Nonlocal Cahn-Hilliard and Isoperimetric Problems: 
Periodic Phase Separation Induced by Competing Long- 
and Short-range Interactions

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1 Introduction

The modeling of pattern morphologies via energy minimization involving long and short- 
range competitions is well-established and ubiquitous in science (cf. [47, 23, 33] and the 
references therein). It also provides mathematicians with a wealth of rich variational problems 
consisting of both local (associated with short-range interactions) and nonlocal (associated 
with long-range interactions) terms (see for example [29] and the references therein).

Here we are concerned with two nonlocal variational problems which describe the simplest 
morphology class within this general setting, that of periodic phase separation. Many physical 
systems exhibit a phase separation which, according to experiments, can be roughly described 
as follows:

\( P_1 \) The phase separation is (nearly) periodic on some fixed (mesoscopic) scale.

\( P_2 \) Within a period cell, the structure seems to want to minimize surface area between 
the two phases.

1.1 The Physical paradigm: Diblock copolymers

A paradigm for \( P_1 \) and \( P_2 \) is provided by microphase separation of diblock copolymers (cf. 
[6, 50, 5]). A diblock copolymer is a linear-chain molecule consisting of two sub-chains joined covalently to each other. One of the sub-chains is made of monomers of type A and the other of type B. Below a critical temperature, even a weak repulsion between unlike monomers A
and B induces a strong repulsion between the sub-chains, causing the sub-chains to segregate. A macroscopic segregation whereby the sub-chains detach from one another cannot occur because the chains are chemically bonded: Rather, a phase separation on a mesoscopic scale with A and B-rich domains emerges. The observed mesoscopic domains, illustrated in Figure 1, are highly regular periodic structures with interfaces strongly resembling surfaces with constant mean curvature; for example lamellar, spheres, cylindrical tubes, and single and double-gyroids (see for example, [6]). The connection between observed structures and triply periodic constant mean curvature surfaces (many of which are stable in a suitable sense) has been well-established in the literature (see for example, [50, 27, 3]).

1.2 Mathematical Paradigms

The Periodic Cahn-Hilliard and Isoperimetric Problems

The simplest mathematical model for capturing \( P1 \) and \( P2 \) would be to a propri inforce the periodicity (a hard constraint) and look at the resulting interfacial problems; thus the long-range interactions no longer play any role. One is thus led to the periodic Cahn-Hilliard and isoperimetric problems. Let

\[
E_\epsilon(u) = \int_{\mathbb{T}^n} \frac{(1 - u^2)^2}{4} + \frac{\epsilon^2}{2} |\nabla u|^2 \, dx
\]
for $u \in H^1(\mathbb{T}^n)$, $\int_{\mathbb{T}^n} u = m$, and

$$E_0(u) = c_0 \int_{\mathbb{T}^n} |\nabla u|$$

for $u \in BV(\mathbb{T}^n, \pm 1)$, $\int_{\mathbb{T}^n} u = m$. Only can easily extend $E_\epsilon$ and $E_0$ to all function in $L^1(\mathbb{T}^n)$ by defining them to be zero if $u$ is not in $H^1(\mathbb{T}^n)$ and $BV(\mathbb{T}^n, \pm 1)$, respectively. By the the Periodic Cahn-Hilliard Problem we mean

$$(PCH) \quad \text{Minimize } E_\epsilon(u) \text{ over all } u \in L^1(\mathbb{T}^n).$$

By the Periodic Isoperimetric Problem we mean

$$(PIP) \quad \text{Minimize } E_0(u) \text{ over all } u \in L^1(\mathbb{T}^n).$$

Both problems are clearly well-posed and the rigorous connection between them (for an appropriate choice of constant $c_0$ and rescaling of $E_\epsilon$) is via Gamma convergence. Our main interest here is not to treat the periodicity as a hard constraint, but rather when it results from an interplay between minimizing nonlocal and local terms. However, we do make a few comments concerning (PCH) and (PIP) based upon recent work with P. Sternberg ([14]). Our primary concern in [14] was the wealth of nontrivial stable local minimizers of (PIP) (stable constant mean curvature surfaces), and whether or not these surfaces have diffuse interface analogues (local minimizers of (PCH)). Our primary tool was the notion of a strict (up to translation) $L^1$ local minimizer in the sense of [30] which carries through to the diffuse problem via Gamma convergence. Hence the relevant question was whether or not stability (i.e. a positive bound on the second variation) was equivalent to being a strict $L^1$ local minimizer. In 2D the situation is greatly simplified, and we proved the equivalence of the two notions. The 3D case remains open.

The Nonlocal Cahn-Hilliard and Isoperimetric Problems

Any model for microphase separation of diblock copolymers cannot be based solely on minimization of $A$-$B$ monomer interfaces, and must take into account the nature of the joined $A$- and $B$-subchain interactions. Indeed, a density functional theory, first proposed by Ohta and Kawasaki [35], entails the minimization of a nonlocal Cahn-Hilliard like energy (cf. [34]) whereby the standard Cahn-Hilliard free energy is augmented by a long-range

\footnote{We should mention here that Pacard and Ritore [37] prove a result connecting critical points of the sharp and diffuse interface problems. With a completely different machinery, they address the problem in great generality. However, our question is different as it pertains to structures which are in some sense minimizing (not just critical points).}

\footnote{The term the Nonlocal Cahn-Hilliard Equation has already been extensively used. It pertains to a different problem and should not be confused with either the variational problem (NLCH) or the gradient flow PDE (4.4).}
interaction term – associated with the connectivity of the sub-chains in the diblock copolymer macromolecule\(^3\). Let \( \Omega \) be the unit cube \((0, 1)^n\) in \( R^n \) and fix \( m \in (-1, 1)\). Minimize

\[
(NLCH) \quad \text{Minimize} \quad \frac{\epsilon^2}{2} \int_{\Omega} |\nabla u|^2 \, dx + \int_{\Omega} \frac{(1 - u^2)^2}{4} \, dx + \frac{\sigma}{2} \int_{\Omega} |\nabla v|^2 \, dx,
\]

over all \( u \in \left\{ u \in H^1(\Omega, [-1,1]) \mid \int_{\Omega} u = m \right\} \). Here, \( v \) is related to \( u \) via the BVP

\[
-\triangle v = u - m,
\]

in the sense of distributions with either Neumann or periodic boundary conditions for \( v \) on \( \partial \Omega \). The relevant parameter regimes are either \( \epsilon \sim \sigma \ll 1 \) or \( \epsilon \ll \sigma \ll 1 \). From the (NLCH), the incentive for pattern formation is clear: The first term wants uniform phases, with the second term penalizing transitions between phases. However the nonlocal term is lowered by oscillations between the two phases. The effect of the three is to set a fine scale (probably periodic) structure.

If one focuses on the strong segregation limit, wherein the interfacial thickness of the phase boundaries tends to zero, one can easily show (cf. [39], [12], [1]) that the relevant sharp interface problem can be written as:

\[
(NLIP) \quad \text{Minimize} \quad \int_{\Omega} |\nabla u| \, dx + \gamma \int_{\Omega} |\nabla v|^2 \, dx,
\]

over all \( u \in \left\{ u \in BV(\Omega, [-1,1]) \mid \int_{\Omega} u = m \right\} \). Here \( \gamma \) is a parameter which will determine the length scale of minimizers, and again \( v \) is related \( u \) via (1.1). We refer to this problem as the Nonlocal Isoperimetric Problem.

We are interested in the following three issues surrounding (NLCH) and (NLIP):

(i) The periodicity resulting from competing local and nonlocal terms

(ii) The geometry of minimizers: the effect of the nonlocal term, perturbation from the phase boundary being area-minimizing / constant mean curvature

(iii) The PDE resulting from the \( H^{-1} \) gradient flow for (NLCH): numerical simulation of steady states and the dynamic stages of the phase separation (spinodal decomposition, coarsening etc.)

In Section 2, we describe a rigorous results addressing issue (i) for (NLIP) – joint work with G. Alberti and F. Otto. In Section 3, we provide a first attempt at a general method for addressing issue (ii) via some recent results on the consequences of stability for (NLIP) – joint work with P. Sternberg. Finally in Section 4, we discuss issue (iii) and present some simulations, focusing more on questions than on answers.

2 The Nonlocal Isoperimetric Problem: Uniform Energy Distribution

For dimension $n = 1$, one can show (either for (NLCH) or (NLIP)) that minimizers are periodic (see [32] for the case $m = 0$ and [41] for general $m$). Heuristically, it is straightforward to see that the scale of the periodicity is set by the nonlocal term; For $\sigma$ sufficiently large, the periodicity on a scale smaller than the domain size is enforced as a weak constraint via interactions between the perimeter and the nonlocal terms. The natural question is to what extent are minimizers periodic in higher-$D$. Here we present a rigorous result to support the conjecture that in higher-$D$, minimizers are “nearly” periodic on a length scale set by $\gamma$, the dimension $n$, and volume fraction $m$. We do so via the energy associated with a minimizer, and its uniform distribution in space.

The length scale for minimizers is set by $\gamma$. Since in this section, we are interested in a qualitative ”periodicity” result on sufficiently large domains, we may just as well set it to unity. We will however need the following equivalent reformulation of (NLIP). Let $\Omega$ be a cube in $\mathbb{R}^n$ and $m \in (-1, 1)$. Then for any $u \in BV(\Omega_L, \pm 1)$, there exits $b \in L^2(\Omega, \mathbb{R}^n)$ such that $\nabla \cdot b = u - m$ in the sense of distributions, with either Neumann (i.e. $b \cdot n = 0$) or periodic boundary conditions for $b$. Minimizing over all such vector fields $b$, we find

$$\min \int_{\Omega} |b|^2 = \int_{\Omega} |\nabla v|^2,$$

where as usual $v$ solves (1.1) on $\Omega$, i.e. $-\Delta v = u - m$, in the sense of distributions with the respective boundary conditions (Neumann or periodic). This reformulation has several advantages. For example, it localizes the functional and facilitates the use of cutting and pasting arguments. It further allows us to demonstrate that the particular choice of boundary conditions do not influence the structure away from the boundary – Theorem 2.1 and its proof will support this.

We now present the reformulation of (NLIP): Let $\Omega_L = [0, L]^n$, $L >> 1$. Consider

$$\min_{u \in \mathcal{A}_{per}} E(u, b, \Omega_L) = \int_{\Omega_L} |\nabla u| + \int_{\Omega_L} |b|^2 dx.$$

$$\mathcal{A}_{per} = \{(u, b) : u \in BV(\Omega_L, \pm 1), b \in L^2(\Omega_L, \mathbb{R}^n)$$

$$\nabla \cdot b = u - m, \text{ periodic b.c. for } b \cdot n\}.$$

Before presenting our result on the uniform distribution of energy for minimizers, we provide some motivation for the result. Suppose the minimizer $(u_p, b_p)$ on $\Omega_L$ ($L >> 1$) was exactly periodic, say $u_p$ represents a lamellar structure with period $p$, where $L = kp$ for some integer $k$ – see Figure 2. It is straightforward to see that there exist a constant $\bar{E}_0$ (which depends on $p$ but not on $k$) such that

$$\frac{E(u_p, b_p, Q_{kp})}{(kp)^n} = \bar{E}_0.$$
Now consider the structure on some smaller cube, \( Q_l \) with size \( l, L > l \gg 1 \), i.e. consider \((u_p, b_p)\) restricted to \( Q_l \) (see Figure 2). Computing its the energy density, we may not get exactly \( \bar{E}_0 \), however one finds

\[
\left| \frac{E(u_{\text{per}}, b_{\text{per}}, Q_l(x))}{l^n} - \bar{E}_0 \right| \leq C, \]

for some constant \( C \).

Our result is to prove the same estimate but for any minimizer:

**Theorem 2.1.** There exist constants \( \sigma^* > 0, C > 0 \) depending only on \( m, n \) s.t. if \((u_0, b_0)\) is a minimizer of \( E \) on \( A_{\text{per}} \), then for every \( l \leq L \) and \( x \in \Omega_L \),

\[
\left| \frac{E(u_0, b_0, Q_l(x))}{l^n} - \sigma^* \right| \leq C, \]

where \( Q_l(x) = \text{cube if size } l, \text{ centered at } x \). In fact,

\[
\sigma^* = \lim_{L \to \infty} \left( \min_{(u_0, b_0) \in A_{\text{per}}} \frac{E(u_0, b_0, Q_L)}{L^n} \right).
\]

Not surprisingly, the same limit (definition of \( \sigma^* \) above) would be achieved for other boundary conditions (Neumann, free, etc.). One can also prove a similar estimate for the separate terms in the energy, in particular for just the surface energy.

**3 The Nonlocal Isoperimetric Problem: Stability and the Second Variation**

The structures which one observes in microphase separation are mostly constant mean curvature (CMC) surfaces. This fact is documented in both the science and mathematics literature (cf. [6], [50], [5], and [46]). If one believes that the variational problem (NLCH), and hence (NLIP), captures the essential physics of the phase separation, then one would expect that minimizers of (NLIP), are close to minimizers of (PIP).
In a series of papers ([40, 42, 43, 44, 45]), Ren and Wei consider the stability of lamellar, ring/cylindrical, and spot/spherical solutions (the latter two considered on a radially symmetric domain). The approach here was to linearize the Euler-Lagrange equation (or gradient flow equation) about one of these critical points, and consider the spectrum of the linearized operator. One obtains stability of several structures for small $\gamma$ and instability for $\gamma$ sufficiently large. Interestingly, they also prove that certain non CMC structures (so called wiggled lamellar and spot solutions) were stable for suitable values of $\gamma$.

As a first step towards a global approach to the consequences of stability, we provide a rigorous statement of the first and second variations on (NLIP). This is joint work with P. Sternberg, see [15] for details. For simplicity let us work with periodic boundary conditions (i.e. $\Omega = \mathbb{T}^n$, the n-dimensional flat torus). See [15] for the analogous results with Neumann boundary conditions. Let

$$E_\gamma(u) := \int_{\mathbb{T}^n} |\nabla u| + \gamma \int_{\mathbb{T}^n} |\nabla v|^2 \, dx$$

We minimize $E_\gamma$ over all $u \in BV(\mathbb{T}^n, \pm 1)$ with $\int_{\mathbb{T}^n} u = m$ and $-\Delta v = u - m$ in $\mathbb{T}^n$.

For $u \in BV(\mathbb{T}^n, \pm 1)$ we can naturally associate a set of finite perimeter $A := \{u = 1\}$ and visa versa. Given any such $u \iff$ set of finite perimeter $A$, we define an appropriate admissible perturbation $A_t$ (i.e. associated with the volume constraint, see Remark 3.4 (i)) and define

$$U(x, t) = \begin{cases} 1 & \text{if } x \in A_t \\ -1 & \text{if } x \in A_t^c. \end{cases}$$

Then we say $A$ is a critical point if

$$\frac{d}{dt} \bigg|_{t=0} E_\gamma(U(\cdot, t)) = 0,$$

and is further stable if:

$$\frac{d^2}{dt^2} \bigg|_{t=0} E_\gamma(U(\cdot, t)) \geq 0.$$

**Regularity Assumption:** When $\gamma = 0$, it is well-known (see for example [24]) that in dimensions $n < 8$, the phase boundary associated with any $L^1$–local minimizer must have constant mean curvature and be an analytic $(n-1)$-dimensional manifold, while in dimensions $n \geq 8$, the same is true off of a (perhaps empty) singular set of Hausdorff dimension at most $n-8$. While the phase boundary associated with a local minimizer for $\gamma > 0$ will, in general, no longer have constant mean curvature, we strongly suspect that this lower-order, compact perturbation will not destroy regularity, so we expect that the phase boundary associated with a local minimizer of $E_\gamma$ is still an analytic $(n-1)$-dimensional manifold in dimensions $n < 8$, and in particular is $C^2$. Where as it quite natural to expect that the nonlocal perturbation does not destroy regularity, a more interesting question is whether or not it can help. That is, can it reduce the Hausdorff dimension of the singular set for $n \geq 8$?
Proposition 3.1. (The First Variation) Let $u$ be a critical point of $E_\gamma$ such that for $A := \{ u = 1 \}, \partial A$ is $C^2$ with mean curvature $H(x)$. Then for $v$ solving $-\Delta v = u - m$ in $\mathbb{T}^n$ there exists a constant $\lambda$ such that

$$(n - 1) H(x) + 4 \gamma v(x) = \lambda \text{ for all } x \in \partial A. \quad (3.2)$$

Remark 3.2. When $\gamma = 0$, one is simply studying critical points of area subject to a volume constraint so, as is well-known, one gets constant mean curvature as a condition of criticality. For $\gamma > 0$ one sees from (3.2) that the curvature will not actually be constant unless it happens that $\partial A$ is a level set of $v$ solving (1.1). This will be the case for a periodic lamellar structure but note, for example, that a sphere (or periodic array of spheres) will never be a critical point – unless one works, as in [42], within the ansatz of radial symmetry.

Next we present the second variation. A formal derivation of essentially the same formula was previously presented in the appendix of [33].

Theorem 3.3. (The Second Variation) Let $u$ be a stable critical point of $E_\gamma$ such that for $A := \{ u = 1 \}, \partial A$ is $C^2$ with mean curvature $H(x)$. Let $\zeta$ be any smooth function on $\partial A$,

$$\int_{\partial A} \zeta(x) d\mathcal{H}^{n-1}(x) = 0.$$

Then for $v$ solving $-\Delta v = u - m$ in $\mathbb{T}^n$ one has the condition

$$J(\zeta) := \int_{\partial A} (|\nabla_{\partial A} \zeta(x)|^2 - \|B_{\partial A}\|^2 \zeta^2) d\mathcal{H}^{n-1}(x) + 8\gamma \int_{\partial A} \int_{\partial A} G(x, y) \zeta(x) \zeta(y) d\mathcal{H}^{n-1}(x) d\mathcal{H}^{n-1}(y) + 4\gamma \int_{\partial A} \nabla v(x) \cdot \nu(x) \zeta^2(x) d\mathcal{H}^{n-1}(x) \geq 0. \quad (3.3)$$

where, $\nabla_{\partial A} \zeta$ is the gradient of $\zeta$ relative to $\partial A$; $B_{\partial A}$ is the second fundamental form of $\partial A$, \text{i.e.} $\|B_{\partial A}\|^2 = \sum_{i=1}^{n-1} \kappa_i^2$, $\kappa_1, \ldots, \kappa_{n-1}$ principal curvatures; and $\nu$ is the unit normal to $\partial A$ pointing out of $A$.

Remark 3.4. (i) We briefly mention the construction of admissible perturbations. The construction is very similar to [48]). We let $X : \mathbb{T}^n \to \mathbb{R}^n$ be $C^2$ with $\int_{\partial A} X \cdot \nu = 0$, and let $\Psi : \mathbb{T}^n \times (-\tau, \tau) \to \mathbb{T}^n$ solve

$$\frac{\partial \Psi}{\partial t} = X(\Psi) \text{ \quad } \Psi(x, 0) = x.$$

Defining $A_t := \Psi(A, t)$, one finds that the perturbation does not (instantaneously) preserve volume up to second order, in fact:

$$\frac{d^2}{dt^2}_{|t=0} |A_t| = \int_{\partial A} (\text{div } X(x))(X(x) \cdot \nu) d\mathcal{H}^{n-1}(x).$$

Hence we modify explicitly $A_t$ via distance function to achieve the admissible perturbation.
(ii) The second variation is computed about a critical point of (NLIP) which is in general, a non-CMC surface. Hence perimeter term contributes an additional:

\[(n - 1)^2 \int_{\partial A} (H - \bar{H}) H \zeta^2 d\mathcal{H}^{n-1}(x),\]

where \(\bar{H}\) denotes the average of \(H(x)\) over \(\partial A\). This term is exactly cancelled when computing the second variation of the nonlocal term.

(iii) We note that the term in the second line of (3.3) is always positive. Hence to apply (3.3) for structures with CMC, one is simply comparing the gradient term of (3.3) to the term involving \(v\). Note that the latter generates the instability: it is negative for highly oscillatory \(\zeta\). Quantifying this comparison yields certain basic statements about, for example, the stability and instability of periodic lamellar structures (see [15]). What would be more interesting is to apply (3.3) to structures whose curvature (or mean curvature) is not constant.

4 The Modified Cahn-Hilliard Equation

It is well-known (see for example [18]) that the Cahn-Hilliard equation can be derived as a gradient flow with respect to the \(H^{-1}\) norm of the Cahn-Hilliard energy (the functional in (PCH)). Ignoring for the moment that the Cahn-Hilliard equation can also be derived from more basic physical principles ([9]), one can compute the same \(H^{-1}\) gradient flow for (NLCH), obtaining the PDE

\[u_t = \Delta \left(-\epsilon^2 \Delta u - u + u^3\right) - \sigma(u - m).\]

(4.4)

We refer to the (4.4) as the Modified Cahn-Hilliard Equation. Note that with the gradient flow computed with respect to \(H^{-1}\), the nonlocal term in NLCH only adds a zero-th order perturbation to the Cahn-Hilliard Equation. This new term, however, now drives the solution to have spatial average \(m\). Consider (4.4) on the unit torus (i.e. implement the appropriate periodic boundary conditions). As with the Cahn-Hilliard equation, (4.4) preserves the total mass \(\int_{\Omega} u\) of the solution provided the initial data \(u_0\) satisfies \(\int_{\Omega} u_0 = m\). Otherwise, one readily sees that the total mass will adjust to \(m\) exponentially fast.

4.1 Simulation of Steady States

One can readily use (4.4) to simulate steady states in 2D. Such simulations have appeared throughout the literature, perhaps the earliest ones were in [7]. For a suitable range of \(m\), one can either start with random initial data or with the constant state \(m\), randomly perturbed by noise. One simulates the expected stripes and spots with interesting geometries traversed on the path to steady state. Figures 3 and Figures 4 show a few such experiments using a pseudo spectral code written by J.F. Williams at SFU.
Figure 3: 2-D simulation with $\epsilon = 0.8, m = 0.5, \sigma = 4$ on a torus of size $2\pi$ with random initial data.

Figure 4: 2-D simulation with $\epsilon = 0.8, m = 0.5, \sigma = 4$ on a torus of size $2\pi$ with single disk initial data.
While the situation in 3D is interesting in its own right, it is also important from the point of view of the diblock copolymer application. The phase diagram for microphase separation has received much attention. Here one seeks to predict the general qualitative nature of the structure (spherical, lamellar, gyroid-like) for the given material parameters. There are three such parameters which describe a diblock copolymer melt: the Flory-Huggins interaction parameter $\chi$, the index of polymerization $N$, and the molecular weight $f = \frac{m+1}{2}$. The exact relationship between these dimensionless parameters and $\epsilon$, $\sigma$ and $m$ is given in [12].

For the phase diagram, the state of art seems to be via the self-consistent mean field theory (cf. [31, 6]), whereby given an ansatz for the basic structure (eg. spherical) with one or two degrees of freedom, one is able to compute the free energy and minimize with respect to the degrees of freedom. One then compares the optimal result within each known ansatz to determine the overall winner. From [12], we see that the variational problem (NLCH) is a consequence of the self-consistent mean field theory and a further crucial linearization step. The latter step has created skepticism as to whether or not the essential physics is preserved in the intermediate to strong segregation regime, wherein the interface thickness is relatively small. My own belief, is that, yes indeed this linearization step throws away a lot of information; however, from the point of view of determination of the phase diagram, all the phases which have been (numerically) predicted purely from the SCMFT (cf. [31]), can be (numerically) generated from the (4.4) theory within a far less ansatz-driven framework. That is, in the end, one only needs to keep a rather crude approximation of the polymer A- and B-chain interactions, in order to determine the basic qualitative geometry for a given set of material parameters.

3D simulations for (4.4) were begun by Teramoto and Nishiura in [49], simulating both gyroids and double-gyroids. However, to my knowledge, no thorough phase diagram calculation has been done via the gradient theory approach of (4.4), and in particular, none within the context of the material parameters $\chi, N, f$ so that one can make direct comparison with both the phase diagram of [31], and with experiments (cf. [20]). A preliminary investigation using the code from J. Lowengrub’s group (Irvine) suggests that his adaptive, finite difference code may work well here. We give one example in Figure 5, a full investigation is in progress.

**Remark 4.1.** Following on Remark 3.2, the steady states which one attempts to simulate in Figures 3 amd 4 should not be exactly circular: For a rectangular domain, criticality alone precludes interfaces having non-zero constant curvature. This could be either a resolution issue and in fact minimizers look more like the wiggled spots of [42]-[45], or simply that these perturbations from constant curvature (in general, CMC) are exponentially small. Investigation of these issues is currently in progress with J.F. Williams (SFU). It might also be interesting for 3D simulations (like the gyroid of Figure 5) to place the final structure into the *Surface Evolver* ([8]) and see qualitatively and quantitatively the perturbation from area-minimizing.
4.2 Dynamics to Steady States: Questions on Spinodal Decomposition and Coarsening

The dynamics associated with the Cahn-Hilliard equation has received a lot of attention. For an appropriate regime on the mass constraint $m$, and starting from the constant state, the dynamics is essentially described via two distinct temporal regimes (see for example [38]):

Stage I (short time scale): Nucleation/spinodal decomposition: here a fine grained mixture of the two phases appears and fairly sharp interfaces between the two phases are formed.

Stage II (longer time scale): Interface coarsening via the Mullins-Sekerka law.

In this latter stage, there is a well-established coarsening rate of $t^{1/3}$, i.e. in Stage II, the length scale of the phases coarsens like $t^{1/3}$.

The asymptotic dynamics as $\epsilon$ tends to zero are also well understood: solutions converge to the solutions of the Mullins-Sekerka free boundary problem (see for example [38, 4]).

For (4.4), the third (long-range term) changes the dynamics but analogous issues remain. Starting with a homogeneous state $m$, there are now three competing mechanisms for the dynamics: (i) Nucleation/spinodal decomposition/formation of phases (still on a fast scale), (ii) setting of the long-range periodicity scale, (iii) coarsening within this scale. A preliminary asymptotic study of first phase was begun in [36]. Naturally the $\sigma$ term plays a role here when it is sufficiently large – it restricts both the spinodal regime (wherein the homogeneous state is unstable to phase separation) and the extent of the phase separation – no longer will completely pure phases of $u = \pm 1$ form.

The next stage of coarsening is more interesting. A (final) length scale is set by $\sigma$ preventing the coarsening from going on forever. It would be interesting to see if there is some temporal regime associated with a new a scaling law for the basic length scale as a function of time – this exponent should depend on $\sigma$. Certainly for this later stage, one
can use the appropriately scaled asymptotic dynamics (as \( \epsilon \) tends to zero) which was initially studied by Nishiura and Ohnishi in [34] (see also [19, 17, 21]). Here one obtains the following modified Mullins-Sekerka free boundary problem. That is, setting \( A^+ = \{ u = 1 \} \), \( A^- = \{ u = -1 \} \), and \( f = \frac{m+1}{2} \), the evolution of \( \partial A^+ \) is described via the free boundary problem for \( \partial A^+ \) and scalar velocity field \( v \):

\[
\begin{align*}
\Delta v &= \gamma \begin{cases} 1 - f & \text{in } A^+ \\ -f & \text{in } A^- \end{cases} \\
v &= H \quad \text{on } \partial A^+ \\
V_n &= \left[ \frac{\partial v}{\partial n} \right]^+ \quad \text{on } \partial A^+ ,
\end{align*}
\tag{4.5}
\]

where the normal velocity \( V_n \) and mean curvature \( H \) are computed with respect to the outward normal to \( A^+ \). This free boundary problem can naturally be connected with (NLIP).

References


