

Spinodal Decomposition for Multicomponent Cahn–Hilliard Systems

Stanislaus Maier-Paape,¹ Barbara Stoth,² and Thomas Wanner³

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We consider the initial-stage phase separation process in multicomponent Cahn–Hilliard systems through spinodal decomposition. Relying on recent work of Maier-Paape and Wanner, we establish the existence of certain dominating subspaces determining the behavior of most solutions originating near a spatially homogeneous state. It turns out that, depending on the initial concentrations of the alloy components, several distinct phenomena can be observed. For ternary alloys we observe the following two phenomena: If the initial concentrations of the three components are almost equal, the dominating subspace consists of two copies of the finite-dimensional dominating subspace from the binary alloy case. For all other initial concentrations, only one copy of the binary dominating subspace determines the behavior. Thus, in the latter case we observe a strong mutual coupling of the concentrations in the alloy during the initial separation process.

KEY WORDS: Multicomponent Cahn–Hilliard equation; spinodal decomposition; multicomponent alloys.

1. INTRODUCTION

Forty years ago J. W. Cahn and J. E. Hilliard^(3, 4) introduced the partial differential equation

$$\begin{aligned} u_t &= -\Delta(\varepsilon^2 \Delta u + f(u)) && \text{in } \Omega \\ \frac{\partial u}{\partial \nu} &= \frac{\partial \Delta u}{\partial \nu} = 0 && \text{on } \partial \Omega \end{aligned} \tag{1}$$

Dedicated to John W. Cahn on the occasion of his 70th birthday.

¹ Institut für Mathematik, Universität Augsburg, 86135 Augsburg, Germany; e-mail: maier@math.uni-augsburg.de.

² Institut für Angewandte Mathematik, Universität Bonn, Wegelerstraße 6, 53115 Bonn, Germany; e-mail: bstoth@iam.uni-bonn.de.

³ Department of Mathematics and Statistics, University of Maryland, Baltimore County, 1000 Hilltop Circle, Baltimore, Maryland 21250; e-mail: wanner@math.umbc.edu.

to model phase separation of a binary alloy contained in a closed vessel $\Omega \subset \mathbb{R}^n$, where $n \in \{1, 2, 3\}$. Their studies were motivated by a phenomenon called *spinodal decomposition*: If a high-temperature homogeneous mixture of two metals is quenched to a certain lower temperature, a sudden phase separation process may set in. The mixture quickly becomes inhomogeneous, forming a fine-grained structure which more or less alternates in properties between the two components. In two and three dimensions, these structures exhibit a complex, irregular geometry, yet with some common characteristic wavelength. This characteristic wavelength appears to be proportional to the small parameter ε in (1) which models interaction length. Subsequently, a coarsening process can be observed, for which the patterns generated by spinodal decomposition serve as initial conditions. For more details on this coarsening process for binary as well as multicomponent alloys we refer the reader to Alikakos, Bates, Chen,⁽¹⁾ Bronsard, Garcke, Stoth,⁽²⁾ Kalies, VanderVorst, Wanner,⁽¹²⁾ Pego,⁽¹⁶⁾ Stoth,⁽¹⁹⁾ and the references therein.

Although spinodal decomposition has drawn considerable interest in the physics community, mathematical treatments of the phenomenon appeared only recently. The first results are due to Grant,⁽⁹⁾ who considers the case of one-dimensional domains Ω . Maier-Paape and Wanner^(13, 14) explain the early stages of spinodal decomposition for general domains Ω , including the geometries and characteristic wavelengths observed in experiments. Only recently, progress has been made in explaining the subsequent stages of spinodal decomposition. See Sander and Wanner.^(17, 18) However, all of these results exclusively consider the scalar-valued Cahn–Hilliard equation (1), i.e., only binary alloys.

Nonetheless, experimental evidence shows that spinodal decomposition can also be observed in multicomponent alloys. Similar to the binary case, a (almost completely) homogeneous mixture of $N \geq 3$ components will quickly separate into a complex structure with some characteristic wavelength. In order to describe this phenomenon, Morral and Cahn⁽¹⁵⁾ introduced a system of partial differential equations which will be described in more detail in the following (see also Hoyt^(10, 11)). Morral and Cahn also provided a heuristic explanation for the decomposition process by discussing the linearized dynamics. More recently, Eyre^(7, 8) performed numerical simulations to shed more light onto multicomponent spinodal decomposition. He observes that the first decomposition stage behaves very similar to the binary case, and consequently refers to the obtained structure as *pseudo-binary*. To the best of our knowledge, there does not exist any rigorous mathematical explanation for this.

In the following, we will provide such an explanation for the early stages of the decomposition process. This is done by carrying over the

results of Maier-Paape and Wanner^(13, 14) to the multicomponent case. In the course of this, we will not only explain the occurrence of the pseudo-binary as observed by Eyre, but also characterize in detail when the pseudo-binary does not appear. These two different phenomena are described at the end of the introduction in Theorems 1.3 and 1.2, respectively.

Consider an alloy consisting of $N \geq 3$ components which is contained in some closed vessel $\Omega \subset \mathbb{R}^n$, where $n \in \{1, 2, 3\}$. Let $u_i(t, x) \in [0, 1]$, $i = 1, \dots, N$, denote the concentration of the i th component at time t and location $x \in \Omega$. Then we have

$$\sum_{i=1}^N u_i(t, x) = 1 \quad \text{for all } t \geq 0 \text{ and } x \in \Omega$$

i.e., the values of the vector $u = (u_1, \dots, u_N)$ describing the composition of the alloy are contained in the *Gibbs simplex* \mathcal{G} defined by

$$\mathcal{G} := \left\{ v \in \mathbb{R}^N : \sum_{i=1}^N v_i = 1, v_i \geq 0, i = 1, \dots, N \right\}$$

With every state $u \in H^1(\Omega, \mathbb{R}^N)$ of the alloy one can associate an energy $E_\varepsilon[u]$ given by

$$E_\varepsilon[u] := \int_{\Omega} \left(\frac{\varepsilon^2}{2} \cdot \sum_{i=1}^N |\nabla u_i|^2 + W(u) \right) dx \tag{2}$$

Since the parameter $\varepsilon > 0$ models interaction length, it is considered to be small. The nonlinear function $W: \mathcal{G} \subset \mathbb{R}^N \rightarrow \mathbb{R}$ represents the Gibbs free energy, one standard example being

$$W(u_1, \dots, u_N) = \sigma \cdot \sum_{i < j} u_i u_j + \sum_{i=1}^N u_i \ln u_i \tag{3}$$

Similar to the binary case we can use (2) to derive gradient dynamics for the concentration vector $u(t, x)$ which conserves the total mass, i.e., for which

$$\frac{1}{|\Omega|} \cdot \int_{\Omega} u_i(t, x) dx = \bar{u}_i \quad \text{for all } t \geq 0 \text{ and } i = 1, \dots, N \tag{4}$$

Define $\mathbf{e} = (1, \dots, 1) \in \mathbb{R}^N$, and let $P: \mathbb{R}^N \rightarrow \mathbf{e}^\perp$ denote the orthogonal projection onto \mathbf{e}^\perp , i.e., let $Pu = u - (u, \mathbf{e})/N \cdot \mathbf{e}$. Setting

$$f(u) := -PD_u W(u) : \mathcal{G} \subset \mathbb{R}^N \rightarrow \mathbf{e}^\perp \tag{5}$$

the dynamical law associated with (2) is given by the *multicomponent Cahn–Hilliard system*

$$\begin{aligned} u_t &= -\Delta(\varepsilon^2 \Delta u + f(u)) && \text{in } \Omega \\ \frac{\partial u}{\partial \nu} &= \frac{\partial \Delta u}{\partial \nu} = 0 && \text{on } \partial\Omega \\ u &\in \mathcal{G} \subset \mathbb{R}^N \end{aligned} \quad (6)$$

For more information we refer the reader to Morral and Cahn,⁽¹⁵⁾ Hoyt,^(10, 11) and Eyre.^(7, 8) An existence and uniqueness result for solutions of (6) can be found in Elliott and Luckhaus.⁽⁶⁾

In the rest of this introduction we want to state two theorems concerning the dominance of certain subspaces in the spinodal decomposition process. These subspaces are related to the linearized equations of (6). For fixed $\bar{u} \in \mathcal{G}$ denote the matrix $B = B_{\bar{u}} := -PD_u^2 W(\bar{u}) \in \mathcal{L}(\mathbb{R}^N, \mathbb{R}^N)$. Notice that $\text{im}(B) \subset \mathbf{e}^\perp$. Then the linearization of (6) at $u \equiv \bar{u}$ is

$$\begin{aligned} v_t &= -\Delta(\varepsilon^2 \Delta v + Bv) && \text{in } \Omega \\ \frac{\partial v}{\partial \nu} &= \frac{\partial \Delta v}{\partial \nu} = 0 && \text{on } \partial\Omega \\ (v, \mathbf{e}) &= 0 \end{aligned} \quad (7)$$

We try to find solutions of (7) of the form $v(t, x) = e^{\lambda t} \cdot w \cdot \psi(x)$, where $w \in \mathbf{e}^\perp \subset \mathbb{R}^N$ is an eigenvector of B and ψ is an eigenfunction of the negative Laplace operator. To be more precise, we use the following notation.

Notation 1.1. We denote by ψ_i , $i \in \mathbb{N}$, the complete $L^2(\Omega)$ -orthonormal set of eigenfunctions of the scalar-valued negative Laplace operator $-\Delta$ in $L^2(\Omega) \cap \{v: \int_\Omega v \, dx = 0\}$, subject to homogeneous Neumann boundary conditions. The corresponding eigenvalues are denoted by $0 < \kappa_1 \leq \kappa_2 \leq \kappa_3 \leq \dots \rightarrow +\infty$.

The \mathbb{R}^N -orthonormal eigenvectors of $B|_{\mathbf{e}^\perp}$ are denoted by $w_1, \dots, w_{N-1} \in \mathbf{e}^\perp$ with corresponding real eigenvalues $\beta_1 \geq \beta_2 \geq \dots \geq \beta_{N-1}$.

Notice that the eigenvalues β_k are real due to the symmetry of $B|_{\mathbf{e}^\perp} \in \mathcal{L}(\mathbf{e}^\perp, \mathbf{e}^\perp)$. For further reference, we state the following identity for the asymptotic distribution of the eigenvalues κ_i (cf. ref. 5 or 14, Lemma 3.1). For this, let $N_n(\mu)$ denote the number of eigenvalues κ_i of $-\Delta$ (subject to

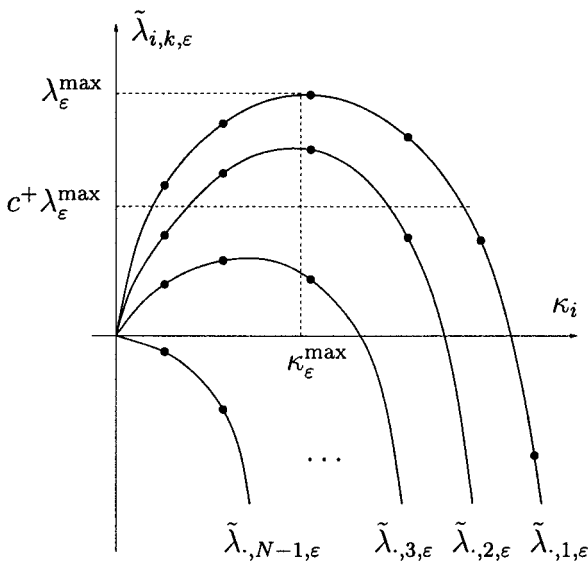


Fig. 1. The eigenvalues $\tilde{\lambda}_{i,k,\varepsilon}$

Neumann boundary conditions) less than $\mu \in \mathbb{R}$ (counting multiplicities), then

$$\lim_{\mu \rightarrow \infty} \frac{N_n(\mu)}{\mu^{n/2}} = c_n |\Omega| \tag{8}$$

with a positive constant c_n depending only on $n = \dim \Omega$.

Using the above notation, the eigenvalues of $A_\varepsilon u := -\Delta(\varepsilon^2 \Delta u + Bu)$, which is the right-hand side of the first equation in (7), are given by

$$\tilde{\lambda}_{i,k,\varepsilon} := \kappa_i(\beta_k - \varepsilon^2 \kappa_i), \quad i \in \mathbb{N}, \quad 1 \leq k \leq N-1 \tag{9}$$

with corresponding eigenfunctions $w_k \cdot \psi_i(x)$ (see Fig. 1). Observe that all eigenvalues are bounded above by $\lambda_\varepsilon^{\max} := \beta_1^2 / (4\varepsilon^2)$.

We want to assume that the equilibrium $u \equiv \bar{u}$ is unstable, i.e., that $\beta_1 > 0$. It then seems intuitively plausible that the eigenfunctions with eigenvalues $\tilde{\lambda}_{i,k,\varepsilon}$ close to $\lambda_\varepsilon^{\max}$ will contribute more to the spatial pattern formation of solutions originating near \bar{u} than the remaining eigenfunctions. Therefore we define

$$\mathbb{Y}_\varepsilon^+ := \text{span}\{w_k \cdot \psi_i : \tilde{\lambda}_{i,k,\varepsilon} \geq c^+ \cdot \lambda_\varepsilon^{\max}\} \tag{10}$$

as a reasonable candidate for a dominating subspace, where $0 \ll c^+ < 1$ is a fixed constant. Let $\kappa_\varepsilon^{\max} := \beta_1 / (2\varepsilon^2)$ denote the point at which $\lambda_\varepsilon^{\max}$ is attained. Then the eigenfunctions ψ_i of $-\mathcal{A}$ such that $w_k \cdot \psi_i$ is contained in \mathbb{Y}_ε^+ for some k are all elements of

$$Y_\varepsilon^+ := \text{span} \left\{ \psi_i : \frac{\kappa_i}{\kappa_\varepsilon^{\max}} \in [1 - \delta, 1 + \delta] \right\} \quad (11)$$

where $0 < \delta \ll 1$ depends on $c^+ < 1$. In fact, Y_ε^+ is the dominating subspace for the binary Cahn–Hilliard model. See refs. 13 and 14. The elements of Y_ε^+ exhibit a common characteristic wavelength of order $O(\varepsilon)$ as $\varepsilon \rightarrow 0$ (cf. ref. 13, Section 4). Leaving out technical details, this can be described as follows.

Let $x_0 \in \Omega$ be a “typical” point for a given element ψ in Y_ε^+ . Then for any ball with radius r and center x_0 which is completely contained in a nodal domain of ψ , the estimate $r \leq C \cdot \varepsilon$ holds with an ε -independent constant C .

This wavelength estimate canonically remains valid for each component of the elements in \mathbb{Y}_ε^+ , since

$$\mathbb{Y}_\varepsilon^+ \subset w_1 Y_\varepsilon^+ \oplus w_2 Y_\varepsilon^+ \oplus \dots \oplus w_{i^*} Y_\varepsilon^+$$

for some $i^* \in \{1, \dots, N-1\}$. The index i^* can be determined from $c^+ < 1$. See for example Fig. 1, where $i^* = 2$. Notice that in general we have $i^* < N-1$, since some of the branches $\mathcal{C}_{k,\varepsilon} = \{\tilde{\lambda}_{i,k,\varepsilon} : i \in \mathbb{N}\}$ may lie completely below $c^+ \cdot \lambda_\varepsilon^{\max}$, as shown in Fig. 1.

The goal of Section 2 is to state and prove a precise formulation of the following: The local evolution near the equilibrium $u \equiv \bar{u}$ of (6) is dominated by the finite-dimensional space \mathbb{Y}_ε^+ . Loosely speaking, we can prove that most solutions of (6) originating near $u \equiv \bar{u}$ will initially stay close to $\bar{u} + \mathbb{Y}_\varepsilon^+$. This is presented as an application of the abstract theory developed in Section 2 of ref. 14, which originally aimed only at describing the early stages of spinodal decomposition for the binary alloy case. Although our resulting main Theorem 2.9 for the multicomponent case is stated at the end of Section 2 in detail, we briefly formulate the two main consequences concerning the dominance of \mathbb{Y}_ε^+ here. (Notice that in Theorem 2.9 the space \mathbb{Y}_ε^+ has to be split up for technical reasons which will become clear later on, i.e., we have $\mathbb{Y}_\varepsilon^+ = \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$.) These results will make clear that depending on the eigenvalues β_k , there are two fundamentally different possibilities for the structure of the dominating

subspace \mathbb{Y}_ε^+ —one corresponding to the pseudo-binary decomposition described by Eyre,^(7,8) the other one to the absence of this strong coupling between the components of the alloy. We begin with the latter phenomenon.

Theorem 1.2. Let $\beta_1 = \beta_2 = \dots = \beta_{i^*} \gg \beta_{i^*+1} \geq \dots \geq \beta_{N-1}$ denote the eigenvalues of B for the linearization of the multicomponent Cahn–Hilliard system (6) at the unstable homogeneous equilibrium $u \equiv \bar{u} \in \mathcal{G}$. Then on a neighborhood of $u \equiv \bar{u}$ with size proportional to $\varepsilon^{\dim \Omega}$ as $\varepsilon \rightarrow 0$, the subspace \mathbb{Y}_ε^+ dominates the behavior of all solutions of (6) originating near $u \equiv \bar{u}$ in a way which is made precise in Theorem 2.9. The space \mathbb{Y}_ε^+ is contained in $w_1 Y_\varepsilon^+ \oplus \dots \oplus w_{i^*} Y_\varepsilon^+$. Thus, each component of \mathbb{Y}_ε^+ exhibits a characteristic wavelength of the order $O(\varepsilon)$.

Let us mention that the size of the neighborhood, on which the above theorem holds, depends on $c^+ < 1$. However, once c^+ has been fixed, the size is proportional to ε^n , where $n = \dim \Omega$. Unfortunately, in the limit $c^+ \rightarrow 1$ one does not obtain an ε^n dependence of the size. This is the reason why in general one cannot neglect the eigenfunctions corresponding to $\beta_2, \dots, \beta_{i^*}$, even in the case $\beta_1 > \beta_2$. Therefore, as long as $\beta_1 \approx \beta_2 \approx \dots \approx \beta_{i^*} \gg \beta_{i^*+1}$, the above theorem cannot be improved.

If on the other hand $\beta_1 \gg \beta_2$, we do observe a strong mutual coupling of the components. More precisely, we have the following result concerning the structure of the dominating subspace \mathbb{Y}_ε^+ , which is responsible for the occurrence of pseudo-binary decomposition.

Theorem 1.3. Suppose the eigenvalues of B satisfy $\beta_1 \gg \beta_2 \geq \dots \geq \beta_{N-1}$. Then on a neighborhood of the unstable equilibrium $u \equiv \bar{u}$ with size proportional to $\varepsilon^{\dim \Omega}$ the subspace $\mathbb{Y}_\varepsilon^+ \subset w_1 Y_\varepsilon^+$ is dominant. In other words, all components of the elements of \mathbb{Y}_ε^+ are constant multiples of each other. Functions of this form are called pseudo-binary.

According to our central Theorem 2.9, spinodally decomposed states will with high probability approach an element in $\bar{u} + \mathbb{Y}_\varepsilon^+$. Therefore, in the situation of the last theorem the observed states exhibit pseudo-binary behavior with a strong mutual coupling between the components. On the other hand, if $\beta_1 \approx \beta_2$, then Theorem 1.2 suggests that there is no longer such a strong coupling, but all components still exhibit the same wavelength. We want to point out that both of these phenomena match the observations made in experiments and in numerical simulations. See for example refs. 7 and 8. Moreover, we believe that our results may be used to determine the relevant parameters ε , β_1 , and i^* from real data. An algorithm and tests how these parameters can be retrieved from numerical data are currently being worked out.

In Section 3 we discuss the implications of the above Theorems 1.2 and 1.3 for the standard example W which is defined in (3). We will identify the regions in \mathcal{G} where Theorem 1.2 or Theorem 1.3 holds, i.e., where $\beta_1 \gg \beta_2$ or $\beta_1 \approx \beta_2$, respectively.

2. SPINODAL DECOMPOSITION

In the following the abstract results of Section 2 in ref. 14 are applied to the Cahn–Hilliard system. We consider

$$\begin{aligned} u_t &= -\Delta(\varepsilon^2 \Delta u - PD_u W(u)) && \text{in } \Omega \\ \frac{\partial u}{\partial \nu} &= \frac{\partial \Delta u}{\partial \nu} = 0 && \text{on } \partial\Omega \\ u &\in \mathcal{G} \subset \mathbb{R}^N \end{aligned} \quad (12)$$

where $\varepsilon > 0$ is a small parameter. The specific assumptions on the domain Ω and the nonlinearity W are stated below.

(A1) Ω denotes a bounded domain in \mathbb{R}^n , where $n \in \{1, 2, 3\}$. Furthermore, we assume that either the boundary of Ω is of class C^2 , or that Ω is a two- or three-dimensional rectangular domain, or that $\Omega \subset \mathbb{R}^2$ is an equilateral triangle.

(A2) Let $W: \mathbb{R}^N \rightarrow \mathbb{R}$ be a smooth function.

Note that in addition to (A2), for the model to be physically realistic, the nonlinearity W restricted to \mathcal{G} has to satisfy further constraints. For instance, an N -well potential like the standard example (3) would guarantee N energetically preferred states for the material. However, since spinodal decomposition is a local phenomenon near a constant solution $u \equiv \bar{u} \in \mathcal{G}$, we do not need this global property.

Similar to the binary case, spinodal decomposition only occurs in a neighborhood of certain homogeneous equilibria $u \equiv \bar{u}$, i.e., we need an additional assumption. Let $\bar{u} \in \mathcal{G}$ and let $B = B_{\bar{u}} := -PD_u^2 W(\bar{u}) \in \mathcal{L}(\mathbb{R}^N, \mathbb{R}^N)$, with eigenvectors $w_k \in \mathbf{e}^\perp$ and corresponding eigenvalues β_k as introduced in Notation 1.1.

(A3) Choose and fix a total mass vector $\bar{u} \in \mathcal{G}$ in the spinodal region, i.e., assume that $B = -PD_u^2 W(\bar{u})$ has a positive eigenvalue $\beta_1 > 0$.

In order to apply the abstract theory of Section 2 in ref. 14, we have to rewrite (12) as an abstract evolution equation $u_t = Au + F(u)$. Rather than considering the original equation (12) for $u \in \mathcal{G}$ with mass constraint

$\int_{\Omega} u \, dx / |\Omega| = \bar{u}$ (see (4)), we introduce the new variable $w = u - \bar{u} \in \mathbf{e}^{\perp}$, which satisfies the mass constraint $\int_{\Omega} w \, dx = 0$. Transforming (12) into an equation for w , and afterwards replacing w by u again, we arrive at

$$\begin{aligned} u_t &= -\Delta(\varepsilon^2 \Delta u - PD_u W(\bar{u} + u)) \quad \text{in } \Omega \\ \frac{\partial u}{\partial \nu} &= \frac{\partial \Delta u}{\partial \nu} = 0 \quad \text{on } \partial\Omega \\ u &\in \mathbf{e}^{\perp} \subset \mathbb{R}^N \\ \int_{\Omega} u \, dx &= 0 \end{aligned} \tag{13}$$

This equation basically is (12), where the nonlinear term is replaced by $f(\bar{u} + u) = -PD_u W(\bar{u} + u)$. Let $\tilde{f}: \mathbb{R}^N \rightarrow \mathbb{R}^N$ be defined as

$$\tilde{f}(u) := f(\bar{u} + u) - D_u f(\bar{u}) u - f(\bar{u}) = f(\bar{u} + u) - Bu - f(\bar{u}) \tag{14}$$

then in fact $\tilde{f}: \mathbf{e}^{\perp} \rightarrow \mathbf{e}^{\perp}$ and $\tilde{f}(0) = D_u \tilde{f}(0) = 0$. Consider the Hilbert space

$$\mathbb{X} = \left\{ u \in \mathbb{L}^2(\Omega) : \int_{\Omega} u \, dx = 0, u \in \mathbf{e}^{\perp} \text{ a.e. in } \Omega \right\} \tag{15}$$

where $\mathbb{L}^2(\Omega) := (L^2(\Omega))^N$. Similarly, we will use $\mathbb{H}^k(\Omega) := (H^k(\Omega))^N$ in the following. Finally, let

$$A_{\varepsilon} u := -\Delta(\varepsilon^2 \Delta u + Bu) \quad \text{and} \quad F(u) := -\Delta \tilde{f}(u) \tag{16}$$

Then the first equation in (13) is of the form

$$u_t = A_{\varepsilon} u + F(u) \tag{17}$$

The evolution equation (17) is of the same form as the one used in the binary case considered in ref. 14. Thus, we can use the abstract theory developed there to prove the dominance properties mentioned in the introduction. For this we only have to verify hypotheses (H1) through (H3) in ref. 14. This will be done in the following subsections. Basically, we have to verify the following three claims.

(H1) The operator $-A_{\varepsilon}$ is sectorial in the Hilbert space \mathbb{X} .

(H2) There exists a decomposition $\mathbb{X} = \mathbb{X}^{--} \oplus \mathbb{X}^{-} \oplus \mathbb{X}^{+} \oplus \mathbb{X}^{++}$ into pairwise orthogonal subspaces, such that all subspaces are finite-dimensional except \mathbb{X}^{--} , and such that the linear semigroup corresponding to $u_t = A_{\varepsilon} u$ satisfies several dichotomy estimates. See Lemma 2.6(b) below.

(H3) The nonlinear mapping $F: \mathbb{X}^\alpha \rightarrow \mathbb{X}$ is C^1 with $F(0) = 0$ and $DF(0) = 0$. Furthermore, it satisfies a global Lipschitz condition with constant L_F , i.e., for all $u, v \in \mathbb{X}^\alpha$ we have

$$\|F(u) - F(v)\|_{\mathbb{X}} \leq L_F \|u - v\|_{\mathbb{X}^\alpha}$$

Here \mathbb{X}^α denotes the fractional power space corresponding to A_ε .

In the course of verifying hypotheses (H1) through (H3), we also calculate several constants introduced in ref. 14, which in turn furnish an upper bound on the Lipschitz constant L_F . (Notice that since the nonlinearity in our example (see (16)) does not satisfy a global Lipschitz condition, we have to employ a standard cut-off technique.) This will eventually determine the size of the neighborhood on which our results are valid.

2.1. Spectral Properties

In the following lemma we collect several properties of the linear operator A_ε which will be needed later on. These results are the obvious generalization of Lemma 3.1 in ref. 14. We will again use Notation 1.1 for the eigenvalues and eigenfunctions of $-\mathcal{A}$.

Lemma 2.1. Let \mathbb{X} be defined as in (15) and assume that (A1), (A2), and (A3) hold. Then the operator $A_\varepsilon: \mathbb{X} \rightarrow \mathbb{X}$ defined by $A_\varepsilon u = -\mathcal{A}(\varepsilon^2 \mathcal{A}u + Bu)$ with domain

$$D(A_\varepsilon) = \left\{ u \in \mathbb{X} \cap \mathbb{H}^4(\Omega) : \frac{\partial u}{\partial \nu}(x) = \frac{\partial \mathcal{A}u}{\partial \nu}(x) = 0, x \in \partial\Omega \right\}$$

satisfies the following assertions.

The operator $-A_\varepsilon$ is self-adjoint and sectorial. The spectrum of A_ε consists of real eigenvalues $\lambda_{1,k,\varepsilon} \geq \lambda_{2,k,\varepsilon} \geq \dots \rightarrow -\infty$, where $k = 1, \dots, N-1$, with corresponding eigenfunctions $\varphi_{1,k,\varepsilon}, \varphi_{2,k,\varepsilon}, \dots$. The eigenvalues $\lambda_{i,k,\varepsilon}$ are obtained from the numbers $\tilde{\lambda}_{i,k,\varepsilon}$ introduced in (9) by ordering $\tilde{\lambda}_{i,k,\varepsilon}$ while keeping k fixed. The eigenfunctions $\varphi_{i,k,\varepsilon}$ are obtained from the eigenfunctions $\tilde{\varphi}_{i,k,\varepsilon} := \psi_i \cdot w_k$ through this ordering process in the obvious way, and they form an $\mathbb{L}^2(\Omega)$ -orthonormal basis of \mathbb{X} . Moreover, the largest eigenvalue $\lambda_{1,1,\varepsilon}$ is of the order

$$\lambda_{1,1,\varepsilon} \sim \lambda_\varepsilon^{\max} := \frac{\beta_1^2}{4\varepsilon^2}, \quad \text{and} \quad \lambda_{1,1,\varepsilon} \leq \lambda_\varepsilon^{\max} \quad (18)$$

Furthermore, let $\tilde{\kappa}_{i,k,\varepsilon}$ denote the reordering of the eigenvalues of $-A$ such that $\lambda_{i,k,\varepsilon} = \tilde{\kappa}_{i,k,\varepsilon}(\beta_k - \varepsilon^2 \tilde{\kappa}_{i,k,\varepsilon})$. Note that for large $i \in \mathbb{N}$ (depending on ε) and all $k = 1, \dots, N - 1$ both $\tilde{\lambda}_{i,k,\varepsilon} = \lambda_{i,k,\varepsilon}$ and $\tilde{\kappa}_{i,k,\varepsilon} = \kappa_i$ are satisfied.

The above lemma implies that A_ε generates an analytic semigroup $S_\varepsilon(t)$ on \mathbb{X} . Furthermore, for every A_ε the fractional power space $\mathbb{X}^{1/2,\varepsilon} = D((-A_\varepsilon + a_\varepsilon I)^{1/2})$ is defined. See the discussion in Subsection 2.1 following (H1) in ref. 14. Although formally this fractional power space depends on ε , the next lemma shows that *algebraically*, i.e., as a vector space, we have $\mathbb{X}^{1/2,\varepsilon} = \mathbb{H}_{av}^2(\Omega)$, where

$$\mathbb{H}_{av}^2(\Omega) := \left\{ u \in \mathbb{H}^2(\Omega) : \int_{\Omega} u \, dx = 0, u \in \mathbf{e}^\perp \text{ a.e. in } \Omega, \text{ and } \frac{\partial u}{\partial \nu} = 0 \text{ on } \partial\Omega \right\}$$

Likewise, the *topological* structure of the fractional power space $X^{1/2,\varepsilon}$ will turn out to be independent of ε —even though the norms $\|\cdot\|_{1/2,\varepsilon}$ are not. This is due to the fact that they are equivalent to the standard $\mathbb{H}^2(\Omega)$ -norm on $\mathbb{H}_{av}^2(\Omega)$, i.e., to the norm

$$\| \| u \| \|_{\mathbb{H}^2(\Omega)} := \sqrt{\sum_{i=1}^N \| u_i \|_{H^2(\Omega)}^2}$$

Moreover, let

$$\| \| u \| \| = \sqrt{\langle (u, u) \rangle} = \| \| u \| \|_{L^2(\Omega)} := \sqrt{\sum_{i=1}^N \| u_i \|_{L^2(\Omega)}^2}$$

With these definitions, we have the following result.

Lemma 2.2. Let both (A1) and (A3) be satisfied, and let $\| \| \cdot \| \|_{1/2,\varepsilon}$ denote the norm on $\mathbb{X}^{1/2,\varepsilon}$ defined by

$$\| \| u \| \|_{1/2,\varepsilon} := \| \| (-A_\varepsilon + a_\varepsilon I)^{1/2} u \| \| \quad \text{for all } u \in \mathbb{X}^{1/2,\varepsilon}$$

where $a_\varepsilon = \beta_1^2/\varepsilon^2$. Moreover, define a norm $\| \| \cdot \| \|_*$ on $\mathbb{H}^2(\Omega)$ by

$$\| \| u \| \|_* = \sqrt{\langle (u, u) \rangle_*} := \sqrt{\| \| u \| \|^2 + \| \| Au \| \|^2} \quad \text{for all } u \in \mathbb{H}^2(\Omega)$$

Then for every $0 < \varepsilon^2 \leq \min\{\beta_1, 2\beta_1^2, 2\beta_1^2/\beta_{N-1}^2\}$ we have

$$\mathbb{X}^{1/2,\varepsilon} = \mathbb{H}_{av}^2(\Omega) \tag{19}$$

and the norm $\|\cdot\|_{1/2, \varepsilon}$ is equivalent to both $\|\cdot\|_{\mathbb{H}^2(\Omega)}$ and $\|\cdot\|_*$. More precisely, there exists a (ε -independent) constant $C > 0$ depending only on the domain Ω such that for all $u \in \mathbb{H}_{av}^2(\Omega)$ the estimates

$$\frac{\varepsilon}{\sqrt{2}} \cdot \|u\|_* \leq \|u\|_{1/2, \varepsilon} \leq \frac{\sqrt{2} \cdot \beta_1}{\varepsilon} \cdot \|u\|_* \tag{20}$$

and

$$\frac{\varepsilon}{C \cdot \sqrt{2}} \cdot \|u\|_{\mathbb{H}^2(\Omega)} \leq \|u\|_{1/2, \varepsilon} \leq \frac{C \cdot \sqrt{2} \cdot \beta_1}{\varepsilon} \cdot \|u\|_{\mathbb{H}^2(\Omega)}$$

are satisfied.

Proof. Similar to our reasoning in the proof of Lemma 3.2 in ref. 14 we find that $\|\cdot\|_*$ and $\|\cdot\|_{\mathbb{H}^2(\Omega)}$ are equivalent, thus we only have to verify (20). Also, functions $u \in \mathbb{X}$ with $\mathbb{L}^2(\Omega)$ -Fourier coefficients $\zeta_{i, k, \varepsilon} = (u, \varphi_{i, k, \varepsilon})$, $i \in \mathbb{N}$, $1 \leq k \leq N - 1$, are contained in $\mathbb{X}^{1/2, \varepsilon}$ if and only if

$$\|u\|_{1/2, \varepsilon}^2 = \sum_{k=1}^{N-1} \sum_{i=1}^{\infty} (a_\varepsilon - \lambda_{i, k, \varepsilon}) \zeta_{i, k, \varepsilon}^2 < \infty \tag{21}$$

On the other hand, a function $u \in \mathbb{X}$ is an element of $\mathbb{H}_{av}^2(\Omega)$ if and only if

$$\|u\|_*^2 = \sum_{k=1}^{N-1} \sum_{i=1}^{\infty} (1 + \tilde{\kappa}_{i, k, \varepsilon}^2) \zeta_{i, k, \varepsilon}^2 < \infty \tag{22}$$

Employing the crucial estimate of the proof of Lemma 3.2 in ref. 14 we obtain the estimate

$$\frac{\varepsilon^2}{2} \cdot (1 + s^2) \leq \varepsilon^2 s^2 - \beta_1 s + a_\varepsilon \leq \frac{\beta_1^2}{\varepsilon^2} \cdot (1 + s^2) \quad \text{for all } s \geq 0 \text{ and } 0 < \varepsilon^2 \leq \beta_1$$

The left-hand side and (9) then imply $\varepsilon^2(1 + \kappa_i^2)/2 \leq a_\varepsilon - \tilde{\lambda}_{i, 1, \varepsilon} \leq a_\varepsilon - \tilde{\lambda}_{i, k, \varepsilon}$, which proves the first inequality in (20) via the obvious reordering of (21) and (22). In order to verify the second inequality in (20) it suffices to prove that

$$\varepsilon^2 \kappa_i^2 - \beta_k \kappa_i \leq \frac{2 \cdot \beta_1^2 \cdot \kappa_i^2 + \beta_1^2}{\varepsilon^2} \quad \text{for } i \in \mathbb{N} \text{ and } 1 \leq k \leq N - 1$$

For $\beta_k \geq 0$ this is a consequence of $0 < \varepsilon^2 \leq \beta_1$, whereas for $\beta_k < 0$ we apply the inequalities $-\beta_k \kappa_i \leq |\beta_{N-1}| \cdot \kappa_i \leq \beta_1^2 \kappa_i^2 / \varepsilon^2 + \beta_1^2 / \varepsilon^2$ using the remaining assumptions on ε . Together with the above characterizations, the identity (19) is now obvious. ■

Since according to the above lemma the fractional power space $\mathbb{X}^{1/2, \varepsilon}$ is independent of ε algebraically, we omit the superscript ε in the following and simply write $\mathbb{X}^{1/2} = \mathbb{H}_{av}^2(\Omega)$. Furthermore, we identify $(\mathbb{X}^{1/2}, \|\cdot\|_{1/2, \varepsilon})$ with the space $(\mathbb{H}_{av}^2(\Omega), \|\cdot\|_*)$, because Lemma 2.2 also shows that the topology of $\mathbb{X}^{1/2, \varepsilon}$ is independent of ε .

2.2. Spectral Gaps and Exponential Dichotomy Estimates

The following lemma proves the existence of suitable spectral gaps in the spectrum of A_ε . They will be used to define the decomposition of \mathbb{X} mentioned in (H2), and therefore eventually furnish the dichotomy estimates (see Lemma 2.6(b)). The size of these gaps turns out to be crucial, because it provides a restriction on the possible size of the global Lipschitz constant of the nonlinearity in (H3).

Lemma 2.3. Assume that assumptions (A1) and (A3) are satisfied and fix two constants $c_* < c^* < 1$.

Then there exist positive constants ε_0 and d_0 which depend only on c_* , c^* , Ω , N , and β_1 such that for arbitrary $0 < \varepsilon \leq \varepsilon_0$ the following holds. The linear operator A_ε has eigenvalues $\lambda_*(\varepsilon)$ and $\lambda^*(\varepsilon)$ satisfying both $\lambda^*(\varepsilon) - \lambda_*(\varepsilon) \geq d_0 \cdot \varepsilon^{n-2}$ and

$$c_* \cdot \lambda_\varepsilon^{\max} \leq \lambda_*(\varepsilon) < \lambda^*(\varepsilon) \leq c^* \cdot \lambda_\varepsilon^{\max}$$

Moreover, the whole interval $(\lambda_*(\varepsilon), \lambda^*(\varepsilon))$ is part of the resolvent set of A_ε , and both the interval $(c_* \cdot \lambda_\varepsilon^{\max}, \lambda_*(\varepsilon))$ and $(\lambda^*(\varepsilon), c^* \cdot \lambda_\varepsilon^{\max})$ contains eigenvalues of the form $\lambda_{i_k, k, \varepsilon}$ for all k satisfying

$$\lambda_{\varepsilon, k}^{\max} := \max\{s(\beta_k - \varepsilon^2 s) : s \geq 0\} \geq c^* \cdot \lambda_\varepsilon^{\max}$$

In particular, they always contain an eigenvalue of the form $\lambda_{i_1, 1, \varepsilon}$.

Proof. Fix two constants $c_* < c_{**} < c^{**} < c^*$. Due to (8) we can choose $\varepsilon_0 > 0$ small enough such that for all $0 < \varepsilon \leq \varepsilon_0$ the following two assertions hold.

- Both in the interval $[c^{**}, c^*] \cdot \lambda_\varepsilon^{\max}$ and in $[c_*, c_{**}] \cdot \lambda_\varepsilon^{\max}$ there is at least one eigenvalue $\lambda_{i_k, k, \varepsilon}$ of A_ε for every k with $\lambda_{\varepsilon, k}^{\max} \geq c^* \cdot \lambda_\varepsilon^{\max}$. Let

$\lambda^{**}(\varepsilon)$ denote the smallest of these eigenvalues in the first interval, and $\lambda_{**}(\varepsilon)$ the largest one in the second interval.

• The number of eigenvalues of A_ε in the interval $(c_{**}, c^{**}) \cdot \lambda_\varepsilon^{\max}$ is bounded above by $N \cdot C \cdot \varepsilon^{-n} - 1$, where C depends only on c_* , c^* , Ω , and β_1 .

Using these facts the proof can be completed similar to the proof of Lemma 3.3 in ref. 14. ■

The above lemma makes it possible to control gaps in the spectrum of A_ε as $\varepsilon \rightarrow 0$. This is used to establish assumption (H2). To this end, choose constants

$$\underline{c}^{--} < \bar{c}^{--} \ll 0 \ll \underline{c}^- < \bar{c}^- < \underline{c}^+ < \bar{c}^+ < 1 \tag{23}$$

where typically the differences $\bar{c}^{--} - \underline{c}^{--}$, $\bar{c}^- - \underline{c}^-$, and $\bar{c}^+ - \underline{c}^+$ are small. Using these constants the following results follow immediately from Lemma 2.3.

Corollary 2.4. Under the assumptions of Lemma 2.3 there exist intervals

$$\begin{aligned} \hat{J}_\varepsilon^{--} &:= [\hat{a}_\varepsilon^{--}, b_\varepsilon^{--}] \subset [\underline{c}^{--}, \bar{c}^{--}] \cdot \lambda_\varepsilon^{\max} \\ J_\varepsilon^- &:= [a_\varepsilon^-, b_\varepsilon^-] \subset [\underline{c}^-, \bar{c}^-] \cdot \lambda_\varepsilon^{\max} \\ J_\varepsilon^+ &:= [a_\varepsilon^+, b_\varepsilon^+] \subset [\underline{c}^+, \bar{c}^+] \cdot \lambda_\varepsilon^{\max} \end{aligned}$$

such that for sufficiently small $\varepsilon > 0$ the following holds.

(a) Each of the intervals \hat{J}_ε^{--} , J_ε^- , J_ε^+ is contained in the resolvent set of A_ε .

(b) With $a_\varepsilon^{--} := (\hat{a}_\varepsilon^{--} + b_\varepsilon^{--})/2$ and $J_\varepsilon^{--} := [a_\varepsilon^{--}, b_\varepsilon^{--}] \subset \hat{J}_\varepsilon^{--} \subset [\underline{c}^{--}, \bar{c}^{--}] \cdot \lambda_\varepsilon^{\max}$, there exists an ε -independent constant $d > 0$ such that the length of each of the intervals J_ε^{--} , J_ε^- , and J_ε^+ is at least $d \cdot \varepsilon^{n-2}$. The constant d depends only on Ω , β_1 , N , and the constants in (23).

(c) The interval $[\underline{c}^{--} \cdot \lambda_\varepsilon^{\max}, a_\varepsilon^{--})$ is not contained in the resolvent set of A_ε . More precisely, this interval contains at least one eigenvalue of A_ε of the form $\lambda_{i_k, k, \varepsilon}$ for every $1 \leq k \leq N - 1$.

Using this result, we can finally define the decomposition of \mathbb{X} needed for applying the abstract theory of Section 2 in ref. 14.

Definition 2.5. Define the intervals $I_\varepsilon^{--} := (-\infty, a_\varepsilon^{--})$, $I_\varepsilon^- := (b_\varepsilon^{--}, a_\varepsilon^-)$, $I_\varepsilon^+ := (b_\varepsilon^-, a_\varepsilon^+)$, and $I_\varepsilon^{++} := (b_\varepsilon^+, \lambda_\varepsilon^{\max}]$, and let $\mathbb{X}_\varepsilon^{--}$, \mathbb{X}_ε^- , \mathbb{X}_ε^+ , and $\mathbb{X}_\varepsilon^{++}$ denote the span of all eigenfunctions of the operator A_ε (see Lemma 2.1) corresponding to eigenvalues in I_ε^{--} , I_ε^- , I_ε^+ , and I_ε^{++} , respectively.

The restrictions of A_ε or of the induced analytic semigroup $S_\varepsilon(t)$, $t \geq 0$, to each of the subspaces defined above will be indicated by the appropriate superscript. With these definitions we can finally verify hypothesis (H2) for the linearization of the Cahn–Hilliard system. As mentioned earlier, this is done for the non-standard norm $\|\cdot\|_*$ on $\mathbb{X}^{1/2} = \mathbb{H}_{av}^2(\Omega)$.

Lemma 2.6. Assume that (A1) and (A3) hold. Let $A_\varepsilon: \mathbb{X} \rightarrow \mathbb{X}$ be the operator defined in Lemma 2.1, let $S_\varepsilon(t): \mathbb{X} \rightarrow \mathbb{X}$, $t \geq 0$, denote the analytic semigroup generated by A_ε , and let $\mathbb{X}^{1/2} = \mathbb{H}_{av}^2(\Omega)$ be the fractional power space introduced in Subsection 2.1 with norm $\|\cdot\|_*$. Moreover, consider the constants and intervals defined in Corollary 2.4 and Definition 2.5. Then the following assertions are satisfied for every $0 < \varepsilon \leq \varepsilon_0$, where ε_0 depends only on the domain Ω , β_1 , $|\beta_{N-1}|$, N , and the constants in (23).

(a) The spaces \mathbb{X}_ε^- , \mathbb{X}_ε^+ , and $\mathbb{X}_\varepsilon^{++}$ are finite-dimensional subspaces of $\mathbb{X}^{1/2}$ with dimensions proportional to ε^{-n} , where n denotes the dimension of the domain Ω . Additionally, $\mathbb{X}_\varepsilon^{--}$, \mathbb{X}_ε^- , \mathbb{X}_ε^+ , and $\mathbb{X}_\varepsilon^{++}$ are pairwise orthogonal with respect to the $\mathbb{L}^2(\Omega)$ -scalar product $((\cdot, \cdot))$, and their restrictions to $\mathbb{X}^{1/2}$ are pairwise orthogonal with respect to $((\cdot, \cdot))_*$.

(b) There exists a positive constant M_ε^{--} so that for every $u^{++} \in \mathbb{X}_\varepsilon^{++}$, $u^+ \in \mathbb{X}_\varepsilon^+$, $u^- \in \mathbb{X}_\varepsilon^-$, $u_*^{--} \in \mathbb{X}_\varepsilon^{--} \cap \mathbb{X}^{1/2}$, and $u^{--} \in \mathbb{X}_\varepsilon^{--}$ the estimates

$$\begin{aligned} \|S_\varepsilon^{++}(t) u^{++}\|_* &\leq e^{b_\varepsilon^+ t} \cdot \|u^{++}\|_* && \text{for } t \leq 0, \\ \|S_\varepsilon^+(t) u^+\|_* &\leq e^{a_\varepsilon^+ t} \cdot \|u^+\|_* && \text{for } t \geq 0, \\ \|S_\varepsilon^+(t) u^+\|_* &\leq e^{b_\varepsilon^- t} \cdot \|u^+\|_* && \text{for } t \leq 0, \\ \|S_\varepsilon^-(t) u^-\|_* &\leq e^{a_\varepsilon^- t} \cdot \|u^-\|_* && \text{for } t \geq 0, \\ \|S_\varepsilon^-(t) u^-\|_* &\leq e^{b_\varepsilon^{--} t} \cdot \|u^-\|_* && \text{for } t \leq 0, \\ \|S_\varepsilon^{--}(t) u_*^{--}\|_* &\leq e^{a_\varepsilon^{--} t} \cdot \|u_*^{--}\|_* && \text{for } t \geq 0, \\ \|S_\varepsilon^{--}(t) u^{--}\|_* &\leq M_\varepsilon^{--} \cdot t^{-1/2} \cdot e^{a_\varepsilon^{--} t} \cdot \|u^{--}\| && \text{for } t > 0 \end{aligned}$$

are satisfied. Moreover, $M_\varepsilon^{--} \leq C_1 \cdot \varepsilon^{-(1+n/2)}$ as $\varepsilon \rightarrow 0$, where $C_1 > 0$ depends only on Ω , β_1 , N , and the constants in (23).

(c) There exists a constant $M_{1/2, \varepsilon} \geq 1$ such that for all $u \in \mathbb{X}_\varepsilon^- \oplus \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$ we have

$$\| \|u\| \leq \| \|u\|_* \leq M_{1/2, \varepsilon} \cdot \| \|u\|$$

as well as $M_{1/2, \varepsilon} \cdot \varepsilon^2 \rightarrow C_2$ as $\varepsilon \rightarrow 0$, where $C_2 > 0$ depends only on β_1 and the constants in (23).

Proof. The assertions of part (a) follow immediately from (8), Corollary 2.4, and Definition 2.5.

As for the proof of (b), let $u \in \mathbb{X}$ be arbitrary, let $\varphi_{i, k, \varepsilon}$ denote the eigenfunctions of A_ε according to Lemma 2.1, and let $u = \sum_{k=1}^{N-1} \sum_{i=1}^\infty \zeta_{i, k, \varepsilon} \varphi_{i, k, \varepsilon}$ denote the Fourier series representation of u in \mathbb{X} , i.e., let $\zeta_{i, k, \varepsilon} := ((u, \varphi_{i, k, \varepsilon}))$, where $((\cdot, \cdot))$ denotes the standard $\mathbb{L}^2(\Omega)$ -scalar product. Then we have an explicit spectral representation of the semigroup $S_\varepsilon(t)$ given by

$$S_\varepsilon(t) u = \sum_{k=1}^{N-1} \sum_{i=1}^\infty e^{\lambda_{i, k, \varepsilon} \cdot t} \cdot \zeta_{i, k, \varepsilon} \cdot \varphi_{i, k, \varepsilon} \quad \text{for } t > 0$$

and if $u \in \mathbb{X}^{1/2}$, then (22) furnishes

$$\| \|u\|_*^2 = \sum_{k=1}^{N-1} \sum_{i=1}^\infty (1 + \tilde{\kappa}_{i, k, \varepsilon}^2) \cdot \zeta_{i, k, \varepsilon}^2 < \infty$$

These two identities already imply the first six inequalities in part (b), similar to the proof of Lemma 3.6 in ref. 14.

In order to prove the seventh inequality, let $u^{--} \in \mathbb{X}_\varepsilon^{--}$ be arbitrary. If $n_1(k) \geq 1$ is chosen in such a way that $\lambda_{i, k, \varepsilon}$ for $i \geq n_1(k)$ and $1 \leq k \leq N-1$ denote all the eigenvalues of A_ε which are contained in I_ε^{--} , then u^{--} has the Fourier series representation $u^{--} = \sum_{k=1}^{N-1} \sum_{i=n_1(k)}^\infty \zeta_{i, k, \varepsilon} \varphi_{i, k, \varepsilon}$ in \mathbb{X} , and for arbitrary $t > 0$ we actually have $S_\varepsilon^{--}(t) u^{--} \in \mathbb{X}^{1/2}$. Due to the choice of the interval I_ε^{--} one further obtains $\lambda_{i, k, \varepsilon} < 0$ for all $i \geq n_1(k)$, and therefore $\tilde{\kappa}_{i, k, \varepsilon} = \kappa_i > \beta_k / \varepsilon^2$ for all $i \geq n_1(k)$. Thus,

$$\| S_\varepsilon^{--}(t) u^{--} \|_*^2 = \sum_{k=1}^{N-1} \sum_{i=n_1(k)}^\infty (1 + \kappa_i^2) \cdot e^{2\lambda_{i, k, \varepsilon} \cdot t} \cdot \zeta_{i, k, \varepsilon}^2$$

Now it is easy to verify that for all $t > 0$, $i \geq n_1(k)$, and $\lambda > \lambda_{n_1(k), k, \varepsilon}$ we have

$$(1 + \kappa_i^2) \cdot e^{2\lambda_{i, k, \varepsilon} \cdot t} \leq \frac{1 + \kappa_i^2}{2e(\lambda - \lambda_{i, k, \varepsilon})} \cdot t^{-1} \cdot e^{2\lambda \cdot t} = \frac{1 + \kappa_i^2}{2e(\lambda - \beta_k \kappa_i + \varepsilon^2 \kappa_i^2)} \cdot t^{-1} \cdot e^{2\lambda \cdot t}$$

see Lemma 2.1. To continue, define the function $h(s) = (1 + s^2) \cdot (\lambda - \beta s + \varepsilon^2 s^2)^{-1}$, where $\lambda = a_\varepsilon^{--} < 0$, $s \in A := \{s > 0 : \lambda - \beta s + \varepsilon^2 s^2 > 0\}$,

and $\beta = \beta_k$ for an arbitrary $1 \leq k \leq N - 1$. Assume first that k is such that $\beta_k \geq 0$. Then h is monotonically decreasing on A , and this implies

$$\begin{aligned} C_{i,k,\varepsilon}^2 &:= \frac{1 + \kappa_i^2}{2e(\lambda - \beta_k \kappa_i + \varepsilon^2 \kappa_i^2)} \leq \frac{1 + \kappa_{n_1(k)}^2}{2e(\lambda - \lambda_{n_1(k),k,\varepsilon})} \\ &\leq \frac{1 + \kappa_{n_1(1)}^2}{2e(a_\varepsilon^{--} - \hat{a}_\varepsilon^{--})} =: (\tilde{M}_\varepsilon^{--})^2 \end{aligned}$$

If $\beta_k < 0$, then h switches exactly once from decreasing to increasing in A , and this yields

$$C_{i,k,\varepsilon} \leq \max\{\tilde{M}_\varepsilon^{--}, \varepsilon^{-1}\} =: M_\varepsilon^{--}$$

Finally, using a calculation which was already employed in the proof of Lemma 3.6(b) in ref. 14 and which uses the estimate $\lambda_{n_1(1),1,\varepsilon} \geq \underline{c}^{--} \cdot \beta_1^2 / (4\varepsilon^2)$ from Corollary 2.4(c), we obtain

$$M_\varepsilon^{--} \leq \varepsilon^{-(1+n/2)} \cdot \max \left\{ \varepsilon^{n/2}, \sqrt{\frac{\varepsilon^4 + \beta_1^2 \cdot (1 + \sqrt{1 - \underline{c}^{--}})^2 / 4}{d}} \right\}$$

where $d > 0$ is defined in Corollary 2.4(b). This finally proves the asymptotic behavior of the constant M_ε^{--} for $\varepsilon \rightarrow 0$.

The proof of part (c) follows the lines of the proof of Lemma 3.6(c) in ref. 14 and is therefore omitted. ■

According to the above lemma the linear part of the Cahn–Hilliard equation (13) satisfies (with respect to the non-standard norm $\|\cdot\|_*$) hypothesis (H2), and trivially (H1), as well as (7) from Section 2 in ref. 14. Moreover, the asymptotic behavior for $\varepsilon \rightarrow 0$ of certain spectral gaps in the spectrum of A_ε , and of the constants M_ε^{--} and $M_{1/2,\varepsilon}$ have been obtained. We close this subsection with the following remark.

Remark 2.7. With the results of Corollary 2.4 and Lemma 2.6 we can deduce the asymptotic behavior of certain constants introduced in ref. 14 Section 2 for $\varepsilon \rightarrow 0$. Although the specific values of these constants are different if compared to the application in ref. 14, Section 3, their dependence on ε is not. Hence, we obtain exactly the same asymptotics, i.e., $C_\varepsilon^{--} \geq C \cdot \varepsilon^n$ and $C_\varepsilon^+ \geq C \cdot \varepsilon^{n-2}$ for $\varepsilon \rightarrow 0$. See Remark 3.7 in ref. 14.

Even though we did not formally introduce these constants, we want to point out that their asymptotic behavior is used to prove that the abstract theory of Section 2 in ref. 14 can be applied to A_ε and a nonlinear

function F , whose Lipschitz constant satisfies $0 \leq L_F \leq C \cdot \varepsilon^n$. For more details see ref. 14, Remark 2.11. All of the constants C above depend only on Ω , β_1 , N , and the constants in (23). ■

2.3. Properties of the Nonlinearity

While in the last subsection we established the validity of hypotheses (H1) and (H2) for the Cahn–Hilliard system, this short subsection is devoted to the verification of hypothesis (H3). We will provide a global Lipschitz constant L_F for an appropriate modification of the nonlinearity F defined in (16).

Assume that assumption (A1) is satisfied and let $g: \mathbb{R}^N \rightarrow \mathbb{R}^N$ denote a smooth function with $g(0) = 0$ and $Dg(0) = 0$. Consider the nonlinear operator G defined by $G(u)(x) := (-\Delta g(u))(x)$. Then G is continuously differentiable from $\mathbb{H}_{av}^2(\Omega)$ to the space $\mathbb{L}^2(\Omega)$ with derivative

$$DG(u)h = -\Delta(Dg(u)h) = \left(-\Delta \sum_{i=1}^N Dg_{ij}(u) \cdot h_i \right)_{1 \leq j \leq N} \quad (24)$$

for $u, h \in \mathbb{H}_{av}^2(\Omega)$. Moreover, $G(0) = 0$ and $DG(0) = 0$. Applying this observation to the function $g = \tilde{f}$ defined in (14) we see that F as defined in (16) is continuously differentiable. However, it does not satisfy the strong global Lipschitz condition of hypothesis (H3).

Fortunately, hypothesis (H3) is valid for some function $\hat{F}: \mathbb{H}_{av}^2(\Omega) \rightarrow \mathbb{L}^2(\Omega)$ which coincides with F on a certain neighborhood of the origin. In order to obtain a global Lipschitz constant $L_{\hat{F}}$ of the order ε^n (as required by Remark 2.7), the size of this neighborhood has to be proportional to ε^n with respect to the $\mathbb{H}^2(\Omega)$ -norm. This can be proved by applying the estimates for $\|DG(u)h\|$ and $N = 1$ from ref. 14, Section 3.3 to the identity (24). This immediately furnishes the following result.

Corollary 2.8. The nonlinear operator F defined in (16) satisfies (H3) with a Lipschitz constant L_F of the order ε^n on an $\mathbb{H}^2(\Omega)$ -neighborhood of 0 with size proportional to ε^n .

2.4. Spinodal Decomposition

In the previous subsections we established all properties of (12) which are necessary to apply the abstract results of ref. 14, Section 2 to the Cahn–Hilliard system—and this can be done exactly as in Subsection 3.4 of

ref. 14. Moreover, since the asymptotic behavior of the involved constants remains basically unchanged, we obtain exactly the same result, of course after adopting the new notation (\bar{u} instead of μ , \mathbb{X}_ε instead of X_ε , $\mathbb{H}_{av}(\Omega)$ instead of $H_{av}(\Omega)$, and so on). Therefore, we refrain from presenting our main theorem again in as detailed a form as in the binary case, and state only an intuitive abbreviated version.

Suppose that three constants $0 < r \ll \varrho \ll R$ are given. We consider initial conditions from the ball

$$B_r(\bar{u}) = \{v \in \bar{u} + \mathbb{X}_\varepsilon^- \oplus \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++} : \|v - \bar{u}\|_* < r\} \subset \bar{u} + \mathbb{H}_{av}^2(\Omega)$$

and their evolution under the dynamics of (12). For initial conditions $v \in B_r(\bar{u})$ we consider *corresponding solutions* of (12) living in some inertial manifold (see Section 3.4 in ref. 14) which is tangential to $\mathbb{X}_\varepsilon^- \oplus \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$ at \bar{u} , and whose projected initial value is v . Let M_r denote the set of all those initial conditions $v \in B_r(\bar{u})$ whose corresponding solution of (12), projected to $\bar{u} + \mathbb{X}_\varepsilon^- \oplus \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$, either remains in the larger ball $B_R(\bar{u})$ for all time, or has distance greater than ϱ from $\bar{u} + \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$ upon exiting $B_r(\bar{u})$. See also Fig. 2. In other words, the initial conditions in M_r cannot be considered as being dominated by the strongly unstable subspace $\mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$.

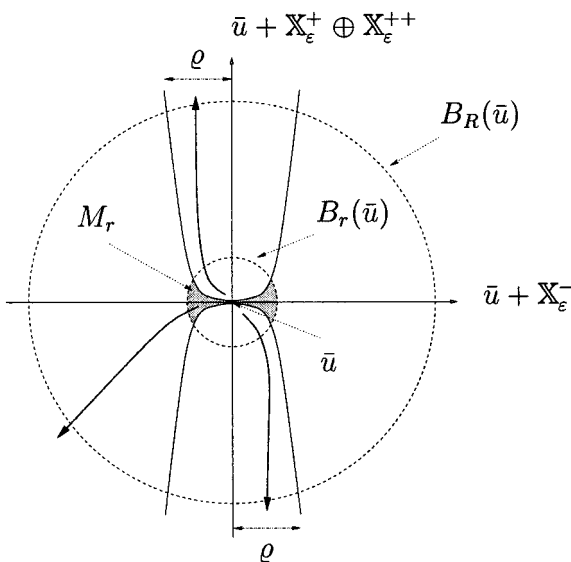


Fig. 2. Situation in the main theorem.

Our main theorem states that the volume (which is the canonical Lebesgue volume of the finite-dimensional space $\mathbb{X}_\varepsilon^- \oplus \mathbb{X}_\varepsilon^+ \oplus \mathbb{X}_\varepsilon^{++}$) of these “bad” initial conditions compared to the volume of all initial conditions in $B_r(\bar{u})$ is arbitrarily small, provided the constants $0 < r \ll \varrho \ll R$ are chosen proportional to ε^n as $\varepsilon \rightarrow 0$.

Theorem 2.9. We consider solutions of the Cahn–Hilliard system (12) and assume that hypotheses (A1), (A2), and (A3) are satisfied. Then there exists a positive constant ε_0 which depends only on Ω , N , β_1 , $|\beta_{N-1}|$, and the constants in (23), such that for arbitrary $0 < \varepsilon \leq \varepsilon_0$ the following holds.

For every $0 < p \ll 1$ there exist constants $0 < r \ll \varrho \ll R$ which depend only on N , β_1 , Ω , and the constants in (23) (r depends additionally on p) and which are all proportional to ε^n as $\varepsilon \rightarrow 0$, such that

$$\frac{\text{vol}(M_r)}{\text{vol}(B_r(\bar{u}))} \leq p \quad (25)$$

Proof. One only has to apply the abstract theory of Section 2 in ref. 14. Hypotheses (H1) through (H3) have been established in Subsections 2.1 through 2.3, and the necessary constants have been calculated, furnishing the ball with size proportional to ε^n on which the result is valid. ■

Remark 2.10. An inspection of the abstract theory in ref. 14, Section 2 shows that the following more general result is true. In Theorem 2.9, choose an ε -dependent constant $\varrho > 0$ which is proportional to $\varepsilon^{k_\varepsilon}$ as $\varepsilon \rightarrow 0$, for some $k_\varepsilon \geq n$, i.e., the constant ϱ depends polynomially on ε . Then for every p as in the theorem, there exists a radius R which is proportional to ε^n , and a radius r which depends polynomially on ε , such that the estimate (25) of Theorem 2.9 remains valid. In other words, if a solution of (12) starts closer to the homogeneous equilibrium \bar{u} , then it will exit closer to the dominating subspace. This result will be needed in order to apply the results of Sander and Wanner⁽¹⁸⁾ to Cahn–Hilliard systems. ■

3. A CASE STUDY FOR TERNARY ALLOYS

3.1. Preliminary Considerations

In this section we describe the implications of our main Theorem 2.9 to ternary alloys. In particular, we consider the Cahn–Hilliard system (6)

with the nonlinearity f defined by (5) for the specific free energy W given in (3). It can readily be verified that in this situation we have

$$f(u_1, u_2, u_3) = \frac{1}{3} \cdot \left(\begin{aligned} &\sigma \cdot (2u_1 - u_2 - u_3) + \ln \frac{u_2 u_3}{u_1^2}, \\ &\sigma \cdot (2u_2 - u_3 - u_1) + \ln \frac{u_3 u_1}{u_2^2}, \\ &\sigma \cdot (2u_3 - u_1 - u_2) + \ln \frac{u_1 u_2}{u_3^2} \end{aligned} \right)$$

If $\bar{u} = (\bar{u}_1, \bar{u}_2, \bar{u}_3)$ denotes any point in the interior of the Gibbs triangle \mathcal{G} , then the linearization of (6) at \bar{u} is given by (7), with

$$B = \frac{1}{3} \cdot \begin{pmatrix} 2\sigma - 2/\bar{u}_1 & -\sigma + 1/\bar{u}_2 & -\sigma + 1/\bar{u}_3 \\ -\sigma + 1/\bar{u}_1 & 2\sigma - 2/\bar{u}_2 & -\sigma + 1/\bar{u}_3 \\ -\sigma + 1/\bar{u}_1 & -\sigma + 1/\bar{u}_2 & 2\sigma - 2/\bar{u}_3 \end{pmatrix} \quad (26)$$

Furthermore, the characteristic polynomial $p(\lambda) = \det(B - \lambda I)$ of the matrix B is given by $p(\lambda) = -\lambda \cdot (\lambda^2 + q_1 \lambda + q_0)$, where

$$q_0 = \sigma^2 + \frac{1 - 2\sigma(\bar{u}_1 \bar{u}_2 + \bar{u}_2 \bar{u}_3 + \bar{u}_3 \bar{u}_1)}{3\bar{u}_1 \bar{u}_2 \bar{u}_3}$$

$$q_1 = -2\sigma + \frac{2(\bar{u}_1 \bar{u}_2 + \bar{u}_2 \bar{u}_3 + \bar{u}_3 \bar{u}_1)}{3\bar{u}_1 \bar{u}_2 \bar{u}_3}$$

Thus, if we let

$$\beta^* = \left(\frac{1}{\bar{u}_1} - \frac{1}{\bar{u}_2} \right)^2 + \left(\frac{1}{\bar{u}_2} - \frac{1}{\bar{u}_3} \right)^2 + \left(\frac{1}{\bar{u}_3} - \frac{1}{\bar{u}_1} \right)^2 \quad (27)$$

then the eigenvalues $\beta_1 \geq \beta_2$ of $B|_{e^\perp}$ are given by

$$\beta_1 = \sigma - \frac{1}{3} \cdot \left(\frac{1}{\bar{u}_1} + \frac{1}{\bar{u}_2} + \frac{1}{\bar{u}_3} \right) + \frac{\sqrt{\beta^*}}{3\sqrt{2}} \quad (28)$$

$$\beta_2 = \sigma - \frac{1}{3} \cdot \left(\frac{1}{\bar{u}_1} + \frac{1}{\bar{u}_2} + \frac{1}{\bar{u}_3} \right) - \frac{\sqrt{\beta^*}}{3\sqrt{2}} \quad (29)$$

In the following two subsections we will first discuss the shape of the spinodal region depending on the parameter σ , and then address the

specific forms of the dominating subspaces for initial concentrations contained on the line $(\bar{u}_1, \bar{u}_2, \bar{u}_3) = (\tau, \tau, 1 - 2\tau)$, for $0 < \tau < 1/2$. We also relate our results to recent numerical simulations of Eyre.^(7, 8)

3.2. Description of the Spinodal Region

As we mentioned in the introduction, the spinodal region is the set of all unstable homogeneous initial conditions \bar{u} in the Gibbs triangle \mathcal{G} . Equivalently, this is the set of all $\bar{u} \in \mathcal{G}$ for which the matrix B defined in (26) has at least one positive eigenvalue, i.e., for which we have $\beta_1 > 0$ (see (28)). Depending on the positive parameter σ , we can qualitatively distinguish the following four cases:

- For $0 < \sigma < 2$, both eigenvalues β_1 and β_2 are negative, the spinodal region is empty, and all homogeneous equilibria in the Gibbs triangle are stable.
- For $2 < \sigma < 8/3$, the spinodal region consists of three components, one along each of the sides of the Gibbs triangle. In those regions, we have $\beta_1 > 0 \geq \beta_2$, as shown in Fig. 3(a).
- For $8/3 < \sigma < 3$, the spinodal region is connected. Its complement consists of four components, one at each of the corners of the Gibbs triangle, and one in the center. Throughout the spinodal region we have $\beta_1 > 0 \geq \beta_2$. This is depicted in Fig. 3(b).
- For $\sigma > 3$, the spinodal region is again connected, but this time it contains everything except three small regions, one at each corner of the Gibbs triangle. In the center of the spinodal region both eigenvalues are positive, otherwise we have $\beta_1 > 0 \geq \beta_2$. See Fig. 3(c).

The transitional forms of the spinodal region at the values 2, 8/3, and 3, can be obtained easily and will therefore not be discussed in more detail.

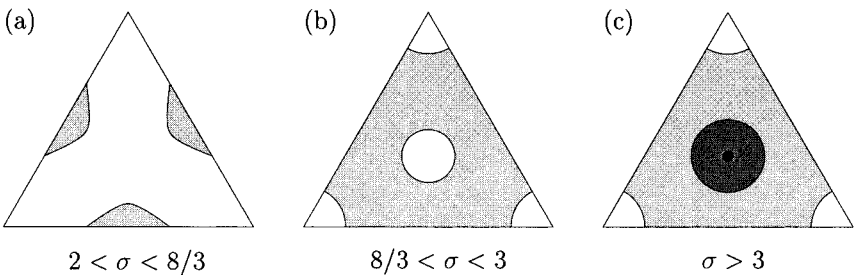


Fig. 3. Qualitative form of the spinodal region for various values of $\sigma > 2$. Dark shading corresponds to $\beta_1 \geq \beta_2 > 0$, lighter shading to $\beta_1 > 0 \geq \beta_2$.

The above discussion shows that only for $\sigma > 2$ the spinodal region is non-empty. For every $\sigma \in (2, 3)$ we have $\beta_1 > 0$ and $\beta_2 \leq 0$ throughout the spinodal region. Thus, for these values of σ , every homogeneous equilibrium in the spinodal region will satisfy the assumptions of Theorem 1.3 from the introduction. In other words, for $\sigma \in (2, 3)$ we always observe a pseudo-binary phase separation, i.e., there exists a strong mutual coupling between the components in the alloy.

The situation is different for $\sigma > 3$. Using (27), (28), and (29) one can easily see that the eigenvalues β_1 and β_2 are equal if and only if $(\bar{u}_1, \bar{u}_2, \bar{u}_3) = (1/3, 1/3, 1/3)$, and for $\sigma > 3$ they are also positive. Due to the discussion at the end of the introduction this implies that in a small neighborhood around the point $(1/3, 1/3, 1/3)$ in the Gibbs triangle, the assertions of Theorem 1.2 hold with $\beta_1 \approx \beta_2$. This is indicated by a small black dot in the center of Fig. 3(c). Outside of this neighborhood, Theorem 1.3 applies again, and we observe pseudo-binary decomposition.

3.3. Dominating Subspaces Along a One-Parameter Curve

In this subsection we want to describe the actual structure of the dominating subspaces \mathbb{Y}_ε^+ introduced in (10), if the initial condition is of the form

$$\bar{u} = (\bar{u}_1, \bar{u}_2, \bar{u}_3) = (\tau, \tau, 1 - 2\tau), \quad \text{for } 0 < \tau < 1/2$$

One can easily check that in this situation the matrix B defined in (26) has the two nontrivial eigenvalues

$$\tilde{\beta}_1 = \sigma - \frac{1}{\tau} \quad \text{and} \quad \tilde{\beta}_2 = \sigma - \frac{1}{3\tau(1 - 2\tau)}$$

with corresponding normalized eigenvectors $\tilde{w}_1 \in \mathbf{e}^\perp$ and $\tilde{w}_2 \in \mathbf{e}^\perp$ given by

$$\tilde{w}_1 = \frac{1}{\sqrt{2}} \cdot \begin{pmatrix} -1 \\ 1 \\ 0 \end{pmatrix} \quad \text{and} \quad \tilde{w}_2 = \frac{1}{\sqrt{6}} \cdot \begin{pmatrix} 1 \\ 1 \\ -2 \end{pmatrix}$$

See also Fig. 4, where $\tilde{\beta}_1$ (solid line) and $\tilde{\beta}_2$ (dashed line) are shown for $\sigma = 4$. Thus we have $\beta_1 = \max\{\tilde{\beta}_1, \tilde{\beta}_2\}$ and $\beta_2 = \min\{\tilde{\beta}_1, \tilde{\beta}_2\}$, which implies

$$\beta_1 = \sigma - \frac{1}{3\tau(1 - 2\tau)} \quad \text{and} \quad \beta_2 = \sigma - \frac{1}{\tau} \quad \text{for } 0 < \tau \leq \frac{1}{3},$$

$$\beta_1 = \sigma - \frac{1}{\tau} \quad \text{and} \quad \beta_2 = \sigma - \frac{1}{3\tau(1 - 2\tau)} \quad \text{for } \frac{1}{3} \leq \tau < \frac{1}{2}$$

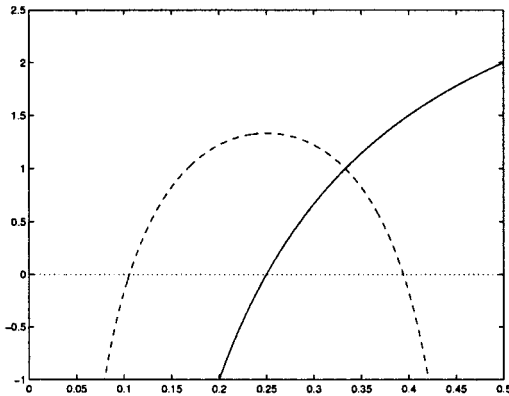


Fig. 4. Eigenvalues of B as functions of τ for $\sigma = 4$. The solid line shows $\tilde{\beta}_1$, the dashed line $\tilde{\beta}_2$.

The corresponding eigenvectors are $w_1 = \tilde{w}_2$ and $w_2 = \tilde{w}_1$ for $0 < \tau \leq 1/3$, as well as $w_1 = \tilde{w}_1$ and $w_2 = \tilde{w}_2$ for $1/3 \leq \tau < 1/2$.

These considerations allow the following descriptions of the dominating subspaces \mathbb{Y}_ε^+ defined in (10).

(i) Assume first that $\bar{u} = (\tau, \tau, 1 - 2\tau)$ is contained in the spinodal region and that $0 < \tau \leq 1/3 - \eta$ for some small $\eta > 0$. Then the dominating subspace is of the form

$$\mathbb{Y}_\varepsilon^+ = \tilde{w}_2 \cdot Y_\varepsilon^+$$

where Y_ε^+ denotes the dominating subspace for the binary Cahn–Hilliard model defined in (11). This situation occurs at the two uppermost points in Fig. 5(b) and (c). During the initial spinodal decomposition process most solutions originating near \bar{u} lead to functions which are close to $\bar{u} + \mathbb{Y}_\varepsilon^+$ (see our main result Theorem 2.9). Thus, the range of these functions is concentrated along the vertical line segments indicated in Fig. 5(b) and (c), which are parallel to \tilde{w}_2 .

(ii) If we have $\bar{u} = (\tau, \tau, 1 - 2\tau)$ in the spinodal region and $1/3 + \eta \leq \tau < 1/2$ for some small $\eta > 0$, then the dominating subspace is of the form

$$\mathbb{Y}_\varepsilon^+ = \tilde{w}_1 \cdot Y_\varepsilon^+$$

This situation occurs at the three lowermost points in Fig. 5(a), (b), and (c). Now the range of most solutions originating near \bar{u} is concentrated along the horizontal line segments shown in Fig. 5(a), (b), and (c), which are parallel to the eigenvector \tilde{w}_1 .

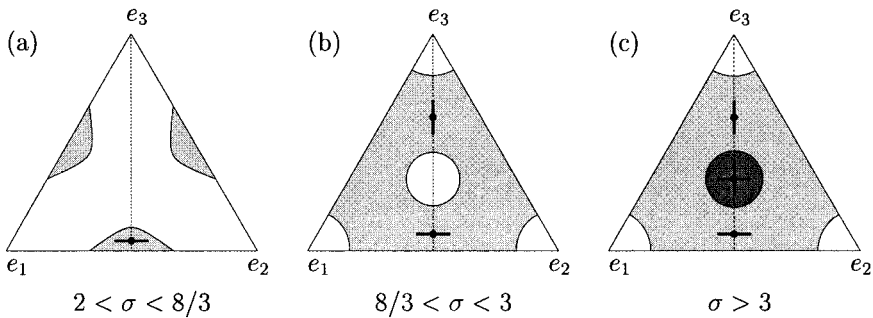


Fig. 5. Directions of the dominating subspaces \mathbb{Y}_ε^+ .

(iii) Finally, if $\bar{u} = (\tau, \tau, 1 - 2\tau)$ is such that $|\tau - 1/3| < \eta$ for some small $\eta > 0$, then the dominating subspace is of the form

$$\mathbb{Y}_\varepsilon^+ = \tilde{w}_1 \cdot Y_\varepsilon^+ \oplus \tilde{w}_2 \cdot Y_\varepsilon^+$$

This situation occurs only at those points in \mathcal{G} which are close to the middle of Fig. 5(c). Unlike in the previous two cases, the range of functions close to $\bar{u} + \mathbb{Y}_\varepsilon^+$ is no longer restricted to a specific line segment. Any direction in the Gibbs triangle is possible, there is no automatic coupling between the components.

The three cases described above can be illustrated using the numerical results of Eyre.^(7,8) Case (i) corresponds to ref. 8, Fig. 3; (ii) is depicted in ref. 7, Fig. 9 and Figs. 2 and 4 of ref. 8; and (iii) can be seen in ref. 7, Fig. 8.

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