Behavior of nematic liquid crystals in a rotating magnetic field

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The effect of a rotating magnetic field on nematic liquid crystals is studied. At certain rotation frequencies an effect is observed which is analogous to electrohydrodynamic instability in a high-frequency electric field (a periodic distribution of orientations arises). The results are in agreement with the experimental ones.[1]

1. In the recent experiments of Prost and Canet[1] it was found that a periodic structure is established in the liquid crystal MBBA (metoxy-benzylidine-butyl-aniline) placed in a homogeneous magnetic field rotating with constant velocity. The rotation axis of the field coincides with the orientation of the director vector on the boundary of the liquid crystal. The present paper is devoted to an explanation of this phenomenon. To this end, a solution is obtained for the corresponding equations of the hydrodynamics of a nematic liquid crystal (NLC). It is shown that the onset of a periodic structure is analogous to the electrohydrodynamic effect in the high-frequency case[2] (the onset of convective instability in an alternating electric field perpendicular to the orientation of the NLC on the surface of a flat layer). Therefore, at a given value of the field H_0, there is an upper limit of the frequency ω_2, at which the appearance of the structure is observed (at high frequencies, the molecules cannot follow the variation of the field and there are no periodic solutions). Unlike the electrohydrodynamic effect, however, the ionic conductivity of the NLC, which are good electrolytes, does not give rise in this case to instabilities in the low-frequency case (in particular, in the static case). There is therefore a lower limit ω_1 of the field rotation frequency in a magnetic field.

The estimates obtained for MBBA are in good agreement with the experimental results[1]. In addition, the problem is solved (in cylindrical geometry) with the rotation axis perpendicular to the orientation of the director vector on the boundary of the NLC. A quasiperiodic radial structure is then produced (with a period that varies slowly with the distance from the center). The character of this structure depends essentially on the boundary condition at $r = 0$. For simplicity, we consider in detail the case when there is one disclination at the center. The existence of such a solution calls for a definite anisotropy of the elastic constants (see, for example,[3]). It is possible to investigate in similar fashion also the case with two disclinations or when the solution is not planar and nonsingular. At a suitable value of the parameters, the quasiperiod is such that the sample should produce strong Bragg scattering in the visible region of the spectrum.

The solutions of the two problems could actually overlap. However, in the view of the mathematical difficulties (nonlinear partial differential equations), we neglect the hydrodynamic flows in the solution of the problem with the rotation axis perpendicular to the initial orientation, and in the first part (rotation axis parallel to the orientation on the boundary) we do not take into account the change of the tangential velocity of rotation of the director over the NLC thickness. It is legitimate to discard the hydrodynamic flows by virtue of the presence in liquid crystals of the small parameter $\mu = K\rho/\eta^2$ (K is the modulus of elasticity, ρ is the density, and η is the average viscosity). A typical value for MBBA is $\mu \sim 10^{-3}$. As to the radial dependences, it is permissible to neglect them only when the threshold values are determined. Of course, all the results can be easily generalized to the inverse case, when there is a homogeneous time-invariant magnetic field and the NLC sample rotates. It should also be noted that the obtained formulas can be used to determine the magnetic and viscoelastic characteristics of NLC. Of particular importance here is the variation of the threshold quantities as functions of the sample thickness, and the presence of a critical thickness below which the effect is not observed at all.

2. We consider a cylindrical vessel of radius R filled with a NLC. In the absence of a field we have (z is the cylinder axis and n is the director)

$$n_z = 1, \quad n_x = n_y = 0.$$

Application of a magnetic field rotating in the xy plane

$$H_z = 0, \quad H_x = H_0 \cos \omega t, \quad H_y = H_0 \sin \omega t,$$

changes the orientation. We seek a solution in the most general form:

$$n_z = \cos\theta, \quad n_x = \sin\theta\cos\psi, \quad n_y = \sin\theta\sin\psi. \tag{1}$$

We note that, as will be shown below, an important role is played here by the non-planar character of the deviation from equilibrium orientation in the absence of a field. This distinguishes our problem immediately from the electrohydrodynamic effect, where allowance for the distortion in the yz plane leads to corrections of next order of smallness to the threshold quantities.

By virtue of the symmetry of the problem, it is convenient to use not rectangular but cylindrical coordinates r, φ, z. The system of electrodynamic equations in these coordinates is derived in the appendix. Taking (1) into account we have:

The incompressibility condition

$$\frac{\partial v_z}{\partial z} + \frac{1}{r} v_1 + \frac{\partial v_1}{\partial r} = 0, \tag{2}$$

v_i are the velocity components.

The Navier-Stokes equation

$$\rho\frac{\partial v_z}{\partial t} = -\frac{\partial P}{\partial z} + \beta_3\frac{\partial^2 v_1}{\partial z^2} + \beta_2\frac{1}{r}\frac{\partial^2 (v_z r)}{\partial r^2},$$

$$\rho\frac{\partial v_1}{\partial t} = -\frac{1}{r}\frac{\partial (Pr)}{\partial r} + \beta_1\frac{1}{r}\frac{\partial^2 (v_z r)}{\partial r^2} + \beta_2\frac{\partial^2 v_1}{\partial z^2} + \alpha_2\frac{\partial^2\theta}{\partial t\,\partial z}; \tag{3}$$

here P is the ordinary pressure, ρ is the density, and β_i and α_i are suitable combinations of the viscosity coefficients (see the Appendix); the equations (3) have been linearized with respect to θ ($\theta \ll 1$), a legitimate procedure when determining the threshold $\theta = 0$ of the stability of the solution (see below).

The equation of motion of the director:

$$\chi_a H_0^2 \theta \cos^2(\omega t - \psi) + K\left[\frac{\partial^2\theta}{\partial z^2} + \frac{\partial^2\theta}{\partial x^2}\right] = \gamma_1 \frac{\partial\theta}{\partial t} + \gamma_1 \frac{\partial v_x}{\partial z}, \tag{4}$$

χ_a is the anisotropy of the diamagnetic susceptibility and γ_1 is the viscosity coefficient. (According to (A.11) of the appendix we have $\gamma_1 = \alpha_2 - \alpha_3 \approx \alpha_2$.)

By virtue of the nonplanar character of the deviation, it is necessary to write also an equation for the azimuthal angle ψ. Neglecting the spatial derivatives of ψ, the corresponding equation can be separated from the system (2)–(4) and assumes a particularly simple form:

$$\tfrac{1}{2}\chi_a H_0^2 \sin 2(\psi - \omega t) = \gamma_1 \partial\psi / \partial t. \tag{5}$$

Equation (5) also takes into account the fact that usually in NLC the friction coefficients satisfy the condition $\gamma_1 + \gamma_2 = 0$. As to the neglect of the coordinate dependence of ψ, this is permissible if the boundary layer is small relative to the entire thickness of the NLC. The size of the wall layer for the orientation of the direction vector can be estimated at

$$h \sim (K / \chi_a H_0^2)^{1/2}. \tag{6}$$

Under the experimental conditions[1] the inequality $h \ll R$ is satisfied with a large margin. This condition is the opposite of the inequality that ensures linearization of the equations in (4). Whether such a procedure is valid depends on the parameters of the viscous wall layer

$$\chi_a H_0^2 / \eta\omega. \tag{7}$$

We therefore get from (6) and (7)

$$K / \chi_a R^2 < H_0^2 < \eta\omega / \chi_a. \tag{8}$$

Actually such a different behavior of ψ and θ is due to the fact that the corresponding equations contain different velocity components. The angle θ is determined by the normal velocity component, which varies little in the wall layer. We can therefore stipulate satisfaction of the boundary condition $v_1 = 0$ on the boundary of the main layer of the liquid crystal. The condition $\theta = 0$ is also satisfied with the same accuracy. On the other hand, the azimuthal angle ψ is connected with the tangential components of the velocity. They vary rapidly in the wall layer[2]:

$$v_t \sim \tilde{r}\tilde{v}_t / h$$

($\tilde{r}$ is the coordinate reckoned from the boundary and $\tilde{v}_t$ is the velocity on the boundary of the main layer). Therefore ψ behaves on the boundary of the main layer in free fashion, i.e., as if slippage were to be present.

The solution of (5) depends on the ratio of the field-rotation frequency to the parameter $\chi_a H_0^2/\alpha_2$. At $\omega < \chi_a H_0^2/2\alpha_2$ we have

$$\psi = \omega t + \text{const} \tag{9}$$

(the vector of the director moves together with the field). In the opposite case $\omega > \chi_a H_0^2/2\alpha_2$ we have

$$\psi = \omega' t; \tag{10}$$

$$\omega' = \omega - \frac{\pi}{\tau}\left[\int_0^\pi \frac{dx}{\omega\tau + \tfrac{1}{2}\sin 2x}\right]^{-1} \tag{11}$$

($\tau \equiv \alpha_2/\chi_a H_0^2$). We have left out from (10) the oscillating terms, which are negligible at large t.

The solution (10) corresponds to the presence of delay, i.e., the director vector rotates more slowly than the field. When (9) and (10) are substituted in (4), the entire dependence on the time drops out in the first case. We therefore have simply a static distortion of the orientation, which rotates stationarily about the z axis and is determined only by the value of the field H_0. On the other hand, in the presence of delay there are, in addition to the trivial solution, also solutions that depend on the time. Introducing the notation $\omega - \omega'$, we can easily see that system (2)–(4) is analogous to the equations obtained by Pikin[2] for the high-frequency electrohydrodynamic effect. It is necessary only to introduce in[2] the following change of notation:

$$v_x \to v_1, \quad v_z \to rv_z, \quad P = rP, \quad \theta \to r\theta.$$

Now we can use directly the solution method developed by Pikin [2]. The only difference lies in the different orders of magnitude of the frequencies that determine the threshold values. It is therefore necessary to take into account the elasticity from the very onset when solving the hydrodynamic equations. Eliminating the velocity and pressure from (2)–(4), we obtain one equation for $\partial\theta/\partial z \equiv T$:

$$\alpha_2^2 \frac{\partial}{\partial t}\frac{\partial^4 T}{\partial z^4} - \alpha_2 \frac{\partial}{\partial t}\left[\rho\frac{\partial}{\partial t}\left[\frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial z^2}\right] - (\beta_1+\beta_4)\frac{\partial^4}{\partial z^2 \partial r^2}\right.$$
$$\left. - \beta_2\frac{\partial^4}{\partial r^4} - \beta_3\frac{\partial^4}{\partial z^4}\right] T + \chi_a\left[\rho\frac{\partial}{\partial t}\left[\frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial z^2}\right] - (\beta_1+\beta_4)\frac{\partial^4}{\partial z^2\partial r^2}\right.$$
$$\left. - \beta_2\frac{\partial^4}{\partial r^4} - \beta_3\frac{\partial^4}{\partial z^4}\right]\left[H^2 T + K\left(\frac{\partial^2 T}{\partial z^2} + \frac{\partial^2 T}{\partial r^2}\right)\right] = 0,$$
$$H^2 \equiv H_0^2 \cos^2 \delta t. \tag{12}$$

The solution of (12) with the least distortion along the radius is

$$T = f(t) e^{iqz} \cos(pr), \tag{13}$$

where $p = \pi/R$. Substituting (13) in (12), we have for f(t) the equation

$$\frac{\partial^2 f}{\partial u^2} + [\lambda + \eta + \nu(1+\cos 2\pi u)]\frac{\partial f}{\partial u} + [\mu\nu(1+\cos 2\pi u)$$
$$- \mu\eta + 2\pi\nu \sin 2\pi u] f = 0. \tag{14}$$

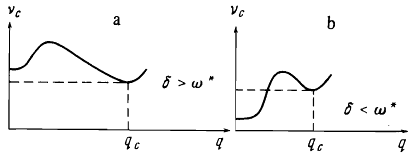
We have introduced here the notation

$$\nu = \frac{\pi}{2}\frac{\chi_a H_0^2}{\alpha_2\delta}, \quad \mu = \frac{\pi[(\beta_1+\beta_4)p^2q^2 + \beta_2 p^4 + \beta_3 q^4]}{\rho\delta(p^2+q^2)},$$
$$\lambda = \frac{\pi\alpha_2}{\rho\delta}\frac{q^4}{p^2+q^2} + \mu, \quad \eta = \frac{K(p^2+q^2)\pi}{\alpha_2\delta}, \quad u = \frac{\delta t}{\pi}. \tag{15}$$

In order for Eq. (14) to have periodic solutions corresponding to the experimentally observed regime[1], it is necessary that at least one root of the characteristic equation be equal to unity. If only the moduli of the roots are equal to unity (the roots are complex), then there are no periodic solutions. In the general case, only a numerical solution is possible. We make use of the fact that the problem has several characteristic parameters with the dimension of frequency

$$\omega_2 = \frac{\pi}{2}\frac{\chi_a H_0^2}{\alpha_2}, \quad \omega_0 = \frac{\beta p^2}{\rho}, \quad \tilde{\omega} = \frac{K p^2}{\beta}. \tag{16}$$

The frequency ω_2 does not depend on the dimensions of the sample and under the conditions of the experiment in [1] we have $\omega_2 \sim 1\ \text{sec}^{-1}$. On the other hand the frequencies ω_0 and $\tilde{\omega}$ (the characteristic relaxation time of the structure) are determined in the main by the dimensions. Therefore, depending on the value R, dif-



ferent cases are possible. If $\omega_2 > \omega_0$, then the solution of (14) corresponding to the periodic time dependence is determined by the condition $\nu \geq \nu_c$. The critical value of the parameter ν_c in the frequency region $\delta > \omega_0$ is obtained numerically in analogy with[2]. In order of magnitude, (with accuracy 10%), $\nu_c = 1$.

Thus, the condition for the presence of the periodic solution is $\delta \leq \omega_2$. On the other hand, in the region $\delta < \omega_0$ we need another solution method, since the series in powers of ω_0/δ diverges. We can qualitatively assume the coefficients λ and μ to be sufficiently large and use a quasiclassical method[2]. Actually, there is no need for a strong inequality $\lambda, \mu \gg 1$, since the series in powers of $1/\lambda$ converges and even the first correction term $\sim 1/4\pi^2\lambda^2$ is small. In the region $\delta < \omega_0$ we have

$$\nu_c = \frac{1}{2}\lambda - \frac{2}{5}[\lambda(\lambda - \mu + \eta)]^{1/2} + \frac{\pi^2}{16}\frac{1}{\lambda - \mu + \eta}\ldots \quad (17)$$

ω_0 becomes larger than ω_2 at sufficiently small radii R. We then have from (17) $\nu_c > 1$, and ν is always smaller than ν_c, so that there are no periodic solutions at all. When $\delta > \omega_2$, there are certainly no solutions of type (13).

Thus, there are limits for the existence of periodic solutions. The lower limit ω_1, corresponding to the delay condition (10), is approximately one-third as large as ω_2. This is observed with good accuracy under the conditions of[1]. In addition, the critical value of ν_c determined from (17) depends on q (see Fig. a). Owing to the presence of the elasticity η, the quantity ν_c has two minima at $q = 0$ and $q = q_c$ (the period of the structure). With decreasing frequency, the minimum at q_c rises, and at a certain ω^* the state with $q = 0$ can become more favored energywise (see Fig. b). The quantity ω^* is determined from (17):

$$\omega^* \approx \beta^{-1/2}\rho^{-1/4}K^{3/4}p^2. \quad (18)$$

The lower limit is determined by the larger of the quantities ω^* and ω_2/π.

We note in concluding this section that physically the analogy with the electrohydrodynamic effect becomes more obvious if we consider a rectangular vessel (with orientation parallel to the long axis) and a field whose direction changes jumpwise by 90° (rectangular pulses). In this case the equations for the different components of the vector of the director can be separated and are analogous[2].

3. We now consider the case when the orientation of the NLC on the boundary is tangential to the surface of the cylinder. As already noted in Sec. 1, we can neglect the hydrodynamic motion of the liquid crystal. We are left then only with the equation of motion of the director vector. We can therefore calculate now the radial dependence of the azimuthal angle. We assume for simplicity that the director vector is located at all times in a plane perpendicular to the cylinder axis (for this purpose it is necessary that the elastic constant corresponding to the rotation (K_{22}) be the smallest. We then have only one equation for ψ:

$$\tau\frac{\partial\psi}{\partial t} = \xi^2\left(\frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\partial^2\psi}{\partial r^2}\right) - \frac{1}{2}\sin 2(\psi - \omega t). \quad (19)$$

Here $\xi = (K/\chi_a H_0^2)^{1/2}$ is the characteristic magnetic length. Were it not for the spatial derivatives in (19), we would have solutions (9) and (10). However, the boundary condition $\psi(R) = \varphi$ gives rise to a radial dependence. We note that in spite of the explicit dependence of the boundary condition on the polar angle φ, neglect of the azimuthal dependence in (19) is legitimate. The reason is that in our approximation we deal only with the quantity $\psi - \varphi$.

Significant deviations from the homogeneous case appear only when $\omega\tau < 1/2$ (since $\tau = \pi/\omega$, the analysis of Sec. 2 is justified). In this case, if

$$\partial^2\psi/\partial r^2 \gg r^{-1}\partial\psi/\partial r,$$

then (19) coincides with the equation considered by de Gennes[4] for the planar problem. The solution can be sought in the form

$$\psi = \omega t + \Phi(r - vt). \quad (20)$$

This corresponds to the presence of a radial structure with a period

$$L = \pi v/\omega. \quad (21)$$

The quantity L (or v) can be determined from energy considerations by equating the energy drawn from the magnetic field to the energy dissipated when the structure (20) moves.

An important role, however, is played by the behavior at $r = 0$. If there is only one disclination at the center, then, putting

$$\partial\psi/\partial r|_{r=0} = 0,$$

we obtain in analogy with[4]

$$L = \xi \ln(\xi\omega_2/r\omega\pi), \text{ if } \pi\omega/\omega_2 < \xi/R, \quad (22)$$

$$L = \xi^2\omega_2/r\omega\pi, \text{ if } \pi\omega/\omega_2 > \xi/R. \quad (23)$$

If (22) is satisfied, then $L \gg \xi$, and in the case (23) we have $L < \xi$. At typical values of the parameters we have $L \sim 1\,\mu$. In the opposite limiting case

$$r^{-1}\partial\psi/\partial r \gg \partial^2\psi/\partial r^2$$

we obtain a first-order equation that has no quasiperiodic solutions. This however, is precisely the case realized when $L \gg \xi$. Therefore, unlike the planar problem[4] where both regimes (22) and (23) can exist, we have here a single condition for the quasiperiodic radial solutions

$$\pi\xi/R < \omega/\omega_2 < 1/\pi. \quad (24)$$

In analogy with the foregoing, this can be regarded as a limitation on the value of the magnetic field H_0. At sufficiently small sample dimensions, $R < \pi^2\xi$, the condition (24) cannot be satisfied and no radial structure should be observed.

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APPENDIX

To derive the hydrodynamic equations in a cylindrical coordinate system, it is necessary to have the metric tensor

$$g_{ij} = \frac{\partial x^\alpha}{\partial y^i}\frac{\partial x^\alpha}{\partial y^j}.$$

It has the following nonzero components:

$$g^{11}=g_{11}=1,\quad g_{22}=1/g^{22}=r^2,\quad g^{33}=g_{33}=1; \tag{A.1}$$

while the nonzero Christofel symbols are

$$\Gamma_{12}{}^2=r^{-1},\quad \Gamma_{22}{}^1=-r,\quad \Gamma_{21}{}^2=r^{-1}; \tag{A.2}$$

The components of the director in terms of the new coordinates are

$$n_3=\cos\theta,\quad n_2=r\sin\theta\sin(\psi-\varphi),\quad n_1=\sin\theta\cos(\psi-\varphi). \tag{A.3}$$

Here θ is the angular deviation of the director vector from the tangent to the cylinder, and ψ is the azimuthal angle. In analogy, the components of the magnetic field are

$$H_1=H_0\cos(\omega t-\varphi),\quad H_2=H_0r\sin(\omega t-\varphi),\quad H_3=0. \tag{A.4}$$

Differentiation is now carried out with the aid of (A.2), for example:

$$v_{i,j}=\partial v_i/\partial y_j-\Gamma_{ij}{}^k v_k. \tag{A.5}$$

To obtain the Navier-Stokes equations, it is necessary to calculate the corresponding forces

$$f_i=-P_{,i}+\sigma'_{ki,k}, \tag{A.6}$$

$$\sigma_{ki}'=\alpha_1 n_k n_i A_{mj} n_m n_j+\alpha_2 n_k N_i+\alpha_3 n_i N_k + \alpha_4 A_{ki}+\alpha_5 n_k n_j A_{ji}+\alpha_6 n_i n_j A_{jk}, \tag{A.7}$$

$$N_i=\partial n_i/\partial t+v_j g^{jj} n_{i,j}+g^{kk}\omega_{ki}n_k, \tag{A.8}$$

$$\omega={}^1/_2\operatorname{rot}\mathbf{v},\quad A_{ij}={}^1/_2(v_{i,j}+v_{j,i}). \tag{A.9}$$

After substituting (A.8)—(A.10) in (A.6), we obtain Eqs. (2) and (3) of the text, where account is taken of the fact that $\alpha_1=\alpha_3=0$ (as is usual in NLC), and where we put

$$\beta_1=\alpha_5+{}^1/_2(\alpha_4+\alpha_6),\quad \beta_2={}^1/_2(\alpha_4+\alpha_6),$$
$$\beta_3={}^1/_2(-\alpha_2+\alpha_4+\alpha_5),\quad \beta_4={}^1/_2(-\alpha_2+\alpha_4-\alpha_5). \tag{A.10}$$

Analogously, we obtain the equation of motion of the director vector. It is necessary also to use the definition of the friction moment:

$$M_i=g^{1/2}g^{jj}g^{kk}e_{ijk}\sigma_{jk}', \tag{A.11}$$

g is the determinant of the metric tensor and e_{ijk} is a unit fully antisymmetrical tensor.

Equations (4) and (5) are then obtained by equating (A.1) to the moment $\mathbf{n}\times\mathbf{h}$ exerted by the surrounding molecules, where $\mathbf{h}$ is the molecular field

$$\mathbf{h}=K\Delta\mathbf{n}-\chi_a(\mathbf{Hn})\mathbf{H} \tag{A.12}$$

(for a more detailed derivation of the hydrodynamic equations see, e.g.,[2]).

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37