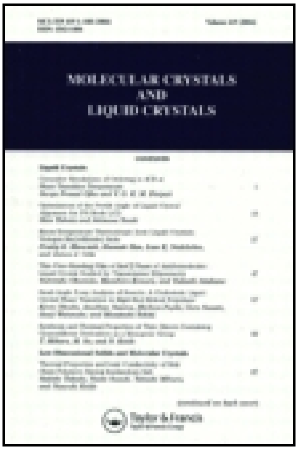

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Surface Waves in Discotic Liquid Crystals

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Surface Waves in Discotic Liquid Crystals

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The surface waves in discotic liquid crystals are studied in the geometry in which the column axes are perpendicular to the free surface of a sample. It is shown that in the low frequency limit, when permeation is essential, only the diffusion mode exists. In the opposite case, besides the diffusion mode, there exists also the propagating elastic mode.

The dynamics of systems with broken translation symmetry have attracted the attention of many investigators. Typical examples of three-dimensional systems with one-dimensional ordering are smectic–A liquid crystals. Phenomena such as sound propagation,[1,2] surface wave propagation[3] and other hydrodynamic properties have by now been sufficiently well described.

The discovery by Chandrasekhar, Sadashiva, and Suresh[4] of discotic liquid crystals produced an example of the real existence of a three-dimensional system with two-dimensional ordering. Later, a series of materials possessing similar properties was discovered and the number of such systems can be expected to increase. Accordingly, interest in the study of the dynamical properties of the discotic plane has grown. The hydrodynamic equations of discotics were obtained and used to study sound propagation,[5,6] light and X-ray scattering[7,8] and stability of the discotic cylinders.[9]

The present work is devoted to the study of the surface waves in incompressible discotic liquid crystals. Such waves can be produced by varying the pressure at the free surface or by thermal fluctuations which cause small displacements of the molecules of the surface layer. Not only is the study of such waves interesting in itself, it also provides the possibility of measuring a series of the characteristic parameters experimentally.

Although the known discotic liquid crystals exhibit a rich polymorphism, we will restrict ourselves to the case of uniaxial discotics whose structure consists of columns forming the hexagonal lattice in a plane perpendicular to their axis. There is no ordering of the molecular structure along the columns. Although this is the simplest type of discotic, we trust that our results will serve to provide at least a qualitatively accurate description. Moreover, we will study only the geometry in which the direction of the column axis is perpendicular to the free surface. The presence of the two-dimensional lattice of columns makes it necessary to introduce two extra hydrodynamic variables: these are the components of a two-dimensional vector $\mathbf{u}(x, y, z)$, which describes the displacement of the column axis from the equilibrium position. The hydrodynamic equations for this case were obtained by Prost and Clark[5] and have the form

$$\begin{aligned} v_{i,i} &= 0 \\ \rho \dot{v}_i &= -P_{,i} + \Phi^i_{j,j} + \sigma_{ij,j} \\ \dot{u}_\beta - v_\beta &= \nu \Phi^\beta_{j,j} \\ i, j &= x, y, z; \qquad \beta = x, y \end{aligned} \tag{1}$$

where v_i—velocity components, u_β—column displacement variables, p—pressure, ρ—density, σ_{ij}—viscosity tensor for a uniaxial crystal[1]

$$\begin{aligned} \sigma_{ij} &= 2\eta_2 v_{ij} + 2(\eta - \eta_2)(v_{iz}\delta_{jz} + v_{jz}\delta_{iz}) + \eta_1 v_{zz}\delta_{iz}\delta_{jz}, \\ v_{ij} &= \tfrac{1}{2}(v_{i,j} + v_{j,i}) \end{aligned} \tag{2}$$

$\Phi^i_j = \delta(\epsilon)/\delta(u_{i,j})$—the thermodynamic forces conjugated to the column displacement gradients; ϵ is the energy density of the discotic liquid crystal. Depending on the definition of $\mathbf{u}$, $\Phi^z_j = 0$.

The first equation in (1) represents the discontinuity equation for the incompressible discotic; the second represents the Navier-Stokes equations; the third describes the permeation process[10] corresponding to the molecules hopping from one column to the other and ν is the permeation coefficient.

The energy density of the discotic has the form

$$\epsilon - \epsilon_0 = \frac{B}{2}(u_{x,x} + u_{y,y})^2 + \frac{D}{2}\left[(u_{x,x} - u_{y,y})^2 + (u_{x,y} + u_{y,x})^2\right]$$
$$+ \frac{K}{2}\left[\left(\frac{\partial^2 u_x}{\partial z^2}\right)^2 + \left(\frac{\partial^2 u_y}{\partial z^2}\right)^2\right]. \tag{3}$$

Here ϵ_0—equilibrium energy density, B, D—elasticity constants, K—Frank modulus corresponding to the bend of the columns. By inserting (2) and (3) into (1) we obtain the system of hydrodynamic equations which describe the discotic.

We will look for the solution to this problem in the form of the surface wave propagating in the x direction

$$\left.\begin{matrix} \mathbf{v}(x, y, z, t) \\ \mathbf{u}(x, y, z, t) \\ P(x, y, z, t) \end{matrix}\right\} = \sum_{\mathbf{q}} \begin{pmatrix} \mathbf{v}_{\mathbf{q}} \\ \mathbf{u}_{\mathbf{q}} \\ P_{\mathbf{q}} \end{pmatrix} \exp(iqx + lqz + i\omega t) \tag{4}$$

and damping deep into the sample ($\mathbf{v}_{\mathbf{q}}, \mathbf{u}_{\mathbf{q}}, P \to 0$ at $z \to -\infty$; $z = 0$ correspond to the free surface).

The equations (1) take, in this case, the following form

$$i\omega u_x - v_x = -\nu A q^2(1 + \lambda^2 q^2 l^4) u_x,$$
$$i\omega\rho v_x = -iqP - Aq^2(1 + \lambda^2 q^2 l^4) u_x + \eta q^2\left(l^2 + 1 - \frac{2\eta_2}{\eta}\right) v_x,$$
$$i\omega\rho v_z = -qlP - \eta q^2\left[1 - l^2\left(3 - \frac{2\eta_2}{\eta} + \frac{\eta_1}{\eta}\right)\right] v_z, \tag{5}$$
$$iv_x + lv_z = 0, \qquad A = B + D, \qquad \lambda = (K/A)^{1/2}.$$

The value l must be found from the solvability condition of this system and must satisfy the wave damping condition when $z \to -\infty$ (that is, the real part of l has to be positive). The permeation effects described by the first of the equations (5) are essential when the column structure is firmly fixed, owing to the existence of boundary pinning effects, or in the case of sufficiently small frequencies. Since we are considering the case of the half-space-sample, i.e. not taking into account the wall influence, the only possible situation in which the permeation effects are essential is the case of low frequencies that mathematically correspond to the condition

$$|\omega| \ll |\nu A q^2(1 + \lambda^2 q^2 l^4).$$

Let us first consider the opposite case, where the permeation effects play no role, i.e., when the first equation (5) has the form

$$i\omega u_x = v_x.$$

The solvability condition of system (5) gives the following equation for the value l

$$-\omega^2\rho(l^2 - 1) + i\omega q^2\left[\eta_1 l^2 - \eta(l^2 - 1)^2\right] + q^2 l^2 A(1 + \lambda^2 q^2 l^4) = 0. \tag{6}$$

The solution of equation (6) in an analytical form with dependence on all parameters of the problem is rather difficult, so we will study two limiting cases corresponding to the different ratios of the values contained in the last bracket.

If $l^4 \ll (\lambda q)^{-2}$ equation (6) transforms to a biquadratic with respect to l

$$l^4 - l^2\left(2 + \frac{\eta_1}{\eta} + \frac{i\omega\rho}{\eta q^2} - \frac{iA}{\eta\omega}\right) + \frac{i\omega\rho}{\eta q^2} + 1 = 0. \tag{7a}$$

If $l^4 \gg (\lambda q)^{-2}$ (taking into account the fact that $(\lambda q)^2 \sim 1$ as $q \sim 10^8$ cm^{-1} and $(\lambda q)^2 \ll 1$ for all smaller q), the value corresponding to this case is large and one can neglect the terms of the order of 1 in (6). It then takes the simple form

$$l^2\left[l^4 - \frac{i\omega\eta l^2}{Kq^2} + \frac{i\omega q^2(\eta_1 + 2\eta) - \omega^2\rho}{Kq^4}\right] = 0. \tag{7b}$$

The solution $l = 0$ has no physical meaning. Therefore the equation (7b) as (7a) gives four roots from which we select only those that satisfy the damping condition, i.e. have a positive real part. To obtain the dispersion relations for ω we use the boundary conditions which in our case are the following

$$\left.\begin{aligned} P - \sigma_{zz} &= -\sigma\frac{\partial^2\xi}{\partial x^2} \\ \sigma_{xz} - \Phi_z^x &= 0 \end{aligned}\right|_{z=0} \tag{8}$$

where the value $\xi(x, t)$ corresponds to the coordinate z of the free surface and satisfies the condition $\xi = v_z/i\omega|_{z=0}$; σ—surface tension coefficient.

Taking into account the properties of the equations (7a), (7b), we will look for the solution in the form

$$v_{qz} = c_1\exp(l_1 qz) + c_2\exp(l_2 qz) \tag{9}$$

where l_1 and l_2 are two solutions of the equations (7a) or (7b).

Putting (1), (2) and (9) into (8) and using the solvability condition of this system with respect to the coefficients c_1 and c_2, we obtain the dispersion equations for (7a) and (7b) correspondingly

$$\begin{aligned}(l_2 - l_1)\Big\{ & n + \frac{2\eta_1}{\eta} + m(1+n)\left(\frac{\eta_1}{\eta} - 3 + p - n\right) + l_1 l_2 \\ & \times\Big[4 - \frac{\eta_1}{\eta} - p + n + m(1+n) \\ & + m\left(\frac{\eta_1}{\eta} + p - n - 3\right)\left(2 + \frac{\eta_1}{\eta} + n - p\right)\Big]\Big\} \\ = & \frac{i\sigma q}{\eta\omega}\left[1 - m\left(2 + \frac{\eta_1}{\eta} + n - p\right)\right](l_2^2 - l_1^2),\end{aligned} \tag{10a}$$

$$\begin{aligned}(l_2 - l_1)\Big\{ & n + \frac{2\eta_1}{\eta} + 4 + m(1+n)\left(\frac{\eta_1}{\eta} - 1 + p - n\right) + l_1 l_2 \\ & \times\Big[2 - \frac{\eta_1}{\eta} - p + n + m\left(\frac{\eta_1}{\eta} - 1 + p - n\right) \\ & \times\left(n - p + 4 + \frac{\eta_1}{\eta}\right)\Big]\Big\} \\ = & \frac{i\sigma q}{\eta\omega}\left[1 - m\left(n - p + 4 + \frac{\eta_1}{\eta}\right)\right](l_2^2 - l_1^2).\end{aligned} \tag{10b}$$

Here we introduce for the sake of simplicity the following notations:

$$m = \frac{iKq^2}{\eta\omega}, \qquad n = \frac{i\omega\rho}{\eta q^2}, \qquad p = \frac{iA}{\eta\omega}.$$

The values l_1 and l_2 also contain the dependence of the frequency ω and all characteristic parameters. The obvious solution $l_1 = l_2$ has no meaning, because $c_1 = -c_2$ in this case and v_x, v_z are identically equal to zero.

Now let us consider the case of the low frequencies where permeation is essential. In this case the first of the equations (5) has the form

$v_x = \nu A q^2(1 + \lambda^2 q^2 l^4) u_x$ and it leads to

$$l^4 - l^2\left(2 + \frac{1}{\nu\eta q^2} + \frac{\eta_1}{\eta} + \frac{i\omega\rho}{\eta q^2}\right) + 1 + \frac{i\omega\rho}{\eta q^2} = 0 \tag{11}$$

By again writing the solution in form (9), using (1), (2) and (8) and taking into account the fact that $\nu\eta q^2 \ll 1$ for all $q < 10^7$ cm^{-1}, we get

$$\begin{aligned} n &+ \frac{2\eta_1}{\eta} + \lambda^4 q^4 s^2(r - n) + \lambda^2 q^2(r^2 + 2rs - 3rs^2 - s^3) \\ &- l_1 l_2\left[r + s + \lambda^4 q^4 s^2(s + 2r) + \lambda^2 q^2(s^3 + 4s^2 r + 5sr^2 + 2r^3)\right] \\ &= -\frac{i\sigma q}{\eta\omega}\left[1 + \lambda^4 q^4 s^2 + \lambda^2 q^2(s^2 + 3sr + 2r^2)\right](l_1 + l_2) \end{aligned} \tag{12}$$

where two extra notations are introduced: $r = (\nu\eta q^2)^{-1}$ and $s = 1 + n$. As the dispersion equations (10a), (10b) and (12) are very complicated and cannot be solved explicitly, we restrict ourselves to the consideration of limiting cases. Such an analysis is rather simple, but it is bulky, so we give final results only, omitting the intermediate calculations. Moreover, we describe only those modes which appear most realistic.

For those discotics known at the present time, the characteristic parameters are: $A \sim 10^8$(dyn/cm^2), $K \sim 10^{-7}$ dyn, $\sigma \sim 10^2$(dyn/cm), $\eta \sim 1 \div 10$(dyn · sec/cm^2).

The permeation coefficient ν for our estimations we take to be the same as in smectic A liquid crystals, i.e. $\nu \sim 10^{-14} \div 10^{-15}$(sec · cm^3/g). In the region of low frequencies, small wave vectors and rather small viscosities, when permeation effects are essential, only the diffusion mode

$$\omega_p \simeq iq^2(\nu\sigma^2/\rho)^{1/3}$$

exists at $q \lesssim \lambda^{-1/3}(\rho\sigma\nu^2)^{-2/9}$ and when the condition on the parameters $(\sigma^2/\rho\nu^2)^{1/3} \ll A$ is fulfilled.

In the case of larger viscosities, this mode changes and has the form

$$\omega_p \simeq iq^2\sigma(\nu/\eta)^{1/2}$$

with the additional conditions on the parameters

$$\frac{\sigma}{A} \ll (\nu\eta)^{1/2} \ll \frac{\sigma\rho}{\eta^2}.$$

From the above it is clear that the existence conditions of the permeation mode are fulfilled better for larger values of A, that is, when the column lattice is more rigid. This seems natural, because the rigid lattice effectively represents the "fixed structure" and the mechanism of the permeation is necessary to produce the flow.

In the case of high frequencies, when the permeation is negligible, we have found the following modes:

1) diffusion mode

$$\omega_D \simeq iq\sigma/\eta$$

which exists at $\sigma\rho/\eta^2 < q < \lambda^{-1}$;

2) propagating mode

$$\omega_{pr} \simeq \pm\frac{(KA)^{1/2}}{\eta}q$$

which exists at $\rho(KA)^{1/2}/\eta^2 < q < \lambda^{-1}, \lambda/\nu\eta$;

3) capillary mode

$$\omega_c \simeq \pm(\sigma/\rho)^{1/2}q^{3/2}$$

which exists at $A/\sigma < q < \sigma\rho/\eta^2$.

The existence conditions of the capillary mode are not likely to be realized in traditional experimental measurements. It is, however, readily conceivable that the capillary mode can manifest itself in the region of temperatures close to the temperature of the discotic–nematic phase transition, because the elastic constants B and D (and consequently A) tend to zero as $T \to T_c$ and other characteristic parameters do not have sharp temperature dependencies. As regards other modes of the surface waves which have been found, both their form and their existence conditions seem quite natural, and, at least qualitatively, give an accurate description of the physical picture of the phenomena.

At the phase transition point, the existence conditions of the permeation mode ω_p deteriorate. This is also quite straightforward, because the decrease in value of A makes the system "softer" and thus more similar to the nematic phase or normal fluid. The propagating mode ω_{pr} connected with the existence of the column lattice, also vanishes, i.e. the system "stops to remember" the lattice structure.

The calculations performed also enable us to write down the expressions for some intermediate cases. However, these results do not qualitatively change the picture obtained and are therefore of no immediate interest. It seems more useful to consider two other geometries: the cases where the columns are parallel to the free surface. It can be assumed that in the case in which the propagation direction of the surface wave coincides with the column axis, results would be obtained similar to those for smectic A. The case of the wave propagation perpendicular to the column axis may, however, prove to be of interest. The corresponding results will be published elsewhere.

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