

## Self-consistent solution of phase separation with competing interactions

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We present a solution of a modified time-dependent Ginzburg-Landau equation in the limit of infinite order-parameter dimension  $N$ . The scalar ( $N = 1$ ) model is believed to describe phase separation in chemically reactive binary mixtures, block copolymers, and other systems where competing short-range and long-range interactions give rise to steady-state, spatially periodic structures. We present exact analytical expressions for the time dependence of the dynamic structure factor  $S(\mathbf{k}, t)$  and the peak position  $k_m(t)$ . We compare the scaling behavior for  $N = \infty$  with that observed in the scalar model.

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Understanding the various mechanisms that give rise to both equilibrium and nonequilibrium pattern formation in complex systems is a problem of long-standing interest [1]. Nonequilibrium pattern formation occurs, e.g., in incompatible binary mixtures undergoing phase separation by spinodal decomposition [2]. During spinodal decomposition, a mixture of two species of molecules  $A$  and  $B$  will become unstable with respect to long-wavelength concentration fluctuations when the mixture is quenched into the unstable region. This process ultimately leads to macroscopic phase separation via an interconnected morphology which coarsens self-similarly with time. In many situations, however, competing phenomena may interfere with phase separation, producing stable, periodic patterns (cf. Fig. 1). For example, magnetic systems and dipolar fluids ordering in the presence of long-range, Coulombic interactions often exhibit striped, lamellar domain patterns [3]. In phase separating mixtures of block copolymers, the finite block length competes with the thermodynamic demixing of the constituent monomers, resulting in equilibrium, microphase-separated lamellar, hexagonal, and micellar structures, depending on the constituent concentrations [4–6]. Conventional phase separation in binary mixtures and polymer blends can also be altered by chemical reactions [7,8]. For example, a reaction such as  $A \rightleftharpoons B$  tends to spatially mix the two species, and when this reaction occurs simultaneously with spinodal decomposition, the phase separation process evolves into a steady-state pattern in which the demixing thermodynamic and mixing reactive processes balance [7].

The theoretical understanding of spinodal decomposition in binary mixtures is based mainly on the Cahn-Hilliard theory [2,9,10]. Consider the following, modified Cahn-Hilliard equation:

$$\frac{\partial \psi(\mathbf{x}, t)}{\partial t} = M \nabla^2 \frac{\delta F\{\psi(\mathbf{x}, t)\}}{\delta \psi(\mathbf{x}, t)} - W \psi(\mathbf{x}, t), \quad (1)$$

where  $\psi(\mathbf{x}, t)$  is the scalar order-parameter field, given, e.g., by the concentration difference  $\phi_A - \phi_B$ ,  $M$  is related to the mobility, and  $F\{\psi(\mathbf{x}, t)\}$  is the standard Cahn-Hilliard free energy functional [2,9],

$$F\{\psi(\mathbf{x}, t)\} = \int d\mathbf{x} \left[ f(\psi) + \frac{\kappa}{2} |\nabla \psi(\mathbf{x}, t)|^2 \right], \quad (2)$$

where  $f(\psi)$ , the local, coarse-grained bulk free energy of mixing, has a double-well structure below the critical point, and  $\kappa$  is related to the interaction range. When  $W = 0$ , Eq. (1) reduces to the usual Cahn-Hilliard equation, describing phase separation in an immiscible binary ( $A$ - $B$ ) mixture. When  $W \neq 0$ , (1) has been proposed to describe phase separation in both symmetric diblock copolymers [5] and chemically reactive binary mixtures [7]. In diblock ( $AB$ ) copolymers,  $W$  is assumed to be inversely proportional to the square of the block length, while in a chemically reactive mixture (e.g.,  $A \rightleftharpoons B$ ),  $W$  is identified with the reaction rate  $\Gamma_{AB}$ . Interestingly, Eq. (1) can be rewritten as [11]

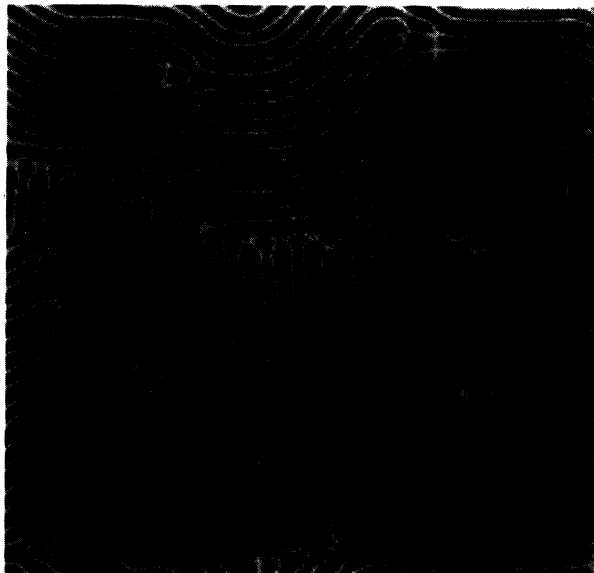


FIG. 1. Example of lamellar domain pattern observed in a variety of systems. This figure is from a numerical simulation of Eq. (1), and shows the equilibrium domain morphology of the concentration field for a system quenched into the two-phase region [7].

$$\frac{\partial \psi(\mathbf{x}, t)}{\partial t} = M \nabla^2 \frac{\delta \mathcal{F}\{\psi(\mathbf{x}, t)\}}{\delta \psi(\mathbf{x}, t)}, \quad (3)$$

where

$$\begin{aligned} \mathcal{F}\{\psi(\mathbf{x}, t)\} &= F\{\psi(\mathbf{x}, t)\} \\ &+ \frac{W}{2M} \int \int G(\mathbf{x}, \mathbf{x}') \psi(\mathbf{x}, t) \psi(\mathbf{x}', t) d\mathbf{x} d\mathbf{x}', \end{aligned} \quad (4)$$

and  $G(\mathbf{x}, \mathbf{x}')$  is the Green's function for Laplace's equation,  $\nabla^2 G(\mathbf{x}, \mathbf{x}') = -\delta(\mathbf{x} - \mathbf{x}')$ , with appropriate boundary conditions, so that, for  $d=3$ ,  $G(\mathbf{x}, \mathbf{x}') = (4\pi|\mathbf{x} - \mathbf{x}'|)^{-1}$ . Thus, the system described by Eq. (1) has a Lyapunov functional [11], given by Eq. (4), that contains both a short-range, attractive interaction whose strength is controlled by  $\kappa$ , and a long-range, repulsive interaction whose strength is controlled by  $W$ . Consequently, the modified Cahn-Hilliard equation that we consider here describes phase separation in systems with *competing* interactions. Experimental, numerical, and computational studies have shown that this competition gives rise to stable, periodic, microphase-separated structures that may exhibit novel scaling behavior [3,5–7].

We consider then the following modified, scalar time-dependent Ginzburg-Landau (TDGL) equation with a conserved order parameter. By taking the local free energy to be of the Landau form,  $f(\psi) = r\psi^2/2 + g\psi^4/4$ , Eq. (1) becomes

$$\begin{aligned} \frac{\partial \psi(\mathbf{x}, t)}{\partial t} &= \nabla^2 [r\psi(\mathbf{x}, t) + g\psi^3(\mathbf{x}, t) - \kappa \nabla^2 \psi(\mathbf{x}, t)] \\ &- W\psi(\mathbf{x}, t), \end{aligned} \quad (5)$$

which for  $W = 0$  gives the usual TDGL equation. Here  $r, g$ , and  $\kappa$  are phenomenological parameters related to the quench depth and interfacial energy, respectively. We seek an exact analytical solution for the evolution of the time-dependent structure factor  $S(\mathbf{k}, t)$  of the system described by Eq. 5 following a quench from high temperature into the unstable region. Such a solution is, at present, intractable in the scalar ( $N = 1$ ) case. However, the unmodified TDGL equation has been solved exactly for an  $N$ -component order parameter in the  $N = \infty$  limit for both conserved and nonconserved order parameter [12]. This solution can also be viewed as a self-consistent approximation of the scalar model [Eq. (5)] in which the term  $\psi^3$  is substituted with  $\langle \psi^2 \rangle \psi$ , where  $\langle \rangle$  represents an average over all initial configurations.

Here we present an exact analytical solution of Eq. (5) for an infinite-component order parameter, after an instantaneous quench from a high temperature to  $r < 0$  ( $T < T_c$ ), following the approach taken in Ref. [12]. Specifically, we derive an exact analytical expression for the time dependence of the dynamic structure factor  $S(\mathbf{k}, t)$  and the peak position  $k_m(t)$  in the spherical model approximation. We show that for values of  $W$  less than a critical value  $W_c$ , the structure factor asymptotically approaches a  $\delta$  function peaked around a constant inverse domain size  $k_{eq}$  which depends on the mobility  $M$ , the interfacial square gradient coefficient  $\kappa$ , and  $W$  as  $k_{eq} = (W/\kappa M)^\alpha$ , where  $\alpha = 1/4$  is the exponent

characterizing domain growth in the unmodified, large- $N$  TDGL model [12]. For  $W \geq W_c$  we show that the peak position of  $S(\mathbf{k}, t)$  approaches an asymptotic value  $k_\infty = (-r/2\kappa)^{1/2}$ , while the amplitude of  $S(\mathbf{k}, t)$  goes to zero. As we will see, the behavior predicted by the solution of the model in the limit of an infinite-component order parameter is consistent in some respects with behavior observed in numerical simulations of the *scalar* model.

We begin by generalizing Eq. (5) to a system with an  $N$ -component order parameter  $\vec{\psi}(\mathbf{x}, t) = (\psi_1(\mathbf{x}, t), \dots, \psi_N(\mathbf{x}, t))$ . For each component  $\alpha$ , we have an equation of motion of the form

$$\begin{aligned} \frac{\partial \psi_\alpha(\mathbf{x}, t)}{\partial t} &= \nabla^2 \left[ -\nabla^2 - 1 + \frac{1}{N} \sum_{\beta=1}^N \psi_\beta^2(\mathbf{x}, t) \right] \psi_\alpha(\mathbf{x}, t) \\ &- W\psi_\alpha(\mathbf{x}, t), \end{aligned} \quad (6)$$

where we have substituted the dimensionless variables  $\mathbf{x}' = \mathbf{x}\sqrt{-r/\kappa}$ ,  $t' = tMr^2/\kappa$ ,  $\psi' = \psi\sqrt{g/-r}$ , and  $W' = W' = W\kappa/Mr^2$ , so that  $W'$  is the only dimensionless parameter, and for simplicity we drop the prime everywhere. We consider an instantaneous quench to  $r < 0$  ( $T < T_c$ ), and assume the initial condition  $\langle \psi_\alpha(\mathbf{x}, t) \rangle = 0$ . In the limit  $N = \infty$ , we recover the spherical model and replace  $\frac{1}{N} \sum_{\beta=1}^N \psi_\beta^2(\mathbf{x}, t)$  by  $\langle \psi_\alpha^2(\mathbf{x}, t) \rangle$  in Eq. (6) [13]. Here  $\langle \rangle$  represents an ensemble average over the initial configurations. Moreover, by assuming translational invariance so that the pair correlation function  $g(\mathbf{x}, \mathbf{x}'; t) \equiv \langle \psi_\alpha(\mathbf{x}, t) \psi_\alpha(\mathbf{x}', t) \rangle = g(\mathbf{x} - \mathbf{x}'; t)$ , the quantity  $\langle \psi_\alpha^2(\mathbf{x}, t) \rangle = g(0, t) \equiv S(t)$  is independent of position  $\mathbf{x}$ . Thus, dropping the label  $\alpha$  and Fourier transforming over space, it is straightforward to obtain the equation of motion of the structure factor  $S(\mathbf{k}, t)$ :

$$\frac{\partial S(\mathbf{k}, t)}{\partial t} = -2k^2 [k^2 - 1 + S(t)] S(\mathbf{k}, t) - 2WS(\mathbf{k}, t), \quad (7)$$

where  $\mathbf{k}$  is a dimensionless wave vector and  $S(\mathbf{k}, t)$  is the spatial Fourier transform of  $\langle \psi_\alpha(\mathbf{x}, t) \psi_\alpha(\mathbf{x}', t) \rangle$ . The total integrated scattering intensity  $S(t)$  must be determined self-consistently:

$$S(t) = \int S(\mathbf{k}, t) \frac{d\mathbf{k}}{(2\pi)^d}, \quad (8)$$

where  $d$  is the spatial dimension of the system.

By replacing  $\frac{1}{N} \sum_{\beta=1}^N \psi_\beta^2(\mathbf{x}, t)$  by  $S(t)$ , we have, in effect, enabled a linearization of Eq. (6) in Fourier space by self-consistently “preaveraging” the nonlinear term in the equation [14]. Consequently, Eq. (7) can be integrated to obtain

$$S(\mathbf{k}, t) = S(\mathbf{k}, 0) e^{-2k^2 [k^2 t + Q(t)] - 2Wt}, \quad (9)$$

where we have defined  $Q(t) \equiv \int_0^t [S(t') - 1] dt'$ . To complete the solution,  $S(t)$  must be computed explicitly by integrating Eq. (9) over  $\mathbf{k}$ . To do so, we note that  $S(\mathbf{k}, t)$  has a maximum at a wave vector  $k_m(t)$  given by

$$k_m^2(t) = -\frac{Q(t)}{2t}, \quad (10)$$

which gives an expression for  $Q(t)$  in terms of the peak position  $k_m(t)$ . The solution for  $S(\mathbf{k}, t)$  can then be written as

$$S(\mathbf{k}, t) = S(\mathbf{k}, 0)e^{[k_m^4(t)-W]L^4(t)}e^{-[k^2-k_m^2(t)]^2L^4(t)}, \quad (11)$$

where we have defined  $L(t) \equiv (2t)^{1/4}$ . However, we still require the solution for  $k_m(t)$ . Using the method of steepest descent to solve for  $\dot{Q}(t)$ , and relating  $\dot{Q}(t)$  to  $k_m(t)$  through Eq. (10), we obtain the following equation when  $k_m^4(t)L^4(t) \gg 1$ :

$$e^{[k_m^4(t)-W]L^4(t)} = P[k_m(t)]^{2-d}L^2(t), \quad (12)$$

where

$$P = \frac{1}{B} \left[ -2k_m^2(t) + 1 - 4tk_m(t) \frac{dk_m(t)}{dt} \right]. \quad (13)$$

Here  $B = K_d S(k_m, 0) \sqrt{\pi}/4$ , with  $K_d = [2^{d-1} \pi^{d/2} \Gamma(d/2)]^{-1}$ . Inserting the right-hand side of Eq. (12) into Eq. (11) allows us to rewrite  $S(\mathbf{k}, t)$  in the final form:

$$S(\mathbf{k}, t) \simeq S(\mathbf{k}, 0) P[k_m(t)]^{-d} [k_m(t)L(t)]^2 \times e^{-[k_m(t)L(t)]^4 \left[ \frac{k^2}{k_m^2(t)} - 1 \right]^2}, \quad (14)$$

where the peak position  $k_m(t)$  is given by the solution of Eq. (12), and the width of the peak is given by  $\Delta k \equiv \langle [k - k_m(t)]^2 \rangle^{1/2} \sim k_m / [k_m(t)L(t)]^2$ . Taking the logarithm of both sides of Eq. (12) and dividing by  $L^4(t) \equiv 2t$  gives an expression for  $k_m(t)$  that is readily analyzed in the limit  $t \rightarrow \infty$ .

Careful analysis shows that there are two distinct solutions depending on the value of  $W$ . If  $W < 1/4$ , then Eq. (12) shows that the peak position reaches a nonzero steady-state value in the limit  $t \rightarrow \infty$ ,

$$k_{\text{eq}} = W^{1/4}. \quad (15)$$

For earlier times such that  $t$  is large and  $W \ll k_m^4(t)$  [and thus  $Wt \ll (d/8) \ln t$ ], we find

$$k_m(t) \sim \left( \frac{d}{4} \ln t \right)^\alpha [L(t)]^{-1}, \quad (16)$$

where  $\alpha = 1/4$ , and thus, apart from logarithmic factors,  $k_m(t) \sim t^{-1/4}$ . Note that this is the large- $N$  solution to the *unmodified* ( $W = 0$ ) TDGL equation in the limit  $t \rightarrow \infty$  [12]. The solution for  $k_m(t)$  is consistent with the scaling form

$$k_m(t, W) = W^{1/4} F(Wt), \quad (17)$$

with  $F(x) \rightarrow \text{const}$  for  $x \rightarrow \infty$  and  $F(x) \rightarrow x^{-1/4}$  for  $x \ll \infty$ .

Inserting Eq. (17) into Eq. (14) gives the exact solution for  $S(\mathbf{k}, t)$ . At late times, where  $[k_m(t)L(t)]^4 = 2Wt$ ,

$P = (1 - 2W^{1/2})/B$ , and  $S(\mathbf{k}, t)$  can be approximated by a Gaussian centered about  $k_{\text{eq}} = W^{1/4}$ , with width  $\Delta k \sim W^{1/4}/[2Wt]^{1/2}$  and amplitude  $A \sim k_{\text{eq}}^{-d} [k_{\text{eq}}L(t)]^2 \sim W^{-d/4} [2Wt]^{1/2}$ , which in the limit  $t \rightarrow \infty$  tends to a  $\delta$  function centered about  $k_{\text{eq}} = W^{1/4}$ . For earlier times, where, from Eq. (16),  $[k_m(t)L(t)]^2 \sim (\ln t)^{1/2}$ , the width of the peak scales with an inverse length *smaller* than  $k_m$  by a logarithmic factor, and  $S(\mathbf{k}, t)$  displays multi-scaling behavior as in the unmodified TDGL model in the  $N = \infty$  limit [12], since the exponent in Eq. (14) is a function of  $k/k_m$ .

The second distinct solution occurs for  $W \geq 1/4$ . In this case we find from Eq. (12) that  $k_m(t)$  asymptotically approaches the limiting value  $k_\infty = 1/\sqrt{2}$  exponentially, [15],

$$k_m^2(t) \simeq k_\infty^2 + \text{const} \times \frac{e^{-2(W-W_c)t}}{t^{3/2}}. \quad (18)$$

Consequently  $P$  and  $S(\mathbf{k}, t)$ , given in Eq. (14), asymptotically vanish in an exponential fashion. Thus an equilibrium, microphase-separated state is only predicted for values of  $W$  less than the critical value  $W_c = Mr^2/4\kappa$ , in dimensional units. For  $W > W_c$ , phase separation is prevented, and  $S(\mathbf{k}, t)$  vanishes as  $k_m$  approaches a limiting value given by  $k_\infty = (-r/2\kappa)^{1/2}$ , resulting in a periodic pattern which fades in time.

We now summarize the results of the scalar model and compare with the results found here. Studies of the modified, scalar TDGL equation [Eq. (5)] have suggested the existence of a critical value of  $W$  above which the homogeneous  $\psi = 0$  state is no longer unstable for  $T < T_c$ . Linearization of that equation gives a cutoff value  $W_c = 1/4$  above which phase separation is prevented [7]. The same value was obtained from a linear stability analysis of the free energy functional of Eq. (4) in  $d = 1$  by Liu and Goldenfeld [11]. Not surprisingly, the same cutoff value is found in the  $N = \infty$  model. Below  $W_c$ , two limiting regimes are found in the scalar model, characterized by small and large  $W$ , respectively. In both regimes, it is found that

$$k_m(t, W) = W^\alpha F(Wt), \quad (19)$$

where  $F(x) \rightarrow \text{const}$  for  $x \rightarrow \infty$  and  $F(x) \rightarrow x^{-\alpha}$  for  $x \ll \infty$ , and

$$S(k, t, W) = [k_m(t)]^{-d} f(k/k_m(t), Wt), \quad (20)$$

where, for  $Wt \ll 1$  and  $t$  large,  $f(x, y) \sim f(x, 0)$ , while for  $Wt \gg 1$ ,  $k_m(t) = k_{\text{eq}} = W^\alpha$  and  $f(x, y) \sim f(x, \infty)$ . Note that, in the scalar model, the position of the peak and the width of the peak instead scale together in the same way.

For small  $W$ ,  $\alpha$  was found to be consistent with a value  $1/3$  in the scalar case [5,7,11]. In this regime, the interfacial thickness is small compared to the domain size, and the composition profile is well approximated by a square-wave-like function [5,11]. For large  $W$ ,  $\alpha$  was found to be roughly  $1/4$  [5,6,11]. In this regime, the interfacial thickness is comparable to the domain size, and the com-

position profile is approximately sinusoidal [11]. In principle, one expects to find  $\alpha = 1/3$  as the true asymptotic exponent of the scalar model, and  $\alpha = 1/4$  as an effective exponent observed simply because the domain size saturates for large  $W$  before the true asymptotic growth regime is reached. However, it has been suggested [5,6,11] that the scalar model applies to diblock copolymer melts, in which  $W \propto 1/n^2$ , where  $n$  is the chain length, and that the two limiting regimes of this model, small  $W$  ( $\alpha = 1/3$ ) and large  $W$  ( $\alpha = 1/4$ ), correspond to the strong and weak segregation limits, respectively, in diblock copolymers [4]. The scalar model also describes chemically reacting binary mixtures, in which  $A \rightleftharpoons B$  and the forward and backward reaction rates are equal and proportional to  $W$  [7], and is expected to describe phase separation in other systems characterized by the Lyapunov functional in Eq. (4).

In the  $N = \infty$  limit, we find that  $k_m$  obeys a scaling law for  $W < 1/4$  given by Eq. (17), similar to Eq. (19) with  $\alpha = 1/4$ , as in the large- $W$  limit of the scalar model.

Moreover, the structure factor in Eq. (14), in the limit  $t \rightarrow \infty$ , is given by a scaling relation similar to Eq. (20) with  $f(x, y)$  approximated by a Gaussian centered at  $x = 1$ , which in the limit  $y \rightarrow \infty$  tends to a  $\delta$  function centered at  $x = 1$ . That is, while the position of the peak of the structure factor tends to a constant, in agreement with the scalar case, the width of the peak continues to shrink to zero. This  $\delta$  function scaling form suggests a sinusoidally varying composition profile in  $d = 1$ . Such a profile has been used to describe the diffuse lamellar structures seen in the large- $W$  limit of the scalar model. Indeed, diffuse interfaces are expected for large  $N$  due to the continuous symmetry of the order parameter. Thus, the  $N = \infty$  TDGL model studied here seems to approximate the scalar model in the large- $W$  limit.

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- [14] In fact, replacing the term  $\psi^3$  with  $\langle \psi^2 \rangle \psi$  in the scalar TDGL equation, rather than the generalized equation, would in effect “preaverage” and thereby self-consistently linearize the scalar model, and we would find the identical solution. However, this “smoothing” procedure is only exact in the limit  $N = \infty$ .
- [15] Note that  $k_\infty$  is simply the value of  $k_m$  given by the usual linear theory, which is the wave vector at which the initial instability grows the fastest.

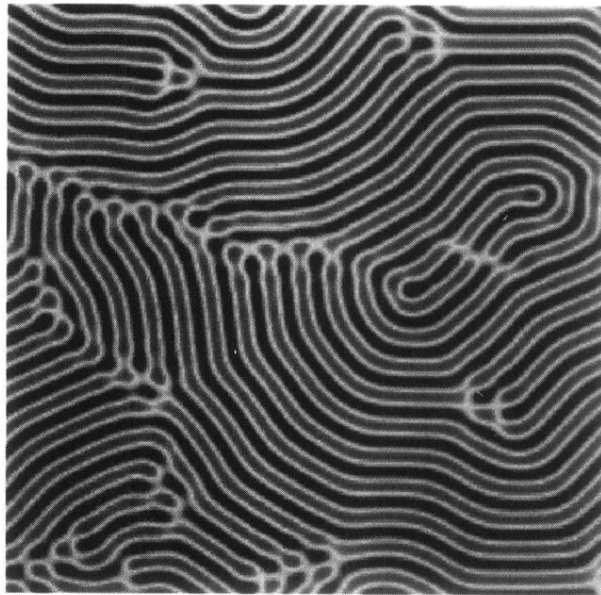


FIG. 1. Example of lamellar domain pattern observed in a variety of systems. This figure is from a numerical simulation of Eq. (1), and shows the equilibrium domain morphology of the concentration field for a system quenched into the two-phase region [7].