New type of Fréedericksz transition in nematic liquid crystals

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An experimental and theoretical study is made of the domain instability in nematic liquid crystals exhibiting a low-frequency inversion of the sign of the dielectric anisotropy. It is shown that the instability arises as a result of the dielectric destabilization of the initial planar orientation of the director of the liquid crystal in an alternating electric field.

INTRODUCTION

The Fréedericksz transition in nematic liquid crystals in an electric field E is a change in the original orientation of the director **n**, with the amplitude of the deformation being proportional to the dielectric moment $\varepsilon_a E^2$ and the threshold voltage $U_{\rm th}$ going as $(\tilde{k}/\varepsilon_a)^{1/2}$, where $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the anisotropy of the dielectric constant, and $\tilde{k}$ is the elastic modulus of the nematic liquid crystal.[1] In the known types of Fréedericksz transitions of nematics in an electric field, the deformation of the director **n** has been homogeneous over the entire area of the cell, i.e., $\mathbf{n} = \mathbf{n}(z)$, where z is the coordinate in the direction perpendicular to the plates of the liquid-crystal cell.[2]

In the present paper we report a new type of Fréedericksz transition in nematic liquid crystals in an electric field; this transition is characterized by an inhomogeneous (periodic) deformation of the director over the entire area of the cell. The effect is observed in an electric field $E(t) = E_0\cos\omega t$ (near the inversion frequency ω_i) in nematics which exhibit an inversion of the sign of the dielectric anisotropy ε_a, in the form of a static domain structure perpendicular to the initial orientation of the nematic. Such an instability was observed earlier in the experiments of de Jeu *et al.*[3,4] but was erroneously interpreted as an electrohydrodynamic instability in the framework of the Goossens model.[5]

The problem is that the initial equations of Ref. 5 take both the orientational and electrohydrodynamic effects into account in the one-dimensional approximation. As a result, a formal estimate of the instability threshold was obtained without analysis of the physical mechanism of the instability (it was assumed[3–5] that the instability is electrohydrodynamic).

Analysis of the more rigorous two-dimensional model proposed in the present paper (this model also incorporates the orientational and electrohydrodynamic terms) shows that the electrohydrodynamic term is not important, and the observed instability is a static modulated structure arising as a result of a purely orientational deformation of the medium—the Fréedericksz effect.

2. THEORY

Let us suppose that a plane-oriented (along the x axis) layer of a nematic liquid crystal exhibiting an inversion of the sign of the dielectric anisotropy is subjected to an alternating electric field

$$E(t) = E_0 \cos\omega t = E_0 \,\mathrm{Re}\,(e^{i\omega t})$$

applied along the z axis (the z axis is perpendicular to the plates of the liquid-crystal cell).

The dielectric properties of the liquid crystal in the dispersion region of $\varepsilon_\parallel$ are described by the Debye model[1]:

$$\varepsilon_\parallel = \varepsilon' - i\varepsilon'', \quad \varepsilon' = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1+\xi^2},$$
$$\varepsilon'' = \frac{(\varepsilon_0 - \varepsilon_\infty)\xi}{1+\xi^2}, \quad \xi = \frac{\omega}{\omega_D}, \tag{1}$$

where ω_D is the Debye relaxation frequency of the dipoles and $\varepsilon_0 > \varepsilon_\perp$ and $\varepsilon_\infty < \varepsilon_\perp$ are, respectively, the low-frequency and high-frequency values of $\varepsilon_\parallel$. The frequency ω_i at which the dielectric anisotropy ε_a changes sign (this frequency is determined from the condition $\varepsilon' = \varepsilon_\perp$) is related to the Debye frequency ω_D by

$$\omega_i = \omega_D[(\varepsilon_0 - \varepsilon_\perp)/(\varepsilon_\perp - \varepsilon_\infty)]^{1/2}. \tag{2}$$

We introduce the notation $\mathbf{n} = (n_x, 0, n_z)$ for the director of the nematic, $\mathbf{V} = (V_x, 0, V_z)$ for the velocity of the liquid, Q for the space charge, and L for the thickness of the liquid-crystal layer.

The behavior of a nematic in an electric field in the dispersion region of $\varepsilon_\parallel$ can be described by a linearized system (analogous to that of Ref. 6) of dynamical equations for the quantities $\psi = \partial n_z/\partial x$, V_z, and Q with allowance for the complex value of the dielectric constant $\varepsilon_\parallel$:

$$\frac{dV_z}{ds} + \frac{V_z}{\tau_V} + \varkappa\frac{d\psi}{ds} - \delta E(t) Q = 0,$$
$$\frac{d\psi}{ds} + \Gamma\psi + \Omega V_z + Q\left(\frac{E(t)}{\eta} + r\right) = 0, \tag{3}$$
$$\frac{dQ}{ds} + \frac{Q}{\tau_e} + \Sigma E(t)\psi = 0,$$

where $q = k_z/k_x$ is the ratio of the wave vectors of the deformations along the z and x axes (to make approximate allowance for the boundary conditions we set $k_z = \pi/L$),[7] $s = \omega t$ is the reduced time,

$$\tau_V = \frac{\rho\omega(1+q^2)}{\xi k_x^2}, \quad \delta = \frac{1}{\rho\omega(1+q^2)}, \quad \varkappa = \frac{\alpha_3 q^2 - \alpha_2}{\rho(1+q^2)},$$
$$\Gamma = \frac{k_{11}k_z^2 + k_{33}k_x^2}{\gamma_1\omega} - \frac{\varepsilon_a\varepsilon_\perp}{4\pi\nu}(1+q^2)\frac{E^2(t)}{\gamma_1\omega} - \frac{i\varepsilon_a k_z f E(t)}{\nu\gamma_1\omega},$$
$$\Omega = \frac{\alpha_3 k_z^2 - \alpha_2 k_x^2}{\gamma_1\omega}, \quad \eta = -\gamma_1\nu\varepsilon_a^{-1}\omega, \quad r = i\frac{4\pi f k_z}{\nu\gamma_1\omega},$$
$$\tau_e = \omega\left(\frac{4\pi\mu}{\nu}\right)^{-1}, \quad \Sigma = \frac{(1+q^2)(\sigma_\parallel\varepsilon_\perp - \sigma_\perp\varepsilon_\parallel)}{\nu\omega},$$

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$$\varepsilon_\parallel=\varepsilon'-i\varepsilon'',\quad \varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp, \tag{4}$$

$$\nu=\varepsilon_\parallel+\varepsilon_\perp q^2,\quad \mu=\sigma_\parallel+\sigma_\perp q^2,\quad \gamma_1=\alpha_3-\alpha_2,$$

$$\zeta={}^1/_2(\alpha_4+\alpha_5-\alpha_2)+(\alpha_1+\alpha_3+\alpha_4+\alpha_5)q^2+{}^1/_2(\alpha_3+\alpha_4+\alpha_6)q^4.$$

Here $\sigma_\parallel$ and $\sigma_\perp$ are the low-frequency values of the conductivity, k_{ii} and α_i are the elastic constants and viscosity coefficients of the nematic, ρ is its density, and $f=e_{11}+e_{33}$ is the sum of the flexoelectric coefficients. We note that the system of equations (3) and (4) can be simplified substantially by using the following relations, which are satisfied experimentally for $\xi=\omega/\omega_D\sim 1$:

$$\varepsilon' E_0,\ \varepsilon'' E_0 \gg 4\pi f k_z,\quad \frac{d\psi}{ds}\sim 0,\quad \frac{dV_z}{ds}\sim 0. \tag{5}$$

With allowance for (5) we have, according to (3):

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

Let us set

$$Q=\sum_{n=1}^{\infty}(Q_n^c\cos ns+Q_n^s\sin ns)$$

and $\psi=\psi_0$ in the second equation in (6) and keep only the first terms of the expansion of Q. Recognizing that the complex dielectric function $\varepsilon_\parallel=\varepsilon'-i\varepsilon''$ characterizes the response of the system to a complex field $E(t)=E_0e^{i\omega t}$, we obtain

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

from which we get

$$Q_1^c=-\Sigma E_0\psi_0\Big/\left(\frac{1}{\tau_e}+i\right),\qquad Q_1^s=iQ_1^c. \tag{8}$$

Substituting (8) into the first of equations (6), taking its real part, and setting the coefficient of ψ_0 equal to zero, we have

$$\bar{E}_0^2=\frac{E_0^2}{2}=\Gamma_0\left\{\left[\Omega\tau_V\delta+\mathrm{Re}\left(\frac{1}{\eta}\right)\right]\mathrm{Re}\left(\frac{\Sigma}{1/\tau_e+i}\right)+\mathrm{Im}\left(\frac{1}{\eta}\right)\mathrm{Im}\left(\frac{\Sigma}{1/\tau_e+i}\right)+\mathrm{Re}(\Gamma_1)\right\}^{-1}, \tag{9}$$

where

$$\Gamma_0=\frac{k_{11}k_z^2+k_{33}k_x^2}{\gamma_1\omega},\qquad \Gamma_1=\frac{\varepsilon_a\varepsilon_\perp(1+q^2)}{4\pi\nu\gamma_1\omega},$$

and $\bar{E}_0$ is the effective value of the field. Changing variables in (9) to the voltage $U=E_0/k_z$, we obtain a function $U(q)$ whose minimum with respect to z gives the desired threshold voltage and instability wave vector:

$$U_{th}=\min_{q>0}U(q)=U_{th}(q_{th}). \tag{10}$$

The $U(q)$ curves calculated with formula (10) are shown in Fig. 1.

We note that according to (4) one has $|1/\eta|\sim\Omega\tau_V\delta\sim|\Gamma_1|$, whereas

$$\left|\frac{\Sigma}{1/\tau_e+i}\right|\sim|\Sigma|\sim\frac{\bar{\sigma}}{\omega},$$

where $\bar{\sigma}$ is the average low-frequency conductivity. The first

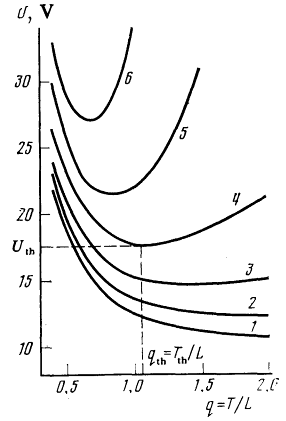

FIG. 1. Voltage at the onset of instability versus the wave-vector ratio $q=k_z/k_x$ of the deformations along the z and x axes; $q\to\infty\,(k_x\to 0)$ corresponds to the Fréedericksz transition. The nematic liquid crystal is in a planar orientation. The numerical calculation was done using Eqs. (9) and (10) for the following values of the parameters: $\varepsilon_\parallel=5.6$, $\varepsilon_\perp=5.25$, $\sigma_\parallel=205$, $\sigma_\parallel/\sigma_\perp=2.17$, $k_{11}=0.85\cdot 10^{-6}$, $k_{33}=1.06\cdot 10^{-6}$, $\alpha_1=0.07$, $\alpha_2=-1.17$, $\alpha_3=-0.017$, $\alpha_4=0.8$, $\alpha_5=0.43$, $n_\parallel=1.784$ cgs esu. The inversion frequency was $f_i=\omega_i/2\pi=16$ kHz, the thickness of the nematic layer was $L=22\,\mu$m. The values of ω/ω_i for curves 1–6 were: 1) 0.85, 2) 0.9, 3) 0.95, 4) 1.0, 5) 1.05, 6) 1.1.

two terms in the denominator of (9) can therefore be neglected with accuracy to terms of higher order in an expansion in $\bar{\sigma}/\omega$, and we have

$$\bar{E}_0^2=\Gamma_0/\mathrm{Re}\,(\Gamma_1). \tag{11}$$

As is seen from (11), the threshold voltage of the instability does not depend on the layer thickness but is determined entirely by the dielectric constants ε_0, $\epsilon_\perp$, ε_∞ and elastic constants k_{11}, k_{33} of the nematic. At the inversion frequency $\omega=\omega_i$, at which $\varepsilon'=\varepsilon_\perp$, the threshold voltage U_{th} and wave vector q_{th} of the instability are [from Eqs. (10) and (11)]:

$$q_{th}=T_{th}/L\sim\left(1+\frac{\varepsilon''^2}{\varepsilon'^2}\right)^{1/4},$$

$$U_{th}=E_{th}L=\frac{\pi}{\varepsilon''(\omega_i)}\left\{8\pi\tilde{k}\varepsilon_\perp\left[1+\left(1+\frac{\varepsilon''^2}{\varepsilon'^2}\right)^{1/2}\right]\right\}^{1/2}, \tag{12}$$

where $\tilde{k}$ is the average value of the elastic modulus of the liquid crystal, and $\varepsilon''(\omega_i)=[(\varepsilon_0-\varepsilon_\perp)(\varepsilon_\perp-\varepsilon_\infty)]^{1/2}$, i.e., the instability vanishes as $\varepsilon_0\to\varepsilon_\perp$ or $\varepsilon_\perp\to\varepsilon_\infty$, and its existence is conditional not only upon the presence of dispersion of the dielectric constant $\varepsilon_\parallel(\omega)$ but also upon a positive dielectric anisotropy of the liquid crystal at low frequencies: $\varepsilon_0=\varepsilon_\parallel(\omega\to 0)>\varepsilon_\perp$. Equation (11) also readily yields the frequency regions $\omega_i(1-\delta\omega_1)<\omega<\omega_i(1+\delta\omega_2)$ in which the instability exists on either side of the inversion frequency. To obtain these we consider the expressions

$$\mathrm{Re}(\Gamma_1)=\frac{\varepsilon_\perp(1+q^2)}{4\pi\gamma_1\omega}\mathrm{Re}\left(\frac{\varepsilon_a}{\nu}\right),$$

$$\mathrm{Re}\left(\frac{\varepsilon_a}{\nu}\right)=\frac{(\varepsilon'-\varepsilon_\perp)(\varepsilon'+\varepsilon_\perp q^2)+\varepsilon''^2}{(\varepsilon'+\varepsilon_\perp q^2)^2+\varepsilon''^2}. \tag{13}$$

Far from the inversion frequency ($\omega\ll\omega_i$) the dielectric loss vanishes ($\varepsilon''\to 0$), and a Fréedericksz transition occurs (the S effect), with a threshold voltage $U_S\sim(\varepsilon'-\varepsilon_\perp)^{-1/2}$. Near the inversion frequency ($\omega\to\omega_i$), where the terms in the numerator of (13) become comparable in order of magnitude,

$$\varepsilon'-\varepsilon_\perp\sim\frac{\varepsilon''^2}{2\varepsilon_\perp},\quad q\sim1,\quad \varepsilon'\sim\varepsilon_\perp, \tag{14}$$

a periodic deformation of the director along the x axis—a domain structure—becomes energetically favorable. It is seen from Fig. 1 that the lower boundary $\omega_i(1-\delta\omega_1)$ for the existence of the domains as determined from (14) is very close to the inversion frequency $\omega_i: \delta\omega_1\sim 0$.

To determine the upper boundary $\omega_i(1+\delta\omega_2)$ for the existence of the instability we equate the denominator of (13) to zero for $q=0$:

$$\varepsilon'-\varepsilon_\perp=-\varepsilon''^2/\varepsilon', \tag{15}$$

and from this expression we obtain, with allowance for (1) and (2),

$$\omega_i(1+\delta\omega_2)\sim(\varepsilon_0/\varepsilon_\infty)^{1/2}\omega_i=\omega_\infty. \tag{16}$$

After using (9), we can evaluate the correction to (16):

$$\omega_\infty\sim\left(\frac{\varepsilon_0}{\varepsilon_\infty}\right)^{1/2}\omega_i +2\pi\sigma_\parallel\frac{\varepsilon''}{\varepsilon'}\frac{(\varepsilon_0-\varepsilon_\infty)}{(\varepsilon_\perp-\varepsilon_\infty)(\varepsilon_0-\varepsilon_\perp)}\left[-\frac{\alpha_2}{\zeta}+\frac{\sigma_\perp}{\sigma_\parallel}-1\right]. \tag{17}$$

We see from (17) that the correction is equal in order of magnitude to $\bar\sigma/\omega_i$ and that the upper frequency boundary ω_∞ for the existence of the instability grows linearly with increasing low-frequency conductivity $\sigma_\parallel$. The frequency dependence of the threshold for the onset of the instability from the planar orientation of the nematic, as calculated with (9) and (10), is shown in Fig. 2a (curve 1). The threshold voltage $U_{\rm th}$ for the formation of the domains increases, and the period $T_{\rm th}$ decreases (curve 1 in Fig. 2b), with increasing frequency of the electric field, and at the inversion frequency $\omega=\omega_i$ the domain period $T_{\rm th}$ is approximately equal to the thickness L of the liquid-crystal layer: $T_{\rm th}\sim L$. The qualitative estimate[5] of the instability threshold (curve 2 of Fig. 2a) that follows from (9) for $k_z=0$ and $T_{\rm th}=L$ understates the threshold values. Near the inversion frequency, $\omega\lesssim\omega_i$, we see that the homogeneous structure of the ordinary Fréedericksz transition (curve 3, Fig. 2a) becomes energetically favorable.

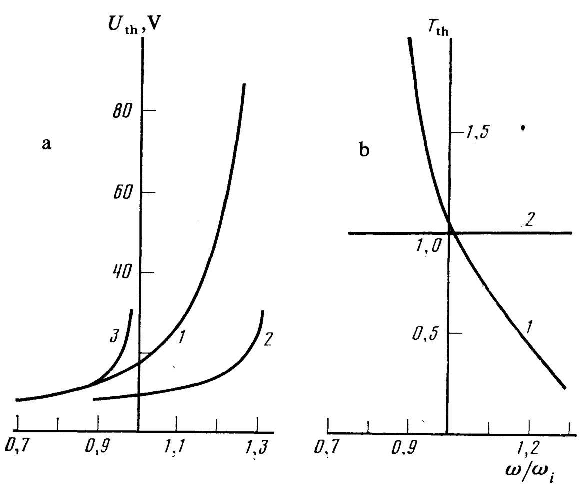

FIG. 2. Calculated frequency dependence of the threshold voltage $U_{\rm th}$ (a) and period $T_{\rm th}$ (b) (the period is in units of L) for the instability arising from the planar orientation of a nematic liquid crystal. The parameters of the nematic are given in the caption to Fig. 1. 1) The two-dimensional theory of Eqs. (9) and (10); 2) the qualitative estimate of Ref. 5; 3) the threshold for the S effect.

We note that all the main results (12)–(17) have been obtained using relation (11), which does not in any way involve the velocity **V** or charge Q of the nematic.

In fact, according to (8) we have

$$|Q|\sim\frac{\bar\sigma}{\omega_i}E_0\psi_0\ll E_0\psi_0,$$
$$|V_z|\sim|\tau_V E_0\,\delta Q|\sim\frac{\bar\sigma}{\omega_i}\frac{E_0^2\psi_0}{\zeta k_x^2}\ll\frac{E_0^2\psi_0}{\zeta k_x^2}, \tag{18}$$

i.e., with accuracy to terms of higher order in $\bar\sigma/\omega_i$ we can assume that the space charge Q and velocity **V** of the liquid crystal are zero [see also the derivation of (11) from (9)]. The only degree of freedom whose perturbation gives rise to the instability in a nematic liquid crystal is the director distribution $\psi=\partial n_z/\partial x$. In view of this let us indicate a simpler method of obtaining relation (11). We introduce the inhomogeneous potential

$$\Phi=-Ez+\Sigma(x,z),\quad |\Sigma|\ll E_0L$$

and write the linearized equation for the rotation of the director with allowance for this inhomogeneous potential:

$$(k_{11}k_z^2+k_{33}k_x^2)\theta-\frac{\varepsilon_a\bar E_0^2\theta}{4\pi}+ik_x\frac{\varepsilon_a}{4\pi}\bar E_0\Sigma=0, \tag{19}$$

where

$$\Sigma=-ik_x\varepsilon_a\bar E_0\theta/(\varepsilon_\parallel k_x^2+\varepsilon_\perp k_z^2),$$

and $\bar E_0$ is the effective field. Substituting into (18) the complex value $\varepsilon_\parallel=\varepsilon'-i\varepsilon''$ and equating the coefficient of θ to zero, we obtain relation (11).

It is seen from (11) and (12) that the threshold characteristics of the instability do not depend on the conductivity or viscosity but are determined solely by the corresponding dielectric and elastic constants of the nematic. The domain structure arises as a consequence of the inhomogeneous static deformation of the director, since the velocity of the macroscopic flow of the liquid crystal is negligibly small (18). This suggests that the instability of the plane-oriented nematic in an alternating electric field near the frequency at which the dielectric anisotropy changes sign is in essence a Fréedericksz transition. The presence of dielectric loss $\varepsilon''(\omega)$ near the inversion frequency has the consequence that a director distribution which is modulated along the x axis, and the nonuniform electric field corresponding to this distribution, are in the present case energetically favored over the ordinary homogeneous Fréedericksz deformation. In particular, the dielectric moment due to the nonuniform field does not go to zero at the point at which the dielectric anisotropy changes sign, where the threshold of the ordinary Fréedericksz transition increases without bound.

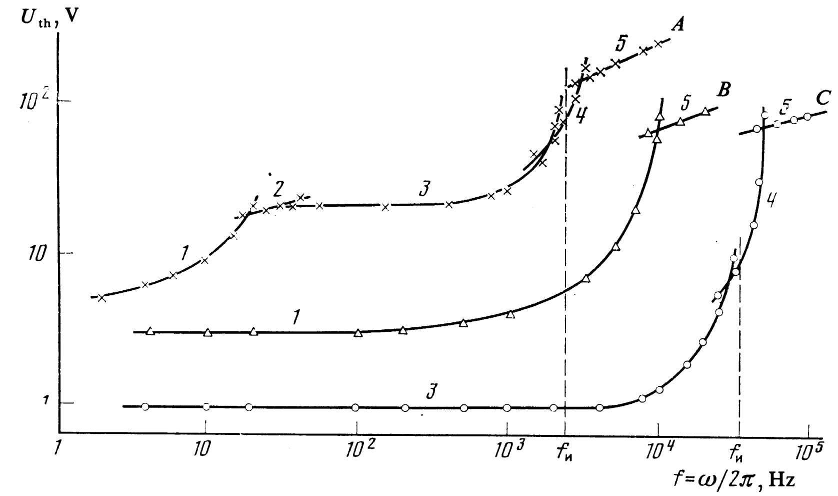

FIG. 3. Frequency characteristics of the threshold voltages for the instabilities in nematic liquid crystals which exhibit an inversion of the sign of the dielectric anisotropy ε_a: A) $\varepsilon_a = +0.05$, $\sigma_\parallel = 126$ sec^{-1}, B) $\varepsilon_a = +0.05$, $\sigma_\parallel = 3400$ sec^{-1}, C) $\varepsilon_a = +6.2$, $\sigma_\parallel = 835$ sec^{-1}.

3. EXPERIMENT AND COMPARISON WITH THEORY

Experimental studies of the domain instability have been carried out on liquid-crystal mixtures exhibiting an inversion of the sign of the dielectric anisotropy ε_a in the frequency regions 3–40 kHz and 0.6–1.0 MHz. The mixtures consisted mainly of *p*-butyl-*p'*-methoxyazoxybenzene and *p*-butyl-*p'*-heptanoyloxyazoxybenzene, taken in a ratio of 2:1 (mixture A), with admixtures of *p*-cyanophenyl o-chloro-*p*-(*p*-*n*-heptylbenzoyloxy) benzoate (1–18 wt.%) or *p'*cyanophenyl *p*-heptylbenzoate (15–40 wt.%).

The threshold characteristics of the instability were studied in sandwich cells with a layer thickness of 15 to 105 μm, for an initial planar director orientation created by rubbing a film, consisting of polyvinyl alcohol with sputtered electrodes of SnO_2, that had been deposited on the plates. The threshold voltage for the formation of the domains was determined by visual inspection in a polarizing microscope and also by diffraction of a laser beam ($\lambda = 0.63\ \mu$m) on the domain structure. The motion of the liquid in the interelectrode gap was observed in a special cell, in the direction perpendicular to the electric field (into the "end" of the cell).

Figure 3 shows the frequency characteristics of the threshold voltages for the instabilities in nematics having an inversion of the sign of the dielectric anisotropy, at various values of the electrical conductivity $\sigma_\parallel$ and ε_a: 1) Williams domains[1] [known as "Kapustin-Williams" domains in the USSR], 2) pre-chevron domains,[1] 3) the Fréedericksz transition, 4) the domain instability under study, 5) an electrohydrodynamic instability of an inertial nature.[8] As is seen in the figure, the instability under study always arises in the region where the dielectric anisotropy changes sign, and its threshold characteristic is practically a continuation of the threshold characteristic of the Fréedericksz transition. With increasing low-frequency dielectric anisotropy $\varepsilon_0 - \varepsilon_\perp$, the threshold voltages for the instability, just as for the Fréedericksz transition, decrease. This is seen by comparing curves A ($\varepsilon_a = +0.05$) and B ($\varepsilon_a = +6.2$). The instability arises only in a poorly conducting nematic, for which the existence region of the Williams domains 1 and pre-chevron domains 2 is bounded on the low-frequency side. At large conductivities of the nematic (in our experiment, for $\sigma_\parallel \gtrsim 10^3$ sec^{-1}) the domain structure 4 and Fréedericksz transition 3 near ω_i do not arise—in this case the Williams domains have a lower threshold voltage and are energetically favorable.

Photographs of the domain instability near the inversion frequency of the dielectric anisotropy are shown in Figs. 4a and 4b for frequencies $\omega \lesssim \omega_i$ and $\omega > \omega_i$, respectively. In terms of both the optical characteristics and the type of director deformation the instability is analogous to the Williams instability. The instability is manifested as linear domains which are oriented perpendicular to the initial direction of the director and which are visible only if the light is polarized in the plane of the director. This indicates that the director suffers a deformation in the *xz* plane. Observations of the behavior of solid particles in the interior of the nematic imply that there is no motion of the liquid. It can thus be concluded that the instability is not electrohydrodynamic, as has been assumed,[3,4] and that the mechanism giving rise to this instability is fundamentally different from that which leads to the Williams domains. In the region $\omega \lesssim \omega_i$ the threshold for the formation of the domain structure is practically the same as the threshold for the ordinary Fréedericksz transition (S effect); here, as the voltage is in-

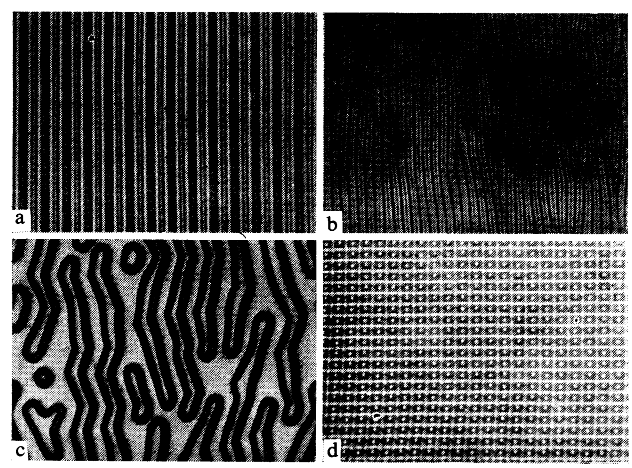

FIG. 4. Instability domain patterns for various external voltages and field frequencies: a) $\omega \lesssim \omega_i$, $U = U_{th}$; b) $\omega > \omega_i$, $U = U_{th}$; c) $\omega \lesssim \omega_i$, $U > U_{th}$; d) $\omega > \omega_i$, $U > U_{th}$.

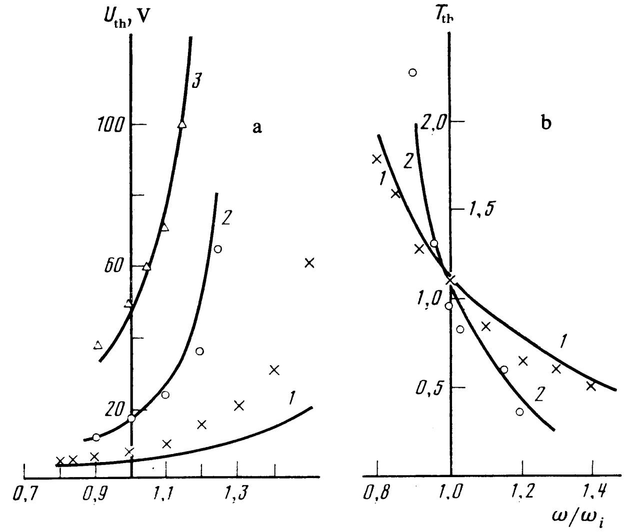

FIG. 5. Frequency characteristics of the threshold voltages U_{th} (a) and period T_{th} (b) (period in units of L) for the instability. 1) $\varepsilon_a = +4.75$; the solid curve is the calculated result and the crosses are experimental data; 2) $\varepsilon_a = +0.35$, the circles are experimental data; 3) $\varepsilon_a = +0.05$, the triangles are experimental data. The other liquid-crystal parameters used in the calculation are given in the caption to Fig. 1.

creased above the threshold value, the domains at first change to a loop-shaped structure[9] (Fig. 4c) and then vanish. If, on the other hand, the field frequency ω is greater than ω_i, a two-dimensional grid with a small period (Fig. 4d) arises at voltages above the threshold.

The threshold voltage of the instability under study is independent of the thickness of the nematic layer over the entire thickness interval studied (5–105 μm). Similarly, changes in the absolute magnitude of the electrical conductivity $\sigma_\parallel$ and its anisotropy $\sigma_\parallel/\sigma_\perp$ had no effect on the threshold characteristics of the instability ($\sigma_\parallel/\sigma_\perp$ was varied from 1.03 to 1.8 in the experiment).

The spatial period T_{th} of the domain structure depended on the frequency of the electric field in the dispersion region $\varepsilon_\parallel$ in accordance with the law $T_{th} \sim \omega^{-\alpha}$, where $\alpha \sim 1/2$, and at the inversion frequency of ε_a was approximately equal to the layer thickness L.

Figure 5 shows a comparison of the theoretical and experimental values of the threshold voltage and instability period as functions of the frequency of the electric field. It is seen that in agreement with the theoretical predictions, the experimental instability threshold increases without bound as $\omega \rightarrow \omega_i(1 + \delta\omega_2)$, with the threshold voltage for the instability decreasing with increasing low-frequency dielectric-constant anisotropy $\varepsilon_0 - \varepsilon_\perp$. The absolute magnitude of the inversion frequency ω_i increases with increasing $\varepsilon_0 - \varepsilon_\perp$, and for this reason the comparison of theory and experiment has been carried out for the relative frequency ω/ω_i. The experimental and theoretical values of the period and threshold voltage of the instability are in good agreement over the entire frequency region. In agreement with our model predictions the point $\varepsilon_a = \varepsilon' - \varepsilon_\perp = 0$ is not a singular point here as it is in the case of the ordinary Fréedericksz transition, so that the instability under study is observed for both positive and negative values of the dielectric anisotropy. The disagreement between the theoretical and experimental curves of $U_{th}(\omega/\omega_i)$, especially in the case of large dielectric anisotropies (Fig. 5), is evidently due to a change in the elastic constants upon doping of the original nematic with admixtures having large values of ε_a. The elastic constants of the mixture could vary with the concentration of the admixture, wherease they were assumed constant in the calculations.

We note in conclusion that on the basis of our comparison of theory and experiment it can be stated with certainty that the domain structure observed in nematic liquid crystals near the inversion frequency arises as a result of a dielectric destabilization of the initial planar orientation of the director of the liquid crystal. This instability is a unique example of a stationary modulated structure in a nematic liquid crystal at a Fréedericksz transition in a uniform external electric field.[7]

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