ARTICLES

Stability of a nonequilibrium interface in a driven phase-segregating system

Claude A. Laberge and Sven Sandow
Department of Physics and Center for Stochastic Processes in Science and Engineering, Virginia Polytechnic Institute and State University, Blacksburg, Virginia 24061-0435
(Received 19 May 1997)

We investigate the dynamics of a nonequilibrium interface between coexisting phases in a system described by a Cahn-Hilliard equation with an additional driving term. By means of a matched asymptotic expansion, we derive equations for the interface motion. A linear stability analysis of these equations results in a condition for the stability of a flat interface. We find that the stability properties of a flat interface depend on the structure of the driving term in the original equation. [S1063-651X(97)01212-9]

PACS number(s): 64.75.+g, 64.70.Ja, 68.10.−m, 47.54.+r

I. INTRODUCTION

Off-equilibrium systems composed of regions with different phases can exhibit a variety of patterns, such as fingers aligned in a certain direction. A familiar example is spinodal decomposition of binary mixtures (for a review see [1,2]). When quenched below its critical temperature, an initially homogeneous system develops domains of the new equilibrium phases. After some time these domains are well defined, i.e., they are separated by sharp interfaces. The system further approaches equilibrium by means of the motion of these interfaces; the initial pattern evolves in the course of time. Another example is the dynamics of driven diffusive systems (for a review see Ref. [3]). Interacting particles undergoing biased diffusion tend to form clusters under certain conditions. Like for the first example there are well-defined interfaces, which undergo some dynamics. In all of these phase segregating systems patterns are selected according to their stability. For example, the instability of a flat interface causes the growth of fingers out of an initially flat interface. For this reason one would like to understand the stability properties of nonequilibrium interfaces.

On a course-grained level these phase-segregating systems are described by means of an order parameter, such as a particle density. Typically, the dynamics of this order parameter can be modeled by means of some nonlinear differential equation. One of the simplest such equations is the Cahn-Hilliard (CH) equation, which was introduced in the context of binary mixtures [4]. The properties of this equation are rather well understood. In particular, the dynamics of the domain boundaries obey a set of linear differential equations, which have been derived by Pego [5] using a matched asymptotic expansion. An instability of the Mullins-Sekerka type has been observed.

Certain modifications of the CH equation were introduced to take into account the effect of external fields, [6–10,5,11,12] (see Refs. [13,14,3] for modified CH equations in the context of driven diffusive systems). One of the questions one would like to answer is the following: How does an external field change the stability properties of a flat interface? This question was addressed by Yeung *et al.* [11] for a certain type of driving term. This driving term has the form of an additional current that is proportional to the field and a conductivity. The conductivity was assumed to be a concave second-order function of the order parameter with the same symmetry as the chemical potential. Yeung *et al.* [11] found that the field modifies the interface dynamics. An interface perpendicular to the external field is stable for one direction of the field and unstable for the other direction.

In this paper we generalize the work of Yeung *et al.* [11]. Like Yeung *et al.* we consider a modified CH equation with an isotropic chemical potential. (Motivating the modified CH equation with a free-energy argument, an isotropic chemical potential is expected for a small enough field. In this paper, we do not address the question of how good an approximation this is, but rather assume that the equation is reasonable for certain physical systems and start our discussion from there.) The driving term in our equation is proportional to the field and an arbitrary conductivity. Our approach is the same as the one from Ref. [11] (see also Ref. [5]). Assuming that the driving field is sufficiently small we perform a matched asymptotic expansion. The resulting set of linear equations describes the motion of an interface between coexisting phases. With the help of these equations we analyze the stability of a flat interface. We find a general stability criterion, which takes a very simple form if we assume that the conductivity is a third-order polynomial in the order parameter. For such a conductivity we show that an interface perpendicular to the external field can be unstable for either direction of the field and that an interface parallel to the external field is always stable.

The paper is organized as follows. In Sec. II we define the model. A matched asymptotic expansion is done in Sec. III. Equations for the motion of an interface are derived. In Sec. IV we perform a linear stability analysis for a flat interface resulting in a condition for the stability of such an interface. This condition is subsequently shown to take a simple form for the case of a conductivity that is a third-order polynomial in the order parameter. Section V summarizes the results.

II. MODEL

We consider a two-dimensional system described by a conserved order parameter $u(\mathbf{R},T)$, where $\mathbf{R}$ denotes a position in space and T the time. The dynamic is assumed to be defined by the following modified Cahn-Hilliard equation [8–12,14]

$$\frac{\partial u}{\partial T} = -\boldsymbol{\nabla}\cdot\mathbf{J}, \tag{1}$$

$$\mathbf{J} = -\boldsymbol{\nabla}\mu + \mathbf{E}\sigma(u), \tag{2}$$

$$\mu = \lambda u\left(u - \frac{u_m}{2}\right)(u-u_m) - \xi^2\nabla^2 u. \tag{3}$$

In the above equations, $\mathbf{J}$ is the current of the order parameter, μ is the chemical potential of the free system, $\mathbf{E}$ is an external field (e.g., an electric field), and $\sigma(u)$ is the conductivity. One may think of the chemical potential as related by means of $\mu = \delta F/\delta u$ to a free-energy functional $F[u] = \int d\mathbf{R}\{f[u(\mathbf{R})] + \xi^2|\nabla u(\mathbf{R})|^2/2\}$, where $f(u)$ is a bulk free energy density the derivative of which is $f'(u) = \lambda u(u-u_m/2)(u-u_m)$. The current has two additive contributions: one that minimizes the free energy and another one defining the transport caused by the field. The first one is specified by the structure of the chemical potential. Assuming $\lambda > 0$ and $u_m > 0$, Eq. (3) defines a chemical potential such that the free energy has minima in regions with $u=0$ or with $u=u_m>0$. The term proportional to ξ^2 results in an increase of energy whenever there is a gradient in u, i.e., ξ^2 stands for a surface tension. The current caused by the field is $\mathbf{E}\sigma(u)$, which depends on the function $\sigma(u)$. The standard conductivity used in the literature [8–12,14] is a concave second-order function of u, which has the same symmetry as the chemical potential, i.e., is symmetric around $u_m/2$. Here we do not restrict ourselves to this type of function, but rather want to see what kind of behavior can be observed for a more general conductivity.

Equations of the type (1)–(3) are usually motivated on a purely phenomenological (see, e.g., [9–12,14]) or mean-field (see, e.g., [8]) level. The standard type of argument results in a mobility as a factor to the total current, i.e., a current of the type $\sigma(u)[-\boldsymbol{\nabla}\mu + \mathbf{E}]$. Assuming furthermore that the variation of $\sigma(u)$ is small enough, $\sigma(u)\boldsymbol{\nabla}\mu$ is then replaced by $\boldsymbol{\nabla}\mu$. We follow the same logic here. It is also worthwhile mentioning that a rigorous derivation of a more general type of equation has been done for a diffusive system with a long-range interaction and Kawasaki dynamics [13]. However, the relation of our equation to the standard driven diffusive system studied in Refs. [14,3] is not well understood. There, some interesting discrepancies between the Monte Carlo simulations and the mean field description have been observed [14].

Since it is more convenient to work with a dimensionless equation, we rescale Eqs. (1)–(3). Simultaneously, we add a constant term to the conductivity and apply a Galilean transformation such that the new current vanishes at both minima of the chemical potential. The adding of a constant does not change the equation for $u(\mathbf{R},T)$ since only the gradient of σ enters into this equation. The Galilean transformation is nothing but a change of the reference frame. We define a length L_0 and a time T_0 obeying

$$1 = L_0^{-2}T_0\lambda u_m^2 \tag{4}$$

as well as a velocity

$$V_g = |\mathbf{E}|u_m^{-1}\sigma(u_m). \tag{5}$$

Now we define dimensionless quantities as

$$\mathbf{r} = L_0^{-1}\mathbf{R} - L_0^{-1}V_g T\mathbf{e}, \tag{6}$$

$$t = T_0^{-1}T, \tag{7}$$

$$\rho = u_m^{-1}u, \tag{8}$$

where

$$\mathbf{e} = \frac{\mathbf{E}}{|\mathbf{E}|} \tag{9}$$

is the unit vector in the direction of the field. Inserting the above definitions into Eqs. (1)–(3) yields the dimensionless equations

$$\frac{\partial\rho}{\partial t} = -\nabla\cdot\mathbf{j}, \tag{10}$$

$$\mathbf{j} = -\nabla\nu + \mathbf{e}\beta(\rho), \tag{11}$$

$$\nu = \rho(\rho-1)(\rho-\tfrac{1}{2}) - \epsilon^2\nabla^2\rho, \tag{12}$$

with

$$\epsilon^2 = L_0^{-2}\xi^2\lambda^{-1}u_m^{-2} \tag{13}$$

and

$$\beta(\rho) = \epsilon^{-1}|\mathbf{E}|\xi\lambda^{-3/2}u_m^{-4}\{\sigma(\rho u_m) - \sigma(0) - \rho[\sigma(u_m) - \sigma(0)]\}. \tag{14}$$

In Eqs. (10)–(12) the operator $\boldsymbol{\nabla}$ stands for derivative with respect to the new coordinate $\mathbf{r}$. The conductivity $\beta(\rho)$ in the new frame has the property

$$\beta(1) = \beta(0) = 0, \tag{15}$$

i.e., it vanishes at both minima of ν. However, it is not necessarily positive in the whole interval $(0,1)$. In fact, later on we will identify a regime where $\beta(\rho) < 0$ for some part of $(0,1)$. For that reason it would be hard to give a direct physical meaning to Eqs. (10)–(14) in terms of the rescaled variables.

The velocity V_g given by Eq. (5) is positive by definition. Consequently, the new frame of reference moves in the direction of the field with respect to the original one.

III. DYNAMICS OF AN INTERFACE

The model defined in the preceding section describes phase segregation, i.e., the order parameter $\rho(\mathbf{r},t)$ evolves such that there are regions in space where $\rho(\mathbf{r},t)$ approxi-

mately takes the values for which the chemical potential is minimal. Since there are two minima of the chemical potential, there are two types of regions, which are separated by transition layers. The transition layer between the different regions, which is seen as a line on a large enough scale, evolves during time. We are going to give an analytical description of its motion for the case where

$$\epsilon \ll 1 \tag{16}$$

and for $0 \leqslant \rho \leqslant 1$,

$$\beta(\rho) = O(\epsilon^0), \quad \beta'(\rho) = O(\epsilon^0) \tag{17}$$

in the rescaled equations (10)–(14). In terms of the original parameters these conditions read

$$L_0^{-1} \xi \lambda^{-1/2} u_m^{-1} \ll 1, \tag{18}$$

$$|\mathbf{E}| \xi \lambda^{-3/2} u_m^{-4} \sigma_{max} \ll 1, \tag{19}$$

where σ_{max} denotes the largest values $\sigma(u)$ and its derivative $\sigma'(u)$ take in $[0,1]$. The first of these conditions can be fulfilled for any choice of the parameters simply by choosing a large enough length scale L_0. However, the second condition poses a restriction on the parameters; it can be understood as a small field condition.

Starting with the rescaled equations (10)–(12) we apply a matched asymptotic expansion. The basic idea is to expand the order parameter and its evolution equation in powers of the small parameter ϵ. We closely follow the line of Refs. [5] and [11]. Since we want to describe the motion of a single interface, we consider an initial configuration with two semi-infinite regions Ω^+ and Ω^- and assume that the order parameter is equal to 1 (0) in Ω^+ (Ω^-) up to corrections of order ϵ. The values 1 and 0 are the minima of the chemical potential. The regions Ω^+ and Ω^- are separated by a transition layer. We assume this layer to have a width of order ϵ like it in the field free case [5,11]. The characteristic time of its motion is

$$\tau = \epsilon t = \epsilon T_0^{-1} T. \tag{20}$$

In order to have a clear definition on any scale we define the interface Γ as the set of points where $\rho = \frac{1}{2}$. (Our results are not sensitive to the choice of the value 1/2.) We are going to expand our equations on the time scale τ first in the bulk of Ω^+ and Ω^- and then in the transition layer, i.e., near the interface Γ.

A. Equations far from the interface

Far from the interface Γ we define the new field

$$\rho^{\pm}(\mathbf{r}, \tau) = \rho(\mathbf{r}, \epsilon^{-1} t) \tag{21}$$

and similarly $\nu^{\pm}(\mathbf{r}, \tau)$ and $\beta^{\pm}(\mathbf{r}, \tau)$, where the superscript denotes the region we are considering. Equations (10) and (11) result in the following equations on the time scale τ:

$$\epsilon \partial_\tau \rho^{\pm}(\mathbf{r}, \tau) = \nabla^2 \nu^{\pm}(\mathbf{r}, \tau) - \mathbf{e} \cdot \nabla \beta^{\pm}(\mathbf{r}, \tau), \tag{22}$$

where $\nu^{\pm}$ and $\beta^{\pm}$ are defined by Eqs. (12) and (14), respectively. Next we expand $\rho^{\pm}(\mathbf{r}, \tau)$ in powers of ϵ as

$$\rho^{\pm}(\mathbf{r}, \tau) = \rho_0^{\pm} + \epsilon \rho_1^{\pm}(\mathbf{r}, \tau) + O(\epsilon^2) \tag{23}$$

where

$$\rho_0^+ = 1, \quad \rho_0^- = 0 \tag{24}$$

according to our previous assumptions. Inserting above expansion into Eqs. (22), (12), and (14) results in an equation for $\rho_1^{\pm}(\mathbf{r}, \tau)$ where the terms of zeroth order in ϵ cancel each other. Comparing terms of first order in ϵ yields the following relation:

$$[\nabla^2 - 2B^{\pm} \mathbf{e} \cdot \nabla] \rho_1^{\pm}(\mathbf{r}, \tau) = 0, \tag{25}$$

with

$$B^+ = \beta'(1), \quad B^- = \beta'(0). \tag{26}$$

B. Equations near the interface

We are going to expand Eqs. (10)–(12) in the transition layer where the chemical potential is not close to its minima. Let us denote by $\mathbf{r}_\Gamma$ a point there, i.e., a point with a distance of order ϵ from Γ, and introduce the quantity $\Phi(\mathbf{r}_\Gamma, \tau)$ as the signed distance between point $\mathbf{r}_\Gamma$ and the interface Γ at the time $t = \epsilon^{-1} \tau$. The sign is chosen such that $\Phi > 0$ in Ω^+ and $\Phi < 0$ in Ω^-. Furthermore, we define

$$\mathbf{m}(\mathbf{r}_\Gamma, \tau) = \nabla \Phi(\mathbf{r}_\Gamma, \tau), \tag{27}$$

$$\kappa(\mathbf{r}_\Gamma, \tau) = \nabla^2 \Phi(\mathbf{r}_\Gamma, \tau), \tag{28}$$

$$V(\mathbf{r}_\Gamma, \tau) = \partial_\tau \Phi(\mathbf{r}_\Gamma, \tau). \tag{29}$$

As can be seen from these definitions, if $\mathbf{r}_\Gamma$ lies on Γ, the vector $\mathbf{m}$ is the unit normal of the interface at $\mathbf{r}_\Gamma$, κ is the curvature there, and V is the normal velocity of Γ on the time scale τ. Signs are such that the vector $\mathbf{m}$ points into Ω^+, the curvature κ is positive if the center of curvature lies in Ω^-, and V is positive if the interface moves towards Ω^-.

Since the transition layer has a width of order ϵ, it is convenient to introduce a variable

$$z = \epsilon^{-1} \Phi(\mathbf{r}_\Gamma, \tau) \tag{30}$$

and for an arbitrary field $f(\mathbf{r}_\Gamma, t)$ a field $\tilde{f}(z, \mathbf{r}_\Gamma, \tau)$ by

$$f(\mathbf{r}_\Gamma, t) = \tilde{f}(\epsilon^{-1} \Phi(\mathbf{r}_\Gamma), \mathbf{r}_\Gamma, \epsilon t). \tag{31}$$

The derivatives occurring in our equations are transformed as follows:

$$\nabla f = [\nabla_r + \epsilon^{-1} \mathbf{m} \partial_z] \tilde{f}, \tag{32}$$

$$\nabla^2 f = [\nabla_r^2 + \epsilon^{-1} \kappa \partial_z + \epsilon^{-2} \partial_z^2] \tilde{f}, \tag{33}$$

$$\partial_t f = [\epsilon \partial_\tau + V \partial_z] \tilde{f}, \tag{34}$$

where ∇_r is the gradient acting on $\mathbf{r}_\Gamma$ only, and arguments were dropped for convenience. Assuming furthermore that all relevant fields near Γ depend only on their relative position with respect to Γ, i.e., that $\tilde{f}(z, \mathbf{r}_\Gamma + \delta \mathbf{m}(\mathbf{r}_\Gamma, \tau), \tau) = \tilde{f}(z, \mathbf{r}_\Gamma, \tau)$ for any small number δ, we find

$$\nabla\Phi(\mathbf{r}_\Gamma,\tau)\cdot\nabla_r\tilde{f}(z,\mathbf{r}_\Gamma,\tau)=0. \tag{35}$$

With the above relations we are ready to transform and expand in powers of ϵ as

$$\rho=\tilde{\rho}=\tilde{\rho}_0+\epsilon\tilde{\rho}_1+\epsilon^2\tilde{\rho}_2+O(\epsilon^3), \tag{36}$$

$$\nu=\tilde{\nu}=\tilde{\nu}_0+\epsilon\tilde{\nu}_1+\epsilon^2\tilde{\nu}_2+O(\epsilon^3), \tag{37}$$

$$\beta=\tilde{\beta}=\tilde{\beta}_0+\epsilon\tilde{\beta}_1+\epsilon^2\tilde{\beta}_2+O(\epsilon^3), \tag{38}$$

where the arguments, which we have dropped for convenience, have to be taken according to Eq. (31). The expansions of ν and σ are related to the one of ρ by means of Eqs. (12) and (14). These relations can be cast in an explicit form using Eqs. (32)–(35). The following ones we are going to use later:

$$\tilde{\nu}_0=\tilde{\rho}_0(\tilde{\rho}_0-1)(\tilde{\rho}_0-\tfrac{1}{2})-\partial_z^2\tilde{\rho}_0, \tag{39}$$

$$\tilde{\nu}_1=[3\tilde{\rho}_0(\tilde{\rho}_0-1)+\tfrac{1}{2}]\tilde{\rho}_1-\kappa\partial_z\tilde{\rho}_0-\partial_z^2\tilde{\rho}_1, \tag{40}$$

$$\tilde{\nu}_2=[3\tilde{\rho}_0(\tilde{\rho}_0-1)+\tfrac{1}{2}]\tilde{\rho}_2+\frac{3}{2}(2\tilde{\rho}_0-1)\tilde{\rho}_1^2 -\nabla_r^2\tilde{\rho}_0-\kappa\partial_z\tilde{\rho}_1-\partial_z^2\tilde{\rho}_2, \tag{41}$$

$$\tilde{\beta}_0=\beta(\tilde{\rho}_0), \tag{42}$$

$$\tilde{\beta}_1=\tilde{\rho}_1\beta'(\tilde{\rho}_0). \tag{43}$$

Relations (42) and (43) hold under the condition (17) only. Inserting now expressions (32)–(34) as well as expansions (36)–(38) into our evolution equations (10) and (11) and comparing terms of order $\epsilon^{-2},\epsilon^{-1},\epsilon^0$, respectively, results in the set of equations

$$0=\partial_z^2\tilde{\nu}_0, \tag{44}$$

$$0=\kappa\partial_z\tilde{\nu}_0+\partial_z^2\tilde{\nu}_1-\mathbf{e}\cdot\mathbf{m}\partial_z\tilde{\beta}_0, \tag{45}$$

$$V\partial_z\tilde{\rho}_0=\nabla_r^2\tilde{\nu}_0+\kappa\partial_z\tilde{\nu}_1+\partial_z^2\tilde{\nu}_2-\mathbf{e}\cdot\nabla_r\tilde{\beta}_0-\mathbf{e}\cdot\mathbf{m}\partial_z\tilde{\beta}_1. \tag{46}$$

The dynamics of the order parameter near the interface is described by the above equations combined with the following boundary conditions: The solution of the above equations should match $\rho^\pm(\mathbf{r}_\Gamma,\tau)$ in $\Omega^\pm$, i.e., outside the transition layer. We demand

$$\lim_{z\to\pm\infty}\tilde{\rho}(z,\mathbf{r}_\Gamma,\tau)=\rho^\pm(\mathbf{r}_\Gamma,\tau). \tag{47}$$

Using $\lim_{z\to\pm\infty}\tilde{\rho}(z,\mathbf{r}_\Gamma,\tau)=\lim_{z\to\pm\infty}\rho(\mathbf{r}_\Gamma+\epsilon z\mathbf{m},\epsilon^{-1}\tau)$ and expanding the above condition in powers of ϵ yields

$$\lim_{z\to\pm\infty}\tilde{\rho}_0(z,\mathbf{r}_\Gamma,\tau)=\rho_0^\pm, \tag{48}$$

$$\lim_{z\to\pm\infty}\tilde{\rho}_1(z,\mathbf{r}_\Gamma,\tau)=\rho_1^\pm(\mathbf{r}_\Gamma,\tau), \tag{49}$$

$$\lim_{z\to\pm\infty}\tilde{\rho}_2(z,\mathbf{r}_\Gamma,\tau)=\rho_2^\pm(\mathbf{r}_\Gamma,\tau)+\lim_{z\to\pm\infty}[z\mathbf{m}\cdot\nabla\rho_1^\pm(\mathbf{r}_\Gamma,\tau)]. \tag{50}$$

Here we used Eqs. (21), (23), and (24).

One can now solve Eqs. (44) and (39) with the boundary condition (48). The result is

$$\tilde{\rho}_0(z,\mathbf{r}_\Gamma,\tau)=[1+e^{-z/\sqrt{2}}]^{-1}, \tag{51}$$

$$\tilde{\nu}_0(z,\mathbf{r}_\Gamma,\tau)=0. \tag{52}$$

Let us now define the following integrals for later use:

$$S=\int_{-\infty}^{\infty}[\partial_z\tilde{\rho}_0(z)]^2dz=\frac{\sqrt{2}}{12}, \tag{53}$$

$$I_n=\int_{-\infty}^{\infty}[\tilde{\rho}_0(z)]^n\beta(\tilde{\rho}_0(z))dz=\sqrt{2}\int_0^1\frac{\rho^{n-1}}{1-\rho}\beta(\rho)d\rho. \tag{54}$$

The first integral was computed and the second one was simplified using Eq. (51). Next we insert $\tilde{\nu}_0=0$ into Eq. (45) and integrate over z. We fix the integration constants by means of the limits $z\to\pm\infty$. In order to compute these limits we expand $\lim_{z\to\pm\infty}\tilde{\nu}(z,\mathbf{r}_\Gamma,\tau)=\lim_{z\to\pm\infty}\nu(\mathbf{r}_\Gamma+\epsilon z\mathbf{m},\epsilon^{-1}\tau)$ in powers of ϵ and use the fact that $\beta(0)=\beta(1)=0$. We obtain

$$\partial_z\tilde{\nu}_1=\mathbf{e}\cdot\mathbf{m}\beta_0. \tag{55}$$

Integrating once more over z and taking again the limits $z\to\pm\infty$ yields a relation between ρ_1^+ and ρ_1^-. Another relation is obtained by multiplying Eq. (45) by $\partial_z\tilde{\rho}_0$ and integrating over z from $-\infty$ to $+\infty$. Combining those two yields

$$\rho_1^+(\mathbf{r}_\Gamma,\tau)=-2S\kappa(\mathbf{r}_\Gamma,\tau)+2I_1\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau), \tag{56}$$

$$\rho_1^-(\mathbf{r}_\Gamma,\tau)=-2S\kappa(\mathbf{r}_\Gamma,\tau)+2(I_1-I_0)\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau), \tag{57}$$

where I_0 and I_1 are the integrals defined by Eq. (54).

In a last step we integrate Eq. (46) and take the limits $z\to\pm\infty$. As a result we obtain an expression for the interface velocity

$$V(\mathbf{r}_\Gamma,\tau)=\frac{1}{2}\mathbf{m}(\mathbf{r}_\Gamma,\tau)[\nabla\rho_1^+(\mathbf{r},\tau)-\nabla\rho_1^-(\mathbf{r},\tau)]\big|_{r=r_\Gamma} +[I_0+2(B^+-B^-)S]\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau)\kappa(\mathbf{r}_\Gamma,\tau) -2[(B^+-B^-)I_1+B^-I_0][\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau)]^2, \tag{58}$$

where $B^\pm$ is defined by Eq. (26), S by Eq. (53) and I_0,I_1 by Eq. (54).

The above equations provide a macroscopic description of the interface dynamics. Macroscopic means that the space resolution is of order ϵ^0, so that the transition layer can be identified with interface Γ, i.e., any point $\mathbf{r}_\Gamma$ can be considered as lying on Γ. Suppose that at a time $t=\epsilon^{-1}\tau$ there is an interface Γ with a unit normal $\mathbf{m}(\mathbf{r}_\Gamma,\tau)$ and a curvature $\kappa(\mathbf{r}_\Gamma,\tau)$ at its points $\mathbf{r}_\Gamma$. Away from the interface the order parameter is given by $\rho^\pm(\mathbf{r}_\Gamma,\tau)=\rho_0^\pm+\epsilon\rho_1^\pm(\mathbf{r}_\Gamma,\tau)$, where

$\rho_0^+=1$, $\rho_0^-=0$, and $\rho_1^\pm(\mathbf{r}_\Gamma,\tau)$ is given by the solution of Eq. (25) with boundary conditions on the interface defined by Eqs. (56) and (57) in terms of $\mathbf{m}(\mathbf{r}_\Gamma,\tau)$ and $\kappa(\mathbf{r}_\Gamma,\tau)$. This solution in turn determines the normal velocity of each interface point by means of Eq. (58).

IV. LINEAR STABILITY ANALYSIS

As derived in the preceding section, on the scale $\tau=\epsilon^{-1}t=\epsilon^{-1}T_0^{-1}T$ the motion of an interface at $\mathbf{r}=\mathbf{r}_\Gamma$ separating the two regions Ω^+ [with a particle density $\rho^+(\mathbf{r},\tau)=1+\epsilon\rho_1^+(\mathbf{r},\tau)+O(\epsilon^2)$] and Ω^- [with $\rho^-(\mathbf{r},\tau)=0+\epsilon\rho_1^-(\mathbf{r},\tau)+O(\epsilon^2)$] is given the normal interfacial velocity V of the interface

$$V(\mathbf{r}_\Gamma,\tau)=\frac{1}{2}\mathbf{m}(\mathbf{r}_\Gamma,\tau)[\nabla\rho_1^+(\mathbf{r},\tau)-\nabla\rho_1^-(\mathbf{r},\tau)]|_{r=r_\Gamma}+[I_0+2(B^+-B^-)S]\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau)\kappa(\mathbf{r}_\Gamma,\tau)-2[(B^+-B^-)I_1+B^-I_0][\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau)]^2, \tag{59}$$

where $\mathbf{m}$ is the local normal to the interface, $\kappa=\nabla\cdot\mathbf{m}$ is the local curvature, and $B^\pm$, S, I_0, and I_1 are constants that depend exclusively on the form of the conductivity σ and are given by Eqs. (26), (53), and (54). The velocity depends on $\rho_1^\pm(\mathbf{r},\tau)$, which satisfy the *linear* partial differential equation

$$[\nabla^2-2B^\pm\mathbf{e}\cdot\nabla]\rho_1^\pm(\mathbf{r},\tau)=0 \tag{60}$$

subjected to the boundary conditions

$$\rho_1^+(\mathbf{r}_\Gamma,\tau)=-2S\kappa(\mathbf{r}_\Gamma,\tau)+2I_1\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau), \tag{61}$$

$$\rho_1^-(\mathbf{r}_\Gamma,\tau)=-2S\kappa(\mathbf{r}_\Gamma,\tau)+2(I_1-I_0)\mathbf{e}\cdot\mathbf{m}(\mathbf{r}_\Gamma,\tau) \tag{62}$$

on the points $\mathbf{r}_\Gamma$ of the interface.

We are interested in the stability of a flat interface against small perturbations in its profile. Let us consider an interface Γ of the form

$$y_\Gamma(x,\tau)=he^{i(kx-\omega\tau)}+V_0\tau,\quad kh\ll1 \tag{63}$$

(see Fig. 1) separating two semi-infinite regions Ω^+ [where $y>y_\Gamma(x,\tau)$] and Ω^- [where $y<y_\Gamma(x,\tau)$]. The direction $\mathbf{e}$ of the external electric field is arbitrary for the moment. To first order in kh, we have

$$m_x=-ikhe^{i(kx-\omega\tau)},\quad m_y=1 \tag{64}$$

for the x and y components of $\mathbf{m}$, respectively. The local curvature κ of the interface is given by

$$\kappa=k^2he^{i(kx-\omega\tau)}, \tag{65}$$

while the normal velocity V takes the form

$$V=-dy_s/d\tau=-V_0+i\omega he^{i(kx-\omega\tau)}. \tag{66}$$

Equation (60) has a solution $\rho_1^\pm(\mathbf{r},\tau)$ of the form

$$\rho_1^\pm(\mathbf{r},\tau)=f^\pm(\zeta)+khe^{i(kx-\omega\tau)}g^\pm(\zeta)+O(kh)^2, \tag{67}$$

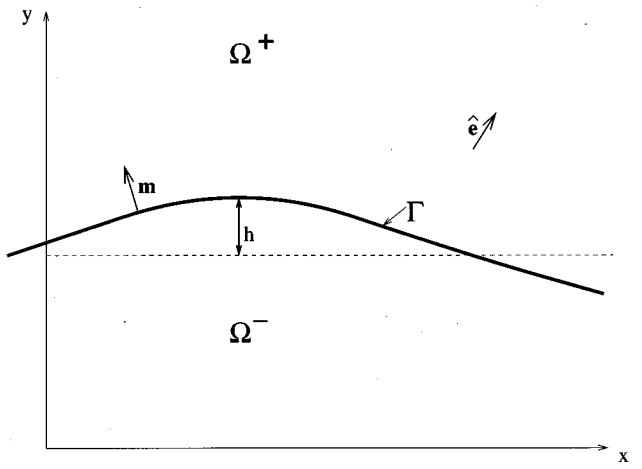

FIG. 1. Schematic picture of the approximately flat interface Γ defined by Eq. (63). The interface separates the semi-infinite regions Ω^+ and Ω^-, in which $\rho\approx1$ and $\rho\approx0$, respectively. In the figure $\mathbf{m}$ denotes the unit normal vector of Γ and h the amplitude of the perturbation around a flat interface. The unit normal vector of the external field is denoted by $\hat{\mathbf{e}}$.

with $\zeta=y-he^{i(kx-\omega\tau)}$. The boundary conditions (61) and (62) are satisfied for

$$f^+(\zeta)=2I_1e_y-C^+(1-e^{2B^+e_y\zeta}), \tag{68}$$

$$g^+(\zeta)=-[2Sk+2ie_xI_1]e^{-\lambda^+\zeta}-2C^+\frac{B^+e_y}{k}\times(e^{-\lambda^+\zeta}-e^{2B^+e_y\zeta}), \tag{69}$$

$$f^-(\zeta)=2(I_1-I_0)e_y-C^-(1-e^{2B^-e_y\zeta}), \tag{70}$$

$$g^-(\zeta)=-[2Sk+2ie_x(I_1-I_0)]e^{-\lambda^-\zeta}-2C^-\frac{B^-e_y}{k}\times(e^{-\lambda^-\zeta}-e^{2B^-e_y\zeta}), \tag{71}$$

where e_x and e_y are the x and y components of the unit vector $\mathbf{e}$ pointing in the direction of the external field $\mathbf{E}$ and $\lambda^\pm$ are solutions of the quadratic equation

$$(\lambda^\pm)^2+2B^\pm e_y\lambda^\pm-k(k+2ie_xB^\pm)=0 \tag{72}$$

subjected to the constraint $\mathrm{Re}(\lambda^+)>0$ [$\mathrm{Re}(\lambda^-)<0$]. The new constants C^+ and C^- are determined by the boundary conditions at $z=\pm\infty$. We will discuss the effects of $C^\pm$ in Sec. IV A.

Inserting $\rho_1^\pm(\mathbf{r},\tau)$ given by Eqs. (67)–(71) into the equation (59) for the normal velocity, we can compare with Eq. (66) to obtain an expression for the constant velocity V_0,

$$V_0=B^+(2I_1e_y-C^+)e_y-B^-[2(I_1-I_0)e_y-C^-]e_y \tag{73}$$

$$=[B^+\rho_1^+(z=+\infty)-B^-\rho_1^-(z=-\infty)]e_y, \tag{74}$$

and for ω,

$$i\omega=\frac{1}{2}k\frac{d}{dz}[g^{+}(z)-g^{-}(z)]|_{z=0}+k^{2}[I_{0}+2S(B^{+}-B^{-})]e_{y}$$
$$+4k[I_{0}B^{-}+I_{1}(B^{+}-B^{-})]ie_{x}e_{y}. \quad (75)$$

Assuming that $C^{\pm}$ are real and writing $\lambda^{\pm}=\lambda_{r}^{\pm}+i\lambda_{i}^{\pm}$ and $\omega=\omega_{r}+i\omega_{i}$, we get

$$\omega_{i}=-k^{2}[I_{0}+2S(B^{+}-B^{-})]e_{y}-k^{2}S(\lambda_{r}^{+}-\lambda_{r}^{-})$$
$$+k[I_{1}\lambda_{i}^{+}-(I_{1}-I_{0})\lambda_{i}^{-}]e_{x}-C^{+}B^{+}(\lambda_{r}^{+}+2B^{+}e_{y})e_{y}$$
$$+C^{-}B^{-}(\lambda_{r}^{-}+2B^{-}e_{y})e_{y}. \quad (76)$$

A flat interface is unstable against small perturbation of the form (63) if the external field, the conductivity, and the boundary conditions at infinity are such that $\omega_{i}>0$. Equation (76) is the main result of this paper.

A. The case of a third-order conductivity

In order to further study the properties of Eq. (76) we assume in this section that the conductivity $\sigma(u)$ can be written as a polynomial of the form

$$\sigma(u)=\sigma_{0}+\sigma_{1}u+\sigma_{2}u^{2}+\sigma_{3}u^{3}. \quad (77)$$

Previous studies [11] of the stability of a flat interface in a driven Cahn-Hilliard system of the form (1)–(3) have limited themselves to the case where $\sigma(u)=u(u_{m}-u)$. While we can reproduce their results, we will observe interesting new effects considering the more general form (77) of $\sigma(u)$.

The constants in Eq. (76) can be expressed explicitly in terms of the system parameters. Using Eqs. (26), (53), (54), and (14) we find

$$B^{+}=A(\sigma_{2}+2\sigma_{3}u_{m}), \quad (78)$$

$$B^{-}=-A(\sigma_{2}+\sigma_{3}u_{m}), \quad (79)$$

$$I_{0}=-\sqrt{2}A(\sigma_{2}+\tfrac{3}{2}\sigma_{3}u_{m}), \quad (80)$$

$$I_{1}=-\sqrt{2}A(\tfrac{1}{2}\sigma_{2}+\sigma_{3}u_{m}), \quad (81)$$

$$S=\frac{\sqrt{2}}{12}, \quad (82)$$

with

$$A=\epsilon^{-1}|\mathbf{E}|\xi\lambda^{-3/2}u_{m}^{-2}>0. \quad (83)$$

All of the above constants are independent of σ_{0} and σ_{1}. The reason for this is that the equations for the order parameter do not depend on σ_{0} and that a change of σ_{1} amounts to a change of the reference frame only.

1. External field parallel to interface

In this case the direction of the field is such that $e_{x}=\pm 1$, $e_{y}=0$. To lowest order in k we get, using $\lambda_{r}^{\pm}\approx\pm k^{1/2}(|B^{\pm}|)^{1/2}$ and $\lambda_{i}^{\pm}=kB^{\pm}e_{x}/\lambda_{r}^{\pm}$,

$$\omega_{i}=k^{3/2}\left[I_{1}\frac{B^{+}}{\sqrt{|B^{+}|}}+(I_{1}-I_{0})\frac{B^{-}}{\sqrt{|B^{-}|}}\right]. \quad (84)$$

Using the explicit expressions (78)–(83), one can show that $\omega_{i}<0$, i.e., that the interface is stable, for any choice of $\{\sigma_{0},\sigma_{1},\sigma_{2},\sigma_{3}\}$. This result does not depend on the sign of e_{x}, nor does it depend on the actual boundary conditions away from the interface through the constants $C^{\pm}$.

2. External field perpendicular to interface

In this case we have $e_{x}=0$ and $e_{y}=+1$ if the external field points into the high density region Ω^{+} or $e_{x}=0$ and $e_{y}=-1$ if it points towards the low density region Ω^{-}. We also have $\lambda_{i}^{\pm}=0$ from Eq. (72).

Depending on the signs of $B^{+}e_{y}$ and $B^{-}e_{y}$ we get different conditions on the possible values of the constants $C^{\pm}$. From Eqs. (68) we see that if $B^{+}e_{y}>0$ ($B^{-}e_{y}<0$) we must take $C^{+}=0$ ($C^{-}=0$) in order for ρ_{1}^{+} (ρ_{1}^{-}) to be finite as $z\to\infty$ ($z\to-\infty$). This means that the system does not support a single interface in this case unless there is some current of particles at infinity (see also the discussion of the one-dimensional model in Ref. [12]).

If $B^{+}e_{y}<0$ ($B^{-}e_{y}>0$) then there are no such constraints on C^{+} (C^{-}) and we chose the special cases $C^{+}=2I_{1}e_{y}$ [$C^{-}=2(I_{1}-I_{0})e_{y}$] which makes ρ_{1}^{+} (ρ_{1}^{-}) vanish at infinity.

We are left with four cases to study for ω_{i}.

Case A: $B^{+}e_{y}>0$ *and* $B^{-}e_{y}<0$. If we assume that $k\ll B^{\pm}$, then $\lambda_{r}^{\pm}\approx\frac{1}{2}(k^{2}e_{y}/B^{\pm})$ and we get, to lowest order,

$$\omega_{i}=-k^{2}e_{y}[I_{0}+2S(B^{+}-B^{-})]. \quad (85)$$

Case B: $B^{+}e_{y}<0$ *and* $B^{-}e_{y}>0$. To lowest order we write $\lambda_{r}^{\pm}=-2B^{\pm}e_{y}[1+k^{2}/4(B^{\pm})^{2}-k^{4}/16(B^{\pm})^{4}]$, and get

$$\omega_{i}=\frac{-k^{4}e_{y}}{4(B^{+})^{2}(B^{-})^{2}}[I_{1}(B^{-})^{2}-(I_{1}-I_{0})(B^{+})^{2}$$
$$+2S(B^{+}-B^{-})B^{+}B^{-}]. \quad (86)$$

Case C: $B^{+}e_{y}<0$ *and* $B^{-}e_{y}<0$. With $\lambda_{r}^{+}=-2B^{+}e_{y}[1+k^{2}/4(B^{+})^{2}]$, $\lambda_{r}^{-}=-k^{2}e_{y}/2B^{-}$, we get

$$\omega_{i}=-k^{2}e_{y}(I_{0}-I_{1}-2SB^{-}). \quad (87)$$

Case D: $B^{+}e_{y}>0$ *and* $B^{-}e_{y}>0$. This is essentially the reverse of the previous case. With $\lambda_{r}^{+}=k^{2}e_{y}/B^{+}$, $\lambda_{r}^{-}=-2B^{-}e_{y}[1+k^{2}/4(B^{-})^{2}]$, and get

$$\omega_{i}=-k^{2}e_{y}(I_{1}+2SB^{+}). \quad (88)$$

Inserting the explicit expressions (78)–(83) for $B^{\pm}$, I_{0}, I_{1}, and S into the above equations for ω_{i}, one arrives at the following conclusion: In all four cases the interface is unstable if

$$(2\sigma_{2}+3\sigma_{3}u_{m})e_{y}>0 \quad (89)$$

and stable otherwise. Condition (89) is the main result of this section.

It is natural to assume the conductivity to be positive in the whole interval $[0,u_{m}]$. Assuming furthermore that $\sigma(u)$

is symmetric around u_m like the chemical potential and vanishes for $u=0$, we are led to the standard expression $\sigma(u)=u(u_m-u)$ [11]. There $\sigma_2<0$, $\sigma_3=0$, and the interface is unstable if $e_y=-1$, i.e., the case where the external field points away from the high density region Ω^+ [11]. Although the assumptions of symmetry and vanishing $\sigma(0)$ are natural for certain Ising-like [8–12,14] or particle-hopping models [13], one can think of more general situations (e.g., the microscopic model in [15]). Then the parameters $\{\sigma_0,\sigma_1,\sigma_2,\sigma_3\}$ can be such that $(2\sigma_2+3\sigma_3u_m)>0$. In this case the interface is unstable for $e_y=+1$, i.e., if the external field points into the high-density region Ω^+.

V. SUMMARY

We have applied a matched asymptotic expansion to a system of equations describing the dynamics of phase segregation in the presence of an external field. The influence of the field on a region of local density u is given by a conductivity $\sigma(u)$. We derived equations for the dynamics of an interface separating two regions in different phases [see Eqs. (25), (56), (57), and (58)] and studied the stability of a flat interface against small perturbations. We found a general condition [see Eq. (76)] for the stability of such an interface and discussed in more detail the case where $\sigma(u)$ can be written as a third-order polynomial in u. In this case the interface is always stable if the field is parallel to it. However, if the field is perpendicular to the interface, the interface can be either stable or unstable, depending on the conductivity and on the direction of the field [see Eq. (89)].

ACKNOWLEDGMENTS

We thank B. Schmittmann and R. K. P. Zia for stimulating discussions. S.S. gratefully acknowledges financial support by the Deutsche Forschungsgemeinschaft. C.A.L. has been supported by the National Science Foundation through Grant No. DMR 94-19393.

[1] J. D. Gunton, M. San Miguel, and P. S. Shani, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8.
[2] J. S. Langer, in *Solids far from Equilibrium*, edited by C. Godrèche (Cambridge University Press, Cambridge, 1991).
[3] B. Schmittmann and R. P. K. Zia, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, London, 1995), Vol. 17.
[4] J. W. Cahn and J. E. Hilliard, J. Chem. Phys. **28**, 258 (1958).
[5] R. L. Pego, Proc. R. Soc. London, Ser. A **422**, 261 (1989).
[6] K. Kitahara, Y. Oono, and D. Jasnow, Mod. Phys. Lett. B **2**, 765 (1988).
[7] J. S. Langer, N. Bar-On, and H. D. Miller, Phys. Rev. A **11**, 1417 (1975).
[8] S. Puri, N. Parekh, and S. Dattagupata, J. Stat. Phys. **75**, 839 (1994).
[9] A. M. Lacasta, A. Hernández-Machado, and J. M. Sancho, Phys. Rev. B **48**, 9418 (1993).
[10] A. J. Bray, Adv. Phys. **43**, 357 (1994).
[11] C. Yeung, J. L. Mozos, A. Hernánez-Machado, and D. Jasnow, J. Stat. Phys. **70**, 1149 (1993).
[12] C. L. Emmot and A. J. Bray, Phys. Rev. E **54**, 4568 (1996).
[13] G. Giacomin and J. L. Lebowitz, Phys. Rev. Lett. **76**, 1094 (1996).
[14] F. J. Alexander, C. A. Laberge, J. L. Lebowitz, and R. K. P. Zia, J. Stat. Phys. **82**, 1133 (1996).
[15] L. K. Wickham and J. P. Sethna, Phys. Rev. B **51**, 15 017 (1995).