

Electrohydrodynamic instability in homeotropically oriented layers of nematic liquid crystals

A. N. Trufanov, M. I. Barnik, L. M. Blinov, and V. G. Chigrinov

Scientific Research Institute of Organic Intermediate Products and Dyes

(Submitted 15 July 1980)

Zh. Eksp. Teor. Fiz. 80, 704-715 (February 1981)

Instability is investigated experimentally in homeotropically oriented layers of nematic liquid crystals with positive dielectric anisotropy. It is shown that the domain pattern observed optically in the nematic phase is a consequence of destabilization of the director by electroconvective "isotropic" flow currents. The mechanism of threshold destabilization of a homogeneous molecular distribution in thin nematic liquid crystal layers is discussed.

PACS numbers: 61.30.Gd

1. INTRODUCTION

It is well known¹⁻⁴ that in thin homeotropically oriented layers of nematic liquid crystals (NLC) with positive dielectric anisotropy ($\epsilon_a = \epsilon_{\parallel} - \epsilon_{\perp} > 0$), under the influence of an external alternating electric field there occurs a threshold-type electrohydrodynamic (EHD) instability,¹⁾ which shows up optically in crossed polaroids in the form of characteristic domain patterns, of the "fingerprint" type at low frequencies and of the "Maltese cross" type at high frequencies. It has been shown^{3,4,7,8} that this instability is produced by electroconvective flow currents, which occur both in the anisotropic and in the isotropic phase of the NLC. For this reason the instability mechanism has been called "isotropic." In the liquid crystal phase, because of the optical anisotropy of the medium, the electroconvective currents show up visually in the form of domain patterns. In isotropic liquids, the electroconvective currents can be observed from the motion of solid particles of foreign impurities.

Starting from experimental data obtained chiefly on thin layers ($L \leq 20 \mu\text{m}$), we suggested earlier^{3,4} that the destabilization of the original homogeneous molecular distribution in oriented layers of NLC occurs directly at the instant of origination of electroconvective currents. In the present paper we have undertaken further experimental and theoretical investigations of the high-frequency mode of isotropic instability, for the purpose of studying the character of the electroconvective currents and the details of the mechanism of destabilization of the NLC director.

2. EXPERIMENTAL METHOD

The experimental investigations of EHD instability were made on cells of the "sandwich" type, with NLC layer thickness 5-105 μm . Homeotropic orientation of the NLC was attained by careful chemical and mechanical purification of the SnO_2 electrodes. The frequency characteristics of the thresholds of the instabilities that occurred in an electric field were investigated during excitation by a sinusoidal voltage in the range 20 Hz-2 kHz. In the nematic phase, the thresholds of instability were followed by means of a polarization microscope with crossed polaroids, on the basis of domain patterns and of the motion of solid particles of

foreign impurities, and also on the basis of the diffraction pattern produced by passage through the cell of radiation from a helium-neon laser ($\lambda = 632.8 \text{ nm}$). In addition, the threshold voltage for domain instability was determined by a photoelectric method, on the basis of the variation of the transmission of light by a cell containing the NLC, placed between crossed polaroids, with the value of the applied voltage. In this experiment, all the light diffracted by the domains was focused on a photosensitive element. In the isotropic phase, the threshold of instability was determined solely from observations on the motion of solid particles of foreign impurities. In each of these methods, the direction of propagation of the light (the direction of observation) and the direction of the applied external electric field were perpendicular to the plane of the cell and coincided with the orientation of the NLC director.

For study of the topology of the flow currents of the liquid in the nematic and isotropic phases of the NLC, we used also cells of special construction, which enabled us to carry out the observation of the process of initiation of EHD instability both by the method described above and also in a direction perpendicular to the electric field (into the "end" of the cell). In these experiments, the thickness of the homeotropically oriented NLC layer (the distance between electrodes) was 30 μm or more, while the dimension of the cell along the direction of observation "into the end" was $\sim 1.5 \text{ mm}$.

The investigations were carried out on *p-n*-methoxybenzylidene-*p'*-butylaniline (MBBA), with additives ($\sim 2-10 \text{ wt.}\%$) of *p'*-cyanophenyl ester of *p-n*-heptylbenzoic acid (CEHBA) for control of the value of ϵ_a . Depending on the composition, the mixtures had dielectric anisotropy in the range ~ 0 to $+2$. In addition, we used in the experiments *p*-octyl-*p'*-cyanobiphenyl (8CB) and *p*-cyanobenzylidene-*p'*-octyloxyaniline (CBOOA), which possess both nematic and smectic-A phases and a high positive dielectric anisotropy ($\epsilon_a \sim 7.5$). Also characteristic of 8CB and CBOOA is the fact that in the nematic temperature range the Leslie coefficient α_3 has a positive sign (8CB),⁹ or its sign changes from negative to positive (CBOOA)¹⁰ with rise of temperature from the isotropic liquid-nematic phase-transition point to the nematic-smectic-A phase-transition point

(t_{NA}). Separate experiments were also carried out on mixture A,¹¹ doped (to obtain $\epsilon_a > 0$), and on *p*-pentyl-*p'*-cyanobiphenyl (5CB).

The electrical conductivity and the anisotropy of the electrical conductivity of all the specimens investigated were, in controlled fashion, regulated or maintained approximately constant ($\sigma_{\parallel} \approx (3-5) \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $\sigma_{\parallel}/\sigma_{\perp} \approx 1.5-1.6$) by doping them with tetrabutylammonium bromide. For these values of the electrical conductivity, the domain instability in the nematic phase had the form of Maltese crosses over the whole range of frequencies above 20 Hz.

The accuracy of determination of the threshold voltage (U_{th}) for instability on the basis of domain patterns in the nematic phase was $\pm 5\%$; and on the basis of the motion of particles in the nematic and isotropic phases, $\pm 10\%$.

3. EXPERIMENTAL RESULTS

Figure 1 shows a microphotograph of the domain pattern observed in crossed polaroids for the high-frequency mode of EHD instability in homeotropically oriented layers of NLC with $\epsilon_a > 0$. It must first of all be noted that the occurrence of a domain pattern in the nematic phase has a threshold character. This is very evident from Fig. 2, which shows the variation of the transmission of light by a cell, placed between crossed polaroids, with the value of the electric field voltage. But visual observation under the microscope shows that the threshold of domain instability is always preceded by currents of liquid flow that can be followed on the basis of the motion of solid particles of foreign impurities. The initiation of liquid flow also has a threshold character: motion of the particles begins at a definite voltage and is independent of their diameter.

We introduce a coordinate system xyz such that the xy plane coincides with the plane of the cell and the z axis with the original direction of the director and with the direction of the external electric field. The domain pattern shown in Fig. 1 is visible in observation along the z direction. The solid particles execute a circular motion in the xy plane. It is characteristic that the circular motion is executed only by particles located in narrow layers next to the electrodes (of the order of several μm). This was established on cells whose construction permitted the carrying out of the observation

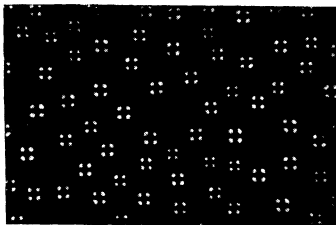


FIG. 1. Form of domain structure of EHD instability in a homeotropically oriented NLC layer with $\epsilon_a > 0$. Doped MBBA, $\epsilon_a = +0.1$, $\sigma_{\parallel} = 4 \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $L = 20 \mu\text{m}$, $f = 100 \text{ Hz}$, $t = 22^\circ\text{C}$. Dimensions of the photograph $600 \times 400 \mu\text{m}$.

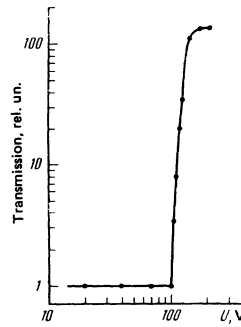


FIG. 2. Variation of light transmission with voltage for a homeotropically oriented NLC layer in crossed polaroids. Doped MBBA, $\epsilon_a = +0.1$, $\sigma_{\parallel} = 3.8 \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $L = 36 \mu\text{m}$, $f = 100 \text{ Hz}$, $t = 22^\circ\text{C}$.

in a direction perpendicular to the electric field. At fixed frequency, with increase of the voltage above the threshold value the velocity of the circular motion of the liquid near both electrodes increases; and upon attainment of a certain critical value, a domain pattern originates in the form of Maltese crosses. Thus, for example, when the NLC layer thickness is $L = 36 \mu\text{m}$, $\epsilon_a = +0.1$, and the temperature $t = 25^\circ\text{C}$, at frequency $f = 500 \text{ Hz}$ the domain pattern originates at voltage $U_{th}^D = 220 \text{ V}$. The velocity of the particles at this instant was $\sim 2-5 \text{ rev/sec}$, which, at the radius $\sim 10 \mu\text{m}$ of the circle along which the particles move (a value averaged over many particles), corresponds to linear velocity $\sim (1-3) \cdot 10^{-2} \text{ cm/sec}$. At low frequencies ($f \sim 100 \text{ Hz}$), the diameter of the circular trajectory of the particle and the transverse dimension of the Maltese cross were comparable with the thickness of the whole NLC layer. With increase of frequency, the distances between the centers of the liquid vortices and of the domains remain practically constant, but their transverse dimensions decrease. In the limits of a single domain (Maltese cross), the particles, and consequently also the liquid, move in mutually opposite directions at the two electrodes. Such correlation of the currents of liquid flow is observed even before the origination of a domain pattern.

We shall present the most important features of the threshold voltages for origination of vortical flow of the

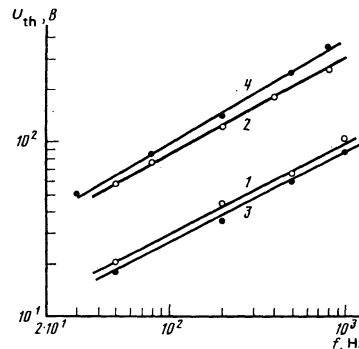


FIG. 3. Frequency dependence of threshold for occurrence of vortical liquid flow (1, 3) and of domain structure (2, 4). Doped MBBA, $\epsilon_a = +0.1$ (1, 2), $\epsilon_a = +1.75$ (3, 4); $\sigma_{\parallel} = 5 \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $L = 36 \mu\text{m}$, $t = 21^\circ\text{C}$.

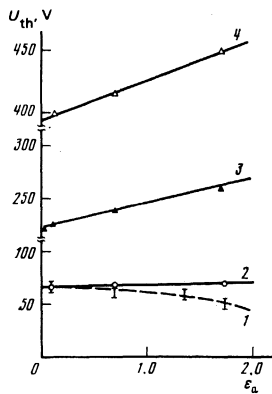


FIG. 4. Variation of threshold for vortical liquid flow (1) and for domain structure (2-4) with the value of the dielectric anisotropy. Doped MBBA, $\sigma_{\parallel} = 5 \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $f = 500 \text{ Hz}$, $t = 21^{\circ}\text{C}$; layer thickness L , in μm : 12(2), 36(1, 3), 66(4).

liquid and of a domain pattern. Figure 3 shows the frequency characteristics of the threshold electric-field voltages of both instabilities for doped MBBA with different values of ϵ_a , at fixed NLC layer thickness. It is seen that the threshold for formation of liquid vortices and of a domain structure increases with increase of frequency, approximately according to the law $U_{\text{th}} \propto f^{1/2}$. From Fig. 3 it also follows that the threshold for origination of a domain structure increases with increase of the value of the dielectric anisotropy ϵ_a , whereas U_{th}^F for formation of vortical flow decreases somewhat. The slight lowering of the threshold of electroconvective flow with increase of ϵ_a is due to the increase of the mean value of the dielectric constant $\epsilon = \epsilon_1 + \epsilon_a/3$.^{3,4} The $U_{\text{th}}(\epsilon_a)$ relations for various NLC layer thicknesses are shown separately in Fig. 4.

The instability thresholds increase linearly with increase of the NLC layer thickness (see Fig. 5). For the domain structure, a relation $U_{\text{th}}^D = \text{const} \cdot L$ is characteristic, whereas the threshold for formation of vortices increases considerably more slowly with increase of thickness, and $U_{\text{th}}^F \rightarrow \text{const}$ when $L \rightarrow 0$. In the small-thickness range, the difference between the thresholds disappears, and it is this case that was

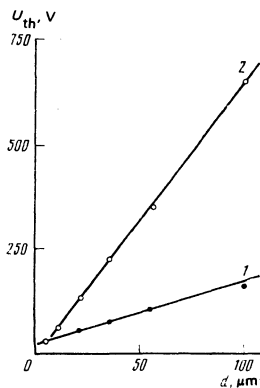


FIG. 5. Variation of threshold for vortical liquid flow (1) and for domain structure (2) with NLC layer thickness. Doped MBBA, $\epsilon_a = +0.1$, $\sigma_{\parallel} = 3.8 \cdot 10^{-11} \Omega^{-1} \text{cm}^{-1}$, $f = 500 \text{ Hz}$, $t = 21^{\circ}\text{C}$.

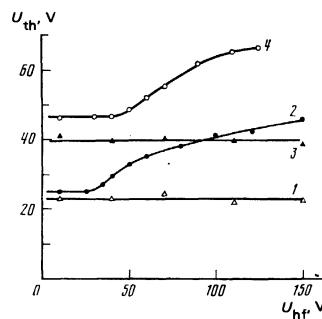


FIG. 6. Effect of a stabilizing high-frequency ($f = 20 \text{ kHz}$) electric field (U_{hf}) on the threshold for occurrence of liquid flow (1, 3) and of domain structure (2, 4). Doped MBBA, $E_a = +2.9$, $L = 20 \mu\text{m}$; 1, 2, $f = 40 \text{ Hz}$; 3, 4, $f = 150 \text{ Hz}$.

investigated experimentally in Refs. 3 and 4.

A high-frequency electric field, additionally applied to the NLC layer, leads to a rise of the threshold for formation of a domain pattern and has no effect on the threshold for origination of liquid vortices. Experimental data demonstrating this phenomenon are presented in Fig. 6. We note that the higher the frequency of the exciting voltage, and therefore also the higher the threshold for domain instability, the higher the values of the voltage of the high-frequency electric field (U_{hf}) are at which increase of the domain-structure threshold begins.

The formation of vortical liquid flows in the thin layers next to the electrodes is not a property solely of the anisotropic phase of the NLC. Figure 7 gives the temperature dependence of the threshold of instability for various thicknesses of the NLC layer, from which it is seen that both thresholds decrease with rise of temperature. At the nematic-isotropic liquid phase-transition point (t_{NI}), the domain structure disappears. But the characteristic circular motion of particles near the electrodes, and therefore also of the liquid, remains; there is practically no discontinuity in the value of U_{th}^F on transition from one phase to the other. As we have already reported,^{3,4} vortical flows are observed also in ordinary isotropic liquids. It should be noted that near the phase-transition point (see Fig. 7), the threshold voltage both for domain structure and for liquid vortices slightly increases.

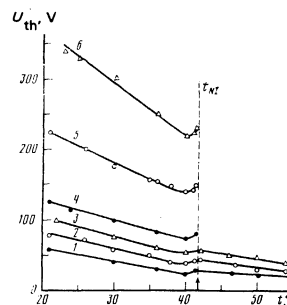


FIG. 7. Temperature dependence of threshold for formation of vortical liquid flow (1-3) and of domain structure (4-6). Doped MBBA, $\epsilon_a = +0.1$, $f = 500 \text{ Hz}$; NLC layer thickness L , μm : 22 (1, 4), 36 (2, 5), 58 (3, 6).

The thickness h of the boundary layers in which electroconvective flow occurs can be estimated experimentally from the experiments on planar NLC layers with $\varepsilon_a > 0$. In this case, EHD instability is preceded by a Fredericisz transition; that is, a reorientation of the NLC to a homeotropic structure (the S-effect⁵). Thus, for example, for doped MBBA with $\varepsilon_a = +0.05$, the threshold voltage of the S-effect is $U_s \approx 20$ V, and electroconvective flow begins at voltage ~ 40 V ($f = 40$ Hz, $L = 65$ μm). But in contrast to layers with an initial homeotropic orientation, in this case the particles in the boundary layers execute a circular motion in the yz plane (in the unperturbed state, the director is oriented along the x axis). Similar electroconvective flow currents are observed in planar layers of NLC with $\varepsilon_a < 0$, which are stable with respect to the reorientation process; these precede "pre-chevron" domains.¹² This means that the electroconvective flow of the liquid originates in thin layers near the electrodes, where the initial planar orientation is still preserved. The thickness of the unperturbed boundary layer, $\xi = L/U(4\pi K_{11}/\varepsilon_a)^{1/2}$, is the dielectric coherence distance (here K_{11} is the elastic constant for splay). With increase of the voltage, the reorientation of the director extends farther and farther from the center of the cell; and at a certain value of it, the coherence distance ξ becomes comparable with h . At this instant, the character of the liquid motion changes: the particles acquire a circular motion in the xy plane; that is, vortical flow currents, characteristic of an ordinary homeotropic NLC layer, are formed. For this specific example, the change of geometry of the liquid flow is achieved at voltage $U \approx 80$ V, to which corresponds $\xi \approx 3$ μm . Hence we find that the thickness of the boundary layer in which electroconvective processes develop is $h \approx \xi \approx 3$ μm . At voltage $U \approx 110$ V, a domain pattern originates; a photograph of it is given in Fig. 8. In this case (that is, with an initial planar orientation), the domains have the form of "beans" and not of Maltese crosses; this is a consequence of the breakdown of cylindrical symmetry in the distribution of the distorted director.

The distortion of the director is generated and is greatest in the boundary layers. With increase of the voltage, it is intensified and extended over the whole NLC layer. The process of development of destabilization of the director is easily visible, for example, by observation in a direction perpendicular to the electric field, if the thickness of the NLC layer in this direction has a value of the order of the transverse di-

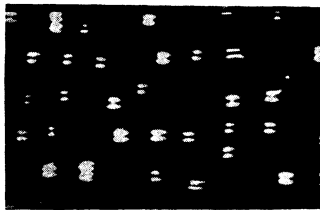


FIG. 8. Form of domain pattern of EHD instability in planarly oriented layer of NLC with $\varepsilon_a > 0$. Doped MBBA, $\varepsilon_a = +0.05$, $L = 22$ μm , $f = 100$ Hz. Dimensions of photograph 600×400 μm .

mension of a Maltese cross. Here the original orientation of the director coincides with the direction of the field. We conducted experiments on cells with distance between electrodes ~ 50 μm and with NLC layer thickness along the direction of observation ~ 45 μm . Similar experiments, both on NLC with $\varepsilon_a > 0$ and on NLC with $\varepsilon_a < 0$, were carried out in Ref. 13, the results of which corroborate our data with respect to the process of destabilization of the director.

The maximum contribution to the optical pattern (regardless of whether this is Maltese crosses or "beans"), as was shown in Ref. 12, is made by the central part of the NLC layer. This becomes understandable from the following simple considerations. Even though the distortion of the director in the center of the layer is small in comparison with the thin boundary regions, nevertheless the difference of phase, which accumulates in the result of induced birefringence, is basically determined by the volume of the specimen.

4. DISCUSSION OF RESULTS

It follows from the analysis of the experimental data that the destabilization of the NLC director, as a result of which a domain pattern originates, is caused by vortical currents of liquid flow that are generated in narrow layers next to the electrodes. The vortical currents owe their origin to electroconvective processes^{4,7,8} that are characteristic both of anisotropic and of isotropic liquids.

We shall discuss the possibility of a theoretical interpretation of the effect of destabilization of the NLC director and origination of a domain pattern. Without going into the mechanism of initiations of vortical currents of flow in the liquid, we shall regard their presence as an experimental fact, which leads to destabilization of the director. In other words, we do not assume the presence of positive feedback between the process of origination of hydrodynamic currents and the destabilization of the director, as occurs in the Carr-Helfrich mechanism. In such a case, our problem reduces in principle to the solution of the problem of hydrodynamic instability of the original orientation of the director, originating in the velocity-gradient field.¹⁴ A calculation of the threshold velocity gradient at which this instability sets in, within the framework of a two-dimensional model, with allowance for a stabilizing external electric or magnetic field, for various possible profiles of the flow velocity of the NLC, was carried out in Refs. 15 and 16. The electroconvective currents in our case have a special geometry, but they destabilize the homeotropically oriented layer of NLC in qualitative agreement with the model.¹⁴⁻¹⁶ Here the external electric field plays a double role. On the one hand, it is the reason for the EHD process; it determines the value of the velocity of the liquid in the vortex and thereby indirectly plays a destabilizing role. On the other hand, it exerts a direct stabilizing influence on the director orientation, since $\varepsilon_a > 0$.

Using the experimental results as a basis and intro-

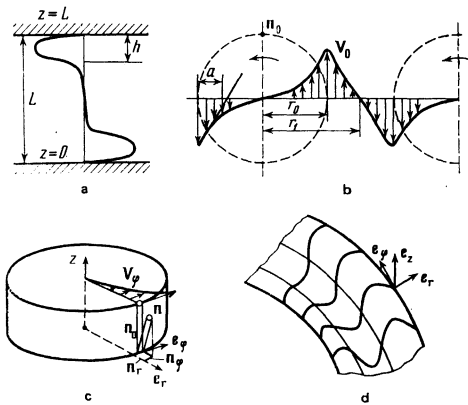


FIG. 9. Model of destabilization of the director by flow: a, profile of the circumferential velocity v_ϕ along the z axis, perpendicular to the electrodes; L is the thickness of the NLC layer, h is the thickness of the layer next to the electrodes in which the chief changes of v_ϕ occur; b, profile of the velocity v_ϕ in the radial direction; v_0 is the velocity of rotation of the cylindrical wall of radius r_0 ($v_\phi \leq v_0$), r_1 is the "total" radius of the vortex; c, deviation of the director \mathbf{n} from the homeotropic orientation \mathbf{n}_0 in a cylindrical coordinate system, $\mathbf{n} = (n_r, n_\phi, n_z)$; d, periodic variation of the orientation n_r, n_ϕ in ring domains with axis coincident with the vortex center.

ducing a cylindrical coordinate system, we shall assume that the principal one of the three components of velocity $\mathbf{v} = (v_r, v_\phi, v_z)$ is the circumferential velocity $v_\phi(r, z)$; that is, $|v_r|, |v_z| \ll v_\phi$. We shall also assume that the distribution of the velocity v_ϕ within a vortex is similar to that that would occur in the rotation of a cylindrical wall of radius r_0 and height h in a stationary liquid.¹⁷ In our case, r_0 is the effective radius of a vortex, and $h \ll r_0$ is the thickness of the boundary layer (see Fig. 9). According to Refs. 4 and 8, the role of h is played by the effective diffusion distance, at which is concentrated the volume charge that causes the electroconvective instability:

$$h \sim (\bar{D}/f)^{1/2} \ll L, \quad (1)$$

where \bar{D} is some mean diffusion coefficient, f is the frequency of the external field, and L is the thickness of the NLC layer.

The Navier-Stokes equation, under the conditions of small Reynolds numbers,¹⁷ reduces for our model, when $r \gg h$, to the form

$$\bar{\eta} \frac{\partial^2 v_\phi}{\partial z^2} + \frac{\partial^2 v_\phi}{\partial r^2} = 0, \quad (2)$$

where $\bar{\eta} = (\alpha_4 + \alpha_5 - \alpha_2)/\alpha_4 \geq 1$ is a dimensionless combination of Leslie viscosity coefficients α_i .⁸ The solution of equation (2) satisfying the boundary conditions $v_\phi|_{z=0, h} = 0$ has the form

$$v_\phi = v_0 \sin \frac{\pi z}{h} \exp \left[\pm \pi \bar{\eta}^{1/2} \frac{(r-r_0)}{h} \right], \quad (3)$$

where the signs \pm correspond to the values of the velocity v_ϕ on the inside and outside, respectively, of the cylindrical wall, and where $v_0 = v|_{r=r_0, z=h/2}$ is the maximum velocity of rotation of the wall itself.

According to (3), the velocity v_ϕ has two gradients, $\partial v_\phi / \partial z$ and $\partial v_\phi / \partial r$, each of which in principle may

cause destabilization of the original homeotropic orientation of the director, $\mathbf{n}_0 \parallel z$ (Fig. 9). The gradient $\partial v_\phi / \partial z$ according to Ref. 18 causes a thresholdless deviation of the director from the z axis by an angle θ , which can be estimated from the equation of rotation of the director.¹⁴ In the linear approximation when $\epsilon_a \sim 0$, we have

$$\theta \sim \alpha_2 v_0 h / \pi \bar{K}, \quad (4)$$

where \bar{K} is some effective coefficient of elasticity. It has been shown experimentally that the magnitude $|\theta| \ll 1$; that is, this effect of thresholdless destabilization makes no contribution to the instability, which has a clearly expressed threshold character (Fig. 2). On substituting in (4) the values $v_0 \sim 10^{-2}$ cm/sec at which a domain pattern appears, we get $h \lesssim 1 \mu\text{m} \ll L$, in full agreement with the estimate (1).

As a whole, as follows from the experiment, the profile of the velocity v_ϕ over the thickness of the layer has a quite complicated character (see Fig. 9). Most surprising is the following fact. Between the vortical flows near the upper ($z \sim L$) and lower ($z \sim 0$) electrodes, there is an unusual mutual coupling, which leads to antiparallel rotation of these vortices, despite the fact that all the variations of velocity are concentrated in very narrow boundary layers. In reality, evidently, there is a nonvanishing gradient $\partial v_\phi / \partial z$ of the circumferential velocity even at the center of the layer; but according to (4), such a gradient should be quite small:

$$(\partial v_\phi / \partial z)_{r=0, z=L-h} \sim h/L \ll 1.$$

Correlations between the velocity distributions at the two electrodes may also be aided by the presence of an axial velocity component v_z , whose value increases with increase of the circumferential velocity v_ϕ :¹⁷ $v_z \sim \rho v_\phi^2 h^2 / r_0 \bar{\alpha}$, where ρ is the density of the NLC, $\bar{\alpha}$ is some effective viscosity, and $|v_z| \ll |v_\phi|$. We note that in those NLC in which the sign of α_3 is positive,^{9,10} a director distribution of the type (4) could promote the appearance of instability because of the influence of nonlinear terms in the equation of rotation of the director, terms that describe a coupling between orientation and flow currents.¹⁹ But the slight dependence of the threshold of the domain pattern on the value and sign of the viscosity (Fig. 10), which have a decisive significance in the case of Ref. 19, provides a basis for supposing that this reason for destabilization may be neglected in the present case.

If the deviation (4) of the director is small, then by

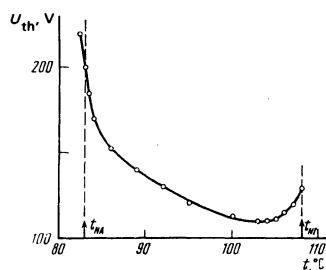


FIG. 10. Temperature variation of the threshold of domain instability for CBOOA; $L = 20 \mu\text{m}$, $f = 2 \text{ kHz}$.

neglecting it in a first approximation one can consider the destabilization of the molecular distribution because of the gradient $\partial v_\varphi / \partial r$. The corresponding equations are written in the linear approximation in the same way as in Ref. 15, with the sole difference that the velocity v_φ in the present case varies according to the nonlinear law (3) and depends on the coordinate z (Fig. 9). The boundary conditions for this problem have the form

$$\mathbf{n}|_{r=r_1} = \mathbf{v}|_{r=r_1} = 0, \quad \mathbf{n}|_{z=0, L} = \mathbf{v}|_{z=0, L} = 0, \quad (5)$$

where $r_1 \geq r_0$ is the distance from the center of the vortex ($r=0$) at which the liquid may be considered stationary. Exact solution of the two-dimensional problem,¹⁵ with use of the cylindrical symmetry of our model and of the boundary conditions (5), when the circumferential velocity v_φ of (3) emerges as the perturbing influence, gives a critical gradient value $(\partial v_\varphi / \partial r)_{\text{crit}}$ dependent on the coordinate r . One can obtain a qualitative solution of this problem by starting from the known model of instability¹⁵ and relying on the following simplifying assumptions, which are fully justified from the point of view of experiment.

a) A vortex of rotating liquid is almost plane, that is, $r_0 \gg h$. On this assumption for values $r \sim r_0$ the cylindrical coordinates (r, φ, z) may be replaced approximately by Cartesian (x, y, z) .

b) The distribution of volume charge in the xy plane, which is the main reason for the "anisotropic" Carr-Helfrich instability, may be neglected. This assumption is correct not only in the isotropic limit with respect to conductivity, $\sigma_{\parallel} \sim \sigma_{\perp}$, but also when $\sigma_{\parallel} / \sigma_{\perp} > 1$, if the frequency of the external field and the anisotropy of the dielectric constant are sufficiently high.⁸

c) The sign and value of the viscosity coefficient α_3 are not decisive (Fig. 10). This means that formulas (32)–(33) of the model of Ref. 15 may not be used to estimate the critical gradient value $(\partial v_\varphi / \partial r)_{\text{crit}}$, since the mechanism of destabilization of the director is not a homogeneous mechanism.^{14,15}

d) The perturbation of the orientation originates initially in a layer of thickness h_1 near the electrodes ($h_1/L \ll 1$) and thereafter spreads over the whole specimen. Obviously $h_1 \leq h$; that is, in a first approximation, for a qualitative estimate, we may suppose that

$$\frac{\partial}{\partial z} (n_{\varphi(r)} v_\varphi) \sim \frac{\partial n_{\varphi(r)}}{\partial z} v_\varphi,$$

neglecting the dependence of the velocity v_φ on the coordinate z .

If the assumptions (a–d) are valid, then the only possible mechanism of threshold destabilization of the director by vortices of liquid flow (3) remains the mechanism of inhomogeneous ("cyclic") instability observed in NLC in a strong stabilizing external field. In our case the domains that originate in consequence of this instability will have a ring shape, with an axis coincident with the center of the vortex, and will differ greatly in width because of the nonlinearity of the gradient $\partial v_\varphi / \partial r$ (Fig. 9). The amplitude of the deviations of the director decreases from the edge of a vortex,

where $\partial v_\varphi / \partial r$ is greatest, to its center, producing an experimentally observable optical pattern of cylindrical rings at the "rim" of the vortex.

For qualitative estimation of the value of the velocity v_0 of the cylindrical wall at which instability can occur, we shall use the model of Ref. 15, bearing in mind that the role of stabilizing magnetic field in the present case is played by the external electric field when $\varepsilon_a > 0$. As effective velocity gradient one must take the quantity

$$s = \left. \frac{\partial v_\varphi}{\partial r} \right|_{r=r_0} \approx \frac{\pi \eta^{1/2}}{h} v_0,$$

and as the characteristic distance at which this gradient acts, the quantity $a \sim h / \pi \eta^{1/2}$ (Fig. 9). To the critical gradient s_{crit} corresponds the minimum value of s for all possible values of the wave vector q_r of inhomogeneous instability:

$$s_{\text{crit}} = \min s(q_r), \quad (6)$$

where the curve $s(q_r)$ is given by the relation (5.9) of Ref. 15 for $q_x = q_r$, $q_z = \pi/2$.

By use of the viscoelastic coefficients of MBBA²⁰ and of the value of the stabilizing moment $\varepsilon_a E^2$ corresponding to our experiment, one can estimate values of $(v_0)_{\text{crit}}$ from (6) and compare them with experiment. In particular, for effective thickness of the layer $h \sim 0.05L$, $\varepsilon_a = 0.1$, and $U_{\text{th}}^D = 220$ V, we get $(v_0)_{\text{crit}}: L \sim (2-8) \cdot 10^{-4}$ cm²/sec for values of α_3 over the range -0.1 to -0.01 cP, which agrees qualitatively with the experimental value $(v_0)_{\text{crit}}: \sim 3 \cdot 10^{-2}$ cm/sec at thickness ~ 40 μm .²¹ Calculation shows also that $(v_0)_{\text{crit}}$ experiences no singularity on change of sign of α_3 (for example, the estimate indicated above at $\alpha_3 = 0$ and $\alpha_3 = 0.01$ cP gives, respectively, $(v_0)_{\text{crit}}: L \sim 9 \cdot 10^{-4}$ cm²/sec and $\sim 10^{-3}$ cm²/sec). The critical gradient increases noticeably with augmentation of the stabilizing effect of the field by increase of ε_a . Thus the experimentally observed increase of the threshold voltage of domains from 120 to 130 V on change of ε_a from $+0.1$ to $+0.75$ ($L = 40$ μm , $f = 200$ Hz) corresponds to an observed increase of the critical velocity of the liquid by a factor of 1.5 to 2. According to calculations, the same variations of the critical voltage and of the value of ε_a will lead to the same change of the critical velocity if we assume the values $h \sim (0.05-0.07)L$ corresponding to experiment.

In conclusion we note that the proposed model of destabilization of a homeotropically oriented NLC layer by electroconvective currents of liquid flow qualitatively explains all the experimentally detected features of domain instability when $\varepsilon_a > 0$. First of all, this model explains the difference between the values of the threshold voltages of the process of destabilization of the NLC as a liquid medium (the origination of vortices of liquid flow) and of the process of destabilization of the NLC director (formation of a domain pattern). The increase of the stabilizing effect of the field on the director, through the dielectric moment $\varepsilon_a E^2$, and the lack of dependence of the threshold voltage for electroconvective liquid flow on ε_a explain the different slopes of the threshold characteristics $U_{\text{th}}(\varepsilon_a)$ (Fig. 4)

for domain and for electroconvective instabilities. On the whole, the assumed mechanism for occurrence of domain instability in a homeotropically oriented NLC, when $\varepsilon_a > 0$, is in qualitative agreement with experiment. There are at present data¹² that make it possible to judge the important role of this mechanism also in a planarly oriented NLC, when $\varepsilon_a < 0$.

The authors express their gratitude to S. A. Pikin for useful discussions, which stimulated the search for a theoretical explanation of the instability, and to N. I. Mashirina for technical assistance in the measurements and in the design of this research.

¹Within the framework of the two-dimensional theory, based on the usual Carr-Helfrich model, this case is considered electrostatically stable.⁵ Such EHD instability directly from the homeotropic orientation, without a preceding Fredericzs transition, occurs only when $\varepsilon_a \approx 0$.⁶ Characteristic of this instability is a small distortion period and a high threshold voltage.

²Similar values of the critical velocity and of the corresponding critical gradient were calculated in Ref. 15 for a strong stabilizing magnetic field.

¹R. A. Kashnow and H. S. Cole, *Mol. Cryst. Liq. Cryst.* **23**, 329 (1973).

²E. A. Kursanov, *Uch. Zap. Ivanovskogo GU* **128**, 59 (1974).

³M. I. Barnik, L. M. Blinov, M. F. Grebenkin, and A. N. Trufanov, *Mol. Cryst. Liq. Cryst.* **37**, 47 (1976).

⁴M. I. Barnik, L. M. Blinov, S. A. Pikin, and A. N. Trufanov, *Zh. Eksp. Teor. Fiz.* **72**, 756 (1977) [*Sov. Phys. JETP* **45**, 396 (1977)].

⁵L. M. Blinov, *Elektro- i magnitooptika zhidkikh kristallov* (Electro- and magneto-optics of Liquid Crystals), M., Nauka, 1978.

⁶M. I. Barnik, L. M. Blinov, M. F. Grebenkin, S. A. Pikin, and V. G. Chigrinov, *Zh. Eksp. Teor. Fiz.* **69**, 1080 (1975) [*Sov. Phys. JETP* **42**, 550 (1975)].

⁷S. A. Pikin, V. G. Chigrinov, and V. L. Indenbom, *Mol. Cryst. Liq. Cryst.* **37**, 313 (1976).

⁸V. G. Chigrinov and S. A. Pikin, *Kristallografiya* **23**, 333 (1978) [*Sov. Phys. Crystallogr.* **23**, 184 (1978)].

⁹K. Skarp, T. Carlsson, T. Lagerwall, and E. Stebler, III *Liq. Cryst. Conf. of Soc. Countries, Budapest, 1979*, Abstracts, D-19.

¹⁰M. Cohen, D. Pieranski, E. Guyon, and C. D. Mitsecu, *Mol. Cryst. Liq. Cryst.* **38**, 97 (1977); A. B. White, P. E. Cladis, and S. Torza, *Mol. Cryst. Liq. Cryst.* **43**, 13 (1977).

¹¹M. I. Barnik, S. V. Belyaev, M. F. Grebenkin, V. G. Rummyantsev, V. A. Seliverstov, V. A. Tsvetkov, and N. M. Shtykov, *Kristallografiya* **23**, 805 (1978) [*Sov. Phys. Crystallogr.* **23**, 451 (1978)].

¹²L. M. Blinov, A. N. Trufanov, B. A. Umanski, and M. I. Barnik, VIII Int. Liq. Cryst. Conf., Kyoto, 1980, Abstracts.

¹³B. R. Jennings and H. Watanabe, *J. Appl. Phys.* **47**, 4709 (1976).

¹⁴P. Pieranski and E. Guyon, *Phys. Rev.* **A9**, 404 (1974).

¹⁵P. Manneville and E. Dubois-Violette, *J. Phys. (Paris)* **37**, 285 (1976).

¹⁶P. Manneville and E. Dubois-Violette, *J. Phys. (Paris)* **37**, 1115 (1976); P. Manneville, *J. Phys. (Paris)* **40**, 713 (1979).

¹⁷H. Schlichting, *Boundary Layer Theory*, McGraw, 1968 (Russ. transl. M. Nauka, 1974).

¹⁸J. Wahl and F. Fischer, *Mol. Cryst. Liq. Cryst.* **22**, 359 (1973); E. Skarp and T. Carlsson, *Mol. Cryst. Liq. Cryst. (Lett.)* **49**, 75 (1978).

¹⁹S. A. Pikin, *Zh. Eksp. Teor. Fiz.* **65**, 2495 (1973) [*Sov. Phys. JETP* **38**, 1246 (1974)]; S. A. Pikin and V. G. Chigrinov, *Zh. Eksp. Teor. Fiz.* **67**, 2280 (1974) [*Sov. Phys. JETP* **40**, 1131 (1974)].

²⁰W. H. de Jeu, *Phys. Lett.* **69A**, 122 (1978).

Translated by W. F. Brown, Jr.