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RAYLEIGH INSTABILITY IN LIQUID-CRYSTAL JETS

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Capillary instability of isothermal incompressible liquid-crystal (LC) jets is considered in the framework of linear hydrodynamics of uniaxial nematic LCs. The free boundary conditions with strong tangential anchoring of the director $\mathbf{n}$ at the surface are formulated in terms of the mean surface curvature $\mathcal{H}$ and the Gaussian surface curvature $\mathcal{G}$. The static version of the capillary instability is shown to depend on the elasticity modulus K, the surface tension σ_0, and the radius r_0 of the LC jet, expressed in terms of the characteristic parameter $\varkappa = K/\sigma_0 r_0$. The problem of the capillary instability in LC jets is solved exactly and a dispersion relation, which reflects the effect of elasticity, is derived. It is shown that the increase of the elasticity modulus results in the decrease of both the cut-off wavenumber k and the disturbance growth rate s. This implies an enhanced stability of LC jets, compared to ordinary liquids. In the specific case where the hydrodynamic and orientational LC modes can be decoupled, the dispersion equation is given in a closed form.

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1. INTRODUCTION

The breakup of liquid jets injected through a circular nozzle into stagnant fluids has been the subject of widespread research over the years. Previous studies that followed the seminal works of Lord Rayleigh have established that the complex jet flow is influenced by a large number of parameters. These include nozzle internal flow effects, the jet velocity profile $\mathbf{V}(\mathbf{r})$, and the physical state of both liquid and gas. Although the hydrodynamic equations are nonlinear, the linear stability theory can provide qualitative descriptions of the breakup phenomena and predict the existence of different breakup regimes.

Using a linear theory, Rayleigh showed [1] that the jet breakup is a consequence of the hydrodynamic instability, or more exactly, the capillary instability. Neglecting the effect of the ambient fluid, the viscosity of the jet liquid, and gravity, he demonstrated that a cylindrical liquid jet is unstable with respect to disturbances characterized by wavelengths larger than the jet circumference. Rayleigh also considered the case of a viscous jet in an inviscid gas and an inviscid gas jet in an inviscid liquid [2]. Weber [3] generalized Rayleigh's result to the case of a Newtonian viscous liquid and showed that the viscosity tends to reduce the breakage rate and increase the drop size. Chandrasekhchar [4] considered the effect of a uniform magnetic field on the capillary instability of a liquid jet. A mechanism of bending disturbances and of buckling, slowly moving, highly viscous jets was presented by Taylor [5]. Further developments of the theory in Newtonian liquids was concerned with additional factors such as the dynamic action of the ambient gas (leading to atomization of the jet), the nonlinear interaction of growing modes that lead to satellite drop formation, and the spatial character of instability [6, 7].

The capillary instability in jets comprised of non-Newtonian suspensions and emulsions presents a different category of cases governed by power-law (pseudoplastic and dilatant) liquids. The effective viscosity of the pseudoplastic liquid decreases with the growth of the strain rate, whereas in dilatant liquids, it increases [7]. The behavior of capillary jets of dilute and concentrated polymer solutions suggests a strong influence of the macromolecular coils on their flow patterns [7]. Free jets of polymeric liquids that exhibit oscillations are reported in [8].

The idea of the Rayleigh instability was applied to tubular membranes in dilute lyotropic phases [9]. Their

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relaxation, following optical excitation, is characterized by a long time and can be described by means of the hydrodynamic approach [10]. Bending deformations of such membranes are governed by the Helfrich energy [11], which depends on the curvature of the tube. Thus, a competition between the surface tension and curvature energy of the membrane immersed into water renders the initial shape of the tube unstable. The hydrodynamic formalism used in [10] and the hydrodynamics of fluids with an inner order such as liquid crystals (LCs) [12] have similar features. In [10], the order parameter is a unit vector normal to the membrane surface. In contrast, the order parameter Q of an LC fluid is defined throughout the space it occupies.

The continuum theory of LC phases has emerged as a rigorous part of the condensed matter theory. The hydrodynamics of the nematic LC phases was developed during the 60–70th in the pioneering works of Ericksen [13, 14], Leslie [15, 16], Parodi [17], and the Harvard group [18][1)], and its predictions were successfully confirmed in many experimental observations. The combination of viscous and elastic properties is likely to produce new evolution patterns of hydrodynamic instabilities, in the context of the Benard–Rayleigh, Marangoni, and electrohydrodynamic effects [19], which cannot occur in ordinary liquids. In particular, we refer to nonsteady state (oscillatory) evolution of the instability that appears via the Hopf bifurcation [20]. The instability of an LC jet poses an additional challenge with respect to the effects listed above. This already applies in the framework of the linear stability theory.

The linear analysis of the capillary instability in a thin nematic LC fiber was recently performed in [21] under the assumption that the director field $\mathbf{n}(\mathbf{r})$ is fixed and does not change even if the fiber shape evolves through the linear instability process. In this analysis, the only influence of the LC nematicity is due to the anisotropy of the elastic surface energy and the anisotropy of viscous LC moduli. The above assumption stipulates the predominance of elastic forces over the surface tension, $\ell \gg r_0$, and over hydrodynamic forces, $Er \ll 1$, where $\ell = K/\sigma$ and $Er = \eta V r_0/K$ denote the anchoring extrapolation length [22] and the Ericksen number [19], respectively, η and K are viscous and elastic moduli, V is the LC velocity, and r_0 stands for the geometric length scale, i.e., the radius of the LC jet. The first condition ($\ell \gg r_0$) is difficult to implement for most of the known nematic LCs with well-studied physical parameters. Indeed, the classical nematic LCs, also known as MBBA and PAA, have the anchoring extrapolation length $\ell \approx 3\cdot10^{-10}$ m (Tables 1 and 2). This value indicates strong anchoring at the surface[2)]. Otherwise, the radius of the jet must be decreased to the molecular scale. In the case of strong director anchoring at the surface, the second requirement, $Er \ll 1$, does not allow a continuous transition to ordinary liquids (a classical Rayleigh–Weber theory) which is an important benchmark in the theory. We note that as the elasticity tends to zero, $K \to 0$, then $Er \to \infty$. Moreover, disregarding the bulk elasticity effects in LCs leaves out the competition between the bulk forces and surface tension that is crucial for the physical picture of thin LC films (see Sec. 3). In this context [21], the Leslie–Ericksen equation of angular motion of the director $\mathbf{n}(\mathbf{r}, t)$ was skipped and the elastically induced nondissipative contributions to the Navier–Stokes equation were not included in the LC hydrodynamics.

A more realistic setup of the problem consists of a rigid boundary condition of strong director anchoring at the free surface of LC jets. The simplest case constitutes a tangential orientation of the director at the surface, $\mathbf{n}\cdot\mathbf{e} = 0$, where $\mathbf{e}$ is a unit normal vector to the jet surface. Such orientation, with strong anchoring and temperature independence, is observed at the free surface of the classical nematic PAA mentioned above [24–26]. Assuming that the scale of deformation of the initial surface is much larger than the molecular length of LCs, we conclude that if the orientation of the director $\mathbf{n}$ is set tangential to the undisturbed surface, then it must also remain tangential when the surface is smoothly disturbed.

The elastic properties of LCs are expected to change the evolution patterns of jets that are made from them. In this paper, we derive a rigorous mathematical model of capillary instability for isothermal incompressible nematic LC jets in the single elastic approximation. This model shows how the combined viscous and elastic properties of LC fluids determine the boundary con-

[1)] The name «Harvard group» was proposed by De Gennes [19] and denotes five authors (see [18]).

[2)] On the basis of a heuristic argument, De Gennes [19] noted that if the anisotropic interaction at a nematic–substrate interface is as large as that acting between nematic molecules, the anchoring energy σ can be roughly estimated as $\sigma \sim K/a$, where K is the Frank modulus and a is the molecular dimension; hence, taking $K \approx 8\cdot10^{-12}$ N and $a \approx 5\cdot10^{-10}$ m, we find $\sigma \approx 1.6\cdot10^{-2}$ N/m, which corresponds to the strong anchoring in virtually all practical cases. An extensive review by Cognard [23] lists sixteen most studied nematic LCs with corresponding σ measured at equilibrium with air (see Table 9 in [23]). All values are in the range between $2.45\cdot10^{-2}$ N/m for MPPB and $4\cdot10^{-2}$ N/m for 5CB, which gives a good support to the qualitative consideration of De Gennes.

Table 1. The basic physical parameters α_i, ρ, K, and σ_0 and their derivatives η_i, β_i, γ_i, B_i, μ_i, λ, and ν_i for nematic liquid crystal 4-metoxybenziliden-4-butilanilin (MBBA) at $25\,^\circ$C taken from [23, 30]

α_1, mPa·s	α_2, mPa·s	α_3, mPa·s	α_4, mPa·s	α_5, mPa·s	α_6, mPa·s
7	−78	−1	84	46	−33
η_1, mPa·s	η_3, mPa·s	η_5, mPa·s	λ	μ_1	μ_2
42	50	104	1.026	1.013	0.013
β_1, mPa·s	β_2, mPa·s	β_3, mPa·s	β_4, mPa·s	γ_1, mPa·s	γ_2, mPa·s
42	25	79	59	77	−79
B_1, mPa·s	B_2, mPa·s	B_3, mPa·s	B_4, mPa·s	$\mathcal{B}$	ϑ, m^2/s
58	104	25	78	5.92	$1.2\cdot10^{-10}$
ρ, kg/m^3	K, N	σ_0, N/m	$\ell = K/\sigma_0$, m	ν_i, m^2/s	ϑ/ν_i
$1.2\cdot10^3$	$9\cdot10^{-12}$	$38\cdot10^{-3}$	$2.4\cdot10^{-10}$	10^{-5}–10^{-4}	10^{-6}–10^{-5}

Table 2. The basic physical parameters α_i, ρ, K, and σ_0 and their derivatives η_i, β_i, γ_i, B_i, μ_i, λ, and ν_i for nematic liquid crystal para-azoxyanisole (PAA) at $122\,^\circ$C taken from [23, 30]

α_1, mPa·s	α_2, mPa·s	α_3, mPa·s	α_4, mPa·s	α_5, mPa·s	α_6, mPa·s
4	−6.9	−0.2	6.8	5	−2.1
η_1, mPa·s	η_3, mPa·s	η_5, mPa·s	λ	μ_1	μ_2
3.4	4.5	13.7	1.06	1.03	0.03
β_1, mPa·s	β_2, mPa·s	β_3, mPa·s	β_4, mPa·s	γ_1, mPa·s	γ_2, mPa·s
3.4	2.25	11.45	4.55	6.7	−7.1
B_1, mPa·s	B_2, mPa·s	B_3, mPa·s	B_4, mPa·s	$\mathcal{B}$	ϑ, m^2/s
4.34	9.36	2.26	11.24	7.11	$1.8\cdot10^{-9}$
ρ, kg/m^3	K, N	σ_0, N/m	$\ell = K/\sigma_0$, m	ν_i, m^2/s	ϑ/ν_i
$1.4\cdot10^3$	$11.9\cdot10^{-12}$	$40\cdot10^{-3}$	$3\cdot10^{-10}$	10^{-6}–10^{-5}	10^{-4}–10^{-3}

ditions at the free surface with strong tangential anchoring of the director and the range where instability prevails.

2. HYDRODYNAMICS OF AN LC JET

In this section, we first formulate the problem of capillary instability and then derive the basic equations that govern the linear hydrodynamics of an LC jet. The incompressible flow of a nematic LC is described by a set of differential equations: the continuity equation, the Navier–Stokes equation for viscoelastic LCs, and the Leslie–Ericksen equation of angular motion of the director $\mathbf{n}(\mathbf{r},t)$. They are supplemented by boundary conditions on the LC free surface with strong tangential anchoring of the director.

The basic notation and linear hydrodynamic equations for uniaxial nematic LCs follow the theory given in [18] (the so-called Harvard group approach), which has become standard in many monographs, e.g., [12, 27]. We note that the Harvard group and

Ericksen–Leslie–Parodi approaches are in full agreement (a detailed discussion is given in [19]).

2.1. Basic notation and variables

The following basic variables describe the nematic LC medium: velocity $\mathbf{V}(\mathbf{r},t)$, pressure $P(\mathbf{r},t)$, and director $\mathbf{n}(\mathbf{r},t)$, $\mathbf{n}^2=1$. The initial values of the functions are denoted by «0», either as a subscript or superscript. The following notation, which is commonly accepted in the theory of LCs, is used henceforth:

1. The free energy density E_d of a deformed nonchiral uniaxial nematic LC, given in the quadratic approximation in terms of the derivatives $\partial\mathbf{n}/\partial x_j$ and in the single elastic approximation, has the form

$$E_d=\frac{K}{2}\left(\mathrm{div}^2\mathbf{n}+\mathrm{rot}^2\mathbf{n}\right), \tag{1}$$

where $K\geq 0$ is known as the Frank elasticity modulus. In the vicinity of a phase transition, $K\propto Q^2$ [19], and in the isotropic phase, it vanishes.

2. The bulk molecular field $\mathbf{F}$ and the Ericksen elastic stress tensor τ_{ki}, which set the equilibrium distribution of the $\mathbf{n}$-field in an LC, are determined by the variational derivatives[3)],

$$\mathbf{F}=\mathbf{M}-\mathbf{n}\langle\mathbf{n},\mathbf{M}\rangle,\quad\text{or}\quad F_i=(\delta_{ij}-n_in_j)M_j, \tag{2}$$

where

$$\begin{aligned}M_i&=\frac{\partial}{\partial x_k}\frac{\partial E_d}{\partial(\partial_k n_i)}-\frac{\partial E_d}{\partial n_i},\\ \tau_{ki}&=\frac{\partial E_d}{\partial(\partial_k n_i)},\quad \partial_k=\frac{\partial}{\partial x_k},\end{aligned} \tag{3}$$

i.e.,

$$\begin{aligned}\mathbf{M}=K\Delta_3\mathbf{n},\quad \tau_{ki}=K\,(&\delta_{ki}\mathrm{div}\,\mathbf{n}+(\mathbf{n}\cdot\mathrm{rot}\,\mathbf{n})n_m\epsilon_{mki}+\\ &+[[\mathbf{n}\times\mathrm{rot}\,\mathbf{n}]\times\mathbf{n}]_m\,\epsilon_{mki}),\end{aligned} \tag{4}$$

where ϵ_{mki} is the completely antisymmetric unit tensor of the third rank (the Levi-Civita tensor).

3. If the deviations of the director $\mathbf{n}=\mathbf{n}^0+\mathbf{n}^1$ from its initial orientation $\mathbf{n}^0$ along the z direction are small, then

$$\begin{aligned}&n_x^0=n_y^0=0,\quad n_z^0=1,\\ &1\gg n_x^1,\,n_y^1\gg n_z^1\sim\left(n_x^1\right)^2,\left(n_y^1\right)^2,\end{aligned} \tag{5}$$

and simple algebra yields the linear approximation

$$F_x=K\Delta_3 n_x^1,\quad F_y=K\Delta_3 n_y^1,\quad F_z=0, \tag{6}$$

where Δ_3 is the three-dimensional Laplacian. Similar considerations regarding the Ericksen stress tensor τ_{ki} give

$$\begin{aligned}\tau_{xx}&=\tau_{yy}=\tau_{zz}=K\;\mathrm{div}\;\mathbf{n}^1,\\ \tau_{xy}&=-\tau_{yx}=K\left(\frac{\partial n_y^1}{\partial x}-\frac{\partial n_x^1}{\partial y}\right),\\ \tau_{yz}&=-\tau_{zy}=K\left(\frac{\partial n_z^1}{\partial y}-\frac{\partial n_y^1}{\partial z}\right),\\ \tau_{zx}&=-\tau_{xz}=K\left(\frac{\partial n_x^1}{\partial z}-\frac{\partial n_z^1}{\partial x}\right).\end{aligned} \tag{7}$$

The stresses given by Eqs. (7) do not contribute to the nondissipative stress tensor $T_{ik}^{(r)}$ used in the linear hydrodynamics of LCs (see Eq. (8) below).

4. The reactive (nondissipative) $T_{ik}^{(r)}$ and dissipative $T_{ik}^{(d)}$ stress tensors are defined as

$$\begin{aligned}T_{ik}^{(r)}=-P\delta_{ik}-\tau_{kj}\frac{\partial n_j}{\partial x_i}-\frac{\lambda}{2}(n_iF_k+n_kF_i)+\\ +\frac{1}{2}(n_iF_k-n_kF_i),\end{aligned} \tag{8}$$

$$\begin{aligned}T_{ik}^{(d)}=2\eta_1\Upsilon_{ik}+(\eta_3-2\eta_1)\left(n_i\Upsilon_{kj}n_j+n_k\Upsilon_{ij}n_j\right)+\\ +(2\eta_1+\eta_5-2\eta_3)n_in_kn_jn_m\Upsilon_{jm},\end{aligned} \tag{9}$$

where the antisymmetric Ω_{ik} (vorticity) and symmetric Υ_{ik} parts of the derivative ∂_kV_i are given by

$$\begin{aligned}\Omega_{ik}&=\frac{1}{2}\left(\frac{\partial V_k}{\partial x_i}-\frac{\partial V_i}{\partial x_k}\right),\\ \Upsilon_{ik}&=\frac{1}{2}\left(\frac{\partial V_k}{\partial x_i}+\frac{\partial V_i}{\partial x_k}\right).\end{aligned} \tag{10}$$

Three independent viscous moduli η_j, the kinetic coefficient λ, and the rotational viscosity γ_1 determine the dissipative stress tensor $T_{ik}^{(d)}$, the forth-rank viscosity tensor η_{ikjm}, and the dissipative function D in the absence of heat fluxes,

$$\begin{aligned}&D=\eta_{ikjm}\Upsilon_{ik}\Upsilon_{jm}+\frac{1}{\gamma_1}\mathbf{F}^2,\quad T_{ik}^{(d)}=\eta_{ikjm}\Upsilon_{jm},\\ &\eta_{ikjm}=\eta_1(\xi_{ij}\xi_{km}+\xi_{kj}\xi_{im})+\\ &\quad+\frac{\eta_3}{2}(n_in_j\xi_{km}+n_kn_j\xi_{im}+n_in_m\xi_{kj}+\\ &\quad\quad+n_kn_m\xi_{ij})+\eta_5n_in_kn_jn_m.\end{aligned} \tag{11}$$

The tensor η_{ikjm} consists of three independent uniaxial invariants [12] and is highly symmetric,

[3)] Here and throughout the paper, unless noted otherwise, we apply the summation rule over indices that are repeated in a tensor product, e.g., $a_{ij}b_{jk}=\sum_j a_{ij}b_{jk}$.

$\eta_{ikjm} = \eta_{kimj} = \eta_{jmik}$. The requirement that D is positive becomes

$$\eta_1 \geq 0, \quad \eta_3 \geq 0, \quad \eta_5 \geq 0, \quad \gamma_1 \geq 0. \tag{12}$$

The parameter λ is close to $+1$ or -1 for rod-like or disk-like molecules, respectively. If the liquid is viscoisotropic, then $\lambda = 0$.

5. The hydrodynamic reactive (nondissipative) $\mathbf{m}^{(r)}$ and dissipative $\mathbf{m}^{(d)}$ fields are defined as

$$\begin{aligned} m_i^{(r)} &= -(\mathbf{V}\cdot\nabla_3)n_i + n_k\Omega_{ki} + \lambda\,\xi_{ij}\Upsilon_{jk}n_k, \\ \mathbf{m}^{(d)} &= \frac{1}{\gamma_1}\mathbf{F}, \end{aligned} \tag{13}$$

where ∇_3 is the three-dimensional gradient operator, $(\nabla_3)^2 = \Delta_3$.

6. The surface tension σ of a nematic LC is given by [28]

$$\sigma = \sigma_0 + \sigma_1 \langle \mathbf{n}, \mathbf{e}\rangle^2, \tag{14}$$

where σ_0 and σ_1 are isotropic and anisotropic surface tension moduli respectively, and $\mathbf{e}$ is a unit normal vector to the LC surface.

7. Another system of viscous moduli α_i (called the Leslie viscosities) relate the dissipative and kinetic moduli as[4)]

$$\begin{aligned} &\eta_1 = \alpha_4/2, \quad \lambda = -\gamma_2/\gamma_1, \\ &\eta_5 = \alpha_1 + \alpha_4 + \alpha_5 + \alpha_6, \\ &\gamma_1 = \alpha_3 - \alpha_2, \quad \gamma_2 = \alpha_3 + \alpha_2, \\ &\eta_3 - 2\eta_1 = \alpha_5 + \alpha_2\lambda, \\ &2\eta_1 + \eta_5 - 2\eta_3 = \alpha_1 + \gamma_2^2/\gamma_1, \end{aligned} \tag{15}$$

with the support of the Onsager–Parodi relation [17] $\alpha_3 + \alpha_2 = \alpha_6 - \alpha_5$. In the vicinity of a phase transition, the viscous moduli α_i have different dependences on the order parameter Q: $\alpha_1 \propto Q^2$, $\alpha_2, \alpha_3, \alpha_5, \alpha_6 \propto Q$, and $\alpha_4 \propto Q^0$ [19].

Tables 1 and 2 (see above) summarize viscosities and other physical parameters that characterize the most frequently used and well studied nematic LCs, also known as MBBA and PAA.

2.2. Basic equations

The complete system of hydrodynamic equations for the isothermal incompressible nematic LC reflects the conservation laws of mass and of the linear and angular momenta.

4) The correct expression for η_5 is given in [18].

1. The continuity equation

$$\operatorname{div}\mathbf{V} = 0. \tag{16}$$

2. The Navier–Stokes equation for viscoelastic LC,

$$\rho\frac{\partial V_i}{\partial t} + \rho(\mathbf{V}\cdot\nabla_3)V_i = \frac{\partial}{\partial x_k}\left(T_{ik}^{(r)} + T_{ik}^{(d)}\right). \tag{17}$$

3. The Leslie–Ericksen equation of angular motion of the director $\mathbf{n}(\mathbf{r}, t)$,

$$\frac{\partial \mathbf{n}}{\partial t} = \mathbf{m}^{(r)} + \mathbf{m}^{(d)}. \tag{18}$$

The last equation is written for a negligible specific angular moment of inertia $\mathcal{J}_{LC}$ of the LC, namely, $\mathcal{J}_{LC} \ll \rho r_0^2$, where r_0 is a characteristic size of the system. This is true in our case, where r_0 is the radius of the jet.

We consider an isothermal incompressible jet flowing along the z axis, out of a nozzle at a velocity $\mathbf{V}$. The initial orientation of the director $\mathbf{n}^0$ is assumed collinear with $\mathbf{V}$. Deviations from the initial values of the director and pressure are defined as $\mathbf{n}^1 = \mathbf{n} - \mathbf{n}^0$ and $P_1 = P - P_0$, respectively, where $P_0 = \sigma_0/r_0$ is the unperturbed pressure within the cylindrical jet. In the linear approximation, $|\mathbf{n}^1| \ll 1$, Eqs. (16)–(18) are simplified as

$$\begin{aligned} &\operatorname{div}\mathbf{V} = 0, \quad \rho\frac{\partial V_i}{\partial t} = -\frac{\partial P_1}{\partial x_i} + \frac{\partial T_{ik}^{(d)}}{\partial x_k} + \\ &\qquad + \frac{1-\lambda}{2}n_i^0 \operatorname{div}\mathbf{F} - \frac{1+\lambda}{2}(\mathbf{n}^0\cdot\nabla_3)F_i, \\ &\frac{\partial n_i^1}{\partial t} = n_k^0\Omega_{ki} + \lambda\xi_{ij}^0\Upsilon_{jk}n_k^0 + \frac{1}{\gamma_1}F_i, \\ &\xi_{ij}^0 = \delta_{ij} - n_i^0 n_j^0, \quad i,j,k = x,y,z. \end{aligned} \tag{19}$$

Choosing $n_z^0 = 1$ gives $F_z = 0$, and hence

$$\frac{\partial V_x}{\partial x} + \frac{\partial V_y}{\partial y} + \frac{\partial V_z}{\partial z} = 0, \tag{20}$$

$$\begin{aligned} \rho\frac{\partial V_x}{\partial t} &= -\frac{\partial P_1}{\partial x} + \left(\beta_1\Delta_2 + \beta_2\frac{\partial^2}{\partial z^2}\right)V_x + \\ &\quad + (\beta_2 - \beta_1)\frac{\partial^2 V_z}{\partial x \partial z} - \frac{\lambda+1}{2}\frac{\partial F_x}{\partial z}, \\ \rho\frac{\partial V_y}{\partial t} &= -\frac{\partial P_1}{\partial y} + \left(\beta_1\Delta_2 + \beta_2\frac{\partial^2}{\partial z^2}\right)V_y + \\ &\quad + (\beta_2 - \beta_1)\frac{\partial^2 V_z}{\partial y \partial z} - \frac{\lambda+1}{2}\frac{\partial F_y}{\partial z}, \\ \rho\frac{\partial V_z}{\partial t} &= -\frac{\partial P_1}{\partial z} + \left(\beta_2\Delta_2 + \beta_3\frac{\partial^2}{\partial z^2}\right)V_z - \\ &\quad - \frac{\lambda-1}{2}\left(\frac{\partial F_x}{\partial x} + \frac{\partial F_y}{\partial y}\right), \end{aligned} \tag{21}$$

$$
\begin{aligned}
&\frac{\partial n_x^1}{\partial t} = \frac{\lambda+1}{2}\frac{\partial V_x}{\partial z} + \frac{\lambda-1}{2}\frac{\partial V_z}{\partial x} + \frac{F_x}{\gamma_1},\\
&\frac{\partial n_y^1}{\partial t} = \frac{\lambda+1}{2}\frac{\partial V_y}{\partial z} + \frac{\lambda-1}{2}\frac{\partial V_z}{\partial y} + \frac{F_y}{\gamma_1},\\
&\frac{\partial n_z^1}{\partial t} = 0,
\end{aligned} \tag{22}
$$

where $\Delta_2 = \partial^2/\partial x^2 + \partial^2/\partial y^2$ is the two-dimensional Laplacian, $\beta_1 = \eta_1$, $\beta_2 = \eta_3/2$, $\beta_3 = \eta_5 - \eta_3/2$, and F_x and F_y are given in (6). Because isotropic viscosity implies that $\beta_i = \beta$, the liquid crystals MBBA and PAA mentioned above are clearly far from being isotropic (see Tables 1 and 2 above).

To make the problem more specific and easier to solve, we consider axisymmetric disturbances in the system of a cylindrical LC jet with radius r_0. In this case,

$$
\frac{\partial V_z}{\partial z} + \frac{\partial V_r}{\partial r} + \frac{V_r}{r} = 0, \tag{23}
$$

$$
\rho\frac{\partial V_r}{\partial t} = -\frac{\partial P_1}{\partial r} + \left[\beta_1\left(\Delta_{2c} - \frac{1}{r^2}\right) + \beta_2\frac{\partial^2}{\partial z^2}\right]V_r + {} + (\beta_2-\beta_1)\frac{\partial^2 V_z}{\partial r\partial z} - \mu_1\frac{\partial F_r}{\partial z}, \tag{24}
$$

$$
\rho\frac{\partial V_z}{\partial t} = -\frac{\partial P_1}{\partial z} + \left[\beta_2\Delta_{2c} + \beta_3\frac{\partial^2}{\partial z^2}\right]V_z - {} - \mu_2\left(\frac{\partial F_r}{\partial r} + \frac{F_r}{r}\right), \tag{25}
$$

$$
\gamma_1\frac{\partial n_r^1}{\partial t} = \gamma_1\mu_1\frac{\partial V_r}{\partial z} + \gamma_1\mu_2\frac{\partial V_z}{\partial r} + F_r, \quad n_z^1 = 0, \tag{26}
$$

where

$$
\begin{aligned}
&\Delta_{2c} = \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r},\\
&F_r = K\left(\Delta_{2c} - \frac{1}{r^2} + \frac{\partial^2}{\partial z^2}\right)n_r^1,\\
&\mu_1 = \frac{\lambda+1}{2}, \quad \mu_2 = \frac{\lambda-1}{2}.
\end{aligned} \tag{27}
$$

Equations (23)–(26) describe ordinary linear hydrodynamic behavior of isotropic incompressible liquids if the LC properties vanish: $K, \gamma_1 \to 0$ and $\beta_i = \beta$. The result is the well-known continuity equation and the linearized Navier–Stokes equation,

$$
\operatorname{div}\mathbf{V} = 0, \quad \rho\frac{\partial \mathbf{V}}{\partial t} = -\nabla P_1 + \beta\Delta_3\mathbf{V}. \tag{28}
$$

2.3. Boundary conditions at the free surface

Boundary conditions at the free surface of an LC state that the jump in normal stress consists of two parts: one depends on the surface tension σ and the other on the elastic disturbance W_{elast} of the uniform director field $\mathbf{n}_0(\mathbf{r})$. Assuming that no tangential stresses exist at the free surface, we can express the boundary conditions at $r = r_0$ as

$$
\left(T_{ik}^{(r)} + T_{ik}^{(d)}\right)e_k + (2\sigma\mathcal{H} + W_{elast})\,e_i + \frac{\partial\sigma}{\partial x_i} = 0, \tag{29}
$$

where e_i are the components of the normal unit vector $\mathbf{e}$ in the reference frame of the LC cylinder and $\mathcal{H} = (1/R_1 + 1/R_2)/2$ denotes the mean surface curvature with the principal radii R_1 and R_2.

The nonhydrodynamic part of the boundary conditions with strong tangential anchoring of the director at the free surface holds if the scale of deformation of the initial surface is much larger than the molecular length of LCs[5)]. This determines tangential behavior of a smoothly disturbed director $\mathbf{n}$ at the free surface, $e_z \ll e_r \sim 1$:

$$
\mathbf{e}\cdot\mathbf{n} = 0 \;\to\; e_z + n_r^1 = 0 \quad \text{at} \quad r = r_0. \tag{30}
$$

The last constraint cancels the gradient term in Eq. (29). We finally obtain the boundary conditions in the linear approximation of the variables n_r^1, V_r, V_z, and P_1,

$$
T_{rr}^{(r)} + T_{rr}^{(d)} + 2\sigma\mathcal{H} + W_{elast} = 0, \quad T_{zr}^{(r)} + T_{zr}^{(d)} = 0. \tag{31}
$$

Substitution of the expressions for the reactive and dissipative stress tensors gives

$$
\begin{aligned}
&2\beta_1\Upsilon_{rr} - P_1 = 2\sigma_0(\mathcal{H}_0 - \mathcal{H}) - W_{elast},\\
&2\beta_2\Upsilon_{zr} = \mu_2 F_r \quad \text{at} \quad r = r_0,
\end{aligned} \tag{32}
$$

where $\mathcal{H}_0 = (2r_0)^{-1}$ is the initial mean curvature of the LC cylinder. The equations for the jet surface disturbed by a wave $\zeta(z,t)$ and its radial velocity $\partial\zeta/\partial t$ are given by

$$
r(z,t) = r_0 + \zeta(z,t), \quad V_r = \frac{\partial\zeta}{\partial t} \quad \text{at} \quad r = r_0, \tag{33}
$$

where $\zeta \ll r_0$ is the radial displacement of a surface point. The principal radii of the surface curvature,

[5)] Strictly speaking, this assumption is correct when the equilibrium distribution of the director field $\mathbf{n}(\mathbf{r})$ is free of singularities. The problem of the minimal surface of an LC drop presents another situation where an essential rearrangement of the field $\mathbf{n}(\mathbf{r})$ at the surface can decrease the total energy by destroying the disclination core within the drop.

in the linear approximation with respect to ζ, and its derivatives can be expressed as

$$\frac{1}{R_1} = \frac{1}{r_0+\zeta} \approx \frac{1}{r_0} - \frac{\zeta}{r_0^2}, \quad \frac{1}{R_2} \approx -\frac{\partial^2\zeta}{\partial z^2}. \tag{34}$$

This transforms the boundary conditions given by Eqs. (30) and (32) into

$$n_r^1 = \frac{\partial\zeta}{\partial z}, \quad V_r = \frac{\partial\zeta}{\partial t}, \tag{35}$$

$$2\beta_2\Upsilon_{zr} = \mu_2 F_r, \tag{36}$$

$$P_1 - 2\beta_1\Upsilon_{rr} = -\sigma_0\left(\frac{\zeta}{r_0^2} + \frac{\partial^2\zeta}{\partial z^2}\right) + W_{elast}. \tag{37}$$

The term W_{elast} deserves further discussion. It reflects the existence of normal stresses at the surface, which arise due to the resistance of the uniformly orientated continuos LC media to the surface disturbance. The term W_{elast} vanishes in undisturbed LC jets and depends linearly on the elastic modulus K, the radius r_0, and the derivatives of ζ. Moreover, the invariance of the problem under inversion of the z axis requires dependence on derivatives of only even orders. An explicit expression for W_{elast} is derived in Sec. 3.1.

3. PLATEAU INSTABILITY IN AN LC CYLINDER

Before considering the sophisticated mathematics of Eqs. (23)–(26) supplemented by boundary conditions (35)–(37), we discuss capillary instability of the LC cylinder. This is done by applying the Plateau considerations [29] on shape of a liquid mass withdrawn from the action of gravity.

We consider an LC cylinder with the surface disturbed in accordance with (33), where $\zeta = \zeta_0\cos kz$, ζ_0 is small compared to r_0, and $k = 2\pi/\Lambda$, with Λ being the disturbance wavelength. The idea of Plateau, applied here, is to find the cut-off wavelength Λ_s of the disturbance that determines breakage of the cylinder into droplets with due decrease of the total energy.

The average volume v over one wavelength Λ in the z direction is given by

$$v = \frac{1}{\Lambda}\int_0^\Lambda dz \int_s ds = \pi\left(r_0^2 + \frac{1}{2}\zeta_0^2\right) \quad \to$$
$$\to \quad r_0 = \sqrt{\frac{v}{\pi}}\left(1 - \frac{1}{4}\frac{\pi\zeta_0^2}{v}\right), \tag{38}$$

where r_0 in the right-hand side is given as a second-order expansion in ζ_0. The total energy $\mathcal{E}$ of the LC cylinder per unit wavelength with a disturbed director field $\mathbf{n}(\mathbf{r})$ is given by

$$\mathcal{E} = \sigma_0\int_s ds + \frac{K}{2}\int_0^\Lambda dz\int_s\left(\mathrm{div}^2\mathbf{n} + \mathrm{rot}^2\mathbf{n}\right)ds. \tag{39}$$

The static director field $\mathbf{n}(\mathbf{r})$ can be found from Eq. (27) and the associated boundary condition (35),

$$\begin{gathered} n_z^0 = 1, \quad F_r = 0 \quad \to \quad \left(\Delta_{2c} - \frac{1}{r^2} + \frac{\partial^2}{\partial z^2}\right)n_r^1 = 0, \\ n_r^1 = \frac{\partial\zeta}{\partial z} \quad \text{at} \quad r = r_0. \end{gathered} \tag{40}$$

Equation (40) has the solution

$$n_r^1(r,z) = -\frac{k\zeta_0}{I_1(kr_0)}I_1(kr)\sin kz, \tag{41}$$

which is finite at $r = 0$, where $I_m(x)$ is the modified Bessel function of order m. The contribution of elastic forces is determined by

$$\mathrm{div}^2\mathbf{n} + \mathrm{rot}^2\mathbf{n} = k^2\left[\frac{k\zeta_0}{I_1(kr_0)}\right]^2 \times \\ \times\left[A_1^2(kr)\sin^2 kz + A_2^2(kr)\cos^2 kz\right], \tag{42}$$

where

$$A_1(q) = \frac{dI_1(q)}{dq} + \frac{1}{q}I_1(q), \quad A_2(q) = I_1(q).$$

Simple integration of Eq. (39) gives

$$\mathcal{E} = 2\pi\sigma_0 r_0\left(1 + \frac{1}{4}k^2\zeta_0^2\right) + \frac{\pi}{2}K\left[\frac{k\zeta_0}{I_1(kr_0)}\right]^2 \times \\ \times\int_0^{kr_0}\left[A_1^2(q) + A_2^2(q)\right]q\,dq. \tag{43}$$

Inserting r_0 from Eq. (38) in the first term above, we obtain

$$\mathcal{E} - 2\sigma_0\sqrt{\pi v} = \sigma_0\frac{\pi\zeta_0^2}{2r_0}\left(\varpi^2 - 1\right) + \frac{\pi}{2}K\left[\frac{\zeta_0\varpi}{r_0 I_1(\varpi)}\right]^2 \times \\ \times\int_0^{\varpi}\left[A_1^2(q) + A_2^2(q)\right]q dq, \quad \varpi = kr_0. \tag{44}$$

The positive root $\varpi_s = k_s r_0$ of the expression in the right-hand side of Eq. (44) determines the cut-off wavelength Λ_s of capillary disturbances that renders the LC cylinder unstable.

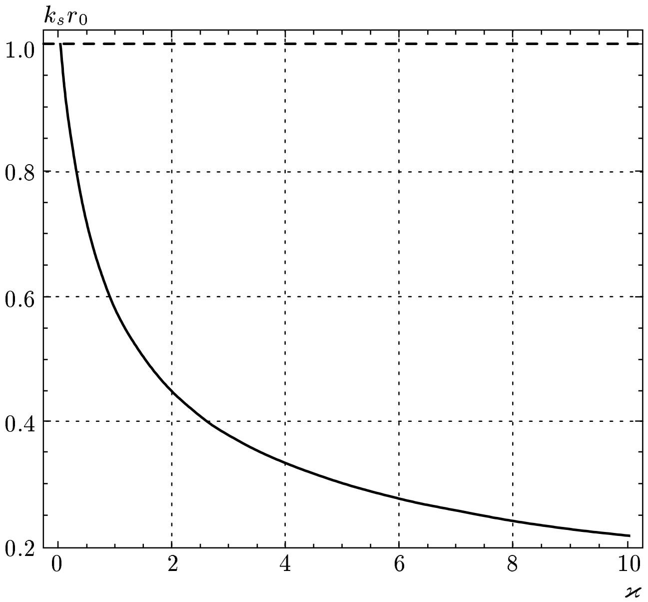

Fig. 1. Universal plots of $k_s r_0$ versus $\varkappa$ for the Plateau instabilities in an LC cylinder (solid line), and in ordinary liquid, $k_s r_0 = 1$ (dashed line)

The quadratic approximation with respect to the derivatives $\partial \mathbf{n}/\partial x_j$ in Eq. (1), which provides the basis for the Frank theory, makes expression (44) correct only in terms of the ϖ^2 approximation. Indeed, the power of ϖ in Eq. (44) must not exceed 2, otherwise the calculation becomes inconsistent. We thus obtain

$$\begin{gathered} \mathcal{E} - 2\sigma_0\sqrt{\pi v} = \sigma_0 \frac{\pi\zeta_0^2}{2r_0}\left(\varpi^2 - 1\right) + \pi K k^2 \zeta_0^2, \\ \varpi_s = \frac{1}{\sqrt{1+2\varkappa}}, \quad \varkappa = \frac{K}{\sigma_0 r_0}, \end{gathered} \tag{45}$$

where the subscript «s» denotes the static nature of the Plateau instability. The asymptotic behavior of $\varpi_s(\varkappa)$ shows two important limits:

$$\begin{gathered} \varpi_s = 1 - \varkappa \quad \text{if} \quad \varkappa \ll 1, \\ \varpi_s = \frac{1}{\sqrt{2\varkappa}}\left(1 - \frac{1}{4\varkappa}\right) \quad \text{if} \quad \varkappa \gg 1. \end{gathered} \tag{46}$$

Figure 1 shows a plot of $k_s r_0$ versus $\varkappa$ for the Plateau instabilities in the LC and in ordinary liquid.

The corresponding asymptotic cut-off wavelength Λ_s is obtained as

$$\begin{gathered} \Lambda_s = 2\pi r_0 \left(1 + \varkappa\right) \quad \text{if} \quad \varkappa \ll 1, \\ \Lambda_s = 2\pi\sqrt{\frac{2K}{\sigma_0}}\,\sqrt{r_0}\left(1 + \frac{1}{4\varkappa}\right) \quad \text{if} \quad \varkappa \gg 1. \end{gathered} \tag{47}$$

This result shows that $k \geq k_s$ increases the total energy $\mathcal{E}$ of the disturbed system, whereas $k < k_s$ decreases it. According to (46), there are two marginal regimes of instability.

1) *The capillary regime* $r_0 \gg K/\sigma_0$. Here, Λ_s is close to the circumference of the cylinder and the elastic deformation contribution $\int E_d dv$ to the total energy $\mathcal{E}$ is negligible. This regime must apply to a wide range of nematic LCs, because the common values of $K \sim 10^{-11}$ J/m [19] and $\sigma_0 \sim 10^{-2}$ J/m^2 [23] lead to $K/\sigma_0 \sim 10^{-9}$ m. This value is evidently smaller than the presently attainable radii of the jet.

2) *The elastic regime* $r_0 \ll K/\sigma_0$. This case reflects the dominance of elastic deformation and predicts an unusual behavior for $\Lambda_s \propto \sqrt{r_0}$. This regime cannot be reached by a simple increase of the elastic moduli because their magnitude is determined by $K \sim \kappa_B T/a$, where $\kappa_B T \approx 4 \cdot 10^{-21}$ J is the Boltzmann thermal energy at room temperature, and $a \approx 5 \cdot 10^{-10}$ m is the molecular length of the LC. In contrast, the effect of surface tension can be diminished by surfactants or by charging the surface of the liquid. In the latter case, the charge can virtually eliminate the effect of surface tension and provide the conditions where the elastic forces predominate.

3.1. W_{elast} and the Gaussian surface curvature

A straightforward way to derive an expression for W_{elast} is to solve the elastic problem for the stresses existing on a deformed axisymmetric surface of an LC cylinder. This is related to the Plateau instability, which obviates the need to repeat the entire procedure.

When we turn from Plateau considerations regarding the static instability of LC cylinders to the capillary instability of LC jets, the question is whether the cut-off wavelengths of the static (Λ_s) and hydrodynamic (Λ_d) problems coincide. This question was skipped by Rayleigh in his studies of isotropic viscous liquids, because the cut-off wavelengths always coincide for ordinary liquids, $\Lambda_s \equiv \Lambda_d$. This identity reflects a deep equivalence principle of the bifurcation point for a nontrivial steady state of a dynamic system and the threshold of static instability related to a minimum of its free energy $\mathcal{E}$ [4].

Using that $\Lambda_s \equiv \Lambda_d$, we construct the term W_{elast} that enters boundary condition (37). For this, we examine and represent the total energy (45) as

$$\mathcal{E} - 2\sigma_0\sqrt{\pi v} = \\ = \frac{\pi\zeta_0 r_0}{2}\left[-\sigma_0\left(\frac{\zeta_0}{r_0^2} - \zeta_0 k^2\right) + 2K\frac{\zeta_0}{r_0}k^2\right]. \tag{48}$$

Next, we compare the expression in the brackets with the right-hand side of Eq. (37). This gives W_{elast} that generates the elastic contribution in (48),

$$W_{elast} = 2K\mathcal{G}, \quad \mathcal{G} = \frac{1}{R_1 R_2} = -\frac{1}{r_0}\frac{\partial^2 \zeta}{\partial z^2}, \tag{49}$$

where $\mathcal{G}$ is the Gaussian surface curvature in accordance with (34). Thus, the final expression for boundary conditions (29) is based on two fundamental invariants of the surface curvature, the mean surface curvature $\mathcal{H}$ and the Gaussian surface curvature $\mathcal{G}$.

4. DISPERSION RELATION

Rayleigh was the first to observe [1] that the instability problem is not so definite, contrary to the Plateau theory. The mode whereby a system deviates from unstable equilibrium must depend on the nature and characteristics of small displacements to which this system is subjected. In the absence of such displacement, any system, however unstable, cannot depart from equilibrium. These characteristics, being hydrodynamic, reflect the effect of viscosity, which predominates over inertia. For ordinary liquids, the mode of the maximum instability, which corresponds to the wavelength $\Lambda_R = 4.508 \cdot 2r_0$, exceeds the circumference of the liquid cylinder. We anticipate that the instability of LC jets has similar features.

The fact that the velocity potential does not exist in an anisotropic viscoelastic liquid dictates a standard approach to this problem that was first elaborated by Rayleigh [2]. We define the Stokes stream function $\Psi(\mathbf{r}, t)$ and the director potential $\Theta(\mathbf{r}, t)$ as

$$V_r = -\frac{1}{r}\frac{\partial \Psi}{\partial z}, \quad V_z = \frac{1}{r}\frac{\partial \Psi}{\partial r}, \quad n_r^1 = \frac{\partial \Theta}{\partial r}, \tag{50}$$

such that continuity equation (23) holds. From the other three equations, (24)–(26), we have

$$\frac{\partial P_1}{\partial r} = (\beta_2 - \beta_1)\frac{\partial^2}{\partial r \partial z}\left(\frac{1}{r}\frac{\partial \Psi}{\partial r}\right) - \frac{1}{r} \times \\ \times \frac{\partial}{\partial z}\left[\beta_1 r \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \Psi}{\partial r}\right) + \beta_2 \frac{\partial^2 \Psi}{\partial z^2} - \rho \frac{\partial \Psi}{\partial t} + \mu_1 r F_r\right], \tag{51}$$

$$\frac{\partial P_1}{\partial z} = \frac{1}{r} \times \\ \times \frac{\partial}{\partial r}\left[\beta_2 r \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \Psi}{\partial r}\right) + \beta_3 \frac{\partial^2 \Psi}{\partial z^2} - \rho \frac{\partial \Psi}{\partial t} - \mu_2 r F_r\right], \tag{52}$$

$$\frac{\partial^2 \Theta}{\partial r \partial t} = \frac{1}{r}\left[\mu_2 r \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \Psi}{\partial r}\right) - \mu_1 \frac{\partial^2 \Psi}{\partial z^2}\right] + \frac{1}{\gamma_1} F_r, \\ F_r = K\left(\Delta_{2c} + \frac{\partial^2}{\partial z^2} - \frac{1}{r^2}\right)\frac{\partial \Theta}{\partial r}. \tag{53}$$

Applying the commutation rules gives

$$\left(\Delta_{2c} - \frac{1}{r^2}\right)\frac{\partial \Theta}{\partial r} = \frac{\partial}{\partial r}\Delta_{2c}\Theta \quad \to \quad F_r = \\ = K\frac{\partial}{\partial r}\left(\Delta_{2c} + \frac{\partial^2}{\partial z^2}\right)\Theta,$$

which facilitates simplification of the above equations. Assuming that an axisymmetric disturbance characterized by the wavelength $2\pi/k$ increases exponentially in time with the growth rate s gives

$$\{\Psi, \Theta, \zeta, P_1, F_r\} = \\ = \{i\psi(r), i\theta(r), \varsigma(r), p(r), if(r)\} e^{st+ikz}. \tag{54}$$

Inserting (54) in (51)–(53) leads to the amplitude equations

$$\frac{1}{k}\frac{\partial p}{\partial r} = \beta_4 \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \psi}{\partial r}\right) - (\beta_2 k^2 + s\rho)\frac{\psi}{r} + \mu_1 f, \\ \beta_4 = 2\beta_1 - \beta_2, \tag{55}$$

$$kp = \frac{1}{r} \times \\ \times \frac{\partial}{\partial r}\left\{r\left[\beta_2 \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \psi}{\partial r}\right) - (\beta_3 k^2 + s\rho)\frac{\psi}{r} - \mu_2 f\right]\right\}, \tag{56}$$

$$s\frac{\partial \theta}{\partial r} = \mu_2 \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \psi}{\partial r}\right) + \mu_1 k^2 \frac{\psi}{r} + \frac{1}{\gamma_1} f, \\ f = K\frac{\partial}{\partial r}\left(\Delta_{2c} - k^2\right)\theta. \tag{57}$$

The new variables in (54) require reformulating boundary conditions (35)–(37) as

$$k\varsigma = \frac{\partial \theta}{\partial r}, \quad s\varsigma = k\frac{\psi}{r}, \\ \frac{\mu_2}{\beta_2} f = \frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial \psi}{\partial r}\right) + k^2 \frac{\psi}{r}, \\ p = 2\beta_1 k \frac{\partial}{\partial r}\left(\frac{\psi}{r}\right) + \varsigma\Gamma, \tag{58}$$

where

$$\Gamma = \sigma_0\left(k^2 - \frac{1}{r_0^2}\right) + 2K\frac{1}{r_0}k^2.$$

The real forms of amplitude equations (55)–(57) and boundary conditions (58) imply that expression (54) divides the five variables into two groups: P_1, ζ and Ψ, Θ, F_r. These groups are shifted with respect to each other by the phase angle $\pi/2$.

4.1. Reduction of the amplitude equations

In this section, we perform the standard procedure for the simplification of amplitude equations (55)–(57). Substituting f from (57) in the other amplitude equations, we obtain

$$\frac{1}{k}\frac{\partial p}{\partial r} = B_1\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial\psi}{\partial r}\right) - \left(B_2 k^2 + s\rho\right)\frac{\psi}{r} + s\gamma_1\mu_1\frac{\partial\theta}{\partial r}, \quad (59)$$

$$kp = \frac{1}{r}\frac{\partial}{\partial r}\left\{r\left[B_3\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial\psi}{\partial r}\right) - (B_4k^2+s\rho)\frac{\psi}{r}\right]\right\} - s\gamma_1\mu_2\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\theta}{\partial r}\right), \quad (60)$$

$$0 = \mu_2\frac{\partial}{\partial r}\left(\frac{1}{r}\frac{\partial\psi}{\partial r}\right) + \mu_1k^2\frac{\psi}{r} + \frac{K}{\gamma_1}\frac{\partial}{\partial r}\times \times\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\theta}{\partial r}\right) - \left(k^2+\frac{s\gamma_1}{K}\right)\theta\right], \quad (61)$$

where

$$B_1 = \beta_4 - \gamma_1\mu_1\mu_2, \quad B_2 = \beta_2 + \gamma_1\mu_1^2, \quad B_3 = \beta_2 + \gamma_1\mu_2^2, \quad B_4 = \beta_3 - \gamma_1\mu_1\mu_2, \quad (62)$$

and $B_2 > 0, B_3 > 0$ by virtue of conditions (12). Let a new stream function χ be defined as $\psi = r\,\partial\chi/\partial r$. The orientational ($\vartheta$) and kinematic ($\nu_i$) viscosities, as well as the other auxiliary functions, are defined by the relations

$$\vartheta = \frac{K}{\gamma_1}, \quad \nu_i = \frac{B_i}{\rho}, \quad u_i^2 = k^2 + \frac{s}{\nu_i}, \quad w^2 = k^2 + \frac{s}{\vartheta}, \quad \frac{\vartheta}{\nu_i} \ll 1 \quad\to\quad u_i^2 \le w^2, \quad (63)$$

where the first inequality in (63) applies to the known nematic LC fluids (see Tables 1 and 2). Using the new notation, we find the first integrals of the amplitude equations,

$$\frac{p}{k} = \left(B_1\Delta_{2c} - B_2u_2^2\right)\chi + s\gamma_1\mu_1\theta, \quad (64)$$

$$kp = \left(B_3\Delta_{2c} - B_4u_4^2\right)\Delta_{2c}\chi - s\gamma_1\mu_2\Delta_{2c}\theta, \quad (65)$$

$$0 = \left(\mu_2\Delta_{2c} + \mu_1k^2\right)\chi + \vartheta\left(\Delta_{2c} - w^2\right)\theta. \quad (66)$$

Next, we eliminate the pressure amplitude p from Eqs. (64) and (65). This gives

$$\left[B_3\Delta_{2c}^2 - \left(B_1k^2 + B_4u_4^2\right)\Delta_{2c} + B_2u_2^2k^2\right]\chi - s\gamma_1\left(\mu_2\Delta_{2c} + \mu_1k^2\right)\theta = 0, \quad (67)$$

$$\left(\mu_2\Delta_{2c} + \mu_1k^2\right)\chi + \vartheta\left(\Delta_{2c} - w^2\right)\theta = 0. \quad (68)$$

Diagonalizing the matrix of operators in (67) and (68), we obtain homogeneous equations for the functions $\chi(r)$ and $\theta(r)$,

$$\left[D_3\Delta_{2c}^3 - D_2\Delta_{2c}^2 + D_1\Delta_{2c} - D_0\right]\begin{pmatrix}\chi\\ \theta\end{pmatrix} = \begin{pmatrix}0\\ 0\end{pmatrix}, \quad (69)$$

where

$$\begin{aligned} D_0 &= k^2\left(\vartheta B_2u_2^2w^2 - s\gamma_1\mu_1^2k^2\right),\\ D_1 &= \vartheta\left(B_1k^2w^2 + B_2k^2u_2^2 + B_4w^2u_4^2\right) + 2s\gamma_1\mu_1\mu_2k^2, \quad (70)\\ D_2 &= \vartheta\left(B_1k^2 + B_3w^2 + B_4u_4^2\right) - s\gamma_1\mu_2^2,\\ D_3 &= \vartheta B_3. \end{aligned}$$

It is easy to verify that all the coefficients D_j are positive if the conditions $B_i > 0$ and $\mu_2 \ll 1$, $\vartheta/\nu_i \ll 1$ are satisfied (for all i). The latter conditions are in a good agreement with numerous observations in nematic LCs [19].

Factoring the polynomial differential operator further (recalling that $D_3 > 0$) gives

$$D_3\Delta_{2c}^3 - D_2\Delta_{2c}^2 + D_1\Delta_{2c} - D_0 = D_3\left(\Delta_{2c} - m_1^2\right)\left(\Delta_{2c} - m_2^2\right)\left(\Delta_{2c} - m_3^2\right). \quad (71)$$

Equation (71) facilitates finding the finite solutions of Eq. (69),

$$\chi(r) = \sum_{j=1}^{3}\frac{C_j}{m_j}I_0(m_jr), \quad \theta(r) = \sum_{j=1}^{3}\frac{G_j}{m_j}I_0(m_jr), \quad (72)$$

where the second fundamental solutions that diverge at $r = 0$ were excluded, C_j and G_j are indeterminate coefficients, and m_j^2 are three generic[6)] roots of the cubic equation

$$D_3m^6 - D_2m^4 + D_1m^2 - D_0 = 0 \quad\to\quad \to\ \sum_{j=1}^{3}m_j^2 = \frac{D_2}{D_3}, \quad \sum_{j\ne k}^{3}m_j^2m_k^2 = \frac{D_1}{D_3}, \quad \prod_{j=1}^{3}m_j^2 = \frac{D_0}{D_3}. \quad (73)$$

[6)] The freedom to choose the physical parameters of the LC seems to admit a degeneration of cubic equation (73), when some of the roots m_j^2 can coincide in different ways. This coincidence is not important because it can occur only at specific wave vectors k^* on which the coefficients D_2, D_1, and D_0 depend. On the other hand, this kind of degeneration might be interesting if k^* is accidentally close to the cut-off wave vector k_d at which the breakage of the LC jet develops.

The coefficients G_j can be expressed through C_j after inserting solutions (72) in Eq. (68):

$$G_j = \frac{1}{\vartheta} g_j C_j, \qquad g_j = \frac{\mu_1 k^2 + \mu_2 m_j^2}{w^2 - m_j^2}, \quad j = 1, 2, 3. \tag{74}$$

The amplitude of the pressure $p(r)$, the stream function $\psi(r)$, and the displacement $\varsigma(r_0)$ of a point on the surface are easily found from Eqs. (57), (64), (68), and (74) as

$$\begin{aligned} p(r) &= k \sum_{j=1}^{3} \frac{l_j}{m_j} C_j I_0(m_j r), \\ l_j &= B_1 m_j^2 - B_2 u_2^2 + \frac{s}{\vartheta} \gamma_1 \mu_1 g_j, \\ \psi(r) &= r \sum_{j=1}^{3} C_j I_1(m_j r), \\ \varsigma(r_0) &= \frac{1}{\vartheta k} \sum_{j=1}^{3} g_j C_j I_1(m_j r_0), \quad j = 1, 2, 3. \end{aligned} \tag{75}$$

Before proceeding, we discuss the distribution of the roots m_j^2 of cubic equation (73) in the complex plane.

First, m_1^2 is always positive because $D_j > 0$, as mentioned above and as follows from the Descartes rule of sign interchange in the sequence of coefficients for real algebraic equations. The other two roots $m_{2,3}^2$ are either positive or complex conjugate with positive real parts. The last case leads to Bessel functions of complex arguments in (72). This fact can indicate that the separation of the two groups of functions P_1, ζ and Ψ, Θ, F_r by the phase angle $\pi/2$ is more elaborate than assumed in (54). Another consequence of the existence of complex conjugate roots m_j^2, which is more important from the physical standpoint, is the appearance of imaginary contributions to the dispersion equation. This can lead to a complex value of the growth rate $s = \overline{s} + i\omega$ as its solution and to the nonsteady (oscillatory) evolution of the jet, e.g.,

$$\zeta(z, t) \propto \varsigma(r_0) e^{\overline{s}t} \cdot e^{i(\omega t + kz)},$$

where ω is the frequency of oscillations.

4.2. Dispersion equation

In what follows, we derive the dispersion equation $s = s(kr_0)$ that determines the evolution of the Rayleigh instability in LC jets. The revised version of boundary conditions (58) at $r = r_0$, which utilizes the new stream function $\chi(r)$, is given by

$$\begin{aligned} s\frac{\partial \theta}{\partial r} &= k^2 \frac{\partial \chi}{\partial r}, \\ s\gamma_1 \mu_2 \frac{\partial \theta}{\partial r} &= B_3 \frac{\partial}{\partial r} \Delta_{2c} \chi + B_5 k^2 \frac{\partial \chi}{\partial r}, \\ \frac{s}{k} p &= 2s\beta_1 \frac{\partial^2 \chi}{\partial r^2} + \Gamma \frac{\partial \chi}{\partial r}, \end{aligned} \tag{76}$$

where $B_5 = \beta_2 + \gamma_1 \mu_1 \mu_2$. Substituting (72) and (75) in (76) and eliminating the coefficients C_1, C_2, and C_3 from the linear equations leads to a (3×3)-determinant equation

$$\det S_{ij} = 0, \tag{77}$$

where

$$\begin{aligned} S_{1j} &= k^2 - \frac{s}{\vartheta} g_j, \\ S_{2j} &= B_3 m_j^2 + B_5 k^2 - \frac{s}{\vartheta} \gamma_1 \mu_2 g_j, \\ S_{3j} &= \Gamma - s \left[\frac{l_j}{m_j} \frac{I_0(m_j r_0)}{I_1(m_j r_0)} - 2\beta_1 m_j \frac{I_1'(m_j r_0)}{I_1(m_j r_0)} \right], \end{aligned} \tag{78}$$

and $I_1'(y) = dI_1(y)/dy$. Equation (77) is an implicit form of the exact dispersion relation, which is highly complex and cannot be solved analytically in the general case. Nevertheless, here we can verify that the cut-off wavelength Λ_d coincides with Λ_s obtained from the Plateau theory. Indeed, the cut-off regime corresponds to boundary conditions (76) when $s = 0$ and is satisfied for $\Gamma = 0$, i.e., $\Lambda_d = \Lambda_s$. The implications of Eq. (77) can be extended further, for the study of different modes of the LC flow, including oscillations, and in order to describe asymptotic behavior of LC jets. This is outside the scope of this paper. In the next section, we consider the case that facilitates decoupling of hydrodynamic and orientational modes, and consequently the solution of the Rayleigh instability problem in a closed form.

5. DECOUPLING OF HYDRODYNAMIC AND ORIENTATIONAL MODES

In this section, we discuss the case where dispersion equation (77) becomes solvable. Here, we encounter another problem: the elasticity of the LC and anisotropy of its viscous properties have the same origin and cannot therefore be considered separately. Nevertheless, we investigate the case where dispersion equation (77) can be simplified. The large number of physical parameters involved (three viscous moduli, two kinetic coefficients, λ and γ_1, orientational (ϑ) and kinematic (ν_i)

viscosities, and the dimensionless parameter $\varkappa$) call for such a treatment.

We consider the LC with rod-like molecules ($\lambda \approx 1$) and low orientational viscosity ϑ,

$$\mu_1 \approx 1, \quad \mu_2 \approx 0, \quad \vartheta \ll \nu_i, \quad k^2 \ll \frac{s}{\vartheta}, \tag{79}$$

where the first three relations apply to the known nematic LC fluids (see Tables 1 and 2). The last inequality in (79) applies to the low-viscosity limit, which was considered for the kinematic viscosity in ordinary liquids by Rayleigh [1].

In this case, characteristic equation (73) reduces to

$$\begin{gathered} m^6 - \frac{s}{\vartheta} m^4 + \frac{s}{\vartheta}\left(\mathcal{B}k^2 + \frac{s}{\overline{\nu_2}}\right) m^2 - \\ - \frac{s}{\vartheta} k^2 \left(k^2 + \frac{s}{\overline{\nu_2}}\right) = 0, \\ \overline{\nu_i} = \frac{\beta_i}{\rho}, \quad \mathcal{B} = \frac{\beta_3 + \beta_4}{\beta_2}. \end{gathered} \tag{80}$$

The three roots m_j^2 of Eq. (73) become

$$\begin{gathered} 2m_{1,2}^2 = \mathcal{B}k^2 + \frac{s}{\overline{\nu_2}} \pm \\ \pm \sqrt{\left(\mathcal{B}^2 - 4\right) k^4 + 2\left(\mathcal{B} - 2\right) k^2 \frac{s}{\overline{\nu_2}} + \left(\frac{s}{\overline{\nu_2}}\right)^2}, \\ m_3^2 = \frac{s}{\vartheta}. \end{gathered} \tag{81}$$

A simple analysis of the last expression shows that the dimensionless parameter $\mathcal{B}$ has the critical value 2 that separates two different evolution scenarios of the LC jet. If $\mathcal{B} > 2$, both roots, m_1^2 and m_2^2, are positive and the capillary instability always appears via trivial bifurcation (steady-state instability). This scenario applies to MBBA and PAA liquid crystals with $\mathcal{B}_{MBBA} = 5.92$ and $\mathcal{B}_{PAA} = 7.11$ (see Tables 1 and 2). In the opposite case, $\mathcal{B} < 2$, we can find the regime where the above roots are complex conjugate. This leads to the oscillatory evolution of the jet, which appears via Hopf bifurcation (see Sec. 4.1).

Significant simplification can be obtained if we assume degeneration of the three viscosities at the critical value $\mathcal{B}_* = 2$. Indeed, if the viscous moduli β_j satisfy the relation

$$\mathcal{B}_*(\beta_j) = 2 \quad \rightarrow \quad 2\beta_1 + \beta_3 = 3\beta_2, \tag{82}$$

the three roots m_j^2 of Eq. (73) are

$$m_{1*}^2 = k^2, \quad m_{2*}^2 = k^2 + \frac{s}{\overline{\nu_2}}, \quad m_3^2 = \frac{s}{\vartheta}. \tag{83}$$

We note that relation (82) cancels the last term in (9). Expressions (83) indicate that the problem is decomposed into two parts, or, in other words, the cross-terms in Eqs. (67) and (68) are dropped. Thus, the first part of the problem is associated with the Rayleigh instability, described by

$$\left(\Delta_{2c} - m_{1*}^2\right)\left(\Delta_{2c} - m_{2*}^2\right)\chi = 0, \tag{84}$$

with boundary conditions that account for elasticity,

$$\begin{gathered} \frac{\partial}{\partial r}\Delta_{2c}\chi + k^2 \frac{\partial \chi}{\partial r} = 0, \\ \frac{s}{k}\, p = 2s\beta_1 \frac{\partial^2 \chi}{\partial r^2} + \Gamma \frac{\partial \chi}{\partial r} \quad \text{at} \quad r = r_0. \end{gathered} \tag{85}$$

The second part is associated with an orientational instability of the director field $\mathbf{n}(\mathbf{r}, t)$,

$$\left(\Delta_{2c} - m_3^2\right)\theta = 0,$$

with the boundary condition

$$s \frac{\partial \theta}{\partial r} = k^2 \frac{\partial \chi}{\partial r} \quad \text{at} \quad r = r_0. \tag{86}$$

The solutions of Eqs. (84) and (86) are

$$\begin{gathered} \chi(r) = \frac{c_1}{m_{1*}} I_0(m_{1*}r) + \frac{c_2}{m_{2*}} I_0(m_{2*}r), \\ \theta(r) = \frac{c_3}{m_3} I_0(m_3 r). \end{gathered} \tag{87}$$

With these solutions, the hydrodynamic pressure $p(r)$, stream function $\psi(r)$, and surface displacement $\varsigma(r_0)$ are obtained as

$$p(r) = -c_1 s\rho I_0(m_{1*}r),$$

$$\psi(r) = r\left[c_1 I_1(m_{1*}r) + c_2 I_1(m_{2*}r)\right],$$

$$\varsigma(r_0) = \frac{c_3}{k} I_1(m_3 r_0),$$

where the only indeterminates are c_1 and c_2, while c_3 can be expressed as their linear combination,

$$c_3\, \frac{s}{k^2} = c_1 \frac{I_1(m_{1*}r_0)}{I_1(m_3 r_0)} + c_2 \frac{I_1(m_{2*}r_0)}{I_1(m_3 r_0)}, \tag{88}$$

provided that $s = s(kr_0)$ satisfies the dispersion relation that follows from (85) and (87),

$$\begin{gathered} s^2 + \frac{2\overline{\nu_1}k^2}{I_0(kr_0)} \times \\ \times \left[I_1'(kr_0) - \frac{2km_{2*}}{k^2 + m_{2*}^2}\, \frac{I_1(kr_0)}{I_1(m_{2*}r_0)} I_1'(m_{2*}r_0)\right] s = \\ = \frac{\sigma_0 k}{\rho r_0^2}\left[1 - k^2 r_0^2 (1 + 2\varkappa)\right] \frac{I_1(kr_0)}{I_0(kr_0)}\, \frac{m_{2*}^2 - k^2}{m_{2*}^2 + k^2}. \end{gathered} \tag{89}$$

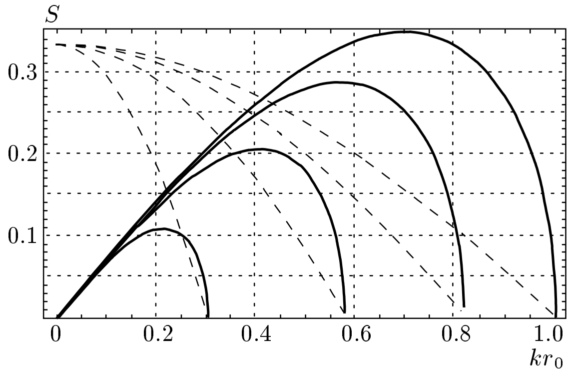

Fig. 2. A plot of the rescaled growth rate S versus kr_0 for low viscosity $\sqrt{\rho r_0^3/\sigma_0}\, s_-(kr_0)$ (solid lines) and high viscosity $(2\beta_2 r_0/\sigma_0)s_+(kr_0)$ (dashed lines) for different values of $\varkappa$ in descending order from top down $\varkappa = 0,\ 0.25,\ 1,\ 5$. If $\vartheta/\nu = 4\varkappa$, then the scaling for both viscous regimes is the same

If $\varkappa = 0$ and $\overline{\nu_1} = \overline{\nu_2}$, Eq. (89) is known as the Weber equation for a viscous isotropic liquid [6]. For low viscosity[7)], $\beta_1 \sim \beta_2 \ll \sqrt{\rho\sigma_0 r_0}$, a Rayleigh-type expression is obtained (see Fig. 2),

$$s_-^2(kr_0) = \frac{\sigma_0 k}{\rho r_0^2}\left[1 - k^2 r_0^2(1+2\varkappa)\right]\frac{I_1(kr_0)}{I_0(kr_0)}, \qquad (90)$$

where the subscript «−» denotes low viscosity.

The maximum s_-^{max} in Eq. (90), which corresponds to the wave number k_-^{max}, leads to evolution of the largest capillary instability. Numerical calculation shows that s_-^{max} and k_-^{max} are both proportional to $(1+2\varkappa)^{-1/2}$,

$$\begin{aligned} s_-^{max} &\approx \frac{1}{3\sqrt{1+2\varkappa}}\sqrt{\frac{\sigma_0}{\rho r_0^3}}, \\ k_-^{max} &\approx \frac{a}{r_0\sqrt{1+2\varkappa}}, \quad a = 0.697. \end{aligned} \qquad (91)$$

When high viscosity prevails, $\beta_1 \sim \beta_2 \gg \sqrt{\rho\sigma_0 r_0}$, the dispersion equation is given by (see Fig. 2)

$$\begin{aligned} s_+(kr_0) &= \frac{\sigma_0}{2\beta_2 r_0^2 k} \times \\ &\times \frac{\left[1 - k^2 r_0^2(1+2\varkappa)\right] I_1^2(kr_0)}{I_0(kr_0)I_1(kr_0) + kr_0\left[I_1'(kr_0)\right]^2}, \\ s_+^{max} &\approx \frac{\sigma_0}{6\beta_2 r_0}, \quad k_+^{max} = 0, \end{aligned} \qquad (92)$$

where the subscript «+» denotes high viscosity. In this limit, similar to ordinary liquids [4], there is no finite mode of the maximum instability for any $\varkappa$. In this case, we have

$$\begin{aligned} &\varsigma(r_0) = \\ &= \frac{k_+^{max}}{s_+^{max}}\left[c_1 I_1(k_+^{max} r_0) + c_2 I_1(m_{2*} r_0)\right] = 0. \end{aligned} \qquad (93)$$

Nevertheless, there exists a continuous range $[0, (1+2\varkappa)^{-1/2} r_0^{-1}]$ of wave numbers k with a finite disturbance growth rate $s_+(kr_0)$, which affects the cylindrical jet.

We note that the dispersion curves shown in Fig. 2 and those in Fig. 5 in [21] appear to be similar, but are characterized by different physical parameters. The reason for this observation is the similarity between Weber equation (89) and dispersion equation (36) in [21], which are obtained from different models. Our approach was to develop a general axisymmetric solution in the framework of the three-dimensional model. This model dates back to the Rayleigh–Weber theory [2, 3] and accounts for the radial inhomogeneity of the disturbed director field. The implicit solutions of Eq. (77) reflect the radial dependence of both the hydrodynamic $\mathbf{V}(r,z,t)$ and orientational $\mathbf{n}(r,z,t)$ modes, and they include all types of the LC jet evolution. A specific case where the hydrodynamic and orientaional modes are decoupled exhibits this radial dependence and yields dispersion equation (89) in explicit form.

In contrast, the one-dimensional analysis of the LC jet evolution, used in [21], is hardly compatible with the distortion of the director field $\mathbf{n}(r,z,t)$, and therefore must be supported by assuming a fixed axial direction of $\mathbf{n}^0$ (see detailed comments in Sec. 1). This endows their model with an inherent «decoupling» that results from the *a priori* elimination of the elastic forces. Obviously, similarity between the dispersion curves mentioned above disappears if we consider the general solution given by (77).

5.1. Hydrodynamic influence on the orientational instability of LCs

We conclude this section with a brief discussion regarding the hydrodynamic influence on the orientational instability of the director field $\mathbf{n}(\mathbf{r},t)$. As the effect of hydrodynamics changes the wave number k_s of the Plateau instability to k_{max}, the flow drives the

[7)] In the theory of viscoisotropic liquid jets, this case is known [7] as pertaining to the range of low Ohnesorge numbers $\mathrm{Oh} = \eta/\sqrt{\rho\sigma_0 r_0}$ that determine a competition between the hydrodynamic and surface tension forces. Expression (92) corresponds to the case of high Ohnesorge numbers.

orientational instability (41) of the director field $\mathbf{n}(\mathbf{r}, t)$. Indeed, in accordance with (87),

$$n_r^1(r,z) = c_3 I_1\left(m_3^{max} r\right), \quad m_3^{max} = \sqrt{\frac{s^{max}}{\vartheta}}. \qquad (94)$$

It is convenient to consider the following two marginal viscous regimes.

1. The low-viscosity limit,

$$\left(m_{3-}^{max} r_0\right)^2 \approx \frac{1}{3\sqrt{1+2\varkappa}} \frac{1}{\sqrt{\varkappa\varepsilon}}, \quad \varepsilon = \frac{\rho K}{\gamma_1^2}, \qquad (95)$$

where $\varepsilon \sim 10^{-6}$–10^{-4} is a small dimensionless parameter.

2. The high-viscosity limit,

$$\left(w_{3+}^{max} r_0\right)^2 \approx \frac{1}{6\varkappa} \frac{\gamma_1}{\beta_2}. \qquad (96)$$

In both limits, the distribution of the director field $\mathbf{n}(\mathbf{r}, t)$ in the jet is always nontrivial and definitely far from static distribution (41).

6. CONCLUSIONS

1. The capillary instability of an LC jet with a strong tangential anchoring of the director at the surface is considered in the framework of linear hydrodynamics of the uniaxial nematic LC. Its static version, which is called the Plateau instability and corresponds to the variational problem of minimal free energy, predicts an essential dependence of the disturbance cut-off wavelength on the dimensionless parameter $\varkappa = K/\sigma_0 r_0$.

2. The hydrodynamic problem of the capillary instability in LC jets is solved exactly and the dispersion relation is derived. This relation, which is represented as a determinant equation, implicitly expresses the dispersion $s = s(k)$ of the growth rate s as a function of the wave number k of axisymmetric disturbances of the jet.

3. The case where the dispersion equation becomes explicitly solvable is considered in detail. It corresponds to the regime where the hydrodynamic and orientational modes become decoupled. Hydrodynamics changes the wave number k_s of the Plateau instability into k_{max}, which produces evolution of the largest capillary instability. Similarly, a hydrodynamic flow influences the static orientational instability of the director field $\mathbf{n}(\mathbf{r}, t)$.

4. The present theory can easily be extended to nonuniaxial nematic LCs that possess finite point symmetry groups $G \subset O(3)$ as distinguished from the uniaxial group $D_{\infty h}$. The corresponding expressions for the free energy density $E_d(G)$ and the dissipative function $D(G)$ were derived in [31].

5. In this work, the effect of external fields was not considered. However, the theory developed here facilitates the treatment of the Rayleigh instability in nematic LCs in the presence of static electromagnetic fields.

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