Stefan and Hele-Shaw type models as asymptotic limits of the phase-field equations

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Using detailed asymptotic analyses of the dynamics of the phase-field model, we show that the major sharp-interface models (Stefan, modified Stefan, Hele-Shaw, etc.) all arise as limiting cases of the phase-field equations. The scaling of the physical parameters in the microscopics leads to distinct macroscopic models with critical differences.

I. INTRODUCTION

In this paper we show, using detailed formal asymptotics, that one can obtain any of the major sharp-interface (discontinuous-gradient) models (e.g., Stefan, modified Stefan models, Hele-Shaw models) as limiting cases of a particular continuous representation of phase transitions which is based on microscopic considerations (see Figs. 1 and 2). Furthermore, the distinctions in the macroscopic sharp-interface models arise from the scaling relationships in the microscopic parameters of the continuous or phase-field model (see Sec. III). We discuss first the three macroscopic sharp-interface problems which have been used in modeling phase transitions.

One considers a material which may be in either of two phases, e.g., solid or liquid, and occupies a region $\Omega \subset \mathbb{R}^N$ in space. We let *u* denote the (dimensionless) temperature and shift it so that u = 0 is the usual equilibrium melting temperature. In the classical Stefan problem, the interface is defined to be

$$\Gamma(t) = \{ x \in \Omega : u(t,x) = 0 \} , \qquad (1.1)$$

while the liquid region Ω_1 is defined by

$$\Omega_1(t) = \{ x \in \Omega: \ u(t,x) > 0 \} , \qquad (1.2)$$

and the solid region Ω_2 analogously for negative u.

The temperature must satisfy the heat diffusion equation

$$u_t = K \Delta u, \quad x \in \Omega_1(t) \text{ or } x \in \Omega_2(t) ,$$
 (1.3)

where K is a dimensionless thermal diffusivity. Across the interface the latent heat condition must be satisfied

$$lv = K \left(\nabla u_s - \nabla u_L \right) \cdot \hat{\mathbf{n}}, \quad x \in \Gamma(t)$$
(1.4)

where $\hat{\mathbf{n}}$ is the unit normal to Γ (in the direction solid to liquid), v the (normal) velocity, and l is a dimensionless latent heat. Together with the temperature condition [by definition of $\Gamma(t)$]

$$u(t,x) = 0, \quad x \in \Gamma(t) \tag{1.5}$$

one must specify external boundary conditions and initial conditions, e.g.,

$$u(t,x) = u_{\partial}(t,x), \quad x \in \partial\Omega, \quad t \in \mathbb{R}^+$$
(1.6)

 $u(0,x) = u_0(x), x \in \Omega$ (1.7)

The mathematical problem¹ then is to find u and Γ , in suitable function spaces, satisfying (1.3)-(1.7).

The physical situation is generally more complicated than indicated by the classical Stefan model, particularly in two or higher dimensions.² One of the physical effects neglected by the classical Stefan model is that of surface tension, which is generally a stabilizing factor. As noted by Gibbs in the last century,³ an immediate consequence of surface tension, as an equilibrium property, to modify the temperature at the interface so that

$$\Delta s [u(t,x)] = -\sigma \kappa(t,x), \quad x \in \Gamma(t)$$
(1.8)

where Δs is the difference in entropy between liquid and solid, σ is the surface tension, and $\kappa(t,x)$ is the sum of principal curvatures at a point on the interface. One may thus study the system [(1.3),(1.4),(1.8)] as an alternative to the classical Stefan model. However, there is now a problem of formulation which is often a practical problem as well. The interface is no longer defined simply by (1.1) but must be "tracked." In some applications, such as linear stability analysis, this is quite convenient; in others such as numerical computations it presents difficulties.

In addition to the surface tension effect, metallurgists observed that the temperature at the interface should be reduced beyond the "supercooling" exhibited by (1.8).^{4,2} Although different expressions for this "kinetic undercooling" term have been used, the most prevalent model has been the linear velocity dependence

$$\Delta s \left[u(t,x) \right] = -\sigma \kappa(t,x) - \alpha \sigma v(t,x) , \qquad (1.9)$$

where α has generally been regarded as an adjustable parameter. The phase-field equations to be discussed in Sec. II have been used to derive (1.9) and relate α to a microscopic relaxation time. This will be discussed in detail in Sec. II. The problem posed by Eqs. (1.3),(1.4),(1.9), which we call the modified Stefan problem, can be studied in the hope of obtaining a more realistic picture of the interface.

Perhaps the most interesting aspect of the differences between the three models is manifested in the stability properties of the interface. The interface for the classical Stefan problem is notoriously unstable⁵ under some conditions. The insertion of the condition (1.8) clearly restricts the magnitude of the curvature and thereby the ex-

39 5887



STEFAN-TYPE MODELS AS LIMITING CASES OF THE PHASE FIELD EQUATIONS

FIG. 1. In the scaling limits shown at left, the phase-field equations are governed by the various Stefan-type limits. The phase-field equations can be used to approximate any of these sharp-interface problems.

tent of the instability. That is, if the temperature is restricted via the initial and boundary conditions, then a large surface tension σ is not compatible with a large curvature κ .

The role of nonzero α in the problem [(1.3),(1.4),(1.9)] is not as manifest. It has been shown⁶ that this is a stabilizing influence, in the sense that an unstable mode will remain unstable but with a smaller amplitude.

Thus, it is clear that the three problems posed above will lead to very different behavior of the interface. In practical terms, a reliable calculation (numerical or analytical) for a particular material is only possible if the appropriate choice of models is made. This in turn depends crucially on parameters such as σ and α .

We have assumed so far that the interface is perfectly sharp, and that the latent heat is released on this set of measure zero.

We note that Eqs. (1.3) and (1.4) can be incorporated in a weak sense⁷ into the single equation

$$u_t + \frac{l}{2}\varphi_t^{(s)} = K\Delta u, \quad \varphi^{(s)} \equiv \begin{cases} +1 & \text{liquid} \\ -1 & \text{solid} \end{cases}.$$
 (1.10)

Within this interpretation it is heuristically evident that each of these three models describes an interface of discontinuity in the phase φ . In Sec. II we will consider the phase-field model of phase transitions and then obtain



HELE-SHAW AND CAHN-ALLEN MODELS AS LIMITING CASES

OF THE PHASE FIELD EQUATIONS

FIG. 2. In the first two scaling limits, the phase-field model is governed by the equations which are known as the quasistatic limit in phase transitions and the Hele-Shaw equations in fluid flow. In the last limit, the Cahn-Allen equation for antiphase domain boundaries is attained.

each of these three models as limiting cases. In Sec. VII we show that two Hele-Shaw models also arise as limits.

II. THE PHASE-FIELD MODEL

As noted earlier, acceptance of the idea that the temperature need not to be zero at the interface (or negative in the solid, etc.) leads immediately to the question of how one distinguishes the two phases. In fact, if there were no macroscopically measurable quantity, let us call it φ , which differed in the two phases, then we probably would not be interested in the interface between phases. In general, this quantity φ is called an "order parameter" or "phase field," and one occurrence of such a function is in (1.10). The inhomogeneous heat equation (1.10) can be expected to remain valid even if φ is continuous. However, the key question is how should one determine φ ? Accepting this second variable involves the implementation of a second equation, which is fundamental in determining the relevant physics. Using the analog of Eq. (1.10) and Landau-Ginzburg theory of phase transitions⁸ one obtains the system^{9, 10}

$$u_t + \frac{l}{2}\varphi_t = K\Delta u \quad , \tag{2.1}$$

$$\alpha\xi^2\varphi_t = \xi^2\Delta\varphi + \frac{1}{a}g(\varphi) + 2u \quad , \tag{2.2}$$

where g is a derivative of a symmetric double-well potential with minima at ± 1 , e.g., $g(\varphi) = \frac{1}{2}(\varphi - \varphi^3)$. The parameters l, K, α , ξ , and a, are dimensionless constants whose physical interpretation can be found in Refs. 9 and 10. We can assume initial conditions

$$u(0,x) = \tilde{u}(x), \quad \varphi(0,x) = \tilde{\varphi}(x), \quad x \in \Omega$$
(2.3)

and boundary conditions, for example,

$$u(0,x) = u_{\partial}(x), \quad \varphi(t,x) = \varphi_{\pm}(x), \quad x \in \partial \Omega$$
 (2.4)

where φ_{\pm} are the largest and smallest roots, respectively, of $a^{-1}f(\varphi) + 2u = 0$. Since *a* will be a small parameter, these roots will be near ± 1 . The asymptotic analysis does not depend crucially on the boundary conditions (2.4) and one can use instead Neumann boundary conditions $\partial \varphi / \partial v = \partial u / \partial v = 0$ (*v* normal to $\partial \Omega$).

The interface in (2.1) and (2.2) is specified as

$$\Gamma(t) = \{ x \in \Omega \colon \varphi(t, x) = 0 \} .$$
(2.5)

Within this formulation, interfacial conditions such as (1.4), (1.5), (1.8), and (1.9) need not be imposed and in fact can be derived from (2.1) and (2.2) as a consequence of the microscopic physics built into these equations.⁹⁻¹⁴

We note that under rather general conditions there exists a unique global solution to Eqs. (2.1)-(2.4) in arbitrary dimension which is smooth.⁹ The situation is quite different for the problems discussed in Sec. I, in which there is no existence theory for the modified Stefan problems and much of the existence theory for the classical Stefan problem is limited to one and two dimensions.^{15,16}

Our aim in this paper is to show using the methods of Refs. 9, 11, and 17 that all of the sharp interface problems discussed in Secs. I and VII arise as particular limits of the phase-field equations (2.1) and (2.2) in the asymptotic analysis as ξ , a, and in some cases α , approach zero.

The asymptotics of the dynamical situation considered previously (e.g., in Ref. 17) concerns the limit as ξ approaches zero with α and a fixed. This is an analysis of the small temperature (i.e., proportional to ξ) limit. Within this limit one does not obtain any of the Stefan or Hele-Shaw limits discussed in Secs. I and VII.

In this paper, however, we show that scaling of the parameters (particularly the new parameter a) is crucial in the limiting behavior of the equations. In particular, one obtains distinct limits with very different behavior as a consequence due to the physical implications of this scaling (see Sec. III).

The objective in considering these limits is to make contact with other macroscopic models which have been or can be studied. The result of this asymptotic analysis is that an arbitrary Stefan-type or Hele-Shaw-type model with any set of physical parameters in any dimension can be approximated with arbitrary accuracy by a suitable set of phase-field equations (2.1), (2.2), and vice versa.

The significance of Secs. IV-VII is that they can be used to study (e.g., numerically) such diverse phenomena as fluid interface and various solidification problems using a single set of equations. One needs only to adjust three parameters to observe the change from one system to another. Some related conjectures for theorems have have been discussed in Refs. 18-20.

Single equation limits can also be obtained as limits. By choosing l=0 and, for initial and boundary conditions, u=0, one can trivialize the role of temperature and thereby obtain the Cahn-Allen antiphase domain boundary model²¹

$$\alpha \xi^2 \varphi_t = \xi^2 \Delta \varphi + \frac{1}{a} g(\varphi) \tag{2.6}$$

as a limiting case (see Fig. 2).

III. THE BASIC IDEAS AND HEURISTICS

Before presenting detailed asymptotics, it is useful to indicate, heuristically, the essential strategy, originally introduced in Refs. 9 and 11 with different scaling. Let r be the coordinate normal to the interface Γ (i.e., r is the distance to the interface if it is in the liquid region, negative distance if it is in the solid region). Suppose that in (2.1)-(2.4), φ varies much more rapidly across the interface than u and it attains φ_+ a short distance toward the liquid side and φ_- on the solid side. Suppose further that φ is approximately in the form $\varphi(r - vt)$, and make the following assumptions on the parameters:

$$\epsilon^2 \equiv \xi^2 a, \ \alpha = \text{fixed}, \ \xi, a \to 0, \ \rho \equiv r/\epsilon$$
 (3.1)

Under these conditions we may write (2.2) using the prototype $g(\phi) \equiv \frac{1}{2}(\phi - \phi^3)$, as

$$-\alpha v \epsilon \phi_{\rho} \simeq \phi_{\rho\rho} + \epsilon \kappa \phi_{\rho} + \cdots + \frac{1}{2} (\phi - \phi^3) + 2au \quad , \tag{3.2}$$

where terms of order ϵ^2 in this expansion have been omitted. If there exists an expansion of the form $\phi = \phi^0 + \epsilon \phi^1 + \cdots$, then (3.2) implies that the O(1) balance is

$$\phi_{\rho\rho}^{0} + \frac{1}{2} [\phi^{0} - (\phi^{0})^{3}] = 0 , \qquad (3.3a)$$

with solution

$$\phi_0(\rho) = \tanh(\rho/2) . \tag{3.3b}$$

Subtracting the O(1) equation (3.3) from (3.2) one has the $O(\epsilon)$ equation [provided $a\epsilon^{-1}$ is O(1) or smaller]

$$L\phi^{1} \equiv \phi^{1}_{\rho\rho} + \frac{1}{2} [1 - (3\phi^{0})^{2}]\phi^{1}$$

$$\simeq \epsilon [-\alpha v \phi^{0}_{\rho} - \kappa \phi^{0}_{\rho} - 2(a/\epsilon)u] \equiv F . \qquad (3.4)$$

Noting that the derivative of the O(1) solution satisfies the homogeneous equation for (3.4), $L\phi_{\rho}^{0}=0$, one has the solvability condition

$$0 = (F, \phi_{\rho}^{0}) = \epsilon \int_{-\infty}^{\infty} \phi_{\rho}^{0} [-\alpha v \phi_{\rho}^{0} - \kappa \phi_{\rho}^{0} - 2(a/\epsilon)u] d\rho . \quad (3.5)$$

Since

$$\int_{-\infty}^{\infty}\phi_{\rho}^{0}=\phi_{+}-\phi_{-}\simeq 2,$$

one has from (3.5) the identity

$$4u(t,x) \simeq -\frac{\epsilon}{a} \sigma_0 \kappa(t,x) - \frac{\epsilon}{a} \alpha \sigma_0 v(t,x)$$
(3.6)

on Γ , where

$$\sigma_0 \equiv \int_{-\infty}^{\infty} (\phi_{\rho}^0)^2 d\rho = \frac{2}{3}$$

Hence, $\epsilon a^{-1} = \xi a^{-1/2}$ emerges as an important scaling factor.

Next, we perform a short calculation to understand the significance of $\epsilon a^{-1}\sigma_0$ in (3.6). Equation (2.2) was derived from $\tau \varphi_t = \delta \mathcal{F} / \delta \varphi$, where the free energy \mathcal{F} is given by

$$\mathcal{F}\{\varphi\} = \int_{\Omega} d\mathbf{x} \left[\frac{\xi^2}{2} (\nabla \varphi)^2 + \frac{1}{8a} (\varphi^2 - 1)^2 - 2u\varphi \right]$$

for this prototype $g(\varphi)$. The surface tension σ is defined⁹ as

$$\sigma \equiv \frac{\mathcal{F}\{\varphi\} - \frac{1}{2}\mathcal{F}\{\varphi_+\} - \frac{1}{2}\mathcal{F}\{\varphi_-\}}{A} \cong \frac{\mathcal{F}\{\varphi^0\}}{A} , \qquad (3.7)$$

where A is the area of the interface. To calculate this to first order, one may multiply Eq. (3.3) by ϕ_r^0 and integrate to obtain

$$0 = \int_{-\infty}^{r} \{\xi^2 \varphi_r^0 \varphi_{rr}^0 + \frac{1}{2} \varphi_r^0 [\varphi^0 - (\varphi^0)^3] \} dr , \qquad (3.8)$$

which is an exact integral, i.e.,

$$\frac{\xi^0}{2}(\varphi_r^0)^2 = \frac{1}{8}(\varphi^2 - 1) .$$
(3.9)

Then the free energy in (3.7) may be written as

$$\sigma \simeq \frac{\mathcal{F}\{\varphi^0\}}{A} = \int_{-\infty}^{\infty} \xi^2 (\varphi_r^0)^2 dr = \frac{\epsilon}{a} \int_{-\infty}^{\infty} (\phi_\rho^0)^2 d\rho \quad . \tag{3.10}$$

Noting that the difference in entropy density between liquid and solid is

$$\Delta s \equiv \frac{-\frac{\partial \mathcal{F}}{\partial u} \{\varphi_+\} + \frac{\partial \mathcal{F}}{\partial u} \{\varphi_-\}}{V} \cong 4 , \qquad (3.11)$$

where V is the volume, the relation (1.9) follows as an O(1) statement within this heuristic derivation, provided $\epsilon a^{-1} = \xi a^{-1/2} = O(1)$ or smaller. If, however, $\xi a^{-1/2}$ approaches zero, then one obtains the usual u = 0 Stefan condition (1.5). Finally, if α approaches zero while $\xi a^{-1/2} = O(1)$, one has the limit (1.8).

In each of the scalings, the interfacial width is ϵ , and the solution φ is approximated by (3.3). Hence, far from the interface, φ is constant (to arbitrary order in ϵ), so that the heat equation (1.3) is valid. Across the interface, as ϵ approaches zero, one obtains, as a result of integration, the latent heat condition (1.4) (see also the end of Sec. VI).

In attaining the three Stefan-type limits discussed in Sec. I, it is clear that there is a crucial interplay between $\epsilon = \xi a^{1/2}$ and $\epsilon a^{-1} = \xi a^{-1/2}$, i.e., in the roles of interfacial thickness and interfacial tension. At a deeper level of physics, one has a competition between the molecular or atomic forces, represented by ξ and the well depth, represented by a^{-1} . The well depth, and in fact, the entire double-well potential, $a^{-1}g(\varphi)$, is obtained from the microscopic physics. One can regard it as a representation of the energy barrier between the two phases which, of course, depends on the particular microscopics considered. The double well can also be regarded (from the point of view of ϕ^4 field theory,²² as a probabilistic measure that expresses the "preference," by the "spins" or particles, to be in the liquid, solid, or interfacial part of the phase diagram. On a more fundamental level, the Landau-Ginzburg free energy incorporates the subtle concept of the correlation length, which is a measure of the distance within which atoms influence one another on a probabilistic basis. The correlation length concept provides yet another approach to understanding the different macroscopic limits.

IV. THE MODIFIED STEFAN LIMIT

The heuristic calculations of Sec. III suggest the following limit which we will verify explicitly.

Proposition 4.1. In the limit ξ , $a \rightarrow 0$ with α and $\xi a^{-1/2}$ fixed, there exists a formal asymptotic solution of the phase-field model [(2.1) and (2.2)] which is governed by the modified Stefan model [(1.3),(1.4), and (1.9)].

We verify this by setting

$$c_1 \equiv \xi a^{-1/2}, \ \epsilon_1 = \xi^2, \ f(\varphi) \equiv c_1^2 g(\varphi) ,$$
 (4.1)

and rewriting Eqs. (2.1) and (2.2) as

$$u_t + \frac{1}{2}\varphi_t = K\Delta u \quad , \tag{4.2}$$

$$\alpha \epsilon_1^2 \varphi_t \equiv \epsilon_1^2 \Delta \varphi + f(\varphi) + 2u \epsilon_1 . \qquad (4.3)$$

At this point one may carry out the asymptotics of (4.2) and (4.3) in the style of Ref. 11 or Ref. 17. We choose a direct application of the latter, which is a complete asymptotic analysis for both variables, in order to display explicitly the inner and outer expansions. For convenience, we discuss two dimensions. For higher dimensions the procedure is essentially the same and the net difference is that the curvature is replaced by the sum of principal curvatures. The interface $\Gamma(t)$ defined by (2.5) is regular provided the initial and boundary conditions are smooth.⁹ Hence, for a sufficiently small neighborhood around $\Gamma(t)$, we may define r(x, y, t) to be \pm the distance from (x, y) to $\Gamma(t)$, such that the positive sign is in the direction of positive φ (i.e., liquid) and vice versa. We define s(x, y, t) as a measure of arc length from some fixed point so that (r,s) is a local coordinate system. The interface $\Gamma(t)$ may be described as the set of points at which (x, y, t) vanishes. Furthermore, in a neighborhood of Γ one has¹⁷

$$|\nabla r| = 1, \quad \Delta r = \kappa . \tag{4.4}$$

We formally expand the variables in their original coordinates to obtain the *outer expansion* as

$$u(x,y,t,\epsilon_{1}) = u^{0}(x,y,t) + \epsilon_{1}u^{1}(x,y,t) + \epsilon_{1}^{2} \cdots + u^{2}(x,y,t) + \cdots, \qquad (4.5)$$

$$\varphi(x,y,t,\epsilon_1) = \varphi^0(x,y,t) + \epsilon_1 \varphi^1(x,y,t) + \epsilon_1^2 \cdots , \quad (4.6)$$

$$r(x,y,t,\epsilon_1) = r^0(x,y,t) + \epsilon_1 r^1(x,y,t) + \epsilon_1^2 \cdots$$
, (4.7)

$$s(x,y,t,\epsilon_1) = s^0(x,y,t) + \epsilon_1 s^1(x,y,t) + \epsilon_1^2 \cdots$$

$$+\varphi^{\underline{2}}(x,y,t)+\cdots \qquad (4.8)$$

5890

Note that the terms on the right-hand side of (4.5) and (4.6) may be discontinuous at r=0 but are smooth for $r\neq 0$.

By "stretching out" the variable r and defining a new variable

$$z = r/\epsilon_1 , \qquad (4.9)$$

we write the inner expansion

$$u(x,y,t,\epsilon_1) \equiv U(z,s,t,\epsilon_1)$$

= $U^0(z,s,t) + \epsilon_1 U^1(z,s,t) + \cdots$, (4.10)

$$\varphi(x, y, t, \epsilon_1) \equiv \phi(z, s, t, \epsilon_1)$$

= $\phi^0(z, s, t) + \epsilon_1 U^1(z, s, t) + \cdots$ (4.11)

The notation¹⁷ $f|_{\Gamma_{\pm}}$ means the limiting value of f as Γ is approached from r > 0 or r < 0, respectively.

The outer expansion. We obtain a sequence of equations (for $r \neq 0$) by substituting the expansions (4.5)-(4.8) into (4.2) and (4.3), as follows: For O(1),

$$u_t^0 + \frac{l}{2}\varphi_t^0 = K\Delta u^0 , \qquad (4.12)$$

$$f(\varphi^0) = 0$$
, (4.13)

for $O(\epsilon_1)$,

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$$u_t^{1} + \frac{l}{2}\varphi_t^{1} = K\Delta u^{1} , \qquad (4.14)$$

$$f'(\varphi^0)\varphi^1 + 2u^0 = 0 , \qquad (4.15)$$

and for $O(\epsilon_1^2)$,

$$u_t^2 + \frac{l}{2}\varphi_t^2 = K\Delta u_t^2 , \qquad (4.16)$$

$$\alpha \varphi_t^0 - \Delta \varphi^0 = f'(\varphi^0) \varphi^2 + \frac{1}{2} f''(\varphi^0) (\varphi^1)^2 + 2u^1 .$$
 (4.17)

The O(1) equations imply the following. Equation (4.13) has solutions ± 1 and 0. Note that this is true for both Dirichlet of Neumann boundary conditions. Hence, for $r \neq 0$, i.e., in the liquid or solid, Eq. (4.12) reduces to the heat equation (1.3). This is the first of three objectives.

The inner expansion. Using the (r,s) coordinate system one may write the Laplacian as

$$\Delta u = u_{rr} + \Delta r u_r + |\nabla s|^2 u_{ss} + \Delta s u_s$$

and the time derivative u_t as $u_t + r_t u_r + s_t u_s$. Scaling the r variable by use of the Eq. (4.9), one may write (4.2) and (4.3) as

$$KU_{zz} + \epsilon \left[-r_t U_z - \frac{l}{2} r_t \phi_z + K \Delta r U_z \right]$$
$$-\epsilon^2 \left[U_t + s_t U_s + \frac{l}{2} \phi_t + \frac{l}{2} s_t \phi_s + K(|\nabla s|^2 U_{ss} + \Delta s U_s) \right] = 0 , \quad (4.18)$$

$$\phi_{zz} + f(\phi) + 2\epsilon U - \epsilon \alpha r_t \phi_z + \epsilon \phi_z \Delta r + \epsilon^2 (|\nabla s|^2 \phi_{ss} + \Delta s \phi_s - \alpha \phi_t - \alpha s_t \phi_s) = 0 .$$
(4.19)

The inner expansion is obtained by matching formal orders in (4.18) and (4.19) as follows: For O(1),

$$U_{zz}^0 = 0$$
, (4.20)

$$\phi_{zz}^0 + f(\phi^0) = 0 . (4.21)$$

The solution to (4.20) is $U^0 = az + b$. The matching condition used in Ref. 17 implies [see Eq. (A10) in the Appendix]

$$U^{0}(\pm\infty,t) = u^{0}(\Gamma^{0}_{\pm},t) . \qquad (4.22)$$

This can only be satisfied if a = 0; otherwise u^0 would be unbounded at the interface. Hence, we conclude that

$$U^0 = b$$
, (4.23)

where b may depend on t or s but not on z. Using the same matching condition again, we have (for both Dirichlet and Neumann conditions on $\partial\Omega$)

$$\phi^{0}(\pm\infty,t) = \varphi^{0}(\Gamma^{0}_{\pm},t) = \pm 1 . \qquad (4.24)$$

By definition of the interface (2.5), one has $\phi(0,t)=0$ so that $\phi^0(0,t)=0$ (a contradiction would result otherwise). Hence, $\phi^0(z) \equiv \psi(z)$ is the unique solution of

$$\psi''(z) + f(\psi) = 0, \quad \psi(\pm \infty) = \pm 1, \quad \psi(0) = 0.$$
 (4.25)

The $O(\epsilon_1)$ balance in (4.18) implies

$$KU_{zz}^{1} = \frac{l}{2}r_{t}^{0}\phi_{z}^{0} = \frac{l}{2}r_{t}^{0}\psi_{z} , \qquad (4.26)$$

since $U_z^0 = 0$ by (4.23). Integrating (4.26), one obtains

$$KU_{z}^{1} = \frac{l}{2}r_{l}^{0}\psi(z) + c_{1}(s,t) . \qquad (4.27)$$

Using the matching condition¹⁷ [see Eq. (A12) in the Appendix] and differentiating with respect to z, one obtains

$$\lim_{z \to \pm \infty} U_z^1(z,t) = u_r^0(\Gamma_{\pm}^0,t) .$$
 (4.28)

Noting that the boundary conditions (4.25) at $\pm \infty$ used in (4.27) imply together with (4.28), the interface condition

$$Ku_r^0|_{\Gamma_{\pm}} = \pm \frac{l}{2}r_t^0 + c_1(s,t) . \qquad (4.29)$$

Since the normal velocity v (dropping the subscript) is given by $-r_t$, we may write $r_t^0 = -v^0$ in (4.29). Subtracting (4.29) with the minus sign from (4.29) with the plus sign, one has the latent heat condition to lowest order as

$$K[u_r^0]_{\Gamma_+} = -lv^0 . (4.30)$$

Having shown that the heat equation (1.3) and the latent heat condition (1.4) both arise as the lowest-order terms we now pursue the evaluation of the temperature at the interface.

The $O(\epsilon_1)$ terms for (4.19) are (recalling the sign of v^0)

$$L\phi^{1} \equiv \phi^{1}_{zz} + f'(\phi^{0})\phi^{1} = -2U^{0} - \alpha v^{0}\psi'(z) - \kappa^{0}\psi'(z) . \quad (4.31)$$

Since ψ' is a solution (and simple eigenvalue) to $L\psi'=0$ (with ψ' and ψ'' vanishing at $\pm \infty$) one has the solvability condition

$$\int_{-\infty}^{\infty} \psi'(z) [-2U^0 - \alpha v^0 \psi'(z) - \kappa^0 \psi'(z)] dz = 0 .$$
 (4.32)

Recalling that U^0 must be constant in z [by (4.23)], one has then

$$4U^{0} = (-\alpha v^{0} - \kappa^{0}) \int_{-\infty}^{\infty} [\psi'(z)]^{2} dz . \qquad (4.33)$$

The first matching condition applies again so that

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$$u^{0}(\Gamma^{0}_{\pm},t) = \left\lfloor \frac{-\alpha v^{0} - \kappa^{0}}{4} \right\rfloor \sigma ,$$

$$\sigma \equiv \int_{-\infty}^{\infty} [\psi'(z)]^{2} dz . \qquad (4.34)$$

We verify that σ as defined above agrees with the definition (3.7). Note that the free energy $\mathcal{F}{\varphi}$ which leads to (4.3) in the form $\tau \varphi_t = \delta \mathcal{F} / \delta \varphi$ is [with $G'(\varphi) \equiv 4g(\varphi)$]

$$\mathcal{F}\{\varphi\} = \int_{\Omega} d\mathbf{x} \left[\frac{\xi^2}{2} (\nabla \varphi)^2 + \frac{1}{4a} G(\varphi) - 2u\varphi \right], \quad (4.35)$$

and the integration trick used in (3.8) leads to

$$\frac{\xi^2}{2}(\varphi_r^0)^2 = \frac{1}{4a}G(\varphi) \ . \tag{4.36}$$

Let Ω' denote that portion of Ω which is within $\delta = \epsilon^p$ $(0 of the interface. Writing <math>\nabla \varphi$ in the (r,s) coordinates, and then transforming to z and utilizing (4.36), one obtains a local version of (4.35) near the interface,

12

$$\frac{\mathcal{F}\{\varphi\}}{A_{\Omega'}} = \int_{z \in \Omega'} dz \left[\left(\frac{d\varphi^0}{dz} \right)^2 + \epsilon_1 O(1) - 2u \left[\varphi^0 + \epsilon_1 \varphi^1 + O(\epsilon_1^2) \right] \right], \quad (4.37)$$

where $A_{\Omega'}$ is the area of Ω' and the O(1) terms involve derivatives of φ^0 with respect to z and s. Note that $\varphi^0 = \psi$ is symmetric, $\partial u / \partial z$ is $O(\epsilon_1)$ and that the integration of φ_z^0 can be extended to $(-\infty, +\infty)$ with an error of less than $O(\epsilon_1)$. Using (4.37) in a local interpretation of (3.7) (i.e., using Ω') one finds the surface tension σ is indeed given by the second equation in (4.34). Similarly, the entropy difference remains 4 using (3.11) with (4.35). Hence, the outer problem at the O(1) level is determined completely by Eqs. (1.3), (4.30), and (4.34), so that Proposition 4.1 has been verified.

V. THE ALTERNATIVE MODIFIED STEFAN LIMIT

We consider the same basic limit as in the previous section but allow α to approach zero in an arbitrary way. Physically this means that the (dimensionless) relaxation time τ is small in comparison with ξ^2 in (2.1) and (2.2).

Proposition 5.1. In the limit $\xi, a, \alpha \rightarrow 0$ with $\xi a^{-1/2}$ fixed, there exists a formal asymptotic solution of the phase-field model [(2.1) and (2.2)] which is governed by

We verify this by using the same formalism as in Sec. IV. We then have the following for the outer expansion and the inner expansion.

The outer expansion. For O(1),

$$u_t^0 + \frac{l}{2}\varphi_t^0 = K\Delta u^0 , \qquad (5.1)$$

$$f(\varphi^0) = 0$$
, (5.2)

for $O(\epsilon_1)$,

$$u_t^{1} + \frac{l}{2}\varphi_t^{1} = K\Delta u^{1} , \qquad (5.3)$$

$$f'(\varphi^0)\varphi^1 + 2u^0 = 0 , \qquad (5.4)$$

for $O(\epsilon_1^2)$,

$$u_t^2 + \frac{l}{2}\varphi_t^2 = K\Delta u^2 , \qquad (5.5)$$

$$-\Delta \varphi^{0} = f'(\varphi^{0})\varphi^{2} + \frac{1}{2}f''(\varphi^{0})(\varphi^{1})^{2} + 2u^{1}.$$
 (5.6)

We see that the outer expansion is identical to Sec. IV up through $O(\epsilon_1)$.

The inner expansion. For O(1),

$$U_{zz}^0 = 0$$
, (5.7)

$$\phi_{zz}^0 + f(\phi^0) = 0 , \qquad (5.8)$$

for $O(\epsilon_1)$,

$$KU_{zz}^{1} - r_{t}^{0}U_{z}^{0} - \frac{l}{2}r_{t}^{0}\phi_{z}^{0} - K\Delta rU_{z}^{0} = 0 , \qquad (5.9)$$

$$L\phi^{1} \equiv \phi_{zz}^{1} + f'(\phi^{0})\phi^{1} = -2U^{0} - \kappa^{0}\phi_{z}^{0} . \qquad (5.10)$$

The analysis of the O(1) outer expansion leads to the same conclusions as in Sec. IV, i.e., φ^0 is ± 1 or 0, and u^0 satisfied the heat equation (1.3). The O(1) inner expansion proceeds similarly and one obtains, again,

 $U^0 = b$ (b independent of z). (5.11)

Since the matching condition

$$\phi^{0}(\pm\infty,t) = \varphi^{0}(\Gamma^{0}_{\pm},t) = \pm 1$$
 (5.12)

and the O(1) inner balance (5.8) are identical, it follows that $\phi^0(z) = \psi(z)$ where ψ is the unique solution to (4.25).

Noting that the $O(\epsilon_1)$ inner balance equation (5.9) is identical to (4.26) (since $U_z^0=0$ again) and that the matching relation (4.28) is valid, one finds that the latent heat condition (4.30) remains valid.

There remains only the evaluation of the U^0 term in (5.11). Since $\phi_z^0(z) = \psi'(z)$ solves $L\psi' = 0$, ψ' must be orthogonal to the inhomogeneous term in (5.10) which now differs from (4.31). The first matching condition still applies and one obtains the result

$$u^{0}(\Gamma^{0}_{\pm},t) = \frac{-\kappa^{0}}{\Delta s} \sigma \equiv \frac{-\kappa^{0}}{\Delta s} \int_{-\infty}^{\infty} [\psi'(z)]^{2} dz \quad (5.13)$$

This completes the argument for Proposition 5.1.

VI. THE CLASSICAL STEFAN LIMIT

The calculations of Sec. III suggest the following.

Proposition 6.1. In the limit $\xi, a, \xi a^{-1/2} \rightarrow 0$, there exists a formal asymptotic solution of the phase-field model [(2.1) and (2.2)] which is governed by the classical Stefan model [(1.3), (1.4), and (1.5)]. We also note that there is more flexibility in the scaling of ξ and a within this limit since the same O(1) condition at the interface can be expected when $\xi a^{-1/2} = \epsilon a^{-1}$ (which is proportional to the surface tension) is of any order smaller than O(1). We choose a particular scaling in order to make the calculations explicit (in one parameter) and easily manageable in other calculations. We let

$$\alpha = \text{fixed}, \quad a \equiv \xi c_0^{-2}, \quad \xi \to 0$$
 (6.1)

Multiplying (2.2) by ξ and setting $\overline{\epsilon}^2 = \xi$ we may rewrite the system (2.1) and (2.2) as

$$u_t + \frac{l}{2}\varphi_t = K\Delta u \quad , \tag{6.2}$$

$$\alpha \overline{\epsilon}^{\,6} \varphi_t = \overline{\epsilon}^{\,6} \Delta \varphi + f(\varphi) + 2u \overline{\epsilon}^{\,2} , \qquad (6.3)$$

where $f(\varphi) = c_0^2 g(\varphi)$. We use the same outer expansion (4.5)-(4.8) now in $\overline{\epsilon}$, and an inner expansion similar to (4.10), (4.11) except that in this case, the "stretched variable" \overline{z} is defined by

$$\overline{z} = r/\overline{\epsilon}^3 . \tag{6.4}$$

In general, the matching relations for this case differ from the previous¹⁷ due to the cubic in (6.4). However, a simple generalization of these relations is derived in the Appendix.

By formally equating powers of $\overline{\epsilon}$, one obtains the outer expansion: For O(1),

$$u_{t}^{0} + \frac{l}{2}\varphi_{t}^{0} = K\Delta u^{0} , \qquad (6.5)$$

$$f(\varphi^0) = 0$$
, (6.6)

and for $O(\overline{\epsilon})$,

$$u_t^{1} + \frac{l}{2} \varphi_t^{1} = K \Delta u^{1} , \qquad (6.7)$$

$$f'(\varphi^0)\varphi^1 = 0$$
 . (6.8)

The O(1) balance is the same as in Sec. IV, so that one obtains the heat equation (1.3) in each region and φ^0 is ± 1 or 0. In the $O(\bar{\epsilon})$ balance above one obtains

$$\varphi^1 = 0 , \qquad (6.9)$$

$$u_t^1 = K \Delta u^1 . \tag{6.10}$$

Using the coordinate system (\overline{z}, s) we write

$$U(\overline{z},s,t,\overline{\epsilon}) \equiv u(x,y,t,\overline{\epsilon}) , \qquad (6.11)$$

$$\phi(z,s,t,\epsilon) \equiv \varphi(x,y,t,\epsilon)$$
,

so that Eqs. (6.2) and (6.3) are transformed into

$$KU_{zz} + \overline{\epsilon}^{3} \left[-r_{t}U_{z} - \frac{l}{2}r_{t}\phi_{z} + K\Delta rU_{z} \right]$$
$$-\overline{\epsilon}^{6} \left[U_{t} + U_{s}s_{t} + \frac{l}{2}\phi_{t} + \frac{l}{2}\phi_{s}s_{t} + K(U_{ss}|\nabla s|^{2} + U_{s}\Delta s) \right] = 0,$$
(6.12)

$$\phi_{zz} + f(\phi), + 2\overline{\epsilon}^2 U - \overline{\epsilon}^3 (\alpha r_i \phi_z + \Delta r \phi_z)$$

$$+\overline{\epsilon}^{\circ}(|\nabla_{s}|^{2}\phi_{ss}+\Delta s\phi_{s}-\alpha\phi_{t}-\alpha\phi_{s}s_{t})=0. \quad (6.13)$$

This yields the inner expansion: For O(1),

$$U_{zz}^{0} = 0$$
, (6.14)

$$\phi_{zz}^0 + f(\phi^0) = 0 , \qquad (6.15)$$

for $O(\overline{\epsilon})$,

$$U_{zz}^1 = 0$$
, (6.16)

$$L\phi^1 \equiv \phi_{zz}^1 + f'(\phi^0)\phi^1 \equiv 0$$
, (6.17)

and for $O(\overline{\epsilon}^2)$,

$$U_{zz}^2 = 0$$
, (6.18)

$$L\phi^{2} \equiv \phi_{zz}^{2} + f'(\phi^{0})\phi^{2} = -f''(\phi^{0})(\phi^{1})^{2}/2 - 2U^{0}$$

=0. (6.19)

We note that the first matching condition used in (4.22) remains valid in this scaling also, so that (6.14) once again has the solution

$$U^0 = b$$
 (b independent of z). (6.20)

Since Eq. (6.15) is identical to (4.21), the first matching relation is unchanged, leading to the same boundary conditions, one concludes that $\phi^0(z) = \psi(z)$ defined by (4.25).

Next, we proceed to evaluate the constant b. Since φ^0 and φ^1 are constant (for $r \neq 0$) and one has

$$\lim_{r \to 0^{\pm}} \frac{\partial}{\partial r} \varphi^{0}(r, s, t) = 0 ,$$

$$\lim_{\bar{z} \to +\infty} \varphi^{1}(\bar{z}, s, t) = 0 .$$
(6.21)

Applying the second matching relation (A11), which differs from the previous version, one has

$$\lim_{z \to \pm \infty} \phi^{1}(z,s,t) = 0 . \qquad (6.22)$$

Thus, Eq. (6.17) with boundary condition (6.22) has the unique (symmetric) solution

$$\phi^1 = \psi' \quad . \tag{6.23}$$

To proceed further, we study the $O(\bar{\epsilon}^2)$ equation (6.19). Since ψ^1 solves the homogeneous equation (6.17) one obtains the solvability condition for (6.19) as

$$\int_{-\infty}^{\infty} \left[-f''[\psi(\overline{z})] \frac{[\psi'(\overline{z})]^2}{2} - 2b \right] \psi'(\overline{z}) d\overline{z} = 0$$

Noting that f, f'' are odd while f', ψ' are even, the first

term vanishes, leading to the result

$$0 = \int_{-\infty}^{\infty} 2b \, \psi'(z) dz = 4b \quad . \tag{6.24}$$

Using (6.20) and the first matching relation (4.22), one has

$$u^{0}|_{\Gamma_{\pm}} = 0$$
 . (6.25)

Finally, we need to obtain the latent heat condition. One can do this by proceeding to third order in $\overline{\epsilon}^3$, since the $lr_t^0/2$ term arises as an $\overline{\epsilon}^3$ term in (6.12). We take the following alternative approach which is more illuminating and less tedious. If the asymptotic methods are valid, then there exists \overline{u} and $\overline{\varphi}$ (obtained from the inner and outer expansions) which satisfies

$$|\bar{u}-u| < C\bar{\epsilon}, \quad |\bar{\varphi}-\varphi| < C\bar{\epsilon}, \quad (6.26)$$

where (u, φ) is the true solution which we know (rigorously) exists.⁹ The function $\overline{\varphi}$ must be ψ (to this order) since it satisfies these requirements on both the inner and the outer regions.

Using the (r,s) moving coordinate system, the Eq. (2.1) for $(\overline{u},\overline{\varphi})$ can be written as

$$\overline{u}_{t} + \overline{u}_{r}r_{t} + \overline{u}_{s}s_{t} + \frac{l}{2}(\overline{\varphi}_{t} + \overline{\varphi}_{r}r_{t} + \overline{\varphi}_{s}s_{t})$$

$$= K(\overline{u}_{rr} + \overline{u}_{ss}|\nabla s|^{2} + \overline{u}_{r}\Delta r + \overline{u}_{s}\Delta s) . \quad (6.27)$$

From the information [(6.20) and (6.24)] about the inner solution, we know that

$$\lim_{r \to 0\pm} \overline{u}(r,s,t) = 0, \quad \lim_{\delta \to 0} \int_{-\delta}^{\delta} |\overline{u}_t(r,s,t)| dt = 0$$
 (6.28)

and similarly for the s derivatives. If we let $\delta \equiv \overline{\epsilon}^{p}$ for any $p \in (0,1)$ then integrating (6.27) in r one obtains

$$K \int_{-\delta}^{\delta} \overline{u}_{rr} dr = \frac{l}{2} r_t^0 \int_{-\delta}^{\delta} \psi_r dr . \qquad (6.29)$$

Note that (6.27) also implies directly that

$$K\bar{u}_{rr} = \frac{l}{2}r_{t}^{0}\psi_{r} + O(1) = \frac{l}{2}r_{t}^{0}\frac{c^{0}}{\bar{\epsilon}} + O(1) , \qquad (6.30)$$

for some $c_0 \in \mathbb{R}^+$, i.e., that \overline{u}_r must have a jump in the limit as $\overline{\epsilon}$ approaches zero. From (6.29) and the antisymmetry of ψ one has

$$K\bar{u}_{r}|_{-\delta}^{\delta} = lr^{0}\psi(\delta) + o(1) = -lv^{0} + o(1) . \qquad (6.31)$$

In terms of the outer expansion, then, one has the latent heat condition,

$$Ku_r^{0|+} = -lv^0$$
 on Γ , i.e., $r = 0$. (6.32)

This completes the analysis of the Stefan limit.

VII. HELE-SHAW-TYPE PROBLEMS

In studying equations that approximate the pressure in a system containing two immiscible fluids, one may use the following system of equations:²³

$$\Delta u = 0 \quad \text{in } \Omega_1, \Omega_2 , \qquad (7.1)$$

$$[\nabla u]_{-}^{+} = -\frac{l}{K}v \quad \text{on } \Gamma , \qquad (7.2)$$

$$u = \left[\frac{-\alpha v - \kappa}{4}\right] \sigma \quad \text{on } \Gamma , \qquad (7.3)$$

subject to suitable initial and boundary conditions, e.g., (1.6) and (1.7). Here u is the pressure instead of temperature and the other physical constants l, K, α have a different meaning,²³ while σ is the interfacial tension between the fluids. We consider the two cases, $\alpha > 0$ and $\alpha = 0$.

Proposition 7.1. In the formal limit $\xi \to 0$ with $a \equiv \xi^2 c_1^{-2}$, $K \equiv \xi^{-2}$, $l \equiv \xi^{-2} c_2^{-2}$, and α, c_1 fixed, there exists a formal asymptotic solution of the phase-field model [(2.1), (2.2)] which is governed by the Hele-Shaw model [(7.1)-(7.3)]. Moreover, if $\alpha \to 0$ also, then the same statement applies with $\alpha = 0$ in (7.3).

Using the parameter $\epsilon_h = \xi^2$ we rewrite (2.1) and (2.2) as

$$\epsilon_h u_l + \frac{c_2^2}{2} \varphi_l = \Delta u \quad , \tag{7.4}$$

$$\alpha \epsilon_h^2 \varphi_t = \epsilon_h^2 \Delta \varphi + f(\varphi) + 2u \epsilon_h , \qquad (7.5)$$

with f = g. Using the same formalism as in Sec. IV, we have the outer expansion: For O(1),

$$\frac{c_2^2}{2}\varphi_t^0 = \Delta u^0 , \qquad (7.6)$$

$$f(\varphi^0) = 0$$
, (7.7)

for $O(\epsilon_h)$,

$$u_t^0 + \frac{c_2^2}{2} \varphi_t^1 = \Delta u^1 , \qquad (7.8)$$

$$f'(\varphi^0)\varphi^1 + 2u^0 = 0 , \qquad (7.9)$$

and for $O(\epsilon_h^2)$,

f

$$u_t^{\ 1} + \frac{c_2^2}{2} \varphi_t^2 = \Delta u^2 , \qquad (7.10)$$

$$f'(\varphi^0)\varphi^2 + 2u^1 = -\frac{1}{2}f''(\varphi^0)(\varphi^1)^2 + \alpha\varphi_l^0 - \Delta\varphi^0$$
. (7.11)

Equations (7.4) and (7.5) can be rewritten in the (z,s) $[z \equiv r/\epsilon_h$ analogously with (4.9)] moving coordinate system and inner variables as

$$U_{zz} + \epsilon_{h} (-c_{2}^{2}r_{t}\phi_{z} + K\Delta rU_{z}) -\epsilon_{h}^{2} \left[r_{t}U_{z} + \frac{c_{2}^{2}}{2}\phi_{t} + \frac{c_{2}^{2}}{2}\phi_{s}s_{t} + (U_{ss}|\nabla s|^{2} + U_{s}\Delta s) \right] +\epsilon_{h}^{3} (U_{t} + U_{s}s_{t}) = 0 , \quad (7.12)$$

$$\phi_{zz} + f(\phi) + 2\epsilon U - \epsilon_h \alpha r_t \phi_z + \epsilon_h \phi_z \Delta r + \epsilon_h^2 (\phi_{ss} |\nabla s|^2 + \phi_s \Delta_s - \alpha \phi_t - \alpha \phi_s s_t) = 0.$$
(7.13)

Matching formal orders in (7.12) and (7.13) we have the inner expansion: For O(1),

$$U_{zz}^0 = 0$$
 , (7.14)

$$\phi_{zz}^{0} + f(\phi^{0}) = 0 , \qquad (7.15)$$

and for $O(\epsilon_h)$,

$$U_{zz}^{1} - \frac{c_{z}^{2}}{2} r_{t}^{0} \phi_{z}^{0} + \Delta r^{0} U_{z}^{0} = 0 , \qquad (7.16)$$

$$\phi_{zz}^{1} + f'(\phi^{0})\phi^{1} + 2U^{0} - \alpha r_{t}^{0}\phi_{z}^{0} + \Delta r^{0}\phi_{z}^{0} = 0 . \qquad (7.17)$$

The O(1) outer expansion has solutions $\varphi^0 = \pm 1$ or 0 and

$$\Delta u^0 = 0 \quad (r \neq 0) \; . \tag{7.18}$$

The inner balance has the same terms as in the modified Stefan model (Sec. IV) and so

$$\phi^0 = \psi$$
 [defined by (4.25)],
 $U^0 = b$ (independent of z). (7.19)

The $O(\epsilon_h)$ inner balance can then be written as

$$U_{zz}^{1} = \frac{c_{2}^{2}}{2} r_{t}^{0} \psi' . \qquad (7.20)$$

Since (7.20) is identical to (4.26) and the first matching relation (4.22) also applies, one obtains (4.30) in an identical way, i.e.,

$$[u_r^0]_{-}^{+} = -\frac{l}{K}v^0 = -c_2^2 v^0 . \qquad (7.21)$$

The temperature at the interface $u^0|_{\Gamma_{\pm}}$ is derived from the second matching relation and (7.15) and (7.17) in the same manner as in Sec. IV so that

$$u^{0}|_{\Gamma_{\pm}} = \left[\frac{-\alpha v^{0} - \kappa^{0}}{4} \right] \int_{-\infty}^{\infty} [\psi'(z)]^{2} dz$$

$$= \left[\frac{-\alpha v^{0} - \kappa^{0}}{4} \right] \sigma .$$

$$(7.22)$$

Hence, (7.18), (7.21), and (7.22) complete the verification of Proposition 7.1 for $\alpha > 0$. In order to obtain (7.3) in the more common form with $\alpha = 0$, one can implement the same idea as in Sec. V combined with the scaling of ξ and a as above.

For simplicity, we have considered the phase-field equations without anisotropy. As discussed in Ref. 11, microscopic anisotropy modifies the phase-field equations. In particular, the temperature relation at the interface is different. The methods of this paper can be applied in the same way. For x - y anisotropy in two dimensions, for example, the result of Proposition 4.1 is then valid with

$$\Delta s \left[u(t,x) \right] = -(\sigma + \sigma'') \kappa(t,x) , \qquad (7.23)$$

where the primes denote derivatives with respect to orientation angle. The velocity terms in Secs. IV and VII are also altered with an angle-dependent term. The role of anisotropy has also been discussed in other approaches to solidification and pattern development (see Refs. 24-26).

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APPENDIX: MATCHING RELATIONS FOR INTERNAL LAYERS OF THICKNESS ϵ^k

We apply standard asymptotic theory to an expansion of an arbitrary function $w(t, x, \epsilon)$ in powers of ϵ but with layers of width ϵ^k $(k \ge 2)$. This is a minor generalization of the case¹⁷ in which the layers are also of width ϵ . We let $Y(t, \epsilon)$ represent the value of x for which w = 0, and set

$$z \equiv \frac{x - Y}{\epsilon^k} \quad . \tag{A1}$$

In addition to the outer expansion

$$w(x,t,\epsilon) = w^{0}(x,t) + \epsilon w^{1}(x,t) + \epsilon^{2} w^{2}(x,t) + \cdots,$$
(A2)

one defines the inner variable W by

$$W(z,t,\epsilon) \equiv w(x,t,\epsilon)$$
, (A3)

and assume the expansions

$$W(z,t,\epsilon) = W^{0}(z,t) + \epsilon W^{1}(z,t) + \cdots, \qquad (A4)$$

$$Y(t,\epsilon) = Y^{0}(t) + \epsilon Y^{1}(t) + \cdots$$
 (A5)

Using (A1) and (A3) one has the basic relation

$$W(z,t,\epsilon) = w(Y(t,\epsilon) + \epsilon^{k}z,t,\epsilon) . \qquad (A6)$$

Expanding the right-hand side in a series in ϵ as in Ref. 17 but noting the ϵ^k rather than ϵ in (A6), one obtains

$$W(z,t,\epsilon) = \sum_{n=0}^{N} \epsilon^{n} P_{n}(z,t) + \epsilon^{N+1} R_{N} , \qquad (A7)$$

$$P_{n}(z,t) \equiv \frac{1}{n!} \frac{\partial^{n}}{\partial \epsilon^{n}} w(Y(t,\epsilon) + \epsilon^{k} z, t, \epsilon) \big|_{\epsilon=0} , \qquad (A8)$$

where R_N is identical to P_N except that *n* is replaced by N+1 and R_N is evaluated at some $\hat{\epsilon} \in [0, \epsilon]$.

The matching is accomplished in the usual way by letting $\epsilon \rightarrow 0$ and $z \rightarrow 0$ in an arbitrary way provided $\epsilon z^{(n+1)/2} \rightarrow 0$. Since it follows that $\epsilon^k z \rightarrow 0$ one has, e.g.,

$$P_{0}(t,z) = w^{0}(Y^{0}(t) + \epsilon Y^{0}(t) + \epsilon^{2} Y^{2}(t) + \cdots + \epsilon^{k} z, t)|_{\epsilon=0}$$

= $w^{0}(Y^{0}_{\pm}(t), t)$, (A9)

where \pm denoted the approach from either side.

The first two matching conditions are then

$$\lim_{z \to \pm \infty} W^0(z,t) = w^0(Y^0_{\pm},t) , \qquad (A10)$$

$$\lim_{z \to \pm \infty} W^{1}(z,t) = w_{x}^{0}(Y_{\pm}^{0}(t),t)Y^{1}(t) + w^{1}(Y_{\pm}^{0}(t),t) .$$
(A11)

$$\lim_{z \to \pm \infty} \left[\frac{W^{1}(z,t) - \{ w^{1}(Y^{0}_{\pm},t) + [z + Y^{1}(t)] w^{0}_{x}(Y^{0}_{\pm},t) \}}{z} \right] = 0.$$
 (A12)

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