STATIONARY FLOW OF A NEMATIC FLUID IN AN EXTERNAL MAGNETIC FIELD

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The time of onset of stationary motion in a layer of a nematic liquid crystal located between conducting planes is determined theoretically. Instability appears at a certain critical potential difference V_{cr}, which is independent of the distance between the planes. The stationary motion is periodic along a selected direction on the plane. The period d is proportional to the distance between the planes. The results are compared with experiment.

INTRODUCTION

ONE of the interesting problems of the physics of the liquid-crystal state is the behavior of the mesophases in external electric and magnetic fields. The phenomenon considered in the present paper was first observed by Williams[1]. It is manifest by a strip-like ("domain") structure of a layer of nematic liquid placed between plane-parallel plates to which a potential difference $V = \varphi_1 - \varphi_2$ is applied. The necessary conditions for the occurrence of this effect are a magnetic field **H** parallel to the plane of the capacitor, or polishing of the plates in a preferred direction (the x axis). Under these conditions, in the absence of an electric field **E**, the nematic mesophase becomes single-domain, the long axes of the molecules are oriented along the x axis: the "director" **n** is parallel to the x axis. The characteristic periodic picture of the black-white bands parallel to the y axis appears at a definite (critical) field E_{cr}.

The cause of this phenomenon apparently lies in the fact that the nematic liquid loses mechanical equilibrium in the electric field and macroscopic motion is produced in it. In considering this motion, which has a periodic character in the zx plane, it is necessary to take into account the volume forces that act on liquid containing extraneous charges, internal-friction forces due to the viscosity of the nematic liquid, and liquid-crystal elastic forces. The elastic forces in the nematic mesophase are connected with the inhomogeneous distribution of the director **n**. The distribution of the director **n** is in this case periodic in the zx plane and gives an observable optical picture of alternating dark and light strips along the y axis.

The nematic liquid may also be in mechanical equilibrium if the potential φ is not constant along the liquid. An important role in the stability of such an equilibrium is played by the anisotropy of the dielectric constant ϵ_{ik} and of the conductivity σ_{ik} in the nematic liquid[2]. Thus, in the case when the conductivity along the x axis is larger than along the y axis (this is precisely the situation in paraazoxyanisole), the equilibrium will be stable only if a definite condition is satisfied. If the potential difference $\varphi_1 - \varphi_2$ is not too large, then the regime of pure electric conductivity is maintained, with the potential a function of only the vertical coordinate z. On the other hand, if the difference $\varphi_1 - \varphi_2$ exceeds a certain critical value, then the space charges produced in the liquid interact with the external electric field and cause internal flow that tends to mix the liquid in such a way as to equalize in it the potential φ.

The instant of the onset of such a stationary motion, analogous to stationary convection[3], is determined theoretically in the present article. In accordance with the geometry of the experiment, we assume that the plates are polished along the x axis, the electric field is directed along the z axis perpendicular to the plates, and the resultant "domains" are parallel to the y axis. All the perturbations depend on the coordinates z and x. A solution of such a two-dimensional problem is given below.

1. EQUATIONS AND BOUNDARY CONDITIONS

We derive the equations describing the stationary flows in a nematic liquid. The liquid is assumed to be incompressible. This means that we neglect the change of the density ρ under the influence of a small change of pressure p along the liquid.

The Navier-Stokes equation in an anisotropic liquid dielectric containing extraneous charges with density ρ_{ext} is

$$\rho v_k \frac{\partial v_i}{\partial x_k} = f_i + \frac{\partial \sigma_{ki}'}{\partial x_k}, \quad (1)$$

where **v** is the velocity of the liquid and f_i is the volume force, equal to[4]

$$f_i = \frac{\partial}{\partial x_i}\left(-p + \frac{1}{8\pi}E_m E_k \rho \frac{\partial \varepsilon_{mk}}{\partial \rho}\right) - \frac{1}{8\pi}E_m E_k \frac{\partial \varepsilon_{mk}}{\partial x_i} + \rho_{ст} E_i, \quad (2)$$

σ'_{ki} is the tensor of the stresses due to the viscosity of the liquid crystal[5]:

$$\sigma_{ki}' = \alpha_1 n_k n_i A_{mj} n_m n_j + \alpha_2 n_k N_i + \alpha_3 n_i N_k + \alpha_4 A_{ki} + \alpha_5 n_k n_j A_{ji} + \alpha_6 n_i n_j A_{jk},$$

$$A_{mj} = \frac{1}{2}\left(\frac{\partial v_j}{\partial x_m} + \frac{\partial v_m}{\partial x_j}\right), \quad \mathbf{N} = \frac{d\mathbf{n}}{dt} + \frac{1}{2}[\mathbf{n}\,\mathrm{rot}\,\mathbf{v}]. \quad (3)^*$$

We write the alternating potential φ in the form $\varphi = -E_0 z + \psi$, $-l/2 \le z \le l/2$, assuming that $\psi \ll V = E_0 l$, where l is the thickness of the layer. We assume also that the deviations of the director **n** from the x axis are small: $n_z = \sin\theta \approx \theta$, $n_x = \cos\theta \approx 1$. Then, writing ρ_{ext} and ϵ_{ik} in the form

*$[\mathbf{n}\,\mathrm{rot}\,\mathbf{v}] = \mathbf{n} \times \mathrm{curl}\,\mathbf{v}$.

$$\rho_{ст} = \frac{1}{4\pi}\frac{\partial}{\partial x_i}(\varepsilon_{ik}E_k),$$
$$\varepsilon_{ik} = \varepsilon_\perp\delta_{ik} + (\varepsilon_\parallel - \varepsilon_\perp)n_in_k,$$

and omitting second-order terms, we obtain from (1)–(3)

$$\frac{\partial}{\partial z}\left(\beta_1\frac{\partial^2 v_x}{\partial x^2} + \beta_2\frac{\partial^2 v_x}{\partial z^2}\right)$$
$$= \frac{\partial}{\partial x}\left\{\beta_3\frac{\partial^2 v_z}{\partial x^2} + \beta_4\frac{\partial^2 v_z}{\partial z^2} + \frac{E_0}{4\pi}\left[E_0(\varepsilon_\parallel - \varepsilon_\perp)\frac{\partial\theta}{\partial x} - \varepsilon_\parallel\frac{\partial^2\psi}{\partial x^2} - \varepsilon_\perp\frac{\partial^2\psi}{\partial z^2}\right]\right\}, \quad (4)$$

where

$$\beta_1 = \alpha_1 + \alpha_5 + {}^1/_2(\alpha_3 + \alpha_4 + \alpha_6), \quad \beta_2 = {}^1/_2(\alpha_3 + \alpha_4 + \alpha_6),$$
$$\beta_3 = {}^1/_2(-\alpha_2 + \alpha_4 + \alpha_5), \quad \beta_4 = {}^1/_2(-\alpha_2 + \alpha_4 - \alpha_5).$$

We write the current density in the form

$$j_i = \sigma_{ik}E_k, \quad \sigma_{ik} = \sigma_\perp\delta_{ik} + (\sigma_\parallel - \sigma_\perp)n_in_k.$$

We have neglected here the diffusion current, which is proportional to l^{-2}, and is small if the layer thickness l is sufficiently large: $l > 3\times10^{-5}$ cm[2]. The layers of the nematic liquid under the experimental conditions have a thickness $\gtrsim 6\times10^{-4}$ cm.

From the equation div j = 0, which the spatial distribution of j in the constant field obeys, we obtain accurate to first-order terms

$$E_0(\sigma_\parallel - \sigma_\perp)\frac{\partial\theta}{\partial x} - \sigma_\parallel\frac{\partial^2\psi}{\partial x^2} - \sigma_\perp\frac{\partial^2\psi}{\partial z^2} = 0. \quad (5)$$

The equation of motion for the director **n** can be written in the form[5]:

$$J\frac{d\Omega}{dt} = [\mathbf{nh}] - \mathbf{\Gamma},$$

where J is the moment of inertia per unit volume, $\Omega = \mathbf{n}\times(d\mathbf{n}/dt)$ is the velocity of rotation, **h** is the molecular field, and **Γ** is the moment of the friction forces. The molecular field **h** is determined as a functional derivative $\mathbf{h}(\mathbf{r}) = -\delta\mathcal{F}/\delta\mathbf{n}(\mathbf{r})$. The free energy $\mathcal{F}$ of the nematic crystal in the continuous-medium model is equal to

$$\mathcal{F} = \frac{1}{2}\int\left\{K_{11}(\operatorname{div}\mathbf{n})^2 + K_{22}(\mathbf{n}\operatorname{rot}\mathbf{n})^2 + K_{33}[\mathbf{n}\operatorname{rot}\mathbf{n}]^2 - \frac{1}{4\pi}\varepsilon_{mk}E_mE_k\right\}d\mathbf{r},$$

where K_{ii} are the elastic constants. The moment of the friction forces in an incompressible liquid crystal is given by[5]

$$\mathbf{\Gamma} = [\mathbf{n}(\gamma_1\mathbf{N} + \gamma_2\hat{A}\mathbf{n})],$$

where the velocity **N** of the internal motion and the tensor $\hat{A} \equiv \{A_{ij}\}$ are defined in (3), and the constants γ_1 and γ_2 are connected with the Leslie constants α_i by the relations $\gamma_1 = \alpha_3 - \alpha_2$, $\gamma_2 = \alpha_6 - \alpha_5$. Substituting these expressions into the equation of motion, we obtain finally in the approximation under consideration

$$\frac{\varepsilon_\parallel - \varepsilon_\perp}{4\pi}E_0\left(E_0\theta - \frac{\partial\psi}{\partial x}\right) + K_{11}\frac{\partial^2\theta}{\partial z^2} + K_{33}\frac{\partial^2\theta}{\partial x^2}$$
$$= \frac{1}{2}(\gamma_1 + \gamma_2)\frac{\partial v_x}{\partial z} + \frac{1}{2}(\gamma_2 - \gamma_1)\frac{\partial v_z}{\partial x}. \quad (6)$$

Equations (4)–(6), together with the continuity equation div **v** = 0, constitute a complete system of equations describing the stationary flow in a nematic liquid. This system of four equations determines the unknown functions **v**, θ, and ψ. The boundary conditions on the solid surfaces are of the form

$$\psi = 0, \quad \mathbf{v} = 0, \quad \partial v_z/\partial z = 0, \quad \theta = 0. \quad (7)$$

The condition $\partial v_z/\partial z = 0$ follows from the continuity equation. The condition $\theta = 0$ follows from the formulation of the problem (experiment), namely, we assume that the surfaces are polished in the direction of the x axis.

2. CRITICAL FIELD

Eliminating the variables **v** and θ from the obtained equations and recognizing that in a nematic liquid (paraazoxyanisole) $\epsilon_\perp \approx \epsilon_\parallel$, $\alpha_1 \approx 0$, $\gamma_2 \approx -\gamma_1$[2], we obtain as a result for one variable ψ an equation of the type

$$\left(\frac{\partial^2}{\partial z^2} + a\frac{\partial^2}{\partial x^2}\right)\left(\frac{\partial^2}{\partial z^2} + b\frac{\partial^2}{\partial x^2}\right)\left(\frac{\partial^2}{\partial z^2} + c\frac{\partial^2}{\partial x^2}\right)\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial x^2}\right)\psi$$
$$= -\frac{\lambda}{l^2}\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial x^2}\right)\frac{\partial^4\psi}{\partial x^4}, \quad (8)$$
$$\lambda = \frac{1}{4\pi}\left(\varepsilon_\perp\frac{\sigma_\parallel}{\sigma_\perp} - \varepsilon_\parallel\right)\frac{\gamma_1}{\beta_2}\frac{V^2}{K_{11}}, \quad a = 1 - \frac{\alpha_2}{\beta_2}, \quad b = \frac{K_{33}}{K_{11}}, \quad c = \frac{\sigma_\parallel}{\sigma_\perp}.$$

The boundary conditions (7) are accordingly transformed into

$$\psi = 0, \quad \frac{\partial^2\psi}{\partial z^2} = 0, \quad \frac{\partial^4\psi}{\partial z^4} = 0, \quad \frac{\partial^5\psi}{\partial z^5} + (b+c)\frac{\partial^5\psi}{\partial x^2\partial z^3} + bc\frac{\partial^5\psi}{\partial x^4\partial z} = 0. \quad (9)$$

We seek ψ in the form $\exp(ikx)\zeta(z)$. The general solution of the equation obtained for $\zeta(z)$ is a linear combination of the functions $\cosh(2\mu_j z/l$ and $\sinh(2\mu_j z/l)$, where $\mu_1^2 = (kl)^2/4$; μ_2^2, μ_3^2, μ_4^2 are the solutions of the algebraic equation

$$(4\mu^2 - ak^2l^2)(4\mu^2 - bk^2l^2)(4\mu^2 - ck^2l^2) + \lambda(kl)^4 = 0. \quad (10)$$

The coefficients of this combination are determined by the boundary conditions (9), which lead to a system of algebraic equations, the condition for the compatibility of which determines the dependence of kl on λ. The inverse function $\lambda(kl)$ has a minimum at a certain value k_0l; the corresponding value $\lambda_0 = \lambda(k_0l)$ determines the criterion for the occurrence of instability, and the value of k_0 determines the periodicity along the x axis. The results of the corresponding calculations, analogous to those of[6], are presented below.

The coefficients a, b, and c are numbers of the order of unity (in paraazoxyanisole, $a \approx 3.8$, $b \approx 2.4$, and $c \approx 1.5$); the critical value of the parameter $\lambda/(kl)^2$ is of the order of 50. Therefore the solution of (10) can be written approximately in the form

$$\mu_2^2 \approx g\frac{(kl)^2}{4}(1-\nu), \quad \mu_3^2 \approx g\frac{(kl)^2}{4}\left(1 + \frac{1-i\sqrt{3}}{2}\nu\right),$$
$$\mu_4^2 \approx g\frac{(kl)^2}{4}\left(1 + \frac{1+i\sqrt{3}}{2}\nu\right), \quad (11)$$

where

$$g = (a+b+c)/3, \quad (g\nu)^3 = \lambda/(kl)^2.$$

Setting up an even and an odd (relative to z) combination of the functions $\cosh(2\mu_j z/l)$ and $\sinh(2\mu_j z/l)$, which satisfy the conditions (9) at $z = \pm l/2$, we find that when (11) is taken into account the compatibility condition is given respectively by the equations

$$-(\nu-1)^{3/2}K\left[1+\frac{b+c}{g(\nu-1)}+\frac{bc}{g^2(\nu-1)^2}\right]\operatorname{tg}\left(\frac{kl}{2}\sqrt{g(\nu-1)}\right)$$
$$=\frac{3}{g^{5/2}}(1-b-c+bc)\operatorname{th}\left(\frac{kl}{2}\right) \tag{12}$$

and

$$+\frac{(AI+BR)\operatorname{sh}(kl\sqrt{g}A)-(BI-AR)\sin(kl\sqrt{g}B)}{\operatorname{ch}(kl\sqrt{g}A)+\cos(kl\sqrt{g}B)};$$
$$(\nu-1)^{3/2}K\left[1+\frac{b+c}{g(\nu-1)}+\frac{bc}{g^2(\nu-1)^2}\right]\operatorname{ctg}\left(\frac{kl}{2}\sqrt{g(\nu-1)}\right)$$
$$=\frac{3}{g^{5/2}}(1-b-c+bc)\operatorname{cth}\left(\frac{kl}{2}\right)$$
$$+\frac{(AI+BR)\operatorname{sh}(kl\sqrt{g}A)+(BI-AR)\sin(kl\sqrt{g}B)}{\operatorname{ch}(kl\sqrt{g}A)-\cos(kl\sqrt{g}B)}. \tag{13}$$

Here A, B, I, K, and R are functions of g and ν:

$$A^2=\tfrac{1}{2}(1+\nu/2+\sqrt{1+\nu+\nu^2}),\qquad B^2=\tfrac{1}{2}(-1-\nu/2+\sqrt{1+\nu+\nu^2}),$$
$$K=1+\left(1-\frac{1}{g}\right)\frac{1}{\nu}+\left(1-\frac{2}{g}+\frac{1}{g^2}\right)\frac{1}{\nu^2},$$
$$I=\frac{1}{\nu^2}\Big[(\nu-1)(\nu^3-1)-\frac{1}{g}(2+\nu)(\nu-1)^2-\frac{1}{g^2}(2\nu^2+2\nu-1)$$
$$+\frac{b+c}{g}\left(\nu^3-1+\frac{2\nu^2-\nu+2}{g}+\frac{\nu-1}{g^2}\right)+\frac{bc}{g^2}\Big((\nu-1)^2$$
$$+2\frac{\nu-1}{g}+\frac{1}{g^2}\Big)\Big],$$
$$R=\frac{\sqrt{3}}{\nu^2}\Big[(\nu+1)(\nu^3-1)+\frac{1}{g}(\nu^3+3\nu+2)-\frac{1}{g^2}(2\nu+1)-\frac{b+c}{g}$$
$$\times\left(\nu^3-1+\frac{\nu+2}{g}-\frac{\nu+1}{g^2}\right)+\frac{bc}{g^2}\left(\nu^2-1+\frac{2}{g}-\frac{1}{g^2}\right)\Big].$$

Equations (12) and (13), with an arbitrary parameter g, are solved in general by numerical methods. However, when $(kl\sqrt{g}/2) \gtrsim 2.3$ (in paraazoxyanisole $g \approx 2.6$ and $k_0 l \sim 3$), we can obtain an approximate (error $\lesssim 3\%$) analytic expression for $\nu(kl)$, for in this case the solution of (12) is the value $\frac{1}{2}kl\sqrt{g(\nu-1)} \approx \pi/2$. From this we get

$$\lambda=(kl)^2(g\nu)^3\approx(kl)^2[g+(\pi/kl)^2]^3. \tag{14}$$

The solution of (13) is the value $kl\sqrt{g(\nu-1)} \approx 2\pi$, which corresponds to large λ, and consequently to large critical fields. We therefore do not consider the corresponding solutions below.

The function $\lambda(kl)$ has a minimum at $kl = k_0 l = \pi\sqrt{2/g}$:

$$\lambda_0=\lambda(k_0 l)\approx\frac{27}{4}\pi^2g^2. \tag{15}$$

From (8) and (15) we find that the critical potential difference at which periodic distribution of **n**, **v**, and φ along the x axis sets in, is equal to

$$V_{cr}\approx 3\pi g\sqrt{\frac{3\pi K_{11}\beta_2\sigma_\perp}{\gamma_1(\varepsilon_\perp\sigma_\parallel-\varepsilon_\parallel\sigma_\perp)}},\quad E_{cr}=\frac{V_{cr}}{l}. \tag{16}$$

The optical image of this distribution has a period

$$d=2\pi/k_0\approx 2l\sqrt{g/2}.$$

COMPARISON WITH EXPERIMENT

Substituting in (16) the values $K_{11} = 7 \times 10^{-7}$ dyne, $\beta_2 = 0.024$ g-cm^{-1} sec^{-1}, $\gamma_1 = 0.03$ g-cm^{-1} sec^{-1}, $\epsilon_\perp = 5.83$, $\epsilon_\parallel = 5.62$, $\sigma_\parallel/\sigma_\perp = 1.5$, and g = 2.6, which correspond to paraazoxyanisole, we find that the critical voltage is $V_{cr} \approx 9.4$ V. The period is equal to $d = 2l$. The experimental values of V_{cr} and d[7-9] are close to the calculated ones. It should be noted here that under the experimental conditions these values can vary by 30%, depending on the investigated sample[9]. In addition, in alternating electric fields the measured (effective) critical voltage is smaller than in constant ones by approximately a factor of $\sqrt{2}$[7,8].

The critical potential difference defined by formula (16) does not depend on the thickness of the layer of the nematic liquid, which also agrees with experiment. We note that the critical temperature difference at which convection sets in depends on the distance between the planes[6].

From Eqs. (5) and (6) and from the equations div **v** = 0 we see that if, for example, $\psi \sim \cos kx$, then $\theta \sim \sin kx$, $v_z \sim \cos kx$, $v_x \sim \sin kx$, i.e., the nematic liquid moves with maximum velocity (perpendicular to the planes of the capacitor) at those points of the x axis where the director **n** does not deviate from this axis. Such a distribution of the vectors **v** and **n** in the zx plane was observed experimentally in[9].

At sufficiently large values of $V(\lambda)$, the stationary flow of the nematic liquid described above can become unstable, just as in the case of convection. Turbulence occurs in paraazoxyanisole when $V - V_{cr} \approx 3$ V[9].

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