

Transient patterns in nematic liquid crystals: Domain-wall dynamics

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We study the dynamics of the late stages of the Fréedericksz transition in which a periodic transient pattern decays to a final homogeneous state. A stability analysis of an unstable stationary pattern is presented, and equations for the evolution of the domain walls are obtained. Using results of previous theories, we analyze the effect that the specific dynamics of the problem, incorporating hydrodynamic couplings, has on the expected logarithmic domain growth law.

I. INTRODUCTION

In the general context of pattern-formation problems, an interesting question concerns the description of the emergence and disappearance of transient spatial structures. Such a situation occurs, for example, in the Fréedericksz transition when a magnetic field whose intensity exceeds a definite threshold is applied to a uniformly aligned nematic liquid crystal.^{1,2} Molecules reorient locally in opposite directions, giving rise to a transient pattern consisting, for a certain geometry, of parallel stripes of a well-defined periodicity. At later times the pattern decays, leading to a uniform reorientation throughout the plane of the sample. The appearance of this transient structure is due to backflow effects which couple the director and velocity fields, resulting in a finite-wavelength instability. The dynamical stages of this pattern-formation phenomenon find formal analogies in the problem of spinodal decomposition.³ Using this analogy a description of this transient process has been given⁴ in terms of the time-dependent structure factor of orientational fluctuations. The dynamics of the structure factor is based on the equations of stochastic nematodynamics.⁴ That description is limited to the dynamical stages during which the pattern forms but it does not account for the late dynamical stages during which the pattern decays giving place to an homogeneous distortion. Our aim in this paper is precisely the study of such late-stage dynamics. In fact, our analysis here is also motivated by a recent digital video experiment² which, in view of the experimental limitations found when following the initial behavior of the reorientational process,⁵ suggest the convenience of focusing on the final evolution towards the new equilibrium configuration. An additional motivation for this study is that a recent work⁶ on propagating fronts in bistable systems establishes a different mechanism to produce a transient periodic pattern of the same nature. Our analysis might also be relevant to describe the decay dynamics of such pattern.

The decay of the periodic pattern is essentially a one-dimensional problem dominated by wall dynamics.⁷

These parallel walls separate the different equivalent orientations of the molecules. The one-dimensional wall dynamics is certainly not a new problem. An early study of one-dimensional domain growth due to wall dynamics is due to Langer⁸ in the context of spinodal decomposition. An extension of this approach which considers the coupling of conserved and nonconserved field variables has been also reported.⁹ These studies are based on a linear stability analysis of a periodic configuration of walls. On the other hand another scheme which consists of the reduction of the original field equations to equations for the wall dynamics has been proposed by Kawasaki and Ohta.¹⁰ From these reduced equations domain growth can be studied.¹¹ The work of Langer,⁸ Kawasaki and Nagai,¹¹ and other similar numerical and analytical studies¹² lead to a domain growth law in which the average domain size grows logarithmically in time. This can also be anticipated from general arguments which result in an exponentially decaying attractive force between walls.¹³ However, concepts like forces between walls are static concepts related to a free energy and they do not determine the detailed dynamics which depends on the specific kinetic operators of any particular system. To insist on that, we remark that both the one dimensional dynamics of a conserved and that of a nonconserved order parameter lead to an asymptotic logarithmic growth, $\ln t^\nu$, but the exponent ν is different in the two cases.¹⁴ This comes from the fact that those dynamics are intrinsically different. These are the kind of interesting questions which merit some consideration and which we address in detail in this paper. Essentially, we are interested in elucidating how the wall dynamics is affected by the peculiar kinetics involved in the pattern formation associated with the Fréedericksz transition. The director field obeys a nonconserved dynamics with a constant kinetic coefficient given by the pure reorientational viscosity. The consideration of the coupling with the velocity flow leads to an effective wave-number-dependent viscosity which modifies the dynamics of the director. The central question is how such effective kinetic coefficient modifies the well-known one-dimensional dynamics of

walls for a nonconserved order parameter.¹⁰⁻¹³ In any case the logarithmic growth law is expected to hold asymptotically.

Our study of the decay of the transient pattern of opposite reorientations starts in Sec. II with a linear stability analysis of a periodic configuration of Fréedericksz walls modeling the internal nematic disposition. This suffices to identify the instability and to obtain a characteristic time of evolution of the pattern. Our results indicate that in the eigenvalue problem backflow effects just lead to a numerical redefinition of a kinetic coefficient. This anticipates the result of Sec. III in which we show that the Fréedericksz wall dynamics can be mapped into the problem of kink dynamics for a scalar nonconserved order parameter with the corresponding change of time scale resulting from the coupling between director and velocity flows. This result is obtained by reducing, for late stages, the original dynamics of the director angle configuration to that of the positions of the Fréedericksz walls. In this context the separation between interaction force between walls and dynamic effects is discussed. From the analysis of the wall dynamics the domain growth problem can be addressed. As expected, the domain growth law is found to be logarithmic but in a time scale renormalized in terms of the parameters involved in the hydrodynamic coupling and with an exponent independent of backflow effects. Some technical calculations are summarized in an appendix.

II. STABILITY ANALYSIS OF A PERIODIC PATTERN

We consider a nematic liquid crystal sample with positive anisotropic magnetic susceptibility χ_a contained between two plates perpendicular to the z axis. The director is initially aligned along the x axis [$\mathbf{n}^0=(1,0,0)$] and a magnetic field is initially applied along the y axis. A periodic pattern consisting of stripes perpendicular to the x axis appears^{1,2} when applying a magnetic field exceeding some critical intensity. The local, equivalent but opposite reorientations of the director can be described⁴ in terms of two variables: $\phi(x,z;t)$, a distortion angle in the x - y plane, and $v_y(x,z;t)$, the corresponding component of the hydrodynamic flow. We focus on the behavior of the amplitude $\bar{\theta}(x,t)$ of the most unstable z mode of $\phi(x,z,t)$. We neglect thermal noise effects since we are interested in the late stages of this reorientational dynamics. In addition, we make the usual approximation of negligible inertia, so that the pair of equations we are finally led to consider are⁴

$$\begin{aligned} \partial_t \bar{\theta}(x;t) = & -(1/\gamma_1)[(K_2 \pi^2/d^2 - K_3 \partial_x^2 - \chi_a H^2) \bar{\theta} \\ & + \frac{1}{2} \chi_a H^2 \bar{\theta}^3] + \frac{1}{2} (\lambda + 1) \partial_x v_y, \\ \frac{1}{2} (\lambda + 1) \partial_x [(K_2 \pi^2/d^2 - K_3 \partial_x^2 - \chi_a H^2) \bar{\theta} + \frac{1}{2} \chi_a H^2 \bar{\theta}^3] \\ & + (v_3 \partial_x^2 - v_2 \pi^2/d^2) v_y = 0, \end{aligned} \quad (2.1)$$

where K_2 and K_3 respectively stand for twist and bend elastic constants; γ_1 , λ , v_2 , and v_3 are viscosity related pa-

rameters; and d is the width of the sample along the z axis.

A closed equation for the angular variable $\bar{\theta}$ is readily obtained by formally inverting the equation for v_y . In doing this algebra it turns out appropriate to consider dimensionless space and time variables defined in terms of field-dependent characteristic length and time scales $x \equiv \xi u$ and $t \equiv \tau_0 s$,

$$\xi^2 \equiv 2 \frac{K_3}{K_2 \pi^2/d^2} \frac{1}{h^2 - 1}, \quad \tau_0 \equiv \gamma_1 \frac{1}{K_2 \pi^2/d^2} \frac{1}{h^2 - 1}, \quad (2.2)$$

where h is the reduced magnetic field $h = H/H_c$, with H_c being the critical field for the Fréedericksz transition $H_c = (K_2 \pi^2/\chi_a d^2)^{1/2}$.

Scaling the dynamical variable correspondingly, $\theta^2 \equiv \bar{\theta}^2/2(1-h^{-2})$, we formally have

$$\partial_s \theta(u;s) = L(\partial_u) [\frac{1}{2} \partial_u^2 \theta + V'(\theta)], \quad (2.3)$$

where the kinetic operator $L(\partial_u)$ explicitly reads

$$L(\partial_u) = 1 + \frac{1}{4} \gamma_1 (1 + \lambda)^2 \left[v_3 \partial_u^2 - v_2 \frac{2K_3}{K_2 (h^2 - 1)} \right]^{-1} \partial_u^2, \quad (2.4)$$

and the potential $V(\theta)$ is given by $V(\theta) = \frac{1}{2} \theta^2 - \frac{1}{4} \theta^4$.

A nonhomogeneous stationary solution of (2.3) is readily identified as the profile connecting the two possible equivalent homogeneous stationary configurations $\bar{\theta}^2 = 2(1-h^{-2})$, of the Fréedericksz instability. This stationary profile is completely independent of the kinetic operator L . It corresponds to the well-known Fréedericksz wall, and reads¹⁵

$$\theta_w(u) = \tanh(u), \quad (2.5)$$

or in the original variables

$$\bar{\theta}_w(x) = [2(1-h^{-2})]^{1/2} \tanh(x/\xi). \quad (2.6)$$

The width of the wall is directly given in terms of the unit length ξ which manifestly diverges at the transition point $h^2=1$. The wall solution (2.6) is obtained in the usual small-amplitude expansion of the magnetic free energy implicit in (2.1). Although a more complete treatment of nonlinear terms might be desirable for large fields, this does not affect our main concern here, which is to describe the effects of hydrodynamic flow on the kinetics of domain walls and on the associated domain growth law.

The early stages of the dynamics described by (2.3) lead to a periodic pattern⁴ which we identify as a regular array of alternate solutions of the sort obtained above. A purely periodic configuration of Fréedericksz walls constitute an unstable stationary solution of (2.3). We are interested in the dynamical decay of this type of solution. In what follows we first discuss the linear stability analysis of such a periodic configuration $\theta_s(u)$: If χ_n and ϵ_n stand for the eigenfunctions and corresponding eigenvalues of this linear stability analysis, respectively, we directly obtain from (2.3)

$$\theta(u; s) - \theta_s(u) = \sum_n \chi_n(u) e^{-\varepsilon_n s}, \quad (2.7)$$

$$L(\partial_u) \left[-\frac{1}{2} \partial_u^2 - 1 + 3\theta_s^2(u) \right] \chi_n(u) = \varepsilon_n \chi_n(u), \quad (2.8)$$

with $3\theta_s^2(u) - 1$ being the term associated with the periodic organization of kinks and antikinks (Fréedericksz walls of opposite sign),

$$3\theta_s^2(u) - 1 = 2 - 3 \sum_{j=0}^{M-1} \text{sech}^2(u - j\rho). \quad (2.9)$$

In particular (2.9) refers to a situation consisting of M walls with spacing ρ . In terms of the original space variable, $l = \rho\xi$ denotes the unscaled spacing while $L = Ml$ corresponds to the typical length of the nematic sample in the x direction. In the calculation below we make an extensive use of the fact that $e^{-\rho} \ll 1$,¹⁶ since we consider situations away from the instability.

The strategy to solve (2.8) follows Langer's work⁸ for a similar problem in the context of the dynamics of the spinodal decomposition. The basic idea consists of transforming (2.8) into a variational problem which is solved by using an appropriate set of trial functions. We define a Schrödinger Hamiltonian-like operator F which corresponds to the left-hand side of (2.8) except for the kinetic operator L . In addition a set of conjugate states $\tilde{\chi}_n(u)$ are introduced satisfying

$$F \equiv -\frac{1}{2} \partial_u^2 - 1 + 3\theta_s^2(u), \quad (2.10)$$

$$L(\partial_u) \tilde{\chi}_n(u) = \chi_n(u). \quad (2.11)$$

Assuming periodic boundary conditions for both sets χ_n and $\tilde{\chi}_n$ it is not difficult to see that the variational problem is well posed¹⁷ and that finally reduces to

$$\varepsilon_{n, \min} \leq (\chi_n, F\chi_n) / (\tilde{\chi}_n, \chi_n). \quad (2.12)$$

The scalar products in (2.12) are to be understood in the usual way,

$$(A, B) = \int_0^{M\rho} A(u)B(u)du. \quad (2.13)$$

We select a set of mutually orthogonal trial functions specified by wave numbers q ,

$$\chi_q(u) = \frac{1}{\sqrt{M}} \sum_{j=0}^{M-1} e^{iq\rho j} \text{sech}^2(u - j\rho), \quad (2.14)$$

with

$$q = (2\pi/M\rho)I,$$

where I is an integer, due to the periodic conditions prescribed for the problem.

The task of computing the numerator in (2.12) is greatly simplified by keeping only first-order overlap integrals, the final result being⁸

$$(\chi_q, F\chi_q) \simeq -64e^{-2\rho}(1 + \cos q\rho). \quad (2.15)$$

The denominator in (2.12) contains the dynamical effects associated with the kinetic operator L . Its evaluation is more involved than that of the numerator. First it is necessary to solve for the auxiliary set of functions $\tilde{\chi}_q$ and

then to perform the corresponding scalar products. As we mentioned earlier the whole procedure is greatly simplified taking ρ as a large parameter. That enables us to make a series of rounding approximations, as, for example, the replacement of $\tanh(u)$ by a step function $\Theta(u)$ which causes errors of order $e^{-\rho}$. We relegate the technical details to the Appendix. The final result reads

$$\Gamma \equiv (\tilde{\chi}_q, \chi_q) \simeq 2\alpha \left[\frac{2}{h^2 - 1} \beta \right]^{1/2} + \frac{4}{3}(1 - \alpha) \quad (2.16)$$

in terms of dimensionless material dependent parameters α and β ,

$$\alpha \equiv \frac{1}{4} \gamma_1 (1 + \lambda)^2 / [v_3 + \frac{1}{4} \gamma_1 (1 + \lambda)^2],$$

$$\beta \equiv (K_3/K_2)v_2 / [v_3 + \frac{1}{4} \gamma_1 (1 + \lambda)^2].$$

At the level of approximation required here, (2.16) neither shows an explicit q dependence nor depends on the spacing parameter ρ . This contrasts with Langer's result in the spinodal decomposition problem. Note that the final expression (2.16) manifestly incorporates hydrodynamic effects through the set of viscosity parameters involved in the backflow currents, but these hydrodynamic effects finally result in a sort of effective numeric renormalization. In other words, except for this constant and viscosity-dependent factor, the final result for the variationally determined eigenvalues of the linear stability problem are clearly analogous to those corresponding to a simple nonconserved dynamics³ in which the kinetic operator L is a numerical constant. The eigenvalues found here are essentially different to those found for dynamics with a conservation law, as in the problem of spinodal decomposition where L is proportional to ∂_u^2 . This point will be further discussed in Sec. III.

The examination of the set of obtained eigenvalues deserves some additional comments. First of all, the ε_q are always negative indicating the unstable nature of the periodic configuration of Fréedericksz walls. In addition one might obtain an estimate of the time scale involved in the relaxation of this unstable configuration in terms of a characteristic time defined by

$$\tau^{-1} \equiv \sum_q |\varepsilon_q| = 64\Gamma^{-1} M e^{-2\rho}. \quad (2.17)$$

A domain growth law can be inferred^{8,9} from this result for the characteristic time as explained in Sec. III. Finally we note that the wave-number dependence of the eigenvalues obtained in (2.15) and (2.16) is such that the maximum absolute value corresponds to the homogeneous mode $q=0$. This means that the corresponding more unstable eigenfunction $\chi_0(u) = (1/M^{1/2}) \sum_{j=0}^{M-1} \text{sech}^2(u - j\rho)$ distorts the array of equally spaced interfaces by equal amounts over each wall. The picture we can thus extract of the relaxation of this unstable configuration corresponds to the disappearance of one of the type of domains inside the other one. This mechanism explains the eventual destruction of the periodic pattern. It differs from the one proposed in problems with a conservation law in where the most unstable mode occurs for $q = \pm\pi/2\rho$.

III. EQUATIONS FOR DOMAIN-WALL DYNAMICS

The effective one-dimensional Eq. (2.3) is the starting point of a wall-dynamics approach to the study of the late-stage dynamics of the Fréedericksz transition. Equation (2.3) is a particular inertialess form of a general class of equations considered by Kawasaki and Ohta.¹⁰ These authors reduce the dynamics contained in such equations to the dynamics of walls, by deriving equations for the positions of the walls. Two basic assumptions are generally invoked in this procedure. First, the effects of collisions between walls are not taken into account. A second assumption is essentially an approximation concerning the range of the interaction between walls which is assumed to be limited to adjoint pairs of walls. From the analysis of Kawasaki and Ohta it follows that the locations $u_i(t)$ of the i th wall satisfies the equation

$$\sum_j (L^{-1}\theta'_{w,i}, \theta'_{w,j}) \dot{u}_j = R(u_{i+1} - u_i) - R(u_i - u_{i-1}), \quad (3.1)$$

where L is the kinetic operator (2.4), $\theta'_{w,i} = -(-1)^i d\theta_w(u)/du$ is the Goldstone mode associated with the zero eigenvalue of the operator obtained from (2.10) replacing θ_s by θ_w , and R admits a potential-dependent generic expression given by

$$\begin{aligned} R(u_{i+1} - u_i) &= -[\theta_{w,i}(u_{i+1} - u_i) - \theta_{w,i}(\infty)] \\ &\quad \times [\theta_{w,i}(\infty) - \theta_{w,i}(-\infty)] \\ &\quad \times V''(\theta_{w,i}(\infty)) \end{aligned} \quad (3.2)$$

For our particular problem it becomes

$$R(u_{i+1} - u_i) = 8 \exp[-2(u_{i+1} - u_i)]. \quad (3.3)$$

Equation (3.1) permits a simple interpretation of the essential ingredients involved in the wall dynamics. The right-hand side represents a balance of forces between a wall and its nearest neighbors. The attractive interaction between walls is given by $R(u)$ and it decreases exponentially with their separation apart. The second ingredient basically corresponds to the kinetic peculiarities of the problem described by the left-hand side. The interaction force $R(u)$ is a static concept independent of the particular kinetics. Therefore $R(u)$ does not take into account the detailed wall dynamics characterized by the kinetic operator L , which in our case contains backflow effects. Indeed, this effective interaction between walls could be generically obtained¹³ linearizing the equation of motion around one of the homogeneous stable states of the system.¹⁸

Let us now turn our attention to the left-hand side of (3.1), which is what is specific of our problem of the dynamics of Fréedericksz walls. According to our characterization of these walls (2.5), it is readily seen that the derivatives of this basic set of functions generate a corresponding set of $\text{sech}^2(u)$ functions. Thus, it is easily understood that in what refers to the particular task of evaluating the scalar products left in (3.1) we are not far

from what we did previously in our linear stability analysis when calculating the denominator (χ_q, χ_q) in (2.12). Indeed, invoking again the limit of large spacing between walls and neglecting exponentially small terms for $e^{-\rho} \ll 1$, the results of the Appendix show that the only term surviving in this limit in the sum of (3.1) corresponds to a diagonal contribution $(L^{-1}\theta'_{w,i}, \theta'_{w,i})$ which numerically coincides with the result for the kinetic coefficient Γ introduced in (2.16),

$$(L^{-1}\theta'_{w,j}, \theta'_{w,i}) \simeq (L^{-1}\theta'_{w,i}, \theta'_{w,i}) \delta_{ij} = \Gamma \delta_{ij}. \quad (3.4)$$

In summary the dynamical equation for the wall positions in the Fréedericksz transition is given by

$$\dot{u}_j = \Gamma^{-1} [R(u_{j+1} - u_j) - R(u_j - u_{j-1})], \quad (3.5)$$

where Γ is the kinetic coefficient calculated in (2.16). Equation (3.5) describes the drift motion of walls due to attractive forces. This equation turns out to be of the same form that for a simple nonconserved dynamics due to the result (3.4). For a conserved dynamics one finds contributions for $i \neq j$ which depend on the distance between the two walls.¹⁰

It is obvious that a stationary solution of (3.5), $u_{j+1} - u_j = u_j - u_{j-1} = \rho$, describes a completely periodic array of domain walls separated a distance ρ . This stationary solution corresponds to the periodic pattern whose stability was analyzed in Sec. II. The direct linear stability analysis of (3.5) around the stationary solution $u_{j+1} - u_j = u_j - u_{j-1} = \rho$, leads to eigenvalues of the form¹⁰

$$\epsilon_q = 4\Gamma^{-1} |R'(\rho)| \sin^2(q\rho/2). \quad (3.6)$$

Equation (3.6) may be then compared with (2.15) and (2.16). Actually, both calculations give the same values for their respective most unstable q modes. The fact that the most unstable q mode $q=0$ in (2.15) and (2.16) and $q = \pm\pi/\rho$ in (3.6) are different admits an easy interpretation if we note that when referred to the stability of the locations of the walls the "dissolution" of one type of domains into the other one, accomplished via the $q=0$ mode for $\theta_s(u)$, has a counterpart here in the opposite movement of wall positions as dictated by the mode $q = \pm\pi/\rho$.

Equation (3.5) is a good basis for studies of late-stage dynamics. As anticipated in Sec. II, the late-stage dynamics of the Fréedericksz transition is then equivalent to the rather well studied dynamics of a simple nonconserved order parameter in one dimension with a redefined kinetic coefficient Γ . In this sense it is possible to state that although backflow effects are responsible for the emergence of the pattern in the early stages, they only affect the late-stage dynamics through a renormalization of its time scale. Once the problem of the dynamics of Fréedericksz walls has been reduced to the standard situation for a nonconserved dynamics, a logarithmic growth law may be obtained by several procedures.

A first simple approach is based on the characteristic time τ , (2.17), introduced through the linear stability analysis of the periodic pattern. It might be interpreted as the rate at which the number M of domains changes in

time,⁸

$$\frac{dM}{dt} \simeq -\tau^{-1}. \quad (3.7)$$

The time dependence of the average domain size $\bar{z} = L/M$ is then obtained from (2.17) and (3.7) as

$$\frac{d\bar{z}}{dt} \sim \Gamma \bar{z} e^{-2\bar{z}}, \quad (3.8)$$

which indicates that asymptotically \bar{z} grows logarithmically in time in a time scale fixed by the renormalized kinetic coefficient Γ . Backflow effects appear only through Γ . The same equation (3.8) can be obtained¹⁹ considering the drift motion of kinks due to their attractive interactions.

A more careful study of the wall dynamics was reported in Ref. 11, where Kawasaki and Nagai presented a kinetic theory for the statistical dynamics of kinks. They obtained a Boltzmann-like equation for a domain-size distribution function $f(z, t)$. Using this scheme the asymptotic time dependence of the average domain size can be calculated. What we obtain, using (3.5) for the drift wall motion, is

$$\bar{z}(t) \sim \frac{1}{2} \ln(32\Gamma t). \quad (3.9)$$

This result is modified when including annihilation of pairs of walls upon contact, leading to an asymptotic law

$$\bar{z}(t) \sim \ln(t^\nu). \quad (3.10)$$

The way the factor ν appears in the theory¹¹ leads us to believe that it should coincide with the one corresponding to a simple nonconserved dynamics. This latter one has been estimated by numerical simulations as $\nu \simeq 3.5$.

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APPENDIX

We rewrite the kinetic operator L in terms of the parameters α and β introduced after (2.16),

$$L(\partial_u) = 1 + \alpha \left[(1-\alpha)\partial_u^2 - \frac{2}{h^2-1}\beta \right]^{-1} \partial_u^2. \quad (A1)$$

Next, we introduce two auxiliary operators L_0 and L_1 ,

$$L_0(\partial_u) \equiv 1 - \frac{(h^2-1)}{2} \frac{1}{\beta} \partial_u^2, \quad (A2)$$

$$L_1(\partial_u) \equiv 1 - \frac{(h^2-1)}{2} \frac{1}{\beta} (1-\alpha)\partial_u^2,$$

such that $L = L_1^{-1}L_0$. The denominator in (2.12) converts then into

$$(\bar{\chi}, \chi) = (L^{-1}\chi, \chi) = (L_1L_0^{-1}\chi, \chi), \quad (A3)$$

since L_0 and L_1 commute. Introducing a derived set of functions $\bar{\chi}$ such that $L_0^{-1}\chi \equiv \bar{\chi}$, and using the Hermitian

property of L_1 we have $(\bar{\chi}, \chi) = (\bar{\chi}, L_1\chi)$. From the definitions in (A2) it then turns out that $(\bar{\chi}, \chi)$ is given by

$$(\bar{\chi}, \chi) = \alpha(\bar{\chi}, \chi) + (1-\alpha)(\chi, \chi). \quad (A4)$$

Our first task will be to solve for $\bar{\chi}_q$,

$$\left[1 - \frac{(h^2-1)}{2} \frac{1}{\beta} \frac{d^2}{du^2} \right] \bar{\chi}_q(u) = \chi_q(u), \quad (A5)$$

in terms of the trial set of functions χ_q given in (2.14). In solving (5) we exhaustively invoke the limit of large ρ , and we impose periodic boundary conditions on $\bar{\chi}_q(u)$ and $d\bar{\chi}_q/du$,

$$\begin{aligned} \bar{\chi}_q(-\rho/2) &= \bar{\chi}_q(M\rho - \rho/2) \\ \int_{-\rho/2}^{M\rho - \rho/2} du \frac{d^2\bar{\chi}_q(u)}{du^2} &= 0, \end{aligned} \quad (A6)$$

where to avoid spurious problems we shift the boundaries away by an amount $\rho/2$ from the interfaces. The final result for $\bar{\chi}_q(u)$ reads

$$\begin{aligned} \bar{\chi}_q(u) &= \left[\frac{2}{h^2-1}\beta \right]^{1/2} \frac{1}{\sqrt{M}} \\ &\times \sum_{j=0}^{M-1} e^{iq\rho j} \exp \left[- \left[\frac{2}{h^2-1}\beta \right]^{1/2} |u - j\rho| \right] \\ &+ O(e^{-\rho}). \end{aligned} \quad (A7)$$

Now, once $\bar{\chi}_q$ has been determined we directly turn to the explicit calculations of the scalar products $(\bar{\chi}_q, \chi_q)$ and (χ_q, χ_q) in (4). In respect to $(\bar{\chi}_q, \chi_q)$ it decomposes into integrals of the type

$$\begin{aligned} \int_{-\rho/2}^{M\rho - \rho/2} du \exp \left[- \left[\frac{2}{h^2-1}\beta \right]^{1/2} |u - j\rho| \right] \\ \times \text{sech}^2(u - k\rho). \end{aligned} \quad (A8)$$

Once again in the limit $e^{-\rho} \ll 1$ it is easily checked that the unique terms of $O(1)$ are those corresponding to diagonal contributions $j = k$. They turn out to be identical and given by

$$\begin{aligned} \int_{-\rho/2}^{M\rho - \rho/2} du \exp \left[- \left[\frac{2}{h^2-1}\beta \right]^{1/2} |u - j\rho| \right] \\ \times \text{sech}^2(u - j\rho) = 2 + O(e^{-\rho}), \end{aligned} \quad (A9)$$

so that

$$(\bar{\chi}_q, \chi_q) = 2 \left[\frac{2}{h^2-1}\beta \right]^{1/2} + O(e^{-\rho}). \quad (A10)$$

Finally, for (χ_q, χ_q) we find again $O(1)$ diagonal contributions,

$$\int_{-\rho/2}^{M\rho - \rho/2} du \text{sech}^4(u - j\rho) = \frac{4}{3} + O(e^{-\rho}), \quad (A11)$$

and first- and higher-order overlap integrals for the non-diagonal products. They are exponentially small corrections to the leading diagonal terms in (11), so that

$$(\chi_q, \chi_q) = \frac{4}{3} + O(e^{-\rho}). \quad (\text{A12})$$

Finally, then

$$(\tilde{\chi}_q, \chi_q) \simeq 2\alpha \left(\frac{2}{h^2 - 1} \beta \right)^{1/2} + \frac{4}{3}(1 - \alpha), \quad (\text{A13})$$

which is the final result quoted in (2.16).

- ¹(a) E. Guyon, R. Meyer, and J. Salan, *Mol. Cryst. Liq. Cryst.* **54**, 261 (1979); (b) F. Lonberg, S. Fraden, A. J. Hurd, and R. B. Meyer, *Phys. Rev. Lett.* **52**, 1903 (1984); (c) A. J. Hurd, S. Fraden, F. Lonberg, and R. B. Meyer, *J. Phys. (Paris)* **46**, 905 (1985); (d) Y. W. Hui, M. R. Kuzma, M. San Miguel, and M. M. Labes, *J. Chem. Phys.* **83**, 288 (1985); (e) M. R. Kuzma, *Phys. Rev. Lett.* **57**, 349 (1986); (f) D. V. Rose and M. R. Kuzma, *Mol. Cryst. Liq. Cryst. Lett.* **4**, 39 (1986); (g) J. P. McClymer and M. M. Labes, *Mol. Cryst. Liq. Cryst.* **144**, 275 (1987).
- ²J. P. McClymer, M. M. Labes and M. R. Kuzma, *Phys. Rev. A* **37**, 1388 (1988).
- ³J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1983), Vol. 8.
- ⁴M. San Miguel and F. Sagués, *Phys. Rev. A* **36**, 1883 (1987); F. Sagués, F. Arias, and M. San Miguel, *Phys. Rev. A* **37**, 3601 (1988).
- ⁵The consideration of a periodic magnetic field as a way of slowing the dynamics could overcome experimental limitations. See M. C. Torrent, F. Sagués, F. Arias, and M. San Miguel, *Phys. Rev. A* **38**, 2641 (1988).
- ⁶G. T. Dee and W. van Saarloos, *Phys. Rev. Lett.* **60**, 2641 (1988).
- ⁷Hydrodynamic effects responsible for domain growth mechanisms in phase separating binary liquids can hardly play a role in this context. For example, Siggia's mechanism [*Phys. Rev. A* **20**, 595 (1979)] is known to be ineffective for $d \leq 2$ [M. San Miguel, M. Grant, and J. D. Gunton, *Phys. Rev. A* **31**, 1001 (1985)].
- ⁸J. Langer, *Ann. Phys. (N.Y.)* **65**, 53 (1971).
- ⁹M. San Miguel, J. D. Gunton, G. Dee, and P. S. Sahni, *Phys. Rev. B* **23**, 2334 (1981).
- ¹⁰K. Kawasaki and T. Ohta, *Physica* **116A**, 573 (1982).
- ¹¹K. Kawasaki and T. Nagai, *Physica* **121A**, 175 (1983).
- ¹²F. De Pasquale, P. Tartaglia, and P. Tombesi, *Phys. Rev. A* **31**, 2447 (1985).
- ¹³P. Coulet, C. Elphick, and D. Repaux, *Phys. Rev. Lett.* **58**, 431 (1987).
- ¹⁴T. Kawakatsu and T. Munahata, *Prog. Theor. Phys.* **74**, 11 (1985).
- ¹⁵F. Brochard, *J. Phys. (Paris)* **33**, 607 (1972).
- ¹⁶For the parameter values considered in Ref. 1(b), where $H/H_c = 10$, we estimate $e^{-\rho} \simeq 10^{-7}$.
- ¹⁷The operators F and L are Hermitian as can be formally verified from their respective definitions, (2.10) and (2.4), and invoking periodic boundary conditions for both sets χ_n and $\tilde{\chi}_n$. On the other hand the eigenvalues ϵ_n are shown to be real and the orthogonality conditions, $(\tilde{\chi}_m, \chi_n) = 0$, are readily checked.
- ¹⁸We point out that the presence of the operator L in (2.3) does not modify the values of the eigenvalues when linearizing around homogeneous states so that the force between walls is not modified.
- ¹⁹T. Nagai and K. Kawasaki, *Physica* **120A**, 587 (1983).