we get

$$F_z = -q\Gamma_\varphi + n_\alpha\Gamma_\theta, \quad F_{x,y} - F_{y,x} = \Delta_\perp\Gamma_\varphi, \quad F_{\alpha,\alpha} = 0.$$

Expressing Γ_φ and Γ_θ in terms of the viscous moments with the aid of the rotation equations (4), we obtain the final form of the equations of motion:

$$(\rho\partial_t\Delta - \bar{\Delta}^2)v_z = -\gamma\Delta_\perp n_\alpha\dot{\theta}_{,\alpha} + (\mu_3 + \alpha_5)v_{zz}\Delta_\perp n_\alpha\theta_{,\alpha} - \alpha_6\partial_z^2(v_{zz}n_\alpha\theta_{,\alpha}) + (\text{rot } \text{rot } \mathbf{f})_z - \gamma\Delta_\perp\{q[\varphi, \dot{v}_z - 1/2(\text{rot } \mathbf{v})_z] + n_x n_y (v_{xx} - v_{yy}) + (n_x^2 - n_y^2)v_{xy} - n_\beta\partial_\beta(\dot{\theta} - n_\alpha v_{z,\alpha} - \theta v_{zz})\}, \quad \beta = x, y,$$

$$[\rho\partial_t - (\eta + \gamma)\partial_z^2 - (\eta - \alpha_i/2)\Delta_\perp](\text{rot } \mathbf{v})_z = -\gamma\Delta_\perp(\varphi, \dot{v}_z + qv_z) + \alpha_6\partial_z[v_{zz}(n_y\theta_{y,x} - n_x\theta_{x,y})] + \partial_z(\text{rot } \mathbf{f})_z - \Delta_\perp\Gamma_\varphi, \quad (5)$$

$$\text{div } \mathbf{v} = 0.$$

The term $-\gamma q^2\Delta_\perp v_z$ in the right-hand side of the equation for v_z gives rise to a seeming increase of viscosity in flow of a cholesteric liquid in a direction normal to cholesteric layers whose positions are fixed in space.

We linearize next the system (4) and (5) with respect to the angular variables and the velocity perturbation $\delta\mathbf{v}$, regarding the velocity of the medium and its compression in an incident sound wave as coefficients that depend on the time and coordinates. We represent θ , φ , and $\delta\mathbf{v}$ as a sum of a stationary term and one that oscillates at the frequency of sound:

$$\theta = \theta^0 + \theta', \quad \varphi = \varphi^0 + \varphi', \quad \delta\mathbf{v} = \mathbf{v}^0 + \mathbf{v}',$$

where the superscript 0 denotes the stationary part, and the prime the oscillating one, and subdivide the system (4), (5) into systems of equations for stationary and nonstationary variables. We consider the distortions of a structure with a minimal free energy, when the angle is represented in the form $\theta = n_\alpha\tau_\alpha$ while τ_α and the angle φ vary slowly along the crystal axis. We shall also regard the velocity perturbations as slowly varying with respect to the functions. We average all the terms in the equations for φ and \mathbf{v} over the

pitch P_0 of the helix; the variables φ' , $(\text{curl } \mathbf{v}')_z$ and $(\text{curl } \mathbf{v}')_0 z$ turn out to be unrelated to the stationary distortions φ^0 and θ^0 , and consequently do not influence the destruction of the cholesteric structures—we henceforth leave out the equations for these variables. Discarding also in the equation for θ' the elastic terms, which are small compared with the viscous ones, and carrying out the necessary transformations, we obtain the following self-consistent system of equations for θ' , v'_z , v_z^0 , θ^0 , φ^0 :

$$\theta'_{,t} - n_\alpha v'_{z,\alpha} - \theta^0 v_{zz} = 0, \quad (6)$$

$$(\rho\partial_t\Delta - \bar{\Delta}^2)v'_z \approx (\mu_3 + \alpha_6)v_{zz}\Delta_\perp \langle n_\alpha\theta'_{,\alpha} \rangle,$$

$$\gamma q v_z^0 - 1/2\gamma(\text{rot } \mathbf{v}^0)_z - K[\Delta_\perp\varphi^0 + \lambda\varphi_{,zz}^0 - 2q\langle n_\alpha\theta'_{,\alpha} \rangle] = 0,$$

$$\gamma(\overline{\theta'_{,z}v_z - n_\alpha v_{z,\alpha} - \theta^0 v_{zz}}) - K\{\Delta_\perp\theta^0 - q^2\theta^0 - 2qn_\alpha\varphi_{,\alpha}^0 + (\lambda - 1)(n_x^2\theta'_{,yy} + n_y^2\theta'_{,xx})\} = 0,$$

$$-\bar{\Delta}^2 v_z^0 = -\gamma\Delta_\perp \langle n_\alpha\theta'_{,\alpha} \rangle v_z + (\mu_3 + \alpha_5)\Delta_\perp v_{zz} \langle n_\alpha\theta'_{,\alpha} \rangle + \langle (\text{rot } \text{rot } \mathbf{f})_z \rangle - \gamma\Delta_\perp\{q[qv_z^0 - 1/2(\text{rot } \mathbf{v}^0)_z] - \langle n_\beta\partial_\beta(\theta'_{,z}v_z - n_\alpha v_{z,\alpha} - \theta^0 v_{zz}) \rangle\}.$$

Here and elsewhere the velocity v_z and its derivatives with respect to z pertain to the acoustic wave in the layer, the angle brackets denote averaging over the pitch of the cholesteric helix, and the superior bar denotes averaging over the period of the oscillations in the sound wave. Terms of the form $v_z^0 v_{zz}$ and $v_{zz}^0 v_z$ have been left out of the equation for v'_z , since their inclusion in the solution of the problem leads to a small relative correction, of order $K\rho/\gamma^2 \ll 1$, to the determination of the threshold amplitude of the velocity.

We assume zero stationary perturbations at the boundaries:

$$\varphi^0|_{z=0, h} = \theta^0|_{z=0, h} = 0, \quad v_z^0|_{z=0, h} = 0,$$

and define the oscillating variables v'_z and θ' , neglecting effects in the boundary layers whose thickness is less than the length of the viscous wave as a particular solution of corresponding inhomogeneous equations. The condition for the existence of a nonzero solution of the system (6) is then tantamount to the condition of domain formation.

We substitute the stationary perturbations in the form of functions that are periodically dependent on the variables x and y , and satisfy the boundary conditions,

$$\varphi^0, \theta^0, v_z^0 \propto \exp(ik_x x + ik_y y) \sin(pz),$$

where k_x and k_y are wave numbers that determine the form of the domain structure, $p = 2\pi/h$, and $\partial_\alpha = ik_\alpha$ and $\Delta_\perp = -k_x^2 - k_y^2 = -k^2$. We assume that the following inequalities are obeyed:

$$p < k < q.$$

Under these assumptions, the sound field in the CLC layer leads according to (6), in the presence of the perturbation θ^0 , to the following oscillations of the velocity v'_z and the angle θ' :

$$v'_z = \frac{\mu_3 + \alpha_6}{2a} \tau_{\alpha,\alpha}^0 (\eta k^2 - \rho\partial_t) v_{zz},$$

$$\theta' = \frac{\mu_3 + \alpha_6}{2a} n_\beta \tau_{\alpha,\alpha\beta}^0 (\eta k^2 - \rho\partial_t) u_{zz},$$

where $a = \rho^2 \omega^2 + \eta^2 k^4$ and u_{zz} is the compression in the sound wave.

The averaged terms in the stationary equations of the system (6) are equal to

$$L_1 = \overline{\langle n_{\alpha} \theta'_{,\alpha'} \rangle v_{zz}} = \frac{\rho k^2 (\mu_3 + \alpha_6)}{4a} \tau_{\alpha,\alpha}^0 \overline{\langle v_{zz} \rangle^2},$$

$$L_2 = \overline{\langle n_{\alpha} \theta'_{,\alpha z} \rangle v_z} = \frac{\rho k^2 (\mu_3 + \alpha_6)}{4a} \tau_{\alpha,\alpha}^0 \overline{\langle v_z v_{z,zz} \rangle},$$

$$L_3 = \overline{\langle (\text{rot rot } \mathbf{f})_z \rangle} = \frac{\rho k^4 \eta (\mu_3 + \alpha_6)}{2a} \tau_{\alpha,\alpha}^0$$

$$\times [2 \overline{\langle v_{zz} \rangle^2} + \overline{\langle v_z v_{z,zz} \rangle}].$$

When the condition $h\omega/c < 1$ is met, the mean values $\overline{\langle v_{zz} \rangle^2}$ and $\overline{\langle v_z v_{z,zz} \rangle}$ vary little over the CLC-layer thickness; we replace them in L_i hereafter by the values averaged over the layer thickness. Substituting L_i in the stationary equations of the system (6), excluding the stream velocities v_z^0 , and carrying out the necessary transformations we arrive at the following system of equations for the angles φ^0 and $\theta^0 = n_{\alpha} \tau_{\alpha}^0$:

$$(k^2 + \lambda p^2) \varphi^0 - [q + \rho k^2 (\mu_3 + \alpha_6) (\mu_3 + \alpha_5) D / 4aqK] ik_{\alpha} \tau_{\alpha}^0 = 0,$$

$$[2q^2 + k^2 + (\lambda - 1) (3k_y^2 + k_x^2) / 4] \tau_x^0 + 2iqk_x \varphi^0 = 0, \quad (7)$$

$$[2q^2 + k^2 + (\lambda - 1) (3k_x^2 + k_y^2) / 4] \tau_y^0 + 2iqk_y \varphi^0 = 0,$$

where

$$D = \left(\frac{2\gamma + 4\eta}{\mu_3 + \alpha_5} - 1 \right) \overline{\langle v_{zz} \rangle^2} + \frac{2\eta - \gamma}{\mu_3 + \alpha_5} \overline{\langle v_z v_{z,zz} \rangle},$$

the angle brackets denote averaging over the CLC layer thickness, and the term containing D in Eqs. (7) describes the torque which is due to the presence of the sound field and which destabilizes the CLC structure.

Equating to zero the determinant of the system (7), we obtain the value of D at which the system has a nonzero solution:

$$D = \frac{4a}{\rho k^2 (\mu_3 + \alpha_5) (\mu_3 + \alpha_6)} B(\psi) K [k^2 + \lambda p^2 - k^2 B(\psi)], \quad (8)$$

where

$$B(\psi) = 2 \frac{q^2}{k^2} \left[\sin^2 \psi \left(2 \frac{q^2}{k^2} + \frac{1}{4} + \frac{3}{4} \lambda + \frac{\lambda - 1}{2} \sin^2 \psi \right)^{-1} \right. \\ \left. + \cos^2 \psi \left(2 \frac{q^2}{k^2} + \frac{1}{4} + \frac{3}{4} \lambda + \frac{\lambda - 1}{2} \cos^2 \psi \right)^{-1} \right],$$

the angle ψ determines the relation between the wave numbers k_x and k_y , viz., $k_x = k \cos \psi$, $k_y = k \sin \psi$, and consequently the form of the produced structure.

The form of the domain structure and the threshold velocity amplitude $v_{0,\text{thr}}$ are obtained by minimizing D with respect to ψ and k . A slight minimum with respect to the angle ψ is reached at $|\sin \psi| = |\cos \psi| = 2^{-1/2}$, when $k_x = k_y$, and the domain structure has the structure of a square grid. Note that the dependence of D on the angle ψ is governed only by the form of the elastic energy (3), whose density g can be represented, with allowance for the second and third equations of the system (7), in the form

$$g = \frac{1}{16} K [k^2 + \lambda p^2 - k^2 B(\psi)] (\varphi^0)^2$$

and which is minimal for perturbations of the square-grid type ($B\psi - \max$), determining by the same token the appearance of just this structure. Domains of the square-grid type, being energywise favored, should result also from other actions, isotropic in the layer planes, on the CLC structure. Such actions are thermal convection, dilatation, and others. The anisotropy of the external action can lead to a different domain structure—such a situation arises if domains in the form of stripes appear in Couette flow in a CLC layer.¹⁴

Putting $\sin^2 \psi = \cos^2 \psi = 1/2$ in Eq. (8) and transforming it, we get

$$D = \frac{K\rho(1+\lambda)\omega^2}{(\mu_3+\alpha_5)(\mu_3+\alpha_6)} \frac{(k^2+4\sigma^4)(k^2+k_0^4)}{4k^4\sigma^4},$$

$$\sigma = \left(\frac{\rho\omega}{2\eta} \right)^{1/2}, \quad k_0 = \left(\frac{4\lambda}{1+\lambda} p^2 q^2 \right)^{1/4},$$

where σ is a quantity close to the modulus of the wave number in a viscous wave in a cholesteric crystal; k_0 is a wave number that coincides, as will be shown below in the analysis of Eq. (12), with the wave number of the domain structure produced when the CLC layer is stretched along the crystal axis.

Minimizing D with respect to the wave number k , we obtain the value of k at the threshold of the onset of domain, and the threshold velocity gradients:

$$k = (2\sigma k_0)^{1/2}, \quad (9)$$

$$\left(\frac{2\gamma+4\eta}{\mu_3+\alpha_5} - 1 \right) \overline{\langle v_{zz} \rangle^2} + \frac{2\eta-\gamma}{\mu_3+\alpha_5} \overline{\langle v_z v_{z,zz} \rangle} \\ = \frac{K\rho(1+\lambda)}{(\mu_3+\alpha_5)(\mu_3+\alpha_6)} \frac{(k_0^2+2\sigma^2)^2}{8\sigma^4}.$$

In the subsequent analysis of the effect we confine ourselves to total reflection of the sound from the boundary $z = h$, assuming the boundary to be solid; the sound field in the layer is then determined by the standing wave

$$v_z = 2v_0 e^{-i\omega t} \sin [\omega(z-h)/c].$$

Averaging in Eq. (9) with allowance for the inequality $h\omega/c < 1$, we obtain the threshold velocity amplitude in the form

$$v_{0,\text{thr}} = c [K\rho(1+\lambda) / (2\alpha_4 + \gamma - \mu_3) (\mu_3 + \alpha_6)]^{1/2} (2\sigma^2 + k_0^2) / 4\sigma^2. \quad (10)$$

At low frequencies, when the inequality $2\sigma^2 = \rho\omega / \eta \ll k_0^2$, is satisfied, the threshold velocity amplitude is inversely proportional to the frequency: $v_{0,\text{thr}} \propto 1/\omega$; at high frequencies, when $2\sigma^2 \gg k_0^2$, the velocity $v_{0,\text{thr}}$ is independent of frequency and is equal to

$$v_{0,\text{thr}} = v_{0,\text{thr}}^{(\infty)} = \frac{1}{2} c [K\rho(1+\lambda) / (2\alpha_4 + \gamma - \mu_3) (\mu_3 + \alpha_6)]^{1/2}. \quad (11)$$

Setting the parameters in (11) equal to values typical of nematic crystals, viz., $c \approx 10^5 \text{ cm} \cdot \text{s}^{-1}$, $K \approx 0.5 \cdot 10^{-6} \text{ dyn}$, $\alpha_4 \approx \gamma \approx \mu_3 \approx 1 \text{ P}$, $\alpha_6 \approx 0$, $\rho \approx 1 \text{ g} \cdot \text{cm}^{-3}$ (Refs. 12 and 15), and putting $\lambda \ll 1$ we obtain for the threshold velocity amplitude the estimate $v_{0,\text{thr}}^{(\infty)} \approx 25 \text{ cm/s}$.

The domain dimension d in an unstretched CLC layer is given by

$$d = 2^{1/2} \pi / k = \pi (\sigma k_0)^{-1/2} = \pi [(1+\lambda) / 4\lambda p^2 q^2 \sigma^4]^{1/4}$$

and its dependence on the layer thickness, the helix pitch,

and the frequency is given by $d \sim h^{1/4} P_0^{1/4} \omega^{-1/4}$.

The wave-number relation $p < k < q$ used in the derivation of (9) holds for frequencies satisfying the inequality

$$4\pi^2 \eta P_0 / \lambda^{1/2} \rho h^3 < \omega < 4\pi^2 \eta h / \lambda^{1/2} \rho P_0^3,$$

restricting further the region of validity of the results obtained for the frequency.

We consider now the action of sound on a CLC layer stretched along the axis. Stretching of the layer takes place, in particular, if the boundaries are not parallel near the Grandjean lines in a medium with paired disclinations, on which the number of cholesteric layers changes. Stretching of the CLC structure does not affect the form of the hydrodynamic equations (1), but alters substantially the Frank elastic energy, for which we obtain a new expression by retaining the third powers of the variables in the expansion of the energy over the angles. This leads to the appearance of a term $K\tilde{\varphi}_z (\nabla_{\perp} \varphi)^2 / 2q$, where $\tilde{\varphi} = -qz\delta' + \varphi$ is the total molecule rotation angle due both to rotation of the molecules through an angle φ and to the displacement of the molecules when the layer is stretched, and δ' is the tension strain. Discarding next the third powers of the angles φ and θ we obtain for G

$$G = \frac{1}{2} K \int_V \{ (\nabla_{\perp} \varphi)^2 (1 - \delta') + \lambda (\nabla_{\parallel} \varphi)^2 + (\nabla \theta)^2 + q^2 \theta^2 + 4q\theta (\mathbf{n} \nabla) \varphi + (\lambda - 1) [n_1^2 (\nabla_2 \theta)^2 + n_2^2 (\nabla_1 \theta)^2] \} dV. \quad (12)$$

The minimum of the elastic energy for perturbations, periodic along the layer, of the angles θ and φ makes possible the formation domains of the square-grid type at a tension strain equal to $\delta'_{\text{thr}} = (1 + \lambda) k_0^2 / 2q^2$; the wave number of this structure is equal to the previously introduced wave number k_0 .

Calculation of the effect for a stretched CLC layer is quite similar to the foregoing calculation for an unstretched layer, but with a free energy in the form (12), and leads likewise to the appearance of square-grid domains. Leaving out the intermediate steps, we present the final expression for the value of D at which domains are produced:

$$D = \frac{K\rho\omega^2}{(\mu_3 + \alpha_6)(\mu_3 + \alpha_5)} \frac{(k^4 + 4\sigma^4)(k^4 + k_0^4 - k^2 k_0^2 \delta)}{4k^4 \sigma^4},$$

where $\delta = 2\delta' / \delta'_{\text{thr}}$ is the reduced tension strain. The value $\delta = 2$ corresponds to the critical stretching, at which a domain structure appears also in the absence of a sound field. We assume hereafter $0 < \delta < 2$.

Considering, as in the preceding case, total reflection of incident sound wave from the boundary $z = h$, and assuming as before that $h\omega/c < 1$, we obtain the following expression for v_0 :

$$v_0 = \frac{1}{4} c \left[\frac{K\rho(1+\lambda)}{(2\alpha_4 + \gamma - \mu_3)(\mu_3 + \alpha_6)} \right]^{1/2} \times \left[\frac{(k^4 + 4\sigma^4)(k^4 + k_0^4 - k^2 k_0^2 \delta)}{k^4 \sigma^4} \right]^{1/2}.$$

We obtain again the wave number k of the domains at the threshold of the effect, and the threshold velocity amplitude $v_{0,\text{thr}}$, by minimizing the expression for v_0 with respect to k ; this yields for k the equation

$$2\kappa^4 - \delta\kappa^3 + (4\sigma^4/k_0^4)\delta\kappa - 8\sigma^4/k_0^4 = 0, \quad \kappa = k^2/k_0^2.$$

At high frequencies, when the inequality $\sigma > 2^{1/2} k_0 / \delta$ is satisfied, we obtain $k = (2/\delta)^{1/2} k_0$ and

$$v_{0,\text{thr}} = 1/4 c [K\rho(1+\lambda)(4-\delta^2)/(2\alpha_4 + \gamma - \mu_3)(\mu_3 + \alpha_6)]^{1/2}. \quad (13)$$

The domain size is in this case

$$d = 1/2 [(1+\lambda)/\lambda]^{1/4} (\delta P_0 h / 2)^{1/2} \sim (P_0 h)^{1/2}$$

and is independent of the sound frequency. The threshold velocity amplitude is independent of the sound frequency, of the layer thickness h , of the cholesteric helix pitch P_0 , and of the frequency ω .

At low frequencies, when the inequality $\sigma < \delta^{1/2} k_0 / 2$ holds, we obtain accordingly $k = (\delta/2)^{1/2} k_0$, and

$$v_{0,\text{thr}} = 1/8 c [K\rho(1+\lambda)(4-\delta^2)/(2\alpha_4 + \gamma - \mu_3)(\mu_3 + \alpha_6)]^{1/2} k_0^2 / \sigma^2.$$

In this case the domain size is

$$d = 1/2 [(1+\lambda)/\lambda]^{1/4} (2P_0 h / \delta)^{1/2}$$

and is also independent of ω ; for the threshold velocity amplitude we get the relation $v_{0,\text{thr}} \sim h^{-1} P_0^{-1} \omega^{-1}$.

The dependence of the domain size on the frequency in the frequency transition region $\sigma \sim k_0$ is shown in Fig. 1, on which are plotted the relative domain size d/d_0 ($d_0 = 2^{1/2} \pi / k_0$ is the size of the domains produced by stretching the CLC layer) vs the dimensionless parameter $\Omega = 4\sigma^4 / k_0^4 \sim \omega^2$ for different values of δ . The dependence of the threshold velocity amplitude on the frequency is shown in Fig. 2, on which is plotted the relative threshold amplitude $v_{0,\text{thr}} / v_{0,\text{thr}}^{(\infty)}$ (here $v_{0,\text{thr}}^{(\infty)}$ is the threshold velocity amplitude at high frequencies and at $\delta = 0$, while $v_{0,\text{thr}}^{(\infty)}$ is given by Eq. (11)).

Let us compare our results with the experimental data of Refs. 9 and 10, where the effect of ultrasound on the CLC structure was investigated. The ultrasound frequency was varied in the range 0.3–3 MHz. The cited papers report the appearance of a domain structure of the square-grid type with sides parallel and perpendicular to the rub-in lines of the boundary surface; the grid appears only on individual sections of the CLC layer. The domain dimensions are proportional to $(P_0 h)^{1/2}$ and decrease weakly with increase of frequency; the threshold velocity amplitude $v_{0,\text{thr}}$ does not depend on the layer thickness h . The experimental picture of the action of ultrasound on a CLC layer agrees qualitatively with the theory developed for a stretched structure at values of the parameter δ close to 2; it appears that in the experiment the stretched regions are bounded by disclinations.

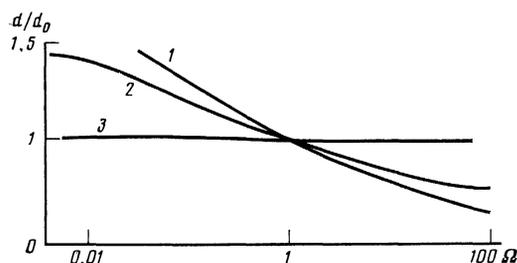


FIG. 1. Reduced domain dimension d/d_0 vs the parameters $\Omega = 4\sigma^4/k_0^4$ at different degrees of layer stretching: 1— $\delta = 0.5$; 2— $\delta = 1$; 3— $\delta = 1.95$.

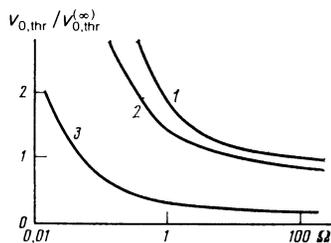


FIG. 2. Reduced threshold velocity $v_{0,thr}/v_{0,thr}^{(\infty)}$ vs the parameter $\Omega = 4\sigma^4/k_0^4$ at different degrees of layer stretching: 1— $\delta = 0.5$; 2— $\delta = 1$; 3— $\delta = 1.95$.

A numerical comparison of the results with the experimental data is impossible without the use of free parameter, since δ remains undetermined in the experiment, and there are no published data on λ for CLC. We confine ourselves here therefore to estimates of δ and λ such that the equations for d and $v_{0,thr}$ agree numerically with the experimental data. At $f = 1.2$ MHz, $h = 4 \cdot 10^{-3}$ cm, and $P_0 = 2 \cdot 10^{-4}$ cm the experimental domain size is $d \approx 1.4 \cdot 10^{-3}$ cm (Ref. 10). Equating the theoretical domain size at $\delta = 2$ to the experimental one we obtain the ratio of the elastic moduli: $\lambda = K_2/K_3 \approx 10^{-2}$. For the same frequency, at $h = 4 \cdot 10^{-3}$ cm and $P_0 = 4 \cdot 10^{-4}$ cm experiment yields $v_{0,thr} \approx 1.2$ cm/s.¹⁰ Taking into account at these data the estimate $\sigma/k_0 \approx 1.25 > 2^{1/2}/\delta \approx 0.7$, we determine the threshold velocity amplitude from Eq. (13). Substituting in this equation the value of the elastic constant and of the viscosity coefficients,

previously chosen for the determination of $v_{0,thr}$ in a non-stretched structure, we estimate the parameter δ at which the theoretical and experimental values of $v_{0,thr}$ agree: the estimate yields $\delta \approx 1.95$.

¹E. N. Kozhevnikov, Zh. Eksp. Teor. Fiz. **82**, 161 (1982) [Sov. Phys. JETP **55**, 96 (1982)].

²E. L. Vinogradova, O. A. Kapustina, V. N. Reshetov, *et al.*, Akust. Zh. **31**, 17 (1985) [Sov. Phys. Acoust. **31**, 10 (1985)].

³P. Pieranski and E. Guyon, Phys. Rev. Lett. **39**, 1280 (1977).

⁴E. N. Kozhevnikov, Zh. Eksp. Teor. Fiz. **91**, 1346 (1986) [Sov. Phys. JETP **64**, 793 (1986)].

⁵P. Pieranski and E. Guyon, Phys. Rev. **A9**, 404 (1974).

⁶O. A. Kapustina and V. N. Lupanov, Abstracts, 4th Internat. Socialist-Block Internat. Conf. on Liquid Crystals, Tbilisi, 1981, Vol. 2, p. 64.

⁷F. F. Legusha, V. A. Murga, and A. N. Slavin, Akust. zh. **29**, 84 (1983) [Sov. Phys. Acoust. **29**, 47 (1983)].

⁸I. N. Gurova, O. A. Kapustina, V. N. Lukanov, and G. S. Chilaya, Ref. 6, p. 47.

⁹O. A. Kapustina, Mol. Cryst. Liq. Cryst. **112**, 1 (1984).

¹⁰M. J. Stephen and J. P. Straley, Rev. Mod. Phys. **46**, 617 (1974).

¹¹T. C. Lubensky, Phys. Rev. **A6**, 452 (1972).

¹²P. G. de Gennes, *Physics of Liquid Crystals*, Oxford, 1977.

¹³N. Scarmuzzo, G. Bartolina, and G. Barbero, Mol. Cryst. Liq. Cryst. **70**, 315 (1981).

¹⁴I. Janossy, J. de Phys. **41**, 437 (1980).

¹⁵V. A. Balandin, A. N. Larionov, and S. V. Pasechnik, Zh. Eksp. Teor. Fiza. **83**, 2121 (1982) [Sov. Phys. JETP **56**, 1230 (1982)].

Translated by J. G. Adashko