Coupled Surface Diffusion and Motion by Mean Curvature from a Diffuse Interface Model

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Abstract

A degenerate Allen-Cahn/Cahn-Hilliard system which was developed to describe simultaneous ordering and phase separation, can also be viewed as a diffuse interface approximation for various problems in materials science in which surface diffusion and motion by mean curvature are coupled. In the original context the low temperature coarsening limit which yields geometric motion in the limit was designed to describe small particles of a disordered phase whose shape evolves by surface diffusion which are embedded along grain boundaries which partition the system into two ordered variants. While an early analysis focused on systems in the proximity of the complete wetting limit [12], a later analysis extended these results to the more generic partial wetting setting [14]. We outline here some determining features into whether a given degenerate Allen-Cahn/Cahn-Hilliard system will be describing complete or partial wetting.

Introduction

Let us consider the degenerate Allen-Cahn/Cahn-Hilliard system

$$(\mathbf{dAC/CH}) \quad \begin{cases} u_t &= 4\epsilon^2 \nabla \cdot [Q(u,\, v)\nabla \mu], \\ \mu &= F_u(u,\, v) - \epsilon^2 \triangle u, \\ v_t &= -\frac{1}{4} Q(u,\, v)[F_v(u,\, v) - \epsilon^2 \triangle v], \end{cases}$$

for $(x,\, t) \in \Omega \times (0,\, T)$ where $\Omega \subset R^3$ and $0 < T < \infty$, with the boundary conditions

$$\mathbf{n} \cdot Q(u,\, v)\nabla \mu = \mathbf{n} \cdot \nabla u = \mathbf{n} \cdot \nabla v = 0,$$

for $(x,\, t) \in \partial\Omega \times (0,\, T)$, which was developed in [6] to model simultaneous phase separation and ordering in binary alloys. Here u denotes an average concentration, v represents a non-conserved order variable, $Q(u, v) \geq 0$ is a mobility, and F is a homogeneous free energy with both entropy and energy terms. The degeneracies of the system derived from the fact that the mobility $Q(u,\, v)$ will be assumed to vanish for certain values of $(u,\, v)$ which correspond to *pure phases*. The remaining assumptions on the form of the functions $Q(u,\, v)$ and $F(u,\, v)$ can reflect both the physical framework which pertain to the binary system under consideration as well as our other goal which is to consider the (dAC/CH) system as a diffuse interface model for coupled motion by mean curvature

and motion by surface diffusion. In the context of binary alloys coupled motion by mean curvature and motion by surface diffusion occurs the coarsening regime in which small grains or particles of disordered phase, whose surface evolves under motion by surface diffusion, are embedded within grain boundaries that are moving by mean curvature. In this context the surface of the disordered grains are known as interphase boundaries (IPBs) and the grain boundaries are known as anti-phase boundaries (APBs). Coupled surface diffusion and motion by mean curvature also occurs in materials science in other contexts, such as in grain boundary motion where the grain boundary is connected to an external surface via a thermal groove and as well as in sintering under suitable assumptions. According to Young's law it is the relative size of m, the ratio between the surface energy of the grain boundary to the surface energy of the IPB which determine the angles at triple junctions where two IPBs connect up with an APB. If $m = 2$, the angle between the two IPBs is zero, which corresponds to complete wetting. If $0 < m < 2$, this angle is greater than zero, which corresponds to partial wetting.

In the present note we outline the formal asymptotics used in the recent papers [12] and [15] where this geometric description was achieved for the first time, which lead one to consider the (dAC/CH) system as a diffuse interface model for coupled motion by mean curvature and motion by surface diffusion, explaining in particular why the former corresponded to a system in the proximity to a complete wetting limit and why the latter succeeds in capturing the more generic situations.

The structure of the (dAC/CH) system

Let us now consider the structure of the degenerate Allen-Cahn/Cahn-Hilliard system, (dAC/CH), and the boundary conditions given in the Introduction, where u, v, and μ are scalar functions representing respectively the concentration, the order parameter, and the chemical potential, and

$$\vec{j} := -Q(u, v)\nabla\mu \tag{1}$$

expresses the mass flux. With regard to the boundary conditions, the first boundary condition represents a no flux boundary condition; the second and third reflect "free" or energy minimizing boundary conditions.

In (dAC/CH), $F(u, v)$ is a *homogeneous free energy* which is assumed to be of the form

$$F(u, v) = \frac{1}{2}\Theta[G(u + v) + G(u - v)] + E(u, v), \tag{2}$$

where Θ is a dimensionless temperature,

$$G(s) = s \ln s + (1 - s) \ln(1 - s)$$

is the entropy density of the system, and E is the interaction energy density of the system which we take to be polynomial,

$$E = \sum_{i,\, j=0}^{k} a_{ij}\, u^i v^j. \tag{3}$$

We remark that in the existence proof in [7], E was taken as a specific quadratic polynomial. The choice of the coefficients a_{ij} reflect the physical properties being modelled by the (dAC/CH) system.

In order that $F(u,\, v)$ be well defined, it is necessary that $(u,\, v) \in B$, where

$$B := \{\, (u,\, v) \;\mid\; 0 < u - v < 1, \quad 0 < u + v < 1 \,\}.$$

In the asymptotics in [12, 14] it was assumed that there exist solutions with some minimal regularity which lie in B. Off hand, this is not so easy to guarantee or to prove. However, at least in one dimension, it was proven in [7] that if the initial data is in $\bar{B}$ and $\overline{u} \in (0,\, 1)$, where $\overline{u}$ denotes the average concentration, $\overline{u} := \frac{1}{|\Omega|}\int_\Omega u\, dx$ and $Q(u,\, v)$ satisfied some degeneracy assumptions, that this is true.

The corners of $\overline{B}$, $\{(0,\, 0),\, (1,\, 0),\, (1/2,\, -1/2),\, (1/2,\, 1/2)\}$ can be considered as representing the *pure phases* mentioned in the Introduction, where $(0,\, 0)$ and $(1,\, 0)$ are pure one component phases and $(1/2,\, \pm 1/2)$ are purely ordered phases. The mobility, $Q(u,\, v)$, is assumed to be degenerate at these corners, or more specifically, one can assume that

$$Q(u,\, v) = P(u,\; v)\widetilde{Q}(u,\, v), \tag{4}$$

where $P(u,\, v) = u(1-u)(\frac{1}{4} - v^2)$ and $\widetilde{Q}(u,\, v)$ is smooth and does not non-vanishing throughout $\overline{B}$. Note that $P(u,\, v)$ has simple roots at the corners of $\overline{B}$. Assuming that v represents an ordering variable and that $v \to -v$ represents the exchange of one phase variant by another energetically and kinetically equivalent one, $F(u,\, v)$ and $Q(u,\, v)$ can be assumed to be even functions v. For simplicity the interaction energy $E(u,\, v)$ can be assumed to be concave with $\mathcal{O}(1)$ coefficients. Moreover, since (dAC/CH) is supposed to model ordering and phase separation, there should be at least two ordered phases and a disordered phase present in the system. This can be accomplished by assuming that $E(u,\, v)$ has two global minimizers located at $(1/2,\, \pm 1/2)$ and two minimizers located at $(0,\, 0)$ and $(1,\, 0)$ which are local but not global minimizers. This implies that when Θ is positive and sufficiently small, $F(u,\, v)$ has 4 minimizers within B which are located transcendentally close, $\mathcal{O}(e^{-c/\Theta})$, to the corners of $\overline{B}$. When Θ is small and positive, there exists a Lagrange multiplier so that the "tilted" free energy $F(u,\, v) + \lambda u$ has exactly three equal depth global minimizers within B which we denote by $\alpha_\pm$ and $\alpha_\pm$ which transcendentally close ($\mathcal{O}(e^{-c/\Theta})$) to the corners of $\overline{B}$, with $\alpha_\pm$ near $(1/2,\, \pm 1/2)$ and α_0 near one of the other corners. We refer to $\alpha_\pm$ as the "ordered phase variants", and to α_0 as the "disordered phase." For simplicity, we set $F + \lambda u \to F \to F$, and refer to F as "the free energy," and without loss of generality, we may set $F = 0$ at $\{\alpha_0,\, \alpha_\pm\}$.

The location of the equilibria and the structure of the free energy surface will play a critical role in determining the behavior of the limiting solutions. In particular, whether the analysis is to predict complete or partial wetting is determined by the properties of certain energy minimizing paths connected the global minimizers of $\alpha_\pm$. We shall elaborate on this point a little later.

The asymptotics

Since a long time coarsening limit is to be described, a slow time scale needs to be introduced which in [12, 15] was found could be taken as $\tau = \epsilon^{7/2}t$. Writing (dAC/CH) in terms of the variable τ and setting $\tau \to t$ for convenience, we obtain

$$\begin{aligned} \epsilon^{5/2}u_t &= 4\epsilon\nabla\cdot[Q(u,\, v)\nabla\mu_u], \\ \mu &= F_u(u,\, v) - \epsilon^2\triangle u, \\ \epsilon^{7/2}v_t &= -\tfrac{1}{4}Q(u,\, v)[F_v(u,\, v) - \epsilon^2\triangle v]. \end{aligned} \tag{5}$$

To consider a low temperature limit, we take $\Theta = \mathcal{O}(\epsilon^{1/2})$, or more specifically,

$$\Theta = \epsilon^{1/2}.$$

We remark that limiting equations of motion for the degenerate Cahn-Hilliard equation were obtained in [5] both in the deep quench limit when $\Theta = 0$ as well as when $0 < \Theta << 1$. To obtain small particles of the disordered phase in the limiting description, we assume that the disordered phase constitutes a minor phase; i.e., that the volume fraction of Ω which is occupied by the disordered phase is small. This is further reflected the assumption

$$(\kappa_i)_{IPB} = \mathcal{O}(\epsilon^{-1/2}), \quad i = 1, 2, \tag{6}$$

where $(\kappa_i)_{IPB}$, $i = 1, 2$ denote the principle curvatures at points along IPBs, which states that the curvature of the particles which contain the disordered phase is very large. Since (5) is mass conservative, the mean concentration of the system as a whole must be close to $1/2$, the concentration of the ordered phase variants. This is quantified by setting

$$1/2 - \bar{u} = \mathcal{O}(\epsilon^{n/2}),$$

where n denotes the dimension of the domain; i.e., $\Omega \subset R^n$. In [12], the case $n = 2$ was considered and in [15], the analysis was given in terms of $n = 3$ though it was relevant to the case $n = 2$ as well. The dependence on the dimension n is required for consistency with (6). In parallel with (6), we set

$$(\kappa_i)_{APB} = \mathcal{O}(\epsilon^{3/2}), \quad i = 1, 2, \tag{7}$$

where $(\kappa_i)_{APB}$, $i = 1, 2$ denote the principle curvatures at points along APBs, which asserts that the curvature of the APBs is very small.

The strategy of the asymptotics is as follows. As in e.g [3, 4] to describe the dynamics in the long time coarsening regime, we consider separately first the behavior of solutions in the "outer region" lying away from the IPBs and APBs, then the behavior of solutions along "inner regions" encasing the IPBs and APBS , using the outer solution for matching. Afterwards the solutions are studied at the triple junctions and where IPBs and APBs intersect $\partial\Omega$.

In the outer region, u and v are taken to assume their values at one of the minimizers, $\{\alpha_0, \alpha_\pm\}$ up transcendentally small corrections, and the flux j is assumed to be transcendentally small. Moreover, μ, and $\mu_v = F_v(u, v) + \epsilon\triangle v$ are assumed to possess regular perturbation expansions in $\epsilon^{1/2}$. These assumptions can shown to be self-consistent, and μ and μ_v can be seen to be $\mathcal{O}(\epsilon^{1/2})$.

The inner regions along IPBs and APBS are assumed to have an $\mathcal{O}(\epsilon)$ width, and regular perturbation expansions in $\epsilon^{1/2}$ are assumed for u, v, j, and μ, as well as for W, the normal velocity. A locally defined orthonormal system, (r, s), is defined, where $r = r(x, t)$ measures normal distance from the mid-surface of the inner region, and $s(x, t) = (s_1(x, t), s_2(x, t))$ corresponds to an orthonormal frame relative to the mid-surface. Afterwards, a scaled variable, $\rho = r/\epsilon$, is introduced. In [15] a normalization condition used in defining the mid-surface is somewhat different for IPBs than for APBs. The asymptotics for IPBS and for the APBs are somewhat similar, but the normalization condition (**??**) which is used for APBs allows us to demonstrate that within the APB inner region, u is an odd function of ρ and v is an even function of ρ, and this symmetry is used extensively in the asymptotic analysis which follows. In [12], the asymptotic analysis along APBs assumed oddness and evenness of u and v, respectively. In both cases, the analysis is somewhat technically involved,

and there is an integration by parts gives rise to many terms, though eventually most all terms can be estimated and shown to be transcendentally small.

In [15] along triple junction lines, a prism is constructed with height ϵ^α and cross-sectional dimensions proportional to ϵ^β, where $7/8 < \beta < 1$ and $\alpha = 2\beta$. Similarly, at intersections of IPBs and APBs with the exterior boundary, a parallelogram construction is employed with similar height and cross-sectional dimensions. It is assumed here that the behavior of the solution in the vicinity of triple junction lines and intersection of IPBs and APBs with the exterior surface is no more singular than elsewhere in Ω. The treatment in [12] is similar, though simpler since it is lower dimensional.

For a summary of the results, see Figure .

Wetting features for some free energies

Some conclusions

the deep quench limit where $\Theta = 0$ is singular.

The form which will be adopted for the entropy contribution will reflect the fact that in the original derivation both $u - v$ and $u + v$ acted as concentration variables. Additional assumptions will be imposed on F in order to guarantee that the system will evolve in a partial wetting regime. Roughly speaking, what is necessary in this context is to require that certain weighted geodesics, which connect the ordered phases on the surface defined by $\sqrt{F(u, v)}$, do not pass via an intermediary globally energy minimizing pure phase, see [13]. We remark that our analysis neglects effects which could lead to anisotropy or grain rotation; we do this for the sake of simplicity in order to allow us to focus on asymptotics which couple the two types of motions. We will amplify our assumptions further in the section which follows. For a discussion of models which are similar and related to AC/CH, we refer the reader to Eguchi & Ninomiya [8] and Chen et. al. [**?**, **?**].

The asymptotic analysis leading to the coupling of motion by mean curvature and surface diffusion is somewhat delicate even in the partial wetting case, since rough time scale arguments would lead one to expect motion by mean curvature to evolve on a much faster scale than motion by surface diffusion, hence their coupling on the same time scale would be seemingly inconsistent. However, the coupling becomes possible when the "aspect ratio" (given by the ratio of a typical length scale for the volumes which evolve by surface diffusion to a typical length scale for the volumes which evolve by motion by mean curvature) is sufficiently small. Ideally, one should like to obtain predictions for the manner in which the length scales of the system increase with time during the long time coarsening regime, such as those obtained for example, by Kohn & Otto [**?**] in the context of the Cahn-Hilliard equation in the deep quench limit. Though this has so far not been accomplished in the present context, this may be possible to undertake within the framework of the limiting equations which we obtain here. We remark that some degree of "self-similarity" in length scale growth must be assumed in order to guarantee the validity of the "aspect ratio" assumption described above during coarsening.

Ideally we should like to be able to rigorously prove a connection between the dynamics of AC/CH and the limiting asymptotic motions which we derive. We note that the connection between the dynamics of diffuse interface equations and limiting asymptotics motions has been rigorously es-

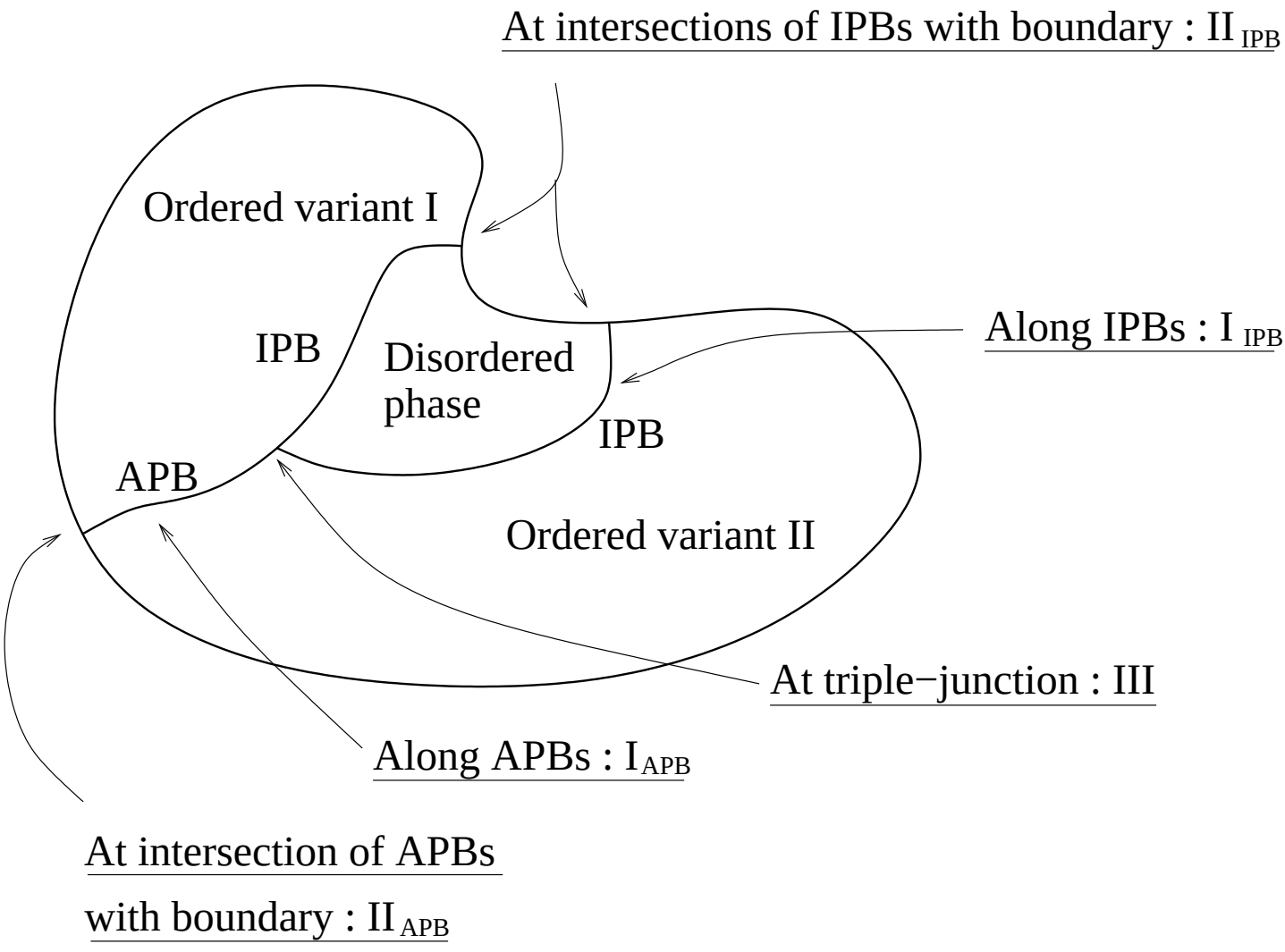

Figure 1: The limiting dynamics

tablished in a number of contexts, see for example Caginalp & Chen [4] and references therein. We point out that in order to accomplish such a program in the present context, a number of nontrivial steps are required. In particular, existence and regularity must be established both for the AC/CH system and for the limiting equations of motion which we obtain.

This has been accomplished for example in [9] for the limiting motion which arises from a system of degenerate Cahn-Hilliard equations.

However, once a clear connection is made between the AC/CH diffuse interface model and the limiting motions as explained in the sections which follow, even though the connection has yet to become rigorously established, the AC/CH system can be looked upon as an approximation of the geometric problem. As such, the system can be implemented as a numerical tool for studying the limiting geometric problem.

Notably, fully practical numerical schemes have been recently developed for the AC/CH system both for the degenerate and the non-degenerate cases [1, 2].

The outline of this paper is as follows. In §2, the basic assumptions and notation for our analysis are given, and the strategy for the asymptotic analysis is presented. *Assumptions in the text are highlighted in italics for the reader's convenience.* In §3, specific ansatzes are introduced for the asymptotic expansions in the outer solution and are demonstrated to be self-consistent. Section §4 contains the derivation of the limiting motions for APBs and IPBs. The laws governing triple junctions are derived in §5, and the laws governing the intersections of APBs and IPBs with the external boundary, $\partial\Omega$, are obtained in §6. In §7, our results are summarized for the limiting geometric problem. We invite the interested reader to turn now to Figure **??** in §7 for a rapid overview of these results.

Preliminaries

We shall assume that the coefficients in (3) are such that there exists a unique minimizing path connecting $\alpha_\pm$ where

$$\liminf_{(u,\,v)\in A_\pm} \int_0^1 \sqrt{F(u,\,v)\{\dot{u}^2+\dot{v}^2\}}\,dt, \tag{1}$$

is attained, where

$$A_\pm := \{\,(u,\,v)\in C^1[0,\,1],\,(u(0),\,v(0))=\alpha_\mp,\,(u(1),\,v(1))=\alpha_\pm,\,(u(t),\,v(t))\in B\quad \forall t\in[0,\,1]\},$$

which does not pass via the global minimizer, α_0. Were the geodesic to pass via α_0, it would be implied (see [13]) that the surface energy of the transition from one phase variant to the other was precisely equal to twice the surface energy of a transition from one of the phase variants to to the disordered phase, and this would correspond to the complete wetting case which is not being treated here. *We shall, in fact, make the slightly stronger assumption that the geodesic path stays boundedly away from α_0.* This assumption can be interpreted as saying that our system will be assumed to be in the generic partial wetting regime and not asymptotically close to complete wetting.

Because the geodesic is an energy minimizing path, and because of the $v\to -v$ symmetry which has been assumed for $F(u,\,v)$, it is easy to show that the geodesic path possesses $v\to -v$ symmetry

and crosses the line $v = 0$ in B only once, since otherwise the energy as defined in (1) may be lowered if this is not the case.

We call to the reader's attention that in the partial wetting case, the geodesic may or may not pass via a local minimizers. If the geodesic passes via a local minimizer, then the system is said to be prewetting. Otherwise, there is said to be absorption, unless there is no variation at all in concentration along the transition from one phase variant to the other. Our analysis should be applicable both when there is prewetting and when there is absorption. *We shall further assume that the geodesic path stays* $\mathcal{O}(1)$ *away from* $(1,\, 0)$. Such an assumption is not unreasonable, since even in the prewetting case, under quite minimal assumptions on $F(u,\, u)$, the geodesic path can be shown not to cross a local minimizer located near $(1,\, 0)$, if such a local minimizer exists. Suppose, for example, that we were to assume that in addition to the $v \to -v$ symmetry, prior to "tilting" by the Lagrange multiplier λ, the free energy $F(u,\, v)$ also exhibits $u \to 1/2 - u$ symmetry. Then it is readily seen that *if* $0 < \bar{u} < 1/2$ *as we shall assume,* an energy minimizing geodesic cannot come too close to $(1,\, 0)$, since this would imply that one could readily construct a path lying to the left of $u = 1/2$ with lower energy, which would contradict the assumption that the original path described was itself energy minimizing.

For the analysis which follows, *we shall also make use of the minimizing paths connecting* $\alpha_\pm$ *with* α_0. Analytically, these are the paths where

$$\liminf_{(u,\, v) \in A_0^\pm} \int_0^1 \sqrt{F(u,\, v)\{\dot{u}^2 + \dot{v}^2\}}\, dt$$

is attained, where

$$A_0^\pm := \{\, (u,\, v) \in C^1[0,\, 1],\, (u(0),\, v(0)) = \alpha_0,\, (u(1),\, v(1)) = \alpha_\pm, \quad (u(t),\, v(t)) \in B \quad \forall t \in [0,\, 1]\}.$$

We shall assume this minimizing path as well as the minimizing path described earlier to be uniformly bounded an $\mathcal{O}(e^{-c/\Theta})$ *distance away from* ∂B. Such an assumption is easy to justify based on energetic considerations when $0 < \Theta \ll 1$. We remark that the limit $\Theta \to 0$ is a singular limit, known in the literature [16] as the deep-quench limit. In this limit, the minimizers of F are located at the corners of $\overline{B}$, and the geodesics may be totally or partially contained in ∂B.

We point out to the reader that inherently the partial wetting case is far more generic than the complete wetting case. In [13], it is demonstrated analytically that for interaction energies of the form $E(u,\, v) = \alpha u(1-u) - \beta v^2$ with $0 < \alpha < \beta$ which were employed in [12], complete wetting is indeed implied when $\bar{u} \in (0,\, 1/2) \cup (1/2,\, 1)$ at $\Theta = 0$. Moreover, some simple examples are given of interaction energies which imply partial wetting.

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