Incommensurable and helicoidal structures in nematic liquid crystals in electric fields

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A generalized thermodynamic potential is employed to describe systems of spindles that form during electrohydrodynamic instability in nematic liquid crystals, and the potential is related to the elasticity theory for smectics. It is shown that the incommensurability between the periods of the initial system and applied modulated field leads to the formation of a two-dimensional lattice of twisted spindles previously observed experimentally.

INTRODUCTION

The $D_{\infty h}$ symmetry of nematic liquid crystals (NLC) should rule out helicoidal structures, provided the NLC is in equilibrium. However, steady-state nonequilibrium dissipative structures such as Williams domains can form in applied electric fields. Our theory of dissipative structures developed below predicts that spindle systems of nontrivial symmetry can form during electrohydrodynamic (EHD) instability and provides a qualitative explanation for the recent experimental findings in Ref. 1.[1)]

Various qualitative and quantitative perturbation-theoretical techniques have been developed for describing the essentially nonlinear behavior of systems with EHD instability. However, the physics of nonlinear isotropic liquid crystals (LC) is very diverse and much remains to be explained. Progress has been hindered partly because of the inherent complexity of these systems and partly because of the lack of a reasonably simple theoretical formalism.

In this paper we introduce new variables which are particularly well suited for describing EHD instability. These variables appear as order parameters in the generalized thermodynamic potential which we employ to find novel spatially periodic structures in a system of EHD spindles.

1. DERIVATION OF THE FUNCTIONAL

In this section we will briefly outline the derivation of the thermodynamic functional; the reader may consult Ref. 2 for details. We start with the system of equations

$$I\frac{d}{dt}\boldsymbol{\Omega}=[\mathbf{nh}]-\boldsymbol{\Gamma},\quad \rho\frac{d}{dt}\mathbf{V}=\mathbf{f}+\mathbf{g};$$
$$\operatorname{div}\mathbf{V}=0,\quad \operatorname{div}\mathbf{D}=4\pi q, \tag{1}$$
$$\operatorname{rot}\mathbf{E}=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t},\quad \frac{\partial q}{\partial t}+\operatorname{div}\mathbf{J}=0,$$

which describe an LC in the hydrodynamic approximation.[3] Here the notation [**AB**] denotes the vector product $\mathbf{A}\times\mathbf{B}$; I is the inertial moment per unit volume of the NLC; $\boldsymbol{\Omega}$ is the angular velocity in the direction of the unit vector **n**; **f** and **g** are the forces associated with the Maxwell and LC stress tensors, respectively; **h** is the molecular field; $\boldsymbol{\Gamma}$ is the torque; **V** is the hydrodynamic velocity of the LC; q, J, and ρ are the charge, current, and mass densities of the LC. The remaining terminology is standard.

Let an NLC film of thickness l lie in the xy plane with an external electric field $\mathbf{E}_0$ applied normal to it (along the z axis). We will assume for definiteness that the unperturbed NLC molecules are oriented by glass surfaces (plates) so that they are aligned along the x axis. It has been found experimentally that LCs become electrodynamically unstable (mechanical equilibrium is lost) for fields E exceeding a critical strength E_c. We are interested in describing the behavior that occurs for E slightly greater than E_c. It will be helpful to introduce the 9-dimensional vector

$$\mathbf{U}\equiv(\mathbf{V},\ \mathbf{n},\ \mathbf{E}) \tag{2}$$

in the analysis of Eqs. (1), which we rewrite symbolically as

$$\hat{L}[\mathbf{U}]=0. \tag{3}$$

The matrix form of the differential operator $\hat{L}$ follows directly upon comparing (1) with (3) and using the definition (2). We will use perturbation theory to solve (3) for $E\approx E_c$ by taking the solution of the linearized form of system (1)

$$\hat{L}_0[\mathbf{U}_0]=0 \tag{4}$$

as the lowest-order approximation. Just as in Ref. 2, Eq. (4) is easily seen to have the solution

$$\mathbf{U}_0(t)=\mathbf{U}_0 e^{-\lambda t}. \tag{5}$$

One of the damped modes is found to have the damping rate

$$\lambda_1=-\varepsilon\lambda_s. \tag{6}$$

Here λ_s is the damping rate in the absence of the external field ($E_0=0$), and

$$\varepsilon=\frac{R-R_c}{R_c},\quad R=\frac{\rho l^2 E_0^2}{\alpha_0^2}, \tag{7}$$

where α_0 is the viscosity. The damping rate λ_1 thus tends to zero as $E_0\to E_c$, and EHD instability can develop. We take ε to be the small parameter in the perturbation expansions.

The simplest type of expansion is of the form

$$\mathbf{U}(t)=\mathbf{U}_0(t)+\varepsilon\mathbf{U}_1(t)+\varepsilon^2\mathbf{U}_2(t)+\dots.$$

However, more accurate perturbation expansion can be derived by recalling that the linear theory implies that a change

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in R by εR will scale the dimensions of the characteristic fluctuations by ε^{-1} and $\varepsilon^{-1/2}$ along the x and y axes, respectively, while the fluctuation times are scaled by ε^{-2}. We thus follow Ref. 4 and define new variables ξ, η, and τ by

$$x=\xi/\varepsilon, \quad y=\eta/\varepsilon^{1/2}, \quad t=\tau/\varepsilon^2 \tag{8}$$

and make the substitutions

$$\partial_x\to\partial_x+\varepsilon\partial_\xi, \quad \partial_y\to\partial_y+\varepsilon^{1/2}\partial_\eta, \quad \partial_t\to\partial_t+\varepsilon^2\partial_\tau. \tag{9}$$

We then expand $\hat{L}$ as a series

$$\hat{L}=\hat{L}_0+\varepsilon^{1/2}\hat{L}_{(1/2)}+\varepsilon^{3/2}\hat{L}_{(3/2)}+\varepsilon^2\hat{L}_{(2)}+\ldots \tag{10}$$

and set the coefficients of each power of ε equal to zero. Carrying out the same procedure for $U(t)$, we thus get a different set of perturbation-theoretic approximations.

We will seek a solution of the nonlinear equation (3) of the form[2)]

$$\mathbf{U}=W\mathbf{U}_0+W^*\mathbf{U}_0^*. \tag{11}$$

As usual in the analysis of hydrodynamic instability, we will derive the equation for the amplitude W by including random noise (thermal fluctuations) in addition to the nonlinear terms. If we confine ourselves to the nonlinearities of lowest order in W, we can truncate the series at ε^3. The Langevin equation for W then takes the form

$$\begin{aligned}C_0\partial_\tau W=&[C_1(R-R_c)-C_2|W|^2]W+C_3\partial_\xi^2 W\\&-iC_4\partial_\xi\partial_\eta^2 W-C_5\partial_\eta^4 W+2\gamma(\xi,\eta,\tau).\end{aligned} \tag{12}$$

Elaborate expressions are available for the dimensionless coefficients C_i in terms of the NLC parameters (cf. Ref. 2). The quantity γ in (12) describes random forces whose correlation can be found by the standard technique.

Equation (12) can be reduced by the familiar procedure[4] to the Fokker-Planck equation for the probability density distribution for the amplitudes W, W^*; following Ref. 2, we can use the latter equation to derive the functional

$$\begin{aligned}\Phi=\int\int dx\,dy\{&-(R-R_c)C_1|W|^2+C_2|W|^4+C_3|\partial_x W|^2\\&+iC_4(\partial_x W\partial_y^2 W^*-\partial_x W^*\partial_y^2 W)+C_5|\partial_y^2 W|^2\},\end{aligned} \tag{13}$$

which approximately describes the properties of the NLC for E_0 slightly above the threshold E_c for EHD stability. Here we have reverted to the original variables x, y, z, t.

If we use the fact that

$$\begin{aligned}&C_3|\partial_x W|^2+iC_4(\partial_x W\partial_y^2 W^*-\partial_x W^*\partial_y^2 W)+C_5|\partial^2 W|^2\\&=C_3\left|\partial_x W-\frac{C_4}{C_3}\partial_y^2 W\right|^2+\left(C_5-\frac{C_4^2}{C_3}\right)|\partial_y^2 W|^2\end{aligned}$$

and recall that $C_4\sim k_0$ (Ref. 2), it is clear that (13) is unchanged if x is replaced by $-x$. The functional is therefore invariant under the replacements $x\to -x_0$, $k_0\to -k_0$, where k_0 is the wave vector of the original (unperturbed) system of spindles aligned along the x axis.

The estimates for MBBA in Ref. 2 show that the coefficients of the first two terms in (13) are numerically much larger than the others, which involve derivatives. We may thus assume that in the long-wavelength approximation $|W|$ remains constant as W varies in space (cf., e.g., the "modulus conservation principle" in Ref. 5). This prompts us to write

$$W=|W_0|e^{i\varphi}, \quad W^*=|W_0|e^{-i\varphi}, \quad |W_0|\approx\text{const}, \tag{14}$$

where

$$\varphi=k_0X(x,y). \tag{15}$$

This substitution reduces the functional (13) to

$$\begin{aligned}\Delta\Phi=k_0^2|W_0|^2\int\int dx\,dy\{&C_3(\partial_x X)^2+2k_0C_4(\partial_x X)(\partial_y X)^2\\&+k_0^2[C_5(\partial_x X)^4+C_5(\partial_y^2 X)^2]\}.\end{aligned} \tag{16}$$

2. HELICOIDAL STRUCTURES

The term with the coefficient C_4 in (16) can give rise to new qualitative behavior; in particular, the spindles may become twisted helicoids. This twisting seems to have been first observed experimentally only recently in Ref. 1, where Williams domains were studied. The domains were produced by a slight modification of the standard technique—an additional voltage, which was spatially modulated along the x axis and exceeded the bias voltage by 0.6 V, was applied to one of the plates. The spindle system was deformed and two-dimensional structures were observed (these structures are clearly seen in Fig. 3 in Ref. 1).

In order to satisfactorily describe the experimental situation using the potential derived above, we must include another term which arises from the effects of the additional voltage on the LNC plate. The form of this term can be found by the following arguments.

Strictly speaking, if a weak external periodic electric field is applied to an NLC in addition to a bias field, the problem must generally be reformulated and the potential rederived. However, the effects of the periodic field can be accommodated by the functional (16) if we note that only the long-wavelength contribution is important, because the period of the applied field is comparable to the distance between the spindles and is much less than the characteristic scale of the inhomogeneities described by (16) (the spindle system is regarded as a continuous medium). The longwave contribution is due to beating associated with the small difference between the periods of the applied external and modulated self-consistent internal electric fields. This method of treating the beat contribution to the long-wavelength part of the dielectric permittivity is familiar from solid-state theory (cf., e.g., Ref. 6).

The first term in the functional (13) gives the dominant contribution to the electric field ($R\propto E_z^2$). We specify the external modulated field to be of the form

$$E_z^b=\Delta E\cos(k_1x) \tag{17}$$

and write

$$E_c=E_1\cos[k_0x+\varphi(x)] \tag{18}$$

for the internal self-consistent field. The phase φ in (18) allows for the effect of the external modulated field and varies slowly over distances $\sim 1/k_0$. We will derive an expression for $\varphi(x)$ below from the condition that the free energy be a minimum.

Under the experimental conditions in Ref. 1,

$$k_0 = {}^3/_2 k_1 + \Delta_0, \quad 0 < \Delta_0 \ll k_0. \tag{19}$$

The expression for E_z^2 contains the product of the fields (17) and (18), averaged over the period $2\pi/k_0$. As a result, an additional term of the form

$$R_c\left(\frac{\Delta E}{E_0}\right) C_1 \mu_1 |W_0|^3 \Delta_0 \sin^2 \frac{\Delta_0 x - \varphi}{2} \tag{20}$$

appears in the functional (13). Here $|W_0|$ is the modulus of the order parameter. Formula (37) in Ref. 2 gives an explicit expression for μ_1; here we simply note the estimate $\mu_1 \approx -1.5 \cdot 10^4$ for MBBA. We observe that (20) differs from the term postulated in Ref. 1, where the important fact that the system is sensitive not to the individual phases but to their difference was overlooked.

Now that we have the full expression for the generalized thermodynamic potential appropriate to the experimental conditions, we can use it to study the two-dimensional structures. We note that the specific sign of Δ_0 is of little importance in the subsequent calculations; only the fact that Δ_0 does not change sign is important. Again, we will assume that the modulus of the order parameter is independent of the spatial coordinates, so that we need examine only the coordinate dependence of the phase.

Following Kantorovich,[7] we seek to minimize the functional by a function of the form

$$W = W_0 e^{i\varphi} e^{ik_y y}, \tag{21}$$

where we have used the fact that the NLC system is unbounded along the y axis. Here $\varphi(x)$ is the desired phase and the parameter k_y is to be determined. The corresponding functional (per unit area of the NLC film) is

$$\Delta\tilde{\Phi} = \frac{1}{l_x}\int_0^{l_x} dx \left\{ 2C_3 W_0^2 \left(\frac{d\varphi}{dx}\right)^2 + 2C_4 k_y^2 W_0^2 \frac{d\varphi}{dx} + 2C_5 k_y^4 W_0^2 - C_6 W_0^3 \Delta_0 \sin^2 \frac{\Delta_0 x - \varphi(x)}{2} \right\}, \tag{22}$$

where $C_6 \equiv R_c C_1 (\Delta E/E_0)|\mu_1|$ and l_x is the characteristic length over which $\varphi(x)$ varies. We minimize the functional (22) by solving the associated Euler equation, which simplifies when expressed in terms of the function $\varphi_1 = -(\varphi - \Delta_0 x)/2$. Function (22) then becomes

$$\Delta\tilde{\Phi} = \frac{1}{l_x}\int_0^{l_x} dx \left\{ \frac{B}{2}\left(\frac{d\varphi_1}{dx}\right)^2 - b\frac{d\varphi_1}{dx} - \alpha \sin^2 \varphi_1 \right\} + 2C_5 W_0^2 k_y^4 \tag{23}$$

where we use the terminology

$$B/2 = 4C_3 W_0^2, \quad b = 8\Delta_0 C_3 W_0^2 + 4C_4 W_0^2 k_y^2, \quad \alpha = C_6 \Delta_0 W_0^3 \tag{24}$$

adopted in Ref. 8. We have thus reduced the problem to the one considered in Ref. 8, with the formal difference that the coefficient b contains a new variational parameter k_y^2.

If we substitute the solution of the Euler equation found in Ref. 8 into (29), we get

$$\Delta\tilde{\Phi} = -4\alpha \left\{ \frac{\pi}{2\varkappa_0} - 1 - (1-\varkappa)\left(1 - \frac{\pi}{4\varkappa_0}\right) \right\} \times \ln^{-1}\frac{8}{1-\varkappa} + 2C_5 W_0^2 k_y^4. \tag{25}$$

Here $\varkappa_0 = (2\alpha B)^{1/2}/b$, and the variational parameter $\varkappa$ introduced in Ref. 8 is related to the first integral of the Euler equations by the formula

$$\frac{B}{2}\left(\frac{d\varphi_1}{dx}\right)^2 + \alpha \sin^2 \varphi_1 = \frac{\alpha}{\varkappa^2}. \tag{26}$$

Unlike the case in Ref. 8, here we must also minimize (25) with respect to the parameter k_y^2.

The value of k_y^2 for which (25) is a minimum is given by

$$k_y^2 = \frac{\pi C_4}{C_5}\left(\frac{\alpha}{2B}\right)^{1/2} (3-\varkappa)\ln^{-1}\frac{8}{1-\varkappa} = \frac{\pi}{4}\frac{C_4}{C_5}\left(\frac{W_0\Delta_0 C_6}{C_3}\right)^{1/2} (3-\varkappa)\ln^{-1}\frac{8}{1-\varkappa}. \tag{27}$$

Substituting into (25) and differentiating with respect to $\varkappa$, we get the condition

$$1 + \frac{2}{1-\varkappa}\ln^{-1}\frac{8}{1-\varkappa}\left[\left(\frac{\pi}{2\varkappa_0} - 1\right) - \frac{\pi^2}{4}\frac{C_4^2}{C_3 C_5}\ln^{-1}\frac{8}{1-\varkappa}\right] = 0 \tag{28}$$

for the minimum. Equation (28) coincides with the result in Ref. 8, apart from the last term in the square brackets. However, this term tends to zero near the phase transition ($\varkappa \to 1$), whereas the first term in the square brackets remains constant.

The phase transition point is thus determined by solving a strictly one-dimensional problem. The corrections for the two-dimensionality along the x axis are small near the phase transition point. A periodic structure therefore exists along the x axis with the same period as was found in Ref. 8; in our notation, the period is

$$l_x \approx -(4/\pi\Delta_0)\ln\delta, \quad \delta = \pi/2\varkappa_0 - 1, \quad 1-\varkappa \approx -2\delta/\ln\delta. \tag{29}$$

We recall that all lengths are divided by the thickness of the NLC film. Unlike Ref. 8, however, in our case the x-periodic structure is also accompanied by a y-periodic structure with period determined by (27). We see from (27) that the latter period is uniquely determined by the spatial structure along the x axis.

We have thus shown that the two-dimensional periodic structures observed in Ref. 1 can form when a weak external periodic electric field is applied to an EHD spindle structure whose period is incommensurable with the period of the field.

We have thus far confined our analysis to purely two-dimensional systems. However, it should be noted that even though the system is bounded along the z axis, spindle deformations of the type considered above may also occur along the z direction. Indeed, because of the preferential alignment of the director along the x axis, each spindle may be regarded as a highly anisotropic "elastic rod"; spindle deformations in the x-y plane will thus inevitably produce deformations along the

z axis. Since the film is bounded along the z axis, realignment of the spindles so that they are no longer parallel to the top and bottom of the film should be accompanied by periodic breakage along the y axis. The combined deformation in both the xy and the yz planes twists the spindles, and the periodic breakage along the y axis can be regarded as a type of "helicoidal" structure in bounded systems. In the foregoing analysis we used the boundedness of the system along the z axis to justify neglecting the z-dependence of the thermodynamic functional. The latter thus actually describes a two-dimensional projection of a three-dimensional problem.

We note that the twisting of the spindles in the yz plane is particularly pronounced if the spindle diameter is appreciably less than the thickness of the plate. This can occur, e.g., when high-frequency chevron-type domains are formed. Such domains cannot be described directly by means of the above functional, because the latter was derived assuming a single amplitude for all of the components of the vector (2) in function space; however, a similar, suitably modified functional should also apply to the high-frequency case. It should be possible to treat the different relaxation times of the vector components (2) in this case by increasing the number of tensor dimensions of the order parameter. Because the spatial periods of the components (2) also differ in the high-frequency case, the beating effects noted above may give rise to incommensurable structures even when no external spatially modulated field is applied. We have already noted that the spindles should then be more highly twisted than was found in the case analyzed above. However, the detailed investigation of chevron domains in terms of generalized thermodynamic potentials constitutes a separate problem.

In closing, we note that in general the thermodynamic potential should also involve a term of the form $(\partial X/\partial Y)^2$ which describes the anisotropy energy of the spindles. This term will be present if the projection from the three-dimensional to the quasi-two-dimensional case is carried out by averaging the functional over the thickness of the plate in order to allow for the rigid boundary conditions for the director. The structures considered above will then appear only above a certain threshold. This case readily be seen if $\alpha \ll 1$; indeed, in this case expression (23) simplifies to

$$\Delta\Phi = \left(-4\Delta_0^2 C_3 - 4\Delta_0 C_4 k_y^2 - \frac{C_4^2}{C_3} k_y^4 + 2C_5 k_y^4\right) W_0^2. \quad (30)$$

If we now include the term associated with the spindle anisotropy, we get

$$\Delta\Phi = [-k_y^2(4\Delta_0 C_4 - C_7) + k_y^4(2C_5 - C_2/C_3) - 4\Delta_0^2 C_3] W_0^2,$$

in place of (30). We note that by the above arguments, the contribution from the anisotropy term is given by the additional term in the curly brackets in (16) and is of the form $C_7(\partial_y X)^2 \propto C_7 k_y^2$. The two-dimensional structures will thus form when $\Delta_0 \geqslant C_7/4C_4$.

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[1)]We are grateful to E. I. Kats for informing us of this work.
[2)]Reference 2 gives the explicit expression for U_0 needed to calculate the coefficients C_i in the functional (cf. below).

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