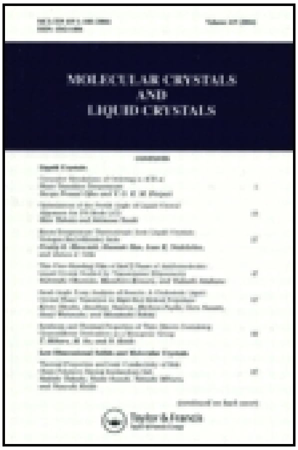

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Freedericksz Transition in Electric Fields Near the Dielectric Sign Reversal Frequency†

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Domain instability in Nematic Liquid Crystals (NLC) in ac electric fields near the dielectric anisotropy sign reversal frequency is investigated both theoretically and experimentally. The initial director is oriented planar, homeotropic or tilted. The electrohydrodynamic model of the instability proposed earlier is found to give an inadequate explanation of the experimental phenomena. The observed instability pattern is shown to be a static modulated structure, taking place as a result of a pure orientational NLC deformation, i.e., a Freedericksz transition.

I. INTRODUCTION

The electrically induced Freedericksz transition in Nematic Liquid Crystals (NLC) is the reorientation of the initial director, with an amplitude proportional to the dielectric torque $\epsilon_a E^2$, and a threshold (critical) voltage $U_c \sim (\tilde{K}/\epsilon_a)^{1/2}$, where $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$ is the NLC dielectric anisotropy and $\tilde{K}$ is the elastic constant[1]. In various types of electrically induced Freedericksz transitions in NLC, investigated up to the present moment, the director deformation remains uniform over the whole cell surface area, i.e., $\bar{n} = \bar{n}(z)$, z being the coordinate perpendicular to the NLC substrates[2].

In the present work, we report on a new type of electrically induced

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Freedericksz transition in NLC, which is characterized by a nonuniform (periodic) director deformation over the cell surface area. The effect is observed in ac electric fields in NLC, possessing a dielectric anisotropy sign reversal (near the reversal frequency ω_τ). The instability pattern arises in the form of a static domain structure, the domain appearance depending on the initial director orientation $\bar{n}_0$. This instability has been observed earlier by de Jeu et al.[3,4] in experiments with planar NLC cells, but the interpretation of the phenomenon using the electrohydrodynamic model of Goossens[5] does not seem to be adequate.

The basic equations used in Ref. 5 are a one-dimensional approach taking into account both orientational and electrohydrodynamic terms. As a result, a formal evaluation of the instability threshold has been obtained without analysing its physical essence (the authors[3-5] consider the instability to be electrohydrodynamic). The analysis of a more rigorous two-dimensional model, carried out in this work, allowing for all the orientational and electrohydrodynamic terms, shows, that the latter do not give an apparent contribution. Thus, the observed instability results in a static modulated structure in the process of a pure orientational NLC deformation, i.e., a Freedericksz effect.

Our work considers all the possible cases of the NLC static modulated structure arising in the electrically induced Freedericksz transition near the dielectric anisotropy sign reversal frequency, for different initial NLC orientations—planar, homeotropic or tilted.

II. THEORY

A. Planar orientation

Consider a layer of the NLC homogeneously oriented along the x axis, possessing a dielectric anisotropy sign reversal. The ac electric field $E = E_0 cos\omega t = E_0 \mathrm{Re}(e^{i\omega t})$ is applied along the axis perpendicular to the cell substrates.

The NLC dielectric properties in the vicinity of the reversal frequency are described by the Debye model[1]:

$$\epsilon_{\|} = \epsilon' - i\epsilon'',$$

$$\epsilon' = \epsilon_\infty + \frac{\epsilon_0 - \epsilon_\infty}{1+\xi^2}, \tag{1}$$

$$\epsilon'' = \frac{(\epsilon_0 - \epsilon_\infty)\xi}{1+\xi^2}, \qquad \xi = \frac{\omega}{\omega_D},$$

where ω^D is the Debye dipole relaxation frequency, $\epsilon_0 > \epsilon_\perp$ is the low frequency and $\epsilon_\infty < \epsilon_\perp$ the high frequency value of $\epsilon_\parallel$ respectively. The dielectric anisotropy ϵ_a sign reversal frequency ω_τ is defined by the relation $\epsilon' = \epsilon_\perp$, i.e., connects with the Debye frequency ω_D as follows:

$$\omega_\tau = \omega_D \left(\frac{\epsilon_0 - \epsilon_\perp}{\epsilon_\perp - \epsilon_\infty} \right)^{1/2} \tag{2}$$

Let the NLC director be denoted as $\bar{n} = (n_x, 0, n_z)$, macroscopic velocity $\bar{V} = (V_x, 0, V_z)$ space charge density Q and the layer thickness L. The NLC behaviour in an electric field near the dielectric anisotropy sign reversal, in the vicinity of the threshold, can be described by linearized dynamical equations (similar to those described in ref. 6), written for the values of $\psi = \partial n_z/\partial x$, V_z and Q and allowing for the complex value of the dielectric constant $\epsilon_\parallel$†:

$$\begin{aligned} &\frac{dV_z}{dS} + \frac{V_z}{\tau_V} + \kappa \frac{d\psi}{dS} - \delta E(t) Q = 0 \\ &\frac{d\psi}{dS} + \Gamma\psi + \Omega V_z + Q\left(\frac{E(t)}{\eta} + \tau \right) = 0 \\ &\frac{dQ}{dS} + \frac{Q}{\tau_e} + \sum E(t)\psi = 0 \end{aligned} \tag{3}$$

where $q = K_z/K_x$ is the wave vector ratio of the corresponding NLC deformations along the z and x axes (the boundary conditions are taken into account by letting $K_z = \pi/L$[7]), and

$$\tau_V = \frac{\rho\omega(1+q^2)}{\zeta K_x^2}, \qquad \delta = \frac{1}{\rho\omega(1+q^2)}, \qquad \kappa = \frac{\alpha_3 q^2 - \alpha_2}{\rho(1+q^2)},$$

$$\Gamma = \frac{K_{11}K_z^2 + K_{33}K_x^2}{\gamma_1\omega} - \frac{\epsilon_a\epsilon_\perp}{4\pi\nu}(1+q^2)\frac{E^2(t)}{\gamma_1\omega} - \frac{i\epsilon_a K_z f E(t)}{\nu\gamma_1\omega},$$

$$\Omega = \frac{\alpha_3 K_z^2 - \alpha_2 K_x^2}{\gamma_1\omega}, \qquad \eta = -\gamma_1 \nu \epsilon_a^{-1}\omega, \qquad \tau = i\frac{4\pi f K_z}{\nu\gamma_1\omega},$$

$$\tau_e = \frac{\omega\nu}{4\pi\mu}, \qquad \Sigma = \frac{(1+q^2)(\sigma_\parallel\epsilon_\perp - \sigma_\perp\epsilon_\parallel)}{\nu\omega}, \tag{4}$$

$$\epsilon_\parallel = \epsilon' - i\epsilon'', \qquad \epsilon_a = \epsilon_\parallel - \epsilon_\perp, \qquad \nu = \epsilon_\parallel + \epsilon_\perp q^2, \qquad \mu = \sigma_\parallel + \sigma_\perp q^2,$$

$$\gamma_1 = \alpha_3 - \alpha_2,$$

$$\zeta = \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2) + (\alpha_1 + \alpha_3 + \alpha_4 + \alpha_5)q^2 + \frac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6)q^4$$

†The domain direction is perpendicular to the initial planar director orientation.

Here $\sigma_{\|}, \sigma_{\perp}$ are the low-frequency conductivities K_{ii}, α_j NLC elastic and viscosity coefficients respectively, ρ is the density of the NLC, and $f = e_{11} + e_{33}$ the sum of the flexoelectric coefficients. We can simplify eqs. (3) and (4), using the following relations, which are valid for $\xi = \omega/\omega_D \sim 1$:

$$\epsilon' E_0, \qquad \epsilon'' E_0 \gg 4\pi f K_z, \qquad \frac{d\psi}{dS} \sim 0, \qquad \frac{dV_z}{dS} \sim 0 \tag{5}$$

According to eq. (5) it follows from eq. (3):

$$\Gamma\psi + QE(t)\left(\frac{1}{\eta} + \Omega\delta\tau_V\right) = 0, \qquad \frac{dQ}{dS} + \frac{Q}{\tau_e} + \Sigma E(t)\psi = 0 \tag{6}$$

Substituting $Q = \sum_{n=1}^{\infty}(Q_n^c \cos nS + Q_n^S \sin nS)$ and $\psi = \psi_0$ into the second equation (6) we retain only the first terms of the Q series. Then, taking into account the complex value of the dielectric constant $\epsilon_{\|} = \epsilon' - i\epsilon''$, which characterized the NLC response to the complex field amplitude $E = E_0 e^{i\omega t}$, we get:

$$-Q_1^c + \frac{Q_1^S}{\tau_e} + i\Sigma E_0\psi_0 = 0, \qquad Q_1^S + \frac{Q_1^c}{\tau_e} + \Sigma E_0 \psi_0 = 0 \tag{7}$$

and

$$Q_1^c = -\frac{\Sigma E_0 \psi_0}{\dfrac{1}{\tau_e} + i}, \qquad Q_1^S = iQ_i^c \tag{8}$$

Substituting eq. (8) into the first of the equations (6), and taking the real part of the latter, we have the following condition for the existence of a nontrivial amplitude ψ_0:

$$\bar{E}^2 = \frac{E_0^2}{2}\Gamma_0\left\{\left[\Omega\tau_V\delta + \mathrm{Re}\left(\frac{1}{\eta}\right)\right] \cdot \mathrm{Re}\left[\frac{\Sigma}{\dfrac{1}{\tau_e} + i}\right]\right.$$

$$\left. + \mathrm{Im}\left(\frac{1}{\eta}\right) \cdot \mathrm{Im}\left[\frac{\Sigma}{\dfrac{1}{\tau_e} + i}\right] + \mathrm{Re}(\Gamma_1)\right\}^{-1} \tag{9}$$

where $\Gamma_0 = (K_{11}K_z^2 + K_{33}K_x^2)/\gamma_1\omega$, $\Gamma_1 = [\epsilon_a\epsilon_\perp(1 + q^2)]/4\pi\nu\gamma_1\omega$ and $\bar{E}$ is the effective field value. From eq. (9) the voltage is $U = \pi\bar{E}/K_z = L\bar{E}$, and we obtain the function $U(q)$ the minimum of which gives the threshold voltage U_c and wave vector q_c of the instability:

$$U_c = \min_{q>0} U(q) = U_c(q_c) \tag{10}$$

The calculated $U(q)$ curves are given in fig. 1.

Let us note, that according to eq. (4) $|1/\eta| \sim \Omega\tau_\nu\delta \sim |\Gamma_1|$ and $|\Sigma/(1/\tau_e + i)| \sim |\Sigma| \sim \bar{\sigma}/\omega_\tau$, where $\bar{\sigma}$ is the average low-frequency conductivity. Then we can omit the first two components in the equation (9), and accurate up to the higher order terms in $\bar{\sigma}/\omega_\tau$ we obtain:

$$\bar{E}^2 = \frac{\Gamma_0}{\mathrm{Re}(\Gamma_1)} \tag{11}$$

It follows from eq. (11) that the instability threshold voltage does not depend on the layer thickness and is defined by dielectric constants ϵ_0, $\epsilon_\perp$, ϵ_∞ and elastic constants K_{11}, K_{33}. At the reversal frequency $\omega = \omega_\tau$, $\epsilon' = \epsilon_\perp$ and the instability threshold voltage U_c and wave vector q_c are the following eqs. (10) and (11):

$$\begin{aligned} q_c &= \frac{T_c}{L} \sim \left(1 + \frac{\epsilon''^2}{\epsilon_\perp^2}\right)^{1/4}, \\ U_c &= E_c L = \frac{\pi}{\epsilon''(\omega_\tau)} \cdot \left\{8\pi\tilde{K}\epsilon_\perp\left(1 + \sqrt{1 + \epsilon''^2/\epsilon_\perp^2}\right)\right\}^{1/2}, \end{aligned} \tag{12}$$

where $\tilde{K}$ is the average NLC elastic constant, $\epsilon''(\omega_\tau) = \sqrt{(\epsilon_0 - \epsilon_\perp)(\epsilon_\perp - \epsilon_\infty)}$, i.e., the instability disappears for $\epsilon_0 \to \epsilon_\perp$ or $\epsilon_\perp \to \epsilon_\infty$, and exists only for the positive low-frequency dielectric anisotropy $\epsilon_0 = \epsilon_\parallel(\omega \to 0) > \epsilon_\perp$. The equation (11) gives also the frequency range near the reversal frequency $\omega_\tau(1 - \delta\omega_1) < \omega < \omega_\tau(1 + \delta\omega_2)$, where this instability can be observed. In order to calculate this range consider:

$$\begin{aligned} \mathrm{Re}(\Gamma_1) &= \frac{\epsilon_\perp(1 + q^2)}{4\pi\gamma_1\omega} \cdot \mathrm{Re}\left(\frac{\epsilon_a}{\nu}\right) \\ \mathrm{Re}\left(\frac{\epsilon_a}{\nu}\right) &= \frac{(\epsilon' - \epsilon_\perp)(\epsilon' + \epsilon_\perp q^2) + \epsilon''^2}{(\epsilon' + \epsilon_\perp q^2)^2 + \epsilon''^2} \end{aligned} \tag{13}$$

Far from the reversal frequency ($\omega \rightarrow 0$) the dielectric losses do not exist ($\epsilon'' \rightarrow 0$) and an ordinary type Freedericksz transition (S-effect) takes place with the threshold $U_S \sim (\epsilon' - \epsilon_\perp)^{1/2}$. In the vicinity of the reversal frequency ($\omega \rightarrow \omega_\tau$), when the components in the numerator of eq. (13) become comparable in value:

$$\epsilon' - \epsilon_\perp \sim \frac{\epsilon''^2}{2\epsilon_\perp}, \qquad q \sim 1, \qquad \epsilon' \sim \epsilon_\perp \tag{14}$$

the periodic director deformation along x-axis (domain structure) is energetically favourable. Fig. 1 shows that the lower frequency limit of the domain existence $\omega_S \sim \omega_\tau(1 - \delta\omega_1)$, defined by eq. (14) is very close to the reversal frequency ω_τ: $\delta\omega_1 \sim 0$.

To define the upper limit of the domains' existence ω_∞, we equate

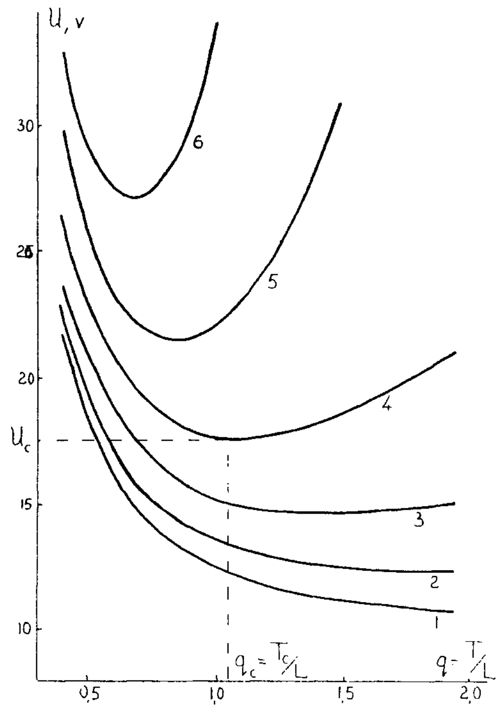

FIGURE 1 The instability threshold voltage vs. wave vector ratio of the deformations along z and x axes, $q = K_z/K_x$, $q \rightarrow \infty$, $K_x \rightarrow 0$ corresponds to the S-effect (the initial director is planar). The curves are calculated by means of eq. (9) for the following NLC parameters: $\epsilon_\parallel = 5.6$, $\epsilon_\perp = 5.25$, $\sigma_\parallel = 205$, $\sigma_\parallel/\sigma_\perp = 2.17$, $k_{11} = 0.85 \cdot 10^{-6}$, $K_{33} = 1.06 \cdot 10^{-6}$, $\alpha_1 = 0.07$, $\alpha_2 = -1.17$, $\alpha_3 = -0.017$, $\alpha_4 = 0.8$, $\alpha_5 = 0.43$, $n_\parallel = 1.784$ CGSE units. The reversal frequency $f_\tau = \omega_\tau/2\pi = 16$ KHz, NLC layer thickness $L = 22$ μm. $\omega/\omega_\tau = 0.85$ (curve 1), 0.9 (2), 0.95 (3), 1,0 (4), 1.05 (5), 1.1 (6).

the denominator of eq. (13) to zero for $q = 0$:

$$\epsilon' - \epsilon_\perp = -\frac{\epsilon''^2}{\epsilon'} \tag{15}$$

and by means of eqs. (2) and (3) obtain:

$$\omega_\infty = \omega_\tau(1 + \delta\omega_2) \sim \left(\frac{\epsilon_0}{\epsilon_\infty}\right)^{1/2} \omega_\tau \tag{16}$$

Using eq. (9) we can calculate a correction to (16):

$$\omega_\infty \sim \left(\frac{\epsilon_0}{\epsilon_\infty}\right)^{1/2} \omega_\tau + 2\pi\sigma_\parallel \cdot \frac{\epsilon''}{\epsilon'} \frac{(\epsilon_0 - \epsilon_\infty)}{(\epsilon_\perp - \epsilon_\infty)(\epsilon_0 - \epsilon_\perp)} \cdot \left[-\frac{\alpha_2}{\zeta} + \frac{\sigma_\perp}{\sigma_\parallel} - 1 \right] \tag{17}$$

It follows from eq. (17) that the correction has the order of $\bar{\sigma}/\omega_\tau$, and due to this, the upper frequency limit of the instability existence ω_∞ increases linearly with rising the low frequency conductivity $\sigma_\parallel$. The frequency dependence of the threshold of the instability, arising from the planar NLC orientation, calculated by eqs. (9) and (10) is shown in fig. 2a (curve 1). The threshold voltage of the domains U_c increases, while the period T_c goes down (curve 1, fig. 2b) for the higher electric field frequencies, so that at the reversal frequency $\omega = \omega_\tau$ the domain period is approximately equal to the NLC layer thickness L: $T_c \sim L$. The qualitative evaluation[5] of the instability threshold (curve 2, fig.

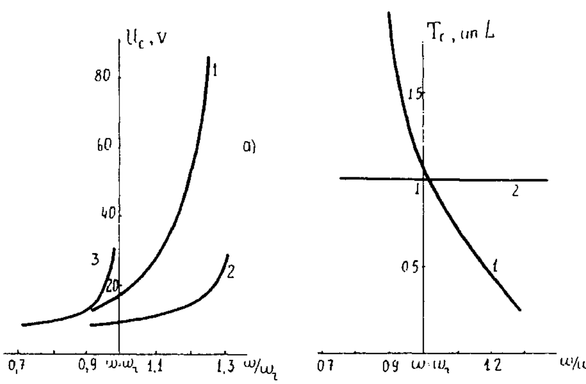

FIGURE 2 The calculated instability threshold characteristics $U_c(a)$ and $T_c(b)$ vs electric field frequency for the planar director of the NLC. NLC parameters are taken as in fig. 1. The curve (1): two-dimensional model eq. (9), (2): qualitative estimation[5], (3): S-effect threshold.

2a), obtained from eq. (9) for $K_z = 0$, $T_c = L$ gives an underestimated threshold value. Fig. 2a shows that near the reversal frequency $\omega \lesssim \omega_\tau$ the uniform structure of the ordinary type Freedericksz effect (curve 3, fig. 2a) becomes energetically unfavourable.

Let us note that all the basic results eqs. (12)–(17) are obtained by using the relation eq. (11), i.e. without taking into account the NLC velocity $\overline{V}$ and charge density Q.

Indeed, according to (8) we have:

$$
\begin{aligned}
&|Q| \sim \frac{\bar{\sigma}}{\omega_\tau} E_0\psi_0 \ll E_0\psi_0, \\
&|V_z| \sim |\tau_V \delta E_0 Q| \sim \frac{\bar{\sigma}}{\omega_\tau} \frac{E_0^2\psi_0}{\zeta K_x^2} \ll \frac{E_0^2\psi_0}{\zeta K_x^2}
\end{aligned}
\tag{18}
$$

i.e., accurate to the higher order terms in $\bar{\sigma}/\omega_\tau$ one can consider the space charge Q and NLC velocity $\overline{V}$ to be equal to zero (see also the reduction of eq. (11) to eq. (9)). The only degree of freedom, whose disturbance results in the instability is the director distribution $\psi = \partial n_z/\partial x$. In view of this we can give another simple method of deriving the relation eq. (11). We introduce a nonuniform potential $\Phi = -Ez + R(x,z)$, $|R| \ll E_c L$ and consider the linearized equation of the director rotation, allowing for this nonuniform potential:

$$
(K_{11}K_z^2 + K_{33}K_x^2)\theta - \frac{\epsilon_a \overline{E}^2}{4\pi}\theta + \frac{iK_x\epsilon_a \overline{E}R}{4\pi} = 0, \tag{19}
$$

where $R = -iK_x\epsilon_a\overline{E}\theta/(\epsilon_\parallel K_x^2 + \epsilon_\perp K_z^2)$, $\overline{\mathrm{E}}$ is the effective field. The condition for a nontrivial value of the director amplitude θ, together with the relation $\epsilon_\parallel = \epsilon' - i\epsilon''$, immediately gives us the necessary result eq. (11).

The relations eq. (11) and (12) show that the threshold instability characteristics do not depend on the NLC conductivity and viscosity and are defined by the corresponding NLC dielectric and elastic constants. The domain structure results from the nonuniform static director deformation, since the NLC velocity is negligible. Starting from this, one can state that the electrically induced instability of the planar NLC sample near the dielectric anisotropy sign reversal frequency can be considered as a new type of a Freedericksz transition. The dielectric losses $\epsilon''(\omega)$ near the reversal frequency favour the NLC director distribution modulated along the x-axis and the corresponding nonuniform electric field, thus giving an energetic gain in comparison with an ordinary type of uniform Freedericksz deformation. In particular, the dielectric torque, caused by a nonuniform field, is not equal to zero at the dielectric anisotropy sign reversal point, where the threshold of an ordinary type Freedericksz transition goes to infinity.

B. Tilted and homoetropic orientation

If the NLC director is tilted or homeotropic $n = (cos\theta_0, 0, \sin\theta_0)$, $0 < \theta_0 \leqslant \pi/2$, then the modulated structure in the Freedericksz transition can arise due to azimuthal director deviations from the plane xz of the initial orientation, as a pattern of linear domains, parallel to the y axis. The geometry of these domains is described in ref. (7). Accordingly we can rewrite the relation eq. (11) in the form:

$$\overline{E}^2 = \frac{\tilde{\Gamma}_0}{\mathrm{Re}(\tilde{\Gamma}_1)}, \tag{20}$$

where $\tilde{\Gamma}_0 = (K_{22}\cos^2\theta_0 + K_{33}\sin^2\theta_0)K_z^2 + K_{11}K_y^2$,

$$\tilde{\Gamma}_1 = -\frac{\epsilon_a(\epsilon_\perp + \epsilon_a\sin^2\theta_0)(1+q^2)}{4\pi\nu}, \quad \nu = \epsilon_\perp + (\epsilon_\perp + \epsilon_a\sin^2\theta_0)q^2$$

Here all the elastic moduli K_{11}, K_{22}, K_{33} play an important role. In the case of a homeotropic director orientation $\theta_0 = \pi/2$ the x and y axes are equivalent and due to this, two perpendicular domain systems exist forming a domain net.

The values of the threshold voltage U_c and domain period T_c of the instability pattern, calculated by means of eqs. (20) and (10), are given in fig. 3 for the homeotropic director orientation. The frequency

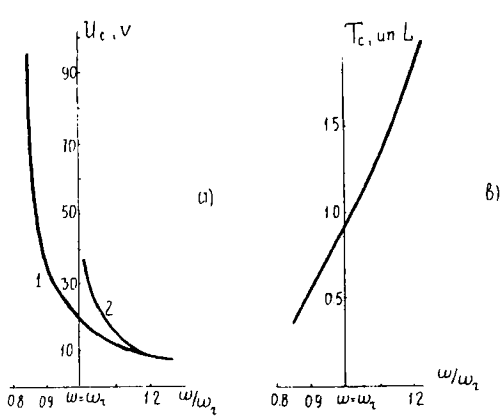

FIGURE 3 The instability threshold characteristics U_c (a) and T_c(b) for the homeotropic NLC orientation. The NLC parameters are taken as in fig. 1. The curve (1): two-dimensional theory for $\theta_0 = \pi/2$ eqs. (20) and (10), (2): B-effect threshold.

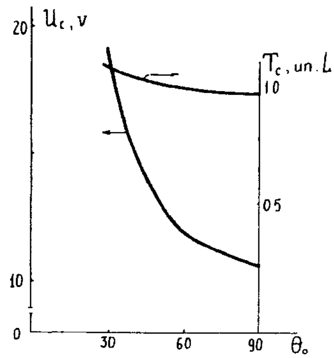

FIGURE 4 Threshold characteristics of the instability arising from the tilted NLC orientation vs. initial tilt angles θ_0 at $\omega = \omega_\tau$. The calculations are made, using eqs. (20) and (10) for $\epsilon_\parallel = 6$, $\epsilon_\perp = 5{,}25$, $K_{11} = 0.677 \cdot 10^{-6}$, $K_{33} = 0.826 \cdot 10^{-6}$, $n_\parallel = \sqrt{\epsilon_\infty} = 1.75$ CGSE units.

dependencies $U_c(\omega)$ and $T_c(\omega)$ in the homeotropic and planar cases are found to be symmetric with respect to the line $\omega = \omega_\tau$ (figs. 2, 3). The threshold of the domain structure becomes infinite at the frequency $\omega = \omega_\infty$ and is equal to the Freedericksz transition threshold (B-effect) at $\omega = \omega_B \sim \omega_\tau$ ($\omega_\infty < \omega_\tau < \omega_B$). The corresponding domain period increases from $T_c \sim 0$ at $\omega = \omega_\infty$ to $T_c \to \infty$ at $\omega = \omega_B$.

If the initial director is tilted ($0 < \theta_0 < \pi/2$) the character of the threshold curves $U_c(\omega)$ and $T_c(\omega)$ are quite similar to that considered above for the homeotropic case. Fig. 4 shows that at a given frequency the instability threshold increases and the period remains the same with decreasing the θ_0 value. The corresponding frequency ω_B of the domain is found to very close to the reversal frequency ω_τ and is defined from the relation:

$$\frac{\epsilon''^2 \sin^2\theta_0}{2\epsilon_\perp} \sim \epsilon_\perp - \epsilon'$$

To evaluate the lower frequency limit ω_∞ we can put $\tilde{\Gamma}_1 = q = 0$ as in the earlier derivation of eqs. (15) and (16). Then the frequency ω_∞ can be obtained from the equation:

$$\epsilon' - \epsilon_\perp = \frac{\epsilon''^2 \sin^2\theta_0}{\epsilon' + (\epsilon' - \epsilon_\perp)\sin^2\theta_0} \tag{21}$$

Equation (21) shows, that if $\theta_0 \to 0$, then $\omega_\infty \to \omega_\tau (\epsilon' \to \epsilon_\perp)$. The calculations of the azimuthal instability threshold by means of eqs. (20),

(10) confirm the fact that $U_c \to \infty$ for $\epsilon_0 \to \epsilon_\perp$. Thus, as in the case of planar orientation, the necessary condition for the existence of such an instability is a positive low frequency dielectric anisotropy $\epsilon_0 - \epsilon_\perp > 0$. Finally we should note, that if the director is tilted ($\theta_0 \neq 0, \pi/2$), a nonthreshold Freedericksz transition can be observed, leading either to modulated or the uniform deformation pattern.

III. EXPERIMENTAL, RESULTS AND DISCUSSION

A. Planar orientation

Experimental investigations of the domain instability were made with liquid crystalline mixtures possessing the dielectric anisotropy sign reversal in the frequency range 3–40 KHz and 0.6–1.0 MHz. The mixtures were prepared on the basis of p-butyl-p′-methoxyazoxybenzole and p-butyl-p′-heptayloxyazoxybenzole in the proportion 2 : 1 (mixture A), doping with n-cyanophenyl ester of c-chloro-p-(p-n-heptylbenzyloxy) benzoic acid (1 : 18 weight percent) or p-cyanophenyl ester of p-heptyl-benzoic acid (15 : 40 weight percent) respectively.

The instability threshold characteristics were investigated in "sandwich" type cells with the thicknesses 15 → 105 μm. The initial planar NLC orientation was prepared by rubbing a polyvinyl alcohol

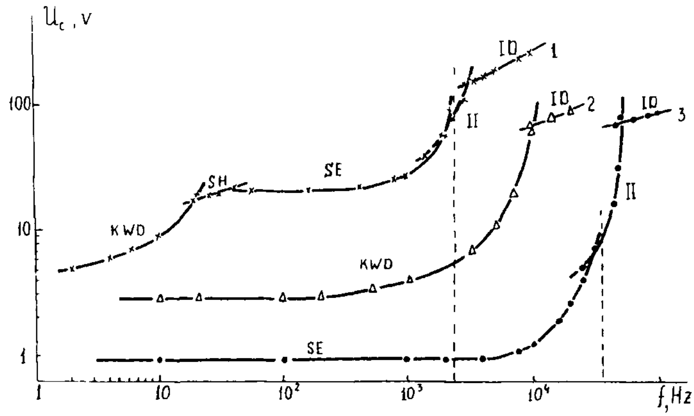

FIGURE 5 Experimental frequency dependencies of the instabilities threshold characteristics of NLC with dielectric anisotropy sign reversal: KWD—Kapustin–Williams domains, SH—chevron domains, SE—S-effect, II—investigated instability, ID—electrohydrodynamic "inertial" instability[8]. For the curves (1) and (2): $\epsilon_0 - \epsilon_\perp = +0.05$; (3): $\epsilon_0 - \epsilon_\perp = +6.2$. The NLC conductivity $\sigma_\parallel = 126\ \text{sec}^{-1}$ (curve 1), 3400 sec^{-1} (2), 835 sec^{-1} (3).

film, coated on the glass substrates after depositing the transparent SnO_2 electrodes. The threshold voltage of the domains was visually registered in a polarizing microscope, as well as by laser beam diffraction ($\lambda = 0.63\mu$) by the domain structure. The observations of the possible liquid motion were carried out in the interelectrode gap of a specially constructed cell in a direction perpendicular to the electric field (i.e., from the "face" of the cell).

Fig. 5 shows the experimental frequency characteristics of the instability threshold voltages in NLC with dielectric anisotropy sign reversal for different values of $\sigma_{\parallel}$ and ϵ_a: KWD—Kapustin–Williams domains[1], SH—chevron domains, SE—Freedericksz transition (S-effect), II—investigated instability, ID—electrohydrodynamic "inertial" instability[8]. As seen from fig. 5, the investigated instability always arises in the vicinity of the reversal frequency, its threshold characteristic practically following that of the S-effect. With increasing low frequency dielectric anisotropy $\epsilon_0 - \epsilon_{\perp}$ the instability threshold voltage goes down, as for the usual Freedericksz effect. The comparison between the curve 1 ($\epsilon_a = +0.05$) and 3 ($\epsilon_a = +6.2$) in fig. 5 confirms this fact. The instability occurs only in a relatively low conductivity NLC, when the KWD and SH domains frequency regions do not spread very extensively. For the higher NLC conductivities ($\sigma_{\parallel} \gtrsim 10^3$ see^{-1} in our experiment) the investigated instability II and Freedericksz effect SE does not exist in the vicinity of the reversal frequency, since Kapustin–Williams domains occur under a low threshold voltage and are energetically more favourable.

The photographs of the domain instability near the dielectric anisotropy sign reversal frequency are shown in fig. 6a and 6b for $\omega \lesssim \omega_\tau$ and $\omega > \omega_\tau$ respectively. The optical image of the instability and the character of the director deformation resembles that of the Kapustin–Williams domains. The instability appears as a pattern of linear domains, oriented perpendicular to the initial director and observed only with light polarized in the director plane. This testifies to the fact that the director deformation arises only in the xz plane. According to our observations of solid dust particles, introduced in the NLC, we come to the conclusion that there is no liquid motion in the cell. Thus, the instability is not of an electrohydrodynamic type, as accepted in refs. 3 and 4, and differs considerably from the Kapustin–Williams domains. In the frequency region $\omega \lesssim \omega_\tau$ the domain threshold practically coincides with the threshold of usual Freedericksz transition (S-effect). Above the threshold the domain pattern transforms into a loop structure[9] (fig. 6c), and, after that, disappears. If $\omega > \omega_\tau$, then under the higher voltages a domain net arises with a comparatively small period in two directions (fig. 6d).

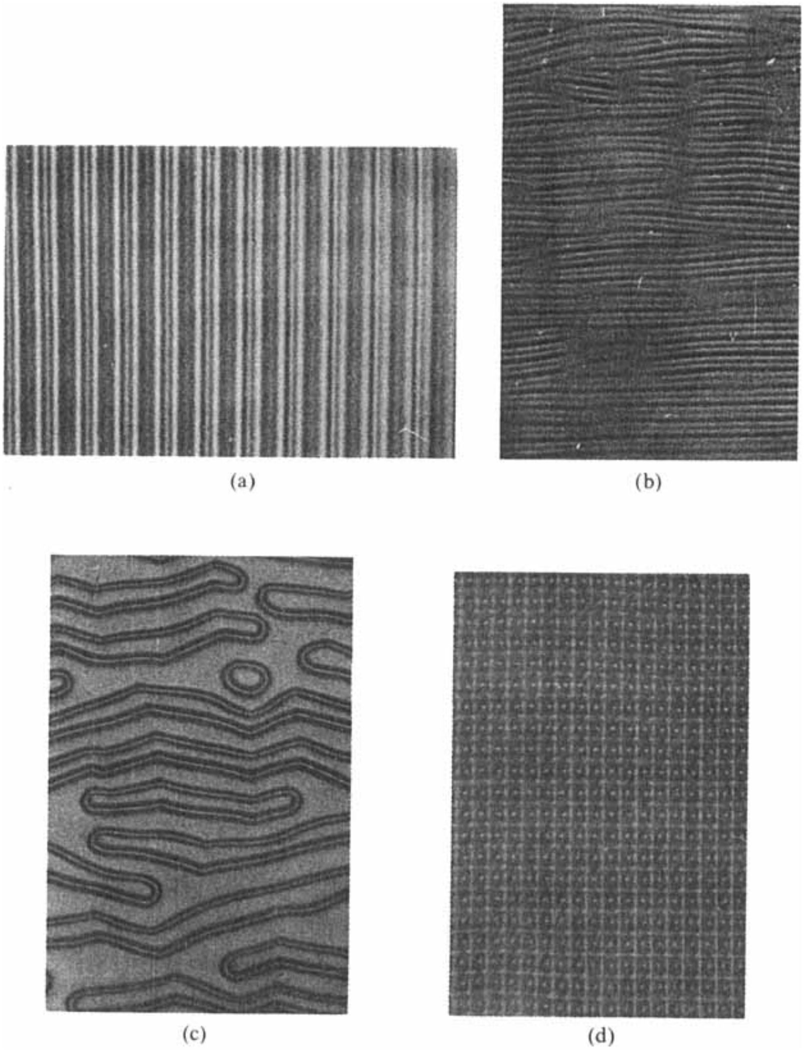

FIGURE 6 The appearance of the domain structure for different voltages and field frequencies. Photograph, (a): $\omega \lesssim \omega_\tau$, $U = U_c$; (b) $\omega > \omega_\tau$, $U = U_c$; (c) $\omega \lesssim \omega_\tau$, $U > U_c$, (d) $\omega > \omega_\tau$, $U > U_c$.

The threshold voltage of the investigated instability does not depend on NLC layer thickness within the range $5 \to 105$ μm. The variation of the absolute conductivity value $\sigma_{\parallel} \ll \omega_\tau$, and the conductivity anisotropy $\sigma_{\parallel}/\sigma_{\perp}$ $(1.03 < \sigma_{\parallel}/\sigma_{\perp} < 1.8)$ also does not exert the slightest influence on the instability threshold characteristics.

The domain period T_c dependence on electric field frequency close to the reversal frequency has the form $T_c \sim \omega^{-\alpha}$ where $\alpha \sim 1/2$, so that

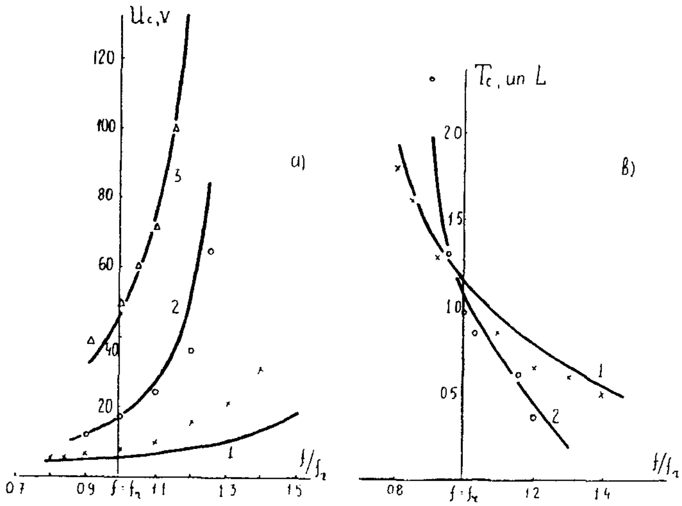

FIGURE 7 The threshold characteristics U_c (a) and T_c (b) of the instability for the planar director orientation. Solid lines are the theoretical calculated curves. For the curve (1): $\epsilon_a = +4.7$ (x-experiment); (2): $\epsilon_a = +0.35$ (o-experiment), (3): $\epsilon_a = +0.05$ (Δ-experiment). The other NLC parameters in the calculation are taken as in fig. 1.

at the reversal frequency the period is approximately equal to the layer thickness L.

The theoretical and experimental frequency threshold characteristics of the instability are given in fig. 7. According to the theoretical predictions, the experimental instability threshold diverges for $\omega \to \omega_\infty = \omega_\tau(1 + \delta\omega_2)$ and goes down with increasing low frequency dielectric anisotropy $\epsilon_0 - \epsilon_\perp$ (curves 3 − 1, fig. 7). The reversal frequency is strongly dependent on the value of $\epsilon_0 - \epsilon_\perp$ (eq. (2)), so we compare the theoretical and experimental data, using the reduced variable ω/ω_τ. Experimental values are in agreement with the theory within the whole frequency range. In view of our model, the point $\epsilon_a = \epsilon' - \epsilon_\perp = 0$ is not singular, as it takes place in usual Freedericksz effect, so the instability is observed both for positive and negative ϵ_a values. The discrepancy between the theoretical and experimental $U_c(\omega)$ curves, especially in the case of large dielectric anisotropy (Figure 7), might be explained by the variation of the elastic moduli in the process of doping the initial mixture to get the desired large ϵ_a values. (The elastic coefficients are changed in accord with the dopant concentrations, while they are taken as constant in the calculations).

B. Homeotropic orientation

Homeotropic NLC layer orientation was made by deep cleaning and careful washing of the electrodes. The quality of the orientation was

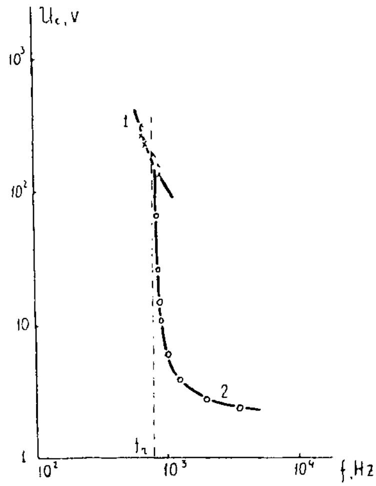

FIGURE 8 The frequency characteristics of the instability (curve 1) and B-effect (curve 2) threshold voltages for the homeotropic NLC orientation $\epsilon_0 - \epsilon_\perp = +1.2$ ($f = 200$ Hz), $\epsilon_\infty - \epsilon_\perp = -0.7$ ($f = 40$ KHz), $T = 21°$C, $L = 70$ μm.

verified visually by placing the cell between the crossed polaroids. The instability was investigated in NLC mixture, consisting of mixture A (90 weight %) and 4-cyano-4′-biphenyl ester of 4(4-heptylbenzyloxy) benzoic acid (10%). The dielectric anisotropy sign reversal frequency of the NLC mixture was 700Hz at 20°C. The dielectric anisotropy $\epsilon_0 - \epsilon_\perp = 1.2$ (f = 200Hz) and -0.7 (f = 40kHz).

The frequency characteristics of the domain threshold voltage of the homeotropically oriented NLC layer is given in fig. 8 (curve 1). The curve 2 in fig. 8 corresponds to the B-effect. According to the experiment, the domain pattern arises in the vicinity of the reversal frequency $\omega = \omega_\tau$, its threshold voltage sharply increasing for $\omega < \omega_\tau$. The domains look like twisting belts with a period approximately equal to the layer thickness at the reversal frequency ω_τ. The threshold characteristics, as in the previous case eq. (12), do not depend on the layer thickness.

CONCLUSION

We have investigated the ac electric field induced domain instability in Nematic Liquid Crystals near the dielectric anisotropy sign reversal frequency. The domain appearance depends on the initial NLC orientation: planar, homeotropic or tilted. Our experiments show that the domain patterns which arise remain static (there is no liquid motion). The threshold characteristics of the effect are defined in the main by the NLC dielectric and elastic constants, and do not depend upon the NLC conductivity and viscosity. The analysis of the two-dimensional model of the instability has shown that the electrohydrodynamic terms do not play an important role, i.e., the observed domain structure occurs due to the pure orientational NLC director deformation. We should state that the investigated instability represents a unique type of a static modulated NLC structure in a Freedericksz transition under a homogeneous external electric field.

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