Domain structure in a normally oriented nematic liquid crystal layer under the action of low-frequency shear

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(Submitted 19 March 1986)
Zh. Eksp. Teor. Fiz. **91**, 1346–1351 (October 1986)

An analysis of nonlinear equations of hydrodynamics for a nematic liquid crystal is used to show that the action of low-frequency shear vibrations on a layer of a normally oriented nematic creates a spatially periodic structure in the form of domains elongated at right-angles to the shear direction. The orientation of molecules in the domains, the domain size, and the threshold amplitude of the vibrations are determined. The results of the calculations are compared with experimental data.

Investigations of the influence of a periodic shear on a layer of a normally oriented nematic liquid crystal have revealed effects both linear and nonlinear in the shear strain. The former include a periodic (at the shear frequency) deflection of molecules from the normal, which results in modulation of a light flux transmitted by a nematic liquid crystal layer.[1] The nonlinear effects give rise to steady-state distortions of the structure both inhomogeneous along the layer in the case of elliptic shear[2] and periodic along the layer, manifested in the form of elongated stripes or domains.[3–8] The domains appear only above a certain threshold; the directions of the domains, their width, and the threshold amplitude of the shear depend on the nature of the shear effect, frequency, and layer thickness. An elliptic shear acting on a nematic liquid crystal layer creates a structure in which the domain directions depend on the ellipticity of the shear.[3] This effect can be explained theoretically by an allowance for the quadratic (in the angles of rotation of the molecules) viscous moments[4]; in the absence of ellipticity the effects should disappear.

A domain structure of different nature has been investigated experimentally[5–8] and in this case a shear strain has been created by moving one of the boundary plates in its plane in just one direction, so that there has been no ellipticity. In this case the domains are orthogonal to the direction of shear. At low frequencies (~ 1 Hz) the threshold amplitude of the shear and the domain width exceed considerably the thickness of a nematic liquid crystal layer.[5] When the frequency is increased by two or three orders of magnitude, the threshold amplitudes are already less than the layer thickness.[6] The investigations reported in Refs. 7 and 8 were carried out at high frequencies when the initial shear perturbation was concentrated near a boundary in a region of thickness of the order of the wavelength of a viscous wave. A theoretical attempt to describe domains in a nematic liquid crystal layer made in Refs. 9 and 10 was concerned with a periodic shear strain, but the picture developed there does not fit that observed experimentally: in particular, the threshold values of the shear amplitude do not agree with the experimental values in respect to magnitude, do not exhibit the experimentally observed dependences on the frequency and on the layer thickness, and fail to give also the domain dimensions.

We shall develop a theory of the appearance of a domain structure in a normally oriented nematic liquid crystal layer subjected to a low-frequency shear (when the wavelength of a viscous wave in a liquid crystal is greater than the layer thickness), created by the motion of one of the plates in its plane. We shall analyze the effect on the basis of equations of hydrodynamics of a nematic liquid crystal retaining the quadratic terms proportional to the product of the angle of rotation of the molecules and the velocity of the liquid. Inclusion of these quadratic terms leads to the following picture of the effect. A periodic shear in the case of a random slowly varying and periodic (along the layer) deviation of molecules from the normal creates eddies oscillating at the shear frequency; the dimensions of the eddies are the same as the spatial period of distortion of the structure. The interaction of oscillatory eddies with the initial shear field gives rise to an average (per oscillation period) moment which increases the deflection of the molecules. At the threshold the effect may be stabilized the Frank elastic moment. The angle of steady-state deflection (rotation) of the molecules governing the appearance of domains and the eddy flow velocities are related via a self-consistent system of equations with coefficients containing the initial shear strain. The condition for solvability of the system determines simultaneously the threshold amplitude of the shear above which a domain structure appears, and the domain dimensions.

The equations describing the rotation of molecules and their motion in a nematic liquid crystal are[11]

$$[\mathbf{n}\{\gamma(\mathbf{N}-\hat{v}\mathbf{n})-\nabla_i\partial g/\partial(\nabla_i\mathbf{n})+\partial g/\partial\mathbf{n}\}]=0,$$
$$\rho\dot{\mathbf{v}}=-\nabla P+\nabla\hat{\sigma}. \qquad (1)$$

Here, $\mathbf{n}$ is the director governing the direction of alignment of molecules ($\mathbf{n}^2=1$); $\mathbf{v}$ is the velocity; $\hat{v}$ is the strain rate tensor; $\mathbf{N}=\dot{\mathbf{n}}-\frac{1}{2}\,\mathrm{curl}[\mathbf{v}\times\mathbf{n}]$ is the rotation of molecules relative to the surrounding liquid; g is the Frank elastic energy, which in the one-constant approximation is described by

$$g={}^1/_2K\sum_i(\nabla n_i)^2,\quad i=x,\ y,\ z,$$

where K is the Frank elastic modulus; ρ is the density; P is the pressure; $\hat{\sigma}$ is the tensor of elastic stresses with the com-

 0038-5646/86/100793-04$04.00

ponents

$$\sigma_{ij}=\alpha_4 v_{ij}+\alpha_2 N_i n_j+\alpha_5 v_{ik} n_k n_j+\alpha_6 v_{jk} n_k n_i;$$

α_i is the Leslie viscosity coefficient. The small viscosity coefficients α_1 and α_3 obtained in writing down the system (1) and the expressions for σ_{ij} are assumed to be zero; the rotational viscosity coefficient is $\gamma \approx \alpha_5 - \alpha_6 \approx -\alpha_2'$.

We shall consider the growth of domains in the shear plane xz governed by the shear direction (x axis) and by the normal to the layer (z axis) on the assumption that the rotation of molecules occurs in the xz plane and the variables are independent of y, in which case we find that $v_y \equiv 0$. This distribution of molecules in the domains will be justified by an estimate of the stability of the structure, made below, against the deflection of molecules out of the shear plane. We shall assume that the initial shear strain is specified as follows: the lower ($z = 0$) boundary is at rest, the motion of the upper ($z = h$, where h is the layer thickness) boundary plate is in its own plane at a frequency ω and with an amplitude u_0, which gives

$$v_x|_{z=h}=\omega u_0 \cos \omega t.$$

We shall consider shear amplitudes which are small compared with the layer thickness: $u_0 < h$. This condition means that the angle φ of deflection of the molecules from the z axis in the shear field is small and allows us to linearize the equations of motion with respect to the angle. At low values of φ the components of the director $\mathbf{n}$ are of the form

$$n_x \approx \varphi, \quad n_y=0, \quad n_z \approx 1.$$

Retaining in the system (1) the terms proportional to the product of the angle φ and the velocity v_x or v_z, we obtain the following system of equations for φ, v_x, and v_z:

$$\begin{gathered}
\gamma\dot{\varphi}-K\Delta\varphi=\gamma v_{x,z}+\gamma\varphi(v_{xx}-v_{zz}),\\
\rho\dot{v}_x-\alpha_4 v_{x,xx}-(\eta+\gamma)v_{x,zz}-\eta v_{z,xz}+\gamma\dot{\varphi}_{,z}=-P_{,x}-(\gamma-\alpha_5)(\varphi v_{x,z})_{,x}\\
+\alpha_5(\varphi v_{xx})_{,x}+\alpha_6(\varphi v_{z,x})_{,x}+\alpha_6(\varphi v_{zz})_{,z}, \qquad (2)\\
\rho\dot{v}_z-(\alpha_4+\alpha_5+\alpha_6)v_{z,zz}-\eta v_{z,xx}-\eta v_{x,xz}=-P_{,z}+(\gamma+\alpha_6)(\varphi v_{x,z})_{,z}\\
+\alpha_6(\varphi v_{xx})_{,x}+\alpha_5(\varphi v_{z,x})_{,z}+\alpha_5(\varphi v_{zz})_{,x},
\end{gathered}$$

where $\eta = \frac{1}{2}(\alpha_4+\alpha_5-\gamma)$; the dot in the above equations represents the total derivative with respect to time and the indices after the comma represent the partial derivatives with respect to the indicated coordinates.

We shall describe the variables φ, v_x, and v_z by the expressions

$$\varphi=\varphi_0+\varphi_1+\varphi', \quad v_\alpha=v_{0\alpha}+v_{1\alpha}+v_\alpha', \quad \alpha=x, z,$$

where φ_1, v_{1x}, and v_{1z} represent the initial perturbation set by the periodic shear of the boundary plate and found by solving the linear equations of hydrodynamics; the index 0 denotes the steady-state part of the corresponding quantity; φ' and v_α' are the additional oscillations of the angle and velocity which appear in the case of steady-state distortion of the structure and are proportional to φ_0.

We shall consider only the frequencies ω satisfying the inequalities

$$K\pi^2/\gamma h^2 \ll \omega \ll \rho c^2/\eta, \quad \omega < \eta/\rho h^2,$$

where c is the velocity of sound in the investigated liquid crystal. The first of these inequalities makes it possible to simplify the equations for φ_1 and φ' by dropping the elastic moments compared with the viscous moments. The second condition allows us to analyze the effect by discussing only incompressible oscillatory flows and to ignore the acoustic modes, the velocities of which are $\rho c^2/\eta\omega \gg 1$ times less the velocities in the flows and which therefore do not affect the growth of domains. It also follows from the second inequality that the initial perturbation can be described by

$$v_{1x}=(\omega u_0 z/h)\cos\omega t, \quad v_{1z}=0, \quad \varphi_1=(u_0/h)\sin\omega t. \qquad (3)$$

Retaining in the system (2) the terms proportional to the product of the angle of rotation of molecules and the velocity, and separating the eddy component of the velocities, we obtain the following system of equations for oscillatory perturbations v_α' and φ':

$$\begin{gathered}
[(\rho/\gamma)\partial_t\Delta-D]v_x'=-\varphi_{0,xz}v_{1x,z},\\
\operatorname{div}\mathbf{v}'=0, \quad \varphi'_{,t}\approx v'_{x,z}-\varphi_{0,x}v_{1x}
\end{gathered} \qquad (4)$$

and in the case of steady-state perturbations φ_0 and $\mathbf{v}_0$, the relevant equations are

$$\begin{gathered}
Dv_{0x}-(K/\gamma)\Delta\varphi_{,z^3}=-[\langle\varphi_1 v'_{x,xz^3}\rangle+\langle\varphi'_{,,zz}v_{1x,z}\rangle+\langle\varphi_1 v'_{z,zzxx}\rangle\\
+\langle\varphi_1(v'_{z,zz}-v'_{z,xx})_{,xx}\rangle]-(\rho/\gamma)\langle(\Delta v_z' v_{1x})_{,z}\rangle,\\
\operatorname{div}\mathbf{v}_0=0, \quad (K/\gamma)\Delta\varphi_0+v_{0x,z}=[2\langle\varphi_1 v'_{z,z}\rangle+\langle\varphi'_{,x}v_{1x}\rangle],
\end{gathered} \qquad (5)$$

where D is the differential operator:

$$D=(\eta/\gamma)\partial_x^4+[(\alpha_4+\gamma)/\gamma]\partial_x^2\partial_z^2+(\eta/\gamma)\partial_z^4,$$

and the angular brackets denote averaging over one oscillation period.

Equations (4) and (5) are simplified by dropping the terms proportional to the product of φ_1 or v_{1x} and the steady-state flow velocity v_{0x}, because the order of smallness of such products is given by $K\rho^2/\gamma \sim 10^{-6} \ll 1$ compared with the other terms. Eliminating from the equations the oscillatory components and the steady-state flow velocities, and then averaging, we obtain the following equation for φ_0:

$$\begin{gathered}
K(D+\partial_z^4)[D^2+(\rho^2\omega^2/\gamma^2)\Delta^2]\varphi_0\\
=(\rho v_0^2/2h)\{(1+4\eta/\gamma)\partial_z^2+2[(\alpha_4+\alpha_5)/\gamma]\partial_x^2\}\partial_z^4\partial_x^2\varphi_0. \qquad (6)
\end{gathered}$$

We shall seek a solution for φ_0 which is periodic along the x axis and satisfies the condition of rigid binding of the molecules to the boundary ($\varphi|_{z=0,h}=0$); this solution φ_0 is

$$\varphi_0=\varphi_{00}\cos kx \sin pz, \qquad (7)$$

where $p = \pi/h$ and the wave number k will be found later from the condition for a minimum (with respect to k) of the shear amplitude u_0 for which Eq. (6) has a nonzero solution and, consequently, a steady-state distortion of the structure is possible. Substituting φ_0 from Eq. (7) into Eq. (6), we find the following expression for the amplitude u_0:

$$u_0=(2K/\rho)^{1/2}(\pi/\omega h^2)F(\xi),$$

where $\xi = k^2/p^2$ and the function $F(\xi)$ is of the form

$$F(\xi)=\{(1+\tilde{D})[\tilde{D}^2+a(1+\xi)^2]/\xi[1+4\eta/\gamma+2\xi(\alpha_4+\alpha_5)/\gamma]\}^{1/2},$$

$$a=\frac{\rho^2\omega^2}{\gamma^2 p^4}=\frac{\rho^2\omega^2 h^4}{\gamma^2\pi^4},\qquad \tilde{D}=\frac{\eta}{\gamma}+\frac{\alpha_4+\alpha_5}{\gamma}\xi+\frac{\eta}{\gamma}\xi^2.$$

Minimizing the function $F(\xi)$ with respect to ξ, we obtain ξ_0 which governs the spatial periodicity of the domain structure at the threshold of the effect, as well as the value of the threshold amplitude $u_{0,\text{th}}$:

$$u_{0,\text{th}}=\left(\frac{2K}{\rho}\right)^{1/2}(\pi/\omega h)F(\xi_0). \tag{8}$$

At the frequencies under consideration the function $F(\xi_0)$ depends weakly on the frequency; the threshold amplitude is then inversely proportional to the frequency ω and to the layer thickness h. The first of these dependences was observed experimentally in Ref. 6, where the appearance of a domain structure was deduced from a diffraction pattern obtained on illumination of a nematic liquid crystal layer with a light beam.

A numerical calculation of $u_{0,\text{th}}$ from Eq. (8) and a comparison of the results obtained with experimental data was made for an MBBA liquid crystal with the following parameters: $\rho = 1$ g/cm^3, $K = 0.7\times 10^{-6}$ dyn, $\alpha_4 = 1.04$ P, $\alpha_5 = 0.46$ P, $\gamma = 0.78$ P (Ref. 11), which gives $\eta/\gamma = 0.32$ and $(\alpha_4+\alpha_5)/\gamma = 1.92$. The values of $k_0/p = \xi_0^{1/2}$ found by numerical calculation (k_0 is the wave number k at the threshold of the effect) and $F(\xi_0)$ are plotted in Fig. 1 as a function of the parameter a. At low frequencies, when $a \ll 1$, the ratio k_0/p is $k_0/p \approx 0.5$. This gives the following expression for the domain size: $d \approx \pi/k_0 \approx 2h$.

Figure 2 shows the theoretical dependences of $u_{0,\text{th}}$ on the frequency $f=\omega/2\pi$ for layers of two thicknesses: $h = 100$ and $20\,\mu$; this figure includes also the experimental values of $u_{0,\text{th}}$ for the same values of h and different frequencies.[6] Curve 1 ($h = 100\,\mu$) is plotted for frequencies $f \lesssim 10^4$ Hz, because the low-frequency condition no longer holds at higher frequencies; curve 2 ($h = 20\,\mu$) is plotted for frequencies $f \gtrsim 10^3$ Hz, because at lower frequencies the condition of smallness of the shear amplitude u_0 is not obeyed. Figure 2 shows that in the range of validity of our calculations the theory and experiment agree well.

We shall now estimate the stability of the new structure against deflection of molecules out of the shear plane by a small angle ψ_0 which is homogeneous in the layer. In the case of homogeneity of ψ the elastic moments associated with ψ are absent and the stability of the structure is governed by the ratio of the viscous moments. If the angle ψ_0 above the threshold of formation of a domain structure is finite, we find that the system (1) leads to the following equation for a homogeneous angle ψ_0, which varies with time more slowly than does the frequency:

$$\psi_{0,t}\overline{\sin^2\varphi_0}\approx-\overline{\langle\varphi_1 v'_{y,x}\rangle\sin\varphi_0\cos\varphi_0}-\overline{\langle\psi' v_{1x,z}\rangle\sin\varphi_0\cos\varphi_0}. \tag{9}$$

Here the bar denotes averaging over the volume of the nematic liquid crystal layer; v_v' is the velocity of the oscillatory flow along the y axis which appears when $\psi\neq 0$; ψ' represents oscillations of the angle ψ at the frequency ω. In the derivation of the system (1) we allowed for the fact that the component n_x and the velocity v_y', which vanish at the boundaries of the layer, can be represented as series in $\sin mpz$ ($m = 1,2,3,\ldots$) and terms of the $n_x v'_{y,z}$ type as a result of averaging over the volume.

For ψ' and v_y' we find that the system (1) yields the following equations:

$$\psi'_{,t}\sin^2\varphi_0=-v'_{y,z}\sin^2\varphi_0-\psi_0 v_{1x,z}\cos\varphi_0\sin\varphi_0+v_{y,z}\cos\varphi_0\sin\varphi_0,$$

$$(\rho\partial_t-\eta\Delta)v_y'=[(\alpha_6/2+\gamma\sin^2\varphi_0)\sin\varphi_0\cos\varphi_0)\varphi_0 v_{1x,z}. \tag{10}$$

Assuming that the angle φ_0 beyond the threshold of the effect varies along x as a series in $\cos mkx$ ($m = 1,2,3,\ldots$) we shall seek the velocity v_y' in the form $v_y' \sim \sin pz \sin kx$. Using the expressions for v_y' and ψ' from Eq. (9) and substituting them in the system (10), we find—after averaging—the following equation for ψ_0:

$$\psi_{0,t}\overline{\sin^2\varphi_0}=S_1\left(\frac{\alpha_6}{2}S_1+\gamma S_3\right)\frac{\rho v_0^2 k^2}{h[\rho^2\omega^2+\eta^2(k^2+p^2)^2]}\psi_0,$$

where the following notation is employed:

$$S_1=\overline{\sin\varphi_0\cos\varphi_0\cos kx\sin pz}>0,$$

$$S_3=\overline{\sin^3\varphi_0\cos\varphi_0\cos kx\sin pz}>0.$$

The behavior of the perturbations ψ_0 depends on the amplitude of the angle of rotation of the molecules φ_{00} in the do-

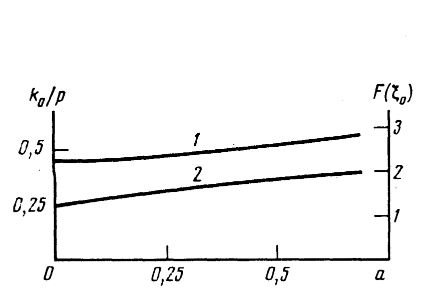

FIG. 1. Dependences of k_0/p (1) and $F(\xi_0)$ (2) on the parameter $a=\rho^2\omega^2 h^4/\gamma^2\pi^4$.

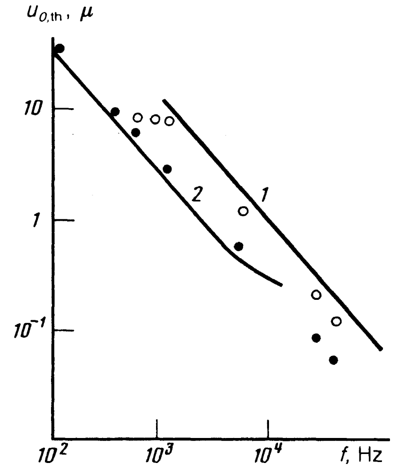

FIG. 2. Theoretical (continuous curves) and experimental dependences of the threshold displacement on the frequency: 1) $h = 20\,\mu$ (○); 2) $h = 100\,\mu$ (●).

mains. In the case of small values of φ_{00}, we find that $|\alpha_6|S_1 > 2\gamma S_3$ and perturbations decay exponentially with time. In the case of high values of φ_{00}, when $|\alpha_6|S_1 < 2\gamma S_3$, the angle ψ_0 rises with time, the molecules are deflected out of the plane of the shear, and the domain structure changes. The threshold value $\varphi_{00,\mathrm{th}}$ can be estimated by representing the angle φ_0 in the form $\varphi_0 \approx \varphi_{00} \cos kx \sin pz$. This gives

$$\varphi_{00,\mathrm{th}} \approx (-8\alpha_6/9\gamma)^{1/2}.$$

In the case of MBBA, we have $\varphi_{00,\mathrm{th}} \approx 0.5$

Inhomogeneous perturbations of ψ at low values of φ_0 may be suppressed not only by viscous moments of the type discussed above, but also by the Frank elastic moments $\Gamma_\psi = K\nabla(\nabla\psi \sin^2 \varphi_0)$. The domain structure is also stable in the presence of such moments. Therefore, directly above the threshold of the effect the molecules are in the shear planes and the domain structure has the form discussed above.

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Translated by A. Tybulewicz