

## Interface dynamics for layered structures

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(Received 24 January 1997; revised manuscript received 12 May 1997)

We investigate the dynamics of large-scale and slow deformations of layered structures. Starting from the respective model equations for a nonconserved system, a conserved system and a binary fluid, we derive the interface equations which are a coupled set of equations for deformations of the boundaries of each domain. A further reduction of the degrees of freedom is possible for a nonconserved system such that internal motion of each domain is adiabatically eliminated. The resulting equation of motion contains only the displacement of the center of gravity of domains, which is equivalent to the phase variable of a periodic structure. Thus our formulation automatically includes the phase dynamics of layered structures. In a conserved system and a binary fluid, however, the internal motion of domains turns out to be a slow variable in the long-wavelength limit because of concentration conservation. Therefore a reduced description only involving the phase variable is not generally justified. [S1063-651X(97)14610-4]

PACS number(s): 68.10.-m, 68.60.Dv, 68.15.+e

### I. INTRODUCTION

Layered structures appear in various systems in nature. Smectic liquid crystals [1], mesophases in a microphase separation of block copolymers [2], magnetic bubbles in magnetic thin films [3], and condensed phases of Langmuir monolayers [4,5] are typical examples in thermal equilibrium.

A layered structure can also be observed in pattern formation far from equilibrium [6]. Turing was the first to predict a stable periodic structure in a reaction-diffusion system. Recently, a Turing instability has been observed experimentally in a chemical reaction in a gel [7]. Other well-known examples are roll patterns in Rayleigh-Benard convection and Williams domains in the electrohydrodynamic convection of liquid crystals.

In order to study dynamics of these structures, a coarse-grained description is often employed. Suppose that there is a periodic layer with period  $\ell$  in the  $x$  direction. This is expressed by a periodic function  $X(x + \phi) = X(x + \ell + \phi)$ , where  $X$  is a physical property and  $\phi$  is a phase and is arbitrary in a uniform extended system. When the periodic structure is slightly deformed, its large-scale slow motion can be described by allowing  $\phi$  to depend on  $x$ ,  $y$ , and  $t$ . Since  $\phi$  is a neutral mode, it is one of the slowest variables in the long-wavelength limit. The theory based on this idea, which is called phase dynamics, was developed in pattern dynamics far from equilibrium [8,9,10].

An essentially similar method can be applied to layered structures in equilibrium [11]. One can express the free-energy increase due to deformation in terms of the local displacement field as in continuum elastic theory. The local displacement field corresponds to the phase variable, and the dynamic equation for it can be written down by using the free-energy functional.

Many of the theories assume a sinusoidal function (and only a few higher harmonics) of  $X(x)$  in the derivation of the phase equation. This is valid when the system is in the vicinity of a bifurcation (critical) point where a uniform state

loses stability, and a periodic structure appears supercritically. However, when the function  $X$  changes abruptly at the interface separating two adjacent layers, we need to develop an alternative method for dynamics. Phase dynamics is still meaningful in this case for long-wavelength deformations, but the derivation of the phase equation is not possible using available theories.

In this paper, we address the above problem. Some years ago, Kawasaki and one of the authors [12] derived a free-energy functional of general periodic structures in terms of the displacement fields without assuming the slow variation of  $X$ . Recently a similar analysis has been applied to a model specific to Langmuir monolayers [13]. Here, on the other hand, we are concerned with dynamics. The static aspects can be obtained as a byproduct by taking the equilibrium limit. It is emphasized that our theory does not necessarily rely on the existence of a free-energy functional or a Lyapunov functional. As a consequence, we can deal with the systems both near equilibrium and far from equilibrium in a unified way.

Our strategy is to derive the interface equation of motion for each domain boundary. Suppose that  $X$  varies within one period as  $X=1$  for  $0 < x < 2w$  and  $X=-1$  for  $2w < x < \ell$ , where  $w (< \ell/2)$  is a constant. Thus there are interfaces at  $x=0$  and  $x=2w$  bounding a domain where  $X=1$ . We assume the interface width is infinitesimal compared to  $w$  and  $\ell$ . It is generally expected that these two interfaces are coupled to each other as well as to those in other domains. We can derive the phase equation of motion from the interface equation by retaining only the average displacement of two interfaces at each domain. The other degree of freedom which makes the center of gravity of a domain time independent represents the internal motion of domains. In some situations, internal deformations of each domain are relevant to the stability. The present theory covers such a domain instability.

The phase variable describes the Goldstone mode associated with the translational symmetry breaking due to a periodic structure in an extended uniform system, and hence it is

a slow variable in the long-wavelength limit. In the terminology of lattice vibrations this corresponds to an acoustic mode. On the other hand, the internal motion of domains is similar to an optical mode, and has a finite energy in the limit of long-wavelength deformation. As will be shown below, this gives rise to a finite relaxation rate of the internal mode for small wave numbers in a nonconserved case. Therefore one may eliminate the internal mode adiabatically. However, as we will show, the situation is entirely different in a conserved case and a binary fluid. When the interface width is infinitesimal, and there is no mass flux through the system boundary, concentration conservation is equivalent to a conservation of total area (volume in three dimensions) of domains. Therefore, the internal mode should also be a slow variable in the long-wavelength limit. Our main concern in this paper is to clarify how the phase dynamics is affected by the extra slow mode originating from conservation.

We start with a time-dependent Ginzburg-Landau (TDGL)-type equation describing the system at the coarse-grained level. A general method is available to derive the interface equation of motion [14] from the TDGL equation. In binary alloys or in crystal growth, on the other hand, it is also well known that the same equation can be obtained by starting at the macroscopic level with the simple diffusion equation for the minority atoms and employing the Gibbs-Thomson relation as the boundary condition at an infinitely sharp interface [15]. Thus one may wonder why the TDGL approach is necessary. There are, of course, several reasons. First of all, most simulations of spinodal decomposition and other phase separation phenomena in binary systems use TDGL equations, since for many purposes it is more efficient than the macroscopic approach. Hence the interfacial analysis based on the TDGL equation is necessary for a complete understanding of the simulation results. Second, the interfacial method is valid beyond the simple diffusion equation which has been used only for macrophase separation and for crystal growth. As mentioned above, we will show that our method can be useful for both thermal equilibrium structures and dissipative structures out of equilibrium.

The organization of this paper is as follows. In Sec. II, we make a general argument on layered structures in equilibrium. In Sec. III, we derive the interface equation of motion for a periodic structure in a nonconserved case. In Sec. IV, we verify that the vanishing of one of the diffusion constants in the phase equation is equivalent to free-energy minimization with respect to the spatial period. The method is extended in Sec. V to a conserved system. In Sec. VI we treat a layered structure in binary fluids. The results obtained are discussed and summarized in Sec. VII. Some of the details of the method used in Sec. III are described in the Appendix.

## II. PERIODIC STRUCTURES

A typical example of a layered structure in a system with a nonconserved order parameter is a magnetic thin film [16]. Experiments [17] show stripe and bubble structures of magnetic domains depending on the magnitude of the external magnetic field normal to the film. The free energy of this system can be written in terms of the local order parameter  $u(\vec{r})$  as

$$F = F_{\text{GL}} + F_{\text{LR}}, \quad (1)$$

where

$$F_{\text{GL}} = \int d\vec{r} \left[ \frac{\epsilon^2}{2} (\vec{\nabla}u)^2 + W(u) - hu \right] \quad (2)$$

and

$$F_{\text{LR}} = \frac{\alpha}{2} \int d\vec{r} \int d\vec{r}' G(\vec{r}, \vec{r}') [u(\vec{r}) - c][u(\vec{r}') - c]. \quad (3)$$

Note that the free energy is generally divided into two parts.  $F_{\text{GL}}$  is the short-ranged part, and is the usual Ginzburg-Landau form with  $W(u)$  a double-well potential in the ordered state and  $h$  the external field. The parameter  $\epsilon$ , which is a measure of an interfacial width, is assumed to be sufficiently small. The other part  $F_{\text{LR}}$  contains the long-range interaction, where  $\alpha$  is the interaction strength and  $c$  is a constant. The form of the kernel  $G(\vec{r}, \vec{r}')$  depends on the system considered. For instance, it is given in a thin magnetic film of a strongly uniaxial material by [16]

$$G(\vec{r}, \vec{r}') = \int d\vec{q} \frac{4\pi}{q} [1 - \exp(-qD)] \exp[i\vec{q} \cdot (\vec{r} - \vec{r}')], \quad (4)$$

where the integral over  $\vec{q}$  is taken in a two-dimensional space parallel to the film surface. Note that the order parameter  $u$  in this case is the magnetization normal to the film which has thickness  $D$ . The spatial period of a modulated structure is larger than  $D$ , and the interface width  $\epsilon$  is much smaller than  $D$ , i.e.,  $\epsilon \ll D \ll \ell$ . Probably the simplest choice of  $G(\vec{r}, \vec{r}')$  which provides us with a periodic structure in equilibrium is the Coulomb Green function

$$-\nabla^2 G_c(\vec{r}, \vec{r}') = \delta(\vec{r} - \vec{r}'). \quad (5)$$

This type of long-range interaction appears in the theory of phase separation of block copolymers [18]. The constant  $c$  in Eq. (3) is equated with the spatial average of  $u$  to avoid a divergence in the long-range interaction.

In terms of Fourier components, the sum of the short- and long-range interactions contains the quadratic part

$$\frac{1}{2} \Gamma(q) u_{\vec{q}} u_{-\vec{q}}, \quad (6)$$

where

$$\Gamma(q) = \epsilon^2 q^2 + \alpha G_q - \tau, \quad (7)$$

with  $G_q$  the Fourier transform of  $G(\vec{r} - \vec{r}')$ . The constant  $\tau$  comes from the bilinear term in the local part  $W(u)$ .

In order for a stable periodic structure to exist, we assume that  $\Gamma$  has a minimum for a finite value of  $q$ , and that the free energy (1) takes the lowest value for such a modulation. Hereafter, to be specific, we consider the case where  $G(\vec{r}, \vec{r}')$  is given by the screened Coulomb interaction

$$-(\nabla^2 - \kappa^2) G(\vec{r}, \vec{r}') = \delta(\vec{r} - \vec{r}'), \quad (8)$$

where  $1/\kappa$  is the screening length which is assumed to be much larger than  $\epsilon$ . In this sense, the interaction  $G(\vec{r}, \vec{r}')$  is still long ranged. The interfacial theory for other forms of

$G(\vec{r}, \vec{r}')$  can be constructed similarly; in particular, the pure Coulomb case follows by taking the  $\kappa \rightarrow 0$  limit of the interface equations.

### III. NONCONSERVED SYSTEM

The time-evolution equation of a nonconserved order parameter  $u$  is determined by the free energy functional (1) according to

$$\frac{\partial u}{\partial t} = - \frac{\delta F}{\delta u}. \tag{9}$$

It is noted that the right-hand side contains the nonlocal long-range interaction. When we derive the interface equation of motion for layers, it is more convenient to express it in a local form. To this end, we write Eq. (9) in the form of a coupled set of equations as

$$\tau_1 \frac{\partial u(\vec{r})}{\partial t} = \epsilon^2 \nabla^2 u + f(u) - v + h, \tag{10}$$

$$\tau_2 \frac{\partial v(\vec{r})}{\partial t} = \nabla^2 v - \kappa^2 v + \alpha u, \tag{11}$$

where we have introduced two parameters  $\tau_1$  and  $\tau_2$  and  $f(u)$ , defined by

$$f(u) = - \frac{dW}{du}. \tag{12}$$

The variable  $v$  in Eq. (11) is an auxiliary field and has no physical meaning for model (9). However the set of equations (10) and (11) appears in pattern formation far from equilibrium as will be mentioned below. Without loss of generality, we may put the diffusion constant for  $v$  equal to unity. In the limit  $\tau_2 \rightarrow 0$ , Eq. (11) can be solved for  $v$  by using  $G(\vec{r}, \vec{r}')$  defined by Eq. (8). Substituting it into Eq. (10) we recover Eq. (9).

For a general form of the kernel  $G(\vec{r}, \vec{r}')$  in the long-range interaction, we may replace  $\nabla^2 v - \kappa^2 v$  by  $\int d\vec{r}' \Gamma(\vec{r}, \vec{r}') v(\vec{r}')$  where  $\Gamma(\vec{r}, \vec{r}')$  is defined through the relation

$$\int d\vec{r}'' G(\vec{r}, \vec{r}'') \Gamma(\vec{r}'', \vec{r}') = \delta(\vec{r} - \vec{r}'). \tag{13}$$

It is remarked that Eqs. (10) and (11) are model equations for pattern dynamics far from equilibrium. For instance, the Belousov-Zhabotinski reaction in the excitable regime can be described, in its simplest approximation, by Eqs. (10) and (11). In fact this set of equations has been extensively studied during the past two decades by applied mathematicians [19,20]. It is well known that Eqs. (10) and (11) undergo a Turing instability when  $\epsilon$  is small so that a periodic structure can be formed. To our knowledge, however, an interfacial approach has not been applied to a periodic solution except for Ref. [21].

We solve Eqs. (10) and (11) in the limit  $\epsilon \rightarrow 0$  to derive the interface equation of motion. We assume that when the diffusion terms are absent, these equations have two stable

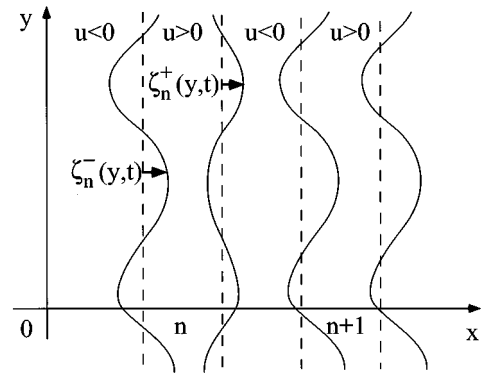


FIG. 1. Gently deformed layered structure. The dotted lines indicate an equilibrium configuration with  $w = \ell/2$ .

uniform solutions  $(u_1, v_1)$  and  $(u_2, v_2)$ . As shown in Fig. 1, we consider a layer of domains in two dimensions where the regions  $u = u_1$  and  $u = u_2$  are arrayed alternatively. We assume the width of a domain in which  $u = u_1$  is equal to  $2w$  and the period of the layer is  $\ell$ .

Now suppose these striped domains are gently curved as in Fig. 1. We introduce the deformations  $\zeta_n^+(y, t)$  [ $\zeta_n^-(y, t)$ ] around the equilibrium position of the right (left) interface in the  $n$ th domain in which  $u = u_1$ . Our aim is to derive the linearized equation of motion for  $\zeta_n^\pm$ . For concreteness, we hereafter assume the following form of  $f(u)$ :

$$f(u) = \frac{1}{2} u(1 - u^2). \tag{14}$$

As was mentioned above, the parameter  $\epsilon$  is a measure of an interface width. Suppose that we are concerned with the motion of a specific interface. In the limit  $\epsilon \rightarrow 0$ , the density  $u$  varies rapidly through the interface, but the spatial variation of  $v$  is much slower. Therefore we may replace  $v$  in Eq. (10) by its value at the interface, which is denoted by  $v_I$  and which may depend on time and position on the interface. Thus Eq. (10) can be written as

$$\tau_1 \frac{\partial u(\vec{r})}{\partial t} = \epsilon^2 \nabla^2 u + f(u) - v_I + h. \tag{15}$$

This is just the TDGL equation with the ‘‘external field’’  $v_I - h$ , and hence the interface equation of motion for this system is readily obtained:

$$\tau_1 V = \epsilon^2 H - 3\epsilon(v_I - h), \tag{16}$$

where, for simplicity, we assumed that the absolute value of  $v_I - h$  is sufficiently small so that the gap of the density  $u$  at the interface is approximately equal to 2.  $V$  is the normal component of the interface velocity, and  $H$  the mean curvature. These are defined as follows. First let us introduce a field variable  $\psi(\vec{r}, t)$  which is positive (negative) in the region  $u > 0$  ( $u < 0$ ) and  $\psi(\vec{r}, t) = 0$  specifies the interface configuration. The velocity  $V$  and the mean curvature  $H$  are expressed, respectively, as

$$V = \frac{1}{|\vec{\nabla} \psi|} \frac{\partial \psi}{\partial t}, \tag{17}$$

$$H = \vec{\nabla} \cdot \vec{n}, \quad (18)$$

with

$$\vec{n} = \frac{\vec{\nabla} \psi}{|\vec{\nabla} \psi|}. \quad (19)$$

Note that  $\vec{n}$  is the *inward* normal of a  $u > 0$  domain.

From the above definition of  $\psi$ , deformations  $\zeta^\pm$  may be written as  $\psi = -x + \zeta^+(y, t)$  and  $\psi = x - \zeta^-(y, t)$ . Substituting these into Eqs. (17), (18), and (16), we obtain, up to linear order in  $\zeta^\pm$ ,

$$\tau_1 \partial \zeta_n^\pm / \partial t = \epsilon^2 \partial^2 \zeta_n^\pm / \partial y^2 \mp 3 \epsilon (v_I - h). \quad (20)$$

The last term indicates that a domain where  $u > 0$  shrinks as  $v_I - h$  is increased.

The remaining problem is to determine  $v_I$  as a functional of  $\zeta_n^\pm$ , so that Eq. (20) becomes a closed set of equations. In what follows, we need to solve Eq. (11) for a given set of  $\zeta_n^\pm$ . This was carried out in a previous paper [21] for general values of  $\tau_2$  to study a dynamical instability of layered domains. Here we consider only the limit  $\tau_2 \rightarrow 0$ . In this limit, the solution is given by

$$v(\vec{r}, t) = \alpha \int d\vec{r}' G(\vec{r}, \vec{r}') u(\vec{r}', t). \quad (21)$$

When the absolute value of  $v_I - h$  is sufficiently small as we have assumed, the uniform solutions  $u_1$  and  $u_2$  may be approximated as  $u_1 = 1$  and  $u_2 = -1$ . Actually only the difference  $u_1 - u_2$  enters in the theory given below. The correction is however, of order  $(v_I - h)^2$ , i.e.,  $u_1 - u_2 = 2 + O[(v_I - h)^2]$ , and hence does not affect the first order correction in Eq. (16). Therefore Eq. (21) can be written as

$$v(\vec{r}, t) = \alpha \int \frac{d\vec{q}}{(2\pi)^2} \int dy' \times \sum_m \left[ \int_{b_m^-}^{b_m^+} dx' \frac{1}{q^2 + \kappa^2} \exp[i\vec{q} \cdot (\vec{r} - \vec{r}')] \right. \\ \left. - \int_{b_m^+}^{b_m^{+1}} dx' \frac{1}{q^2 + \kappa^2} \exp[i\vec{q} \cdot (\vec{r} - \vec{r}')] \right], \quad (22)$$

where we have defined

$$b_m^\pm = m\ell \pm w + \zeta_m^\pm. \quad (23)$$

The value  $v_I$  in the  $n$ th domain, which generally takes a different value at the right and the left interfaces, is given by

$$v_I^\pm = v(b_n^\pm, y, t). \quad (24)$$

Now we calculate  $v_I$  to first order in  $\zeta^\pm$ . Some further details are provided in the Appendix. The zeroth-order solution is given from Eq. (A2) in the Appendix by

$$v_I^{(0)\pm} = \frac{\alpha}{\ell} \sum_Q \frac{2}{Q(Q^2 + \kappa^2)} \sin 2Qw, \quad (25)$$

where reciprocal-lattice vectors are given by  $Q = 2\pi n/\ell$  with  $n$  integers ( $n = 0, \pm 1, \pm 2, \dots$ ). The first-order solution is given from Eq. (A3) in the Appendix by

$$v_I^{(1)+} = -2\alpha \zeta_n^+(y) \frac{1}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} (1 - \cos 2Qw) \\ + 2\alpha \int \frac{dk}{2\pi} e^{in\ell k} \int \frac{dp}{2\pi} \frac{1}{\ell} \\ \times \sum_Q \frac{e^{ipy}}{(k-Q)^2 + p^2 + \kappa^2} [\zeta_{k,p}^+ - e^{-2iQw} \zeta_{k,p}^-], \quad (26)$$

$$v_I^{(1)-} = 2\alpha \zeta_n^-(y) \frac{1}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} (1 - \cos 2Qw) \\ + 2\alpha \int \frac{dk}{2\pi} e^{in\ell k} \int \frac{dp}{2\pi} \frac{1}{\ell} \\ \times \sum_Q \frac{e^{ipy}}{(k-Q)^2 + p^2 + \kappa^2} [e^{2iQw} \zeta_{k,p}^+ - \zeta_{k,p}^-]. \quad (27)$$

In the derivation of Eqs. (26) and (27), we introduced the Fourier transform

$$\zeta_n^\pm(y) = \int \frac{dk}{2\pi} \int \frac{dp}{2\pi} \zeta_{k,p}^\pm \exp[ik(n\ell \pm w) + iyp], \quad (28)$$

and used the Poisson formula

$$\sum_{n=-\infty}^{\infty} \exp(ikn\ell) = \frac{1}{\ell} \sum_Q \delta(k - Q). \quad (29)$$

Substituting these into Eq. (20), we obtain the interface equation of motion. The zeroth-order solution is particularly important, which reads

$$h = \frac{2\alpha}{\ell} \sum_Q \frac{1}{Q(Q^2 + \kappa^2)} \sin 2Qw. \quad (30)$$

This specifies the ratio  $w/\ell$  of these unknown parameters. It is readily verified that when  $h = 0$ ,  $w/\ell = \frac{1}{4}$ , as it should for the symmetric case. Equation (30) is equivalent to  $\partial \zeta_n^\pm / \partial t = 0$  without deformations. Hence this is an equilibrium condition for a nonconserved system.

From the first-order equation, we obtain the coupled set of linear equations for  $\zeta^\pm$ ,

$$\tau_1 \frac{d}{dt} \zeta_{k,p}^+ = - \left[ \epsilon^2 p^2 - \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} (1 - \cos 2Qw) \right] \zeta_{k,p}^+ \\ + \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{(k-Q)^2 + p^2 + \kappa^2} \\ \times [-\zeta_{k,p}^+ + e^{-2iQw} \zeta_{k,p}^-], \quad (31)$$

$$\begin{aligned} \tau_1 \frac{d}{dt} \zeta_{k,p}^- = & - \left[ \epsilon^2 p^2 - \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} (1 - \cos 2Qw) \right] \zeta_{k,p}^- \\ & + \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{(k-Q)^2 + p^2 + \kappa^2} \\ & \times [e^{2iQw} \zeta_{k,p}^+ - \zeta_{k,p}^-]. \end{aligned} \quad (32)$$

In order to analyze the motion of long-wavelength deformations, we make the following transformation:

$$\zeta_n^{(1)}(y) = \frac{1}{2} [\zeta_n^+(y) + \zeta_n^-(y)], \quad (33)$$

$$\zeta_n^{(2)}(y) = \frac{1}{2} [\zeta_n^+(y) - \zeta_n^-(y)]. \quad (34)$$

In real space,  $\zeta_n^{(1)}$  describes the deviation of the center of gravity of each domain, while  $\zeta_n^{(2)}$  is the internal deformation of a domain. Note that  $\zeta_n^{(1)} \neq 0$ , but  $\zeta_n^{(2)} = 0$  for a uniform translation of a domain. Thus  $\zeta_n^{(1)}$  is a phase variable.

Since we are concerned with large-scale deformations of a modulated structure, we may apply a gradient expansion to Eqs. (31) and (32). That is, we expand the coefficients of these equations in terms of  $k$  and  $p$ . Retaining the lowest nontrivial order, we obtain, from Eqs. (31) and (32),

$$\begin{aligned} \tau_1 \frac{d}{dt} \zeta_{k,p}^{(1)} = & - \left[ \epsilon^2 p^2 + \frac{6\epsilon\alpha}{\ell} (3k^2 - p^2) \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} \right. \\ & \left. \times (1 - \cos 2Qw) \right] \zeta_{k,p}^{(1)} \\ & + \frac{12i\epsilon\alpha}{\ell} k \zeta_{k,p}^{(2)} \sum_Q \frac{Q}{(Q^2 + \kappa^2)^2} \sin 2Qw \\ & - \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{(Q^2 + \kappa^2)^3} (1 - \cos 2Qw) p^4 \zeta_{k,p}^{(1)}, \end{aligned} \quad (35)$$

$$\begin{aligned} \tau_1 \frac{d}{dt} \zeta_{k,p}^{(2)} = & - \left[ \epsilon^2 p^2 + \frac{12\epsilon\alpha}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} \cos 2Qw \right] \zeta_{k,p}^{(2)} \\ & - \frac{6\epsilon\alpha}{\ell} (3k^2 - p^2) \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} \\ & \times (1 + \cos 2Qw) \zeta_{k,p}^{(2)} \\ & - \frac{12i\epsilon\alpha}{\ell} k \zeta_{k,p}^{(1)} \sum_Q \frac{Q}{(Q^2 + \kappa^2)^2} \sin 2Qw. \end{aligned} \quad (36)$$

Note that the right-hand side of Eq. (35) vanishes in the limit  $k, p \rightarrow 0$ , whereas that of Eq. (36) remains finite in this limit. Hence we may assume  $\zeta^{(2)}$  relaxes more rapidly, and we may put  $d\zeta_{k,p}^{(2)}/dt = 0$  in Eq. (36). We may then substitute the solution into Eq. (35) to obtain

$$\tau_1 \frac{d}{dt} \zeta_{k,p}^{(1)} = - \frac{3\epsilon\ell}{4} [D_\perp p^2 + D_\parallel k^2 + K p^4] \zeta_{k,p}^{(1)}, \quad (37)$$

where

$$D_\perp = \frac{4\epsilon}{3\ell} - \frac{8\alpha}{\ell^2} \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} (1 - \cos 2Qw), \quad (38)$$

$$\begin{aligned} D_\parallel = & \frac{24\alpha}{\ell^2} \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} (1 - \cos 2Qw) \\ & - \frac{16\alpha}{\sum_Q \frac{\cos 2Qw}{Q^2 + \kappa^2}} \left( \frac{1}{\ell} \sum_Q \frac{Q \sin 2Qw}{(Q^2 + \kappa^2)^2} \right)^2, \end{aligned} \quad (39)$$

and

$$K = \frac{8\alpha}{\ell^2} \sum_Q \frac{1}{(Q^2 + \kappa^2)^3} (1 - \cos 2Qw). \quad (40)$$

In Eq. (37), we have factored out  $3\epsilon\ell/4$ . The reason will become clear in Eq. (50) below. Equation (37) with the coefficients given by Eqs. (38), (39), and (40) is the general form for layered structures. The coefficient  $K$  is always positive and  $D_\parallel$  is positive at least when  $w \approx \ell/4$ . Thus the layered structure in the present model system does not cause any linear instabilities in this regime. The coefficient  $D_\perp$  must be identically zero when the spatial period  $\ell$  is the equilibrium period. This will be verified separately in Sec. IV.

#### IV. DETERMINATION OF THE PERIOD

When the system has a free-energy functional, the equilibrium period should be determined by its minimization. In this section we show that this is identical to the condition  $D_\perp = 0$ . Although this is a well-known fact (see, e.g., Ref. [12]), a verification is necessary to confirm the validity of the present approach.

We consider a one-dimensional system with the system size  $L (\gg \ell)$ , and write the free energy per unit length as

$$\hat{F} = \frac{1}{2L} \int dx \int dx' s(x-x') u(x) u(x') + \frac{1}{L} \int dx W(u), \quad (41)$$

where  $s(x-x')$  is the nonlocal interaction. In the case of Eq. (1) it is given by

$$s(x-x') = \epsilon^2 \frac{\partial^2}{\partial x \partial x'} \delta(x-x') + \alpha G(x-x'). \quad (42)$$

The second term of Eq. (41) is the local part. We assume that the solution of  $\delta F/\delta u = 0$  is spatially periodic.

Now suppose that the system is expanded uniformly so that the period becomes  $\ell(1+\delta)$  ( $\delta \ll 1$ ). Thus  $u$  and  $L$  are transformed as  $u(x) \rightarrow u[x/\ell(1+\delta)]$ ,  $L \rightarrow L(1+\delta)$ . The free energy changes as

$$\begin{aligned} \hat{F} = & \frac{1+\delta}{2L} \int dy \int dy' s[(y-y')(1+\delta)] u(y) u(y') \\ & + \frac{1}{L} \int dy W(u), \end{aligned} \quad (43)$$

where  $x/(1+\delta)=y$ . The free energy increase  $\Delta\hat{F}$  is, therefore, given up to  $O(\delta)$  by

$$\begin{aligned}\Delta\hat{F} &= \frac{\delta}{2L} \int dy \int dy' s(y-y') u(y) u(y') \\ &+ \frac{\delta}{2L} \int dy \int dy' (y-y') \left[ \frac{ds(y-y')}{d(y-y')} \right] u(y) u(y') \\ &= -\frac{\delta}{2L} \int dy \int dy' (y-y') s(y-y') \left[ \frac{du(y)}{dy} \right] u(y').\end{aligned}\quad (44)$$

This may be rewritten in terms of the Fourier components as

$$\Delta\hat{F} = -\frac{\delta}{2L} \int \frac{dq}{2\pi} q \left[ \frac{ds_q}{dq} \right] u_q u_{-q}. \quad (45)$$

If  $u$  is the equilibrium solution,  $\Delta\hat{F}$  must be identically zero.

When  $s(x-x')$  is given by Eq. (42),  $s_q = \epsilon^2 q^2 + \alpha/(q^2 + \kappa^2)$ , so that we have

$$\Delta\hat{F} = -\frac{\delta}{L} \int \frac{dq}{2\pi} \left( \epsilon^2 q^2 - \frac{\alpha q^2}{(q^2 + \kappa^2)^2} \right) u_q u_{-q}. \quad (46)$$

Let us calculate Eq. (46) explicitly. First of all, we define the equilibrium interfacial energy  $\sigma$ . When  $f(u)$  is given by Eq. (12), it is calculated as

$$\sigma = \epsilon^2 \int dx \left( \frac{du}{dx} \right)^2 = \frac{2\epsilon}{3}, \quad (47)$$

where the integral domain over  $x$  is restricted to the vicinity of an interface. Note that the interface width dependence of  $\sigma$  is different from that in ordinary critical phenomena. However, this is only apparent, and is due to the definition of the free energy (2), where  $\epsilon$  appears as a coefficient of the gradient term.

By using Eq. (47), we note that

$$\frac{\epsilon^2}{L} \int \frac{dq}{2\pi} q^2 u_q u_{-q} = \frac{\epsilon^2}{L} \int dx \left( \frac{du}{dx} \right)^2 = 2 \frac{\sigma}{\ell}. \quad (48)$$

The factor of 2 comes from the fact that there are two interfaces in each domain.

Since we have assumed that  $u=1$  for  $0 < x < 2w$  and  $u=-1$  for  $2w < x < \ell$  away from the interfaces, we obtain

$$\begin{aligned}\frac{\alpha}{L} \int \frac{dq}{2\pi} \left( \frac{q^2}{(q^2 + \kappa^2)^2} \right) u_q u_{-q} &= \frac{8\alpha}{\ell^2} \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} \\ &\times (1 - \cos 2Qw).\end{aligned}\quad (49)$$

Putting this together with Eq. (48), we finally obtain

$$\frac{4\epsilon}{3\ell} - \frac{8\alpha}{\ell^2} \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} (1 - \cos 2Qw) = 0. \quad (50)$$

This precisely agrees with  $D_{\perp}=0$  given by Eq. (38).

## V. CONSERVED SYSTEM

In a phase separation of binary mixtures, the order parameter is a local concentration and is a conserved quantity. The time evolution of a conserved system is modeled by

$$\frac{\partial u}{\partial t} = \nabla^2 \frac{\delta F}{\delta u}. \quad (51)$$

This equation with the free energy given by Eq. (1) was introduced to study a microphase separation of block copolymers [22].

When the long-range interaction is absent in Eq. (1), Eq. (51) is called the Cahn-Hilliard equation for spinodal decomposition, and the interface equation of motion has been derived. However, an interfacial analysis for multilayered systems given by Eq. (51) has not previously been obtained, to our knowledge.

First we reformulate the diffusive model (51) by introducing an auxiliary field  $v$ ,

$$\tau_1 \frac{\partial u(\vec{r})}{\partial t} = \epsilon^2 \nabla^2 u + f(u) - \alpha \int d\vec{r}' G(\vec{r}, \vec{r}') u(\vec{r}', t) - v + h, \quad (52)$$

$$\tau_2 \frac{\partial v(\vec{r})}{\partial t} = \nabla^2 v + \frac{\partial u(\vec{r})}{\partial t}, \quad (53)$$

where the long-range interaction has been put in Eq. (52). If the constant  $\alpha$  were zero in Eq. (52), the above set of equations would be essentially the same as the so-called phase-field model for melt growth [23,24]. In this case, the variable  $v$  is the local temperature, and the last term in Eq. (53) stands for the latent heat production upon crystallization.

Equation (51) can be recovered as follows. We operate  $\nabla^2$  on both side of Eq. (52). Putting  $\tau_2=0$  in Eq. (53), we may write  $\nabla^2 v$  in terms of  $u$ , and hence Eq. (52) becomes a closed equation for  $u$ . Taking the limit  $\tau_1 \rightarrow 0$ , we obtain Eq. (51). Thus the expression of the diffusive model as Eqs. (52) and (53) enables us to analyze the interface dynamics almost parallel to the nonconserved case.

However, there are two important differences. One is the fact that  $v_I$  now contains the time derivative of  $\zeta^{\pm}$  because of  $\partial u/\partial t$  in (53). Putting  $\tau_1=0$ , Eq. (15) becomes

$$0 = \epsilon^2 H - 3\epsilon(v_I - h). \quad (54)$$

It should be noted, however, that the contribution from the third term in Eq. (52) is also included in  $v_I$ . That is,  $v_I$  consists of two parts  $v_I = v_I^{(1)} + v_I^{(2)}$ , where  $v_I^{(1)}$  comes from the third term in Eq. (52) and is given by Eqs. (26) and (27), whereas  $v_I^{(2)}$  comes from the fourth term in Eq. (52), which will be evaluated below. Equation (53) can be solved as

$$v(\vec{r}, t) = \int d\vec{r}' G_c(\vec{r}, \vec{r}') \frac{\partial u(\vec{r}', t)}{\partial t}. \quad (55)$$

It should be noted that the Green function in Eq. (55) is not the screened one, Eq. (8), but the Coulomb Green function (5). Using the fact that

$$\frac{\partial u}{\partial t} = -2 \sum_m \{ \dot{\zeta}_m^- \delta[x - m\ell + w - \zeta_m^-(y)] - \dot{\zeta}_m^+ \delta[x - m\ell - w - \zeta_m^+(y)] \}, \quad (56)$$

where the factor 2 comes from the gap of  $u$  at the interfaces, we readily obtain, up to linear order in  $\zeta^\pm$ ,

$$\hat{v}_I^{(2)+} = \frac{2}{\ell} \sum_{Q(=0)} \frac{1}{(k-Q)^2 + p^2} [\dot{\zeta}_{k,p}^+ - e^{-2iQw} \dot{\zeta}_{k,p}^-], \quad (57)$$

$$\hat{v}_I^{(2)-} = \frac{2}{\ell} \sum_{Q(=0)} \frac{1}{(k-Q)^2 + p^2} [e^{2iQw} \dot{\zeta}_{k,p}^+ - \dot{\zeta}_{k,p}^-], \quad (58)$$

where  $\hat{v}_I^{(2)\pm}$  is the Fourier component of  $v_I^{(2)\pm}$ . In these expressions we have emphasized that the  $n=0$  component is included in the summations.

The other difference from the nonconserved case is that we have to take account of the conservation law in the interface equation of motion. In the nonconserved case, we consider the zeroth solution separately as Eq. (30), which determines the ratio  $w/\ell$ . However, in a conserved system this ratio is fixed uniquely in the strong segregation limit by the average volume fraction. In fact, when there is no current across the system boundary, the conservation law requires

$$\frac{d}{dt} \sum_n \int dy \zeta_n^{(2)} = 0. \quad (59)$$

The constant  $h$  in Eq. (52) plays the role of a Lagrange multiplier associated with condition (59).

Substituting Eqs. (57) and (58) into Eq. (54), and retaining the zeroth-order term, we obtain

$$\begin{aligned} & \frac{6\epsilon}{\ell} \sum_{Q(=0)} \frac{1}{(k-Q)^2 + p^2} [(1 - \cos 2Qw) \dot{\zeta}_{k,p}^{(1)} - i \sin 2Qw \dot{\zeta}_{k,p}^{(2)}] \\ &= - \left[ \epsilon^2 p^2 + \frac{6\epsilon\alpha}{\ell} (3k^2 - p^2) \right. \\ & \quad \times \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} (1 - \cos 2Qw) \left. \right] \zeta_{k,p}^{(1)} \\ & \quad + \frac{12i\epsilon\alpha}{\ell} k \zeta_{k,p}^{(2)} \sum_Q \frac{Q}{(Q^2 + \kappa^2)^2} \sin 2Qw \\ & \quad - \frac{6\epsilon\alpha}{\ell} \sum_Q \frac{1}{(Q^2 + \kappa^2)^3} (1 - \cos 2Qw) p^4 \zeta_{k,p}^{(1)}, \end{aligned} \quad (60)$$

$$\begin{aligned} & \frac{6\epsilon}{\ell} \sum_{Q(=0)} \frac{1}{(k-Q)^2 + p^2} [(1 + \cos 2Qw) \dot{\zeta}_{k,p}^{(2)} + i \sin 2Qw \dot{\zeta}_{k,p}^{(1)}] \\ &= - \left[ \epsilon^2 p^2 + \frac{12\epsilon\alpha}{\ell} \sum_Q \frac{1}{Q^2 + \kappa^2} \cos 2Qw \right] \zeta_{k,p}^{(2)} - \frac{6\epsilon\alpha}{\ell} \\ & \quad \times (3k^2 - p^2) \sum_Q \frac{1}{(Q^2 + \kappa^2)^2} (1 + \cos 2Qw) \zeta_{k,p}^{(2)} \\ & \quad - \frac{12i\epsilon\alpha}{\ell} k \zeta_{k,p}^{(1)} \sum_Q \frac{Q}{(Q^2 + \kappa^2)^2} \sin 2Qw - C \delta(k) \delta(p), \end{aligned} \quad (61)$$

where

$$C = \frac{2\alpha}{\ell} \sum_Q \frac{\sin 2Qw}{Q(Q^2 + \kappa^2)} - h. \quad (62)$$

The right-hand sides of Eqs. (60) and (61) have been expanded in powers of  $k$  and  $p$ , and are the same as those in Eqs. (35) and (36), respectively. The unknown constant  $h$  or, equivalently,  $C$ , is determined by Eq. (59). That is, in the limit  $k, p \rightarrow 0$  in Eq. (61) we obtain

$$C = - \frac{12\epsilon\alpha}{\ell} \sum_Q \frac{\cos 2Qw}{Q^2 + \kappa^2} \zeta_{0,0}^{(2)}. \quad (63)$$

Note from Eq. (63) that the ‘‘zeroth’’-order term is actually not strictly zeroth-order, but turns out to be linearly dependent on  $\zeta^{(2)}$ .

It should be noted that the summations on the left-hand side of Eqs. (60) and (61) contain the  $Q=0$  component. In particular, Eq. (61) produces the factor  $1/(k^2 + p^2)$ , which originates from the conservation law and is dominant in the long-wavelength limit. Thus Eqs. (60) and (61) are approximated as

$$\dot{\zeta}_{k,p}^{(1)} = -\Gamma^{(1)} [(D_\perp p^2 + Dk^2 + Kp^4) \zeta_{k,p}^{(1)} - ikB_2 \zeta_{k,p}^{(2)}], \quad (64)$$

$$\dot{\zeta}_{k,p}^{(2)} = -\Gamma^{(2)} (k^2 + p^2) [B_1 \zeta_{k,p}^{(2)} + ikB_2 \zeta_{k,p}^{(1)}], \quad (65)$$

where

$$D = \frac{24\alpha}{\ell^2} \sum_Q \frac{1 - \cos 2Qw}{(Q^2 + \kappa^2)^2}, \quad (66)$$

$$\Gamma^{(1)} = \frac{\ell^2}{8 \sum_{Q \neq 0} \frac{1 - \cos 2Qw}{Q^2}}, \quad (67)$$

$$\Gamma^{(2)} = \frac{\ell^2}{16}, \quad (68)$$

$$B_1 = \frac{16\alpha}{\ell^2} \sum_Q \frac{\cos 2Qw}{Q^2 + \kappa^2}, \quad (69)$$

$$B_2 = \frac{16\alpha}{\ell^2} \sum_Q \frac{Q \sin 2Qw}{(Q^2 + \kappa^2)^2}. \quad (70)$$

We have omitted the  $C$  term, and  $D_\perp$  and  $K$  are given by Eqs. (38) and (40), respectively. The factor  $\Gamma^{(2)}$  has been factored out such that the  $B_2$  term in Eqs. (64) and (65) have the same coefficient. In this way we have a coupled set of equations for  $\zeta^{(1)}$  and  $\zeta^{(2)}$ , whose relaxation rates vanish in the limit  $k, p \rightarrow 0$ .

There are two cases where the relaxation of  $\zeta^{(2)}$  is much faster than that of  $\zeta^{(1)}$ . One is the limit  $\kappa \rightarrow 0$ , where  $B_1$  becomes very large because of the  $Q=0$  component. Note that  $D$  remains finite. The other is the case that  $D_\perp = 0$  and  $k^2 \ll p^2$ . In these situations we can eliminate  $\zeta^{(2)}$  by putting  $\dot{\zeta}^{(2)} = 0$ :

$$\frac{d}{dt} \zeta_{k,p}^{(1)} = -\Gamma^{(1)} [D_\parallel k^2 + K p^4] \zeta_{k,p}^{(1)}, \quad (71)$$

where  $D_\parallel$  is the same as in Eq. (39).

## VI. BINARY FLUIDS

Equation (51) has been applied to a microphase separation of diblock copolymers. When the sizes of the two blocks are nearly equal, the system exhibits a lamellar structure in the microphase-separated state. The kinetics of this phase separation have been studied mainly by omitting any hydrodynamic effects. However, in some situations, such as when the system is subjected to shear flow, hydrodynamic effects are known to cause an instability of a lamellar structure [25].

Another example in fluids is the formation of a periodic structure within a Langmuir monolayer [26,27]. However, since this is an adsorbed system between an air-water interface, the hydrodynamics are different from that in bulk [28]. One needs a separate treatment of its interface dynamics which we do not enter here.

In this section, we consider the conserved system coupled with the local velocity field  $\vec{v}$ ,

$$\frac{\partial u(\vec{r})}{\partial t} + \vec{v} \cdot \vec{\nabla} u = \nabla^2 [-\epsilon^2 \nabla^2 u - f(u) + \phi], \quad (72)$$

$$\frac{\partial \vec{v}}{\partial t} = \eta \nabla^2 \vec{v} + (\vec{\nabla} u) [-\epsilon^2 \nabla^2 u - f(u) + \phi], \quad (73)$$

$$\nabla^2 \phi - \kappa^2 \phi + \alpha u = 0, \quad (74)$$

where  $\eta$  is the shear viscosity. In order to avoid confusion of the velocity field  $\vec{v}$  with the auxiliary field  $v$ , we have introduced the notation  $\phi$  instead of  $v$  for the auxiliary density. We have imposed the incompressibility condition  $\vec{\nabla} \cdot \vec{v} = 0$  and eliminated the pressure term. Thus  $\vec{v}$  has only transverse components. The nonlinear convective term  $\vec{v} \cdot \vec{\nabla} \vec{v}$  has been ignored in Eq. (73).

A general form of the interface equation can be derived as follows. We temporarily ignore the diffusive coupling of the order parameter given by the right-hand side of Eq. (72), and consider only the hydrodynamic coupling  $\vec{v} \cdot \vec{\nabla} u$ . At the final

stage we will take account of the diffusive effect given by the right-hand side of Eq. (72). Thus the interface velocity  $V$  is given from Eq. (72) by

$$V = -\vec{v}_I \cdot \vec{n}, \quad (75)$$

where the unit normal vector  $\vec{n}$  is directed inward to a  $u=1$  domain, and  $\vec{v}_I$  is the fluid velocity at the interface. Putting  $\partial \vec{v} / \partial t = 0$ , as usual assuming velocity fields relax rapidly compared to interface motion, we can solve Eq. (73). Substituting this into Eq. (75), we obtain

$$V(a) = \int da' \sum_{\alpha, \beta} n^\alpha(a) T^{\alpha\beta}(a, a') n^\beta(a') \times [\sigma H(a') - 2\alpha \phi_I(a')], \quad (76)$$

where  $a$  and  $a'$  stand for the position on the interface. The tensor  $T^{\alpha\beta}$  is the Oseen tensor, whose Fourier transform is given by

$$\hat{T}_q^{\alpha\beta} = \frac{1}{\eta q^2} \left[ \delta_{\alpha, \beta} - \frac{q^\alpha q^\beta}{q^2} \right]. \quad (77)$$

The constant  $\sigma$  is the interfacial energy defined by Eq. (47). The value  $\phi_I$  is the same as  $v_I$  given by Eqs. (26) and (27). In this case the zeroth-order constant part of  $\phi_I$  does not contribute to Eq. (76) because of the relation

$$\int da' \sum_{\alpha, \beta} n^\alpha(a) T^{\alpha\beta}(a, a') n^\beta(a') = 0. \quad (78)$$

Hence we may consider only the first-order corrections given by Eqs. (26) and (27) for  $\phi_I$  in Eq. (76).

The interface equation of motion can be derived from Eq. (76). Note that  $\vec{n}$  has only the  $x$  component to lowest order, and  $n^x = -1$  for a right interface and  $n^x = 1$  for a left interface. Using the facts that  $V = \pm \partial \zeta^\pm / \partial t$  and  $H = \pm \partial^2 \zeta^\pm / \partial y^2$ , we obtain

$$\begin{aligned} \frac{d}{dt} \vec{\zeta} = & \frac{2}{3\epsilon\ell} \sum_{Q=(0)} T_{k-Q, p}^{xx} N(Q) \left[ -\epsilon^2 p^2 \right. \\ & + \frac{6\epsilon\alpha}{\ell} \sum_{Q'} \frac{1}{Q'^2 + \kappa^2} (1 - \cos 2Q'w) \\ & \left. - \frac{6\epsilon\alpha}{\ell} \sum_{Q'} \frac{1}{(k-Q')^2 + p^2 + \kappa^2} M(Q') \right] \vec{\zeta}, \quad (79) \end{aligned}$$

where  $\vec{\zeta}$  is a column vector with  $\zeta_1 = \zeta_{k,p}^{(1)}$  and  $\zeta_2 = \zeta_{k,p}^{(2)}$ , and  $M(Q)$  is a  $2 \times 2$  matrix with the components



$M_{11}=1-\cos 2Qw$ ,  $M_{12}=-i \sin 2Qw$ ,  $M_{21}=i \sin 2Qw$  and  $M_{22}=1+\cos 2Qw$ . The  $2 \times 2$  matrix  $N(Q)$  has the components  $N_{11}=1+\cos 2Qw$ ,  $N_{12}=i \sin 2Qw$ ,  $N_{21}=-i \sin 2Qw$  and  $N_{22}=1-\cos 2Qw$ .

Now we expand Eq. (79) in powers of  $k$  and  $p$ , where it should be noted that the summation over  $Q$  contains the  $Q=0$  component. Using the relations

$$T_{k,p}^{xx} = \frac{p^2}{\eta(k^2+p^2)^2}, \quad (80)$$

$$T_{k-Q,p}^{xx} \approx \frac{p^2}{\eta Q^4}, \quad (81)$$

Eq. (79) becomes, in the long-wavelength limit,

$$\dot{\zeta}_{k,p}^{(1)} = -\frac{p^2}{\eta(k^2+p^2)^2} [(D_{\perp} p^2 + Dk^2 + Kp^4) \zeta_{k,p}^{(1)} - ikB_2 \zeta_{k,p}^{(2)}], \quad (82)$$

$$\dot{\zeta}_{k,p}^{(2)} = -\frac{\gamma p^2}{\eta} [B_1 \zeta_{k,p}^{(2)} + ikB_2 \zeta_{k,p}^{(1)}], \quad (83)$$

where

$$\gamma = \frac{1}{2} \sum_{Q \neq 0} \frac{1 - \cos 2Qw}{Q^4}. \quad (84)$$

The reason for the appearance of the factor  $p^2$  in Eq. (83) is clear. We have so far ignored the diffusive effect in Eq. (72), and considered only the hydrodynamic interactions. Therefore, the concentration must be conserved within each domain, and hence the effective diffusion exists only along the  $y$  axis in Eq. (83), i.e., along the interface.

By comparing Eqs. (82) and (83), we note that if  $k^2 \ll p^2$  and  $K \ll \gamma B_1$  for  $D_{\perp} = 0$ , the relaxation of  $\zeta^{(2)}$  is rapid so that we can eliminate it to obtain

$$\frac{d}{dt} \zeta_{k,p}^{(1)} = -\frac{p^2}{\eta(k^2+p^2)^2} [D_{\parallel} k^2 + Kp^4] \zeta_{k,p}^{(1)}. \quad (85)$$

This is also justified when  $\kappa \rightarrow 0$ , since the constant  $B_1$  becomes infinite in this limit. The coefficients are the same as those in the previous sections. If necessary, we may add to Eq. (85) the contribution from the diffusive effect given by Eq. (71). This equation has been obtained by several authors [29,30]. However, under general conditions, the phase variable  $\zeta^{(1)}$  has to be coupled with the other slow mode  $\zeta^{(2)}$  as in the set of equations (82) and (83).

## VII. DISCUSSION

We have presented a systematic method to develop the equations governing interface dynamics in layered structures.

It is emphasized that the basic equations (10) and (11) and (52) and (53) do not possess any Lyapunov functional when  $\tau_1$  and  $\tau_2$  are finite. Although we have taken a limit in which these parameters vanish, this is simply because we have demonstrated our theory mainly for the systems near equilibrium. The interface dynamics can be formulated without taking these limits as has already been shown for a nonconserved reaction-diffusion system in Ref. [21]. In the present paper, we have extended the methods to conserved systems and binary fluids.

The motion of domains can be divided into two parts. One is the collective motion of center of gravity of each domain, which is described by a phase variable  $\zeta^{(1)}$ . The other is the internal motion of each domain denoted by  $\zeta^{(2)}$ .

Equations of motion for  $\zeta^{(1)}$  and  $\zeta^{(2)}$  in conserved and nonconserved systems take the following forms:

$$\frac{\partial \zeta^{(1)}}{\partial t} = -\Gamma^{(1)} \frac{\delta H}{\delta \zeta^{(1)}}, \quad (86)$$

$$\frac{\partial \zeta^{(2)}}{\partial t} = -\Gamma^{(2)} (i\vec{\nabla})^a \frac{\delta H}{\delta \zeta^{(2)}}, \quad (87)$$

where  $a=0$  for a nonconserved case and  $a=2$  for a conserved case, and  $H$  [which should not be confused with the mean curvature defined by Eq. (18)] is given by

$$H = \frac{1}{2} \int dx dy \left[ D \left( \frac{\partial \zeta^{(1)}}{\partial x} \right)^2 + D_{\perp} \left( \frac{\partial \zeta^{(1)}}{\partial y} \right)^2 + K \left( \frac{\partial^2 \zeta^{(1)}}{\partial y^2} \right)^2 + B_1 (\zeta^{(2)})^2 + 2B_2 \frac{\partial \zeta^{(1)}}{\partial x} \zeta^{(2)} \right]. \quad (88)$$

This Hamiltonian is invariant under the transformation  $x \rightarrow -x$ , since  $\zeta^{(1)}$  changes its sign whereas  $\zeta^{(2)}$  is invariant. Using the same reasoning, the coupling term  $(\partial \zeta^{(1)}/\partial y) \zeta^{(2)}$  should not exist. In the case of binary fluids, we can write the equations formally as

$$\frac{\partial \zeta^{(1)}}{\partial t} = \frac{1}{\eta(\nabla^2)^2} \frac{\partial^2}{\partial y^2} \frac{\delta H}{\delta \zeta^{(1)}}, \quad (89)$$

$$\frac{\partial \zeta^{(2)}}{\partial t} = \frac{\gamma}{\eta} \frac{\partial^2}{\partial y^2} \frac{\delta H}{\delta \zeta^{(2)}}. \quad (90)$$

These sets of equations show that although  $\zeta^{(2)}$  has a ‘‘mass’’ term with coefficient  $B_1$  in  $H$ , it does not produce a finite relaxation rate of  $\zeta^{(2)}$  in the long-wavelength limit except in the nonconserved case. In the conserved system and binary fluids, the variable  $\zeta^{(2)}$  is generally a slow mode in the limit of long-wavelength deformations because of the

conservation law. As we have remarked,  $\zeta^{(2)}$  can be eliminated only when the deformation normal to the layers is spatially sufficiently weak compared with the undulation of the layers. In fact, Eqs. (89) and (90) give us the relaxation rate  $\Omega^{(2)}$  of  $\zeta^{(2)}$  approximately as

$$\Omega^{(2)} = \frac{\gamma p^2 B_1}{\eta} \left[ 1 - \frac{B_2^2}{\gamma B_1^2} \frac{k^2}{(k^2 + p^2)^2} \right], \quad (91)$$

where the second term is the correction due to the coupling with  $\zeta^{(1)}$ , which is, however, not necessarily small for  $p \rightarrow 0$ .

Note that the case of pure Coulomb long-range interactions has to be handled carefully with the limit  $\kappa \rightarrow 0$ . In all cases studied here, when  $\kappa$  is sufficiently small, the internal mode  $\zeta^{(2)}$  can be eliminated adiabatically, and the phase variable description involving  $\zeta^{(1)}$  is complete.

In this paper, we have restricted ourselves to a one-dimensional layered structure. However, the theory is easily extended to those in higher dimensions such as a hexagonal structure of disk-shaped domains and a face-centered-cubic structure of spherical domains. Evaluation of nonlinear terms is also straightforward, although the calculations are more involved.

#### ACKNOWLEDGMENTS

T.O. is grateful to Masao Doi, Ayako Tetsuka, and Yumino Hayase for valuable discussions at the early stage of this work. This work was supported by a Grant-in-Aid of Ministry of Education, Science and Culture of Japan. D.J. is grateful for the support of the NSF under Grant. No. DMR92-17935.

#### APPENDIX

Here we summarize some of the steps in the derivation of the coupled set of interface equations of motion in Sec. III. From Eqs. (22) and (24), the value of  $v$  at an interface is given explicitly by

$$\begin{aligned} v_l^\pm = & \alpha \int \frac{d\vec{q}}{(2\pi)^2} \int dy' \sum_m \left[ \int_{b_m^-}^{b_m^+} dx' \frac{1}{q^2 + \kappa^2} \right. \\ & \times \exp[iq_x(b_n^\pm - x') + iq_y(y - y')] - \int_{b_m^+}^{b_{m+1}^-} dx' \frac{1}{q^2 + \kappa^2} \\ & \left. \times \exp[iq_x(b_n^\pm - x') + iq_y(y - y')] \right]. \quad (A1) \end{aligned}$$

Note that  $b_n^\pm$  contains the deformations  $\zeta_n^\pm(y)$  as in Eq. (23).

The zeroth order  $v_l^{(0)\pm}$  is obtained by ignoring  $\zeta_n^\pm$ . From Eq. (A1), it is given by

$$\begin{aligned} v_l^{(0)\pm} = & \alpha \int \frac{d\vec{q}}{(2\pi)^2} \\ & \times \int dy' \sum_m \left[ \int_{m\ell-w}^{m\ell+w} dx' \frac{1}{q^2 + \kappa^2} \exp[iq_x(n\ell \pm w) \right. \\ & - iq_x x' + iq_y(y - y')] \\ & - \int_{m\ell+w}^{(m+1)\ell-w} dx' \frac{1}{q^2 + \kappa^2} \exp[iq_x(n\ell \pm w) - iq_x x' \\ & \left. + iq_y(y - y')] \right] \\ = & \alpha \int \frac{dq_x}{2\pi} \sum_m \left[ \int_{m\ell-w}^{m\ell+w} dx' \frac{1}{q_x^2 + \kappa^2} \exp[iq_x(n\ell \pm w) \right. \\ & - iq_x x'] - \int_{m\ell+w}^{(m+1)\ell-w} dx' \frac{1}{q_x^2 + \kappa^2} \exp[iq_x(n\ell \pm w) \\ & \left. - iq_x x'] \right]. \quad (A2) \end{aligned}$$

The integral over  $x'$  in Eq. (A2) is readily carried out. By using the Poisson summation formula (29), one can obtain the zeroth solution (25).

In order to calculate the first order correction  $v_l^{(1)\pm}$ , one needs to expand  $\zeta_m^\pm$  and  $\zeta_n^\pm$  contained in  $b_m^\pm$  and  $b_n^\pm$ , respectively, in Eq. (A1). Hence the correction consists of two parts:

$$\begin{aligned} v_l^{(1)\pm} = & \alpha \zeta_n^\pm(y) \int \frac{dq_x}{2\pi} \sum_m \left[ \int_{m\ell-w}^{m\ell+w} dx' \frac{iq_x}{q_x^2 + \kappa^2} \exp[iq_x(n\ell \right. \\ & \left. \pm w) - iq_x x'] - \int_{m\ell+w}^{(m+1)\ell-w} dx' \frac{iq_x}{q_x^2 + \kappa^2} \exp[iq_x(n\ell \right. \\ & \left. \pm w) - iq_x x'] \right] + \alpha \int \frac{d\vec{q}}{(2\pi)^2} \\ & \times \int dy' \sum_m \frac{1}{q^2 + \kappa^2} \exp[iq_x(n\ell \pm w - m\ell) \\ & + iq_y(y - y')] [\zeta_m^+(y') e^{-iq_x w} - \zeta_m^-(y') e^{iq_x w} \\ & - \zeta_{m+1}^-(y') e^{-iq_x(\ell-w)} + \zeta_m^+(y') e^{-iq_x w}]. \quad (A3) \end{aligned}$$

Evaluation of the term which contains the factor  $\zeta_n^\pm(y)$  is straightforward, which gives us the first term in Eqs. (26) and (27). The integrals over  $q_y$  and  $y'$  in the remaining term of Eq. (A3) can be easily carried out by introducing the Fourier transform (28). Using the formula (29), one finally obtains the second term in Eqs. (26) and (27).

- [1] P. G. de Gennes and J. Prost, *The Physics of Liquid Crystals* (Clarendon, Oxford, 1993).
- [2] F. S. Bates and G. H. Fredrickson, *Annu. Rev. Phys. Chem.* **41**, 525 (1990).
- [3] M. Seul and D. Andelman, *Science* **267**, 476 (1995).
- [4] C. M. Knobler, *J. Phys.: Condens. Matter* **3**, S17 (1991).
- [5] H. Moewald, R. M. Kenn, D. Degenhardt, K. Kjaer, and J. Als-Nielsen, *Physica A* **168**, 1277 (1990).
- [6] *Spatio-Temporal Patterns in Nonequilibrium Complex Systems*, edited by P. E. Cladis and P. Palffy-Muhoray, NATO Advanced Research Workshop (Addison-Wesley, New York, 1995).
- [7] Q. Ouyang and H. L. Swinney, *Nature (London)* **352**, 610 (1991).
- [8] Y. Pomeau and P. Manneville, *J. Phys. Lett.* **40**, 609 (1979).
- [9] Y. Kuramoto, *Chemical Oscillations, Waves and Turbulence* (Springer-Verlag, Berlin, 1984).
- [10] M. C. Cross and A. C. Newell, *Physica D* **10**, 299 (1984).
- [11] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, New York, 1995).
- [12] K. Kawasaki and T. Ohta, *Physica A* **139**, 223 (1986).
- [13] A. Deutsch and S. A. Safran, *Phys. Rev. E* **54**, 3906 (1996).
- [14] K. Kawasaki and T. Ohta, *Prog. Theor. Phys.* **68**, 129 (1982); *Physica A* **118**, 175 (1983).
- [15] *Dynamics of Curved Fronts*, edited by R. Pelce (Academic, New York, 1988); D. Kessler, J. Koplik, and H. Levine, *Adv. Phys.* **37**, 255 (1988).
- [16] T. Garel and S. Doniach, *Phys. Rev. B* **26**, 325 (1982).
- [17] M. Seul and R. Wolfe, *Phys. Rev. A* **46**, 7534 (1992).
- [18] T. Ohta and K. Kawasaki, *Macromolecules* **19**, 2621 (1986).
- [19] J. J. Tyson and P. C. Keener, *Physica D* **32**, 327 (1988).
- [20] B. S. Kerner and V. V. Osipov, *Usp. Fiz. Nauk.* **160**, 1 (1990) [*Sov. Phys. Usp.* **33**, 679 (1990)].
- [21] T. Ohta, A. Ito, and A. Tetsuka, *Phys. Rev. A* **42**, 3225 (1990).
- [22] M. Bahiana and Y. Oono, *Phys. Rev. A* **41**, 6763 (1990).
- [23] M. Caginalp, *Ann. Phys. (N.Y.)* **172**, 136 (1986).
- [24] R. Kobayashi, *Physica D* **63**, 410 (1993).
- [25] G. H. Fredrickson, *J. Rheol.* **38**, 1045 (1994).
- [26] D. Andelman, F. Brochard, and J.-F. Joanny, *J. Chem. Phys.* **86**, 3673 (1987).
- [27] H. M. McConnell and V. T. Moy, *J. Phys. Chem.* **92**, 4520 (1988).
- [28] E. I. Kats and J. Lajzerowicz, *Zh. Éksp. Teor. Fiz.* **110**, 899 (1996) [*Sov. Phys. JETP* **83**, 495 (1996)].
- [29] M. Goulian and S. T. Milner, *Phys. Rev. Lett.* **74**, 1775 (1995). The viscosity  $\eta'$  in this paper has not been considered in the present model.
- [30] A. Onuki and J. Fukuda, *Macromolecules* **28**, 8788 (1995).