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Phase instability and coarsening in two dimensions

Chaouqi Misbah[1,*] and Paolo Politi[2,3,†]
[1]*Laboratoire de Spectrométrie Physique, UMR, 140 Avenue de la Physique, Université Joseph Fourier Grenoble and CNRS, 38402 Saint Martin d'Heres, France*
[2]*Istituto dei Sistemi Complessi, Consiglio Nazionale Delle Richerche, Via Madonna Del Piano 10, 50019 Sesto Fiorentino, Italy*
[3]*INFN Sezione di Firenze, via G. Sansone 1, 50019 Sesto Fiorentino, Italy*

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Instabilities and pattern formation is the rule in nonequilibrium systems. Selection of a persistent length scale or coarsening (increase in the length scale with time) are the two major alternatives. When and under which conditions one dynamics prevails over the other is a long-standing problem, particularly beyond one dimension. It is shown that the challenge can be defied in two dimensions, using the concept of phase diffusion equation. We find that coarsening is related to the λ dependence of a suitable phase diffusion coefficient, $D_{11}(\lambda)$, depending on lattice symmetry and conservation laws. These results are exemplified analytically on prototypical nonlinear equations.

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I. INTRODUCTION

Pattern formation (or morphogenesis) is abundant in nature, both in inanimate and living systems. Examples [1] are encountered in many branches of science: physics (e.g., sand ripples), chemistry (chemical spots, reminiscent of those on animal skins, e.g., jaguar), biology (asters of macromolecules during cell division), and so on. Patterns arise often due to the loss of stability of an initially structureless state. The diversity and richness of morphogenesis is concomitant to the nonlinear and nonequilibrium nature of these systems.

A classification of dynamics and patterns that prevail for a given nonlinear system is a challenging task. For example, given a system described by nonlinear equations, it is not obvious to state *a priori* whether one would observe the selection of a pattern with a specific length scale during time or rather coarsening (increase in length scale with time) would prevail.

Coarsening is also the primary scenario in phase-separation processes [2] and it attracts a continuous interest [3–10] motivated by the demand for a deeper understanding. This goal may be attained following two main directions: either investigating rigorous solutions to specific models [4,7] or developing general approaches which are valid for wide classes of models and/or equations. Regarding the second strategy, the only example we are aware of is due to Bray *et al.* [2,11], who analyzed some models which are derivable from a potential: comparing the global rate of energy change with the energy dissipation, the temporal growth law for the typical length scale, $\lambda(t)$, could be derived. The present Rapid Communication can be seen as complementary, in two respects. First, we do not impose to the equations to be derivable from a potential. Second, we do not assume that coarsening does occur, we are able to determine if and when it occurs instead. Note that our study does not address the other kind of coarsening which consists in an increase in a domain with a certain orientation (or pattern) and/or length scale at the expense of another domain [12].

A few years ago we established a coarsening criterion in one dimension [8], for certain classes of nonlinear equations, generalizations of the celebrated models A and B of the dynamics [13]. The criterion demands to analyze the stationary periodic states, which are found to solve Newton's equations where the spatial variable plays the role of time and the fictitious particle oscillates in a potential well of arbitrary shape (different equations give rise to different potentials). The oscillation period of the particle corresponds, in the mechanical analogy, to the wavelength λ of the periodic steady state, and it is a function of the amplitude A, $\lambda=\lambda(A)$. The criterion simply states that coarsening occurs if and only if $d\lambda/dA>0$.

Our analysis anticipated the existence of an "interrupted coarsening" scenario, which has recently been invoked [14] in the context of wind driven sand dunes. It is clear that these physical phenomena (as well as, e.g., mound coarsening at the nanoscale in crystal growth [15]) demand for the construction of a full two-dimensional (2D) approach, which is the basic goal of the present Rapid Communication.

However, in our former one-dimensional (1D) criterion, the spatial variable plays the role of time (see above). Therefore, it is obvious that this concept is limited to one spatial dimensional systems and extensions of the criterion to higher dimensions seemed to present a significant challenge. It is shown here that the challenge to find general criteria for understanding if a two-dimensional model shows coarsening or not can be defied, following the concept of phase diffusion equation [16]. Before discussing the details, we give a simple overview of the method and results.

II. METHOD

A perfectly periodic steady state is defined through a pair of wave vectors $\mathbf{q}_1,\mathbf{q}_2$, and it is a function of the phases $\phi_{1,2}=\mathbf{q}_{1,2}\cdot\mathbf{x}$. Perturbations make $\mathbf{q}_{1,2}$ acquire slow dependencies on time and space, $\mathbf{q}_i=\mathbf{q}_i(\mathbf{X},T)$. The dynamical response to perturbations is described by phase equations, which have,

*chaouqi.misbah@ujf-grenoble.fr
†paolo.politi@isc.cnr.it

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at the lowest order, the form of linear diffusion equations:

$$\partial_T \phi_1(\mathbf{X},T) = D_{11}^{11}\frac{\partial^2\phi_1}{\partial X_1^2} + D_{11}^{12}\frac{\partial^2\phi_2}{\partial X_1^2} + D_{22}^{11}\frac{\partial^2\phi_1}{\partial X_2^2} + \cdots,$$

$$\partial_T \phi_2(\mathbf{X},T) = D_{11}^{21}\frac{\partial^2\phi_1}{\partial X_1^2} + D_{11}^{22}\frac{\partial^2\phi_2}{\partial X_1^2} + D_{22}^{21}\frac{\partial^2\phi_1}{\partial X_2^2} + \cdots. \quad (1)$$

Setting $\phi_{1,2}=\phi_{1,2}^0 \exp(\omega T + i\mathbf{K}\cdot\mathbf{X})$, standard Fourier analysis allows to determine the stable or unstable character of phase dynamics. Coarsening is related to instability, which is signaled by a positive ω. The requirement $\omega>0$ for some $\mathbf{K}$ implies a condition on the diffusion coefficients D_{kl}^{ij}, which are functions of steady-state properties only. The central result is that the condition $\omega>0$ finally takes the form $d\mathcal{A}/d\lambda>0$, where $\mathcal{A}$ is some function of the stationary periodic solution having wavelength λ.

In one dimension, $\mathcal{A}$ is shown to correspond to the amplitude of the stationary solution [8]. In two dimensions, $\mathcal{A}$ takes different expressions, depending on the underlying symmetry of the periodic stationary solutions, and it does not seem to have a simple physical interpretation. Nevertheless, the condition $d\mathcal{A}/d\lambda>0$ thus have a simple reading: coarsening occurs if and only if $\mathcal{A}$ is an increasing function of λ. That is to say the coarsening criterion can still be established only upon inspecting *steady-state solutions*. Even more importantly, the coarsening law, $\lambda(t)$, which describes the time dependence of the typical size of the pattern, is obtained through a relation of the form $D(\lambda)t\sim\lambda^2$ (or equivalently $\omega\sim 1/t$), where D is a typical diffusion constant. In the limit of large time, a power-law behavior, $\lambda\simeq t^n$, is expected.

We shall exemplify the method on classical model equations. For instance, we show that $n=\frac{1}{2}$ for the nonconserved real Ginzburg-Landau (GL) equation (model A) and $n=\frac{1}{3}$ for the conserved Cahn-Hilliard (CH) equation (model B). As will be recognized, the interesting message is that the methodology does not evoke whether the model equation are variational or not. Thus the study should work with any other equation, be it of potential (i.e., variational) nature or not.

III. GENERALIZED GINZBURG-LANDAU-TYPE MODELS

We now apply and discuss our method for a nonconserved class of equations,

$$\partial_t u = A(u) + \nabla^2 u, \quad (2)$$

which reduces, for $A(u)=u-u^3$, to the famous real GL (or Allen-Cahn) equation. This equation is known to exhibit perpetual coarsening, with $n=\frac{1}{2}$ [2]. Here we only require $A(u)\approx u$ for small u, otherwise $A(u)$ can be any function of u.

A steady periodic state $u_0(\mathbf{x})$ depends on the fast spatial variables $\mathbf{x}=(x_1,x_2)$, or equivalently $\phi_{1,2}=\mathbf{q}_{1,2}\cdot\mathbf{x}$ which are the fast (constant) phases. Phase modes are studied by perturbing the perfectly periodic steady state. The perturbations of interest depend on slow spatial and temporal variables. The slow character of relevant perturbations reflects the property of the Goldstone mode [if $u_0(\mathbf{x})$ is a periodic solution, so is $u_0(\mathbf{x}+\mathbf{R_0})$], which has infinite relaxation time and is a quasidangerous mode, making slowly varying perturbations to be most persistent. Let us encode the slow modulation by a dimensionless parameter $\epsilon\ll 1$. We define slow spatial and time scales as

$$X_1=\epsilon x_1, \quad X_2=\epsilon x_2, \quad T=\epsilon^2 t, \quad (3)$$

where the factor ϵ^2 is an ansatz dictated by the fact that phase rearrangement occurs via diffusion. We introduce the slow phases $\psi_{1,2}=\epsilon\phi_{1,2}$ so that $\mathbf{q}_i=\nabla_{\mathbf{x}}\phi_i=\nabla_{\mathbf{X}}\psi_i$, where $\mathbf{X}=(X_1,X_2)=\epsilon\mathbf{x}$.

In general terms, at order ϵ we can write [8]

$$\partial_t = \epsilon\left[\frac{\partial\psi_1}{\partial T}\partial_{\phi_1} + \frac{\partial\psi_2}{\partial T}\partial_{\phi_2}\right], \quad (4)$$

$$\nabla_{\mathbf{x}} = \mathbf{q}_1\partial_{\phi_1} + \mathbf{q}_2\partial_{\phi_2} + \epsilon\nabla_{\mathbf{X}}. \quad (5)$$

The above expressions for the derivative, along with the standard expansion $u=u_0+\epsilon u_1$, are substituted into Eq. (2). The zeroth-order problem yields the differential equation obeyed by the two-dimensional profile of steady states,

$$\mathcal{N}[u_0] \equiv A(u_0) + \nabla_0^2 u_0 = 0, \quad (6)$$

where the subscript in the ∇^2 operator is taken to mean that it is evaluated for $\epsilon=0$ in Eq. (5).

The next order in ε has the form [16,17]

$$\mathcal{L}[u_1] = g(u_0, \partial_T\psi_i, \partial_{X_iX_j}\psi_k), \quad (7)$$

where $\mathcal{L}=\mathcal{L}_0=[A'(u_0)+\nabla_0^2]$ is the linear operator obtained as Frechét derivative of $\mathcal{N}$ and

$$g = \frac{\partial\psi_1}{\partial T}\frac{\partial u_0}{\partial\phi_1} + \frac{\partial\psi_2}{\partial T}\frac{\partial u_0}{\partial\phi_2} - [2(\mathbf{q}_1\partial_{\phi_1} + \mathbf{q}_2\partial_{\phi_2})\cdot\nabla_{\mathbf{X}}u_0 + (\nabla_{\mathbf{X}}\cdot\mathbf{q}_1)\partial_{\phi_1}u_0 + (\nabla_{\mathbf{X}}\cdot\mathbf{q}_2)\partial_{\phi_2}u_0]. \quad (8)$$

The expression for g can be rewritten as

$$g = (\partial_T\psi_1)v_1 + (\partial_T\psi_2)v_2 - 2q_{ki}(\psi_l)_{ij}\frac{\partial v_k}{\partial q_{lj}} - (\psi_l)_{ii}v_l, \quad (9)$$

where $v_i=\partial_{\phi_i}u_0$, q_{ki} is the ith component of $\mathbf{q}_k$, and $(\psi_l)_{ij}=\partial^2\psi_l/\partial X_i\partial X_j$.

In order to avoid secular terms in Eq. (7), if the homogeneous equation $\mathcal{L}_0^\dagger[w]=0$ has a nonvanishing solution, w must be orthogonal to g [18], $\langle w,g\rangle=0$ (the precise definition of the inner product is given below).

There exist two nonvanishing solutions w_1, w_2 in two dimensions, resulting thus in two solvability conditions. Since $\mathcal{L}_0^\dagger=\mathcal{L}_0$ for Eq. (2), we easily get $w_i=v_i$. This leads to the sought-after phase diffusion equations,

$$\langle v^2\rangle\partial_T\psi_1 + \langle v_1v_2\rangle\partial_T\psi_2 = \sum_{ijl}(\psi_l)_{ij}A_{ij}^l, \quad (10)$$

$$\langle v_1v_2\rangle\partial_T\psi_1 + \langle v^2\rangle\partial_T\psi_2 = \sum_{ijl}(\psi_l)_{ij}B_{ij}^l, \quad (11)$$

where $\langle v^2\rangle=\langle v_1^2\rangle=\langle v_2^2\rangle$ and

$$A_{ij}^{l} = 2\sum_{k} q_{ki}\left\langle v_1 \frac{\partial v_k}{\partial q_{lj}}\right\rangle + \delta_{ij}\langle v_1 v_l\rangle, \tag{12}$$

$$B_{ij}^{l} = 2\sum_{k} q_{ki}\left\langle v_2 \frac{\partial v_k}{\partial q_{lj}}\right\rangle + \delta_{ij}\langle v_2 v_l\rangle. \tag{13}$$

There are five Bravais lattices in 2D (oblique, rectangular, centered rectangular, hexagonal, and square). We have performed explicitly the calculation for squares, hexagons, and rectangles. Here, we shall focus on the first two symmetries. For square symmetry, $\mathbf{q}_1 = q(1,0)$, $\mathbf{q}_2 = q(0,1)$, and $\langle v_1 v_2\rangle = 0$. Finally, we get

$$\frac{\partial \psi_1}{\partial T} = D_{11}\frac{\partial^2 \psi_1}{\partial X_1^2} + \frac{\partial^2 \psi_1}{\partial X_2^2} + D_{12}\frac{\partial^2 \psi_2}{\partial X_1 X_2},$$

$$\frac{\partial \psi_2}{\partial T} = \frac{\partial^2 \psi_2}{\partial X_1^2} + D_{11}\frac{\partial^2 \psi_2}{\partial X_2^2} + D_{12}\frac{\partial^2 \psi_1}{\partial X_1 X_2}, \tag{14}$$

with $D_{11} = D_{11}^{\mathrm{sq}} = \partial_q(q\langle v^2\rangle)/\langle v^2\rangle$ and $D_{12} = D_{12}^{\mathrm{sq}} = D_{11}^{\mathrm{sq}} - 1$. A linear stability analysis is performed by setting $\psi_{1,2} = \psi_{1,2}^0 \exp(\omega T + i\mathbf{K}\cdot\mathbf{X})$ in Eqs. (14), which yields

$$\omega^2 + \omega(1 + D_{11})K^2 + D_{11}K^4 = 0, \tag{15}$$

whose roots are $\omega_1 = -K^2$ and $\omega_2 = -D_{11}K^2$.

While $\omega_1 < 0$, the sign of $\omega_{\mathrm{NC}} = \omega_2$ depends on the sign of $\partial_q \mathcal{A} = \partial_q(q\langle v^2\rangle)$. Therefore, ω_2 is positive, implying phase instability if and only if $\mathcal{A} = q\langle v^2\rangle$ is an increasing function of the wavelength λ.

For hexagonal symmetry, $\mathbf{q}_{1,2} = q(\frac{1}{2}, \pm\frac{\sqrt{3}}{2})$ and $\psi_{1,2}$ must be linearly combined in order to get Eqs. (14), with a different

$$D_{11} = D_{11}^{\mathrm{hex}} = \frac{\partial_q(q\langle v^2\rangle^2)}{\langle v^2\rangle^2}, \tag{16}$$

albeit the relation $D_{12}^{\mathrm{hex}} = D_{11}^{\mathrm{hex}} - 1$ still holds.

IV. CAHN-HILLIARD-TYPE MODELS

Let us now consider a conserved equation, a generalized form of the well-known CH equation,

$$\partial_t \mu = -\nabla^2[A(u) + \nabla^2 u]. \tag{17}$$

We only provide the result. We formally obtain Eq. (7), but now $\mathcal{L} = -\nabla_0^2 \mathcal{L}_0$ and $g = (\partial_T \psi_1)v_1 + (\partial_T \psi_2)v_2 + \nabla_0^2 \nabla_1^2 u_0$, where $\nabla_1^2 u_0$ is a shorthand notation for the expression in square parentheses in Eq. (8). $\mathcal{L}$ is not self-adjoint because $\mathcal{L}^\dagger = -\mathcal{L}_0 \nabla_0^2$ and its kernel w_i is such that $\mathcal{L}^\dagger w_i = 0 = -\mathcal{L}_0 \nabla_0^2 w_i$ so that $\nabla_0^2 w_i = v_i = \partial u_0/\partial \phi_i$.

The orthogonality conditions $\langle w_i g\rangle = 0$ are a bit more challenging to analyze and will be discussed elsewhere. Suffice it here to say that the relevant eigenvalue reads as

$$\omega_{\mathrm{C}} = I_{\mathrm{C}}\omega_{\mathrm{NC}}, \tag{18}$$

where ω_{C} and ω_{NC} are the eigenvalues associated with the unstable mode for the conserved and nonconserved versions of a given model, and $I_{\mathrm{C}} = \langle v^2\rangle/\langle wv\rangle$ is the factor taking into account the conservation law.

TABLE I. Summary of coarsening exponents for different models and base symmetries.

	Square	Other symmetries
Standard GL	1/3	1/2
Modified GL	1/2	1/2
Standard CH	1/4	1/3

V. COARSENING EXPONENT

The coarsening law is determined on the basis of the relation $|\omega(q)| \sim 1/t$, where $\omega = \omega_{\mathrm{NC}} = -D_{11}q^2$ or $\omega = \omega_{\mathrm{C}} = I_{\mathrm{C}}\omega_{\mathrm{NC}}$. A major role is played by the quantity

$$\langle v^2\rangle = \frac{1}{(2\pi)^2}\int_0^{2\pi} d\phi_1 \int_0^{2\pi} d\phi_2 \left(\frac{\partial u_0}{\partial \phi_1}\right)^2 = \frac{1}{(2\pi)^2}\int_0^{\lambda} dx_1 \int_0^{\lambda} dx_2 \left(\frac{\partial u_0}{\partial x_1}\right)^2. \tag{19}$$

Consider first the Ginzburg-Landau (nonconserved) equation. In this case, u_0 is a constant except along domain walls so $\langle v^2\rangle$ scales linearly with λ. More precisely, $\langle v^2\rangle = a/q + b$. Reporting this expression into D_{11} yields $n = \frac{1}{3}$ for square symmetry and $n = \frac{1}{2}$ for hexagonal symmetry. Preliminary analysis for rectangular symmetry leads to $n = \frac{1}{2}$ as well. In general had $\langle v^2\rangle$ scaled as $1/q^\alpha$ $(\alpha > 1)$ we would have obtained $n = \frac{1}{2}$ for all symmetries. Therefore, GL scaling ($\langle v^2\rangle \sim 1/q$) obtained for the square symmetry seems to be singular: all other cases provide $n = \frac{1}{2}$.

The computation of the coarsening exponent for conserved models requires estimation of $I_{\mathrm{C}} = \langle v^2\rangle/\langle wv\rangle$. Since $\nabla^2 w = v$, we infer (after integration by parts) $I_{\mathrm{C}} \sim \langle v^2\rangle/\langle u_0^2\rangle \sim q$. Therefore, for the standard Cahn-Hilliard model, we get $n = \frac{1}{4}$ for square symmetry and $n = \frac{1}{3}$ for other symmetries. Table I summarizes the results. Note that standard GL and CH refer to $A(u) = u - u^3$ so that $\langle v^2\rangle \sim q^{-1}$. In contrast, modified GL equation means $\langle v^2\rangle \sim q^{-\alpha}$, with $\alpha > 1$.

VI. DISCUSSION

We have found that the 2D coarsening exponent for the standard GL and CH equations is the expected one ($n = \frac{1}{2}$ and $n = \frac{1}{3}$, respectively) for all symmetries but the square one, which exhibits a slower coarsening. The peculiar behavior of square symmetry is at present not understood. It must be noted, however, that if $\langle v^2\rangle \sim q^{-\alpha}$ ($\alpha > 1$, as occurs with the modified GL) then $n = \frac{1}{2}$ for any symmetry. It seems thus that the peculiar behavior of square symmetry is quite specific to the considered equation rather than general. Nonetheless, a deeper understanding of this fact merits higher attention in the future.

It is worth stressing once again that our approach allows us to reduce the study of dynamical properties (coarsening) to the behavior of quantities (D_{11}, D_{12}) depending on steady-state properties only. Our analysis has been, for ease of presentation, exemplified on the two classical equations,

namely, the GL and CH ones, but the method can be applied to other equations. In particular, our approach (as it is evident in Ref. [17]) does not require that the equation is derivable from a potential.

While for the GL and CH equations we could extract analytically the coarsening exponent, it may prove necessary that for other equations there would be a need for numerical solutions of the steady-state problem in order to evaluate the diffusion constants. This is a quite simple task numerically even at higher dimensions (where full time-dependent studies are difficult or even unfeasible if the asymptotic regime is to be ascertained).

The amplitude branch is stable, in contrast to the phase one when coarsening is to take place. There are five different branches corresponding to different symmetries (five Bravais lattices). The coarsening of the square branch is slower (only for the classical GL and CH equations) than that of the other symmetries. It is natural to expect the fastest growing structure to prevail. Symmetries other than the square one offer, for these classical equations, a faster channel toward coarsening.

A particularly important question concerns relevance of slower coarsening peculiar to square symmetry. A possibility might be offered [19] by the Rayleigh-Bénard convection, which may develop either rolls or hexagons, with different local symmetries in different spatial regions. These regions coarsen in time and the coarsening rate might be different for domains of rolls and hexagons.

We have investigated steady-state *periodic solutions*, while during coarsening, even in 1D, no long-range order is observed. The questions thus arises about importance of periodic solutions. Locally in space the amplitude is adiabatically slaved to the phase so that (for a given wave vector) it reaches the quasisteady solution. If steady-state solutions were stable (no coarsening), then the system would generically choose the periodic solution (possibly with defects), as is known for many pattern forming systems. Thus, having shown here that the periodic solutions are unstable with respect to phase fluctuations, we expect that the pattern should coarsen. One cannot exclude, however, the existence of nonperiodic (such as disordered) stable solutions for nonlinear extended systems with an average wavelength which does not grow in time. We are not aware of any such scenario, however.

Finally, it is noteworthy that the phase diffusion equations share the same structures as those for the displacement field of 2D crystals [20] (see also the supplementary materials [21]). Conservation law imposes $\partial_t \phi_i = -\partial_k J_{ik}$ (this is the analog of the dynamical equation in continuum media) where J_{ik} is the phase current. J_{ik} is proportional to the gradient of the phase, $\partial_l \phi_m$, and the proportionality coefficient is a rank four tensor, $J_{ik} = \Lambda_{kilm} \partial_l \phi_m$ (this is the analog of Hooke's law). Λ should be invariant under the symmetry group of the considered crystal. The phase equations has thus the same structure as the elasticity problem of crystals.

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