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Stability of a nonequilibrium steady-state interface

A. Hernández-Machado* and David Jasnow
Department of Physics and Astronomy, University of Pittsburgh, Pittsburgh, Pennsylvania 15260
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We study the interfacial modes of a driven diffusive model under suitable nonequilibrium conditions leading to possible instability. The external field parallel to the interface, which sets up a steady-state parallel flux, enhances the growth or decay rates of the interfacial modes. More dramatically, asymmetry in the model can introduce an oscillatory component into the interfacial dispersion relation. In certain circumstances, the applied field behaves as a singular perturbation.

Phase transitions under a variety of nonequilibrium conditions have been receiving attention, and this promises to be a valuable study. Recently a series of computer simulations[1-3] have characterized the bulk features of the nonequilibrium phase transition of a lattice-gas model of interacting particles subject to an external field, which drives a steady particle current. Theoretical discussions have involved both discrete[2,4-6] and continuous models.[7-11] The computer simulations indicate[1-3,12] the existence of a sharp phase transition at a temperature which depends on the strength of the external field, below which the system undergoes phase separation into two phases (particle rich and poor). The external field is responsible for the anisotropic striplike configuration of the system; the interface separating the two phases lies *parallel* to the external field, and a constant particle flux parallel to the interface is carried by each of the phases.

The simulations and the related work noted above lie within the general context of nonequilibrium steady-state systems; in the present case they are also referred to as driven diffusive systems. The possibility of using such systems to model superionic conductors has been widely explored in the literature.[13] In a different context there is theoretical[14] and experimental[15] work on critical fluids under shear flow.

A study of the stability of the two-phase interface present in the simulations[1-3] will be the subject of this short paper.[16] Such a study is an essential ingredient in understanding patterns which might be produced under a variety of nonequilibrium conditions. Considerable progress has been made in problems of pattern formation,[17-20] and some insights derived from such studies might be applicable to the present class of driven systems. In a reverse sense, the systems discussed here are particularly simple in description, and there may be implications of the study of such systems for other pattern forming processes.

The system of interest is easily described. One considers an Ising lattice gas with equal numbers of particles and holes. The dynamics are particle conserving, and an applied field **E** is oriented parallel to the x axis. Periodic boundary conditions are assumed, particularly in the x direction. In the simulations,[1-3] relative to a nearest-neighbor particle-hole exchange along a bond perpendicular to **E**, the applied field favors a particle-hole exchange in which the particle moves along **E**, and inhibits particle motion opposite **E**. The situation in the ordered phase is shown schematically in Fig. 1. We have generalized this model in two ways. First, we note that the applied field **E** does not of itself induce unstable behavior. To study the effect of the applied field **E** on an interfacial instability, we consider the interface driven by a particle flux *perpendicular* to the interface. This flux of particles (from the particle poor to the rich phase) may be imagined as resulting from a rapid quench deeper into the ordered phase.[21] The model without **E** is just the so-called symmetric model,[22,23] which has an interfacial instability for sufficiently long wavelength for any nonvanishing perpendicular flux. Such instability, which contains a Mullins-Sekerka-type instability[18] in perhaps its purest form, lies at the heart of a variety of interfacial growth processes. The present work considers the effect of the applied field **E** on the well-studied features of the symmetric model. As a second generalization we consider the effect of asymmetry.

The most noticeable effect introduced by the external field **E** is the appearance of an oscillatory component in the growth or decay of interfacial modes when asymmetry is present. On the other hand, the external field **E** does not modify the critical wave number for which a particu-

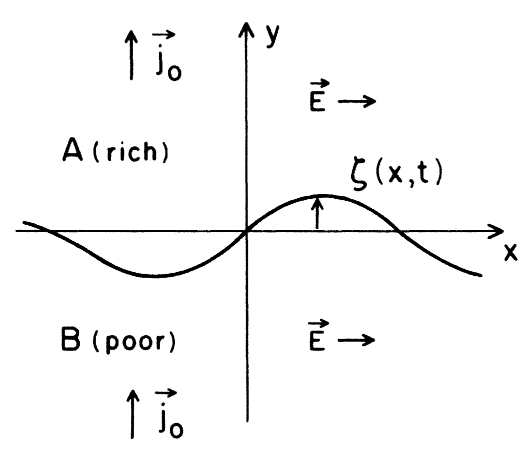

FIG. 1. Schematic illustration indicating the phase separated system under the influence of the external field **E** and the perpendicular current $\mathbf{j}_0$. The flat interface lies in the plane $y=0$; a small amplitude distortion is indicated.

lar mode is stable or unstable, but one finds that the growth or decay rates are increased by the presence of the applied field. The analytic dependences on **E** are interesting; the applied field may have to be considered a singular perturbation (see below).

The continuum model we use to study the linear stability of the interface has been used extensively in studies of the bulk properties.[7-11] It corresponds to a generalization of the usual time-dependent Ginzburg-Landau description of a system with concentration as the conserved order parameter [model B (Ref. 24)]. A new current proportional to the external field is added, and the coupling to the order parameter is produced by a concentration-dependent conductivity. This term has been introduced on a phenomenological basis,[7] and it brings up an interesting conceptual point. For such a system with periodic boundary conditions, as noted by Katz *et al.*,[2] the external field cannot be included in the Hamiltonian but rather enters the transition probabilities. From this conceptual viewpoint the situation is slightly different from the inclusion of a field gradient (say, gravitational) in a closed system with walls. The continuum equations of motion can be derived from the cell-model description of Oono and Puri[25] or, one expects, from the full master equation describing the discrete lattice model with conserving dynamics (i.e., Kawasaki exchange[26]).

We are interested here in the simplest *macroscopic* description, which means that we consider a sharp interface compared to the length scale of any interfacial undulations, and we neglect effects of noise. The nonlinearities involving the interface are introduced in the standard fashion via thermodynamic boundary conditions. In this approach, the equation for the concentration field is

$$\partial_t c_a = -\nabla \cdot \mathbf{j}_a \ , \tag{1}$$

$$\mathbf{j}_a = -D(\nabla c_a) + \mathbf{E}\sigma(c_a) \ , \tag{2}$$

where $c_a(\mathbf{r},t)$ and $\mathbf{j}_a(\mathbf{r},t)$ are the concentration and current in the particle rich and poor phases (A and B, respectively), D is the diffusion coefficient (which we take equal in both phases), $\mathbf{E} = E\hat{\mathbf{x}}$ is the applied field, and $\sigma(c)$ is the conductivity.[27] We consider effects of the external field on the stability of the flat interface, taken to be the plane $y=0$. The configuration is shown in Fig. 1.

The flat, translationally invariant (in the x direction) stationary solution of Eqs. (1) and (2) near the interface takes the form

$$c_{a0}(y) = c_{a0} - \frac{j_0}{D} y \ , \tag{3}$$

where c_{a0} is the equilibrium (or steady state) concentration for the phase $a = A$ or B. We have assumed a constant flux $j_0\hat{\mathbf{y}}$ driving the interface, imagined induced, as discussed in Ref. 21, by quenching the system deeper into the ordered state. On the other hand, there is a net flux of particles in the x direction due to the external field. The total stationary flux is then

$$\mathbf{j}_{a0}(\mathbf{r}) = j_0\hat{\mathbf{y}} + \mathbf{E}\sigma(c_{a0}(y)) \ . \tag{4}$$

A perturbation of the interface of the form

$$y_{\text{interface}} = \zeta(x,t) = \zeta \exp(ikx + \omega t) \ , \tag{5}$$

is introduced in the standard fashion; the amplitude ζ is the small parameter in the linearization. The sign of $\mathrm{Re}(\omega)$ will determine the stability of the perturbation of wave number k. One searches for a solution of the form

$$c_a(\mathbf{r},t) = c_{a0}(y) + c_{a1}(\mathbf{r},t) \ , \tag{6}$$

where the second term is the correction, which in a linear analysis, is of order ζ. In the linear regime the equation for $c_{a1}(\mathbf{r},t)$ is

$$\partial_t c_{a1} = D(\nabla^2 c_{a1} - Q_a \partial_x c_{a1}) \ , \tag{7}$$

where $Q_a = E(d\sigma/dc)_a/D$. We have assumed an expansion of the form $\sigma(c_a(\mathbf{r},t)) \approx \sigma(c_{a0}(y)) + (d\sigma/dc)_a \times c_{a1}(\mathbf{r},t)$, and have evaluated the derivative at c_{a0} (valid for small j_0). Note that the (inverse length) parameters Q_a explicitly depend on the phase $a = A,B$. Strictly speaking, within a symmetric Ising model one should take $Q_A = -Q_B$, but we allow further generality for more realistic systems or those operating some distance below the phase separation temperature.[28] One seeks a solution of the form

$$c_{a1}(\mathbf{r},t) = A_a \exp(ikx \mp q_a y + \omega t) \ , \tag{8}$$

where, as noted, A_a is $O(\zeta)$. In Eq. (8) the minus and plus signs correspond to the A and B phases, respectively, so the solution must have $\mathrm{Re}(q_a) > 0$. For simplicity, in the following discussion we consider $k > 0$. To determine the constants A_a, q_a, and the dispersion relation for ω we need additional conditions. The first is the Gibbs-Thomson relation, which is a statement of local equilibrium and introduces the effect of capillarity, namely,

$$c_a(\mathbf{r},t)\,|_{\text{interface}} - c_{a0} = -\Gamma K \ , \tag{9}$$

where Γ is proportional to the surface tension (but generally also involves an appropriate thermodynamic derivative[23]). K is the curvature of the interface, which for small displacements is just

$$K \approx -\frac{\partial^2}{\partial x^2}\zeta(x,t) \ . \tag{10}$$

The second condition is provided by the continuity equation

$$(\Delta c)V_n = \hat{\mathbf{n}} \cdot (\mathbf{j}_A - \mathbf{j}_B) \ , \tag{11}$$

which yields the motion of the interface due to a flux imbalance. Here $\hat{\mathbf{n}}$ is the normal directed toward the A phase, v_n is the normal velocity, and Δc is the equilibrium miscibility gap. Straightforward algebraic manipulations yield in first order the relations

$$A \equiv A_a = \left[\frac{j_0}{D} - \Gamma k^2\right]\zeta \ , \tag{12}$$

$$(\Delta c)\omega\zeta = DA(q_A + q_B) \ , \tag{13}$$

$$\omega = D(-k^2 + q_a^2 - iQ_a k) \ . \tag{14}$$

The first two equations are quite standard for the symmetric model, and at this order the external field (**E**) enters only the last equation. In this quasistatic approximation, one neglects ω in Eq. (14); for simplicity we make this approximation recognizing that there will be differences in detail, for example, at extremely long wavelengths. The external field changes the values of q_a and the dispersion relation; in general q_a and ω are complex. We will return to this point below.

The equations determining the real and imaginary parts of $q_a = q_{a1} + iq_{a2}$ become

$$q_{a1}^2 - q_{a2}^2 = k^2 \ , \tag{15a}$$

$$2q_{a1}q_{a2} = Q_a k \ . \tag{15b}$$

As noted above, the real part of q_a must be positive. The particle-rich (A) phase is expected to have $Q_A < 0$, and the poor (B) phase is expected to have the opposite sign. Hence, it is expected that the imaginary parts q_{A2} and q_{B2} carry opposite sign with $q_{A2} < 0$. The unique solution is

$$q_{a1} = \frac{k}{\sqrt{2}} \left\{ 1 + \left[1 + \left(\frac{Q_a}{k} \right)^2 \right]^{1/2} \right\}^{1/2} \equiv \frac{k}{\sqrt{2}} \bar{q}_{a1} \ , \tag{16a}$$

$$q_{a2} = \mp \frac{k}{\sqrt{2}} \left\{ 1 + \left[1 + \left(\frac{Q_a}{k} \right)^2 \right]^{1/2} \right\}^{1/2} \equiv \frac{k}{\sqrt{2}} \bar{q}_{a2} \ . \tag{16b}$$

The upper (lower) sign corresponds to the A (B) phase. Finally the dispersion relation becomes from Eq. (13)

$$(\Delta c)\omega = (j_0 - \Gamma D k^2) \frac{k}{\sqrt{2}} \sum_a (\bar{q}_{a1} + i\bar{q}_{a2}) \ . \tag{17}$$

This is the central result of this paper, and some comments are in order at this point.

When the external field vanishes (i.e., $Q_a = 0$) we recover the usual dispersion relation for the symmetric model,[21-23] $(\Delta c)\omega = 2k(j_0 - \Gamma D k^2)$. The presence of the external field E changes the k dependence of the real part of ω, but it does not affect the critical wave number determined by $j_0 - \Gamma D k_c^2 = 0$. Since the imaginary parts q_{a2} are nonvanishing, the external field introduces oscillatory spatial decay of perturbations into the bulk material. A second and perhaps more interesting possibility is the nonvanishing imaginary part in the frequency ω. This means the usual decay or growth in the symmetric model is now modulated. Here a qualifying remark is necessary: Strictly within the framework of a symmetric lattice gas, one expects the two coexisting steady-state phases A and B to have equal conductivity. This makes $\sum_a Q_a = 0$, which implies $q_B = q_A^*$, making the imaginary, oscillatory part of ω vanish. Generally speaking some asymmetry in σ (as well as in the coexistence curve) is expected away from the phase separation temperature, and this makes itself felt in an oscillatory component. In most of what follows we assume the general situation.[29]

Another aspect, noted above, is that the external field introduces a new length scale into the problem, which we can render dimensionless through the ratio Q_a/k. In the limit of small field $Q_a/k \ll 1$, the dispersion relation becomes

$$(\Delta c)\omega = 2k(j_0 - \Gamma D k^2) \times \left[1 + \frac{1}{16} \frac{Q_A^2 + Q_B^2}{k^2} + \frac{i}{4k}(Q_A + Q_B) \right] \ . \tag{18}$$

One sees that the decay ($k > k_c$) or growth ($k < k_c$) rate is increased. For a strictly symmetric lattice gas with ordered phases having equal conductivities, the imaginary part vanishes, and the correction to the usual symmetric model dispersion relation is $O(E^2)$. Attempting to find the dispersion relation (for $j_0 = 0$) from a Ginzburg-Landau equation, say following the perturbative approach of Jasnow and Zia,[30] is extremely difficult.[31] However, in the nonsymmetric case, $Q_A + Q_B \neq 0$, the imaginary part, being $O(E)$, can be analyzed using such methods.

In the large field limit $Q_a/k \gg 1$, one finds

$$(\Delta c)\omega = (j_0 - \Gamma D k^2) \left(\frac{k}{2} \right)^{1/2} \times [\,|Q_A|^{1/2} + |Q_B|^{1/2} + i(|Q_B|^{1/2} - |Q_A|^{1/2})] \ . \tag{19}$$

A new length scale is, as noted, introduced by $E \neq 0$. From Eqs. (18) and (19) one sees that for *fixed* wavelength, sufficiently small E may be treated perturbatively. On the other hand, for *fixed E*, its effect at sufficiently long wavelength is not perturbative. So note, that when the perpendicular driving flux is removed (i.e., setting $j_0 = 0$), there is no instability, but relaxation to the steady state has $\mathrm{Re}(\omega) \sim -k^{5/2}$ as compared to the familiar $(-k^3)$ behavior. In the weak-field case the relaxation preserves the leading $(-k^3)$ dependence as seen in Eq. (18).

In this short paper we have performed a linear stability analysis on the interfacial modes of a driven diffusive model with interface established in steady-state conditions *parallel* to the applied field E. As an additional feature we have imagined the interface subjected to a steady *perpendicular* flux $\mathbf{j}_0$ produced, say, by a rapid quench deeper into the ordered region. At sufficiently long enough wavelengths the interface is driven unstable by $\mathbf{j}_0$. We find that the parallel applied field modifies the growth or decay of the interfacial modes in two essential ways. First, it introduces the possibility of an oscillatory component when the system is not completely symmetric. Second, the applied field introduces a new length scale into the problem, which causes the applied field to behave like a singular perturbation modifying the leading k dependence of the interfacial growth or decay rate.

Although the applied field of the driven diffusive model of Katz, Lebowitz, and Spohn[2] is introduced in a conceptually different fashion than a usual field gradient (say, gravitational) entering the normal Ginzburg-Landau Hamiltonian, away from the boundaries and for short times the effects of the two types of fields is the same. Hence, it is possible that a simple kinetic model, such as the one studied here, can also be of use in problems of crystal growth in anisotropic situations and in the presence of suitably oriented external fields.

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*Permanent address: Departamento de Estructura y Constituyentes de la Materia, Universidad de Barcelona, Av. Diagonal, 647, E-08028, Barcelona, Spain.

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[27]We imagine the scales are such that a local conductivity is sensible.

[28]Since we are considering a two-phase system, the linear terms in Eq. (7) cannot be removed by a coordinate transformation.

[29]To be consistent one should consider allowing slight asymmetry in the diffusion coefficient D, but this does not of itself introduce an oscillatory component into the symmetric model ($E=0$). No substantial changes are introduced into our analysis if the diffusion coefficients differ in the two phases.

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[31]It is inherently a second-order calculation about the ϕ^4 interface. Hence, all eigenvalues and eigenfunctions of the ϕ^4 fluctuation operator enter the calculation. We thank R. K. P. Zia for informing us of some progress in this direction.