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Surface modes at the nematic-isotropic interface

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We examine surface modes at the nematic-isotropic interface using the generalized dynamical Landau-de Gennes theory. We assume an isothermal, infinite, unbounded nematic-isotropic system characterized by a scalar order parameter, both phases having the same density and viscosity, respectively. The generalized dispersion relation is obtained and analyzed in particular cases. Order parameter relaxation dominates in the short wavelength limit, while in the long wavelength limit viscous damping becomes important. We study the crossover between the two regimes and estimate the extent of this region for the liquid crystal 8CB.

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I. INTRODUCTION

The diffuse theory of the interface separating two bulk phases was developed in the late 19th Century by van der Waals [1]. Previously, interfaces had been modeled by Young, Laplace, and Gauss as surfaces. In this formulation the interface is regarded as a singular surface on which associated physical mechanisms are localized and represented as boundary conditions to be applied at the surface [2,3]. The use of this so-called ‘‘sharp interface’’ description of phase boundaries in practical problems requires the solution of a free boundary problem. Free boundary problems are known to be extremely hard mathematically and indeed form a separate class in their own right.

In contrast, diffuse interface theories recognize that, in reality, the interface has a finite thickness (usually small compared with typical macroscopic length scales) in which physical quantities, such as density or composition, vary between their values in the adjacent bulk phases. Diffuse interface models may be based on an extended thermodynamics involving gradients of the thermodynamic variables to account for nonlocal effects. Originally such theories were formulated to investigate near-critical fluids. However, they have subsequently been refined and developed to account for a wide range of physical situations, such as liquid crystals [4], superconductivity [5], spinodal decomposition [6], and ordering transitions in alloys [7–9]. Rowlinson and Widom [10] provide a thorough account of their historical development.

The phase-field models provide an example of a diffuse interface model in which the phase of the system at a particular point in space and time is defined by the value of a hypothetical order parameter. The phase model of the first-order phase transition associated with the solidification of a pure material was first proposed by Langer [11] and subsequently developed by a number of researchers [12–19].

The phase-field formulation replaces the free-boundary problem associated with the sharp interface model of an interface by a coupled pair of nonlinear reaction-diffusion equations. The spatial and temporal variation of the order parameter phase field is governed by the time-dependent Ginzburg-Landau (TDGL) equation. The second equation (for temperature) is based on a modification of the heat equation to allow a source term that accounts for latent heat production at a moving interface. The original derivation of the two equations was justified by requiring the free energy of the system to decrease monotonically in time. Subsequently, Penrose and Fife [14] and others [15,16] applied the arguments of irreversible thermodynamics to the derivation of the phase-field equations, establishing that they are consistent with non-negative local entropy production.

Thermotropic liquid crystal surfaces present a case in which the phase-field model can be used particularly fruitfully. On the one hand, in this case the phase-field order parameter is no longer an *ad hoc* invention, but known to be a physically measurable quantity. On the other, the very fluidity of liquid crystals permits sensible time scales for useful experiments. The relevant phase field theory of nematic-isotropic phase transition [20,21] turns out to be just the dynamical generalization of the familiar Landau-de Gennes theory of liquid crystal interface [4,22]. The necessary hydrodynamic coupling has been included in the more complete dynamical Landau-de Gennes theory of Hess [23] and subsequently by Olmsted and Goldbart [24].

Apart from its intrinsic interest, the dynamics of liquid crystal surfaces presents an interesting case study because the naive sharp interface and naive diffuse interface limits lead to very different conclusions concerning the mode structure at the nematic-isotropic interface. The diffuse interface theory, solved assuming that the order parameter and velocity fields do not interact, gives rise to a purely diffusive surface wave whose mode structure is identical to the bulk diffusive mode induced by a TDGL theory [21]. On the other hand, the sharp interface theory yields a modified capillary wave, with a large propagating component. In this paper we make significant progress in reconciling these points of view.

In what follows we analyze the the surface eigenmotions of the nematic-isotropic interface using the Hess-Olmsted-Goldbart (HOG) model. We consider an equilibrium planar nematic-isotropic interface at T_{NI} (nematic-isotropic phase transition temperature) as the base state of the system. The front is then perturbed with a small amplitude monochro-

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matic wave and linear stability theory is used to obtain the dispersion relation. The paper is organized as follows. In Sec. II we describe the basic model and give the governing equations. In Sec. III we present an asymptotic analysis of the equations of motions and the generalized dispersion relation. Limiting cases of the dispersion relation are analyzed in Sec IV. Finally in Sec V, we draw some conclusions and present some directions for future work.

II. EQUATIONS OF MOTION

The local state of a uniaxial nematic liquid crystal is described by a traceless symmetric second rank tensor $Q_{\alpha\beta}$

$$Q_{\alpha\beta}=\frac{3}{2}\phi\left(n_\alpha n_\beta-\frac{1}{3}\delta_{\alpha\beta}\right), \tag{1}$$

where the unit vector $\vec{n}$ is the usual nematic director, and ϕ is now the scalar order parameter.

In this paper, although not in future studies, we shall suppose $\vec{n}$ to be fixed in space and time, and the relevant physics is given by the scalar order parameter $\phi(\vec{r},t)$. We note that this is an idealization which is in general not true during relaxation, and not even true statically close to an interface. However, in our view, previous studies of related systems [21] suggest that the lowest frequency and slowest relaxation modes do approximately satisfy this condition when the surface is homeotropic. Thus our idealized problem–that of the dynamics of a nonconserved order parameter weakly coupled to a conserved order parameter (the density) – contains a good part of the essential physics of the nematic-isotropic interface, and is significantly easier to analyze.

Within the mesoscopic approach the free energy functional is given by

$$\mathcal{F}(Q,T)=\int[f(Q,T)+f_F(\nabla Q)]dV, \tag{2}$$

where

$$f(Q,T)=a(T-T^*)Q_{\alpha\beta}Q_{\beta\alpha}-BQ_{\alpha\beta}Q_{\beta\gamma}Q_{\gamma\alpha}+C(Q_{\alpha\beta}Q_{\beta\alpha})^2 \tag{3}$$

is the bulk Landau-de Gennes-free energy density [25,26], and

$$f_F(\nabla Q)=\frac{1}{2}L_1(\partial_\alpha Q_{\beta\gamma})^2+\frac{1}{2}L_2(\partial_\alpha Q_{\alpha\beta})^2 \tag{4}$$

is the distortion (or Frank) [27] free energy density. The elastic constants L_1 and L_2 are related to the Frank-Oseen elastic constants by the relations $K_1=K_3=9\phi_b^2(L_1+L_2/2)/2$ and $K_2=9\phi_b^2L_1/2$, where ϕ_b is the bulk nematic order parameter. In the so-called "one-constant approximation" ($K_1=K_2=K_3=K$) and $\phi_b=1$, the Landau-de Gennes-free energy density becomes

$$f_F=\frac{1}{9}K(\partial_\alpha Q_{\beta\gamma})^2. \tag{5}$$

In this paper we use this Landau-de Gennes form, but retaining the Frank-Oseen elastic constant so that comparison with bulk nematic quantities may be more easily made.

We suppose that the diffusion is sufficiently rapid that the system remains in thermal equilibrium. We therefore ignore the equation for energy conservation and assume an isothermal system at a temperature specified in the formulation of the problem. We suppose the system to be incompressible, i.e., the velocities of motion considered are much less than the sound velocity c, and for the circular frequency ω and wave number k, the inequality $\omega \ll ck$ is valid. In subsequent work we shall relax these approximations, and also include the full symmetry allowed for within the Landau-de Gennes formalism.

Using these approximations, the HOG coupled equations of motion for the fluid velocity and the order parameter become [23,24],

$$\nabla\cdot\vec{v}=\vec{0}, \tag{6}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)\vec{v}_\alpha=\partial_\gamma\sigma_{\alpha\gamma}, \tag{7}$$

$$(\partial_t+\vec{v}\cdot\nabla)Q_{\alpha\beta}=\kappa^a_{\alpha\beta}Q_{\gamma\beta}-Q_{\alpha\gamma}\kappa^a_{\gamma\beta}+\lambda\kappa^s_{\alpha\beta}+\frac{1}{\gamma_1}H^s_{\alpha\beta}. \tag{8}$$

The total stress tensor $\sigma_{\alpha\beta}$ is given by

$$\sigma_{\alpha\beta}=-p\,\delta_{\alpha\beta}+\sigma^s_{\alpha\beta}+\sigma^a_{\alpha\beta}+\sigma^d_{\alpha\beta}. \tag{9}$$

$$\sigma^s_{\alpha\beta}=2\,\eta\kappa^s_{\alpha\beta}-\lambda H^s_{\alpha\beta}, \tag{10}$$

$$\sigma^a_{\alpha\beta}=H^s_{\alpha\gamma}Q_{\gamma\beta}-Q_{\alpha\gamma}H^s_{\gamma\beta}, \tag{11}$$

$$\sigma^d_{\alpha\beta}=-\frac{\delta\mathcal{F}}{\delta\partial_\alpha Q_{\gamma\rho}}\partial_\beta Q_{\gamma\rho}. \tag{12}$$

Here $H_{\alpha\beta}\equiv-\delta\mathcal{F}/\delta Q_{\alpha\beta}$ is the molecular field, η is the viscosity not coupled to the rotation, γ_1 is the rotational viscosity and λ is a dimensionless coupling constant between orientational order and flow which turns out to be the ratio of rotational viscosities. The tensor field $\kappa_{\alpha\beta}\equiv\partial_\alpha v_\beta$ is the velocity gradient tensor, and the superscripts *s*, *a*, and *d*, denote symmetric-traceless, asymmetric, and distortional portions of a tensor, respectively.

Using Eqs. (1)–(5) and considering a two-dimensional flow with horizontal and vertical velocity components u and w in the x and z directions, respectively, the basic Eqs. (6)–(8) take the form

$$\partial_x u+\partial_z w=0, \tag{13}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)u=-\partial_x p-K\nabla^2\phi\partial_x\phi+\eta\nabla^2u+\frac{1}{2}\lambda\partial_x\times(K\nabla^2\phi-f_\phi), \tag{14}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)w=-\partial_z p-K\nabla^2\phi\partial_z\phi+\eta\nabla^2w-\lambda\partial_z\times(K\nabla^2\phi-f_\phi), \tag{15}$$

$$(\partial_t+\vec{v}\cdot\nabla)\phi=\frac{1}{\gamma_1}(K\nabla^2\phi-f_\phi)+\frac{1}{2}\lambda(-\partial_x u+\partial_z w),\tag{16}$$

where $f_\phi=df/d\phi=3a(T-T^*)\phi-\frac{9}{4}B\phi^2+9C\phi^3$. The material properties η, λ, and γ_1 are assumed to be constant.

There are two typical lengths in the problem. These are as follows.

(i) The microscopic correlation length l_ϕ associated with order parameter changes. This gives the interface width, or equivalently the dimension of the order parameter profile.

(ii) A macroscopic length related to the capillary number and the Reynolds number. We consider the thin interface limit of the problem. The physics involves capillary forces, viscous dissipation, and fluid inertia. Assuming that no other physics enters, the control parameters are the interfacial tension α, the viscosity η, and the mass density ρ. From these can be constructed only one length $l_\eta=\eta^2/\rho\alpha$.

For usual nematics, $l_\phi\approx10^{-6}$ cm and $l_\eta\approx10^{-2}$ cm. The ratio of these two lengths $\epsilon=l_\phi/l_\eta\approx10^{-4}$ constitutes the small parameter of the theory.

We rewrite Eqs. (13)–(16) in dimensionless form by measuring length in units of l_η and time in units of $t_*=l_\eta^2/D\approx10^{-3}$ s, where $D=\eta/\rho\approx0.1$ cm^2/s is the viscous diffusion constant. The resulting velocity unit is then $v_*=D/l_\eta\approx10$ cm/s.

We introduce the dimensionless quantities: $\overline{\phi}=6C\phi/B$, $\overline{\tau}=24a(T-T^*)C/B^2$, $\overline{f}=24^2C^3f/B^4$, $\overline{\rho}=24^2C^3v_*^2\rho/B^4$, $\overline{p}=24^2C^3p/B^4$, $\overline{\lambda}=6C\lambda/B$, $\overline{\eta}=24^2C^3\eta/t_*B^4$, $\overline{\gamma_1}=l_\eta v_*\gamma_1/K$, $\epsilon^2=16CK/B^2l_\eta^2=l_\phi^2/l_\eta^2$. Omitting the bar notation, the governing Eqs. (13)-(16) can be written as

$$\partial_x u+\partial_z w=0,\tag{17}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)u=-\partial_x p-\epsilon^2\nabla^2\phi\partial_x\phi+\eta\nabla^2u+\frac{1}{2}\lambda\partial_x\times(\epsilon^2\nabla^2\phi-f_\phi),\tag{18}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)w=-\partial_z p-\epsilon^2\nabla^2\phi\partial_z\phi+\eta\nabla^2w-\lambda\partial_z\times(\epsilon^2\nabla^2\phi-f_\phi),\tag{19}$$

$$\gamma_1\epsilon^2(\partial_t+\vec{v}\cdot\nabla)\phi=(\epsilon^2\nabla^2\phi-f_\phi)+\frac{1}{2}\lambda\gamma_1\epsilon^2(-\partial_x u+\partial_z w),\tag{20}$$

where $f_\phi=2\tau\phi-6\phi^2+4\phi^3$.

This well-known free energy density describes a first-order nematic-isotropic phase transition. For $\tau=\tau_{NI}=1$, the two phases, nematic ($\phi_{nem}=1$) and isotropic ($\phi_{iso}=0$) coexist in equilibrium ($f_{nem}=f_{iso}$).

In this paper we shall take the base state of the system to be a stationary planar nematic-isotropic interface situated at $z=0$, such that the nematic lies in the region $z<0$ and the isotropic phase lies in the region $z>0$. The front is then perturbed with a small amplitude monochromatic wave and linear stability theory will be used to obtain the dispersion relation. x is the direction of the wave propagation along the interface. For ease of presentation we discuss two-dimensional perturbations to the system, with the understanding that, since the system is isotropic in x and y directions (neglecting the biaxiality of the nematic phase), the results carry over to the three-dimensional case if the wave number is interpreted as the modulus of the two-dimensional wave vector.

III. ASYMPTOTICS

We now seek solutions of Eqs. (17)–(20) for $\epsilon\ll1$, which are essentially diffusive in the inner region, but which are governed by viscous hydrodynamic behavior in the outer region. In the inner region ϕ varies rapidly, whereas in the outer region advective transport of the order parameter dominates.

A. Outer region

Since in the outer region ϕ is a constant in each phase ($\phi=\phi_{nem}=1$ for $z<0$ and $\phi=\phi_{iso}=0$ for $z>0$), $\vec{v}$ obeys the equations,

$$\partial_x u+\partial_z w=0,\tag{21}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)u=-\partial_x p+\eta\nabla^2u,\tag{22}$$

$$\rho(\partial_t+\vec{v}\cdot\nabla)w=-\partial_z p+\eta\nabla^2w.\tag{23}$$

Thus, the outer problem is equivalent to the Navier-Stokes equations subject to the incompressibility condition. This problem has been much studied in the literature [28–30]. The solution corresponding to the stationary planar interface is given by, $u_0=w_0=0$ and $p_0=$const. We now impose a small periodic perturbation to the interface in the x direction with wave number k,

$$\begin{aligned}u&=u_0+u_1=0+U(z)\exp[i(kx-\omega t)]\\&=U_0\exp[qz+i(kx-\omega t)],\end{aligned}\tag{24}$$

$$\begin{aligned}w&=w_0+w_1=0+W(z)\exp[i(kx-\omega t)]\\&=W_0\exp[qz+i(kx-\omega t)],\end{aligned}\tag{25}$$

$$\begin{aligned}p&=p_0+p_1=\text{const}+P(z)\exp[i(kx-\omega t)\\&=\text{const}+P_0\exp[qz+i(kx-\omega t)].\end{aligned}\tag{26}$$

The circular frequency ω is, in general, a complex number whose imaginary part defines the time scale ($t=-1/\text{Im}\omega$) for the relaxation of the perturbation with characteristic length scale $2\pi/k$.

Substituting these forms into Eqs. (21)–(23), and linearizing, yields

$$0=\partial_x u_1+\partial_z w_1,\tag{27}$$

$$\rho\partial_t u_1=-\partial_x p_1+\eta\nabla^2u_1,\tag{28}$$

$$\rho\partial_t w_1=-\partial_z p_1+\eta\nabla^2w_1.\tag{29}$$

In this case we get a system of algebraic equations for the amplitudes

$$ikU_0+qW_0=0,$$

$$\{i\omega+\nu(q^2-k^2)\}U_0-\frac{ik}{\rho}P_0=0,$$

$$\{i\omega+\nu(q^2-k^2)\}W_0-\frac{q}{\rho}P_0=0, \tag{30}$$

where $\nu=\eta/\rho$ is the kinematic viscosity. In order to obtain the characteristic equation connecting q, k, and ω, we put the determinant of the system (30) equal to zero and obtain

$$(q^2-k^2)[i\omega+\nu(q^2-k^2)]=0, \tag{31}$$

with solutions $q=\pm k$ and $q=\pm l$ where $l=k\sqrt{1-i\omega/(\nu k^2)}$.

For a nematic-isotropic system (which here is merely a simple binary liquid) of large fluid depths, wavelike solutions of Eqs. (27)–(29) in the nematic (region $z<0$), are of the form

$$u_1=(ikAe^{kz}-lCe^{lz})e^{i(kx-\omega t)}, \tag{32}$$

$$w_1=(kAe^{kz}+ikCe^{lz})e^{i(kx-\omega t)}. \tag{33}$$

Similarly, in the isotropic phase (region $z>0$) (using primed quantities),

$$u_1'=(ikA'e^{-kz}+lC'e^{-lz})e^{i(kx-\omega t)}, \tag{34}$$

$$w_1'=(-kA'e^{-kz}+ikC'e^{-l^z})e^{i(kx-\omega_k t)}. \tag{35}$$

In Eqs. (32)–(35) the velocities are determined by two contributions: (i) the potential flow described by the terms with amplitudes A and A', which correspond to the velocity potentials, $\psi=Ae^{kz}e^{i(kx-\omega_k t)}$ and $\psi'=A'e^{-kz}e^{i(kx-\omega t)}$; (ii) the rotational flow represented by a vector potential with amplitudes $C(k^2-l^2)$, $C'(k^2-l^2)$ describing fluids of finite viscosity.

The dispersion relation $\omega(k)$ is derived by consistency relations on the quantities which appear in Eqs. (34) and (35). In the full diffuse interface approach these are provided by matching the results in this section with those of an analogous analysis in the ''inner region'' close to the interface, and we carry out this analysis below. In the classical sharp interface approach, however, the inner region shrinks to zero thickness. Now, as is well known, these consistency relations are determined by the boundary conditions at the sharp boundary $z=0$ on the nematic-isotropic interface, where the stress tensor and velocity must be continuous [31]. The dispersion relation can be written as

$$\omega_\eta^2=\frac{l-k}{l}\omega_0^2, \tag{36}$$

where $\omega_0^2=\alpha k^3/2\rho$ is the capillary wave dispersion relation for ideal fluids [31], and α is the interfacial tension. The subscript η means that the imaginary part of ω_η describes the viscous damping of the capillary waves. The details of the conditions are presented in the Appendix.

B. Inner region

To look for the solutions in the inner region, we rewrite Eqs. (17)–(20) in terms of inner variables $\zeta=x/\epsilon$ and $\xi=z/\epsilon$,

$$\partial_\zeta\tilde{u}+\partial_\xi\tilde{w}=0, \tag{37}$$

$$\tilde{\rho}(\partial_t+\tilde{\vec{v}}\cdot\boldsymbol{\nabla})\tilde{u}=-\partial_\zeta p-\nabla^2\tilde{\phi}\partial_\zeta\tilde{\phi}+\eta\nabla^2\tilde{u}+\frac{1}{2}\lambda\partial_\zeta(\nabla^2\tilde{\phi}-f_{\tilde{\phi}}), \tag{38}$$

$$\tilde{\rho}(\partial_t+\tilde{\vec{v}}\cdot\boldsymbol{\nabla})\tilde{w}=-\partial_\xi p-\nabla^2\tilde{\phi}\partial_\xi\tilde{\phi}+\eta\nabla^2\tilde{w}-\lambda\partial_\xi(\nabla^2\tilde{\phi}-f_{\tilde{\phi}}), \tag{39}$$

$$\tilde{\gamma}_1(\partial_t+\tilde{\vec{v}}\cdot\boldsymbol{\nabla})\tilde{\phi}=\nabla^2\tilde{\phi}-f_{\tilde{\phi}}+\frac{1}{2}\lambda\tilde{\gamma}_1(-\partial_\zeta\tilde{u}+\partial_\xi\tilde{w}), \tag{40}$$

where the velocity unit is now l_ϕ/t_*, $\tilde{\rho}=24^2C^3l_\phi^2\rho/B^4t_*^2$, $\tilde{\gamma}_1=l_\phi^2\gamma_1/Kt_*^2$.

Consider first an equilibrium planar nematic-isotropic interface perpendicular to the ξ axis. The density and horizontal momentum equations [(37) and (38)] are satisfied identically, and the remaining equations give

$$\partial_\xi p_0=\partial_\xi^2\tilde{\phi}_0\partial_\xi\tilde{\phi}_0-\lambda\partial_\xi(\partial_\xi^2\tilde{\phi}_0-f_{\tilde{\phi}}^0), \tag{41}$$

$$\partial_\xi^2\tilde{\phi}_0=f_{\tilde{\phi}}^0, \tag{42}$$

where the subscript 0 refers to the equilibrium interface and we have defined $f_{\tilde{\phi}}^0=f_{\tilde{\phi}}(\tilde{\phi}_0)$. Using Eq. (42) and integrating Eq. (41) we obtain

$$p_0=p_\infty-\frac{1}{2}(\partial_\xi\tilde{\phi}_0)^2, \tag{43}$$

where p_∞ is the common value of the pressure in the bulk phases where the gradient of the order parameter tends to zero. The order parameter profile $\tilde{\phi}_0(\xi)$ is the solution of Eq. (42) with the boundary conditions $\tilde{\phi}_0(-\infty)=1$ and $\tilde{\phi}_0(\infty)=0$. Integrating Eq. (42) once, and imposing the boundary conditions, gives $(\partial_\xi\tilde{\phi}_0)^2=2f(\tilde{\phi}_0)$. This result can be used to obtain the energy per unit area of the interface, that is the interfacial tension, as

$$\tilde{\alpha}=\int_{-\infty}^{\infty}\left[f(\tilde{\phi}_0)+\frac{1}{2}(\partial_\xi\tilde{\phi}_0)^2\right]d\xi=\int_{-\infty}^{\infty}(\partial_\xi\tilde{\phi}_0)^2d\xi. \tag{44}$$

There is a one-parametric class of functions satisfying Eq. (42), with vanishing derivatives in every order at infinity, namely,

$$\tilde{\phi}_0(\xi)=\frac{1}{2}\left(1-\tanh\frac{\xi-\xi_0}{\sqrt{2}}\right). \tag{45}$$

The freedom of choosing ξ_0 anywhere represents the fact that the position of the interface is arbitrary. We fix the center of the interface (defined by $\tilde{\phi}_0=1/2$) to be at $\xi=0$. This extra condition gives ξ_0.

We may perturb this base state as follows:

$$\tilde{u}=0+\tilde{U}(\xi)\exp[i\tilde{k}\zeta-\omega t)],$$

$$\tilde{w}=0+\tilde{W}(\xi)\exp[i(\tilde{k}\zeta-\omega t)],$$

$$p=p_0(\xi)+P(\xi)\exp[i(\tilde{k}\zeta-\omega t)],$$

$$\tilde{\phi}=\tilde{\phi}_0(\xi)+\tilde{\Phi}(\xi)\exp[i(\tilde{k}\zeta-\omega t)], \tag{46}$$

where $\tilde{k}=\epsilon k$. Substituting Eqs. (46) into Eqs. (37)–(40) and linearizing in perturbations, give

$$0=i\tilde{k}\tilde{U}+d_\xi\tilde{W}, \tag{47}$$

$$-i\tilde{\rho}\omega\tilde{U}=-i\tilde{k}P-i\tilde{k}\tilde{\mathbf{\Phi}}d_\xi^2\tilde{\phi}_0+\eta(d_\xi^2-\tilde{k}^2)\tilde{U}-\frac{1}{2}i\tilde{k}\lambda H\tilde{\mathbf{\Phi}}, \tag{48}$$

$$\begin{aligned}-i\tilde{\rho}\omega\tilde{W}=&-d_\xi P-d_\xi^2\tilde{\phi}_0 d_\xi\tilde{\mathbf{\Phi}}-(d_\xi^2-\tilde{k}^2)\tilde{\mathbf{\Phi}}d_\xi\tilde{\phi}_0\\&+\eta(d_\xi^2-\tilde{k}^2)\tilde{W}+\lambda d_\xi H\tilde{\mathbf{\Phi}},\end{aligned} \tag{49}$$

$$-i\tilde{\gamma}_1\omega\tilde{\mathbf{\Phi}}=-H\tilde{\mathbf{\Phi}}+\frac{1}{2}\lambda\tilde{\gamma}_1(d_\xi\tilde{W}-i\tilde{k}\tilde{U})-\tilde{\gamma}_1\tilde{W}d_\xi\tilde{\phi}_0, \tag{50}$$

where $H=-d_\xi^2+\tilde{k}^2+f^0_{\tilde{\phi}\tilde{\phi}}$ can be conveniently thought of as a quantum mechanical Hamiltonian operator [33].

We now note that if we ignore the interaction between velocity and order parameter fluctuations, the physics is completely given by the the eigenvalue equation Eq. (50). This is the TDGL limit in which the inner region dictates the physics entirely. Eq. (50) becomes, $H\tilde{\Phi}=i\gamma_1\omega\tilde{\Phi}$. Note that $f^0_{\tilde{\phi}\tilde{\phi}}$ is positive at $\xi=\pm\infty$ (where $\tilde{\phi}_0=0$ and $\tilde{\phi}_0=1$) and negative at $z=0$ (where $\tilde{\phi}_0=1/2$). For our case $f(\tilde{\phi})=\tilde{\phi}^2(1-\tilde{\phi})^2$, one has $f^0_{\tilde{\phi}\tilde{\phi}}=2-12\tilde{\phi}_0(\xi)+12\tilde{\phi}_0^2(\xi)$, which equals -1 at $\xi=0$, and tends to $+2$ for $\xi\to\pm\infty$. It follows that $f^0_{\tilde{\phi}\tilde{\phi}}$ represents a potential well which must have at least one bound state. In fact, since $\tilde{k}=0$ corresponds to a uniform translation of the interface, we know that $\omega_{\tilde{k}=0}=0$ is the eigenvalue with the eigenfunction $d_\xi\tilde{\phi}_0$ [this can be easily checked by differentiating Eq. (42) with respect to ξ]. Also, since this function has no node, it must be the ground state. Since the $\tilde{k}$ dependence of H is simply the additive constant $\tilde{k}^2$, it follows that $d_\xi\tilde{\phi}_0$ is the ground state eigenfunction—the so-called ‘‘slow mode’’—for all $\tilde{k}$ with eigenvalue [33],

$$\omega_{\tilde{\phi}}=-i\frac{\tilde{k}^2}{\tilde{\gamma}_1}=-i\frac{k^2}{\gamma_1}, \tag{51}$$

where the subscript $\tilde{\phi}$ means that the imaginary part of $\omega_{\tilde{\phi}}$ describes the relaxation rate of the order parameter.

To find the general dispersion relation, we first eliminate $\tilde{U}$ and P from (47)–(50), obtaining

$$\begin{aligned}(i\tilde{\rho}\omega-\eta\tilde{k}^2)\tilde{W}=&\left(\frac{i\tilde{\rho}\omega}{\tilde{k}^2}-2\eta\right)d_\xi^2\tilde{W}+\frac{\eta}{\tilde{k}^2}d_\xi^4\tilde{W}-H\tilde{\Phi}d_\xi\tilde{\phi}_0\\&-\frac{3}{2}\lambda d_\xi(H\tilde{\Phi}),\end{aligned} \tag{52}$$

$$H\tilde{\Phi}-i\tilde{\gamma}_1\omega\tilde{\Phi}=\lambda\tilde{\gamma}_1 d_\xi\tilde{W}-\tilde{\gamma}_1\tilde{W}d_\xi\tilde{\phi}_0. \tag{53}$$

Using $Hd_\xi\tilde{\phi}_0=\tilde{k}^2 d_\xi\tilde{\phi}_0$ and the fact that the linear operator H is self-adjoint, integrating Eq. (52) over all ξ, multiplying Eq. (53) by $d_\xi\tilde{\phi}_0$ and integrate, and eliminating $\int_{-\infty}^{\infty}\tilde{\Phi}d_\xi\tilde{\phi}_0 d\xi$, the following result is obtained:

$$\begin{aligned}(i\tilde{\rho}\omega-\eta\tilde{k}^2)(i\tilde{\gamma}_1\omega-\tilde{k}^2)&\int_{-\infty}^{\infty}\tilde{W}d\xi\\&=-\tilde{k}^2\tilde{\gamma}_1\int_{-\infty}^{\infty}\tilde{W}(d_\xi\tilde{\phi}_0)(2)d\xi.\end{aligned} \tag{54}$$

Considering the relations between ‘‘inner’’ and ‘‘outer’’ quantities (due to different scaling), the Eq. (54) can be written as

$$\begin{aligned}2(i\rho\omega-\eta k^2)(i\gamma_1\omega-k^2)&\int_{-\infty}^{\infty}Wdz\\&=-k^2\gamma_1\alpha[W(0^+)+W(0^-)],\end{aligned} \tag{55}$$

where we have used the matching condition

$$\lim_{\xi\to\pm\infty}\tilde{W}(\xi)=\lim_{z\to\pm0}W(z)=\frac{1}{\epsilon}W(0^\pm), \tag{56}$$

Using Eqs. (33) and (35), the integral in Eq. (55) can be evaluated to give

$$\int_{-\infty}^{\infty}Wdz=A-A'+i\frac{k}{l}(C+C'). \tag{57}$$

Considering the continuity of horizontal velocity at the interface $U(0)=U'(0)$, we obtain the generalized dispersion relation

$$\omega^2+\frac{ik^2}{\gamma_1}\omega=\omega_0^2\frac{l-k}{l}, \tag{58}$$

which can be written in a more compact form as

$$\omega^2-\omega_\phi\omega=\omega_\eta^2, \tag{59}$$

where we have used Eqs. (36) and (51).

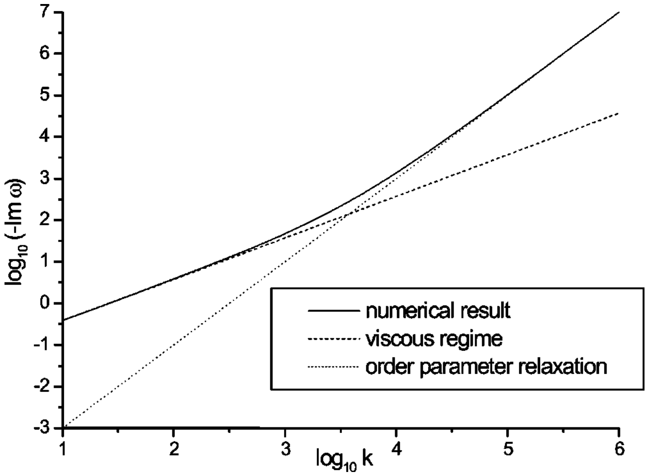

FIG. 1. The "phase" diagram $(\log_{10}(1/t),\log_{10}k)$. The general dispersion relation Eq. (58) (continuous curve), the Navier-Stokes dispersion relation Eq. (36) (dashed curve), and the order parameter relaxation dispersion relation Eq. (51) (dotted curve).

IV. ANALYSIS OF THE DISPERSION RELATION

From the asymptotic analysis presented in Sec. III, it follows that in the short wavelength limit $(k\to\infty)$ the interface must necessarily be regarded as diffuse. In this case, the relaxation of the order parameter is the important process, and this is governed primarily by processes in the inner region in our asymptotic analysis. However, in the large wavelength limit $(k\to 0)$ the interface is essentially sharp. Now the viscous damping process occurring in outer region dominates. The transition between these two regimes takes place when $\omega_\phi=\omega_\eta$, which gives,

$$k_c=\frac{\alpha\gamma_1^2}{2\rho K}\left[\left(1-\frac{K}{\nu\gamma_1}\right)^{-1/2}-1\right], \qquad (60)$$

where we have used dimensional quantities (for 8CB the experimental values are [32]: $K=10^{-6}$ dyn, $\alpha=1.5\times10^{-2}$ erg/cm^2, $\gamma_1=\eta=0.1$ P, and $\rho=1$ g/cm^3). Using these values we obtain the critical wave number $k_c=3750$ cm^{-1} and the corresponding critical wavelength $\lambda_c=17$ μm. For viscous damping, $k<k_c$, and for the order parameter relaxation, $k>k_c$.

The numerical solution of Eq. (59) is presented in Fig. 1. There is a continuous transition between the viscous damping and order parameter relaxation regimes. To define the width of the crossover region between the two regimes, we use the difference between numerical and asymptotic profiles ones. These are plotted in Fig. 2. The width of the interface, defined as usual as width at half height, takes the value $\Delta k=7480$ cm^{-1}.

We now analyze the viscous damping regime in more detail. A numerical study of the solution of Eq. (36) in a wide k range shows that $\omega(k)$ is a unique function of k for all k. Moreover, in the low- and high- viscosity limits, this solution assumes the asymptotic forms given below.

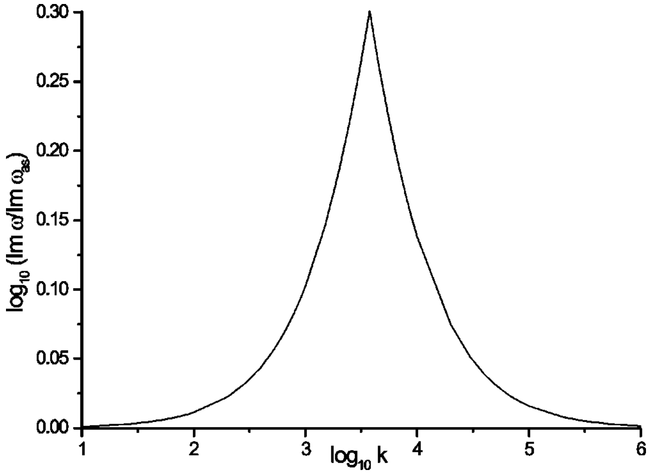

FIG. 2. The difference between numerical $-\mathrm{Im}(\omega)$ (continuous curve in Fig. 1) and corresponding asymptotic values (dashed curve in Fig. 1 for $k<k_c$ and dotted curve in Fig. 1 for $k>k_c$) as a function of $\log_{10}k$.

A. Weak damping approximation $(|\omega_0|\gg\nu k^2)$

In this limit ω_η differs from ω_0 only by a small quantity γ_η. Thus in this regime it can be approximated by $\omega_\eta=\omega_0-i\gamma_\eta$, by analogy with a damped oscillator. Using this approximation for l and ω_η we now obtain

$$\gamma_\eta=(1-i)\left(\frac{\nu\omega_0}{2}\right)^{1/2}k. \qquad (61)$$

The real part of γ_η gives a small correction to the imaginary part of ω_η. Inserting ω_0 in Eq. (61) we notice that the damping coefficient γ_η increases with k as $k^{7/4}$.

B. Strong damping approximation $(|\omega_0|\ll\nu k^2)$

In this case $l/k\approx 1-i\omega_\eta/2\nu k^2$. From Eq. (36) we now obtain two solutions for the dispersion relation: $\omega_\eta=0$ and $\omega_\eta=-i\omega_0^2/2\nu k^2=-i\alpha k/4\eta$. However, for $\omega_\eta=0$, $\vec{v}=0$, and a finite static distortion of the interface does not satisfy balance of forces.

The crossover between these two régimes takes place at $|\omega_0|=\nu k^2$, which gives

$$k_*=\frac{\alpha}{2\rho\nu^2}, \qquad (62)$$

where $k_*\approx 0.75$ cm^{-1} for 8CB. For weak damping, $k\ll k_*$, and for strong damping, $k\gg k_*$.

Thus, the transition in Fig. 1 takes place between a strong damping régime (for which $-\mathrm{Im}\omega_\eta$ grows linearly with k) and a pure order parameter relaxation régime. This explains the linear character of the corresponding viscous régime curve.

V. CONCLUSIONS

In this paper we have examined surface modes at the nematic-isotropic interface using the HOG generalized dynamical Landau-de Gennes theory [23,24]. We have assumed an isothermal system characterized by a scalar order parameter, both phases having the same density and viscosity, respectively. Input parameters include the viscosity η, the rotational viscosity γ_1, a dimensionless coupling constant between orientational order and flow λ which is the analog of the ratio between the rotational viscosities and a microscopic length scale ϵ associated with order parameter changes.

As we have already observed, this model does not appear at first sight to include orientational degrees of freedom. Nevertheless, it represents an important idealization of the liquid crystal problem, a conserved density is coupled to a nonconserved order parameter whose statics is driving the phase transition.

We have considered the equilibrium planar nematic-isotropic interface as the base state of the system. The front was then perturbed with a small-amplitude monochromatic plane wave and the linear stability of the front was examined to obtain the general dispersion relation (58). The numerical study of Eq. (58) in a wide k range shows that for any k only one solution exists and this solution assumes asymptotic forms corresponding to viscous damping ($k \to 0$) and order parameter relaxation ($k \to \infty$) processes. We have defined a crossover point between these two regimes occurring at $k = k_c$, where k_c is given by Eq. (60). Furthermore, for 8CB, we have estimated the magnitude of k_c, as well as the size of the crossover region, and also the crossover k_* between weak and strong damping order parameter relaxation regimes.

We have developed a formalism which successfully links both the short wavelength limit, for which the interface is essentially diffuse and order parameter relaxation is the important process, and the long wavelength limit, for which the interface is essentially sharp and viscous damping is important. In future studies we shall use this method to examine cases in which: (i) the difference in density between the phases is included, in which case gravity-driven capillary waves dominate the long-wave surface spectrum, (ii) the heat equation which explicitly takes account of latent heat production at interface is considered, (iii) a full orientational order parameter is included, and (iv) the NI interface interacts with a neighboring hard wall. The essential effect of the field would be to locate the interface and as a consequence a displacement of the interface would cost energy; the Goldstone soft mode with the eigenfunction $d_\xi \tilde{\phi}_0$ then becomes hard.

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APPENDIX: THE BOUNDARY CONDITIONS AT THE SHARP INTERFACE

(i) The continuity of the stress tensor in the vertical direction gives

$$\sigma_{zz} - \sigma'_{zz} = \alpha \frac{\partial^2 \varsigma}{\partial x^2}, \tag{A1}$$

where $\sigma_{zz} = -p + 2\eta \partial w / \partial z$ and $\sigma'_{zz} = -p' + 2\eta \partial w' / \partial z$ are corresponding stresses for the nematic and isotropic phases at the interface, and $\varsigma = iw/\omega$ is the instantaneous displacement of an interface point with horizontal coordinate x from the mean interfacial plane, with $\langle \varsigma \rangle = 0$. The pressure is expressed as $p = -\rho g \varsigma - \rho \partial \psi / \partial t$, $p' = -\rho g \varsigma - \rho \partial \psi' / \partial t$ and the interfacial pressure $\alpha \partial^2 \varsigma / \partial x^2$ is due to the interfacial tension α.

(ii) The continuity of stress tensor components involving the horizontal direction gives $\sigma_{xz} = \sigma'_{xz}$, where $\sigma_{xz} = \eta(\partial u / \partial z + \partial w / \partial x)$ and $\sigma'_{xz} = \eta(\partial u' / \partial z + \partial w' / \partial x)$ are the corresponding components for the nematic and isotropic liquid, respectively.

(iii) The continuity of vertical and horizontal velocity gives, $w = w'$ and $u = u'$. Using the velocity expressions (28)–(31), these conditions lead to the dispersion relation (36).

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[1] For an English translation, see J.S. Rowlinson, J. Stat. Phys. **20**, 197 (1979).
[2] J.S. Langer, Rev. Mod. Phys. **52**, 1 (1980).
[3] B. Caroli, C. Caroli, and B. Roulet, in *Solids Far From Equilibrium*, edited by C. Godrèche (Cambridge University Press, Cambridge, 1992), p. 155.
[4] P.G. de Gennes, Mol. Cryst. Liq. Cryst. **12**, 193 (1971).
[5] V.L. Ginzburg and L.D. Landau, Sov. Phys. JETP **20**, 1064 (1950).
[6] J.W. Cahn and J.E. Hilliard, J. Chem. Phys. **28**, 258 (1958); J.W. Cahn, Acta Metall. **9**, 795 (1961).
[7] J.W. Cahn and S.M. Allen, J. Phys. (Paris), Colloq. **C7-51** (1977).
[8] S.M. Allen and J.W. Cahn, Acta Metall. **27**, 1085 (1979).
[9] R.J. Braun, J.W. Cahn, G.B. McFadden, and A.A. Wheeler, Philos. Trans. R. Soc. London, Ser. A **355**, 1787 (1997).
[10] J.S. Rowlinson and B. Widom, *Molecular Theory of Capillarity* (Clarendon Press, Oxford, 1989).
[11] J.S. Langer, (unpublished notes, 1978); J.S. Langer, in *Directions in Condensed Matter Physics*, edited by G. Grinstein and G. Mazenko (World Scientific, Philadelphia, 1986), pp. 165-186.
[12] G. Caginalp, in *Applications of Field Theory to Statistical Mechanics*, edited by L. Garrido (Springer, Berlin, 1985), pp. 216-226; G. Caginalp and P.C. Fife, Phys. Rev. B **33**, 7792 (1986); G. Caginalp, Arch. Ration. Mech. Anal. **92**, 205

(1986); Phys. Rev. A **39**, 5887 (1989).

[13] J.B. Collins and H. Levine, Phys. Rev. B **31**, 6119 (1985).

[14] O. Penrose and P.C. Fife, Physica D **43**, 44 (1990).

[15] A.P. Umantsev, J. Chem. Phys. **96**, 605 (1992).

[16] S.-L. Wang, R.F. Sekerka, A.A. Wheeler, B.T. Murray, S.R. Coriell, R.J. Braun, and G.B. McFadden, Physica D **69**, 189 (1993).

[17] A. Karma and W.-J. Rappel, Phys. Rev. E **53**, 3017 (1996).

[18] For a recent review of phase field models, see, e.g., A.A. Wheeler, J. Stat. Phys. **95**, 1245 (1999).

[19] D.M. Anderson, G.M. McFadden, and A.A. Wheeler, Physica D **135**, 175 (2000).

[20] V. Popa-Nita and T.J. Sluckin, J. Phys. II **6**, 873 (1996); V. Popa-Nita, T.J. Sluckin, and A.A. Wheeler, *ibid.* **7**, 1225 (1997).

[21] P. Ziherl, A. Šarlah, and S. Žumer, Phys. Rev. E **58**, 602 (1998); A. Šarlah and S. Žumer, *ibid.* **60**, 1821 (1999).

[22] For a review of these models, see, e.g., T.J. Sluckin and A. Poniewierski, in *Fluid Interfacial Phenomena*, edited by C. A. Croxton, (New York, Wiley, 1986), Chap. 5, and B. Jérôme, Rep. Prog. Phys. **54**, 391 1991.

[23] S. Hess, Z. Naturforsch. A **31a**, 1507 (1976).

[24] P.D. Olmsted and P. Goldbart, Phys. Rev. A **41**, 4578 (1990); **46**, 4966 (1992).

[25] P.G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd ed. (Oxford University Press, Oxford 1993).

[26] P. Oswald, P. Pieranski, *Les Cristaux Liquides* (Gordon and Breach Science New York, 2000).

[27] F.C. Frank, Discuss. Faraday Soc. **25**, 19 (1958).

[28] V.G. Levich, *Physicochemical Hydrodynamics* (Prentice-Hall, Englewood Cliffs, N.J.)

[29] U-Ser Jeng, L. Esibov, L. Crow, and A. Steyerl, J. Phys.: Condens. Matter **10**, 4955 (1998).

[30] J. Jäckle, J. Phys.: Condens. Matter **10**, 7121 (1998).

[31] L.D. Landau and E.M. Lifshitz, *Fluid Mechanics* (Pergamon Press, Oxford, 1959).

[32] O.N. de Mesquita, Braz. J. Phys. **28**, 257 (1998).

[33] J.S. Langer, Ann. Phys. (N.Y.) **41**, 108 (1967); J. Zittartz, Phys. Rev. **154**, 529 (1967); A.J. Bray, Phys. Rev. E **58**, 1508 (1998); W. van Saarloos, Phys. Rep. **301**, 9 (1998).