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Sharp limit of the viscous Cahn–Hilliard equation and thermodynamic consistency

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Abstract Diffuse and sharp interface models represent two alternatives to describe phase transitions with an interface between two coexisting phases. The two model classes can be independently formulated. Thus there arises the problem whether the sharp limit of the diffuse model fits into the setting of a corresponding sharp interface model. We call a diffuse model admissible if its sharp limit produces interfacial jump conditions that are consistent with the balance equations and the second law of thermodynamics for sharp interfaces. We use special cases of the viscous Cahn–Hilliard equation to show that there are admissible as well as non-admissible diffuse interface models.

Keywords Cahn–Hilliard equation · Thermodynamics · Phase transitions · Asymptotic expansions

1 Introduction

We consider two coexisting phases in a body that are represented by two regions with high and low density. The two regions are separated by a moving phase boundary. In order to describe the evolution of that body, two alternative models are available, which are called diffuse interface model and sharp interface model.

A diffuse interface model describes the phase boundary as an interfacial layer. Within the layer, a so-called phase field smoothly changes from a high to a low value but with a steep gradient [1, 2]. On the other hand, the sharp interface model describes the interfacial layer by the evolution of a hypersurface [3–5].

Both alternatives can be established independently of each other. However, the physical basis of a sharp interface model consists of simpler assumptions, which are more directly related to experiments than the corresponding assumptions that are needed to establish a diffuse interface model. For this reason we consider the sharp model as a reference for a diffuse model in the same physical context.

In this context it is important that the diffuse model embodies the same physics as the sharp model. For this reason we study the properties of the diffuse model for small values of the interfacial thickness and then carry out the sharp limit of the diffuse model. To this end we use the method of formal matched asymptotic analysis, which is a well established tool for the understanding of diffuse models [1, 2, 6, 7]. If a sharp limit exists, we will obtain sharp interface equations as a limiting case of the diffuse model. In a second step we compare the limiting equations of the diffuse model with the corresponding equations of the sharp interface model. If we

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find coincidence, we call the diffuse model admissible. If there is no coincidence, the diffuse model is called non-admissible and has to be rejected.

In this paper the viscous Cahn–Hilliard model serves as an example [1, 8, 9]. For the same variables that appear in the viscous Cahn–Hilliard model, we establish a corresponding sharp interface model.

Comparing the limiting equations of the diffuse model with the equations of the sharp model leads to an interesting observation. Both models are equipped with local non-negative entropy productions. This property is preserved in the sharp limit of the diffuse model, i.e. the sharp limit of the entropy production is non-negative as well. In a next step we use that limit function to calculate the interfacial entropy production. We observe that we do not necessarily obtain a local non-negative interfacial entropy production. In more detail, the two main propositions of this paper are:

1. The sharp limit of the classical Cahn–Hilliard equation without viscosity leads to a negative interfacial entropy production.
2. The sharp limit of the viscous Cahn–Hilliard equation contains a parameter that can be adjusted so that a non-negative interfacial dissipation function results.

This unexpected behavior is a consequence of the fact that the Cahn–Hilliard model without viscosity has a local entropy production that is much smaller in the transition layer than in the bulk regions.

Thus the designer of phase field models has to consider this: Even for the simplest diffuse interface model, it is necessary to check whether the phase field model is admissible in the sense from above and represents a correct physical behavior.

The paper is organized as follows:

In Sect. 2 we derive the two kinds of models in the context of thermodynamics. In Sect. 3 we present and discuss our main results. Finally Sects. 4 and 5 contain a detailed description of the applied matched asymptotic analysis.

2 Description of diffuse and sharp interface models

In this sections we derive the viscous Cahn–Hilliard model and a corresponding sharp interface model in the context of thermodynamics in a one-dimensional setting.

2.1 Thermodynamics of the diffuse interface setting

The derivation of the viscous Cahn–Hilliard model starts from a more general model where variations of temperature are allowed.

Basic variables We consider a body $\Omega \subset \mathbb{R}$ whose thermodynamic state at time $t \geq 0$ is described in every point $x \in \Omega$ by two variables. These are the concentration $u: [0, \infty) \times \Omega \rightarrow \mathbb{R}$ and the internal energy density $e: [0, \infty) \times \Omega \rightarrow \mathbb{R}$.

Conservation laws The description of the phenomena relies on the conservation laws for mass and energy

$$\partial_t u + \partial_x f = 0 \quad \text{and} \quad \partial_t e + \partial_x q = 0. \quad (1)$$

The functions $f: [0, \infty) \times \Omega \rightarrow \mathbb{R}$ and $q: [0, \infty) \times \Omega \rightarrow \mathbb{R}$ are the diffusion flux and the heat flux, respectively. These quantities are related to e and u by constitutive functions that are compatible with second law of thermodynamics.

The second law of thermodynamics The following four axioms embody a simplified version of the second law of thermodynamics. The Axioms I and II contain universal statements, whereas Axioms III and IV describe the properties for the material at hand.

- I It exists an entropy density/entropy flux pair (η, φ) that satisfies an equation of balance

$$\partial_t \eta + \partial_x \varphi = \xi. \quad (2)$$

- II The entropy flux has to be determined so that the entropy production ξ is equipped with the following properties:

- (i) ξ is non-negative for every solution of the system of balance equations (1),
(ii) ξ is represented by a sum of binary products *flux* \times *driving force*:

$$\xi = \sum_{A=1}^N F_A D_A \geq 0. \quad (3)$$

- (iii) ξ is zero in equilibrium.

III Our constitutive model relies on an entropy density that is given by a concave constitutive function of the general form

$$\eta = h(e, u, u_x) \quad \text{with} \quad u_x = \partial_x u. \quad (4)$$

IV (Absolute) temperature and the chemical potential are defined by

$$\frac{1}{T} = \frac{\partial h}{\partial e} \quad \text{and} \quad \frac{\mu}{T} = - \left(\frac{\partial h}{\partial u} - \partial_x \frac{\partial h}{\partial u_x} \right). \quad (5)$$

Exploitation of the second law of thermodynamics Next we present a possible constitutive model that is compatible with the four axioms. At first we calculate the entropy production. To this end we insert the entropy function (4) into (2), carry out the time derivative and substitute the time derivatives of e and u by the corresponding equations of balance. After rearranging of terms we obtain

$$\dot{\xi} = \partial_x \left(\varphi - \frac{q}{T} + \frac{\mu f}{T} + \frac{\partial h}{\partial u_x} \partial_t u \right) + q \partial_x \left(\frac{1}{T} \right) - f \partial_x \left(\frac{\mu}{T} \right). \quad (6)$$

If we were to proceed in the usual way, we would end up with the classical Cahn–Hilliard model without viscosity. In order to introduce viscosity, we consider the identity

$$\frac{1}{T} (\partial_t u)^2 + \partial_x \left(\frac{f \partial_t u}{T} \right) - f \partial_x \left(\frac{\partial_t u}{T} \right) = 0, \quad (7)$$

which follows by multiplying the concentration balance (1)₁ with $\partial_t u/T$. Next we multiply the identity by a constant γ and add the expression to the entropy production,

$$\dot{\xi} = \partial_x \left(\varphi - \frac{q}{T} + \frac{(\mu + \gamma \partial_t u) f}{T} + \frac{\partial h}{\partial u_x} \partial_t u \right) + q \partial_x \left(\frac{1}{T} \right) - f \partial_x \left(\frac{\mu + \gamma \partial_t u}{T} \right) + \frac{\gamma}{T} (\partial_t u)^2. \quad (8)$$

Now we choose the entropy flux as

$$\varphi = \frac{q}{T} - \frac{(\mu + \gamma \partial_t u) f}{T} - \frac{\partial h}{\partial u_x} \partial_t u. \quad (9)$$

This choice implies the entropy production

$$\dot{\xi} = q \partial_x \left(\frac{1}{T} \right) - f \partial_x \left(\frac{\mu + \gamma \partial_t u}{T} \right) + \frac{\gamma}{T} (\partial_t u)^2. \quad (10)$$

Thus there is entropy production due to (i) heat conduction, (ii) diffusion and (iii) viscosity. The non-negativity of $\dot{\xi}$ is guaranteed by $\gamma \geq 0$ and the simple constitutive laws

$$q = a \partial_x \left(\frac{1}{T} \right) \quad \text{and} \quad f = -M \partial_x \left(\frac{\mu + \gamma \partial_t u}{T} \right) \quad \text{with} \quad a > 0, \quad M > 0. \quad (11)$$

Balance equation of the free energy For applications the energy density e is usually replaced by the temperature T as a variable. That substitution is accompanied by a Legendre transform of the entropy density. We introduce the free energy density $\psi = e - Th$ as a function of (T, u, u_x) . In terms of ψ , the chemical potential and the entropy flux can be written as

$$\mu = \frac{\partial \psi}{\partial u} - T \partial_x \left(\frac{1}{T} \frac{\partial \psi}{\partial u_x} \right) \quad \text{and} \quad \varphi = \frac{q}{T} - \frac{(\mu + \gamma \partial_t u) f}{T} + \frac{1}{T} \frac{\partial \psi}{\partial u_x} \partial_t u. \quad (12)$$

In the isothermal case, i.e. $T = \text{const.}$, the entropy balance can easily be written as a balance for the free energy. It results from a combination of the balance equations for energy and entropy and of the relations (12). The balance equation for the free energy comes out as

$$\partial_t \psi + \partial_x \left(f(\mu + \gamma \partial_t u) - \frac{\partial \psi}{\partial u_x} \partial_t u \right) = -T \xi \quad \text{with} \quad \xi = \frac{1}{M} f^2 + \frac{\gamma}{T} (\partial_t u)^2 \geq 0. \quad (13)$$

The viscous Cahn–Hilliard equation We proceed with the isothermal case where our problem is reduced to solve a single PDE for the concentration. The PDE follows from the diffusion flux (11)₂ and the balance law (1)₁. We obtain

$$\partial_t u = \frac{M}{T} \partial_{xx} (\mu + \gamma \partial_t u) \quad \text{with} \quad \mu = \frac{\partial \psi}{\partial u} - \partial_x \left(\frac{\partial \psi}{\partial u_x} \right). \quad (14)$$

The free energy density is chosen to be of van der Waals type, viz.

$$\psi = F(u) + \frac{\beta}{2} |\partial_x u|^2, \quad (15)$$

with the positive constant β and a double well function $F: [0, 1] \rightarrow \mathbb{R}$. In this case (14) becomes the well-known viscous Cahn–Hilliard equation, [1, 8, 9],

$$\partial_t u = -\partial_x f \quad \text{with} \quad f = -\frac{M}{T} \partial_x (\mu + \gamma \partial_t u) \quad \text{and} \quad \mu = F'(u) - \beta \partial_{xx} u. \quad (16)$$

We study the viscous Cahn–Hilliard equation on the domain $\Omega = [0, 1]$ with the initial and boundary values

$$u(0, x) = u_0(x), \quad \partial_x u(t, 0) = \partial_x u(t, 1) = 0, \quad f(t, 0) = f(t, 1) = 0. \quad (17)$$

It is known that the parameters β and γ control the width of the diffuse interface [1, 8, 9]. In order to handle only with one small parameter, we introduce the parameter $\varepsilon > 0$. The parameters β and γ are related to ε by the following substitutions

$$\beta \rightarrow \varepsilon^2 \beta \quad \text{and} \quad \gamma \rightarrow \varepsilon^2 \gamma. \quad (18)$$

2.2 Thermodynamics of the sharp interface setting

A further approach to describe the evolution of two coexisting phases is a sharp interface setting. Note, this approach is independent of a diffuse interface model. In the next paragraphs we derive a sharp interface model in the context of thermodynamics.

Basic variables We decompose the body $\Omega = [0, 1]$ into two bulk regions $\Omega^+(t)$ and $\Omega^-(t)$, which are separated by the interface, i.e. by the point x_I in the 1D case. The time-dependent function $\hat{x}_I: [0, \infty) \rightarrow \Omega$ determines the motion of x_I .

In the two bulk regions Ω^\pm the thermodynamic state is described by the concentration $u: [0, \infty) \times \Omega^\pm \rightarrow \mathbb{R}$ and the (internal) energy density $e: [0, \infty) \times \Omega^\pm \rightarrow \mathbb{R}$. Furthermore the interface is equipped with interfacial concentration $u_I: [0, \infty) \rightarrow \mathbb{R}$ and the interfacial internal energy density $e_I: [0, \infty) \rightarrow \mathbb{R}$.

The objective of the sharp interface setting is the determination of the densities u and e in the two bulk regions, the surface densities u_I, e_I of the interface and the interface position \hat{x}_I .

For a generic function χ existing in Ω^+ and Ω^- , respectively, we introduce the notation

$$\chi^\pm = \lim_{x \in \Omega^\pm \rightarrow x_I} \chi(x) \quad \text{and} \quad \llbracket \chi \rrbracket = \chi^+ - \chi^-. \quad (19)$$

χ^\pm are the limiting bulk values of the function χ at the interface. The double bracket $\llbracket \chi \rrbracket$ denotes the difference of χ across the interface.

Conservation laws In the bulk regions Ω^\pm we have the conservation laws for concentration and energy as in the diffuse setting, viz.

$$\partial_t u + \partial_x f = 0 \quad \text{and} \quad \partial_t e + \partial_x q = 0. \quad (20)$$

Across the interface the corresponding conservation laws read

$$\partial_t u_1 + \llbracket \dot{m}_1 \rrbracket = 0 \quad \text{and} \quad \partial_t e_1 + \llbracket e\dot{m} + q \rrbracket = 0. \quad (21)$$

The mass fluxes at the interface are defined as

$$\dot{m}_1^\pm = f^\pm - u^\pm \dot{x}_I, \quad \dot{m}_2^\pm = -f^\pm - (1 - u^\pm) \dot{x}_I \quad \text{and} \quad \dot{m}^\pm = \dot{m}_1^\pm + \dot{m}_2^\pm = -\dot{x}_I. \quad (22)$$

In the sharp interface setting we need constitutive laws for the fluxes f and q in the bulk regions and for the mass fluxes \dot{m}_1^\pm and \dot{m}_2^\pm at the interface. Next we derive explicit constitutive functions that are compatible with the second law of thermodynamics.

Remarks The described sharp interface model is a simplified version of a model for a two-constituent body with two partial mass densities ρ_1 and ρ_2 and two velocities v_1 and v_2 . The flux f is related to the velocity v_1 by $f = \rho_1 v_1$. The simplification concerns the total mass density $\rho = \rho_1 + \rho_2$ and the barycentric velocity $v = (\rho_1 v_1 + \rho_2 v_2)/\rho$. We assume $\rho = 1$ and $v = 0$. Thus the concentration $u = \rho_1/\rho$ is the only mass variable. The two assumptions imply special representations (22) of the mass fluxes $\dot{m}_1 = \rho_1(v_1 - \dot{x}_I)$ and $\dot{m}_2 = \rho_2(v_2 - \dot{x}_I)$ across the interface.

The second law of thermodynamics for the bulk In the sharp interface setting, the universal part of the second law of thermodynamics is the same as in the diffuse setting. Exclusively, the constitutive axiom III is changed:

III The entropy density is given by a concave constitutive function of the general form

$$\eta = h(e, u). \quad (23)$$

Note that the space derivative of u does not appear in the sharp interface setting.

Exploitation of the second law of thermodynamics for the bulk The resulting constitutive equations can be read off from the corresponding equations of the diffuse setting. The entropy flux is now chosen by

$$\varphi = \frac{q - \mu f}{T}, \quad (24)$$

and the entropy production is represented by two dissipative mechanisms:

$$\xi = q \partial_x \left(\frac{1}{T} \right) - f \partial_x \left(\frac{\mu}{T} \right). \quad (25)$$

Consequently, the thermodynamically consistent constitutive laws for the fluxes of heat and diffusion are

$$q = a \partial_x \left(\frac{1}{T} \right) \quad \text{and} \quad f = -M \partial_x \left(\frac{\mu}{T} \right) \quad \text{with} \quad a > 0, \quad M > 0. \quad (26)$$

Balance equation of the free energy In analogous way as in the diffuse interface setting we introduce the free energy density $\psi = e - Th$ as a function of (T, u) . In terms of ψ , the chemical potential and the entropy flux can be written as

$$\mu = \frac{\partial \psi}{\partial u} \quad \text{and} \quad \varphi = \frac{q}{T} - \frac{\mu f}{T}. \quad (27)$$

In the isothermal case, i.e. $T = \text{const.}$, the entropy balance can easily be written as a balance for the free energy. The balance equation for the free energy in the sharp interface setting comes out as

$$\partial_t \psi + \partial_x (f \mu) = -T \xi. \quad (28)$$

The second law of thermodynamics for the interface The constitutive laws for the mass fluxes $\dot{m}_{1,2}^\pm$ must satisfy the second law of thermodynamics for the interface. In an analogous manner to the bulk we give the corresponding axioms that constitute the second law of thermodynamics for the interface.

I It exists an entropy density η_I satisfying an equation of balance

$$\partial_t \eta_I + \llbracket \eta \dot{m} + \varphi \rrbracket = \xi_I. \quad (29)$$

II The interfacial entropy production ξ_I is equipped with the following properties:

- (i) ξ_I is non-negative for every solution of the system of balance equations (21)_{1,2},
- (ii) ξ_I is represented by a sum of binary products *flux* \times *driving force*:

$$\xi_I = \left[\left[\sum_{A=1}^N F_A D_A \right] \right] \geq 0. \quad (30)$$

(iii) ξ_I is zero in equilibrium.

III Our constitutive model relies on an entropy density that is given by a concave constitutive function of the general form

$$\eta_I = h_I(e_I, u_I). \quad (31)$$

IV Interfacial temperature and interfacial chemical potential are defined by

$$\frac{1}{T_I} = \frac{\partial h_I}{\partial e_I} \quad \text{and} \quad \frac{\mu_I}{T_I} = -\frac{\partial h_I}{\partial u_I}. \quad (32)$$

Axioms I and II are universal statements about the properties of entropy density and entropy production. The Axiom III assumes that the entropy is a constitutive quantity that must be given by a material-dependent function. For the case at hand that constitutive function is used in Axiom IV to give definitions of temperature and the chemical potential at the interface.

Remarks Recall that the described sharp interface model is a simplified version of a binary mixture. Thus there are two chemical potentials, whereas only a single chemical potential has appeared up to now. This fact is a consequence of the simplifying assumption that we only consider a single mass variable, namely u in the bulk and u_I on the interface. However, even in this case, two chemical potentials play a role in the theory. These are introduced here according to the definitions

$$\mu_1 = \psi + (1 - u)\mu, \quad \mu_2 = \psi - u\mu \quad \text{in the bulk,} \quad (33)$$

and

$$\mu_{I,1} = \psi_I + (1 - u_I)\mu_I, \quad \mu_{I,2} = \psi_I - u_I\mu_I \quad \text{on the interface.} \quad (34)$$

The quantity ψ_I is the interfacial free energy and is defined as $\psi_I = e_I - T_I \eta_I$.

Exploitation of the interfacial second law of thermodynamics for the interface Next we calculate the interfacial entropy production ξ_I . To this end we start with the entropy balance (29) and insert here the function (31) and the entropy flux (24) for the bulk. The time derivatives are eliminated by the balance laws (21), and after some rearrangements we obtain the structure of the interfacial entropy as it is stated by Axiom II:

$$\xi_I = \left[\left[(e\dot{m} + q) \left(\frac{1}{T} - \frac{1}{T_I} \right) \right] \right] - \left[\left[\dot{m}_1 \left(\frac{\mu_1}{T} - \frac{\mu_{I,1}}{T_I} \right) \right] \right] - \left[\left[\dot{m}_2 \left(\frac{\mu_2}{T} - \frac{\mu_{I,2}}{T_I} \right) \right] \right]. \quad (35)$$

The representations (22) of the mass fluxes imply the continuity of the flux \dot{m} , i.e. $\dot{m}^+ = \dot{m}^-$. Therefore the entropy production can also be written as

$$\xi_I = \left[\left[(e\dot{m} + q) \left(\frac{1}{T} - \frac{1}{T_I} \right) \right] \right] - \left[\left[\dot{m}_1 \left(\frac{\mu}{T} - \frac{\mu_I}{T_I} \right) \right] \right] - \dot{m} \left[\left[\frac{\mu_2}{T} \right] \right] \geq 0. \quad (36)$$

Thermodynamically consistent relations can be read off from this expression for the five independent fluxes q^\pm , \dot{m}_1^\pm and \dot{m} .

In this study we are only interested in the special case of a continuous mass flux \dot{m}_1 , i.e.

$$\llbracket \dot{m}_1 \rrbracket = 0. \quad (37)$$

Thus only one constitutive relation for the mass flux \dot{m}_1 has to be chosen. The continuity of the mass flux simplifies both the interfacial mass balance (21)₁ and the representation of the interfacial entropy production. Thus we have a time-independent interfacial mass density u_1 , i.e. $\partial_t u_1 = 0$, and an entropy production (35) of the form

$$\xi_I = \left[\left(e\dot{m} + q \right) \left(\frac{1}{T} - \frac{1}{T_1} \right) \right] - \dot{m}_1 \left[\frac{\mu_1}{T} \right] - \dot{m}_2 \left[\frac{\mu_2}{T} \right] \geq 0. \quad (38)$$

A simple possibility to satisfy (38) is given by the heat fluxes

$$q^\pm = e^\pm \dot{m} + a^\pm \left(\frac{1}{T^\pm} - \frac{1}{T_1} \right) \quad \text{with} \quad a^\pm > 0 \quad (39)$$

and by the kinetic relations

$$\begin{pmatrix} \left[\frac{\mu_1}{T} \right] \\ \left[\frac{\mu_2}{T} \right] \end{pmatrix} = - \begin{pmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{pmatrix} \begin{pmatrix} \dot{m}_1 \\ \dot{m}_2 \end{pmatrix}, \quad (40)$$

with a positive definite matrix L of kinetic coefficients.

For a more detailed description of sharp interface models, we refer the interested reader to [3–5].

Balance equation of the interfacial free energy We introduce the free energy density $\psi_1 = e_1 - T_1 h_1$ as a function of (T_1, u_1) . The chemical potential can be written as

$$\mu_1 = \frac{\partial \psi_1}{\partial u_1}. \quad (41)$$

Now we consider again the isothermal case, i.e. $T^+ = T^- = T_1$, and substitute in (29) the interfacial entropy by ψ_1 . The time derivative of e_1 will be eliminated by (21)₂. After some rearrangement of terms we obtain the balance equation of the interfacial free energy,

$$\partial_t \psi_1 + \llbracket \psi \dot{m} + \mu f \rrbracket = -T \xi_I \quad \text{with} \quad T \xi_I = - \llbracket \dot{m}_1 (\mu_1 - \mu_{1,1}) \rrbracket - \llbracket \dot{m}_2 (\mu_2 - \mu_{1,2}) \rrbracket \geq 0. \quad (42)$$

A useful alternative to represent the jump bracket in (42)₁ is

$$\llbracket \psi \dot{m} + \mu f \rrbracket = \llbracket \dot{m}_1 \mu_1 + \dot{m}_2 \mu_2 \rrbracket. \quad (43)$$

The identity (43) relies on (22) and (33).

Under a continuous mass flux \dot{m}_1 , the balance equation (42)₁ and the identity (43) imply a time-independent free energy and simplified entropy production,

$$\partial_t \psi_1 = 0 \quad \text{and} \quad T \xi_I = -\dot{m}_1 \llbracket \mu_1 \rrbracket - \dot{m}_2 \llbracket \mu_2 \rrbracket \geq 0 \quad (44)$$

The sharp interface model In an analogous manner to the diffuse setting we proceed with the isothermal case. The diffusion in the bulk is described by both the balance law (20) and the diffusion flux (26)₂,

$$\partial_t u = -\partial_x f \quad \text{with} \quad f = -\frac{M}{T} \partial_x \mu \quad \text{and} \quad \mu = \frac{\partial \psi}{\partial u} \quad \text{for} \quad x \in \Omega^\pm, \quad (45)$$

with a positive constant $M > 0$. It is understood that we may have different free energy functions ψ in Ω^+ and Ω^- . However, for simplicity this fact is not indicated here. Moreover, we use the free energy function

$$\psi = F(u) \quad (46)$$

with the same function F that appears in the diffuse setting.

The initial boundary value problem in the sharp interface setting considers an initial density and boundary values for the fluxes at the external boundaries

$$u(0, x) = u_0(x) \quad \text{and} \quad f(t, 0) = f(t, 1) = 0. \quad (47)$$

The boundary values at the interface I rely on jump conditions. For an isothermal interface with a continuous mass flux \dot{m}_1 the jump conditions are

$$\begin{pmatrix} \llbracket \mu_1 \rrbracket \\ \llbracket \mu_2 \rrbracket \end{pmatrix} = - \begin{pmatrix} L_{11} & L_{12} \\ L_{21} & L_{22} \end{pmatrix} \begin{pmatrix} \dot{m}_1 \\ \dot{m}_2 \end{pmatrix}. \quad (48)$$

The coefficient matrix L is positive definite.

Remarks Here we have restricted the sharp interface model to the special case of a continuous mass flux \dot{m}_1 ,

$$\llbracket \dot{m}_1 \rrbracket = 0. \quad (49)$$

In that case the definition (22) of the mass fluxes yields that the mass flux \dot{m}_2 is also continuous at the interface,

$$\llbracket \dot{m}_2 \rrbracket = 0. \quad (50)$$

Further we obtain from the continuity of the mass fluxes and the interfacial mass balance law (21)₁ we obtain the constancy of the interfacial concentration u_I , i.e.

$$\partial_t u_I = 0. \quad (51)$$

The kinetic relations (48) are the boundary conditions for the diffusion equations (45) at the interface. The evolution of the interface follows from the continuity of the mass flux \dot{m}_1 . There results

$$0 = \llbracket \dot{m}_1 \rrbracket = \llbracket f \rrbracket - \dot{x}_I \llbracket u \rrbracket. \quad (52)$$

This equation is known as Stefan condition and it serves to determine the interface speed.

3 The sharp limit of the viscous Cahn–Hilliard model

In order to describe a phase transition between two coexisting phases we have introduced two model variants. A diffuse interface model represented by the viscous Cahn–Hilliard equation and a sharp interface model with equations in the bulk phases and jump conditions at the interface. Our main objective is a comparison of the two settings.

The comparison of the models is based on the following idea: The diffuse model depends on the small parameter ε that controls the width of the diffuse interface. Therefore the concentration in the diffuse setting depends on the parameter ε . We indicate that dependency by writing u^ε . The method of formal asymptotic analysis relies on different expansions of u^ε in formal series. The outer expansion approximates the function u^ε in the bulk phases, while the inner expansion approximates u^ε in the transition layer. The expansions imply sharp interface equations that we can compare with the corresponding equations of the sharp interface setting.

In this section we present the main results of the asymptotic analysis followed by a detailed discussion. The details of the cumbersome calculations are found in the Sects. 4 and 5.

3.1 Formal asymptotic expansions

In the diffuse setting the interface between two bulk phases is defined by a hypersurface where the solution of the diffuse model assumes a given value u_* :

$$I^\varepsilon(t) = \{x \in (0, 1) : u^\varepsilon(t, x) = u_*\}. \quad (53)$$

The position of the interface is denoted by $x_I^\varepsilon(t)$. We have

$$u^\varepsilon(t, x_I^\varepsilon(t)) = u_*. \quad (54)$$

The sharp limit of the diffuse interface model relies on formal series in the small parameter ε . In the bulk phases we write

$$\chi^\varepsilon(t, x) = \chi^{(0)}(t, x) + \varepsilon\chi^{(1)}(t, x) + \varepsilon^2\chi^{(2)}(t, x) + \mathcal{O}(\varepsilon^3), \quad (55)$$

and call this the outer expansion of a generic function χ^ε . In the ε -neighborhood of the interface we introduce an inner coordinate z according to

$$x = x_1^\varepsilon(t) + \varepsilon z, \quad (56)$$

and define $\tilde{\chi}^\varepsilon(t, z) = \chi^\varepsilon(t, x_1^\varepsilon(t) + \varepsilon z)$. The formal series

$$\tilde{\chi}^\varepsilon(t, z) = \tilde{\chi}^{(0)}(t, z) + \varepsilon\tilde{\chi}^{(1)}(t, z) + \varepsilon^2\tilde{\chi}^{(2)}(t, z) + \mathcal{O}(\varepsilon^3) \quad (57)$$

is called the inner expansion. The resulting jump conditions at the interface are derived by means of the inner expansion.

3.2 Admissible approximation of a sharp interface model

We define requirements so that a diffuse model represents an admissible approximation of a sharp model.

The diffuse model (16)–(18) is an admissible approximation of the sharp model (45)–(48) up to order $\mathcal{O}(\varepsilon^k)$ if it has for every initial density u_0 a solution u^ε with the following properties:

1. The expansions (55) and (57) exist.
2. The outer expansions (55) satisfy the bulk equations (45) of the sharp model up to the order $\mathcal{O}(\varepsilon^k)$.
3. The expansions (55) and (57) imply representations of the interfacial density u_1 , the interfacial dissipation function ξ_1 and the kinetic coefficient matrix L . These quantities satisfy the jump conditions (48) of the sharp model up to $\mathcal{O}(\varepsilon^k)$.

3.3 The leading order equations

In the bulk phases the sharp limit of the viscous Cahn–Hilliard model yields in the leading order $\mathcal{O}(1)$

$$\partial_t u^{(0)} = -\partial_x f^{(0)} \quad \text{with} \quad f^{(0)} = -\frac{M}{T} \partial_x \mu^{(0)} \quad \text{for} \quad x \in \Omega^\pm, \quad (58)$$

where $\mu^{(0)} = F'(u^{(0)})$ is the corresponding chemical potential. The resulting free energy balance for the free energy $\psi^{(0)} = F(u^{(0)})$ in the bulk phases is given by

$$\partial_t \psi^{(0)} + \partial_x (f^{(0)} \mu^{(0)}) = -T \xi^{(0)} \quad \text{with} \quad \xi^{(0)} = \frac{1}{M} (f^{(0)})^2 \geq 0 \quad \text{for} \quad x \in \Omega^\pm. \quad (59)$$

In the same order we obtain the following conditions at the interface:

$$\partial_t u_1^{(0)} = 0, \quad \llbracket \dot{m}_{1,2}^{(0)} \rrbracket = 0, \quad \llbracket \mu_{1,2}^{(0)} \rrbracket = 0. \quad (60)$$

We can even show that the interfacial concentration is zero, i.e. $u_1^{(0)} = 0$. Moreover we obtain for the interfacial free energy balance in the leading order

$$\partial_t \psi_1^{(0)} = 0 \quad \text{and} \quad T \xi_1^{(0)} = -\llbracket \dot{m}_1^{(0)} \mu_1^{(0)} + \dot{m}_2^{(0)} \mu_2^{(0)} \rrbracket = 0. \quad (61)$$

Additionally the interfacial free energy in the leading order is zero, i.e. $\psi_1^{(0)} = 0$.

Thus, in the leading order, the sharp limit of the diffuse model agrees with the sharp interface model. The order $\mathcal{O}(1)$ approximation of the diffuse model is admissible. However, there is no interfacial dissipation and we find a sharp interface that is in local equilibrium.

Remarks 1. The representations of the chemical potentials $\mu_{1,2}^{(0),\pm}$ and of the mass fluxes $\dot{m}_{1,2}^{(0),\pm}$ in terms of the solutions of the viscous Cahn–Hilliard model are

$$\mu_1^{(0),\pm} = F(u^{(0),\pm}) + (1 - u^{(0),\pm})F'(u^{(0),\pm}), \quad \mu_2^{(0),\pm} = F(u^{(0),\pm}) - u^{(0),\pm}F'(u^{(0),\pm}), \quad (62)$$

$$\dot{m}_1^{(0),\pm} = f^{(0),\pm} - \dot{x}_I^{(0)} u^{(0),\pm}, \quad \dot{m}_2^{(0),\pm} = -f^{(0),\pm} - \dot{x}_I^{(0)} (1 - u^{(0),\pm}). \quad (63)$$

The derivation of these terms is given in Sect. 4.

2. The equations (60)₃ represent two algebraic equations for the interface values $u^{(0),\pm}$. Its graphic solution is called *Maxwell construction*.

3.4 The higher-order equations

In the bulk phases the sharp limit of the viscous Cahn–Hilliard model yields in the order $\mathcal{O}(\varepsilon)$

$$\partial_t u^{(1)} = -\partial_x f^{(1)} \quad \text{with} \quad f^{(1)} = -\frac{M}{T} \partial_x \mu^{(1)} \quad \text{for} \quad x \in \Omega^\pm, \quad (64)$$

where $\mu^{(1)} = F''(u^{(0)})u^{(1)}$ is the $\mathcal{O}(\varepsilon)$ contribution to the chemical potential. The resulting free energy balance for the free energy $\psi^{(1)} = F'(u^{(0)})u^{(1)}$ in the bulk phases in order $\mathcal{O}(\varepsilon)$ reads

$$\partial_t \psi^{(1)} + \partial_x (f\mu)^{(1)} = -T\xi^{(1)} \quad \text{with} \quad \xi^{(1)} = \frac{1}{M} (f^2)^{(1)} \quad \text{for} \quad x \in \Omega^\pm. \quad (65)$$

At the interface we obtain in the order $\mathcal{O}(\varepsilon)$ a constant interfacial density and continuous mass fluxes:

$$\partial_t u_1^{(1)} = 0 \quad \text{and} \quad \llbracket \dot{m}_{1,2}^{(1)} \rrbracket = 0. \quad (66)$$

However, in the order $\mathcal{O}(\varepsilon)$, the chemical potentials are not continuous anymore. Now they are given by kinetic relations:

$$\begin{pmatrix} \llbracket \mu_1^{(1)} \rrbracket \\ \llbracket \mu_2^{(1)} \rrbracket \end{pmatrix} = - \begin{pmatrix} K_{11} & K_{12} \\ K_{21} & K_{22} \end{pmatrix} \begin{pmatrix} \dot{m}_1^{(0)} \\ \dot{m}_2^{(0)} \end{pmatrix}. \quad (67)$$

The matrix K of kinetic coefficients in (67) is symmetric and has the representation

$$K = \begin{pmatrix} -2A + B + \gamma C & -A + B + \gamma C \\ -A + B + \gamma C & B + \gamma C \end{pmatrix}, \quad (68)$$

where the definitions of A , B and C are introduced in the following Remark. The interfacial free energy balance and the entropy production in the order $\mathcal{O}(\varepsilon)$ are

$$\partial_t \psi_1^{(1)} = 0 \quad \text{and} \quad -T\xi_1^{(1)} = \dot{m}_1^{(0)} \llbracket \mu_1^{(1)} \rrbracket + \dot{m}_2^{(0)} \llbracket \mu_2^{(1)} \rrbracket. \quad (69)$$

The balance laws and the constitutive functions of the order $\mathcal{O}(\varepsilon)$ satisfy the sharp interface setting. But up to now there is one property missing so that the diffuse model is also an admissible approximation in the order $\mathcal{O}(\varepsilon)$ of the sharp model. Recall that there is no dissipation in the leading order, i.e. $\xi_1^{(0)} = 0$. Hence the non-negativity of the interfacial entropy production must be guaranteed by $\xi_1^{(1)}$.

The non-negativity of the dissipation function $\xi_1^{(0)}$ is related to the properties of the matrix K . We insert the kinetic relations (67) in the dissipation function (69)₂ and obtain

$$T\xi_1^{(1)} = K_{11}(\dot{m}_1^{(0)})^2 + (K_{12} + K_{21})\dot{m}_1^{(0)}\dot{m}_2^{(0)} + K_{22}(\dot{m}_2^{(0)})^2. \quad (70)$$

Obviously, $\xi_1^{(1)}$ is non-negative for every mass flux $\dot{m}_{1,2}^{(0)}$ iff the matrix K is positive semi-definite.

The non-negativity of the interfacial dissipation $\xi_1^{(1)}$ and the properties of the matrix K will be discussed in the next sections.

Remarks 1. Above we have used some abbreviations to keep the equations readable. Here, we express all quantities in terms of the inner and expansion of the concentration u . The derivation of the equation from above is given in Sect. 4, where we will see that the quantities A , B and C in the definition of the matrix K are represented by

$$A = \frac{T}{M} \int_{-\infty}^0 (\tilde{u}^{(0)} - u^{(0,-)}) dz + \frac{T}{M} \int_0^{+\infty} (\tilde{u}^{(0)} - u^{(0,+)}) dz, \quad (71)$$

$$C = \int_{-\infty}^{+\infty} (\partial_z \tilde{u}^{(0)})^2 dz \quad (72)$$

$$B = \frac{T}{M} \int_{-\infty}^0 (\tilde{u}^{(0)})^2 - (u^{(0,-)})^2 dz + \frac{T}{M} \int_0^{+\infty} (\tilde{u}^{(0)})^2 - (u^{(0,+)})^2 dz. \quad (73)$$

The chemical potentials $\mu_{1,2}^{(1),\pm}$ and mass fluxes $\dot{m}_1^{(1),\pm}$ are defined as

$$\mu_1^{(1),\pm} = (1 - u^{(0),\pm})F''(u^{(0),\pm})u^{(1),\pm} + (\partial_x(F(u^{(0)}) + (1 - u^{(0)})F'(u^{(0)}))^\pm x_1^{(1)}, \tag{74}$$

$$\mu_2^{(1),\pm} = -u^{(0),\pm}F''(u^{(0),\pm})u^{(1),\pm} + (\partial_x(F(u^{(0)}) - u^{(0)}F'(u^{(0)}))^\pm x_1^{(1)}, \tag{75}$$

$$\dot{m}_1^{(1),\pm} = f^{(1),\pm} - \dot{x}_1^{(0)}u^{(1),\pm} - \dot{x}_1^{(1)}u^{(0),\pm} + (\partial_x f^{(0)} - \dot{x}_1^{(0)}\partial_x u^{(0)})^\pm x_1^{(1)}, \tag{76}$$

$$\dot{m}_2^{(1),\pm} = -f^{(1),\pm} + \dot{x}_1^{(0)}u^{(1),\pm} + \dot{x}_1^{(1)}u^{(0),\pm} - \dot{x}_1^{(1)} - (\partial_x f^{(0)} - \dot{x}_1^{(0)}\partial_x u^{(0)})^\pm x_1^{(1)}. \tag{77}$$

2. The quantities A , B and C of the matrix K depend on the concentration $\tilde{u}^{(0)}$, which has to be derived from the inner equation

$$\partial_z(\tilde{\mu}^{(0)} - \beta\partial_{zz}\tilde{u}^{(0)}) = 0 \quad \text{with } z \in (-\infty, +\infty) \text{ and } \tilde{\mu}^{(0)} = F'(\tilde{u}^{(0)}). \tag{78}$$

The necessary boundary conditions are

$$\tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} u^{(0),\pm}(t, x_1^{(0)}), \quad \partial_z \tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0, \quad \partial_{zz} \tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0. \tag{79}$$

The derivation of the inner equation and boundary conditions is described in detail in Sect. 4.

3. The interfacial densities $u_1^{(1)}$ and $\psi_1^{(1)}$ are given by their inner and outer leading parts,

$$u_1^{(1)} = \int_{-\infty}^0 (\tilde{u}^{(0)} - u^{(0),-}) dz + \int_0^{+\infty} (\tilde{u}^{(0)} - u^{(0),+}) dz, \tag{80}$$

$$\psi_1^{(1)} = \int_{-\infty}^0 (F(\tilde{u}^{(0)}) - F(u^{(0),-}) + \frac{\beta}{2}(\partial_z \tilde{u}^{(0)})^2) dz + \int_0^{+\infty} (F(\tilde{u}^{(0)}) - F(u^{(0),+}) + \frac{\beta}{2}(\partial_z \tilde{u}^{(0)})^2) dz. \tag{81}$$

4. Note the remarkable symmetry of the matrix K . It reflects the often postulated Onsager symmetry of kinetic coefficients. Apparently that symmetry is embodied in the viscous Cahn–Hilliard model.

3.5 Entropy production in the sharp limit of the viscous Cahn–Hilliard equation.

The question whether the sharp limit of the viscous Cahn–Hilliard equation is an admissible approximation up to the order $\mathcal{O}(\varepsilon)$ of the sharp model is immediately related to the sign of the interfacial entropy production.

The relation (70) yields that the entropy production $\xi_1^{(1)}$ is non-negative for every value of the mass fluxes $\dot{m}_{1,2}^{(0)}$ iff the matrix K is positive semi-definite. Thus it is only necessary to discuss the properties of the matrix K in the following.

To simplify the discussion on the definiteness of the matrix K we consider the double well function

$$F(u) = \frac{1}{2}u^2(u - 1)^2 \tag{82}$$

and set the Cahn–Hilliard coefficient $\beta = 1$, the temperature $T = 1$ and the mobility $M = 1$.

The exploitation of the jump conditions (60) with (62) leads to

$$u^{(0),-} = 0 \quad \text{and} \quad u^{(0),+} = 1. \tag{83}$$

According to the inner equation (78), the concentration $\tilde{u}^{(0)}$ is given by a simple traveling wave with shift parameter α :

$$\tilde{u}^{(0)} = \frac{1}{2} + \frac{1}{2} \tanh\left(\frac{1}{2}(z + \alpha)\right). \tag{84}$$

By means of (83) and (84) we exploit (80) and obtain the interfacial concentration $u_1^{(1)} = \alpha$.

Then we use (54) to relate the shift parameter α to the interface position x_1^ε . To this end recall that in the inner coordinate the interface is located at $z = 0$. Moreover in the leading order of the inner solution the condition (54) reads

$$\tilde{u}^{(0)}(t, z = 0) = u_* . \quad (85)$$

Using (84) it follows that the shift parameter α is determined by the level set value u_* . However, from the definition (54) of the interface position it is clear that the choice of the value u_* is arbitrary. Thus α is arbitrary as well. Vice versa for each parameter $\alpha \in \mathbb{R}$ there is a level set value u_* .

Next we use the definitions (71)–(73) to calculate the quantities A , B and C in the definition (68) of the matrix K . We obtain

$$A = \alpha , \quad B = \alpha - 1 \quad \text{and} \quad C = \frac{1}{6} . \quad (86)$$

The matrix K then assumes the explicit form

$$K = \begin{pmatrix} -\alpha - 1 + \gamma/6 & -1 + \gamma/6 \\ -1 + \gamma/6 & \alpha - 1 + \gamma/6 \end{pmatrix} . \quad (87)$$

Now it is easy to test whether the matrix K is positive semi-definite. At first we consider the classical Cahn–Hilliard model, i.e. we set $\gamma = 0$. In this case the eigenvalues $\lambda_{1,2}$ of K are

$$\lambda_{1,2} = -1 \pm \sqrt{1 + \alpha^2} . \quad (88)$$

We observe that one eigenvalue is negative and we conclude that the Cahn–Hilliard model without viscosity is not an admissible approximation of a sharp model in the order $\mathcal{O}(\varepsilon)$.

The case with viscosity, i.e. $\gamma \neq 0$, yields

$$\lambda_{1,2} = -\left(1 - \frac{\gamma}{6}\right) \pm \sqrt{\left(1 - \frac{\gamma}{6}\right)^2 + \alpha^2} . \quad (89)$$

For $\alpha \neq 0$ even here there is a negative eigenvalue. However, for $\alpha = 0$ and $\gamma \geq 6$ the eigenvalues are $\{0, \gamma/3 - 2\}$. Thus for $\gamma \geq 6$ there are no negative eigenvalues. The condition (85) uniquely determines the level set value u_* . For $\alpha = 0$ we obtain $u_* = \frac{1}{2}$ and a vanishing interfacial concentration, i.e. $u_1^{(1)} = 0$.

We conclude that the sharp limit of the viscous Cahn–Hilliard model generates a non-negative interfacial entropy production and is thus an admissible approximation of order $\mathcal{O}(\varepsilon)$ of a sharp interface model.

3.6 Relation between the inner entropy production of the diffuse model and the interfacial entropy production of the sharp model

We proceed with a discussion of the peculiar result of Sect. 3.5 but from a different point of view. Here we start from the inner entropy production $\tilde{\xi}^\varepsilon$ and study its relation to the interfacial entropy production ξ_1^ε that was calculated in Sect. 3.4.

The inner version of the entropy production (13)₂ reads

$$\tilde{\xi}^\varepsilon = \frac{1}{M} (\tilde{f}^\varepsilon)^2 + \frac{\gamma}{T} (\varepsilon \partial_t \tilde{u}^\varepsilon - \dot{x}_I^\varepsilon \partial_z \tilde{u}^\varepsilon)^2 \geq 0 . \quad (90)$$

Obviously, the limit $\varepsilon \rightarrow 0$ will not change the sign of the leading term. We obtain

$$\tilde{\xi}^{(0)} = \frac{1}{M} (\tilde{f}^{(0)})^2 + \frac{\gamma}{T} (\dot{x}_I^{(0)} \partial_z \tilde{u}^{(0)})^2 \geq 0 . \quad (91)$$

Next we relate the non-negative entropy production $\tilde{\xi}^{(0)}$ to the interfacial entropy production $\xi_1^{(1)}$. In Sect. 5.4 we show that this relation is given by

$$\xi_1^{(1)} = \int_{-\infty}^0 \tilde{\xi}^{(0)} - \xi^{(0),-} dz + \int_0^{+\infty} \tilde{\xi}^{(0)} - \xi^{(0),+} dz . \quad (92)$$

We observe that the sign of $\tilde{\xi}^{(0)}$ does not determine the sign of $\xi_1^{(1)}$.

Next we use a graphic interpretation of the relation (92) to derive its sign. To this end we use a further result of the asymptotics of Sect. 4

$$-\dot{x}_I^{(0)}\tilde{u}^{(0)}(z) + \tilde{f}^{(0)}(z) = c_0, \tag{93}$$

which is the basis for the Stefan condition (60)₂. The constant c_0 is given by $c_0 = \dot{m}_1^{(0),\pm}$.

Then (93) is used to eliminate the flux in (91) yielding

$$\tilde{\xi}^{(0)} = \frac{1}{M}(\dot{x}_I^{(0)}\tilde{u}^{(0)} + c_0)^2 + \frac{\gamma}{T}(\dot{x}_I^{(0)}\partial_z\tilde{u}^{(0)})^2 \geq 0. \tag{94}$$

We exploit this representation by means of the explicit density $\tilde{u}^{(0)}$ from the last section, (84). In this case it is also easy to determine the interface values of the bulk dissipation $\xi^{(0),\pm} = \lim_{z \rightarrow \pm\infty} \tilde{\xi}^{(0)}$. There results an explicit representation of $\tilde{\xi}^{(0)}$ depending on the parameter $c_0/\dot{x}_I^{(0)}$.

As before we consider at first the classical Cahn–Hilliard case with $\gamma = 0$. A short calculation shows that there are two different regimes depending on the value of $c_0/\dot{x}_I^{(0)}$. In the first regime $\tilde{\xi}^{(0)}$ is non-monotone and its global minimum is smaller than the entropy production $\xi^{(0),\pm}$ of the bulk phases at the interface. In the second regime $\tilde{\xi}^{(0)}$ is monotone. The two cases are depicted in Fig. 1.

Obviously the interfacial entropy production must be non-negative for any choice of the parameter $c_0/\dot{x}_I^{(0)}$. This is violated in the non-monotone case that is represented in the left part of Fig. 1. At first glance one might try to shift the parameter α to generate a positive value of $\xi_1^{(1)}$. However, this shift will lead to a conflict in the monotone case shown in Fig. 1 right.

The viscous Cahn–Hilliard equation, i.e. $\gamma \neq 0$, is more complicated, because $\xi_1^{(1)}$ additionally depends on the gradient of $\tilde{u}^{(0)}$. The Fig. 2 again shows two possible variants. Also here it is possible to choose $c_0/\dot{x}_I^{(0)}$

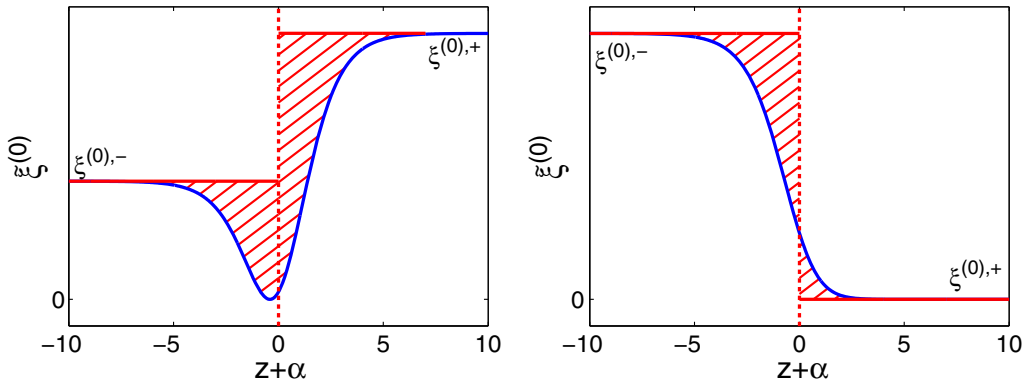


Fig. 1 Blue $\tilde{\xi}^{(0)}$, shaded areas indicate $\xi_1^{(1)}$, left $\gamma = 0$ and $c_0/\dot{x}_I^{(0)} = -0.4$, right $\gamma = 0$ and $c_0/\dot{x}_I^{(0)} = -1$ (color figure online)

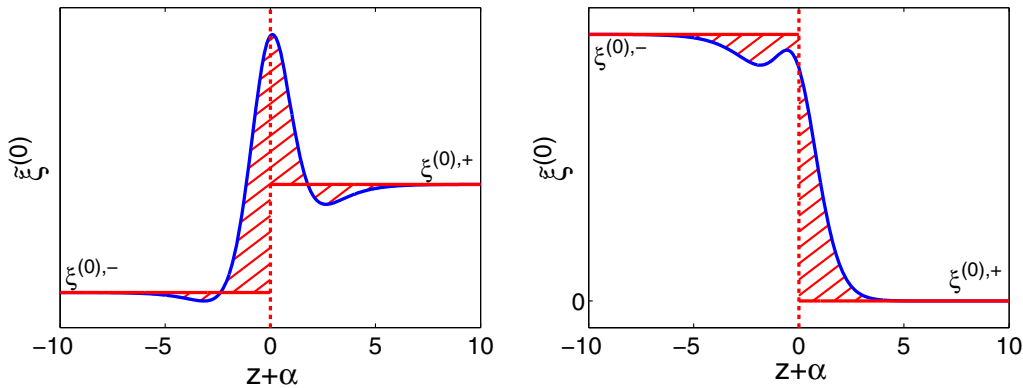


Fig. 2 Blue $\tilde{\xi}^{(0)}$, shaded areas indicate $\xi_1^{(1)}$, left $\gamma = 10$ and $c_0/\dot{x}_I^{(0)} = -0.4$, right $\gamma = 10$ and $c_0/\dot{x}_I^{(0)} = -1$ (color figure online)

so that the entropy production becomes negative. However, the proof in the last section shows that for $\alpha = 0$ and $\gamma > 6$ every choice of $c_0/\dot{x}_I^{(0)}$ leads to a non-negative entropy production.

4 Formal asymptotic analysis of the viscous Cahn–Hilliard model

The viscous Cahn–Hilliard model (16) contains two constitutive constants β and γ . We are interested here in the case where these constants are small. This is indicated by the substitutions $\beta \rightarrow \varepsilon^2\beta$ and $\gamma \rightarrow \varepsilon^2\gamma$ with β and γ now of order $\mathcal{O}(1)$. Thus we consider

$$\partial_t u = -\partial_x f \quad \text{with} \quad f = -\frac{M}{T}\partial_x(\mu + \gamma\varepsilon^2\partial_t u) \quad \text{and} \quad \mu = F'(u) - \beta\varepsilon^2\partial_{xx}u. \quad (95)$$

This equation is equipped with a free energy inequality, viz.

$$\partial_t \left(F(u) + \frac{\beta\varepsilon^2}{2}|\partial_x u|^2 \right) + \partial_x \left(f(\mu + \gamma\varepsilon^2\partial_t u) - \beta\varepsilon^2\partial_x u \partial_t u \right) = -T\xi, \quad (96)$$

where the entropy production is given by

$$\xi = \frac{1}{M}f^2 + \frac{\gamma\varepsilon^2}{T}(\partial_t u)^2 \geq 0. \quad (97)$$

The objective of this section is a study of the sharp limit $\varepsilon \rightarrow 0$ of the viscous Cahn–Hilliard equation. Moreover, we prove the representations of the sharp limit equations of the Cahn–Hilliard model, which are summarized and discussed in the previous section.

4.1 The rules of formal asymptotic analysis

We assume that an initial and boundary value problem for the viscous Cahn–Hilliard model (95) has a solution $u^\varepsilon(t, x)$ with the following properties:

1. u^ε develops a transition layer with smooth but steep gradient between two adjacent bulk phases.
2. We choose a value u_* lying in the transition layer and assume the existence of an interface that is defined by

$$I^\varepsilon(t) = \{x \in (0, 1) : u^\varepsilon(t, x) = u_*\}. \quad (98)$$

3. The interface I^ε at the position $x_1^\varepsilon \in C^1([0, \infty), \mathbb{R})$ generates two regions Ω^\pm with $\Omega^- = [0, x_1^\varepsilon]$ and $\Omega^+ = [x_1^\varepsilon, 1]$.
4. Away from I^ε , i.e. in the bulk phases, we assume the existence of an *outer expansion*

$$u^\varepsilon(t, x) = u^{(0)}(t, x) + \varepsilon u^{(1)}(t, x) + \mathcal{O}(\varepsilon^2). \quad (99)$$

5. In the ε -neighborhood of I^ε we introduce an inner coordinate z according to

$$x = x_1^\varepsilon + \varepsilon z, \quad (100)$$

and we define the inner variable by

$$\tilde{u}^\varepsilon(t, z) = u^\varepsilon(t, x_1^\varepsilon(t) + \varepsilon z). \quad (101)$$

6. The definition (101) implies the following transformation of derivatives:

$$\partial_x u^\varepsilon = \frac{1}{\varepsilon}\partial_z \tilde{u}^\varepsilon \quad \text{and} \quad \partial_t u^\varepsilon = \partial_t \tilde{u}^\varepsilon - \frac{1}{\varepsilon}\dot{x}_1^\varepsilon \partial_z \tilde{u}^\varepsilon. \quad (102)$$

7. Near to I^ε we assume the existence of an *inner expansion*

$$\tilde{u}^\varepsilon(t, z) = \tilde{u}^{(0)}(t, z) + \varepsilon \tilde{u}^{(1)}(t, z) + \varepsilon^2 \tilde{u}^{(2)}(t, z) + \mathcal{O}(\varepsilon^3). \quad (103)$$

8. Correspondingly, we assume the existence of an expansion of the interface position

$$x_1^\varepsilon(t) = x_1^{(0)}(t) + \varepsilon x_1^{(1)}(t) + \mathcal{O}(\varepsilon^2). \tag{104}$$

9. Matching conditions between inner and outer quantities are based on (101). We insert here the expansions (99), (103) and (104) and consider the limit $\varepsilon \rightarrow 0$, $z \rightarrow \infty$ with $\varepsilon z^n \rightarrow 0$ ($n \in \mathbb{N}_0$) to obtain the asymptotic correspondences

$$\tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} u^{(0),\pm}(t, x_1^{(0)}), \tag{105}$$

$$\partial_z \tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0, \tag{106}$$

$$\partial_{zz} \tilde{u}^{(0)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0, \tag{107}$$

$$\tilde{u}^{(1)}(t, z) - \partial_x u^{(0),\pm}(t, x_1^{(0)})(z + x_1^{(1)}) \xrightarrow{z \rightarrow \pm\infty} u^{(1),\pm}(t, x_1^{(0)}), \tag{108}$$

$$\partial_z \tilde{u}^{(1)}(t, z) \xrightarrow{z \rightarrow \pm\infty} \partial_x u^{(0),\pm}(t, x_1^{(0)}), \tag{109}$$

$$\partial_{zz} \tilde{u}^{(1)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0, \tag{110}$$

$$\partial_z \tilde{u}^{(2)}(t, z) - \partial_{xx} u^{(0),\pm}(t, x_1^{(0)})(z + x_1^{(1)}) \xrightarrow{z \rightarrow \pm\infty} \partial_x u^{(1),\pm}(t, x_1^{(0)}), \tag{111}$$

$$\partial_{zz} \tilde{u}^{(2)}(t, z) \xrightarrow{z \rightarrow \pm\infty} \partial_{xx} u^{(0),\pm}(t, x_1^{(0)}), \tag{112}$$

$$\partial_{zzz} \tilde{u}^{(2)}(t, z) \xrightarrow{z \rightarrow \pm\infty} 0, \tag{113}$$

$$\partial_t \tilde{u}^{(0)}(t, z) - \partial_x u^{(0),\pm}(t, x_1^{(0)}) \dot{x}_1^{(0)} \xrightarrow{z \rightarrow \pm\infty} \partial_t u^{(0),\pm}(t, x_1^{(0)}). \tag{114}$$

In order to obtain these matching conditions we follow the scheme that is described in [1,2]. It starts by inserting the inner and outer expansions into (101) and (102)₂, respectively.

$$\begin{aligned} & \tilde{u}^{(0)}(t, z) + \varepsilon \tilde{u}^{(1)}(t, z) + \varepsilon^2 \tilde{u}^{(2)}(t, z) + \mathcal{O}(\varepsilon^3) \\ &= u^{(0)}(t, x_1^{(0)}) + \varepsilon \left(u^{(1)}(t, x_1^{(0)}) + \partial_x u^{(0)}(t, x_1^{(0)})(x_1^{(1)} + z) \right) \\ & \quad + \varepsilon^2 \left(u^{(2)}(t, x_1^{(0)}) + \partial_x u^{(1)}(t, x_1^{(0)})(x_1^{(1)} + z) \right) \\ & \quad + \frac{\varepsilon^2}{2} \left(\partial_{xx} u^{(0)}(t, x_1^{(0)})(x_1^{(1)} + z)^2 + 2\partial_x u^{(0)}(t, x_1^{(0)})x_1^{(2)} \right) + \mathcal{O}(\varepsilon^3) \end{aligned} \tag{115}$$

The matching conditions (105)–(114) follow by comparing terms of power ε^n .

4.2 The bulk equations according to the outer setting

We insert the outer expansion (99) into the viscous Cahn–Hilliard equation (95) and obtain in the leading order $\mathcal{O}(1)$

$$\partial_t u^{(0)} = -\partial_x f^{(0)} \quad \text{with} \quad f^{(0)} = -\frac{M}{T} \partial_x \mu^{(0)} \quad \text{and} \quad \mu^{(0)} = F'(u^{(0)}) \quad \text{for} \quad x \in \Omega^\pm. \tag{116}$$

In the next order $\mathcal{O}(\varepsilon)$ we obtain

$$\partial_t u^{(1)} = -\partial_x f^{(1)} \quad \text{with} \quad f^{(1)} = -\frac{M}{T} \partial_x \mu^{(1)} \quad \text{and} \quad \mu^{(1)} = F''(u^{(0)})u^{(1)} \quad \text{for} \quad x \in \Omega^\pm. \tag{117}$$

The initial boundary value problem for (116) needs equations that describe the jumps of the variables across the interface between Ω^+ and Ω^- . Furthermore we need equations describing the motion of the interface. These equations will be derived next.

4.3 Equations of the inner setting

The jump conditions and the interface motion are encoded by the viscous Cahn–Hilliard equation (95) in the inner setting. It results from a transformation of the derivatives in (95) by means of the rules (102):

$$\partial_t \tilde{u}^\varepsilon - \frac{\dot{x}_I^\varepsilon}{\varepsilon} \partial_z \tilde{u}^\varepsilon + \frac{1}{\varepsilon} \partial_z \tilde{f}^\varepsilon = 0. \quad (118)$$

Here we introduce the inner expansion (103) and obtain in the leading order ε^{-2}

$$\partial_z(\tilde{\mu}^{(0)} - \beta \partial_{zz} \tilde{u}^{(0)}) = 0 \quad \text{with} \quad \tilde{\mu}^{(0)} = F'(\tilde{u}^{(0)}). \quad (119)$$

In the next order, viz. ε^{-1} , we have

$$-\dot{x}_I^{(0)} \partial_z \tilde{u}^{(0)} + \partial_z \tilde{f}^{(0)} = 0 \quad (120)$$

with the zeroth order flux

$$\tilde{f}^{(0)} = -\frac{M}{T} \partial_z \left(\tilde{\mu}^{(1)} - \beta \partial_{zz} \tilde{u}^{(1)} - \gamma \dot{x}_I^{(0)} \partial_z \tilde{u}^{(0)} \right) \quad \text{and} \quad \tilde{\mu}^{(1)} = F''(\tilde{u}^{(0)}) \tilde{u}^{(1)}. \quad (121)$$

Finally we identify the equations of order $\mathcal{O}(1)$,

$$\partial_t \tilde{u}^{(0)} - (\dot{x}_I \partial_z \tilde{u})^{(1)} + \partial_z \tilde{f}^{(1)} = 0 \quad (122)$$

with the first-order flux

$$\tilde{f}^{(1)} = -\frac{M}{T} \partial_z \left(\tilde{\mu}^{(2)} - \beta \partial_{zz} \tilde{u}^{(2)} + \gamma \partial_t \tilde{u}^{(0)} - \gamma (\dot{x}_I^{(1)} \partial_z \tilde{u}^{(0)} + \dot{x}_I^{(0)} \partial_z \tilde{u}^{(1)}) \right) \quad (123)$$

and the second-order chemical potential

$$\tilde{\mu}^{(2)} = \frac{1}{2} F'''(\tilde{u}^{(0)}) (\tilde{u}^{(1)})^2 + F''(\tilde{u}^{(0)}) \tilde{u}^{(2)}. \quad (124)$$

4.4 The interface conditions of the leading order problem

Next we derive the interface conditions of the leading order problem (116) from the equations of the inner setting (118)–(121). At first we integrate equation (119) and obtain

$$\tilde{\mu}^{(0)} - \beta \partial_{zz} \tilde{u}^{(0)} = c \quad \text{with} \quad c = \mu^{(0),\pm}. \quad (125)$$

The equality (125)₂ is a consequence of the matching conditions (105) and (107). Thus $\mu^{(0)}$ is continuous at the interface,

$$\llbracket \mu^{(0)} \rrbracket = 0 \quad \text{with} \quad \mu^{(0),\pm} = F'(u^{(0),\pm}). \quad (126)$$

Next we multiply (119) by $\tilde{u}^{(0)}$ to generate a further integral, which follows from the identity $\partial_x(F(u^\varepsilon) - u^\varepsilon F'(u^\varepsilon)) = -u^\varepsilon \partial_x F'(u^\varepsilon)$. The integral reads

$$-\tilde{\mu}_2^{(0)} - \beta (\tilde{u}^{(0)} \partial_{zz} \tilde{u}^{(0)}) + \frac{\beta}{2} (\partial_z \tilde{u}^{(0)})^2 = d \quad \text{with} \quad d = \mu_2^{(0),\pm}. \quad (127)$$

The integration constant (127)₂ follows from the matching conditions (105)–(107). Thus, the continuity of $\mu_2^{(0)}$ has been established,

$$\llbracket \mu_2^{(0)} \rrbracket = 0 \quad \text{with} \quad \mu_2^{(0),\pm} = F(u^{(0),\pm}) - u^{(0),\pm} F'(u^{(0),\pm}). \quad (128)$$

The Eqs. (126) and (128) imply the continuity of $\mu_1^{(0)} = F(u^{(0)}) + (1 - u^{(0)}) F'(u^{(0)})$,

$$\llbracket \mu_1^{(0)} \rrbracket = 0 \quad \text{with} \quad \mu_1^{(0),\pm} = F(u^{(0),\pm}) + (1 - u^{(0),\pm}) F'(u^{(0),\pm}). \quad (129)$$

Finally the last missing interface condition in the leading order follows from (120). The integration of (120) leads to

$$-\dot{x}_1^{(0)} \tilde{u}^{(0)} + \tilde{f}^{(0)} = m \quad \text{with} \quad m = -\dot{x}_1^{(0)} u^{(0),\pm} + f^{(0),\pm}. \tag{130}$$

As before we have determined the integration constant m by the matching conditions (105)–(107).

A comparison of $-\dot{x}_1^{(0)} u^{(0),\pm} + f^{(0),\pm}$ with the sharp interface definition (22) of the mass flux identifies the corresponding mass flux of the sharp limit procedure:

$$\dot{m}_1^{(0),\pm} = f^{(0),\pm} - \dot{x}_1^{(0)} u^{(0),\pm}. \tag{131}$$

We conclude that the diffuse model implies a continuous mass flux $\dot{m}_1^{(0)}$ in the sharp limit. Thus we write

$$\llbracket \dot{m}_1^{(0)} \rrbracket = 0. \tag{132}$$

The continuity of the second mass flux $\dot{m}_2^{(0)} = -f^{(0)} - \dot{x}_1^{(0)}(1 - u^{(0)})$ at $x_1^{(0)}$ follows directly

$$\llbracket \dot{m}_2^{(0)} \rrbracket = 0 \quad \text{with} \quad \dot{m}_2^{(0),\pm} = -f^{(0),\pm} - \dot{x}_1^{(0)}(1 - u^{(0),\pm}). \tag{133}$$

4.5 The interface concentration in the leading order

Up to now there is no sharp limit expression for the interfacial concentration u_I . We may establish consistency of the two models by the statement: $u_I^{(0)}$ is constant in time. Next we will show that the interfacial concentration $u_I^{(0)}$ even assumes the value zero.

To this end we consider the total mass in the sharp interface model. Here the total mass consists of the two masses of the bulk phases and of the interfacial mass. We have

$$M = \int_0^{x_I} u \, dx + \int_{x_I}^1 u \, dx + u_I. \tag{134}$$

In the diffuse interface setting we decompose the total mass $M = \int_0^1 u^\varepsilon \, dx$ into the corresponding contributions and hereafter we expand:

$$\begin{aligned} M &= \int_0^{x_I^\varepsilon - \varepsilon} u^\varepsilon \, dx + \varepsilon \int_{-1}^{+1} \tilde{u}^\varepsilon \, dz + \int_{x_I^\varepsilon + \varepsilon}^1 u^\varepsilon \, dx \\ &= \int_0^{x_I^{(0)}} u^{(0)} \, dx + \int_{x_I^{(0)}}^1 u^{(0)} \, dx + \varepsilon \int_{-1}^{+1} \tilde{u}^{(0)} \, dz \\ &\quad + \varepsilon \left(u^{(0),-}(x_I^{(1)} - 1) + \int_0^{x_I^{(0)}} u^{(1)} \, dx - u^{(0),+}(x_I^{(1)} + 1) + \int_{x_I^{(0)}}^1 u^{(1)} \, dx \right) + \mathcal{O}(\varepsilon^2). \end{aligned} \tag{135}$$

In the leading order we obtain

$$M = \int_0^{x_I^{(0)}} u^{(0)} \, dx + \int_{x_I^{(0)}}^1 u^{(0)} \, dx. \tag{136}$$

As before we compare the sharp interface result (134) with the diffuse interface result (136). We conclude that the total mass is already used up in the bulk phases. Thus it follows

$$u_I^{(0)} = 0. \tag{137}$$

4.6 The interface conditions of the first-order problem, Part 1: Interfacial mass balance

Here we derive the interface condition for the higher-order problem (117). In preparation we rewrite the bulk equation (116)₁ at the \pm sides of the interface. To this end we use the two identities

$$\partial_t u^{(0),\pm}(t, x_1^{(0)}) = \dot{u}^{(0),\pm}(t, x_1^{(0)}(t)) - \dot{x}_1^{(0)}(\partial_x u^{(0)})^\pm(t, x_1^{(0)}) \quad (138)$$

and obtain (116)₁ in the form

$$\dot{u}^{(0),\pm} - \dot{x}_1^{(0)}(\partial_x u^{(0)})^\pm + (\partial_x f^{(0)})^\pm = 0. \quad (139)$$

This equation is now subtracted from the order $\mathcal{O}(1)$ equation (122) of the inner setting. After some rearrangements we have

$$\partial_t(\tilde{u}^{(0)} - u^{(0),\pm}) + \partial_z \left(-(\dot{x}_1 \partial_z \tilde{u})^{(1)} + \tilde{f}^{(1)} - (-\dot{x}_1^{(0)} \partial_x u^{(0)} + \partial_x f^{(0)})^\pm (z + x_1^{(1)}) \right) = 0. \quad (140)$$

Next we integrate (140) along the z coordinate, namely from $-\infty$ to 0 on the $-$ side and from 0 to $+\infty$ on the $+$ side of the interface. Then we add the resulting equations. Here we apply the matching conditions (105)–(113) to the term under the z derivative of (140). By this procedure we deduce from (140) the interfacial mass balance

$$\partial_t u_1^{(1)} + \llbracket \dot{m}_1^{(1)} \rrbracket = 0. \quad (141)$$

The interface concentration $u_1^{(1)}$ and the mass fluxes $\dot{m}_1^{(1),\pm}$ of the order $\mathcal{O}(\varepsilon)$ are defined as

$$u_1^{(1)} = \int_{-\infty}^0 (\tilde{u}^{(0)} - u^{(0),-}) dz + \int_0^{+\infty} (\tilde{u}^{(0)} - u^{(0),+}) dz, \quad (142)$$

$$\dot{m}_1^{(1),\pm} = f^{(1),\pm} - \dot{x}_1^{(0)} u^{(1),\pm} - \dot{x}_1^{(1)} u^{(0),\pm} + (\partial_x f^{(0)} - \dot{x}_1^{(0)} \partial_x u^{(0)})^\pm x_1^{(1)}. \quad (143)$$

Furthermore the representation (142) shows that $u_1^{(1)}$ does not depend on time because $u^{(0),\pm}$ is determined by the time-independent equations (126) and (128). Thus the integration constant c in (125) is independent of time. Then also $\tilde{u}^{(0)}$ is time independent. We conclude from (141)

$$\partial_t u_1^{(1)} = 0 \quad \text{and} \quad \llbracket \dot{m}_1^{(1)} \rrbracket = 0. \quad (144)$$

We directly obtain from the continuity of the mass flux $\dot{m}_1^{(1)}$:

$$\llbracket \dot{m}_2^{(1)} \rrbracket = 0 \quad \text{with} \quad \dot{m}_2^{(1),\pm} = -f^{(1),\pm} + \dot{x}_1^{(0)} u^{(1),\pm} + \dot{x}_1^{(1)} u^{(0),\pm} - \dot{x}_1^{(1)} - (\partial_x f^{(0)} - \dot{x}_1^{(0)} \partial_x u^{(0)})^\pm x_1^{(1)}. \quad (145)$$

4.7 The interface conditions of the first-order problem, Part 2: Kinetic relations.

The first-order problem (117) needs two further interface conditions. These are the *kinetic relations*. We write equation (130) as

$$-\dot{x}_1^{(0)}(\tilde{u}^{(0)} - u^{(0),\pm}) + \tilde{f}^{(0)} - f^{(0),\pm} = 0. \quad (146)$$

Here we insert the flux $\tilde{f}^{(0)}$ according to (121) and $f^{(0)} = -\frac{M}{T} \partial_x \mu^{(0)}$ from (116) to obtain

$$-\dot{x}_1^{(0)}(\tilde{u}^{(0)} - u^{(0),\pm}) - \frac{M}{T} \partial_z \left(\tilde{\mu}^{(1)} - \partial_x \mu^{(0),\pm} (z + x_1^{(1)}) - \beta \partial_{zz} \tilde{u}^{(1)} - \gamma \dot{x}_1^{(0)} \partial_z \tilde{u}^{(0)} \right) = 0. \quad (147)$$

As before we integrate this equation along the z coordinate and apply again the matching conditions (105)–(113) to the various terms under the z derivative. The result is

$$\llbracket \mu^{(1)} \rrbracket = A \dot{m}^{(0)} \quad \text{with} \quad A = \frac{T}{M} u_1^{(1)}. \quad (148)$$

The derivation of the second missing interface condition starts with (120). Multiplication of equation (120) by $\tilde{u}^{(0)}$ yields

$$-(-\dot{x}_I^{(0)}\tilde{u}^{(0)} + \tilde{f}^{(0)})\partial_z\tilde{u}^{(0)} + \partial_z(-\dot{x}_I(\tilde{u}^{(0)})^2 + \tilde{u}^{(0)}\tilde{f}^{(0)}) = 0. \quad (149)$$

By (130) and (131) we have $\dot{m}_1^{(0)} = -\dot{x}_I^{(0)}\tilde{u}^{(0)} + \tilde{f}^{(0)}$ and furthermore $\dot{m}^{(0)} = -\dot{x}_I^{(0)}$. Recall that $\dot{m}_1^{(0)}$ and $\dot{m}^{(0)}$ are independent of z . We insert the fluxes $\dot{m}_1^{(0)}$ and $\dot{m}^{(0)}$ in (149). Afterward we can easily integrate (149) and obtain with an appropriately chosen integration constant

$$-\dot{m}_1^{(0)}(\tilde{u}^{(0)} - u^{(0),\pm}) + \dot{m}^{(0)}((\tilde{u}^{(0)})^2 - (u^{(0),\pm})^2) + (\tilde{u}^{(0)}\tilde{f}^{(0)} - u^{(0),\pm}f^{(0),\pm}) = 0. \quad (150)$$

Herein the flux $\tilde{f}^{(0)}$ is inserted according to (121). The flux $f^{(0)} = -\frac{M}{T}\partial_x\mu^{(0)}$ results from (116). In both fluxes, we replace the space derivatives of $F'(u)$ by derivatives of $F(u) - uF'(u)$. This substitution relies on the identity $\partial_x(F(u^\varepsilon) - u^\varepsilon F'(u^\varepsilon)) = -u^\varepsilon\partial_x F'(u^\varepsilon)$ implying

$$u^{(0)}\partial_x\mu^{(0)} = \partial_x\mu_2^{(0)} \quad \text{and} \quad \tilde{u}^{(1)}\partial_z\tilde{\mu}^{(0)} + \tilde{u}^{(0)}\partial_z\tilde{\mu}^{(1)} = \partial_z\tilde{\mu}_2^{(1)}. \quad (151)$$

Thus (150) becomes

$$\begin{aligned} &-\dot{m}_1^{(0)}(\tilde{u}^{(0)} - u^{(0),\pm}) + \dot{m}^{(0)}((\tilde{u}^{(0)})^2 - (u^{(0),\pm})^2) + \frac{M}{T}(\partial_z\tilde{\mu}_2^{(1)} - \partial_x\mu_2^{(0),\pm}) \\ &+ \frac{M}{T}\beta\tilde{u}^{(0)}\partial_{zzz}\tilde{u}^{(1)} + \frac{M}{T}\tilde{u}^{(1)}\partial_z\tilde{\mu}^{(0)} + \gamma\frac{M}{T}\dot{x}_I^{(0)}\tilde{u}^{(0)}\partial_{zz}\tilde{u}^{(0)} = 0. \end{aligned} \quad (152)$$

Finally we use (119)₁, viz. $\partial_z(\tilde{\mu}^{(0)} - \partial_{zz}\tilde{u}^{(0)}) = 0$, to obtain

$$\begin{aligned} &-\dot{m}_1^{(0)}(\tilde{u}^{(0)} - u^{(0),\pm}) + \dot{m}^{(0)}((\tilde{u}^{(0)})^2 - (u^{(0),\pm})^2) + \frac{M}{T}(\partial_z\tilde{\mu}_2^{(1)} - \partial_x\mu_2^{(0),\pm}) \\ &+ \beta\frac{M}{T}\partial_z(\tilde{u}^{(0)}\partial_{zz}\tilde{u}^{(1)} + \tilde{u}^{(1)}\partial_{zz}\tilde{u}^{(0)} - \partial_z\tilde{u}^{(0)}\partial_z\tilde{u}^{(1)}) \\ &+ \gamma\frac{M}{T}\dot{x}_I^{(0)}\partial_z(\tilde{u}^{(0)}\partial_z\tilde{u}^{(0)}) - \gamma\frac{M}{T}\dot{x}_I^{(0)}(\partial_z\tilde{u}^{(0)})^2 = 0. \end{aligned} \quad (153)$$

Integration of this equation along the z coordinate yields

$$\llbracket\mu_2^{(1)}\rrbracket = A\dot{m}_1^{(0)} - (B + \gamma C)\dot{m}^{(0)} \quad (154)$$

with the abbreviations

$$C = \int_{-\infty}^{+\infty} (\partial_z\tilde{u}^{(0)})^2 dz, \quad (155)$$

$$B = \frac{T}{M} \int_{-\infty}^0 (\tilde{u}^{(0)})^2 - (u^{(0),-})^2 dz + \frac{T}{M} \int_0^{+\infty} (\tilde{u}^{(0)})^2 - (u^{(0),+})^2 dz. \quad (156)$$

A simple rearrangement shows that the kinetic relations (148) and (154) can be rewritten in the symmetric form:

$$\llbracket\mu_1^{(1)}\rrbracket = -K_{11}\dot{m}_1^{(0)} - K_{12}\dot{m}_2^{(0)}, \quad (157)$$

$$\llbracket\mu_2^{(1)}\rrbracket = -K_{21}\dot{m}_1^{(0)} - K_{22}\dot{m}_2^{(0)}, \quad (158)$$

with the symmetric coefficient matrix

$$K = \begin{pmatrix} -2A + B + \gamma C & -A + B + \gamma C \\ -A + B + \gamma C & B + \gamma C \end{pmatrix}. \quad (159)$$

5 Formal asymptotic analysis of the interfacial free energy inequality

In this section we derive the interfacial free energy inequality in an analogous manner as we have derived the jump conditions for the concentration. We start from the free energy inequality (96) of the diffuse model and insert the outer and the inner expansions of the concentration u . After this we compare the resulting equations with the corresponding equations of the sharp model. That comparison allows to identify the interfacial entropy production of the diffuse model.

5.1 The free energy inequality in the outer setting

We insert the outer expansion (99) into the free energy inequality (96) of the viscous Cahn–Hilliard equation and the dissipation function (97). In the leading order we obtain the balance law

$$\partial_t \psi^{(0)} + \partial_x (f^{(0)} \mu^{(0)}) = -T \xi^{(0)} \quad (160)$$

where the free energy and the entropy production are given by

$$\psi^{(0)} = F(u^{(0)}) \quad \text{and} \quad \xi^{(0)} = \frac{1}{M} (f^{(0)})^2 \geq 0. \quad (161)$$

In the next order the free energy balance reads

$$\partial_t \psi^{(1)} + \partial_x (f \mu)^{(1)} = -T \xi^{(1)}, \quad (162)$$

where the free energy and the entropy production are given by

$$\psi^{(1)} = F'(u^{(0)}) u^{(1)} \quad \text{and} \quad \xi^{(1)} = \frac{1}{M} (f^2)^{(1)}. \quad (163)$$

5.2 The free energy inequality in the inner setting

At first we transform the free energy balance (96) by means of the rules (102) to obtain

$$\begin{aligned} & \frac{\partial}{\partial t} \left(\tilde{F}^\varepsilon + \frac{\beta}{2} (\partial_z \tilde{u}^\varepsilon)^2 \right) - \frac{\dot{x}_1^\varepsilon}{\varepsilon} \frac{\partial}{\partial z} \left(\tilde{F}^\varepsilon + \frac{\beta}{2} (\partial_z \tilde{u}^\varepsilon)^2 \right) \\ & + \frac{1}{\varepsilon} \frac{\partial}{\partial z} \left(\tilde{f}^\varepsilon (\tilde{\mu}^\varepsilon + \varepsilon \gamma (\varepsilon \partial_t \tilde{u}^\varepsilon - \dot{x}_1^\varepsilon \partial_z \tilde{u}^\varepsilon)) - \beta \partial_z \tilde{u}^\varepsilon (\varepsilon \partial_t \tilde{u}^\varepsilon - \dot{x}_1^\varepsilon \partial_z \tilde{u}^\varepsilon) \right) = -T \tilde{\xi}^\varepsilon. \end{aligned} \quad (164)$$

The transformed dissipation function (97) reads in inner variables:

$$\tilde{\xi}^\varepsilon = \frac{1}{M} (\tilde{f}^\varepsilon)^2 + \frac{\gamma}{T} (\varepsilon \partial_t \tilde{u}^\varepsilon - \dot{x}_1^\varepsilon \partial_z \tilde{u}^\varepsilon)^2 \geq 0. \quad (165)$$

In (164) we introduce the inner expansion (103). Due to (119) and (120) we conclude that the terms of the orders ε^{-2} and ε^{-1} of (164) are identically satisfied. The next order $\mathcal{O}(1)$ yields

$$\begin{aligned} & \partial_t \left(\tilde{F}^{(0)} + \frac{\beta}{2} (\partial_z \tilde{u}^{(0)})^2 \right) - \partial_z \left[\dot{x}_1^{(0)} \tilde{F}^{(0)} \right]^{(1)} + \partial_z \left[\tilde{f}^{(0)} \tilde{\mu}^{(0)} \right]^{(1)} \\ & + \frac{\beta}{2} \partial_z \left[\dot{x}_1^{(0)} (\partial_z \tilde{u}^{(0)})^2 \right]^{(1)} - \beta \partial_z (\partial_t \tilde{u}^{(0)} \partial_z \tilde{u}^{(0)}) - \gamma \partial_z (\dot{x}_1^{(0)} \tilde{f}^{(0)} \partial_z \tilde{u}^{(0)}) = -T \tilde{\xi}^{(0)}, \end{aligned} \quad (166)$$

where the dissipation function is given by

$$\tilde{\xi}^{(0)} = \frac{1}{M} (\tilde{f}^{(0)})^2 + \frac{\gamma}{T} (\dot{x}_1^{(0)} \partial_z \tilde{u}^{(0)})^2 \geq 0. \quad (167)$$

In (166) we have used the abbreviations

$$\left[\dot{x}_1^\varepsilon \tilde{F}^\varepsilon \right]^{(1)} = \dot{x}_1^{(0)} \tilde{F}^{(1)} + \dot{x}_1^{(1)} \tilde{F}^{(0)} \quad (168)$$

$$\left[\tilde{f}^\varepsilon \tilde{\mu}^\varepsilon \right]^{(1)} = \tilde{f}^{(1)} (\tilde{\mu}^{(0)} - \beta \partial_{zz} \tilde{u}^{(0)}) + \tilde{f}^{(0)} (\tilde{\mu}^{(1)} - \beta \partial_{zz} \tilde{u}^{(1)}) \quad (169)$$

$$\left[\dot{x}_1^\varepsilon (\partial_z \tilde{u}^\varepsilon)^2 \right]^{(1)} = \dot{x}_1^{(1)} (\partial_z \tilde{u}^{(0)})^2 + 2 \dot{x}_1^{(0)} \partial_z \tilde{u}^{(0)} \partial_z \tilde{u}^{(1)}. \quad (170)$$

5.3 The interfacial free energy inequality of the leading order

The leading orders, i.e. ε^{-2} and ε^{-1} , of the diffuse free energy inequality in inner variables do not give any new information because the leading orders of the inequality are identically satisfied. This is a consequence of previous results. In fact, the continuity of both the mass fluxes (132) and (133) and the chemical potentials (128) and (129) yields in the leading order

$$\dot{m}_1^{(0)} \llbracket \mu_1^{(0)} \rrbracket + \dot{m}_2^{(0)} \llbracket \mu_2^{(0)} \rrbracket = 0. \tag{171}$$

A comparison of (171) with the free energy inequality (44) of the sharp interface model leads to the conclusion: There is no dissipation in the leading order, i.e.

$$\xi_1^{(0)} = 0. \tag{172}$$

Finally we show that the free energy $\psi_I^{(0)}$ in the sharp limit is zero. This follows in an analogous manner to the reasoning of $u_1^{(0)} = 0$. The total free energy Ψ of the system is an additive quantity. Therefore it can be represented in the sharp interface setting by

$$\Psi = \int_0^{x_I} \psi \, dx + \int_{x_I}^1 \psi \, dx + \psi_I. \tag{173}$$

In the diffuse interface setting we decompose the total free energy $\Psi = \int_0^1 \psi^\varepsilon \, dx$ into the corresponding contributions and hereafter we expand:

$$\begin{aligned} \Psi &= \int_0^{x_I^{(0)}} \psi^{(0)} \, dx + \int_{x_I^{(0)}}^1 \psi^{(0)} \, dx + \varepsilon \int_{-1}^{+1} \tilde{\psi}^{(0)} \, dz \\ &+ \varepsilon \left(\psi^{(0),-}(x_I^{(1)} - 1) + \int_0^{x_I^{(0)}} \psi^{(1)} \, dx - \psi u^{(0),+}(x_I^{(1)} + 1) + \int_{x_I^{(0)}}^1 \psi^{(1)} \, dx \right) + \mathcal{O}(\varepsilon^2). \end{aligned} \tag{174}$$

In the leading order we obtain

$$\Psi = \int_0^{x_I^{(0)}} \psi^{(0)} \, dx + \int_{x_I^{(0)}}^1 \psi^{(0)} \, dx. \tag{175}$$

As before we compare the sharp interface result (173) with the diffuse interface result (175). We conclude that the total free energy Ψ has only bulk contributions. Thus it follows

$$\psi_I^{(0)} = 0. \tag{176}$$

5.4 The higher-order interfacial free energy balance inequality

From the inner equation (166) we subtract the corresponding bulk equations (160) at the interface. The result can be written as

$$\begin{aligned} &\partial_t \left(\tilde{F}^{(0)} - F^{(0),\pm} + \frac{\beta}{2} (\partial_z \tilde{u}^{(0)})^2 \right) \\ &- \partial_z \left[\dot{x}_I^\varepsilon \tilde{F}^\varepsilon \right]^{(1)} + \partial_x (\dot{x}_I^{(0)} F^{(0),\pm}) + \partial_z \left[\tilde{f}^\varepsilon \tilde{\mu}^\varepsilon \right]^{(1)} - \partial_r (f^{(0),\pm} \mu^{(0),\pm}) \\ &+ \frac{\beta}{2} \partial_z \left[\dot{x}_I^\varepsilon (\partial_z \tilde{c}^\varepsilon)^2 \right]^{(1)} - \beta \partial_z (\partial_t \tilde{c}^{(0)} \partial_z \tilde{c}^{(0)}) - \gamma \partial_z (\dot{x}_I^{(0)} \tilde{f}^{(0)} \partial_z \tilde{c}^{(0)}) = -T (\tilde{\xi}^{(0)} - \xi^{(0),\pm}). \end{aligned} \tag{177}$$

As before we integrate this equation along the z coordinate and apply the matching conditions (105)–(113) to the various terms under the z derivative and obtain

$$\partial_t \psi_I^{(1)} + \llbracket (\psi \dot{m})^{(1)} \rrbracket + \llbracket (f \mu)^{(1)} \rrbracket = -T \xi_1^{(1)}. \tag{178}$$

Herein the free energy $\psi_1^{(1)}$ is defined as

$$\psi_1^{(1)} = \int_{-\infty}^0 \left(\tilde{F}^{(0)} - F^{(0),-} + \frac{\beta}{2} (\partial_z \tilde{u}^{(0)})^2 \right) dz + \int_0^{+\infty} \left(\tilde{F}^{(0)} - F^{(0),+} + \frac{\beta}{2} (\partial_z \tilde{u}^{(0)})^2 \right) dz \quad (179)$$

and the dissipation function $\xi_1^{(1)}$ has the representation

$$\xi_1^{(1)} = \int_{-\infty}^0 \tilde{\xi}^{(0)} - \xi^{(0),-} dz + \int_0^{+\infty} \tilde{\xi}^{(0)} - \xi^{(0),+} dz. \quad (180)$$

Next we show that the interfacial dissipation function is given by

$$T \xi_1^{(1)} = -\llbracket \mu_1^{(1)} \dot{m}_1^{(0)} + \mu_2^{(1)} \dot{m}_2^{(0)} \rrbracket. \quad (181)$$

We start from (167) and use the relations (120), (151)₂ and (119) to obtain after a straightforward calculation the identity

$$\begin{aligned} T \tilde{\xi}^{(0)} &= -\dot{m}_1^{(0)} \partial_z \tilde{\mu}_1^{(1)} - \dot{m}_2^{(0)} \partial_z \tilde{\mu}_2^{(1)} \\ &\quad + \beta \dot{x}_I^{(0)} \partial_z \left(\tilde{u}^{(0)} \partial_{zz} \tilde{u}^{(1)} + \tilde{u}^{(1)} \partial_{zz} \tilde{u}^{(0)} - \partial_z \tilde{u}^{(0)} \partial_z \tilde{u}^{(1)} \right) + \beta \dot{m}_1^{(0)} \partial_{zzz} \tilde{u}^{(1)} \\ &\quad + \gamma \dot{x}_I^{(0)} \dot{m}_1^{(0)} \partial_{zz} \tilde{u}^{(0)} + \gamma (\dot{x}_I^{(0)})^2 \partial_z (\tilde{u}^{(0)} \partial_z \tilde{u}^{(0)}) \end{aligned} \quad (182)$$

In an analogous manner we obtain for the bulk dissipation function (161) the identity

$$T \tilde{\xi}^{(0)} = -\dot{m}_1^{(0)} \partial_x \mu_1^{(0)} - \dot{m}_2^{(0)} \partial_x \mu_2^{(0)}. \quad (183)$$

Now we subtract from (182) the equation (183). After this we integrate along the inner coordinate z and apply again the matching conditions (105)–(113). Finally we end up with the proposition (181).

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