

# Undamped domain structure oscillations in nematic liquid crystals in dc and ac electric fields

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Low-frequency oscillations of domain structures of the two-dimensional mosaic type domains, Williams-Kapustin domains, are described. Criteria for the formation of the oscillations and the basic laws for the dependence of the oscillation frequency and domain size on the voltage are obtained experimentally and theoretically.

In recent years there has been a significant growth of interest in investigations of nematic liquid crystal-smectic liquid crystal phase transitions and there has been intensive study of the behavior of the physical parameters of the nematic in the immediate neighborhood of the phase transition. It has been discovered that in this case a “helium analog” occurs, i.e., the behavior of various quantities is analogous to the behavior of a number of parameters of helium II in the neighborhood of the transition to the superfluid state.<sup>1</sup> For example, the variation of the elastic moduli has the form<sup>2</sup>  $\delta k_i \propto (T - T_c)^{-\alpha}$ , where  $1/2 < \alpha < 2/3$ . The behavior of the pitch  $p$  of the cholesteric spiral in the neighborhood of the cholesteric liquid crystal-smectic  $A$  phase transition is described in a similar way:  $P \propto (T - T_c)^{-\alpha}$ , where  $\alpha \approx 0.67$ . Another notable effect in the vicinity of the phase transition is the vanishing or the change of sign of the coefficient of viscosity  $\alpha_3$ ; this leads to a number of new effects in the flow of a nematic liquid crystal. Up to the present time the exotic nature of the above-mentioned effects consisting of the possibility to observe them only in the vicinity of the phase transition and at high temperatures, and thus they have been of purely theoretical interest. Therefore devising methods of producing nematics having short-range smectic order and existing over a wide temperature range, and the study of the dynamic effects in these materials under the action of electric fields, is an important problem, which was in fact solved in the course of this investigation.

## THE LIQUID CRYSTALS STUDIED, THEIR STRUCTURE AND PRINCIPAL PHYSICAL PROPERTIES

For a number of nematic liquid crystals it has been shown quite persuasively that when complexes of the type macrocyclic polyether (MCPE)—alkali-metal salt are dissolved in them at concentrations above a critical concentration, then smectic liquid crystals are formed.<sup>3</sup> In this investigation we have studied mixtures of the nematic liquid crystal 4-methoxybenzylidene-4-butylaniline (MBBA), MCPE-18-crown-6, and KCl. To change the sign of the anisotropy of the electrical conductivity  $\sigma_a = \sigma_{||} - \sigma_{\perp}$  in the liquid crystal we added tetrabutylammonium bromide, and to change the sign of the dielectric anisotropy of the MBBA the nematic liquid crystal was doped with 4-cyanobenzylidene-4-n-octyloxyaniline having a substantial positive dielectric constant anisotropy  $\epsilon_a$ . The x-ray diffraction structural studies carried out on a KRM-1 low-angle x-ray camera for these

mixtures such as e.g. MBBA + MCPE + KCl indicate the following: If the concentration of 18-crown-6 + KCl is less than a critical concentration  $c_0 \approx 0.15\%$ , then the structure of the mesophase is the same as the classical nematic phase. However, for  $c > c_0$  clusters which are recorded at low angles begin to form and diffraction maxima corresponding to the dimensions  $a_1 \approx 270 \text{ \AA}$ ,  $a_2 \approx 125 \text{ \AA}$ , and  $a_3 \approx 57 \text{ \AA}$  are observed. In addition to these maxima, others are observed at large angles ( $3.8^\circ$  and  $6.2^\circ$ ) corresponding to the dimensions  $a_4 \approx 22 \text{ \AA}$  and  $a_5 \approx 13 \text{ \AA}$ . The spacing  $a_4 \approx 22 \text{ \AA}$  coincides with the length of the MBBA molecules. Therefore these results allow us to suppose that in the region of the critical concentration there is a transition from the nematic phase to a liquid crystal having a pseudolayered structure with the dimensions of the cybotactic groups (e.g. for MBBA about  $270 \text{ \AA}$ ) having a smectic structure. In the case of symmetric liquid crystal molecules, as for instance in *para*-azoxyanisole or the alkoxybenzoic acid series, clusters are not observed.

The investigations of the coefficients of viscosity  $\alpha_i$  ( $i = 1, \dots, 6$ ) by the method described in Ref. 4 showed that these coefficients differed from those of MBBA by 5–10%. The situation is similar also for the elastic constants  $k_{11}$ ,  $k_{22}$ , and  $k_{33}$  determined from investigations of Lorentz broadening of the spectral lines of laser radiation<sup>5</sup>: they differ from those of the original liquid crystal by about 10–15%. The coefficient of viscosity undergoes substantial changes with increasing concentration of 18-crown-6 + KCl. In the present experiments the method of measuring this coefficient was similar to that described by Guyon and Pieranski.<sup>6</sup> The essence of the method is as follows: a nematic liquid crystal of a given orientation was placed in a capillary, one of whose plates could be moved (see the insert in Fig. 1) such that the direction of motion coincided with the initial orientation of the director. The coefficient  $\alpha_3$  was calculated from the value of the maximum angle of inclination  $\theta$  of the director, this angle being related to the coefficients of viscosity  $\alpha_3$  and  $\alpha_2$  by the relation  $\tan^2 \theta = \alpha_2 / \alpha_3$ . The angle  $\theta$  was determined from measurements of the birefringence, equal to

$$\Delta n = d^{-1} \int_0^d \Delta n_0 \sin^2 \theta dz,$$

where  $\Delta n_0 \approx n_0 - n_e$ ,  $n_0$  and  $n_e$  are the refractive indices of the ordinary and extraordinary light beams, respectively, and  $d$  is the thickness of the crystal. The dependence of  $\alpha_3$  on

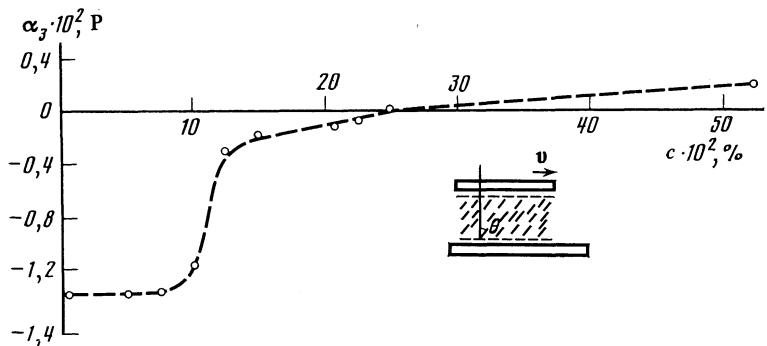


FIG. 1. Coefficient of viscosity  $\alpha_3$  as a function of the concentration  $c$  of the complex MCPE in MBBA. The measuring method is illustrated in the insert.

the concentration of 18-crown-6 + KCl is shown in Fig. 1. It follows from this figure that there is an abrupt change in  $\alpha_3$  in the vicinity of the concentration  $c \approx 0.1\%$ . According to this discussion, the variation of  $\alpha_3$  at concentrations below  $c_0$  can be due to the formation and increase in number of clusters. At  $c_0 \approx 0.25\%$  the coefficient  $\alpha_3$  changes sign and in the vicinity of  $c_0$  we have  $\alpha_3 \propto c - c_0$ .

The dielectric constants of the mixtures nematic liquid crystal + 18-crown-6 + KCl were measured with an R-383 ac bridge, and the electrical conductivity was calculated from  $\tan\delta$ , the tangent of the loss angle. To study the anisotropy of the dielectric constant, the liquid crystal was oriented by a magnetic field  $H \approx 3$  kOe. As a result of the investigations it was determined that the anisotropy of the dielectric constant in the mixtures essentially did not vary. Thus, in MBBA  $\epsilon_a \approx -0.4$ , and in contrast to the pure nematic liquid crystal, the value of  $\epsilon_a$  stayed practically constant up to the phase transition to the isotropic liquid. The conductivity ( $\sigma_{\parallel}$  and  $\sigma_{\perp}$ ) varies with the concentration of MCPE + KCl, but its anisotropy is greater than zero,  $\sigma_a > 0$ ; for MBBA,  $\sigma_{\parallel}/\sigma_{\perp} \approx 1.3$  for voltages  $U < 20$  V.

The domain structures of the nematic liquid crystals were studied with an "Amplival Pol. U" microscope, and the photometric studies of them were carried out with an SFN-10 photographic attachment which made it possible to study areas down to  $1 \mu\text{m}$  in size. Time-dependent processes were recorded on an N-306 plotter or a S8-11 storage oscilloscope. The dc electric field was applied from a B5-11 power supply and the ac field from a G3-33 audio oscillator. The nematic liquid crystal was placed in a planar cell formed of two glass plates having a conducting coating of  $\text{SnO}_2$  and separated by spacers of thickness  $d$ .

#### EXPERIMENTAL RESULTS FOR THE FEATURES OF THE ELECTROHYDRODYNAMIC INSTABILITY OF PSEUDOLAYERED MESOPHASES

##### a) Investigations of electrohydrodynamic instability of nematic liquid crystals in the vicinity of the point $\alpha_3 \approx 0$

The electrodynamic instability of nematic liquid crystals where the coefficient of viscosity  $\alpha_3 < 0$  has been investigated quite thoroughly. There exist isolated investigations of nematic liquid crystals for which, evidently,  $\alpha_3 \geq 0$ , but where  $\epsilon_a$  has different signs.<sup>7</sup> We have undertaken an investigation of the instability of nematic liquid crystals, produced by the methods described above and having short-

range smectic order. In the experiments it was discovered that when an electric field is applied to such a layer of nematic liquid crystal, above a certain critical threshold voltage  $U_{1c}$ , a banded domain structure, Williams-Kapustin domains, are formed. The value of  $U_{1c}$  in the present case is practically independent of the thickness of the sample and the frequency dependence of  $U_{1c}$  is the same as that known from the published literature for nematic liquid crystals with  $\epsilon_a < 0$  and  $\epsilon_a \approx 0$  and it is well described by the theory of Dubois-Violette and DeGennes.<sup>1</sup> However, the character of the instability observed above  $U_{1c}$  is substantially different from the instability in nematic liquid crystals for which  $\alpha_3 < 0$ . These differences depend on the anisotropies  $\epsilon_a$  and  $\sigma_a$  of the nematic liquid crystals, and under the action of dc and ac electric fields have their own particular features, which we should discuss separately.

Above the threshold voltage the equilibrium orientation of the director is governed by the balance of the torques: the torque induced by the conductivity, the torque  $m_o$  of the elastic forces, and the dielectric torque  $m_e$ .<sup>1</sup> The question of the maximum angle of the steady-state orientation of the director has been discussed by various authors. The principal criterion is the vanishing of the torque induced by the velocity gradient  $S$ :

$$m_o = -(\alpha_2 \cos^2 \theta + \alpha_3 \sin^2 \theta) S.$$

The condition for the equilibrium orientation is  $\tan^2 \theta = \alpha_2/\alpha_3$ , and for  $\alpha_3 \approx 0$  we have  $\theta \approx \pi/2$ , but in fact the maximum value of  $\theta$  will be less. This is because of a decrease in the effective electrical conductivity caused by the appearance of a space charge. Carroll has shown<sup>8</sup> that the saturation angle is  $\theta \approx \arctan (\theta_{\parallel}/\sigma_{\perp})^{1/2}$  and a change in sign of  $\alpha_3$  has no important effect. Therefore it is possible to attain the maximum angle  $\theta \approx \pi/2$  for  $\alpha_3 \geq 0$  only under the condition  $\epsilon_a/\epsilon_{\parallel} \gg \sigma_a/\sigma_{\parallel}$  and a substantial positive dielectric anisotropy  $\epsilon_a > 0$ . This case evidently has been studied previously.<sup>9</sup>

##### b) Oscillating domain structures in nematic liquid crystals with $\alpha_3 > 0$ and $\sigma_a > 0$ . DC fields

For the case  $\alpha_3 > 0$  and  $\sigma_a > 0$  the form of the domain structures when the second critical voltage  $U_{2c}$  is reached differs from that described above. (We should mention that here we are considering only those values of  $\sigma_a$  and  $\epsilon_a$  for which electrohydrodynamic instabilities arise). The overall symmetry of the domain structures at the voltage  $U_{2c}$  de-

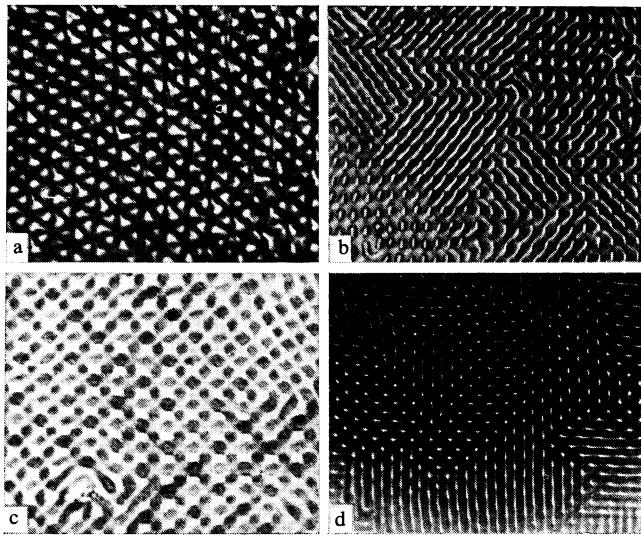
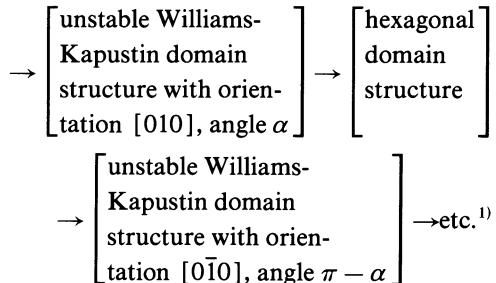


FIG. 2. Oscillating domain structures in dc fields: a)  $U = U_{2c}$ ,  $\alpha_3 \approx 0.01$  P; b)  $U = 10$  V,  $\alpha_3 \approx 0.01$  P, and in ac fields: c)  $\alpha_3 \approx 0.01$  P,  $U = U_{2c}$ ; d)  $U = 10$  V,  $\alpha_3 \approx 0.01$  P,  $U \gg U_{3c}$ .

pends to a large extent on  $\alpha_3$ . For  $\alpha_3 > 0$  the Williams-Kapustin domains are transformed into hexagonal domains, and for  $\alpha_3 \approx 0$  their symmetry is close to trigonal (Fig. 2). In this case there is no change in the domain structure when the voltage is increased, but when  $U \gg U_{2c}$ , turbulent motion of the mesophase sets in.

With increasing coefficient of viscosity  $\alpha_3 > 0$  the symmetry of the two-dimensional structures changes and becomes quasitrigonal. At  $U = U_{3c}$  the two-dimensional domain lattice loses its stability and undamped oscillations arise in the sample with the formation of Williams-Kapustin domains. The orientation of the structures of the Williams-Kapustin domains constitutes an angle of  $\alpha$  and  $\pi - \alpha$  with the initial orientation of the director (at  $U = 0$ ) and coincides with the orientations of the two systems of domain lattices. The period of the Williams-Kapustin domains is one-half the period of one of these systems. The dimension  $L$  of the re-

gions (blocks) of the Williams-Kapustin domains depends on the applied voltage, the thickness  $d$  of the sample, and the temperature  $T$ . Under stationary external conditions ( $U, T, d$ ) = const, the structures of the Williams-Kapustin domains are unstable and transform into a domain lattice and then back into a Williams-Kapustin domain structure. A diagram of the transformations can be written down in the following form:



The frequency of the domain orientation depends on the applied voltage and the thickness of the crystal and weakly on the temperature. These dependences are shown in Fig. 3. Empirical formulas describing the oscillation frequency  $\Omega$  and the dimension  $L$  of the domains in the Williams-Kapustin domain structure showed that they depend on the voltage in the following way:

$$\Omega \approx A[U^2 - (U_{1c} + a_0)^2]^{1/2}, \quad L \approx B[U^2 - (U_{1c} + a_0)^2]^{-1/2}, \quad (1)$$

where  $A$ ,  $B$ , and  $a_0$  are constants that depend on the concentration of MCPE. Here, as follows from investigations of the thickness dependence of the oscillations,  $A \propto d^{-2}$  and  $B \propto d^2$  (Fig. 3).

### c) Oscillating domain structures of nematic liquid crystals for the case $\alpha_3 > 0$ and $\sigma_a > 0$ . AC fields

The case of ac electric fields differs from that which has already been examined in that at the voltage  $U_{2c}$  the Williams-Kapustin domain structures are transformed into a square domain lattice. When the voltage is changed to  $U_{3c}$  the domain structures lose stability and unstable Williams-

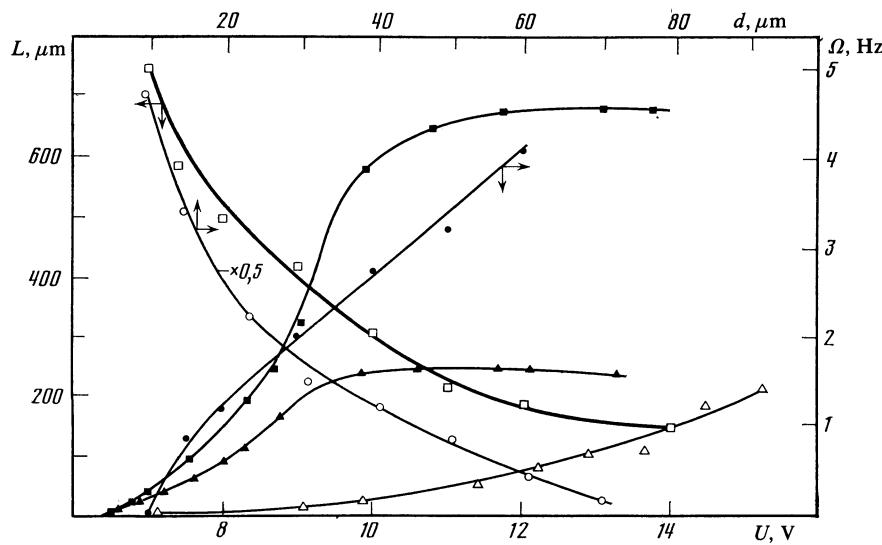


FIG. 3. Voltage dependence of oscillation frequency  $\Omega$  ( $\bullet$ ) and domain size  $L$  ( $\square$ ) in blocks for  $\alpha_3 > 0$  and  $\sigma_a > 0$ . Dependence of  $\Omega$  on the thickness of the crystal ( $\circ$ ), and the voltage dependence of  $\Omega$  for a nematic liquid crystal with  $\alpha_3 < 0$  and  $\sigma_a < 0$ : ( $\blacksquare$ )  $\varepsilon_a \approx -0.1$ ,  $\sigma_1/\sigma_{\parallel} \approx 1.23$ ; ( $\blacktriangle$ )  $\varepsilon_a \approx 0$ ,  $\sigma_1/\sigma_{\parallel} \approx 1.1$ ; ( $\triangle$ )  $\varepsilon_a \approx 0.3$ ,  $\sigma_1/\sigma_{\parallel} \approx 1.03$ .

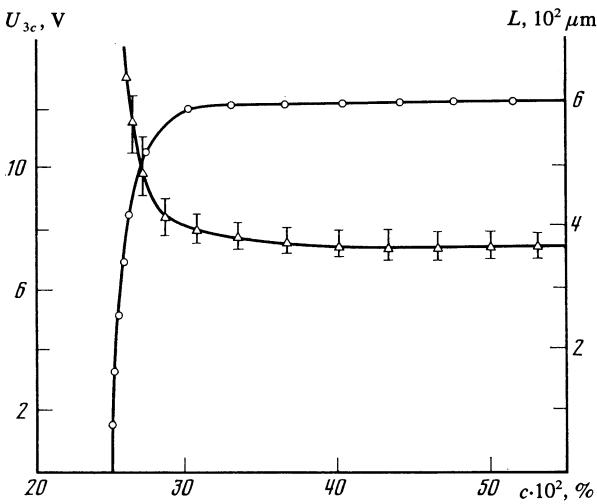
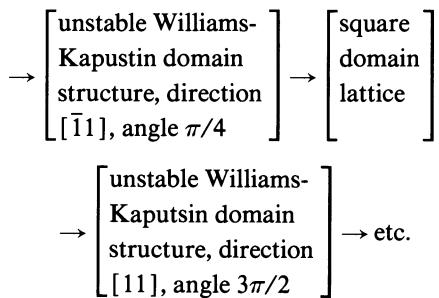


FIG. 4. Dependence of the threshold  $U_{3c}$  ( $\Delta$ ) and of the size  $L$  of domains in blocks ( $\circ$ ) on the concentration of MCPE + KCl in MBBA.

Kapustin domain structures are formed. The undamped oscillations in this case are similar to those in the dc field case, with the orientation of the domains at angles of  $45^\circ$  and  $135^\circ$  to the direction of the director at  $U = 0$  (see Fig. 2). The diagram of the transformations in this case looks like:



The expressions for the oscillation frequency and the domain size  $L$  have a form similar to those presented above (1) for the dc case, but with different coefficients  $A$  and  $B$ . It should be especially noted that while in the vicinity of  $\alpha_3 \approx 0$  the domains are stable for dc fields, in the case of ac fields the oscillations persist to  $\alpha_3 \approx 0$ . This behavior is evidently governed by the symmetry of the domain mosaic. In the ac field case it was possible to measure the frequency  $\Omega$  all the way to  $\alpha_3 = 0$ . It was found that  $L \rightarrow 0$  as  $\alpha_3 \rightarrow 0$ , but with increasing  $\alpha_3$  the domain size in the oscillating structures increases and thereafter is a weak function of  $\alpha_3$  (Fig. 4).

By means of polarization-dependent optical studies of the domains with an optical microscope, it was established that when  $U > U_{3c}$ , in the process of domain oscillation and the formation of Williams-Kapustin domain structures, the director is oriented normal to the axis of the domain, as is the case for Williams-Kapustin domains existing at  $U \approx U_{1c}$ , i.e., there is a rotation of the director relative to the initial orientation, by the angles  $\alpha$  and  $\pi - \alpha$  for dc fields, and  $\pi/4$  and  $-\pi/4$  for ac fields.

A change in the dielectric anisotropy of the nematic liquid crystals from  $-1$  to  $+1$  did not cause the effect to disappear; rather the frequency of the oscillations was prac-

tically independent of the value of  $\epsilon_a$ . It follows from this result that the oscillation processes described here are mainly governed by the sign of  $\alpha_3$  while the characteristic dimensions of the domain structures that arise are determined by  $\epsilon_{||}$ ,  $\epsilon_{\perp}$ ,  $\sigma_{||}$ , and  $\sigma_{\perp}$ , the elastic moduli  $k_{ii}$ , etc. The experimental results obtained here indicate that the conclusions of Penz,<sup>10</sup> who regarded the formation of oscillations as a consequence of the positive sign of the anisotropy of the dielectric constant,  $\epsilon_a > 0$ , are wrong.

#### d) Oscillating domain structures. The case $\alpha_3 < 0$ and $\sigma_a < 0$

In this case, an unstable, nearly square, domain lattice is formed as the voltage is increased to  $U_{2c}$ . The thresholds for the formation of this lattice and for the oscillations coincide, and the diagram of the formation of the Williams-Kapustin domains is the same as that described above. The Williams-Kapustin domains form parallel bands oriented normal to the initial orientation of the director. The dependences of the oscillation frequencies on the voltage at various temperatures differ from those for the case  $\alpha_3 > 0$  and  $\sigma_a > 0$ : at low voltage and in the vicinity of the critical voltage the oscillation frequency goes as the square of the voltage,  $\Omega \propto U^2$  (see Fig. 3), and with increasing voltage the frequency saturates. Corresponding to this result, the size of the Williams-Kapustin domains is on the order of the length of the diagonal of the initial domain lattice.

#### NATURE OF DOMAIN STRUCTURE OSCILLATIONS IN NEMATIC LIQUID CRYSTALS

It follows from the material presented here that the oscillation effect is essentially governed by the coefficient of viscosity  $\alpha_3$  and the anisotropy  $\sigma_a$  of the electrical conductivity. No less important is the symmetry of the domain structures that appear at  $U_{2c}$ ; the domain structures with symmetry close to trigonal are stable.

We shall examine the behavior of a mesophase when there exist a stationary domain structure and perturbations of the velocities and orientation of the director. In such a formulation of the problem the general equations that describe the evolution of the mesophase structure in time and space are nonlinear and in a practical sense impossible to solve. However, if it is assumed that the perturbations are small, then the equations for the velocity, the director, and the field can be linearized. We shall seek a solution of a linearized system of equations of the nematic liquid crystal dynamics for the velocity  $\mathbf{v}_0 + \mathbf{v}$ ,  $\mathbf{v} = (v_x, v_y, v_z)$  and the potential  $\Psi$ . The total potential is  $\Phi = -Ez + \Psi(x, y, z)$  where  $|\Psi| \ll Ed$  and  $E$  is the constant field applied along the  $z$  axis. We shall assume that  $\epsilon_a \approx 0$  and  $k_{11} = k_{22} = k_{33} = k$ . Then the volume charge is

$$\rho_{cr} = -(\epsilon_{\perp}/4\pi) \nabla^2 \Psi,$$

and the bulk elastic energy of the nematic liquid crystal is

$$F = \frac{k}{2} \int_V (\nabla \mathbf{n})^2 dr = \frac{k}{2} \int_0^d (\nabla \mathbf{n})^2 dz.$$

The director

$$\mathbf{n} = (\cos \theta \cos \varphi, \cos \theta \sin \varphi, \sin \theta) \approx (1, \varphi, \theta),$$

( $|\varphi|$ ,  $|\theta| \ll 1$ ) has an initial planar orientation. Let us define

$$\mathbf{h} = k\nabla^2\mathbf{n} = -\delta F/\delta\mathbf{n}, \quad \Gamma = [\mathbf{n}(\gamma_1\mathbf{N} + \gamma_2\tilde{A}\mathbf{n})],$$

where

$$\mathbf{N} = d\mathbf{n}/dt + \frac{1}{2}[\mathbf{n} \text{ rot } \mathbf{v}], \quad A_{ik} = \frac{1}{2}(\partial v_i / \partial x_k + \partial v_k / \partial x_i).$$

Then we have  $\mathbf{n} \times \mathbf{h} = \Gamma$  or  $\Gamma_z = h_z$ ,  $\Gamma_y = h_y$ , i.e., the equations for the rotation of the director are

$$\begin{aligned} k\nabla^2\theta &= \gamma_1\partial_t\theta + \alpha_3\partial_zv_x + \alpha_2\partial_xv_z, \\ k\nabla^2\varphi &= \gamma_1\partial_t\varphi + \alpha_3\partial_yv_x + \alpha_2\partial_xv_y. \end{aligned} \quad (2)$$

Here and subsequently we use the notation

$$\partial_t X = dX/dt, \quad \partial_{x,y,z}^{m+n} v = d^{m+n}v/dx^m dx^n.$$

Then we write the dynamical equations for the nematic liquid crystal:

$$\begin{aligned} \text{div } \mathbf{v} &= 0, \quad f_i + \frac{\partial \sigma_{ki}}{\partial x_k} = 0, \\ \mathbf{f} &= \left( -\frac{\partial P}{\partial x}, -\frac{\partial P}{\partial y}, -\frac{\partial P}{\partial z} - \rho_{cr}E \right), \end{aligned} \quad (3)$$

where  $f_i$  is the volume force,  $P$  is the pressure of the liquid, and

$$\begin{aligned} \sigma_{ik}' &= \alpha_1 n_h n_i A_{mj} n_m n_j + \alpha_2 n_h N_i + \alpha_3 n_i N_h \\ &\quad + \alpha_4 A_{ki} + \alpha_5 n_h n_j A_{ji} + \alpha_6 n_i n_j A_{jk} \end{aligned}$$

is the viscous stress tensor. After eliminating the pressure  $P$  these equations have the form

$$\begin{aligned} &[ [\alpha_1 + \alpha_6 + \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2)] \partial_{xy}^3 + \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6) (\partial_{y^2}^3 + \partial_{z^2}^3) ] v_x \\ &+ [ \frac{1}{2}(\alpha_6 - \alpha_4 - \alpha_3) \partial_{xy}^3 + \frac{1}{2}(\alpha_2 - \alpha_4 - \alpha_5) \partial_{xz}^3 - \frac{1}{2}\alpha_4 \partial_{xz}^3 ] v_y \\ &+ \frac{1}{2}(\alpha_6 - \alpha_3) \partial_{yz}^3 v_z + (\alpha_3 \partial_{y^2}^3 - \alpha_2 \partial_{xz}^3) \varphi + \alpha_3 \partial_{yz}^3 \theta = 0, \\ &[ \frac{1}{2}(\alpha_2 - \alpha_4 - \alpha_5) - \alpha_1 - \alpha_6 ] \partial_{xz}^3 - \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6) (\partial_{y^2}^3 + \partial_{z^2}^3) ] v_x \\ &+ \frac{1}{2}(\alpha_3 - \alpha_6) \partial_{xy}^3 v_y \\ &+ [ \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2) \partial_{x^2}^3 + \frac{1}{2}\alpha_4 \partial_{yz}^3 + \frac{1}{2}(\alpha_4 + \alpha_2 - \alpha_6) \partial_{xz}^3 ] v_z \\ &+ (\alpha_2 \partial_{xz}^3 - \alpha_3 \partial_{yz}^3) \theta - \alpha_3 \partial_{yz}^3 \varphi - (\epsilon_\perp/4\pi) E \partial_x \nabla^2 \Psi = 0, \quad (3') \\ &\partial_x v_x + \partial_y v_y + \partial_z v_z = 0. \end{aligned}$$

Finally we write down the Maxwell equation  $\partial \rho_{st} / \partial t + \text{div } \mathbf{j} = 0$ , where  $\mathbf{j} = \sigma \mathbf{E}$  is the conduction current. The equation has the form

$$(\epsilon_\perp/4\pi) \partial_t \nabla^2 \Psi + \sigma_a (\partial_x^2 \Psi - E \partial_x \theta) = 0. \quad (4)$$

Expressions (1), (2), and (3) comprise a system of six linearized equations for the quantities  $\theta$ ,  $\varphi$ ,  $v_x$ ,  $v_y$ ,  $v_z$ , and  $\Psi$  as a function of  $x$ ,  $y$ ,  $z$ , and the time  $t$ .

We shall seek a solution in the form of a wave with wave vector  $\mathbf{k} = (k_x, k_y, q = \pi/d)$ :

$$\{\theta, \varphi, v_x, v_y, v_z, \Psi\} \sim \exp[i(k_x x + k_y y + qz - \Omega t)]. \quad (5)$$

Then, from (1) – (3), after eliminating  $v_z$  we have five algebraic equations.

The dispersion equation that results from this system of equations for  $k_x \approx k_y \approx k$  (a domain net) has the form

$$\det |a_{ik}| = 0, \quad (6)$$

where

$$\begin{aligned} -a_{11} &= \eta_1 k q^2 + (\eta_1 + \beta_1) k^3, \quad a_{12} = \eta_3 k q^2 + (\eta_2 + \eta_3) k^3, \\ a_{13} &= \alpha_3 \Omega k q, \quad a_{14} = (\alpha_3 - \alpha_2) \Omega k^2, \quad a_{15} = 0; \\ a_{21} &= \eta_1 q^3 + (\eta_1 + \beta_2) k^2 q + (\eta_2 + \eta_3) k^4/q, \\ a_{22} &= \eta_3 k^2 q + (\eta_2 + \eta_3) k^4/q, \\ a_{23} &= (\alpha_2 k^2 - \alpha_3 q^2) \Omega, \quad a_{24} = -\alpha_3 \Omega k q, \quad a_{25} = \epsilon_\perp k \kappa^2 E / 4\pi; \\ a_{31} &= a_{32} = 0, \quad a_{33} = ik \sigma_a E, \quad a_{34} = 0, \\ a_{35} &= \sigma_\perp q^2 + (\sigma_\parallel + \sigma_\perp) k^2 - i \epsilon_\perp \Omega \kappa^2 / 4\pi; \\ a_{41} &= i(\alpha_2 k^2 - \alpha_3 q^2) / q, \quad a_{42} = i \alpha_2 k^2 / q, \\ a_{43} &= i \Omega \gamma_1 - k \kappa^2, \quad a_{44} = a_{45} = 0; \\ a_{51} &= i \alpha_3 k, \quad a_{52} = i \alpha_2 k, \quad a_{53} = 0, \quad a_{54} = k \kappa^2 - i \Omega \gamma_1, \quad a_{55} = 0; \\ \gamma_1 &= \alpha_3 - \alpha_2, \quad \beta_1 = \alpha_1 + \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_5), \quad \eta_1 = \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6), \\ \eta_2 &= \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2), \quad \eta_3 = \alpha_4 / 2, \\ \beta_2 &= \alpha_1 + \alpha_3 + \alpha_4 + \alpha_5, \quad \kappa^2 = q^2 + 2k^2. \end{aligned}$$

Letting  $\Omega \rightarrow 0$  we obtain the condition for the threshold  $U_{2c}$  at which this net appears and for the threshold wave vector:

$$\frac{\epsilon_\perp E^2}{4\pi q^2} = \frac{\sigma_\perp k \eta_1}{\alpha_3 \sigma_a} f(x), \quad (7)$$

where we introduce the dimensionless function

$$\begin{aligned} f(x) &= x(x+\alpha)(x+\beta)/(x+\gamma)(x-\Lambda), \\ x &= q^2/k^2, \quad \alpha = 3 + \eta_2/\eta_3 + \beta_2/\eta_1, \quad \beta = \sigma_\parallel/\sigma_\perp + 1, \\ \Lambda &= (\alpha_2/\alpha_3)/(\eta_1/\eta_3 + 1), \quad \gamma = \eta_2/\eta_3 + 1. \end{aligned} \quad (8)$$

This function has a minimum at  $x_c \approx 2\Lambda \gg 1$ . The values of the threshold voltage and wave vector are

$$\begin{aligned} U_{2c}^2 &\approx \frac{16\pi^3}{\epsilon_\perp} \frac{\sigma_\perp k \eta_1}{\alpha_3 \sigma_a} \Lambda \left( 1 + \frac{\alpha + \beta - \gamma}{2\Lambda} \right), \\ k_c &\approx q \left[ 2 \frac{\alpha_2}{\alpha_3} \left( \frac{\eta_1}{\eta_3} + 1 \right) \right]^{-1/2}, \quad q \sim \frac{\pi}{L}. \end{aligned} \quad (9)$$

It follows from (9) that the threshold value of the voltage for the formation of a domain net,  $U_{2c} \rightarrow \infty$  as  $\alpha_3 \rightarrow 0$ , a result that agrees with experiment. The characteristic dimension of the domain net is  $L \sim \pi/k_c$ , which exceeds the thickness  $d$  of the liquid crystal layer by a factor  $q/k_c = L/d$ . The condition for the formation of a domain net with a large period is

$$1) \quad \sigma_a < 0, \quad \alpha_3 < 0; \quad 2) \quad \sigma_a > 0, \quad \alpha_3 > 0, \quad (10)$$

where it is required that

$$\alpha_2/\alpha_3 \gg 1. \quad (11)$$

To determine the voltage dependence of the oscillation frequency we write down the dispersion equation (6) for non-zero oscillation frequency  $\Omega$  in the vicinity of the critical voltage  $U_{2c}$ :

$$\begin{aligned} k_0 + ik_1 \Omega + k_2 \Omega^2 - i \Omega^3 &= 0, \\ k_0 \sim k_0'(E^2 - U_{2c}^2), \quad k_1 \sim \omega_0^2 > 0, \quad k_2 \sim \omega_0 > 0, \\ k_0' \sim \frac{kq^2 \alpha_3 \sigma_a}{4\pi^2 \eta_1 \Lambda} &> 0, \quad \omega_0 = \frac{4\pi \sigma_\perp}{\epsilon_\perp} + \frac{kq^2}{\gamma_1}, \end{aligned} \quad (12)$$

For  $E = E_{2c}$  we have  $k_0 = 0$ , and equation (12), in addition

to the null solution ( $\Omega_1 = 0$ ) has two solutions that are damped in time:

$$\Omega_{2,s} = \pm \frac{\sqrt{3}}{2} \omega_0 - \frac{i\omega_0}{2}.$$

However, for  $E > E_{2c}$ , once a disturbance is started it begins to grow with time, and a new solution to (12) corresponding to this situation is

$$\tilde{\Omega}_1 = -ik_0/\omega_0.$$

If the disturbance increases sufficiently slowly with increasing external field, then the dispersion equation (12) remains valid, and consequently, at some value  $E_{3c} \gtrsim E_{2c}$  of the external field undamped oscillations of the domain structure can arise, which depend on the voltage and have a frequency

$$\Omega' \sim \frac{k_0}{\omega_0^2} \sim \frac{\alpha_3 \sigma_a}{4\pi^2 \eta_1 k \Lambda} \left( \frac{4\pi\sigma_\perp \gamma_1}{\varepsilon_\perp k q^2} + 1 \right)^{-2} (U^2 - U_{3c}^2). \quad (13)$$

According to expression (13) the oscillation frequency of the domain structure is  $\Omega' \propto U^2 - U_{3c}^2$  and for sufficiently thick nematic liquid crystal layers is inversely proportional to the square of the thickness of the layer, which agrees with experiment (see Fig. 3).

We note that all the estimates given here are for the case  $\alpha_3 < 0$  and  $\alpha_2 < 0$ , i.e., for the usual signs of the viscosity coefficients of nematic liquid crystals. To extend this theory to the case of positive values  $\alpha_3 > 0$  (see Fig. 1) it is necessary

to have supplementary information on the sign of the coefficient of viscosity  $\alpha_2$ . In this case, generally speaking, there are other possibilities for the appearance of undamped oscillations in addition to those discussed above; specifically, oscillations can arise in which there are large angles of deviation of the director from the initial orientation. Here the creation and annihilation of disclinations play a principal role.

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<sup>1</sup>Here and subsequently  $[h k l]$  and  $[h k]$  are the directions of the crystallographic axes of the domain lattice.

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