New type of orbital waves in nematic liquid crystals

V. L. Golo

Mechanomathematical Faculty, M. V. Lomonosov State University, Moscow

E. I. Kats

L. D. Landau Institute of Theoretical Physics, Academy of Sciences of the USSR, Chernogolovka, Moscow Province

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A study is made of the spectrum of normal modes of nematic liquid crystals allowing for the inertia of the motion of the director. A state of a liquid crystal with a rotating director is considered and the equations of motion are averaged for such rotation. This gives a closed system of equations which contains only slow variables. The mode spectrum of this average system is investigated. In particular, an orbital wave with a dispersion law $\omega \propto q^2$ is found; it is analogous to magnon modes in a ferromagnet with the easy-axis anisotropy. The feasibility of experimental observation of these waves in experiments on inelastic scattering of light is discussed.

I. INTRODUCTION

Lyotropic liquid crystals (particularly those of biological origin) are attracting increasing attention (see, for example, the review by Vedenov and Levchenko¹). A characteristic feature of such systems is that fairly large molecular complexes of different shapes play the role of individual molecules. In the case of such large complexes (containing up to 10⁴ separate molecules) it may be wrong to ignore the inertia or delay in reorientation (in contrast to ordinary thermotropic liquid crystals, for which the instant-response approximation may be justified). The dynamics of such liquid crystals with an inertia of the director has never been investigated (to the best of our knowledge), although some special cases have been tackled.² As a rule, in problems of this kind the motion of the director has been restricted to some special geometry and no allowance has been made for the interaction with other hydrodynamic degrees of freedom. For example, Ericksen³ considered twist waves in a nematic liquid crystal. The dynamics of nematic liquid crystals with an inertia is not a trivial problem and the additional normal modes which appear because of an allowance for the inertia do not simply reduce to twist waves. The attention to this point was first drawn by the present authors and Leman.⁴ We considered only the spatially homogeneous case and we demonstrated that two types of dynamic behavioreither unstable and random or quasistationary in a specific long-lived mode-are possible, depending on the external conditions and initial data.

An allowance for the inhomogeneities, i.e., for the gradients of hydrodynamic variables, complicates greatly the problem. A full analysis of nonlinear equations of hydrodynamics of liquid crystals with an inertia is difficult even on a computer. On the other hand, nonlinearities are usually important in the dynamics in those cases when strongly fluctuating modes are encountered in the linear problem [this is true, for example, of smectic liquid crystals discussed by one of us (EIK) and Lebedev⁵]. The presence of strongly fluctuating modes means that the problem includes "dangerous" nonlinear interactions, which are not in the hydrodynamic limit of wavelengths and frequencies. Such interactions do not occur in nematic liqud crystals and, therefore, a weak inhomogeneity can be allowed for by considering only the linearized equations of motion. Even in such a linear rise the system of equations for the hydrodynamics of liquid crystals with an inertia of the director rotation is still too complex and cumbersome to be tackled analytically.

We shall now obtain some estimates before going over to further simplifications. We are interested mainly in new effects associated with the existence of a moment of inertia I(per unit volume) of a liquid crystal. The quantity I is governed by the dimensions of the complexes forming a liquid crystal and their density. The inertial effects in the equations of motion $(I\omega^2)$ should be at least comparable with the dissipation proportional to $\eta \omega$ (ω is the frequency and η is a certain characteristic viscosity). Liquid crystals are unlikely to have very low values of the viscosity η . Usually these values are $\eta \approx 9.1$ P (and sometimes even larger). In special cases (solutions etc.) we can expect smaller values in the range $\eta \sim 10^{-2} - 10^{-3}$ P. Therefore, even when the moment of inertia amounting to $I \sim 10^{-7} - 10^{-8}$ g/cm is of giant magnitude from the molecular point of view, the inertial effects should at best begin from frequencies $\omega \sim \eta/I \sim 10^5 - 10^7$ \sec^{-1} . These restrictions are of very general validity. This is due to the fact that any hydrodynamic equations are expanded in terms of small freugencies and wave vectors. Therefore, an allowance for higher derivatives in the equations (in our case, $I\omega^2$) is permissible only in the case of particularly small coefficients of the lower powers of the frequency or wave vector (in our case, this applied to $\eta\omega$). There may be several such specially small parameters in the problem of a liquid crystal: they may be the anisotropy of the molecular shape, low concentration of anisotropic particles, or proximity to a transition to the isotropic phase (I is independent of such)proximity and the rotational viscosity tends to zero at the transition point).

We can therefore observe inertial effects (for example, on the basis of the inelastic scattering of light) by investigating a liquid crystal in a state with a sufficiently fast rotation of the director. The inertia of the director should appear against the background of this state. If we are interested in the effects which are slow compared with the director rotation frequency, then the equations of dynamics can be averaged over this rotation. The resultant average system of equations is used below to study the normal modes, fluctuations, etc. against the background of a rotating director of a liquid crystal. Such a state of a liquid crystal described by averages is in fact a new object which differs from the initial uniaxial liquid crystal.

We shall assume that in this state a liquid crystal is nevertheless close to thermodynamic equilibrium in the sense that all the suprathermal noise induced by the director rotation is less than the thermal effects associated with averaging of the thermodynamic characteristics over rotation. A rigorous investigation of the validity of this hypothesis will be provided in a separate communication. Clearly, this new object (or, more exactly, the new state of a liquid crystal) does exist as long as dissipation does not reduce the rotation velocity so much that the inertial effects become unimportant and the averaging procedure becomes incorrect. For time intervals of this kind we can expect the usual viscous relaxation of the director motion. Therefore, the effects discussed below occur during the lifetime of a rotating liquid crystal, which is $\sim 10^{-4}$ - 10^{-3} sec (see also Ref. 4).

II. DERIVATION OF AVERAGE EQUATIONS

We shall be concerned solely with nematic liquid crystals, although (subject to some modifications) all the inertial effects considered below do occur also in cholesteric and smectic phases. The linearized equations of hydrodynamics for nematic liquid crystals are well known (see, for example, Ref. 6):

$$\frac{\partial \mathbf{L}}{\partial t} = [\mathbf{n} \times \mathbf{h}] - [\mathbf{n} \times \mathbf{R}], \quad \rho \frac{\partial \xi_i}{\partial t} = -\frac{\partial P}{\partial x_i} + \frac{\partial \Sigma_{ji}}{\partial x_j},$$

$$\rho T \frac{\partial s}{\partial t} = \Sigma_{ji} \frac{\partial \xi_i}{\partial x_j} + \mathbf{R} \frac{d\mathbf{n}}{dt} + \kappa_{ji} \frac{\partial^2 T}{\partial x_i \partial x_j}.$$
(1)

Here, L is the angular momentum associated with the angular rotational velocity of the director:

$$\partial \mathbf{n}/\partial t = [\mathbf{\Omega} \times \mathbf{n}], \quad \mathbf{L} = I\mathbf{\Omega}$$
 (2)

(it is usual to ignore the moment of inertia I and we then have L = 0) and **h** represents an external field **H** as well as an internal field associated with intermolecular forces (Frank energy):

$$[\mathbf{n} \times \mathbf{h}] = \chi_{a}(\mathbf{H}\mathbf{n}) [\mathbf{n} \times \mathbf{H}] + \left\{ \int d^{3} r E, \mathbf{L} \right\}, \tag{3}$$

where χ_a is the anisotropic part of the susceptibility; $\{\ldots\}$ is the Poisson bracket; *E* is the density of the Frank elastic energy.

The dissipation **R** in the system (1) is defined as follows: $R_i = \gamma_i N_i + \gamma_2 n_j A_{ji}$, (4)

where γ_1 and γ_2 are the rotational viscosity coefficients,

$$\mathbf{N} = [(\mathbf{\Omega} - \boldsymbol{\omega}) \times \mathbf{n}], \ A_{ij} = \frac{1}{2} \left(\frac{\partial \xi_i}{\partial x_j} + \frac{\partial \xi_j}{\partial x_i} \right), \quad \boldsymbol{\omega} = \frac{1}{2} \operatorname{rot} \xi,$$
(5)

and ξ is the velocity.

978 Sov. Phys. JETP 60 (5), November 1984

The following quantities occur also in the system (1): P is the pressure, Σ_{ij} is the dissipative part of the stress tensor given by

 $\sum_{ji} = \alpha_1 n_i n_j A_{km} n_k n_m + \alpha_2 n_i N_j + \alpha_3 n_j N_i$

$$+\alpha_4A_{ij}+\alpha_5n_in_kA_{hj}+\alpha_6A_{ih}n_kn_j, \qquad (6)$$

and α_i are the Leslie viscosity coefficients. They are not all independent, but are related by the Onsager relationships: $\alpha_2 + \alpha_3 = \alpha_6 - \alpha_5$; $\gamma_1 = \alpha_3 - \alpha_2$; $\gamma_2 = \alpha_2 + \alpha_3$.

The rest of the notation in Eq. (1) as follows: ρ is the density, T is the absolute temperature, s is the entropy density, and x_{ij} is the thermal conductivity tensor described by

$$\kappa_{ij} = \kappa_0 \delta_{ij} + \kappa_a n_i n_j. \tag{7}$$

As is usual, in the process of linearization it is necessary to introduce small deviations from equilibrium values:

$$\delta \pi = P - P_0, \quad \delta \rho \equiv \rho - \rho_0, \quad \delta \sigma \equiv s - s_0. \tag{8}$$

We shall use the simplest form of the equation of state:

$$\delta \pi = c^2 \delta \rho + \theta \delta \sigma_{\bullet} \tag{9}$$

Here, c is the adiabatic velocity of sound; $\theta \equiv (\partial P / \partial \sigma)_{\rho}$. We shall give also the Poisson brackets⁷ necessary for the calculation of **h** in Eq. (3):

$$\{L_{\alpha}(1), L_{\beta}(2)\} = -e_{\alpha\beta\gamma}L_{\gamma}\delta(1-2), \{L_{\alpha}(1), n_{\beta}(2)\} = -e_{\alpha\beta\gamma}n_{\gamma}\delta(1-2), \{n_{\alpha}(1), n_{\beta}(2)\} = 0.$$
(10)

As already pointed out in the Introduction, a direct solution of the system (1) in its general form is hardly possible. Therefore, we shall average these equations over a "base" solution representing homogeneous rotation of the director:

$$\mathbf{n} = \mathbf{u}\cos\psi + \mathbf{v}\sin\psi. \tag{11}$$

In this solution the angle ψ represents rotation of **n** from an arbitrary direction in a plane defined by the unit vectors **u** and **v**. We shall assume that ψ is a fast variable (roughly speaking, we shall postulate that $\psi \propto \Omega t$) and we shall carry out averaging with respect to this variable. The orbital momentum L is defined as follows:

$$\mathbf{L} = L\mathbf{w}, \quad \mathbf{w} = [\mathbf{u} \times \mathbf{v}]. \tag{12}$$

The set of three vectors \mathbf{u} , \mathbf{v} , and \mathbf{w} forms a base reference set. The base solution (11) corresponds to the conditions $\mathbf{L} = \text{const}$ and $\mathbf{L} \cdot \mathbf{n} = 0$. In describing homogeneous solutions we must allow for the fact that the base reference set and the phase ψ vary from point to point (see Ref. 8). The deviation of ψ from a certain average value ψ_0 at a given moment ($\psi = \psi_0 + \delta \psi$) describes spatial dephasing of the director rotation. It is convenient at this stage to adopt the following procedure from Ref. 8. Let us assume that X is some hydrodynamic variable of interest to us and $R_{ij} = \delta_{ij} - \alpha_k e_{kij}$ is the matrix of an infinitesimally small rotation by an angle α_k , which is generally spatially inhomogeneous. Then, such rotation alters the variable X as follows:

$$\mathbf{X} \to \mathbf{X} + [\boldsymbol{\alpha} \times \mathbf{X}]. \tag{13}$$

The spatial derivative of X is obtained from Eq. (13):

$$\nabla_i \mathbf{X} = [\mathbf{A}_i \times \mathbf{X}], \tag{14}$$

where

$$\mathbf{A}_i = \boldsymbol{\nabla}_i \boldsymbol{\alpha}. \tag{15}$$

Similarly, we find from Eqs. (14) and (15) that

$$\nabla_i \nabla_j \mathbf{X} = [\nabla_j \times \mathbf{A}_i \mathbf{X}] + \mathbf{A}_j (\mathbf{A}_i \mathbf{X}) - \mathbf{X} (\mathbf{A}_i \mathbf{A}_j).$$

Hence, in the linear approximation we obtain

$$\nabla_i \nabla_j \mathbf{X} = [\nabla_j \times \mathbf{A}_i \mathbf{X}]. \tag{16}$$

We can easily demonstrate also that $[X \times \nabla_i X] = A_i X^2 - X (A_i X).$

There is some degree of arbitrariness in the choice of the values of A_i . In particular, it is convenient to introduce the gauge

 $A_i X = 0.$

In this gauge, we have

$$[\mathbf{X} \times \nabla_i \mathbf{X}] = \mathbf{A}_i. \tag{17}$$

In particular, in the gauge $A_i \cdot w = 0$ it follows from Eqs. (16) and (17) that

$$\nabla_i \mathbf{w} = [\mathbf{A}_i \times \mathbf{w}], \quad \Delta \mathbf{w} = [\nabla_j \times \mathbf{A}_j \mathbf{w}], \quad [\mathbf{w}_i \times \nabla_i \mathbf{w}] = \mathbf{A}_i.$$
(18)

In the same way we obtain

$$\Delta \mathbf{u} = -\mathbf{w} (\mathbf{u} \Delta \mathbf{w}), \quad \Delta \mathbf{v} = -\mathbf{w} (\mathbf{v} \Delta \mathbf{w}). \tag{19}$$

In principle, Eqs. (16)–(19) solve the problem of averaging of the system (1). The details of this procedure are described in the Appendix. Here we shall give the final results. The average equations of motion of the director describing the dynamics of the orbital momentum are as follows:

$$L \frac{\partial w_{u}}{\partial t} + \frac{K}{2} \Delta w_{v} = \frac{1}{4} \gamma_{i} \left(\mathbf{u} \operatorname{rot} \boldsymbol{\xi} - 2 \frac{\partial w_{v}}{\partial t} \right) - \frac{\gamma_{2}}{2} w_{p} A_{pk} v_{k},$$

$$L \frac{\partial}{\partial t} w_{v} - \frac{K}{2} \Delta w_{u} = \frac{1}{4} \gamma_{i} \left(\mathbf{v} \operatorname{rot} \boldsymbol{\xi} + 2 \frac{\partial w_{u}}{\partial t} \right) + \frac{\gamma_{2}}{2} w_{p} A_{pk} u_{k},$$

$$I \frac{\partial^{2}}{\partial t^{2}} \delta \psi - K \Delta \delta \psi + \gamma_{i} \frac{\partial}{\partial t} (\delta \psi) = \frac{\gamma_{i}}{2} (\mathbf{w} \operatorname{rot} \boldsymbol{\xi}).$$
(20)

The notation here is the same as in the initial system (1) and we have introduced moreover the following quantities: K is the Frank constant (for simplicity, we shall use the one-constant approximation); $\partial w_u / \partial t \equiv \mathbf{u} \cdot \partial \mathbf{w} / \partial t$; $\Delta w_u \equiv \mathbf{u} \cdot \Delta \mathbf{w}$ and similar definitions of $\partial w_v / \partial t$ and Δw_v ; $\delta \psi$ describes the dephasing:

$$L - L_0 = \delta L = I \delta \Omega = I \partial \delta \psi / \partial t,$$

and L_0 is the precessing moment¹⁾

 $\partial L_0/\partial t = -\gamma_1 L_0/I.$

The average Navier-Stokes equations are

$$\rho \frac{\partial \xi_{i}}{\partial t} = -c^{2} \frac{\partial}{\partial x_{i}} \delta \rho + \theta \frac{\partial}{\partial x_{i}} \delta \sigma + \alpha_{i} (ijkm) \frac{\partial A_{km}}{\partial x_{j}} + (\alpha_{2} - \alpha_{3}) \frac{L}{2I} (\operatorname{rot} \mathbf{w})_{i} - \frac{\alpha_{2}}{4} (\delta_{im} - w_{i}w_{m}) (\operatorname{rot} \operatorname{rot} \boldsymbol{\xi})_{m} - \frac{\alpha_{3}}{4} e_{ipq} (\delta_{iq} - w_{j}w_{q}) \frac{\partial}{\partial x_{j}} (\operatorname{rot} \boldsymbol{\xi})_{p} + \alpha_{4} \frac{\partial A_{ij}}{\partial x_{j}} + \frac{\alpha_{5}}{2} (\delta_{ik} - w_{i}w_{k}) \frac{\partial}{\partial x_{j}} A_{kj} + \frac{\alpha_{6}}{2} (\delta_{jk} - w_{j}w_{k}) \frac{\partial}{\partial x_{j}} A_{ik}.$$
(21)

We have introduced here (ijkm) for the average $\langle n_i n_j n_k n_m \rangle$.

This average obeys an analog of the Wick theorem:

$$(ijkm) = \frac{1}{8} \left[(\delta_{ij} - w_i w_j) (\delta_{km} - w_k w_m) + (\delta_{ik} - w_i w_k) (\delta_{jm} - w_j w_m) + (\delta_{im} - w_i w_m) (\delta_{jk} - w_j w_k) \right].$$
(22)

Finally, we shall give the average equation for the heat flux:

$$\rho T \frac{\partial}{\partial t} \delta \sigma = \gamma_{i} \frac{2L}{I} \frac{\partial}{\partial t} \delta \psi + \left[\left(\varkappa_{0} + \frac{\varkappa_{a}}{2} \right) \delta_{ij} - \frac{\varkappa_{a}}{2} w_{i} w_{j} \right] \\ \times \frac{\partial^{2}}{\partial x_{i} \partial x_{j}} [\lambda_{i} \delta \rho + \lambda_{2} \delta \sigma].$$
(23)

We have used above the thermodynamic relationship

$$\delta T = \lambda_1 \delta \rho + \lambda_2 \delta \sigma; \quad \lambda_1 = (\partial T / \partial \rho)_{\sigma}, \quad \lambda_2 = (\partial T / \partial \sigma)_{\rho}.$$

The system of equations (20)–(23) gives complete information on the dynamics of the average system and in particular it gives all its normal modes. In the next section we shall consider these problems in greater detail. Here, we shall stress that the system of the average equations (20)–(23) is linearized in respect of the usual hydrodynamic variables (ρ , s, ξ) and in respect of the orbital momentum L, but all the nonlinear powers of the director components are included.

III. NORMAL MODES OF THE AVERAGE SYSTEM

Although the equations of motion for the hydrodynamic variables of the average system (L, ξ , σ , ρ) are still very complex (for example, determination of the spectrum requires a calculation of a 8×8 determinant of fairly general form), we can nevertheless draw conclusions of qualitative nature on some slowly varying (compared with the director rotation period) characteristics of the dynamics of nematic liquid crystals.

First of all, we must point out that it is incorrect to assume simply that $\xi = 0$ in these equations. The point is this: if $\xi = 0$, then the Navier-Stokes equations yield the relationship

$$-c^{2}\frac{\partial}{\partial x_{i}}\delta\rho+\theta\frac{\partial}{\partial x_{i}}\delta\sigma+(\alpha_{2}-\alpha_{3})\frac{L}{2I}(\operatorname{rot} \mathbf{w})_{i}=0, \qquad (24)$$

which imposes important restrictions on the motion of the momentum in this case. Therefore, in considering the director waves^{2,3} it is not quite correct to ignore the flow of the liquid. The physical meaning of this conclusion is quite clear. The rotation of the director gives rise to a flow of the liquid and, therefore, the absence of such flow imposes serious restrictions on the motion of the director described by Eq. (24).

Similarly, if in the heat conduction equation we assume that $\delta \rho = \delta \alpha = 0$ (isentropic motion of an incompressible liquid crystal), it then follows from Eq. (23) that $\delta \psi = \text{const.}$ In its turn, the dephasing equation then limits the possible liquid flows:

Once again the meaning of this restriction is that entropy is generated because of dephasing of the director rotation.

We shall investigate in greater detail the dispersion law of new orbital waves in the average system by introducing some additional simplifications. We shall consider the limit of low viscosities. Specifically, we shall ignore all the dissipative terms in the equations for the momentum and in the Navier-Stokes equations that do not contain a large (in our approximation) coefficient $\sim L/I$. In fact, this condition is satisfied since is corresponds to the inequality

 $\alpha_i \ll (L \omega K)^{4/2} \approx 1 \mathbf{P}$

(for characteristic frequencies of the orbital waves). In this approximation the system (20)–(23) becomes very simple and it shows that, in addition to the usual hydrodynamic modes of an isotropic liquid (with slightly renormalized parameters), the new orbital waves are of the acoustic and magnon types.

In fact, in this limit it follows from Eqs. (20)-(23) that

$$L\frac{\partial}{\partial t}w_{u} + \frac{K}{2}\Delta w_{v} = 0, \qquad L\frac{\partial}{\partial t}w_{v} - \frac{K}{2}\Delta w_{u} = 0,$$

$$I\frac{\partial^{2}}{\partial t^{2}}\delta\psi - K\Delta\delta\psi = 0, \qquad \rho T\frac{\partial}{\partial t}\delta\sigma = \gamma_{1}\frac{2L}{I}\frac{\partial}{\partial t}\delta\psi, \qquad (25)$$

$$\rho\frac{\partial\xi_{i}}{\partial t} = -c^{2}\frac{\partial}{\partial x_{i}}\delta\rho + \theta\frac{\partial}{\partial x_{i}}\delta\sigma + (\alpha_{2} - \alpha_{3})\frac{L}{2I}(\operatorname{rot} \mathbf{w})_{i}.$$

In this system the heat conduction equation is coupled to the equation for the dephasing $\delta \psi$. It follows from these two equations that the spectrum of the normal modes of the system includes two $\delta \psi$ oscillation modes. If we include the corrections associated with the viscosity coefficient γ_1 , the spectrum becomes

$$\omega_{1,2} = i \frac{\gamma_1}{2I} \pm \left(\frac{K}{I} q^2 - \frac{{\gamma_1}^2}{4I^2}\right)^{1/2}.$$
 (26)

If $q < (\gamma_1^2/IK)^{1/2} \sim 10^3 - 10^4$ cm⁻¹, then for the selected parameters Eq. (26) yields pure diffusion modes, whereas for the opposite inequality we have two weakly damped modes of the acoustic type:

$$\omega_{1,2} = \pm (K/I)^{\frac{1}{2}} q + i\gamma_{1}/2I.$$
(27)

Writing down the dispersion equation for the system (25), we find that a 4×4 block for the conventional hydrodynamic variables of an isotropic liquid (sound plus two modes describing transverse shear) is separated from the relevant determinant and the rest of the determinant gives rise to the two phase orbital modes mentioned above, a mode associated mainly with heat conduction and also two orbital modes associated with transverse deviations of the orbital momentum w_u and w_v . They obey a magnon-type dispersion equation:

$$\omega_{s,i} = \pm \frac{K}{2L} q^2 \left(1 \mp \frac{i \gamma_i}{L} \right). \tag{28}$$

These waves are weakly damped in the approximation used by us to derive the system (25).

We shall now assume that the geometry of the system is such that the dependence on just one coordinate z is significant and the flow of the liquid is possible only along this coordinate: $\xi_1 = \xi_2 = 0$.

The system of average equations (20)–(23) together with the equation of continuity then becomes $(\psi'' \equiv d^2 \psi/dz^2, \text{ etc.})$:

$$\frac{\partial^2 \psi}{\partial t^2} = -\frac{\gamma_1}{I} \frac{\partial \psi}{\partial t} + \frac{K}{I} \psi'',$$

$$L\frac{\partial w_{u}}{\partial t} = -\frac{K}{2} w_{v}'' - \gamma_{2} w_{3} \frac{\partial \xi_{3}}{\partial z} v_{3} + \gamma_{4} \frac{\partial w_{v}}{\partial t},$$

$$L\frac{\partial w_{v}}{\partial t} = -\frac{K}{2} w_{u}'' + \gamma_{2} w_{3} \frac{\partial \xi_{3}}{\partial z} u_{3} - \gamma_{4} \frac{\partial w_{u}}{\partial t},$$

$$\rho_{0} \frac{\partial \xi_{3}}{\partial t} = -\frac{\partial \pi}{\partial z} + \left[\alpha_{4} + \frac{3}{8} \alpha_{4} (1 - w_{3}^{2})^{2} + \frac{1}{2} (\alpha_{5} + \alpha_{6}) (1 - w_{3}^{2})\right] \frac{\partial^{2} \xi_{3}}{\partial z^{2}}$$

$$\rho_{0} T_{0} \frac{\partial \sigma}{\partial t} = \frac{2L}{I} \gamma_{4} \frac{\partial}{\partial t} \delta \psi + \lambda_{4} \left[\kappa_{0} + \frac{1}{2} \kappa_{a} (1 - w_{3}^{2})\right] \frac{\partial^{2} \rho}{\partial z^{2}}$$

$$+ \lambda_{2} \left[\kappa_{0} + \frac{1}{2} \kappa_{a} (1 - w_{3}^{2})\right] \frac{\partial^{2} \sigma}{\partial z^{2}},$$

$$\frac{\partial}{\partial t} \delta \rho + \rho_{0} \frac{\partial \xi_{3}}{\partial z} = 0, \quad \delta \pi = c^{2} \delta \rho + \theta \delta \sigma.$$

$$(29)$$

However, as pointed out above, it is not possible simply to assume that $\xi_1 = \xi_2 = 0$ in the average equations for a nematic liquid crystal with a rotating director. There is a restriction which follows from the Navier-Stokes equations and applies to the ξ_1 and ξ_2 velocity components that are assumed to vanish in the case of one-dimensional flow. Naturally, this restriction occurs also in the case of a low but finite viscosity. A simple analysis shows that the magnon-type orbital waves are in this case completely impossible. On the other hand, the mode (26) for dephasing does occur also in the one-dimensional case [as it follows directly from Eq. (29)].

A similar situation occurs also in the two-dimensional case. We shall consider two-dimensional flow described by $\xi_3 = 0$ ($\xi_1, \xi_2 \neq 0$) in a plane perpendicular to the equilibrium direction of the orbital momentum. We shall introduce a coordinate system such that $\mathbf{x} || \mathbf{u}, \mathbf{y} || \mathbf{v}$, and $\mathbf{z} || \mathbf{w}$ and assume that there is no dependence on z (two-dimensional case). Equations (20)–(23) then assume the following form (for a liquid crystal regarded as incompressible):

$$L \frac{\partial}{\partial t} w_{u} + \frac{K}{2} \Delta w_{v} = \frac{1}{4} \gamma_{i} (\operatorname{rot} \xi)_{u} + \frac{\gamma_{i}}{2} \frac{\partial}{\partial t} w_{v},$$

$$L \frac{\partial}{\partial t} w_{v} - \frac{K}{2} \Delta w_{u} = \frac{1}{4} \gamma_{i} (\operatorname{rot} \xi)_{v} - \frac{\gamma_{i}}{2} \frac{\partial}{\partial t} w_{u},$$

$$I \frac{\partial^{2}}{\partial t^{2}} \delta \psi - K \Delta \delta \psi + \gamma_{i} \frac{\partial}{\partial t} \delta \psi = \frac{\gamma_{i}}{2} \left(\frac{\partial \xi_{i}}{\partial x_{2}} - \frac{\partial \xi_{2}}{\partial x_{i}} \right),$$

$$\rho \frac{\partial \xi_{i}}{\partial t} = -c^{2} \frac{\partial}{\partial x_{i}} \delta \rho - \theta \frac{\partial}{\partial x_{i}} \delta \sigma + \alpha \Delta \xi_{i},$$

$$\rho T \frac{\partial}{\partial t} \sigma - \gamma_{i} \frac{2L}{I} \frac{\partial}{\partial t} \delta \psi - \left(\varkappa_{0} + \frac{1}{2} \varkappa_{a} \right) \Delta (\lambda_{i} \delta \rho + \lambda_{2} \delta \sigma) = 0,$$
(30)

where

$$\alpha \equiv \frac{1}{8}\alpha_1 + \frac{1}{4}\alpha_2 - \frac{1}{4}\alpha_3 + \frac{1}{2}\alpha_4 + \frac{1}{4}\alpha_5 + \frac{1}{4}\alpha_6 > 0.$$

Once again the usual hydrodynamic block (sound and damped transverse shear) can be separated in the dispersion equation. The equation for the dephasing is related only to the component (curl ξ)₃. The Navier-Stokes equations are easily obtained for this component:

$$\rho \frac{\partial}{\partial t} (\operatorname{w} \operatorname{rot} \xi) = \alpha \Delta (\operatorname{w} \operatorname{rot} \xi).$$
(31)

Therefore, the third equation of the system (30)-(31) gives again two orbital phase waves of Eq. (26) with the acoustictype dispersion. The magnon orbital modes are forbidden by the condition (24), which in this case has the form

 $(rot w)_{3} = 0$.

IV. CONCLUSIONS

We shall now consider the possibility of observing the new orbital waves predicted in the preceding section. Ways of creating, in principle, states with a rotating director necessary for the application of our procedure (by applying a rotating magnetic field, investigating transient regimes of a high-frequency electrohydrodynamic instability, or using pulsed fields), were considered by us in Ref. 4 and we shall not repeat the arguments given there. We shall also mention that all conclusions of our investigation are valid for time intervals 10^{-3} - 10^{-4} sec representing the "lifetime" of such rotational states (see Ref. 4).

Inelastic scattering of light provides a direct method for observing these orbital waves. In principle, the positions, profiles, and intensities of lines in the spectrum can be used to determine all the parameters of the orbital waves and to compare the results found in this way with our theoretical predictions.

It should be pointed out that the properties of nonequilibrium quasiequilibrium systems, particularly fluctuations of the various parameters of such systems, are currently attracting considerable interest (see, for example, Refs. 9–12). However, a calculation of the spectrum of light scattered inelastically by our nonequilibrium system meets with difficulties, which may be even of fundamental nature.

The point is that the scattering of light involves fluctuations of the permittivity tensor and these in turn are governed by fluctuations of the hydrodynamic variables ρ, ξ, σ , ψ , w_{μ} , and w_{ν} . Since we are interested only in new orbital waves, we shall consider only the variables ψ , w_u , and w_v . An allowance for ρ , ξ , and σ in such situations when the orbital waves appear at all (see above) does not alter the situation in the qualitative sense. We shall therefore consider fluctuations of the variables ψ , w_{μ} , and w_{ν} . Fluctuations of conventional equilibrium systems are governed by thermodynamic relationships, particularly by the nature of the free energy, and the correlation functions occurring in the scattered-light intensity satisfy the fluctuation-dissipation theorem.⁹ However, we are dealing with the average system and with fluctuations against the background obtained by dynamic averaging. If we are interested only in the positions of lines in the spectrum, i.e., in the pole denominators of the corresponding correlation functions, then this aspect is quite unimportant because one-time correlation functions governed by fluctuations of dynamically averaged quantities ψ , w_{μ} , and w_{ν} occur only in the numerators of the corresponding expressions for the scattering intensity.

If we nevertheless wish to write down some closed expressions for the correlation functions occurring in the scattering cross section, we can make a natural (in our opinion) assumption that fluctuations in the average system are averaged on the same scale of frequencies by thermodynamic relationships. In fact, this assumption simply means the transfer of the frequency scale which is used in hydrodynamic averaging, from the atomic level (as in the equilibrium case) to characteristic frequencies of the director rotation in our case.

We have in fact made this assumption in going to the limit of low viscosities in Eqs. (20)–(23). We have then ignored the unrenormalized dissipative coefficients originating from the averaging over the atomic frequencies and retained only the renormalized viscosity associated with the frequency scale $\sim \Omega$. When this assumption is made, the subsequent procedure in the calculation of the correlation functions becomes standard.^{13,14} We have to calculate the simultaneous correlation functions of hydrodynamic quantities so that we need to write down the energy of an inhomogeneous state in terms of the variables w and ψ . Subject to the assumptions made above, we can find this energy by averaging the Frank energy:

$$E=\frac{K}{2}\int (\nabla \mathbf{n})^2 d^3r.$$

Proceeding in the same way as in the derivation of the system of equations (20)–(23), we find that such dynamic averaging gives

$$E = \frac{K}{4} \int [(\nabla w_1)^2 + (\nabla w_2)^2 + 2(\nabla \psi)^2] d^3r.$$
 (32)

We can describe simultaneous correlation functions by calculating the relevant functional integral with the Gibbs distribution function $\exp(-E/T)$. However, the Gibbs distribution can be obtained if the energy is expressed in canonically conjugate variables. In the present case representing the linear approximation $(w_3 \approx 1, w_1, w_2 < 1)$, which is the only one necessary for our purposes, the variables w_1 and w_2 are conjugate to one another, whereas ψ is conjugate to L, for which the energy E of Eq. (32) is completely independent. We can demonstrate this by calculating the Poisson brackets of these quantities in the linear approximation. We can easily see that

$$\{w_1, w_2\} = \text{const}, \{w_1, \psi\} = \{w_2, \psi\} = 0.$$

Therefore, the variables w_1 , w_2 , and ψ in Eq. (32) are separable, and L is generally a cyclic variable. Consequently, the simultaneous correlation function $\langle w_i w_j \rangle$ and $\langle \delta \psi \delta \psi \rangle$ can be found directly from Eq. (32).

We shall now consider time-dependent correlation functions, which indeed govern the light scattering cross section:

$$C_{\alpha\beta}(\mathbf{r}, t) = \langle w_{\alpha}(\mathbf{r}, t) w_{\beta}(0, 0) \rangle$$
(33)

[a similar expression applies also in the case when $\langle \delta \psi(\mathbf{r},t) \delta \psi(0,0) \rangle$]. The equation for the correlation function $C_{\alpha\beta}$ can be obtained directly from Eq. (20) (for simplicity, we shall ignore a low viscosity $\sim \gamma_1/L$):

$$L\frac{\partial}{\partial t}C_{\alpha\beta}(\mathbf{r},t) = -e_{\alpha\gamma}\frac{K}{2}\Delta C_{\gamma\beta}(\mathbf{r},t), \qquad (34)$$

where

$$e_{\alpha\gamma} = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix} .$$

We shall now apply the time Laplace tranformation and the coordinate Fourier tranformation:

$$\mathcal{C}_{\alpha\beta}(\mathbf{q},p) = \int dt C_{\alpha\beta}(\mathbf{q},t) e^{ipt}, \quad \text{Im } p > 0.$$

It then follows from Eq. (34) that

$$LC_{\alpha\beta}(\mathbf{q},t=0) - ipL\tilde{C}_{\alpha\beta}(\mathbf{q},p) - e_{\alpha\gamma} \frac{K}{2} q^2 \tilde{C}_{\gamma\beta}(\mathbf{q},p) = 0, \quad (35)$$

so that $\hat{C}_{\alpha\beta}(\mathbf{q}, p)$

$$=L\left(iLp\delta_{\alpha\gamma}-\frac{K}{2}q^{2}e_{\alpha\gamma}\right)C_{\gamma\beta}(\mathbf{q},t=0)\left(-L^{2}p^{2}+\frac{K^{2}}{4}q^{4}\right)^{-1}.$$
 (36)

The initial condition for the correlation function $C_{\alpha\beta}$ should be selected in accordance with the fluctuation-dissipation theorem in such a way that in the static limit we obtain the correlation functions $\langle w_i w_j \rangle$ and $\langle \delta \psi \delta \psi \rangle$ that follow from Eq. (32). Then, Eq. (36) is the solution of the problem of the inelastic scattering of light. The scattering cross section is governed, as is well known, by fluctuations of the permittivity tensor:

$$d\sigma/d\omega \sim \langle \delta \varepsilon_{\alpha\beta} \delta \varepsilon_{\gamma\delta}^* \rangle e_{\alpha} e_{\gamma} e_{1\beta} e_{1\delta}, \qquad (37)$$

where $\delta \varepsilon_{\alpha\beta}$ is a fluctuation of the permittivity tensor associated with fluctuations of the hydrodynamic variables of our problem; **e** and **e**₁ are the polarization vectors of the incident and scattered light. The relationship between $\delta \varepsilon_{\alpha\beta}$ and the fluctuations of **w** can be obtained directly by the above-described method of dynamic averaging of the familiar relationship $\varepsilon_{ij} = \varepsilon_0 \delta_{ij} + \varepsilon_a n_i n_j$, where ε_0 is the isotropic part and ε_a is the permittivity anisotropy.

We can easily see that

$$\langle \varepsilon_{ij} \rangle = (\varepsilon_0 + \frac{i}{2} \varepsilon_a) \delta_{ij} - \frac{i}{2} \varepsilon_a w_i w_j.$$
(38)

Hence and from Eq. (37) it follows that

$$\frac{d\sigma}{d\omega} \approx \frac{1}{2} \varepsilon_a^2 \left(-L^2 \omega^2 + \frac{K^2}{4} q^4 \right)^{-1} \operatorname{Re} C_{\alpha\beta\gamma\delta} e_\alpha e_\gamma e_{i\beta} e_{i\delta}, \quad (39)$$

where

$$C_{\alpha\beta\gamma\delta} = w_{\alpha}w_{\beta}\left(iL\omega\delta_{\gamma\gamma_{1}} - \frac{K}{2}q^{2}e_{\gamma\gamma_{1}}\right)C_{\gamma_{1}\delta_{1}}(\mathbf{q}, t=0),$$

and the brackets in the indices indicate symmetrization.

In principle, the contribution of the $\delta \psi$ mode can be allowed for in a similar manner. However, the relationship between ε_{ij} and $\delta \psi$ is indirect because of the coupling of the variables $\delta \psi$, α , and ρ .

We shall now summarize our results. We considered the equations of dynamics of a nematic liquid crystal with an inertia of the director and we averaged them over the fast motion of the director. The averaging gave a new thermodynamic system and the orbital waves found by us are small deviations of this system from an equilibrium state. In averaging the director rotation of the phase we effectively ignored all fast (compared with the rotation period) fluctuations. This average system is characterized by its own entropy production law and the correlation functions of random functions should be obtained, following the general method,⁹ by employing the transport coefficients which occur in this law.

The principal physical conclusion of our investigation is that the spectrum of light scattered by a rotating nematic liquid crystal has new lines corresponding to the normal modes (26) and (28) of the average system. The positions of these lines depend on the actual values of the parameters and also on the wave vector transferred in the process of scattering. For the average values of the quantities I, Ω , K, γ_1 , and $q \sim 10^4$ cm⁻¹ used in the derivation of the average equations the line corresponding to the phase model lies in the frequency range $\sim 10^5 \text{ sec}^{-1}$ and the magnon mode of the orbital momentum oscillations lies at frequencies $\sim 10^4 \text{ sec}^{-1}$. Unfortunately, we do not know the true parameters of our model. Therefore, our estimates should be treated as tentative. In any case, suitable experiments would provide a method for the determination of the moment of inertia per unit volume of a nematic single crystal, which is the main parameter that governs the feasibility of propagation of new orbital waves. We should also point out that there are grounds for assuming that waves of this type are responsible for the propagation of pulses in biological systems.¹⁵

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APPENDIX

We shall begin from averaging the equation for the momentum:

$$\frac{\partial L_{\alpha}}{\partial t} = K e_{\alpha\beta\gamma} n_{\beta} \Delta n_{\gamma} - \gamma_1 [\mathbf{n} \times \mathbf{N}]_{\alpha} - \gamma_2 [\mathbf{n}, \times \mathbf{n} \hat{A}]_{\alpha}.$$
(A.1)

Here, the first term on the right-hand side is obtained by calculation of the Poisson brackets occurring in Eq. (3):

$$[\mathbf{n} \times \mathbf{N}]_{i} = \Omega_{i} - \frac{i}{2} (\delta_{ij} - n_{i} n_{j}) (\operatorname{rot} \boldsymbol{\xi})_{j},$$

$$[\mathbf{n} \times \mathbf{n} \hat{A}]_{i} = e_{ijk} n_{j} n_{p} A_{pk}.$$
(A.2)

Averaging with the aid of the base solution (11) gives

$$\langle \delta_{ij} - n_i n_j \rangle = \frac{i}{2} (\delta_{ij} + w_i w_j), \qquad (A.3)$$

$$\langle n_j n_p \rangle = \frac{1}{2} (\delta_{jp} - w_j w_p). \tag{A.4}$$

Moreover,

$$\langle e_{\alpha\beta\gamma}n_{\beta}\Delta n_{\gamma}\rangle = \frac{i}{2} [\mathbf{u}(\mathbf{u}\Delta\mathbf{w}) - \mathbf{v}(\mathbf{v}\Delta\mathbf{w}) - \frac{i}{2}\mathbf{w}\Delta\psi]_{\alpha}.$$
 (A.5)

In this way we obtain the following average equation for the momentum:

$$\left\langle \frac{\partial}{\partial t} L_i \right\rangle = -\frac{K}{2} \left[u_i (\mathbf{v} \Delta \mathbf{w}) - v_i (\mathbf{u} \Delta \mathbf{w}) - w_i \Delta \psi \right] - \frac{\gamma_1}{I} \langle L_i \rangle + \frac{\gamma_1}{4} (\delta_{ij} + w_i w_j) (\operatorname{rot} \xi)_j - \frac{\gamma_2}{2} e_{ijk} A_{pk} (\delta_{jp} - w_j w_p). \quad (A.6)$$

Direct calculations indicate that

$$\frac{1}{I} \langle L_i \rangle = \langle [\mathbf{n}\mathbf{n}]_i \rangle = \psi w_i + \frac{1}{2} ([\mathbf{u} \times \mathbf{\dot{u}}] + [\mathbf{v} \times \mathbf{\dot{v}}])_i,$$
$$\frac{1}{I} \langle L_i \rangle = \psi w_i - \psi ([\mathbf{u} \times \mathbf{\dot{v}}] - [\mathbf{v} \times \mathbf{\dot{u}}])_i$$

V. L. Golo and E. I. Kats 982

(the dot denotes differentiation with respect to time). In the last expression we omitted the second derivatives of the slow variables \mathbf{u} and \mathbf{v} .

We can thus see that the system (20) follows from Eq. (A.6).

The Navier-Stokes equations can be obtained by averaging the following terms:

$$A_{1} = \left\langle \frac{\partial}{\partial x_{j}} (n_{i}n_{j}n_{k}n_{m}A_{km}) \right\rangle, \quad A_{2} = \left\langle \frac{\partial}{\partial x_{j}} n_{i}N_{j} \right\rangle,$$
$$A_{3} = \left\langle \frac{\partial}{\partial x_{j}} n_{j}N_{i} \right\rangle, \quad A_{4} = \left\langle \frac{\partial}{\partial x_{j}} n_{i}n_{k}A_{kj} \right\rangle, \quad (A.7)$$
$$A_{5} = \left\langle \frac{\partial}{\partial x_{j}} A_{ki}n_{k}n_{j} \right\rangle.$$

Retaining in Eq. (A.7) only the terms linear in respect of the gradients, we find from Eq. (11) that

$$A_{i} = \frac{1}{8} \frac{\partial A_{km}}{\partial x_{j}} \left[\left(\delta_{ij} - w_{i} w_{j} \right) \left(\delta_{km} - w_{k} w_{m} \right) + \left(\delta_{ik} - w_{i} w_{k} \right) \right. \\ \left. \times \left(\delta_{jm} - w_{j} w_{m} \right) + \left(\delta_{im} - w_{i} w_{m} \right) \left(\delta_{jk} - w_{j} w_{k} \right) \right].$$
(A.8)

A direct calculation gives

$$A_{2} = \left\langle \frac{\partial}{\partial x_{j}} (n_{i}e_{jpq}(\Omega_{p} - \omega_{p})n_{q}) \right\rangle$$

$$= -\frac{\partial \omega_{p}}{\partial x_{j}} e_{jpq} \langle n_{i}n_{q} \rangle + \left\langle \frac{\partial}{\partial x_{j}} e_{jpq}n_{i}\Omega_{p}n_{q} \right\rangle$$

$$= -\frac{1}{4} (\delta_{im} - w_{i}w_{m}) (\text{rot rot } \xi)_{m}$$

$$+ e_{jpq} \frac{\partial \Omega_{p}}{\partial x_{j}} \langle n_{i}n_{q} \rangle + e_{jpq}\Omega_{p} \left\langle \frac{\partial}{\partial x_{j}} (n_{i}n_{q}) \right\rangle$$

$$= -\frac{1}{4} (\delta_{im} - w_{i}w_{m}) (\text{rot rot } \xi)_{m} + \frac{L}{2I} (\text{rot } \mathbf{w})_{i}$$

$$- \frac{L}{2I} w_{i} \left[\mathbf{w}, \frac{\partial}{\partial x_{j}} \mathbf{w} \right]_{j} - \frac{L}{2I} w_{i} \left[v_{j} \left(\mathbf{u} \frac{\partial}{\partial x_{j}} \mathbf{w} \right) - u_{j} \left(\mathbf{v} \frac{\partial}{\partial x_{j}} \mathbf{w} \right) \right].$$
(A.9)

The last two terms in Eq. (A.9) cancel out and we obtain

$$A_{2} = \frac{L}{2I} (\operatorname{rot} \mathbf{w})_{i} - \frac{1}{4} (\delta_{im} - w_{i} w_{m}) (\operatorname{rot} \operatorname{rot} \xi)_{m}. \quad (A.10)$$

We similarly find that

+

$$A_{s} = -\frac{L}{2I} (\operatorname{rot} \mathbf{w})_{i} - \frac{1}{4} e_{ipq} (\delta_{jq} - w_{j}w_{q}) \frac{\partial}{\partial x_{j}} (\operatorname{rot} \boldsymbol{\xi})_{p},$$

$$A_{4} = \frac{1}{2} (\delta_{ik} - w_{i}w_{k}) \frac{\partial A_{ij}}{\partial x_{j}}, \quad A_{5} = \frac{1}{2} (\delta_{kj} - w_{k}w_{j}) \frac{\partial}{\partial x_{j}} A_{ik}.$$
(A.11)

It should be pointed out that A_1 obeys the Wick theorem, i.e., the average $\langle n_i n_j n_k n_m \rangle$ is assumed to be a sum of all possible pair combinations of this set of four quantities. We can therefore see that Eqs. (A.8)-(A.11) yield the Navier-Stokes equations quoted in the main text.

Finally, averaging of the heat conduction equation requires calculation of the following nontrivial averages:

$$B_{i} = \langle \alpha_{2}n_{i}N_{j} + \alpha_{3}n_{j}N_{i} \rangle,$$

$$B_{2} = \left\langle N_{i}\frac{dn_{j}}{dt} \right\rangle, \quad B_{3} = \left\langle n_{i}A_{ij}\frac{dn_{j}}{dt} \right\rangle.$$
 (A.12)
Following the preceding procedure, we find that

$$B_{1} = \alpha_{2} \langle n_{i} [e_{jpq} (\Omega_{p} - \omega_{p}) n_{q}] \rangle + \alpha_{3} \langle n_{j} [e_{ipq} (\Omega_{p} - \omega_{p}) n_{q}] \rangle$$

$$= (\alpha_{2} - \alpha_{3}) \frac{L}{2I} e_{ijp} w_{p},$$

$$B_{2} = \langle e_{ipq} (\Omega_{p} - \omega_{p}) n_{q} e_{ijk} \Omega_{j} n_{k} \rangle = \frac{L}{I} \left[\frac{L}{I} - \frac{1}{2} \operatorname{w} \operatorname{rot} \xi \right].$$

(A.13)

Next, in the same approximation, we find that $B_3=0.$ (A.14)

The cross terms in the heat conduction equation (29) vanish because of the Onsager relationship $\alpha_2 - \alpha_3 + \gamma_1 = 0$.

In this way Eqs. (A.13) and (A.14) yield the heat conduction equation quoted in the main text.

- ¹⁾ It should be pointed out that at such high rotation velocities ($\sim 10^7$ sec⁻¹) the terms nonlinear in respect of the orbital momentum may be important in the dissipative function and this may change the momentum relaxation law and, consequently, the lifetime of a state of a liquid crystal with a rotating director.
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