HIGH-FREQUENCY ELECTRO-HYDRODYNAMICAL EFFECT IN LIQUID CRYSTALS

S. A. PIKIN

Institute of Crystallography, USSR Academy of Sciences

Submitted June 8, 1971

Zh. Eksp. Teor. Fiz. 61, 2133–2139 (November, 1971)

The electro-hydrodynamical effect in liquid crystals is examined for frequencies of the alternating electric field which are much larger than the inverse relaxation time of the volume charge. It is established that for a fixed frequency ω the effect exists in the region of field amplitudes $E \geq E_{min}(\omega)$, and for a fixed electric field amplitude E—the effect exists in the frequency range $\omega \leq \omega_{max}(E)$. The dependence of the wave vector of the generated periodic structure on the frequency ω is found.

1. Depending on the frequency of the variable electric field $E(t) = E \sin \omega t$ the electro-hydrodynamical effect in liquid crystals has a different qualitative character. The onset of stationary flow of a nematic liquid, analogous to stationary convection, under the influence of low-frequency electric fields (the frequency ω is small in comparison with the inverse relaxation time τ_e^{-1} of the volume charge in the anisotropic liquid) was considered in [1]. In this case the volume charges, being generated in the liquid crystal due to the anisotropy of the conductivity, play an essential role.

The formation of a periodic structure in a layer of liquid crystal, placed between the plates of a capacitor, in fields with $\omega \gg \tau_e^{-1}$ is investigated in the present article. Here one can neglect the volume charges and regard the liquid as electrically neutral. In this connection the mechanism for the formation of a banded structure which is periodically changing with time becomes a purely dielectric mechanism.[2,3] Such an effect, which has been observed experimentally in [2–4], has the following characteristics: 1) For a fixed amplitude E there is a range of frequencies $\tau_e^{-1} < \omega \leq \omega_{max}$ in which the corresponding electro-optical effect is observed; for $\omega > \omega_{max}$ the effect vanishes; 2) the thickness of the layer of anisotropic liquid significantly influences the threshold characteristics of the generated structure; 3) the threshold value of the amplitude E_{min} depends on the frequency and increases with increasing ω (the proportional relation $E_{min}^2 \sim \omega$ is theoretically derived in [4]); 4) the period d_c of the banded structure may markedly depend on the frequency ω and may be much smaller than the layer thickness l. These characteristic features of the high-frequency electro-hydrodynamical effect are accounted for in the present article. The threshold characteristics E_{min} and d_c are found in a unique manner from the minimum of the functional dependence E(d) which we assume for the regime under consideration.

2. We take the geometry of the liquid-crystal layer to be the same as in the problem[1]: The molecules are oriented on the average along the x axis, parallel to the plane of the electrode, and the electric field is directed along the z axis, normal to the plane of the electrode; a small deviation of the long axes of the molecules from their average orientation takes place in the zx plane and is characterized by the angle θ. One can show that for negative anisotropy of the dielectric permittivity ϵ_{ik} ($\epsilon_a = \epsilon_{\|} - \epsilon_{\perp} < 0$), taking account of the deviations of the axes of the molecules from the zx plane and also taking the dependence on the y coordinate into account leads to second-order quantities in the equations. According to [1] and the assumptions made above, the system of linearized equations describing a layer of anisotropic liquid has the following form.

The equation of continuity for an incompressible fluid:

$$\partial v_x / \partial x + \partial v_z / \partial z = 0. \tag{1}$$

The Navier-Stokes equations:

$$\begin{aligned} \rho \frac{\partial v_x}{\partial t} &= -\frac{\partial P}{\partial x} + \beta_1 \frac{\partial^2 v_x}{\partial x^2} + \beta_2 \frac{\partial^2 v_x}{\partial z^2} + \alpha_3 \frac{\partial^2 \theta}{\partial t \partial z} \\ \rho \frac{\partial v_z}{\partial t} &= -\frac{\partial P}{\partial z} + \beta_3 \frac{\partial^2 v_z}{\partial x^2} + \beta_4 \frac{\partial^2 v_z}{\partial z^2} + \alpha_2 \frac{\partial^2 \theta}{\partial t \partial x}, \end{aligned} \tag{2}$$

where ρ is the density of the liquid, P is the pressure, and the viscosity constants β_j are related to the Leslie constants α_i by the relations

$$\beta_1 = \alpha_1 + \alpha_5 + \tfrac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6), \quad \beta_2 = \tfrac{1}{2}(\alpha_3 + \alpha_4 + \alpha_6),$$
$$\beta_3 = \tfrac{1}{2}(-\alpha_2 + \alpha_4 + \alpha_5), \quad \beta_4 = \tfrac{1}{2}(-\alpha_2 + \alpha_4 - \alpha_5).$$

Neglecting the small moment of inertia of the liquid, the equation of motion of the director is given by:

$$\begin{aligned} &\frac{1}{4\pi} \varepsilon_a E(t) \left(\theta E(t) - \frac{\partial \psi}{\partial x} \right) + K_{11} \frac{\partial^2 \theta}{\partial z^2} + K_{33} \frac{\partial^2 \theta}{\partial x^2} \\ &= \frac{\gamma_1 + \gamma_2}{2} \frac{\partial v_x}{\partial z} + \frac{\gamma_2 - \gamma_1}{2} \frac{\partial v_z}{\partial x} + \gamma_1 \frac{\partial \theta}{\partial t}, \end{aligned} \tag{3}$$

where ψ denotes the small deviation of the potential $\varphi = -E(t)z + \psi$ from the value $-E(t)z$, $-l/2 \leq z \leq l/2$, $\gamma_1 = \alpha_3 - \alpha_2$, $\gamma_2 = \alpha_6 - \alpha_5$, and K_{11} and K_{33} are the elastic constants.

Maxwell's equation:

$$\varepsilon_a E(t) \frac{\partial \theta}{\partial x} - \varepsilon_{\|} \frac{\partial^2 \psi}{\partial x^2} - \varepsilon_{\perp} \frac{\partial^2 \psi}{\partial z^2} = 0. \tag{4}$$

One can write the boundary conditions for the system of equations (1) through (4) in the form

$$\theta = 0, \quad \psi = 0, \quad v_x = v_z = 0 \text{ for } z = \pm l/2. \tag{5}$$

The first condition in (5) is achieved by special processing of the solid surface (by polishing the plates along the x axis).

Out of the parameters of the problem one can form the following dimensionless combinations which appear in Eqs. (1) through (4):

$$-\frac{\varepsilon_a E^2}{8\pi\beta\omega}, \quad \frac{K}{\beta\omega}\left(\frac{\pi}{l}\right)^2, \quad \frac{K}{\beta\omega}\left(\frac{\pi}{d}\right)^2, \quad \frac{\beta}{\rho\omega}\left(\frac{\pi}{l}\right)^2, \quad \frac{\beta}{\rho\omega}\left(\frac{\pi}{d}\right)^2,$$

where K and β are the average constants of elasticity and viscosity. Since $\rho K \ll \beta^2$ in a liquid crystal, and the threshold values of the quantities $(-\epsilon_a E^2/8\beta\omega)$, $(\beta/\rho\omega) \times (\pi/l)^2$, or $(\beta/\rho\omega)(\pi/d)^2$ are larger than or of the order of unity, then one can neglect the terms containing K_{11} and K_{33} in the equations which determine the threshold characteristics of the generated structure. We also take into consideration that in a nematic liquid $\gamma_2 = \alpha_3 + \alpha_2$,[5] $\alpha_1 \approx 0$, $\alpha_3 = (\gamma_1 + \gamma_2)/2 \approx 0$, $\alpha_2 \approx \beta_2 - \beta_3$, and $|\epsilon_a| \ll \epsilon_\perp$.

It is necessary to take the elasticity of the liquid crystal into account in a thin boundary layer, whose thickness in order of magnitude is given by

$$h \sim \left(-\frac{8\pi K}{\varepsilon_a E^2}\right)^{1/2} \leqslant \left(\frac{\rho K}{\beta^2}\right)^{1/2} l \ll (l, d).$$

In this layer θ rapidly changes from zero on the solid surface to a certain value $\tilde{\theta}$, which is obtained from the solution of the equations for the base layer. Since $h \ll d$, in the boundary layer Eq. (3) takes the form

$$\frac{1}{4\pi}\varepsilon_a E^2(t)\theta + K_{11}\frac{\partial^2\theta}{dz^2} = -\alpha_2\frac{\partial\theta}{\partial t}. \qquad (6)$$

The solution of Eq. (6), having a periodic dependence on the time, is given by the series

$$\theta(t) = \sum_{n=-\infty}^{\infty} a_n \operatorname{sh}(q_n\tilde{z})\operatorname{sh}^{-1}(q_n\tilde{h})\exp\left[in\omega t + \left(\frac{\varepsilon_a E^2}{16\pi\alpha_2\omega}\right)\sin(2\omega t)\right],$$

where

$$q_n^2 = -\frac{\varepsilon_a E^2}{8\pi K_{11}} - i\frac{\alpha_2 n\omega}{K_{11}}, 0 \leqslant \tilde{z} = \frac{l}{2} - |z| \leqslant \tilde{h}, \quad h < \tilde{h} \ll (l, d).$$

The coefficients a_n are determined from the condition that on the boundary of the base layer the obtained series must be equal to the function $\tilde{\theta}(t)$, which is the value of the solution of the equations for the base layer when $|z| = (l/2) - \tilde{h}$:

$$a_n = \frac{\omega}{2\pi}\int_0^{(2\pi/\omega)} \tilde{\theta}(t)\exp\left[-in\omega t - \left(\frac{\varepsilon_a E^2}{16\pi\alpha_2\omega}\right)\sin(2\omega t)\right]dt.$$

With the aid of Eqs. (1) and (2), where $\alpha_3 = 0$, we find that the component v_x of the velocity tangential to the solid surface varies rapidly in the boundary layer according to the law $v_x \sim (\tilde{z}/\tilde{h})\tilde{v}_x$, where $\tilde{v}_x$ is the value of the velocity v_x on the boundary of the base layer, which is obtained from the solution of the equations for the base layer. The variation of the velocity component v_z normal to the surface is small in the boundary layer and amounts to a quantity of the order of $(h/d)\tilde{v}_x$. The distribution of the electric field potential near the solid surface can be determined in similar fashion.

Eliminating the function ψ, v_x, and v_z from Eqs. (1)–(4), we find that in the base layer the function $\Xi = \partial\theta/\partial x$ must satisfy the equation

$$\left\{\frac{\partial^2}{\partial t^2}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)^2 - \frac{\partial}{\partial t}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)^2\left[\frac{1}{\rho}\left(\beta_2\frac{\partial^2}{\partial z^2} + \beta_3\frac{\partial^2}{\partial x^2}\right) - \frac{\varepsilon_a E^2(t)}{4\pi\alpha_2}\right] - \frac{\varepsilon_a E^2(t)}{4\pi\alpha_2}\frac{1}{\rho}\left(\beta_2\frac{\partial^2}{\partial z^2} + \beta_3\frac{\partial^2}{\partial x^2}\right)\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial z^2}\right)\right\}\Xi = 0. \qquad (7)$$

Since the change of the velocity v_z in the boundary layer is small and the true behavior of the normal component of the velocity does not have any singularities in this layer, one can also require fulfilment of the condition v_z on the boundary of the base layer. As a consequence, on the boundary of the base layer the function Ξ must satisfy the condition $\Xi = 0$ with the same degree of accuracy, according to Eqs. (1)–(5).

3. We seek the solution of Eq. (7) in the form $\Xi = f(t)\exp(ikx)\cos(p_{+n}z)$ or $\Xi = f(t)\exp(ikx)\sin(p_{-n}z)$, where f(t) is a periodic function of the time and

$$p_{+n} = \frac{(2n+1)\pi}{l-2\tilde{h}}(n = 0, \pm 1, \ldots), \quad p_{-n} = \frac{2n\pi}{l-2\tilde{h}}(n = \pm 1, \pm 2, \ldots).$$

Since $\tilde{h} \ll l$, we shall neglect below the quantity $\tilde{h}$. The least distorted structure of a liquid crystal corresponds to the wave vector $p = p_{+0} \approx \pi/l$. As a result, by substituting $E(t) = E \sin \omega t$ and introducing the dimensionless variable $\xi = \omega t/\pi$ we obtain the following equation for the function f:

$$\frac{\partial^2 f}{\partial \xi^2} + [\lambda + \nu(1-\cos(2\pi\xi))]\frac{\partial f}{\partial \xi} + [\mu\nu(1-\cos(2\pi\xi)) + 2\pi\nu\sin(2\pi\xi)]f = 0, \qquad (8)$$

where

$$\nu = \frac{\varepsilon_a E^2}{8\alpha_2\omega}, \quad \mu = \frac{\pi}{\rho\omega}(\beta_2 p^2 + \beta_3 k^2), \quad \lambda = \mu + \frac{\pi\alpha_2}{\rho\omega}\frac{k^4}{k^2+p^2}.$$

In order for Eq. (8) to have a periodic solution corresponding to the experimentally observed oscillating regime, it is necessary and sufficient if only one of the roots of the characteristic equation is given by $\sigma = 1$.[6] In the present case the characteristic equation has the form

$$\sigma^2 - (a_{1,1} + a_{2,2})\sigma + a_{1,1}a_{2,2} - a_{1,2}a_{2,1} = 0, \qquad (9)$$

where

$$a_{i,r+1} = \sum_{s=0}^{\infty} f_{s,i}^{(r)}(1) \qquad (i = 1, 2, r = 0, 1),$$

$$f_{s,i}^{(r)}(\xi) = \frac{\partial^r}{\partial\xi^r}f_{s,i}(\xi), \qquad f_{0,1}(\xi) = 1, \qquad f_{0,2}(\xi) = \xi,$$

$$f_{s+1,i}(\xi) = -\int_0^\xi (\xi - u)\{[\mu\nu(1-\cos(2\pi u)) + 2\pi\nu\sin(2\pi u)]f_{s,i}(u) + [\lambda + \nu(1-\cos(2\pi u))]f_{s,i}^{(1)}(u)\}du.$$

Equating the root of Eq. (9) to the value $\sigma = 1$, we obtain the function $\nu(\mu, \lambda)$, where according to Eq. (8) μ is a function of λ and of the ratio $(\omega_0/\omega) \sim (\beta p^2/\rho\omega)$. The equation for ν as a function of μ and λ is represented by a series in powers of ν with coefficients which depend on μ and λ. The solution of this equation is found numerically. The excitation threshold associated with a fixed frequency ω corresponds to the minimum value $\nu_c(\omega_0/\omega)$ of the function ν when the value of the parameter λ is equal to $\lambda_c(\omega_0/\omega)$. In the case $\omega > \omega_0$, $\beta_3 \approx 2\beta_2$ (in paraazoxyanisole) the threshold characteristics of the oscillatory regime with frequency 2ω are given by $\nu_c \approx 1$ and $\lambda_c \approx 1$.

Using the characteristic equation one can verify that the regime under consideration is energetically the most favorable, that is, it corresponds to the smallest threshold values for the quantities ν or E. The non-periodic solutions, which correspond to complex values of σ ($\sigma = e^{i\eta}$, $|\sigma| = 1$, $f(t + \pi/\omega) = f(t)e^{i\eta}$) are less favorable since in this case $\nu_c > 2$. Thus, in the range of frequencies $\omega > \omega_0 \sim \beta p^2/\rho$ the threshold amplitude E_{min} of the electric field, at which the oscillatory regime with frequency ω appears, is given by

$$E_{min}(\omega > \omega_0) \approx 2\sqrt{2\alpha_2\omega / \varepsilon_a}. \tag{10}$$

For a fixed amplitude E the considered effect will obviously exist in the range of frequencies $\omega \le \omega_{max} \approx \epsilon_a E^2/8\alpha_2$. For $E = E_{min}(\omega > \omega_0)$ the wave vector k of the generated structure, which is periodic along the x axis, is given by the formula

$$k_c^2(\omega > \omega_0) \approx \rho\omega / \pi\beta_2 > p^2, \tag{11}$$

and the period $d_c = \pi/k_c$ is correspondingly given by

$$d_c(\omega > \omega_0) \approx \pi\sqrt{\pi\beta_2/\rho\omega} < l. \tag{12}$$

From Eqs. (10)–(12) it is clear that in contrast to the low-frequency electro-hydrodynamical effect,[1] for larger frequencies of the electric field the threshold voltage $V_{min} = lE_{min}$ is proportional to the thickness of the liquid-crystal layer, but the period d_c of the structure does not depend on the layer thickness l.

Since $\omega_0 \sim l^{-2}$, the frequency ω_0 for a sufficiently large thickness of the liquid-crystal layer becomes of the order of magnitude of the inverse relaxation time of the volume charge. In this case for $\omega \lesssim \omega_0 \sim \tau_e^{-1}$ the mechanism for the formation of the banded structure connected with the anisotropy of the conductivity[1,4] is switched on, but for $\omega > \tau_e^{-1}$ the dielectric mechanism leads to the frequency dependences (10)–(12).

If the layer thickness l is such that $\omega_0 \gg \tau_e^{-1}$, then there exists a frequency range $\tau_e^{-1} < \omega < \omega_0$ in which the dielectric mechanism for the formation of the banded structure leads to a behavior of the threshold characteristics which differs from (10)–(12). For $\omega < \omega_0$ the parameters λ and μ are much larger than unity and in this case the solution of Eq. (8) is written in the form $f(\xi) = \exp(\lambda g(\xi))$, where $g(\xi)$ is represented by the series $g = g_0 + (1/\lambda) g_1 + \ldots$ in powers of $(1/\lambda)$. In the approximation which is analogous to the quasiclassical approximation, the dependence $\nu(\mu, \lambda)$ corresponding to the oscillatory regime with frequency 2ω is found from the "quantization" condition

$$\oint\left\{\left[\nu\sin^2(\pi\xi) - \frac{1}{2}\lambda - \delta + \sqrt{\delta(\lambda+\delta)}\right]\right. \\ \left.\times\left[\frac{1}{2}\lambda + \delta - \sqrt{\delta(\lambda+\delta)} - \nu\sin^2(\pi\xi)\right] + \pi\nu\sin(2\pi\xi)\right\}^{1/2} d\xi \approx \pi, \tag{13}$$

where $\delta = \mu - \lambda$, and the integral is taken over one complete period of the "classical motion." Since this integral is small for $\lambda_c \gg 1$, $\delta_c \ll \lambda_c$, and $\nu_c \sim (\lambda_c/2)$, then by rewriting $\sin(\pi\xi)$ in the form $1 - \frac{1}{2}\pi^2(\frac{1}{2} - \xi)^2$ from Eq. (13) we find that

$$\nu \approx \frac{\lambda}{2} - 0.4\sqrt{\delta\lambda} + \left(\frac{\pi}{4}\right)^2\frac{1}{\delta}, \tag{14}$$

where

$$\lambda \approx \frac{\pi\beta}{\rho\omega}(p^2 + 2k^2), \quad \delta \approx \frac{\pi\beta}{\rho\omega}\frac{k^4}{p^2}, \quad \beta = \beta_2 \approx \frac{\beta_3}{2}.$$

From here we obtain

$$\nu_c \approx \frac{\pi}{2}\frac{\beta p^2}{\rho\omega} + 0.5\pi\left(\frac{\beta p^2}{\rho\omega}\right)^{1/3} \qquad k_c^2 \approx 0.6p^2\left(\frac{\rho\omega}{\beta p^2}\right)^{2/3} \tag{15}$$

or

$$V_{min}(\omega < \omega_0) \approx 2\pi\beta(\pi/\rho|\varepsilon_a|)^{1/2}\left[1 + 0.5\left(\frac{\rho\omega}{\beta p^2}\right)^{2/3}\right], \tag{16}$$

$$d_c(\omega < \omega_0) \approx 1.3l\left(\frac{\beta p^2}{\rho\omega}\right)^{1/6}.$$

According to expression (16) the threshold voltages and the period of the structure vary substantially more slowly in the frequency range $\omega < \omega_0$ than they do in the range $\omega > \omega_0$, and also the difference of the potentials V_{min} here weakly depends on the layer thickness l, and the period d is a quantity of the order of l.

Formulas (15) and (16) are valid in the absence of volume charge for frequencies which are larger than the inverse relaxation time of the structure, $\tau_s^{-1} \sim (Kp^2/\beta)$, which is due to the influence of the walls. The electric field can be regarded as constant in the frequency range $\omega \ll \tau_s^{-1}$. However, Eqs. (1)–(4) do not have a stationary solution except for the trivial solution, since there is no volume electric charge in the liquid according to the conditions of the problem (for $\tau_e^{-1} \ll \tau_s^{-1}$). In actual fact the oscillatory regime under consideration becomes energetically unfavorable at a certain frequency $\omega = \omega^*$. For $\omega < \omega^*$ the smallest value of ν corresponds to arbitrarily small values of the wave vector k.

In order to calculate the critical frequency ω^*, in Eqs. (7) and (8) it is necessary to keep the term $(-K_{11}p^2)$ of the series with the quantity $(\epsilon_a E^2(t)/4\pi)$. Consequently Eq. (14) is written in the form

$$\nu \approx \frac{\lambda}{2} - 0.4\sqrt{\lambda(\delta+\zeta)} + \left(\frac{\pi}{4}\right)^2\frac{1}{\delta+\zeta}, \tag{17}$$

where λ and δ are the same as in (14) and $\zeta = (\pi K_{11}p^2/\beta\omega)$. From (17) we find that the critical frequency is given by

$$\omega^* \approx 5(\beta^2/\rho K_{11})^{1/4}(K_{11}p^2/\beta).$$

According to (16), for $\omega = \omega^*$ the threshold values of the voltage and the period of the structure are given by

$$V^* \approx 2\pi\beta\left(\frac{\pi}{\rho|\varepsilon_a|}\right)^{1/2}\left[1 + 1.6\left(\frac{\rho K_{11}}{\beta^2}\right)^{1/3}\right], \quad d^* \approx 0.8\left(\frac{\beta^2}{\rho K_{11}}\right)^{1/4} l.$$

The values k = 0 or d = ∞ give the same value for the threshold difference of the potentials for $\omega = \omega^*$, but they correspond to an undistorted structure of the liquid crystal, which must also be realized for frequencies of the electric field smaller than the critical frequency.

4. The obtained results qualitatively agree with the experimental data. In order to make a quantitative comparison it is necessary to have more complete information about the parameters of specific substances. The form of the boundary conditions at frequencies ω much larger than the inverse relaxation time of the volume charge is not important, since in this case the solid surface influences the behavior of the anisotropic liquid only in the thin boundary layer.

In conclusion I thank A. I. Larkin for fruitful discussions and L. K. Vistin' for acquainting me with the experimental data prior to its publication.

[1] S. A. Pikin, Zh. Eksp. Teor. Fiz. 60, 1185 (1971) [Sov. Phys.-JETP 33, 641 (1971)].

[2] L. K. Vistin' and A. P. Kapustin, Optika i spektroskopiya 24, 650 (1968) [Optics and Spectroscopy 24, 348 (1968)].

[3] G. H. Heilmeier and W. Helfrich, Appl. Phys. Letters 16, 155 (1970).

[4] Orsay Liquid Crystal Group, Phys. Rev. Lett. 25, 1642 (1970).

[5] O. Paradi, Journal de Physique 31, 581 (1970).

[6] Giovanni Sansone, Ordinary Differential Equations (Russ. transl.), IIL, Vol. 1, 1953.

Translated by H. H. Nickle
223