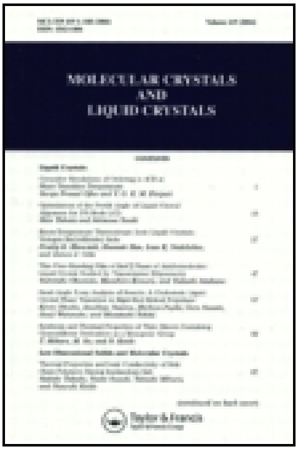

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New Types of Instabilities in Liquid Crystals with Tilted Orientation

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New Types of Instabilities in Liquid Crystals with Tilted Orientation

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We have considered theoretically a new kind of instability formed of rolls perpendicular to Williams' domains when the molecules are initially tilted with respect to the substrate plane. The threshold voltage versus the tilt angle, dielectric constants, electroconductivity, viscosity coefficients and the frequency of an external field is calculated within the framework of the two-dimensional model. The contribution of electrokinetic processes observed in an isotropic liquid to the effect under consideration is discussed and the respective estimates for the threshold are given.

1 DOMAIN STRUCTURE

Williams' domains is a classic example of the electrohydrodynamic (EHD) instability in nematics. They are observed as a periodic distortion of the nematic alignment along an axis X at the initial molecular orientation along X and the external electric field along Z.[1] In this case the molecules remain in the (X, Z) plane and stripes are directed along Y.

There is a possibility for another type of instability if the molecules are initially tilted (a tilt angle θ) with respect to an axis X in the (X, Z) plane.[2,3] This new type of instability is connected with the director rotations around an axis X (X-rotations) or an axis Z (Z-rotations) on small angles (Figure 1). In this case a distortion is periodic along Y and stripes are directed along X. The threshold voltage V_c depends strongly on the tilt angle $\theta : V_c \to \infty$ at $\theta \to 0$; in the case of X-rotations V_c coincides with Williams' threshold at $\theta = \pi/2$.[4]

II BASIC EQUATIONS

The theoretical model, which describes the EHD instability, is the system of linear equations for pertubations of the director orientation ψ, the electric potential U, the charge density Q and the flow velocity ($V_x = 0$, $V_y \neq 0$,

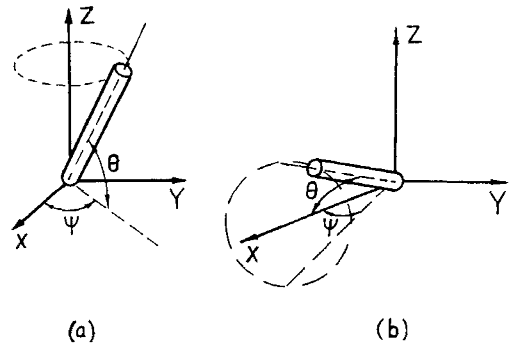

FIGURE 1 Director deviations in the pertubed situation in presence of an electric field along Z: (a) Z-rotations, (b) X-rotations. In the unpertubed situation the director is tilted parallel to the (X, Z) plane ($\psi = 0$) with $\theta \neq 0$.

$V_z \neq 0$). These pertubations depend on y, z and the time t. After excluding the pressure we shall write the acceleration equation for the incompressible fluid in the form:

$$\sin\theta\left(\alpha_2 \frac{d^3\psi}{dz^2 dt} - \alpha_3 \frac{d^3\psi}{dy^2 dt}\right) + \tfrac{1}{2}(\alpha_4 + (\alpha_3 + \alpha_6)\sin^2\theta)\frac{d^4 V_z}{dy^4}$$
$$+ (\alpha_1 \sin^4\theta + \alpha_4 + (\alpha_5 + \alpha_3)\sin^2\theta)\frac{d^4 V_z}{dy^2 dz^2}$$
$$+ \tfrac{1}{2}(\alpha_4 + (\alpha_5 - \alpha_2)\sin^2\theta)\frac{d^4 V_z}{dz^4} = -E\frac{d^2 Q}{dy^2}, \quad (1)$$

where α_j are the Leslie coefficients, $\alpha_3 - \alpha_2 = \gamma_1$. The charge density is given by Poisson's equation

$$4\pi Q + \varepsilon_\perp \frac{d^2 U}{dy^2} + (\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\sin^2\theta)\frac{d^2 U}{dz^2} = (\varepsilon_\parallel - \varepsilon_\perp)E\sin\theta\frac{d\psi}{dy} \quad (2)$$

where $(\varepsilon_\perp, \varepsilon_\parallel)$ and $(\sigma_\perp, \sigma_\parallel)$ are the components of dielectric tensor and the conductivity tensor, respectively. The equation for the electric current has the form

$$\frac{dQ}{dt} + (\sigma_\parallel - \sigma_\perp)E\sin\theta\frac{d\psi}{dy} = \sigma_\perp \frac{d^2 U}{dy^2} + (\sigma_\perp + (\sigma_\parallel - \sigma_\perp)\sin^2\theta)\frac{d^2 U}{dz^2} \quad (3)$$

The equation for a deviation ψ in case of X-rotations reads

$$K_{11}\frac{d^2\psi}{dy^2}+(K_{22}\cos^2\theta+K_{33}\sin^2\theta)\frac{d^2\psi}{dz^2}$$
$$=\frac{\varepsilon_{\parallel}-\varepsilon_{\perp}}{4\pi}\left(E\sin\theta\frac{dU}{dy}+\psi E^2\right)+\sin\theta\left(\alpha_2\frac{dV_y}{dz}+\alpha_3\frac{dV_z}{dy}\right)+\gamma_1\frac{d\psi}{dt}\quad(4)$$

(in case of Z-rotations the term $\psi E^2(\varepsilon_{\parallel}-\varepsilon_{\perp})/4\pi$ is absent). The velocity component V_y is excluded from Eq. (4) due to the equation of incompressibility:

$$\frac{dV_z}{dz}+\frac{dV_y}{dy}=0\quad(5)$$

One has to consider the boundary conditions together with Eqs. (1)–(5). We have at $z=\pm 1/2$ (1 is the layer thickness):

$$V_z=0,\qquad \frac{dV_z}{dz}=0,\qquad U=0.\quad(6)$$

We shall write the boundary conditions for ψ taking into account the finite anchoring energy W:

$$K\frac{d\psi}{dz}+W\psi=0,\qquad K=\pm(K_{22}\cos^2\theta+K_{33}\sin^2\theta)\quad\text{at } z=\pm 1/2\quad(7)$$

III RESULTS IN CONSTANT FIELD

At low frequencies the threshold characteristics (the period d_c along Y and the voltage V_c) are found exactly by the computer calculations of the boundary problem, Eqs. (1)–(7). The exact solution of this problem have the form

$$(\psi, V_y, V_z, U)\sim\exp(iqy)\sum_{j=1}^{4}a_j\cos(p_jz),\quad(8)$$

where the wavelengths $p_j(q, V)$ are chosen according to the Eqs. (1)–(5). The threshold voltage is a minimum $V=V_c$ at $q=q_c$ on the curve $V(q)$, which is obtained by inserting the solutions (ψ, V_y, V_z, U) into Eqs. (6) and (7).

At strong anchoring ($W\gg K/1$) we have $p\sim\pi/1$ and $q_c\gg p$. In this case one can estimate $V_c=E_c 1$ and $d_c=\pi/q_c$:

$$V_c\approx\frac{4\pi^{3/2}}{(a\sin^2\theta+c)}K_{11}^{1/2}(b\sin^2\theta+g)^{1/2},$$
$$d_c\approx 1\left(\frac{a\sin^2\theta+c}{2(b\sin^2\theta+g)}\right)^{1/2}.\quad(9)$$

If we have X-rotations (θ is not very small) the constants a, b, c, g are given by

$$a = 2\left(\varepsilon_{\parallel} - \varepsilon_{\perp}\frac{\sigma_{\parallel}}{\sigma_{\perp}}\right)^2 \frac{\alpha_3}{\alpha_4} - (\varepsilon_{\parallel} - \varepsilon_{\perp})\left(\frac{\sigma_{\parallel}}{\sigma_{\perp}} - 1\right),$$

$$b = -\frac{2\alpha_2}{\alpha_4}\left(\varepsilon_{\perp}\frac{\sigma_{\parallel}}{\sigma_{\perp}} - \varepsilon_{\parallel}\right) + 2\left(\frac{\sigma_{\parallel}}{\sigma_{\perp}} - 1\right)(\varepsilon_{\parallel} - \varepsilon_{\perp}), \tag{10}$$

$$c = \varepsilon_{\perp} - \varepsilon_{\parallel}, \qquad g = (\varepsilon_{\parallel} - \varepsilon_{\perp})\left(\frac{\sigma_{\parallel}}{\sigma_{\perp}} + 2\right).$$

In case of Z-rotations (or $\varepsilon_{\perp} = \varepsilon_{\parallel}$) we have $c = g = 0$ and

$$V_c \approx \frac{4\pi^{3/2}}{a \sin\theta}(bK_{11})^{1/2},$$

i.e. the threshold voltage is infinite at $\theta = 0$.

The estimations Eqs. (9) and (10) show that V_c is determined basically by the dielectric anisotropy $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$, the surface energy W (per unit area) and the tilt angle θ. The Leslie coefficient α_3 is important at $\Delta\varepsilon \approx 0$ only ($V_c \sim |\alpha_3|^{-1}$ in this case). At weak anchoring and $\Delta\varepsilon \neq 0$ it is necessary to

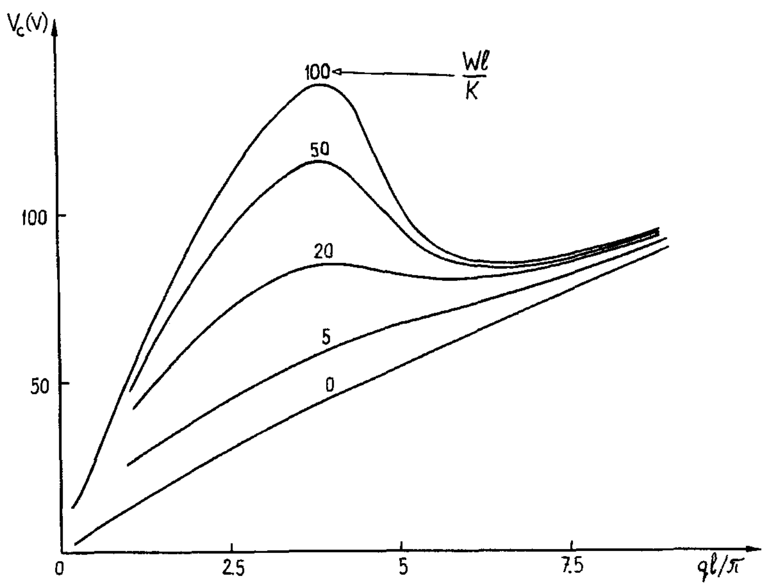

FIGURE 2 Typical voltage versus wave-vector of static distortions (X-rotations), when $K_{11} = 0.677.10^{-6}$, $K_{22}/K_{11} = 0.81$, $K_{33}/K_{11} = 1.22$, $l = 20$ μm, $\alpha_1 = 0.07$, $\alpha_2 = -1.112$, $\alpha_3 = -0.02$, $\alpha_4 = 0.84$, $\alpha_5 = 0.46$, $\varepsilon_{\parallel} = 5.2$, $\varepsilon_{\perp} = 5.25$, $\sigma_{\parallel}/\sigma_{\perp} = 1.5$ cgs units.

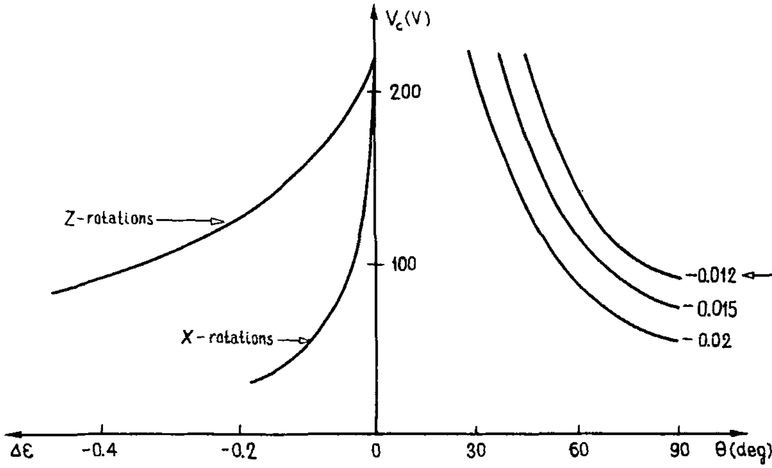

FIGURE 3 Threshold voltage for X-rotations versus tilt angle. Threshold voltage versus dielectric anisotropy: upper curve for Z-rotations; lower curve for X-rotations ($\theta = 30$ deg, $\alpha_3 = -0.02$ cgs units). The rest constants are the same as in Figure 2.

take into account the field effects. At $\theta = \pi/2$ the Eqs. (9) give estimates of the threshold for a homeotropic texture with $\Delta\varepsilon \geq 0$.

The results of computer calculations for small frequencies are shown in Figures 2 and 3. Figure 2 shows that the existence of threshold for the static distortions depends on W. The dependence of the threshold V_c on the parameters $\Delta\varepsilon$ and θ are shown in Figure 3. One can see that the threshold depends strongly on the coefficient α_3 at $|\Delta\varepsilon| \ll 1$. The threshold for the static X-rotations exists only at certain values of $\Delta\varepsilon$. The threshold V_c calculated at $\theta = \pi/2$, $\Delta\varepsilon = 0$, $W \sim 10^3$ and $q_c = 14\pi/1$ coincides with the experimental value $93V$.[4]

IV TIME DEPENDENT MODEL

The variation of the threshold V_c with frequency f is determined as in case of the chevron regime.[5] The equations are transformed into equations for the curvature $\phi = d\psi/dy$ and for the charge Q:

$$\frac{d\phi}{dt} = \frac{\phi}{T^*} + \frac{QE}{\eta} = 0,$$
$$\frac{dQ}{dt} + \frac{Q}{\tau} + \sigma^* E\phi = 0, \qquad (11)$$

where it is assumed that $q_c \gg p$; $\tau = (\varepsilon_\perp/4\pi\sigma_\perp)$ describes dielectric relaxation,

$$\sigma^* = \sin\theta\left(\sigma_\parallel - \sigma_\perp\frac{\varepsilon_\parallel}{\varepsilon_\perp}\right)$$

is the effective conductivity,

$$\eta = -\frac{\alpha_2}{\sin\theta(1 - (\varepsilon_\parallel/\varepsilon_\perp) - 2(\alpha_3/\alpha_4))}$$

is the effective viscosity,

$$T^* = \frac{-\alpha_2}{K_{11}q^2 + \left(\dfrac{\varepsilon_\parallel}{\varepsilon_\perp} - 1\right)\dfrac{E^2}{4\pi}(\varepsilon_\perp + (\varepsilon_\parallel - \varepsilon_\perp)\sin^2\theta)}$$

is the relaxation time for molecular orientation. The dependence $E(q)$ is obtained here from the condition of periodicity for the functions Q and ϕ. The threshold characteristics are found for the step-like electric field:

$$E(t) = \begin{cases} E & \text{at } t_0 \le t < t_0 + (T/2) \\ -E & \text{at } t_0 + (T/2) \le t < t_0 + T \end{cases}$$

The dependence of V_c on $f = 1/T$ calculated for $p = \pi/1$ and $p = 0$ is depicted on Figure 4. Evidently the value $p = \pi/1$ corresponds to a strong anchoring[6] and the value $p = 0$ corresponds to a weak anchoring. At high frequencies the curves 2 and 3 are close, i.e. the influence of the boundary

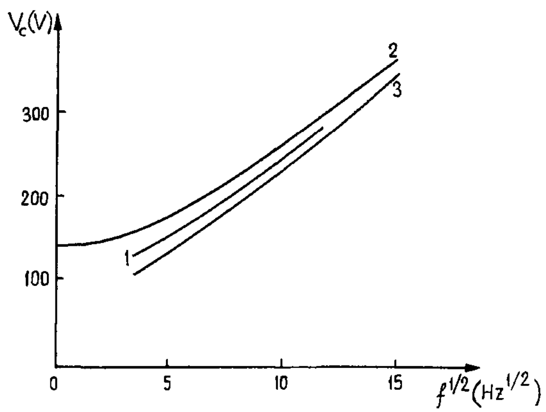

FIGURE 4 Threshold voltage versus frequency. Curve 1: $\theta = 30$ deg, $\varepsilon_\parallel = 4.75$, $\varepsilon_\perp = 5.25$, $p = \pi/1$; curve 2: $\theta = 90$ deg, $\varepsilon_\parallel = \varepsilon_\perp = 5.25$, $p = \pi/1$; curve 3: $\theta = 90$ deg, $\varepsilon_\parallel = \varepsilon_\perp = 5.25$, $p = 0$. The rest constants are: $K_{11} = 0.677.10^{-6}$, $K_{22} = 0.5.10^{-6}$, $K_{33} = 0.826.10^{-6}$, $1 = 20\,\mu$m, $\sigma_\parallel/\sigma_\perp = 1.4$, $\sigma_\parallel = 90$, $\alpha_1 = 0.07$, $\alpha_2 = -0.78$, $\alpha_3 = -0.012$, $\alpha_4 = 0.84$, $\alpha_5 = 0.46$ cgs units.

conditions is decreasing. These curves describe the dielectric regime in which the proportionality $V_c \sim q_c \sim f^{1/2}$ takes place. Such frequency dependence was observed experimentally for the new type of EHD instability in MBBA.[3]

The instabilities described above which occur only in anisotropic liquids have in general a more complex character due to well-known electro-convective effects observed in an isotropic liquid too. The role of these effects depends on internal parameters of the substance. In particular a liquid crystal must be considered as an electrolite with typical electrokinetic processes. It is possible to estimate the contribution of these processes to the effect under consideration, taking into account an inhomogeneous distribution of the volume charge Q_0 before an instability. In case of weak electric currents such distribution has the form:[7]

$$\frac{dQ_0}{dz} = -\nu E \tag{12}$$

where $\nu \sim \sigma/D$, D is the average diffusivity, σ is the average electroconductivity. Thus the second part of Eq. (11) has the additional term

$$V_z(dQ_0/dz) = -\nu E V_z$$

which is very important if $\nu \gg (\Delta\sigma \cdot \gamma_1/K)$, $\Delta\sigma = \sigma_{\parallel} - \sigma_{\perp}$. In the last case the main cause of the instability is a destruction of the distributions Q_0. The qualitative consideration which is similar to Eq. (11) (where the first equation is absent now and $V_z = \alpha_4^{-1} E q^2 (q^2 + p^2)^{-2}$ in the second one) shows that the instability threshold is

$$E_c^2 = \frac{16\pi\sigma\alpha_4 p^2}{\varepsilon\nu}$$

Here $p \sim \pi/h$; at low frequencies ($\omega = 2\pi f < 4\pi\sigma/\varepsilon$) h is the thickness of electric double layer or the Debye radius, i.e. $h^2 \sim (D\varepsilon/4\pi\sigma)$; at higher frequencies ($\omega > 4\pi\sigma/\varepsilon$) h is the diffusion length, i.e. $h^2 \sim D/\omega$.

At certain parameters of a substance the new type of EHD instability has the lower threshold[3] than the familiar Williams' domains and chevrons. The type of instability considered above is also possible in smectics C. The experimental data confirm this. In smectics C the threshold voltage must increase near the temperature of the phase transition smectic C $\rightleftharpoons$ smectic A.

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