

Instabilities and pattern formation in driven diffusive systems

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The instabilities of a phase boundary are investigated in a system corresponding to the two-dimensional attractive lattice-gas model driven by a uniform external field. On the analogy of Mullins-Sekerka instability the planar interface becomes unstable due to the enhanced material transport along the interface. This phenomenon explains why the particles are condensed on strips parallel to the driving field at low temperatures.

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In driven diffusive systems the ordering process is affected by the application of a uniform electric field (for a review see the paper by Schmittmann [1]). In such nonequilibrium systems one can observe inhomogeneous material transport related to the density fluctuations or to the presence of an interface separating two phases (or domains). The inhomogeneous material transport may result in pattern formation.

In this paper we investigate the effect of enhanced material transport along the interface on the pattern formation. The stability of the interface parallel to the external field has been shown by several authors [2,3]. Very recently Leung [4] and Yeung *et al.* [5] have shown that the external field leads to an instability of those interfaces which are not parallel to the field. This phenomenon is analogous to the Mullins-Sekerka instability which starts the pattern-forming process in solidification [6,7]. Yeung *et al.* [5] have derived the interface description from the Cahn-Hilliard equation and found an instability mechanism due to the enhanced surface current. Now, a much simpler formalism is presented which allows us to study the late stage of pattern formation. Its consequences underline the importance of interfacial phenomena when analyzing the ordering process in driven systems [8].

For simplicity we concentrate on a two-dimensional system corresponding to a lattice-gas model with attractive nearest-neighbor interaction. In equilibrium the particles are condensed below a critical temperature T_c . In a finite box with periodic boundary conditions the condensed phase may form a strip or droplet region depending on the average concentration [9]. The effect of a strong driving field on this system has been studied by Katz *et al.* [10]. Using Monte Carlo simulations these authors found that the particles segregate into strip regions with boundaries parallel to the driving field at low temperatures. Since then, different approaches (Monte Carlo simulations, dynamic mean-field theory, field-theoretic renormalization-group analysis, etc.) have been developed to study this driven system [1,10–14].

In this work our attention is focused on the interface where the dominant part of the particle transport is localized at low temperatures. This fact may be easily visualized in Monte Carlo simulations when displaying the time evolution of particle distribution. In this description the thickness of the boundary layer is neglected. Furthermore, the conductivity of this layer is assumed

to be independent of its direction. The position of the interface at a time t is given by the expression

$$y = \zeta(x, t). \quad (1)$$

This interface separates the condensed phase [$y < \zeta(x, t)$] from the empty region [$y > \zeta(x, t)$]. Neglecting the material transport in the bulk the interface motion is controlled by the particle current $j(x, t)$ along the interface. Thus, the interface motion is determined by the particle conservation,

$$c_0 \partial_t \zeta = -\partial_x j(x, t), \quad (2)$$

where c_0 is the concentration difference between the two phases, and ∂_t and ∂_x denote the partial derivatives with respect to time and the x coordinate. It is worth mentioning that negative c_0 refers to a situation where the condensed phase is positioned above the interface. The current $j(x, t)$ is assumed to be proportional to the gradient of the chemical potential $\mu(x, t)$ along the interface. More precisely,

$$j(x, t) = -\sigma \frac{d\mu}{ds} = \frac{-\sigma \partial_x \mu}{[1 + (\partial_x \zeta)^2]^{1/2}}, \quad (3)$$

where s refers to the derivation with respect to arc length along the curve $\zeta(x, t)$. The coefficient σ characterizes the (isotropic) interfacial conductivity.

In the present simplified description the chemical potential has two terms,

$$\mu(x, t) = -E\zeta + \mu_{\text{stab}} \quad (4)$$

where the first term takes the effect of the vertical driving field E into consideration. It is easy to see that the external field produces material transport toward the peak, leading to its growth. This phenomenon has been observed by Yeung *et al.* [5,15] in the numerical simulations of the corresponding Cahn-Hilliard equation. On the analogy of capillarity during solidification [6,7], a second term is introduced to stabilize the smooth interface. Now we choose μ_{stab} to be proportional to the interface curvature, viz.,

$$\mu_{\text{stab}} = -\eta \frac{\partial_{xx}^2 \zeta}{[1 + (\partial_x \zeta)^2]^{3/2}}, \quad (5)$$

where η characterizes the strength of the stabilizing force. Henceforth the curvature is understood to be negative if the center of curvature lies on the condensed phase side of the interface. This simple stabilizing force induces material transport from the peak (negative curvature) towards the valley (positive curvature), and this process will smooth the curve if $\eta > 0$.

After some mathematical manipulations one can obtain the following partial differential equation which describes the time evolution of the interface in the presence of a driving field:

$$\partial_t \zeta = -\frac{\sigma}{c_0} \partial_x \frac{1}{[1 + (\partial_x \zeta)^2]^{1/2}} \partial_x \left[E \zeta + \eta \frac{\partial_{xx}^2 \zeta}{[1 + (\partial_x \zeta)^2]^{3/2}} \right]. \quad (6)$$

Similar equations of motion have recently been derived to describe deposition processes in which the surface diffusion constitutes the dominant relaxation mechanism [16–18]. For example, in the continuity equation introduced by Villain [16] an evaporation rate dependent on the shape of surface is substituted for the term proportional to E in Eq. (6). Thus, Eq. (6) may be considered as an isotropic (curvature-dependent) description of this deposition process. This analogy may be very fruitful in future analysis of the effect of noise [16–19].

Equation (6) has a trivial solution

$$\zeta_0(x, t) = y_0 + mx \quad (7)$$

corresponding to a standing planar interface with a slope m .

To examine the linear stability of the planar interface, we look for a solution in the form

$$\zeta(x, t) = \zeta_0 + \delta e^{\lambda t + i k x}, \quad (8)$$

where δ and k are the amplitude and wave number of the periodic perturbation. In the limit $\delta \rightarrow 0$, Eq. (6) may be linearized and the corresponding equation gives an expression for the amplification rate λ :

$$\lambda = \frac{\sigma}{c_0} \left[\frac{E}{(1 + m^2)^{3/2}} - \frac{\eta}{(1 + m^2)^2} k^2 \right] k^2. \quad (9)$$

The m dependence in the second term of Eq. (9) is a consequence of the reduction of wave number for finite slope. This inconvenience may be avoided by introducing the real wave number measured along the interface: $q = k/(1 + m^2)^{1/2}$. The amplification rate may be expressed by q ,

$$\lambda = \frac{\sigma}{c_0} \left[\frac{E}{(1 + m^2)^{1/2}} - \eta q^2 \right] q^2. \quad (10)$$

This result demonstrates that the planar interface is unstable against infinitesimal fluctuations at all wave numbers less than q_0 where $q_0^2 = E(1 + m^2)^{-1/2}/\eta$. The amplification rate λ has a maximum [$\lambda_{\max} = \sigma E^2/4(1 + m^2)c_0\eta$] for $q = q_{\max} = q_0/\sqrt{2}$. It seems reasonable to guess that the periodicity of the pattern emerging initially from this instability may be characterized by $q_{\max}$. Note that the short-wavelength perturbations are sup-

pressed by the stabilizing term. At the same time the planar interface becomes stable when reversing the external field ($E < 0$). These results are in close agreement with the linear stability analysis of the corresponding Cahn-Hilliard equation performed by Yeung *et al.* [5] for interfaces normal to E . Similar instabilities have been found in deposition processes [16–19].

The present description suggests that the destabilization vanishes in the limit $m \rightarrow 0$. This prediction agrees with the results of Monte Carlo simulations [10,14] carried out in a two-dimensional lattice-gas model with attractive interaction. The Monte Carlo simulations are performed under the influence of very large external fields with periodic boundary conditions. It is observed that the particles are condensed into strips parallel to the field below a critical temperature. These strips “wrap around” the system; therefore the interfacial instability cannot affect this state. This might be the reason why the multistrip states are found to be stable for a long time. Here it is worth mentioning that the multistrip state is expected to approach the single-strip state according to the theory of gambler’s ruin [20], because this process is analogous to a particular situation where each gambler (domain) can win or lose a certain sum (area) from his adversaries.

The stability of the interfaces parallel to the external field has already been clarified by several authors [2,3,5]. It is easy to see that the nonlinear terms of Eq. (6) become essential if the interface is parallel to the applied field. Analysis of the nonlinear behavior, however, is beyond the scope of the present paper.

One can easily check that Eq. (6) has a stationary solution traveling with a constant velocity v , namely,

$$\zeta = \pm(vt + \sqrt{r^2 - x^2}). \quad (11)$$

These solutions are not affected by the stabilizing term. From this type of solution one can construct a circular domain moving along the field, and its velocity is determined by the radius r and the maximum current at the periphery, that is,

$$v = E\sigma/c_0 r. \quad (12)$$

In agreement with the particle-hole symmetry in lattice-gas models, there exists another solution corresponding to a circular bubble in the condensed phase moving against the field. For $\eta = 0$ the semicircular solutions [Eq. (11)] may be connected to each other by inserting a rectangular region with sides parallel to the external field; the velocity of the resulting pattern is determined by the tip radius.

Under the same conditions ($\eta = 0$), one can construct a finger-like solution (the sides are parallel to the applied field) with a tip of constant radius whose growth rate along the field is equivalent to the velocity of the corresponding circular domain. This moving pattern is an analogy of the Ivantsov solution in the area of solidification [21,7]. Based on a brief numerical analysis of Eq. (6) the stabilizing term ($\eta \neq 0$) smooths the sudden change of the curvature. Evidently, an anti-finger-like solution may also be constructed whose bottom travels against

the field. The composite of the finger- and anti-finger-like solutions may be considered as a late stage of the time evolution initiated by the above-mentioned instability. The finger growth process continuously produces interfaces parallel to the driving field. Furthermore, two condensed regions separated by walls parallel to the driving field may be connected by a traveling "bridge" with circular profiles corresponding to Eq. (11). The motion of such "bridges" (as well as the finger growth and drop motion) have already been observed by Yeung *et al.* [15].

The above processes split the large domains into strips with a constant velocity [see Eq. (12)]. It is expected that the formation of large (isotropic) domains is prevented in driven systems because the domains grow with a decreasing velocity during the ordering process. The competition between the traditional domain growth and interfacial instability has been observed in the driven half-filled lattice-gas model with repulsive nearest-neighbor interaction which exhibits an anisotropic, stationary, polydomain structure at low temperatures [22]. As a consequence, the singularity of specific heat is suppressed. In this system the particle motion is also dominant at the interface and the more complicated mechanism results in a faster interface evolution. In these simulations the system sizes are chosen to be much larger than the corresponding domain sizes. By this means we could avoid the formation of closed strips which switches the interfacial instability off. The present work, in fact, was inspired by the hypothesis that similar phenomena may appear in driven lattice gases with attractive interaction(s).

The dynamics of phase separation in the presence of a field has already been studied with simulations of the Cahn-Hilliard equation for driven diffusive systems [15,23]. Neglecting the thermal fluctuations the authors have observed the formation of extremely anisotropic domain structure from a random initial state. The role of thermal fluctuations, however, becomes significant in the vicinity of T_c because the lengths of striplike domains are strongly affected by wall fluctuations. This phenomenon may be observed in some snapshots (see, e.g., Fig. 1 in Ref. [14]) displaying the particle distributions in Monte Carlo simulations in large systems. Unfortunately, the Monte Carlo simulations in driven lattice gases are concentrated at the large-field limit and systematic studies are carried out in small systems ($L < 100$). In this case the interface thickness becomes large and particle transport is saturated; consequently, the field dependence is not significant. Furthermore, the defect density is increased by the driving field and this process results in higher transport in the bulk phase. In the light of the present results it is worth performing Monte Carlo simulations for large systems in the low-field limit.

The instability as well as the ensuing finger growth process are not restricted to two-dimensional driven systems. Similar phenomena are expected for higher dimensions. The above analysis is not yet complete; our attention has focused on the interfacial material transport which provides a driving force for the pattern formation. For this purpose we have neglected some phenomena (e.g., anisotropic features, bulk conduction and diffusion, fluctuations, saturation in transport, nonlinearity, etc.) which limit the validity of the above results. The main conclusions, however, seem to be valid, though further analysis is required to understand the behavior of driven diffusive systems.

In summary, the effect of interfacial material transport on pattern formation is studied in those systems where the particles segregate into a condensed phase under the influence of an external field. A simple model is introduced to describe the time evolution of the interfaces. The planar interface is found to be unstable against infinitesimal fluctuation in the long-wavelength region in agreement with the results of Yeung *et al.* [5]. This phenomenon is analogous to the Mullins-Sekerka instability. Here the instability is driven by the interfacial material transport induced by the external field. The present analysis is restricted to the low-field limit and it suggests that the wavelength of the emerging fluctuations is proportional to $1/\sqrt{E}$. On the analogy of the Ivantsov solution, we have found a circular solution traveling with a constant velocity. This solution explains the motion of droplets, bubbles, and "bridges" as well as the finger growth process observed in numerical simulations. From the analytical calculations we can conclude that the increasing periodic perturbations evolve into a finger growth process which splits the large domains into strips parallel to the external field. Consequently, there is a competition between the traditional domain growth mechanism and the interfacial instability. It is an exciting question whether the interfacial instability prevents the formation of a monodomain state in (sufficiently large) driven lattice gases with attractive interaction(s). The present results support the stability of an extremely anisotropic, "self-organizing," polydomain state in driven systems satisfying the above conditions.

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