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Sharp Interface Limits of the Cahn-Hilliard Equation with Degenerate Mobility

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Phase field models are a common framework to describe the mesoscale kinetics of phase separation and pattern-formation. Replacing a sharp interface by a diffuse order parameter profile, phase field models avoid the numerically challenging interface tracking, and are versatile enough to capture topological changes. Although models can be constructed starting from a systematic coarse-graining of the microscopic dynamics, their use as a numerical tool to solve free boundary problems requires careful consideration of what their correct asymptotic long-time sharp interface limits are.

One of the simplest models for spinodal demixing of binary alloys is the Cahn-Hilliard equation, where the order parameter u is conserved and satisfies

$$u_t = -\nabla \cdot \mathbf{j}, \quad \mathbf{j} = -M(u)\nabla\mu, \tag{1a}$$

$$\mu = -\epsilon^2\nabla^2 u + f'(u), \tag{1b}$$

$$f(u) = \frac{1}{4}(1-u^2)^2, \tag{1c}$$

where $M(u)$ is the mobility function; $\mathbf{j}$ is the flux; μ is the chemical potential; ϵ is the interfacial tension which determines the width of the interface, and $f(u)$ is the bulk free energy.

Whilst the asymptotic sharp interface limit $\epsilon \to 0$ for $M(u) = \text{const}$ has been shown by Pego (1989) (and proven rigorously in Alikakos et al. (1994)) to reduce to the Mullins-Sekerka problem (Mullins and Sekerka, 1963) at the long time scale $t = O(1/\epsilon)$, the sharp interface limit for a degenerate mobility function, which vanishes at the pure phases, *e.g.* $M(u) = \frac{1}{2}(1-u^2)$, has been recently a matter of controversy.

A body of prior works (e.g. Bhate et al. (2000); Yeon et al. (2006); Jiang et al. (2012)) used the Cahn-Hilliard equation with degenerate mobility as a basis for solving the surface diffusion free boundary problem, where the interface velocity is proportional to the surface Laplacian of the mean curvature, *i.e.*

$$v_n = \mathcal{M}\Delta_s\kappa, \tag{2}$$

with $\mathcal{M}$ a constant and κ the mean curvature. This phase field approach to surface diffusion model has been the key framework for modelling a myriad of complex physical processes such as electromigration in metals (Mahadevan and Bradley, 1999), heteroepitaxial growth (Rätz et al., 2006) and more recently solid-solid dewetting (Jiang et al., 2012). Heuristically, the degeneracy of the mobility function at the pure phases suppresses the flux normal to the interface and therefore the diffusion from or into the bulk. However, recently, Gugenberger et al. (2008) have suggested an inconsistency in the matching condition used and cast this "conventional wisdom" into doubt.

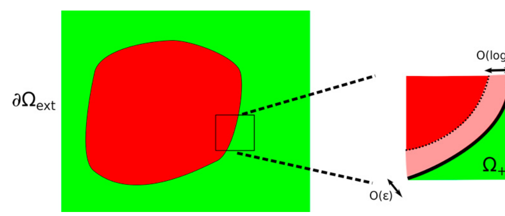

Fig. 1. Illustration of the asymptotic structure of the degenerate mobility case. The dotted line indicates the contact line free boundary beyond which $u = 1$, and the solid line denotes the interface ζ. There are interior layers about the contact line and interface respectively, and an outer layer outside the precipitate (the green region Ω_+).

To resolve this conundrum, we revisit the matched asymptotic analysis of Equation (1). Our analysis reveals several aspects of the asymptotic structure of the solution which were not taken into account previously. First, unlike the constant mobility case, degeneracy in the mobility introduces a free boundary where the order parameter reaches $u = \pm 1$. The dynamics of the free boundary "contact line" couples with the dynamics of the interface, and requires an $O(\epsilon \log(1/\epsilon))$ small contact line region to complete the asymptotic analysis via exponential asymptotics (see Figure 1 for the asymptotic structure of the solution).

Second, we found that with the commonly used quadratic mobility $M(u) = \frac{1}{2}(1-u^2)$, mass flux from and into the bulk region (where $u = \pm 1$) still appears in the leading order dynamics. As a result of that, the interface velocity in the sharp interface model has two contributions: one from surface diffusion (Equation (2)) which is local to the interface, and another contribution from nonlinear bulk diffusion. More precisely, our analysis leads to the sharp interface limit

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$$\begin{aligned} \nabla \cdot (\mu_1 \nabla \mu_1) &= 0, \quad \text{in } \Omega_+, \\ \mu_1 &= \frac{2}{3}\kappa, \quad \text{on } \zeta, \\ \nabla_n \mu_1 &= 0, \quad \text{on } \partial\Omega_{\text{ext}}, \\ v_n &= \frac{2}{3}\Delta_s \kappa + \frac{1}{4}\mu_1 \nabla_n \mu_1 \quad \text{on } \zeta, \end{aligned} \tag{3}$$

with the definition of Ω_+, Ω_{ext} and ζ identical to Figure 1. Unlike pure surface diffusion, the mass flux contribution to normal velocity in Equation (3), which satisfies a porous-medium equation, couples precipitates with each other. This results in coarsening where larger precipitates grow at the expense of smaller ones.

Third, not all degenerate mobilities are the same. For higher order mobilities $M_\alpha(u) = (1 - u^2)^\alpha$, $\alpha > 1$, our asymptotics suggests that the normal mass flux does not enter into v_n at leading order, so that the surface diffusion model is recovered in the asymptotic limit. (Note that $M'_\alpha(1)$ diverges for $\alpha < 1$, hence we restrict our attention to the case $\alpha > 1$.)

Fourth, the sharp interface limit is dependent on the precise form of the free energy. Our asymptotic matching relies on a continuous free energy function, which can be derived via a Landau-Ginzburg expansion based on symmetry arguments for a second order phase transition. The Landau-Ginzburg formalism assumes that the free energy is an analytic function. Indeed, the double obstacle free energy, which is the low temperature limit of the lattice gas entropy and diverges at the pure phases, was shown to give surface diffusion flow even for $M_1(u) = (1-u^2)$ (Cahn et al., 1996). This outlines an underappreciated piece of physics: different free energy functions for phase transitions, though being in the same equilibrium universality class, may have drastically different dynamics.

Numerical solutions of Equation (1) agree well with our asymptotic results. Taken together, our asymptotic framework conclusively closes the conundrum in the literature and proposes the phase field approximation to a new free boundary problem involving coupled surface diffusion and nonlinear bulk diffusion. Phase field approaches to surface diffusion cannot be realised with the Cahn-Hilliard equation with a degenerate mobility $M(u) = 1 - u^2$ and polynomial free energy, as was repeatedly assumed in the literature — a higher order degeneracy in the mobility or a double obstacle free energy maybe needed.

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