

The Cahn-Hilliard equation with a concentration dependent mobility: motion by minus the Laplacian of the mean curvature.

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Abstract

We show by using formal asymptotics that the zero level set of the solution to the Cahn-Hilliard equation with a concentration dependent mobility approximates to lowest order in ϵ an interface evolving according to the geometric motion,

$$V = -\frac{\pi^2}{16}\Delta_s\kappa, \quad (0.1)$$

where V is the normal velocity, Δ_s is the surface Laplacian and κ is the mean curvature of the interface, both in the deep quench limit and when the temperature θ is $\mathcal{O}(\epsilon^\alpha)$, $\alpha > 0$ where ϵ^2 is the coefficient of gradient energy. Equation (0.1) may be viewed as motion by surface diffusion, and as a higher order analogue of motion by mean curvature predicted by the bistable reaction-diffusion equation.

1 Introduction.

The Cahn-Hilliard equation

$$u_t = -\nabla \cdot \mathbf{J} \quad (1.1a)$$

$$\mathbf{J} = -M(u)\nabla w \text{ and } w = -\gamma\Delta u + \Psi'(u) \quad (1.1b)$$

arises as a phenomenological model for isothermal phase separation in a binary alloy, see Cahn [6],[7] and Hilliard [17] for a derivation, [23, 14, 21, 31] for general analysis, and the reviews given in [10, 22]. Here u is the difference in the mass fraction of the two components of the alloy, hence $-1 \leq u \leq 1$ and the extreme values ± 1 correspond to the pure components. The mass flux is \mathbf{J} and w is a generalized chemical potential. The homogeneous free energy for a mean field model of a mixture at a fixed (scaled) absolute temperature is

$$\Psi(u) = \frac{\theta}{2}\{(1+u)\ln(1+u) + (1-u)\ln(1-u)\} + \frac{1}{2}(1-u^2). \quad (1.2)$$

Equations (1.1) hold for $(x, t) \in \Omega \times (0, T]$ where Ω is a bounded open domain in \mathbb{R}^n ($n = 1, 2, 3$). As considerable work has been done on the one-dimensional case [24, 15, 2], and since it is only in the context of higher dimensions in which geometric motion of the interface is seen, we shall restrict our attention to \mathbb{R}^n ($n = 2, 3$). We supplement (1.1) with no-flux and Neumann boundary conditions

$$\mathbf{n} \cdot \mathbf{J} = \mathbf{n} \cdot M(u)\nabla w = 0 \quad (1.3a)$$

$$\mathbf{n} \cdot \nabla u = 0 \quad (1.3b)$$

on $\partial\Omega$, and thus mass is conserved

$$\frac{d}{dt} \int_{\Omega} u = 0. \quad (1.4)$$

For θ below a critical temperature $\theta_{crit} = 1$, Ψ has the form of a double well potential with absolute minima at the binodal values $u_+ = -u_- = \beta(\theta)$ where $\beta(\theta)$ is the (unique) positive root of

$$\frac{2}{\theta} = \frac{\phi(\beta)}{\beta} \quad (1.5)$$

where $\phi(r) \equiv \ln(1+r) - \ln(1-r)$.

The non-negative function $M(\cdot)$ is the mobility, which we take to be

$$M(u) = 1 - u^2, \quad (1.6)$$

and remark that the concentration dependence in the mobility appeared in fact in the derivation [6, 17]. See also [8, 30]. We further remark that in many physical situations atom movement is confined to the interface region. Interface or surface diffusion dominates when the mobile species are found only at the interface, either because there is little of the required disorder in the abutting phases, or the density of mobile species is very low, as it might be in a vapor or poor-solvent phase. We note that existence of a weak solution to (1.1), (1.2) and (1.6) has been recently proved in [11].

The purpose of this paper is to employ formal asymptotics to derive an equation of motion for the interfaces separating the $\pm\beta(\theta)$ phases of the alloy. In particular, by assuming the temperature θ to satisfy $\theta(\epsilon) = \mathcal{O}(\epsilon^\alpha)$, $\alpha > 0$, and by rescaling the time $t \rightarrow \epsilon^2 t$, and setting $\gamma = \epsilon^2$, we show by formal asymptotics that as $\epsilon \rightarrow 0$, Ω has the decomposition

$$\Omega = \Omega_\epsilon^+(t) \cup \Omega_\epsilon^I(t) \cup \Omega_\epsilon^-(t)$$

such that

$$u \approx \beta(\theta) \text{ for } x \in \Omega_\epsilon^+(t) \quad \text{and} \quad u \approx -\beta(\theta) \text{ for } x \in \Omega_\epsilon^-(t)$$

and $\Omega_\epsilon^I(t)$ corresponds to a narrow interfacial region of thickness ϵ which contains $\Gamma_\epsilon(t)$, the zero level surface of u . Furthermore, $\Gamma_\epsilon(t)$ approximates a surface $\Gamma(t)$ which evolves according to the equation of motion

$$V = -\frac{\pi^2}{16} \Delta_s \kappa. \quad (1.7)$$

Here V is the normal velocity of $\Gamma(t)$, κ is the mean curvature of $\Gamma(t)$ and Δ_s is the surface Laplacian. This equation was first proposed by Mullins for the motion of the surface of a crystal when all mass transport is by curvature driven diffusion along the surface, and has recently been examined in a more general mathematical and physical context, [20, 8, 30, 9, 12]. In the case of the evolution of closed curves in the plane, local existence for (1.7) has been proven and results for the global behaviour when the initial data is close to a circle have been obtained, [12].

This equation of motion is in contrast to the case in which $M(u) = 1$ and the temperature θ is independent of ϵ , where it has been shown first by formal asymptotics (Pego [25]) and later analytically by Alikakos, Bates and Chen [1] (in the case of that $\Psi(\cdot)$ is a smooth potential, e.g. $\Psi(u) = \frac{1}{4}(u^2 - a^2)^2$) that on a time scale $\tau = \epsilon t$ the motion of $\Gamma_\epsilon(t)$ is determined by the Mullins-Sekerka free boundary problem

$$\Delta w = 0 \quad \text{for } x \in \Omega \setminus \Gamma_\epsilon, \quad V = -[\mathbf{n} \cdot \nabla w]_-^+ \quad \text{and } w|_{\Gamma_\epsilon} = \kappa \quad \text{for } x \in \Gamma_\epsilon \quad (1.8)$$

where $[\cdot]_-^+$ denotes the jump across Γ_ϵ .

Our equation (1.7) gives rise to a (local) geometric motion and can be compared to motion by mean curvature

$$V = \alpha \kappa \quad (1.9)$$

which has been derived by Rubinstein, Sternberg and Keller [27] as a limit of the Allen-Cahn equation [3],

$$u_t = -\alpha w \quad w = -\gamma \Delta u + \Psi'(u). \quad (1.10)$$

As there is a rich literature on the A-C equation and motion by mean curvature, we refer the reader to the reviews [29, 16], and the references contained therein. Here we wish only to remark that both equations (1.7) and (1.9) are curve shortening (as is the motion prescribed by the Mullins-Sekerka free boundary problem (1.8)), however (1.7) is mass conserving, while (1.9) is not.

The outline of this paper is as follows: Section §2 presents a number of preliminary properties of the Cahn-Hilliard equation. Afterwards, in Section §3 it is demonstrated that equation (1.7) may be obtained by formal asymptotics in the deep quench limit, and in Section §4, the equation (0.1) is again obtained to lowest order in the case in which $\theta = \mathcal{O}(\epsilon^\alpha)$, $\alpha > 0$. After a few short summary remarks in Section §5, an Appendix is given in Section §6 in which a derivation for the variables needed for the inner expansion is presented.

2 Preliminaries.

The system (1.1) with boundary conditions (1.3) give rise to an initial value problem for the degenerate parabolic equation

$$u_t = \nabla \cdot \{M(u) \nabla \{-\gamma \Delta u + \Psi'(u)\}\}. \quad (2.1)$$

Solutions of (2.1) satisfy the energy equation

$$\int_{\Omega} M(u) |\nabla w|^2 dx + \frac{d}{dt} \mathcal{E}(u) = 0, \quad (2.2)$$

where \mathcal{E} is the gradient energy functional

$$\mathcal{E}(u) \equiv \int_{\Omega} \left\{ \frac{\gamma}{2} |\nabla u|^2 + \Psi(u) \right\} dx. \quad (2.3)$$

It follows that solutions for large time should approach critical points of the gradient energy functional that satisfy

$$-\gamma \Delta u + \Psi'(u) = w \quad x \in \Omega, \quad \mathbf{n} \cdot \nabla u = 0 \quad x \in \partial\Omega \quad (2.4)$$

$$\int_{\Omega} u \, dx = \int_{\Omega} u_0 \, dx = m, \quad w = \frac{1}{|\Omega|} \int_{\Omega} \Psi'(u) \, dx.$$

As non-minimizing critical points can be shown to be linearly unstable, solutions can be expected to approach minimizers of \mathcal{E} . In the limit $\epsilon \rightarrow 0$, minimizers can be characterized as satisfying $\Psi'(u) = 0$ *a.e.*, a minimal perimeter condition for the interface, and

$$w = \frac{1}{2} \epsilon \kappa \int_{-1}^1 \Psi^{1/2}(u) \, du + o(\epsilon), \quad (2.5)$$

see [18, 19, 28, 4]. We assume in this paper that at long times, our solutions should roughly mimic the characteristic features of minimizers.

In Section §4, we assume that $\gamma = \epsilon^2$ and that $\theta = \theta(\epsilon) = \mathcal{O}(\epsilon^\alpha)$, $\alpha > 0$, and we study the long time approach to layered steady solutions of (2.1) which satisfy

$$u = \begin{cases} \beta_\epsilon & x \in \Omega_\epsilon^+ \\ -\beta_\epsilon & x \in \Omega_\epsilon^- \end{cases}, \quad (2.6a)$$

and

$$\Omega = \Omega_\epsilon^+ \cup \Omega_\epsilon^I \cup \Omega_\epsilon^- \quad (2.6b)$$

where Ω_ϵ^I has a narrow thickness $\sigma(\epsilon) = \mathcal{O}(\epsilon)$. Here $\beta_\epsilon = \beta_\epsilon(\theta)$ is the positive root of (1.5) and satisfies

$$\beta_\epsilon = 1 - T.S.T..$$

(The terminology *T.S.T.* stands for transcendentally small terms in ϵ .) Hence, in our asymptotics for this case, we assume the decomposition (2.6b) to be valid, (2.1) to hold throughout Ω , and (2.6a) to hold at least through the first few orders in the outer region; i.e.

$$u(x, t) = \pm \beta_\epsilon + \mathcal{O}(\epsilon^m) \quad \text{for } x \in \Omega_\epsilon^+ \cup \Omega_\epsilon^- \quad (2.7)$$

for some $m \geq 3$. Moreover, we find that $w = \mathcal{O}(\epsilon)$ throughout Ω , and in Ω_ϵ^I , we obtain results supporting the predictions of (2.5).

For fixed ϵ we call the limit $\theta = 0$ in (1.1) the deep quench limit. In the case $M(u) = 1$, it was shown by Elliott and Luckhaus [13] that one recovers in the limit the problem:

$$u_t = \nabla \cdot \{M(u) \nabla w\} \quad (2.8a)$$

$$w + \gamma \Delta u + u \in \partial I_{[-1,1]}(u) \quad (2.8b)$$

which was studied by Blowey and Elliott, [4, 5]. Here $\partial I_{[-1,1]}(\cdot)$ is the subdifferential of the indicator function $I_{[-1,1]}$ for the interval $[-1,1]$. The structure of solutions to (2.8) is such that $u \in C^1(\Omega)$, Ω is decomposed into $\Omega = \Omega_\epsilon^+(t) \cup \Omega_\epsilon^I(t) \cup \Omega_\epsilon^-(t)$ and

$$|u| < 1, \quad w = -\gamma \Delta u - u \quad x \in \Omega_\epsilon^I(t) \quad (2.9)$$

$$u \equiv \pm 1 \quad x \in \Omega_\epsilon^\pm(t). \quad (2.10)$$

Equation (2.8a) holds everywhere in Ω . In Section §3, we shall assume the existence of a deep quench limit to (1.1) and (1.3) with $M(u) = 1 - u^2$. In this instance, equation (2.10) is an ansatz in $\Omega_\epsilon^+ \cup \Omega_\epsilon^-(t)$ and (2.8a) and (2.9) hold in $\Omega_\epsilon^I(t)$.

3 Interfacial motion in the deep quench limit.

In this section we consider the derivation of an equation for the interface motion in the context of the deep quench limit problem (2.8). Thus we suppose that there exists an annular domain $\Omega_\epsilon^I(t)$ with outer boundary $\Gamma_\epsilon^+(t)$ and inner boundary $\Gamma_\epsilon^-(t)$. The domain is decomposed such that

$$\Omega = \Omega_\epsilon^+(t) \cup \Omega_\epsilon^I(t) \cup \Omega_\epsilon^-(t), \quad (3.1)$$

$$\partial\Omega_\epsilon^+(t) = \partial\Omega \cup \Gamma_\epsilon^+(t), \quad \partial\Omega_\epsilon^-(t) = \partial\Gamma_\epsilon^-,$$

where

$$u(x, t) = \pm 1 \quad x \in \Omega_\epsilon^\pm(t) \quad (3.2a)$$

$$\epsilon^2 u_t = \nabla \cdot (M(u) \nabla w) \quad x \in \Omega_\epsilon^I(t) \quad (3.2b)$$

$$w(t) = -\epsilon^2 \Delta u - u \quad x \in \Omega_\epsilon^I(t) \quad (3.2c)$$

$$\lim_{x \rightarrow \Gamma_\epsilon^\pm(t)} \frac{\partial u}{\partial n} = 0 \quad (3.2d)$$

$$\lim_{x \rightarrow \Gamma_\epsilon^\pm(t)} M(u) \frac{\partial w}{\partial n} = 0. \quad (3.2e)$$

We assume that there is a unique smooth zero level surface of u and denote it by $\Gamma^\epsilon(t) = \{x = \phi_\epsilon(s, t), s \in S\}$, see Appendix. The coordinate transformation described in the Appendix is employed with the scaling

$$\rho = r/\epsilon \quad (3.3)$$

where r is the variable normal to $\Gamma^\epsilon(t)$.

It is assumed that there exist expansions

$$u(x, t; \epsilon) = U(\rho, s, t; \epsilon) = U^0 + \epsilon U^1 + \epsilon^2 U^2 + \mathcal{O}(\epsilon^3) \quad (3.4a)$$

$$w(x, t, \epsilon) = W(\rho, s, t; \epsilon) = \epsilon W^1 + \epsilon^2 W^2 + \epsilon^3 W^3 + \mathcal{O}(\epsilon^4) \quad (3.4b)$$

$$\phi_\epsilon(s, t) = \phi^0 + \epsilon \phi^1 + \epsilon^2 \phi^2 + \mathcal{O}(\epsilon^3). \quad (3.4c)$$

From (6.3) it follows that if $\psi(x, t) = \Psi(\rho, s, t)$ then

$$\frac{\partial \psi}{\partial t} = \Psi_t + \Psi_s^T \frac{\partial s}{\partial t} + \frac{1}{\epsilon} \Psi_\rho \frac{\partial d}{\partial t}$$

$$\begin{aligned}\nabla_x \psi &= (J_x s) \Psi_s + \frac{1}{\epsilon} \Psi_\rho \nabla_x d \\ \Delta_x \psi &= \Delta_s \Psi + \frac{1}{\epsilon} \Psi_\rho \Delta_x d + \frac{1}{\epsilon^2} \Psi_{\rho\rho}.\end{aligned}$$

Furthermore it follows from (6.2) that

$$\begin{aligned}-\frac{\partial d^\epsilon}{\partial t} &= V^0 + \epsilon V^1 + \mathcal{O}(\epsilon^2) \\ \Delta_x d^\epsilon &= \kappa^0 + \epsilon \kappa^1 + \mathcal{O}(\epsilon^2) \\ \nabla_x d^\epsilon &= \mathbf{n}_0\end{aligned}$$

where V^0 , κ_0 , and \mathbf{n}_0 are respectively the normal velocity, mean curvature of and unit normal to $\Gamma^\epsilon(t)$. We recall that $(J_x s)^T \nabla_x d = 0$.

We proceed to derive expansions for the equations (3.2 b,c) written as

$$\epsilon^3 U_t = \epsilon \nabla \cdot (M(U) \nabla W) \quad (3.6a)$$

$$W = -\epsilon^2 \Delta U - U \quad (3.6b)$$

and holding in the domain $\Omega_\epsilon^I(t)$ which is described in the transformed coordinates by

$$\rho \in (Y_\epsilon^-(s, t), Y_\epsilon^+(s, t)) \quad s \in S. \quad (3.7)$$

Here we suppose that $\Gamma_\epsilon^\pm(t)$ are described by the graphs

$$\rho = Y_\epsilon^\pm(s, t) = Y_0^\pm(s, t) + \epsilon Y_1^\pm(s, t) + \mathcal{O}(\epsilon^2) \quad s \in S. \quad (3.8)$$

Thus the boundary conditions (3.2 a,d,e) imply that

$$\lim_{\rho \rightarrow Y_0^\pm} U^0(\rho, s, t) = \pm 1, \quad \lim_{\rho \rightarrow Y_0^\pm} U_\rho^0(\rho, s, t) = 0, \quad \lim_{\rho \rightarrow Y_0^\pm} U^1(\rho, s, t) = 0 \quad (3.9a)$$

$$\lim_{\rho \rightarrow Y_0^\pm} \{M(U^0) W_\rho^1\}(\rho, s, t) = 0, \quad \lim_{\rho \rightarrow Y_0^\pm} \{U^1 M'(U^0) W_\rho^1 + M(U^0) W_\rho^2\}(\rho, s, t) = 0 \quad (3.9b)$$

3.1 Zero Order

It follows that to zero order equations (3.6 a,b) become

$$0 = M(U^0) W_{\rho\rho}^1 + M'(U^0) W_\rho^1 U_\rho^0 \quad (3.10a)$$

$$0 = U_{\rho\rho}^0 + U^0. \quad (3.10b)$$

Equation (3.10b) with boundary condition (3.9a) has the unique monotone increasing solution

$$U^0(\rho, s, t) = \sin(\rho) \quad |\rho| < \frac{\pi}{2} \quad (3.11b)$$

$$Y_0^\pm = \pm \frac{\pi}{2}. \quad (3.11a)$$

Clearly (3.10a) implies that $(M(U^0) W_\rho^1)_\rho = 0$ and hence $M(U^0) W_\rho^1 = a(s, t)$. And from the boundary condition (3.9b), we require $a(s, t) = 0$. Thus for $\rho \in (Y_0^-, Y_0^+)$,

$$W_\rho^1(\rho, s, t) = 0, \quad W^1(\rho, s, t) = \mu(s, t). \quad (3.12)$$

3.2 First Order

The equations are (using (3.12)):

$$0 = M(U^0)W_{\rho\rho}^2 + M'(U^0)W_{\rho}^2U_{\rho}^0 \quad (3.13a)$$

$$W^1 = -U_{\rho\rho}^1 - \kappa^0 U_{\rho}^0 - U^1. \quad (3.13b)$$

It follows from (3.13a) and an argument identical to the zero order case that

$$W_{\rho}^2(\rho, s, t) = 0 \quad \rho \in (Y_0^-, Y_0^+). \quad (3.14)$$

The problem for determining U^1 is thus

$$LU^1 \equiv U_{\rho\rho}^1 + U^1 = -\mu - k^0 U_{\rho}^0 \equiv f, \quad \rho \in (-\pi/2, \pi/2)$$

$$U^1(\pm \frac{\pi}{2}, s, t) = 0.$$

For this to be solvable we require the right hand side f to be orthogonal to the kernel of L subject to homogeneous boundary conditions. Since the kernel is simply span $\{U_{\rho}^0\}$, we multiply (3.13b) by U_{ρ}^0 and integrate. This yields the compatibility equation for $\mu(s, t)$,

$$\mu(s, t) \int_{-\pi/2}^{\pi/2} U_{\rho}^0 d\rho = -\kappa^0 \int_{-\pi/2}^{\pi/2} U_{\rho}^{02} d\rho + \int_{-\pi/2}^{\pi/2} U_{\rho}^0 (U_{\rho\rho}^1 + U^1) d\rho$$

which yields

$$\mu(s, t) = -\frac{\pi}{4} \kappa^0. \quad (3.15)$$

Here we have used the boundary conditions (3.9) and the equation (3.10b).

3.3 Second Order

The equations are, using (3.12) and (3.14)

$$-U_{\rho}^0 V^0 = M(U^0)(W_{\rho\rho}^3 + \Delta_s W^1) + M'(U^0)W_{\rho}^3 U_{\rho}^0 \quad (3.16a)$$

$$W^2 = -[U_{\rho\rho}^2 + U^2 + \Delta d^1 U_{\rho}^0 + \Delta d^0 U_{\rho}^1]. \quad (3.16b)$$

It follows from (3.16a) that

$$-U_{\rho}^0 V^0 = (M(U^0)W_{\rho}^3)_{\rho} + M(U^0)\Delta_s W^0$$

and integrating with respect to ρ from $-\pi/2$ to $\pi/2$, we obtain

$$-2V^0 = \int_{-\pi/2}^{\pi/2} M(U^0) \Delta_s W^0 d\rho = \int_{-\pi/2}^{\pi/2} (1 - \sin^2 \rho) d\rho \Delta_s \mu(s, t)$$

yielding

$$V^0 = -\frac{\pi}{4} \Delta_s \mu(s, t). \quad (3.17)$$

Taking (3.15) and (3.17) together we obtain the desired equation of motion of the interface.

4 Interfacial motion for $0 < \theta \ll 1$.

In this section, the temperature is considered to be small but positive. In particular, it is assumed that

$$\theta = \mathcal{O}(\epsilon^\alpha)$$

for some $\alpha > 0$.

We undertake the matching in terms of the concentration and the concentration fluxes, and we verify *a posteriori* that these assumptions are consistent with the requirement of continuity of the chemical potential and with the expected limiting estimate (2.5) for the size of the chemical potential within the inner region. Therefore, in the outer region we expand

$$u = u^0 + \epsilon u^1 + \epsilon^2 u^2 + \dots \quad (4.1a)$$

$$\mathbf{j} = \mathbf{j}^0 + \epsilon \mathbf{j}^1 + \epsilon^2 \mathbf{j}^2 + \dots, \quad (4.1b)$$

and a similar expansion will be undertaken in the inner region. As in Section §3, we rescale the time $t \rightarrow \epsilon^2 t$, set $\gamma = \epsilon^2$, and study the equation

$$\epsilon^3 u_t = \epsilon \nabla \cdot \{M(u) \nabla \{-\epsilon^2 \Delta u + \Psi'(u)\}\}. \quad (4.2)$$

Note also that as we have assumed that $\theta = \mathcal{O}(\epsilon^\alpha)$ for $\alpha > 0$, its respective place in an ϵ -expansion is somewhat undetermined. We resolve this issue by maintaining it as an $\mathcal{O}(1)$ term in constructing the expansion, but taking into consideration its ϵ -dependence in the matching.

4.1 The outer solution.

As stated in Section §2, both equation (2.1) (rescaled as (4.2)) and the ansatz (2.7) should hold in the outer region. We show below that these assumptions are compatible.

At $\mathcal{O}(1)$,

$$\begin{aligned} 0 &= \nabla \cdot \mathbf{j}^0, \quad \mathbf{j}^0 = (\theta - (1 - u^{02})) \nabla u^0 \quad x \in \Omega_\epsilon^+ \cup \Omega_\epsilon^- \\ \mathbf{n} \cdot \nabla u^0 &= \mathbf{n} \cdot \mathbf{j}^0 = 0 \quad x \in \partial\Omega_\epsilon^+ \cup \partial\Omega_\epsilon^- \cap \partial\Omega. \end{aligned}$$

Note that the boundary conditions pertain to points belonging to the exterior boundary of the domain. Clearly a possible solution is $u^0 = \text{constant}$. In accordance with the ansatz that our solution is nearly equilibrated in the outer solution, we assume that

$$u^0 = \pm\beta(\theta) \quad a.e.$$

and this implies that $\mathbf{j}^0 = 0$.

At $\mathcal{O}(\epsilon)$,

$$\begin{aligned} 0 &= \nabla \cdot \mathbf{j}^1, \quad \mathbf{j}^1 = (\theta - (1 - u^{02})) \nabla u^1 \quad x \in \Omega_\epsilon^+ \cup \Omega_\epsilon^- \\ \mathbf{n} \cdot \nabla u^1 &= \mathbf{n} \cdot \mathbf{j}^1 = 0 \quad x \in \partial\Omega_\epsilon^- \cap \partial\Omega. \end{aligned}$$

From the above equations, we see that $u^1 = 0$ and hence $\mathbf{j}^1 = 0$ is a possible solution at this level.

A similar analysis at order $\mathcal{O}(\epsilon^2)$ yields that $u^2 = 0$ is also consistent with the equations for the outer region.

4.2 The inner solution.

In order to obtain a solution in the inner region Ω_ϵ^I , we introduce again the variables ϕ_ϵ, ρ and s discussed in Section §3 and the Appendix, and we shall assume that $\rho \in I_\epsilon$ where I_ϵ is defined by

$$I_\epsilon \equiv (-\epsilon^{-1}, \epsilon^{-1}).$$

However, in what follows we distinguish between the intervals I_ϵ and $(-\infty, \infty)$ only where necessary. We shall denote by $U = U(\rho, s, t)$ and by $\mathbf{J} = \mathbf{J}(\rho, s, t)$ the concentration and the concentration fluxes expressed in terms of the inner variables, and shall assume expansions for U and \mathbf{J} .

$$\begin{aligned} U &= U^0 + \epsilon U^1 + \epsilon^2 U^2 + \dots, \\ \mathbf{J} &= \epsilon^{-1} \mathbf{J}^{-1} + \mathbf{J}^0 + \epsilon \mathbf{J}^1 + \epsilon^2 \mathbf{J}^2 + \dots \end{aligned}$$

To obtain the desired equation for the motion of the interface, we proceed to develop the expansion through four orders of magnitude.

At $\mathcal{O}(\epsilon^{-1})$,

$$0 = (\theta U_\rho^0 - (1 - U^{02})U_\rho^0 - (1 - U^{02})U_{\rho\rho\rho}^0)_\rho.$$

Integrating this equation once with respect to ρ yields

$$0 = \theta U_\rho^0 - (1 - U^{02})U_\rho^0 - (1 - U^{02})U_{\rho\rho\rho}^0 + C(s, t).$$

Noting that

$$\theta U_\rho^0 - (1 - U^{02})U_\rho^0 - (1 - U^{02})U_{\rho\rho\rho}^0 = \mathbf{J}^{-1} \cdot \mathbf{n}$$

and that noting that $\mathbf{j} = \mathcal{O}(\epsilon^2)$ in the outer region, it follows upon imposing the matching conditions on the concentration fluxes that $C \equiv 0$. We now wish to seek monotone solutions of

$$0 = \theta U_\rho^0 - (1 - U^{02})U_\rho^0 - (1 - U^{02})U_{\rho\rho\rho}^0 \quad (4.3)$$

which also satisfy the matching conditions,

$$\lim_{\epsilon \rightarrow 0} \left[U^0\left(\frac{\eta}{\epsilon^{1/2}}, s, t\right) - u^0(\phi(s, t) + \epsilon^{1/2}\eta\mathbf{n}, t) \right]_{|s, t, \eta \text{ fixed}} = 0$$

where $\eta = r\epsilon^{1/2}$, see (6.1). The most obvious way to satisfy the boundary conditions on the internal I_ϵ is to choose as our solution the unique solution of (4.3) which corresponds to the heteroclinic orbit connecting the two values $\pm\beta(\theta) = \pm(1 - T.S.T.)$, since all other solutions of (4.3) will have finite support and will lie strictly within the interval $(-1, 1)$. This solution satisfies the equation

$$0 = \theta \int_0^{U^0} \ln\left(\frac{1 + \tilde{U}}{1 - \tilde{U}}\right) d\tilde{U} - U^{02} - (U_\rho^0)^2 + k_0 \quad (4.4)$$

where k_0 is chosen so that the minimum of

$$\Psi = \theta \int_0^{U^0} \ln\left(\frac{1 + \tilde{u}}{1 - \tilde{u}}\right) d\tilde{U} - U^{02}$$

will be attained when $U_\rho^0 = 0$.

We note that since $\theta = \theta(\epsilon)$, $U^0 = U^0(\rho, \epsilon)$. In fact, one may write

$$U^0(\rho, \epsilon) = \bar{U}^0(\rho) + \mathcal{O}(\epsilon)$$

where

$$\bar{U}^0(\rho) = \begin{cases} 1 & \rho > \frac{\pi}{2}, \\ \sin(\rho) & -\frac{\pi}{2} \leq \rho \leq \frac{\pi}{2}, \\ -1 & \rho < -\frac{\pi}{2}. \end{cases}$$

Thus, $U^0(\rho, \epsilon)$ may be viewed as a regularized version of the solution $U^0(\rho, s, t)$ obtained in Section §3. It is because of the ϵ -dependence in the solution $U^0(\rho, \epsilon)$ that it is necessary to consider the interval I_ϵ and not directly the interval $(-\infty, \infty)$. Thus, in particular, we are led to look for solutions which are uniformly bounded (with respect to ϵ) in $L^2(I_\epsilon)$ where

$$\|U\|_{L^2(I_\epsilon)} = \frac{\epsilon}{2} \int_{-\epsilon^{-1}}^{\epsilon^{-1}} U^2(\rho) d\rho,$$

and when the integral $\int_{-\infty}^{\infty} \cdot d\rho$ is indicated, we mean $\lim_{\epsilon \rightarrow 0} \int_{-\epsilon^{-1}}^{\epsilon^{-1}} \cdot d\rho$.

At $\mathcal{O}(1)$ using (4.3), we obtain

$$\mathcal{L}[U^1] = ((1 - U^{02})\kappa^0 U_{\rho\rho}^0)_\rho \quad (4.5)$$

where

$$\mathcal{L}[U^1] \equiv \{\theta U_\rho^1 - (1 - U^{02})U_\rho^1 - (1 - U^{02})U_{\rho\rho\rho}^1 + 2U^0 U^1 U_\rho^0 + 2U^0 U^1 U_{\rho\rho\rho}^0\}_\rho = (\mathbf{n} \cdot \mathbf{J}^0)_\rho.$$

The existence (and uniqueness) of the solutions to (4.5) depends on the orthogonality of the right hand side of equation (4.5) to the null solutions of the adjoint equation. It is easy to check that the adjoint equation is given by

$$0 = \theta Q_{\rho\rho} - ((1 - U^{02})Q_\rho)_\rho - ((1 - U^{02})Q_\rho)_{\rho\rho\rho} - 2U^0(2U_\rho^0 + U_{\rho\rho\rho}^0)Q_\rho$$

and that its null solutions are

$$Q_1 = 1, \quad Q_2 = \int^\rho \frac{1}{(1 - U^{02})} d\tilde{\rho}, \quad Q_3 = \int^\rho \frac{U^0}{(1 - U^{02})} d\tilde{\rho}$$

and

$$Q_4 = \int^\rho \frac{v}{(1 - U^{02})} d\tilde{\rho} \text{ where } v_{,\rho} = U_\rho^0 p, \text{ and } p_\rho = \frac{1}{(U_\rho^0)^2}.$$

However, $Q_2, Q_3,$ and Q_4 are sufficiently singular in ϵ that there is no necessity for the right hand side of (4.5) to be orthogonal to these solutions. Therefore, it remains only to require that the right hand side of (4.5) be orthogonal to Q_1 . In particular, integrating the right hand side of (4.5) between $-\infty$ and ∞ with respect to the weight function $Q_1 = 1$, we obtain that

$$[(1 - U^{02})\kappa^0 U_{\rho\rho}^0]_{-\infty}^{\infty} = 0,$$

which can be seen by (4.4) to hold.

Integrating the resultant equation for U^1 once with respect to ρ and noting that matching gives that

$$\begin{aligned}\lim_{\epsilon \rightarrow 0} \left[\mathbf{n} \cdot \mathbf{J}^0 \left(\frac{\eta}{\epsilon^{1/2}}, s, t \right) \right]_{\eta \text{ fixed}} &= \lim_{\epsilon \rightarrow 0} \left[\mathbf{n} \cdot \mathbf{j}^0(\phi(s, t) + \epsilon^{1/2} \eta \mathbf{n}, t) \right]_{\eta \text{ fixed}} = 0 \\ \lim_{\epsilon \rightarrow 0} \left[(U^0)^2 \left(\frac{\eta}{\epsilon^{1/2}}, s, t \right) \right]_{\eta \text{ fixed}} &= \lim_{\epsilon \rightarrow 0} \left[(u^0)^2(\phi(s, t) + \epsilon^{1/2} \eta \mathbf{n}, t) \right]_{\eta \text{ fixed}} = 1\end{aligned}$$

we may conclude that U^1 is determined by the equation

$$(1 - U^{02}) \kappa^0 U_{\rho\rho}^0 = \mathbf{n} \cdot \mathbf{J}^0. \quad (4.6)$$

At order $\mathcal{O}(\epsilon)$, noting that U^0 is independent of s and using (4.3) and (4.6), we find that the relevant equations are

$$\mathcal{L}[U^2] = (1 - U^{02}) (\kappa^0)^2 U_{\rho\rho}^0 - ((2U^0 U^1) \kappa^0 U_{\rho\rho}^0)_\rho + ((1 - U^{02}) \kappa^1 U_{\rho\rho}^0)_\rho. \quad (4.7)$$

Again we impose the condition that the right hand side of the equation (4.7) be orthogonal to Q_1 . However, in this case, this condition may be taken into account rather simply as follows. We note that the function U^0 is odd and that the function U^1 prescribed by (4.6) may easily be seen to be even, hence by looking at the terms on the right hand side of (4.7), we find that the first two terms are odd and hence trivially orthogonal to the constant function $Q_1 = 1$. Lastly, it was shown above that

$$\int_{-\infty}^{\infty} ((1 - U^{02}) U_{\rho\rho}^0)_\rho d\rho = 0.$$

Thus, (4.7) may be simplified to give

$$(\mathbf{n} \cdot \mathbf{J}^1)_\rho = \mathcal{L}[U^2] = ((1 - U^{02}) \kappa^0 U_{\rho\rho}^1)_\rho - ((2U^0 U^1) \kappa^0 U_{\rho\rho}^0)_\rho. \quad (4.8)$$

Since $u^1 = 0$ and $\mathbf{j}^1 = 0$, we have in addition the matching conditions

$$\begin{aligned}\lim_{\epsilon \rightarrow 0} \left[\mathbf{n} \cdot \mathbf{J}^1 \left(\frac{\eta}{\epsilon^{1/2}}, s, t \right) \right]_{\eta \text{ fixed}} &= \lim_{\epsilon \rightarrow 0} \left[\mathbf{n} \cdot \mathbf{j}^1(\phi(s, t) + \epsilon^{1/2} \eta \mathbf{n}, t) \right]_{\eta \text{ fixed}} = 0 \\ \lim_{\epsilon \rightarrow 0} \left[U^1 \left(\frac{\eta}{\epsilon^{1/2}}, s, t \right) \right]_{\eta \text{ fixed}} &= \lim_{\epsilon \rightarrow 0} \left[u^1(\phi(s, t) + \epsilon^{1/2} \eta \mathbf{n}, t) \right]_{\eta \text{ fixed}} = 0,\end{aligned}$$

hence (4.8) may be integrated explicitly to yield

$$\begin{aligned}\mathbf{n} \cdot \mathbf{J}^1 &= (1 - U^{02}) \kappa^0 U_{\rho\rho}^1 - (2U^0 U^1) \kappa^0 U_{\rho\rho}^0 \\ &\quad - 2U^0 U^1 \kappa^0 U_\rho^1 - (U^1)^2 \kappa^0 U_\rho^0 - 2U^0 U^1 \kappa^0 U_{\rho\rho\rho}^1 - (U^1)^2 \kappa^0 U_{\rho\rho\rho}^0.\end{aligned}$$

We remark that it is not necessary to solve for U^2 explicitly, since this expression will not be necessary in order to obtain the asymptotic description of the evolution of the interface.

At order $\mathcal{O}(\epsilon^2)$, imposition of the orthogonality constraint implies that

$$\begin{aligned}
V^0\{U^0(\infty) - U^0(-\infty)\} = & \int_{-\infty}^{\infty} \left\{ -(1 - U^{02}) \Delta_s \kappa^0 U_{\rho}^0 + \theta \{ \Delta_s U^1 \} - (1 - U^{02}) \Delta_s U^1 - (1 - U^{02}) \Delta_s U_{\rho\rho}^1 \right\} d\rho \\
& + \int_{-\infty}^{\infty} \left\{ -(1 - U^{02}) (\kappa^0)^2 U_{\rho\rho}^1 + 2U^0 U^1 (\kappa^0)^2 U_{\rho\rho}^0 + 2U^0 U^1 \kappa^0 U_{\rho}^1 + (U^1)^2 \kappa^0 U_{\rho}^0 + 2U^0 U^1 \kappa^0 U_{\rho\rho\rho}^1 \right. \\
& \quad \left. + U^{12} \kappa^0 U_{\rho\rho\rho}^0 \right\} d\rho \\
& + \kappa^0 \int_{-\infty}^{\infty} \left\{ \theta U_{\rho}^2 - (1 - U^{02}) U_{\rho}^2 - (1 - U^{02}) U_{\rho\rho\rho}^2 + 2U^0 U^2 U_{\rho}^0 + 2U^0 U^2 U_{\rho\rho\rho}^0 \right\} d\rho \\
& + \kappa^1 \int_{-\infty}^{\infty} \left\{ (1 - U^{02}) \kappa^0 U_{\rho\rho}^0 - ((1 - U^{02}) U_{\rho\rho}^1)_{\rho} + 2U^1 (U^0 U_{\rho\rho}^0)_{\rho} + 2U^0 U^1 U_{\rho\rho}^0 \right\} d\rho \\
& + \kappa^2 \int_{-\infty}^{\infty} \left\{ (1 - U^{02}) U_{\rho\rho}^0 \right\}_{\rho} d\rho.
\end{aligned}$$

Noting now that the third integral is simply equal to $\kappa^0 \int_{-\infty}^{\infty} \mathbf{n} \cdot \mathbf{J}^1 d\rho$ and using the expression for $\mathbf{n} \cdot \mathbf{J}^1$ given above, we find that the sum of the second and third integrals vanishes. The fourth integral vanishes since the integrand is easily seen to be odd and hence orthogonal to $Q_1 = 1$. The fifth integral vanishes as discussed previously.

In order to evaluate the first integral, noting that $\mathcal{L}[U_{\rho}^0] = 0$, it follows from (4.4) and (4.5) that U^1 may be expressed in the form $U^1 = \kappa^0 g U_{\rho}^0$ where $g = g(\rho)$. Substituting this expression in the first integral on the right hand side of (4.9), and recalling (4.3), we obtain that

$$V^0\{U^0(\infty) - U^0(-\infty)\} = -\Delta_s \kappa^0 \left\{ \int_{-\infty}^{\infty} (1 - U^{02}) U_{\rho}^0 d\rho + \int_{-\infty}^{\infty} (1 - U^{02}) (g_{\rho\rho} U_{\rho}^0 + 2g_{\rho} U_{\rho\rho}^0) d\rho \right\}. \quad (4.9)$$

It is easy to check that $g = g(\rho)$ satisfies the equation

$$g_{\rho\rho} U_{\rho}^0 + 2g_{\rho} U_{\rho\rho}^0 = -U_{\rho}^0 + \frac{1}{[U^0]_{\pm}^{\pm}} \int_{-\infty}^{\infty} U_{\rho}^{02} d\rho. \quad (4.10)$$

Using the expression (4.10) in (4.9), we obtain finally the equation for the motion of the interface.

$$V^0\{U^0(\infty) - U^0(-\infty)\} = -\Delta_s \kappa^0 \left\{ \frac{1}{2} \int_{-\infty}^{\infty} U_{\rho}^{02} d\rho \cdot \int_{-\infty}^{\infty} (1 - U^{02}) d\rho \right\}, \quad (4.11)$$

or, in other words, motion by the Laplacian of the mean curvature. We note that the second integral in the equation (4.11) does not converge unless we apply the definition of the integral described earlier, in which case these expressions can be evaluated to yield in the limit

$$V^0 = -\frac{\pi^2}{16} \Delta_s \kappa^0.$$

4.3 The chemical potential.

Let us now consider the behavior of the chemical potential. Returning now to the solution given in the outer region, we find that formally evaluating w to lowest order, we find that

$w^0 = w(u^0) = 0$. However, it is not so easy to obtain a next order approximation, since if we write $u = u^0 + \tilde{u}$, then \tilde{u} satisfies the equation

$$\tilde{w} = -\gamma\Delta\tilde{u} + \theta(1 - u^{02})^{-1}\tilde{u} - \tilde{u},$$

where \tilde{w} is the correction to w^0 , and since $\theta(1 - u^{02})^{-1}$ is transcendently large and \tilde{u} is (at least) algebraically small, the order of the correction to w^0 is not clear.

Suppose we consider now the behavior of the chemical potential in the inner region. To lowest order,

$$W^0 = -U_{\rho\rho}^0 + \frac{\theta}{2}\{\ln(1 + U^0) - \ln(1 - U^0)\} - U^0,$$

which can easily be seen to vanish by looking at equation (4.4). However, here it is possible to obtain the correction,

$$\tilde{W} = -\kappa^0 U_{\rho}^0 - U_{\rho\rho}^1 + \theta(1 - U^{02})^{-1}U^1 - U^1,$$

which, using (4.10), (4.12), can be evaluated to yield

$$\tilde{W} = -\frac{1}{2}\epsilon\kappa^0 \int_{-\infty}^{\infty} U_{\rho}^{02} d\rho,$$

or more explicitly,

$$\tilde{W} = -\frac{\epsilon\pi}{4}\kappa^0. \quad (4.15)$$

This result is in line with the expected results described in the Preliminaries in Section §2, as well as (3.15) which was obtained in Section §3. The continuity up to zero order, and the order of the correction also supports the expansion in ϵ of W undertaken in Section §3.

5 Conclusion.

The geometric motion given by minus the surface Laplacian of the curvature has been previously derived by Mullins in the context of surface diffusion for single component alloys in a vapor environment [20] and by Davi and Gurtin [9] in the general thermodynamic framework of [16]. It has also been obtained formally as H^{-1} gradient flow [30]. In the present paper, this motion has been derived formally here in the context of phase separation in binary alloys, described by Cahn-Hilliard equation with concentration dependent mobility, both in the deep quench limit and at asymptotically low temperatures, assuming a time scale of $\epsilon^2 t$ as opposed to the ϵt time scale employed in the derivation of the Mullins-Sekerka equation for the Cahn-Hilliard equation with constant mobility [25]. In order to prove this formal result rigorously one would need an extension of the techniques of Alikakos, Bates and Chen [1] to deal with the degenerate nature of (1.1).

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Appendix.

Let $\Gamma(t)$ be a smooth closed evolving interface in $\Omega \subset \mathbb{R}^n$, $n = 2$ or 3 with interior $\Omega^-(t)$ and exterior $\Omega^+(t)$. Let $\phi : S \times [0, T] \rightarrow \mathbb{R}^n$ be a parametric representation of $\Gamma(t)$ where S is an

orientated manifold of dimension $n - 1$. Thus $x \in \Gamma(t)$ provided a unique $s \in S$ exists such that $x = \phi(s, t)$. We assume that there is an annular neighborhood

$$\mathcal{N}(t) = \{x \in \Omega \mid \text{dist}(x, \Gamma_\epsilon(t)) < d_0\}$$

of $\Gamma_\epsilon(t)$ such that for each $x \in \mathcal{N}(t)$ there is a unique orthogonal projection from x to $\Gamma_\epsilon(t)$. Thus there is a unique pair $(s, r) \in S \times (-d_0, d_0)$ solving for each $x \in \mathcal{N}(t)$,

$$x = \phi(s, t) + r\mathbf{n}(s, t) \tag{6.1}$$

where \mathbf{n} is a unit normal to $\Gamma_\epsilon(t)$ pointing into $\Omega^+(t)$ and $r = d(x, t)$ where $d(\cdot, t)$ is the distance function to $\Gamma_\epsilon(t)$ signed to be positive in $\Omega^+(t)$ and negative in $\Omega^-(t)$. It follows that

$$\begin{aligned} \nabla_x d &= \mathbf{n}, \\ \frac{\partial d}{\partial t} &= -V, \end{aligned} \tag{6.2}$$

$$\Delta_x d = \kappa(s(x, t), t) + d\kappa_s(s(x, t), t) + \mathcal{O}(d^2)$$

where κ is the mean curvature of $\Gamma^+(t)$ signed to be positive if $\Omega^+(t)$ is convex and V is the normal velocity of $\Gamma_\epsilon(t)$.

Because of (6.1) we may transform the coordinates from (x, t) to (s, r, t) and setting $\Psi(s, r, t) = \psi(x, t)$ we have

$$\begin{aligned} \frac{\partial \psi}{\partial t} &= \Psi_t + \Psi_s^T \frac{\partial s}{\partial t} + \Psi_r \frac{\partial d}{\partial t} \\ \nabla_x \psi &= (J_{xs})^T \Psi_s + \Psi_r \nabla_x d \\ \Delta_x \psi &= \Delta_s \Psi + \Psi_r \Delta_x d + \Psi_{rr}. \end{aligned} \tag{6.3}$$

Here J_{xs} is the Jacobian matrix $\partial s_i / \partial x_j$ and

$$\Delta_s \Psi := \text{Tr}[(J_{xs})^T \Psi_{ss} (J_{xs})] + \Delta_x s^T \nabla_s \Psi$$

is the surface Laplacian of Ψ . It holds, by the orthogonality of $\nabla_x d$ with respect to the surface, that

$$(J_{xs})^T \nabla_x d \equiv \mathbf{0}.$$

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