

Phase Boundaries in Alloys with Elastic Misfit

Harald Garcke

Abstract. We study a mathematical model describing phase transformations in alloys with kinetics driven by mass transport and stress. To describe the dynamics, a Cahn-Hilliard system taking elastic effects into account is studied. Existence and uniqueness results for the resulting singular elliptic-parabolic system are given.

In the Cahn-Hilliard model, phase boundaries are described by a diffuse interface with small positive thickness. In the stationary case we identify the sharp interface free boundary problem that arises when the interfacial thickness tends to zero. In particular, we obtain a geometric partition problem generalizing variants of isoperimetric problems to situations where elastic interactions cannot be neglected.

1. Introduction

Many problems involving free boundaries arise in the theory of phase transitions. In this context the interfaces separating two or more phases are the boundaries that have to be determined as part of the problem. We will consider phase transformations in alloys with kinetics driven by mass transport and stress. Different phases in such systems are characterized by certain distinct mixtures of the alloy components. For example if the alloy consists of just two types of atoms, A and B , and if two phases are possible, then one phase, which is called the α phase, has a higher concentration of A atoms than the other which is called the β phase. The interest now lies in a mathematical description of interfaces between different phases. We will first mathematically study a theory describing the evolution in such systems and then we will analyze stationary solutions that arise as absolute minimizer of an underlying energy.

Classical mathematical theories for phase transformations describe interfaces as hypersurfaces across which certain quantities, e.g. the mass density or certain components of the stress tensor, suffer jump discontinuities. These models lead to free boundary problems. The domains occupied by the individual phases are a priori unknown, i.e. the boundaries of the phase regions are free boundaries that have to be determined. In the interior of the phase domains, partial differential

equations have to hold and on the boundaries certain conditions, e.g. jump conditions ensuring mass or energy conservation, are prescribed. Through the boundary conditions the solutions in the individual phases are coupled. Models of this type are called sharp interface models and a classical example is the Stefan problem describing melting and solidification (see e.g. the book of Meirmanov [11]).

In another approach phase boundaries are modeled by transition layers with a small positive thickness. In these theories usually all quantities describing the physical system are continuous. Some of the quantities involved might have steep gradients in the interfacial layers. That are those quantities that would jump in a sharp interface model. Models of this type are called diffuse interface models and an example are the phase field models that are the diffuse interface counterpart of Stefan problems (see e.g. Caginalp [1]).

To study sharp interface models analytically or computationally one has to somehow resolve the hypersurfaces making up the free boundary. This can lead to serious difficulties when the phase boundaries develop singularities and/or undergo topological changes during the evolution. Diffuse interface models instead use quantities that are smooth in the whole domain and at least conceptionally they handle topological changes without difficulties.

Our goal in the first part of this contribution is to mathematically analyze a diffuse interface model describing the motion of interfaces in alloys in which stresses caused by an elastic misfit have a pronounced effect on the evolution (see e.g. Leo, Lowengrub and Jou [10] and Garcke, Rumpf and Weikard [6] for numerical simulations on the basis of such models). The evolution will be governed by a fourth order parabolic system resulting from mass balance laws for the alloy components coupled to an elliptic system describing mechanical equilibrium. In a second part we will show that the energies related to the diffuse interface model converge in the sense of Γ -limits to the energy related to the sharp interface model when the interfacial thickness tends to zero. Moreover, we can show that minimizers converge (along subsequences) and it is also possible to pass to the limit in the first variation formula. The results on this limit are the first rigorous results relating a diffuse interface model to a sharp interface model in the case of alloys with elastic misfit.

This contribution is a review of results obtained by the author in his habilitation thesis [5]. Readers who are interested in more details may contact the author directly for a copy of the thesis.

2. The Diffuse Interface Model

Let us consider an alloy that consists of N components. We denote by c_k ($k = 1, \dots, N$) the concentration of component k . Therefore the vector $\mathbf{c} = (c_k)_{k=1, \dots, N}$ has to fulfill the constraint $\sum_{k=1}^N c_k = 1$, i.e. \mathbf{c} lies in the affine hyperplane

$$\Sigma := \{\mathbf{c}' = (c'_k)_{k=1, \dots, N} \in \mathbb{R}^N \mid \sum_{k=1}^N c'_k = 1\}.$$

To describe elastic effects we define the displacement field $\mathbf{u}(\mathbf{x})$, i.e. a material point \mathbf{x} in the undeformed body will be at the point $\mathbf{x} + \mathbf{u}(\mathbf{x})$ after deformation. Since in phase separation processes the displacement gradient usually is small, we consider an approximative theory based on the linearized strain tensor

$$\mathcal{E}(\mathbf{u}) = \frac{1}{2} (\nabla \mathbf{u} + (\nabla \mathbf{u})^t).$$

A free energy of alloy systems taking elastic interactions into account is of the form

$$E(\mathbf{c}, \mathbf{u}) = \int_{\Omega} \left\{ \frac{\gamma}{2} |\nabla \mathbf{c}|^2 + \Psi(\mathbf{c}) + W(\mathbf{c}, \mathcal{E}(\mathbf{u})) \right\} \quad (1)$$

where $\Omega \subset \mathbb{R}^n$, $n \in \mathbb{N}$, is a bounded domain with Lipschitz boundary and $\gamma > 0$ is a small positive constant. The first term in the energy is the gradient part penalizing rapid spatial variations in the concentrations. The second summand Ψ is the chemical energy of the system and in the case of phase separation Ψ is a non-convex function of \mathbf{c} . The non-convexity in Ψ gives rise to the appearance of different phases. Roughly speaking one can say that local minima of Ψ correspond to different phases in the system. The last term in the free energy takes elastic effects into account. The dependence on \mathbf{c} reflects the fact that the stress free strain depends on the concentration. As the concentration is typically inhomogeneous in space, the stress free strain will vary in space.

To describe evolution phenomena, we consider mass diffusion for the individual components leading to diffusion equations for the concentrations. Mechanical equilibrium is attained on a much faster time scale than diffusion takes place. Therefore, we will assume a quasi-static equilibrium for \mathbf{u} , i.e. for all times

$$\nabla \cdot \mathcal{S} = 0,$$

where

$$\mathcal{S} = W_{,\mathcal{E}}(\mathbf{c}, \mathcal{E}(\mathbf{u}))$$

is the stress tensor. Here and below we will denote by $W_{,\mathcal{E}}$, $W_{,\mathbf{c}}$, $\Psi_{,\mathbf{c}}, \dots$ the derivative with respect to the variable appearing as a subscript. We point out that \mathbf{c} in general is time dependent, and hence the solution of the elastic system will also depend on time.

The diffusion equations for the concentrations c_k ($k = 1, \dots, N$) are based on mass balances for the individual components. To define the evolution equations for the concentrations we need to introduce the vector of generalized chemical potential differences \mathbf{w} which is given by the first variation of the energy E with respect to \mathbf{c} . Since values of \mathbf{c} are restricted to lie on the affine hyperplane Σ , we need to take this constraint into account when computing the first variation. One obtains

$$\mathbf{w} = \mathbf{P} (-\gamma \Delta \mathbf{c} + \Psi_{,\mathbf{c}}(\mathbf{c}) + W_{,\mathbf{c}}(\mathbf{c}, \mathcal{E}(\mathbf{u})))$$

where \mathbf{P} is the euclidian projection of \mathbb{R}^N onto

$$T\Sigma = \{\mathbf{d}' = (d'_k)_{k=1,\dots,N} \in \mathbb{R}^N \mid \sum_{k=1}^N d'_k = 0\}$$

which is the tangent space to Σ . Using the projection \mathbf{P} is one way of taking the Lagrange multiplier associated to the constraint $\sum_{k=1}^N c_k = 1$ into account. Since \mathbf{w} serves as a diffusional potential, the balance law for mass becomes $\partial_t \mathbf{c} = \Delta \mathbf{w}$. Altogether we obtain the system of equations

$$\partial_t \mathbf{c} = \Delta \mathbf{w}, \quad (2)$$

$$\mathbf{w} = \mathbf{P}(-\gamma \Delta \mathbf{c} + \Psi_{,\mathbf{c}}(\mathbf{c}) + W_{,\mathbf{c}}(\mathbf{c}, \mathcal{E}(\mathbf{u}))), \quad (3)$$

$$\nabla \cdot W_{,\mathcal{E}}(\mathbf{c}, \mathcal{E}(\mathbf{u})) = 0. \quad (4)$$

In the case of two components the equations (2)–(4) were first stated by Larché and Cahn [9] for $\gamma = 0$ and for nonzero γ by Onuki [12].

We will consider the system (2)–(4) together with initial conditions for \mathbf{c}

$$\mathbf{c}(\mathbf{x}, 0) = \mathbf{c}^0(\mathbf{x}) \quad (5)$$

and boundary conditions

$$\nabla \mathbf{w} \cdot \mathbf{n} = 0, \quad (6)$$

$$\nabla \mathbf{c} \cdot \mathbf{n} = 0, \quad (7)$$

$$\mathcal{S} \cdot \mathbf{n} = 0 \quad (8)$$

where \mathbf{n} is the outer unit normal to Ω .

Two simple properties of the above system are:

i) *mass conservation*: i.e., for all $t > 0$ it holds

$$\int_{\Omega} \mathbf{c}(\mathbf{x}, t) d\mathbf{x} = \int_{\Omega} \mathbf{c}^0(\mathbf{x}) d\mathbf{x},$$

and

ii) *decay of the free energy*: for all $t_2 > t_1 \geq 0$ it holds

$$E((\mathbf{c}, \mathbf{u})(t_2)) + \int_{t_1}^{t_2} |\nabla \mathbf{w}|^2 = E((\mathbf{c}, \mathbf{u})(t_1)).$$

3. Existence for the Diffuse Interface System

To keep the presentation relatively simple, we present the existence result for a specific case which is of physical interest and still contains all mathematical difficulties. A chemical energy Ψ derived from a mean-field theory is the sum of a logarithmic entropy term and a pairwise interaction term and has the form

$$\Psi(\mathbf{c}) = \sum_{k=1}^N c_k \ln c_k + \frac{1}{2} \mathbf{c} \cdot \mathbf{A} \mathbf{c}, \quad (9)$$

where the entries of the matrix $\mathbf{A} = (A_{kl})_{k,l=1,\dots,N}$ are the constant parameters that describe pairwise interactions between the components. A typical form for the free energy density is

$$W(\mathbf{c}, \mathcal{E}) = \frac{1}{2} (\mathcal{E} - \mathcal{E}^*(\mathbf{c})) : \mathcal{C}(\mathbf{c}) (\mathcal{E} - \mathcal{E}^*(\mathbf{c})) . \quad (10)$$

Here, $\mathcal{C}(\mathbf{c})$ is the concentration dependent elasticity tensor mapping symmetric tensors in $\mathbb{R}^{n \times n}$ into itself. We require $\mathcal{C}(\mathbf{c})$ to be symmetric, positive definite and bounded. The quantity $\mathcal{E}^*(\mathbf{c})$ is the symmetric stress free strain (or eigenstrain) at concentration \mathbf{c} . This is the value the strain tensor attains if the material were uniform with concentration \mathbf{c} and unstressed. For simplicity, we assume that \mathcal{E}^* grows at most linearly in \mathbf{c} . Finally, for matrices \mathbf{A} and \mathbf{B} we use the notation $A : B = \text{tr}(A^t B)$.

The main purpose of this section is to present a result on existence of weak solutions to the problem (2)–(8). Main difficulties in proving such a result are the facts that $\Psi_{,\mathbf{c}}$, a term entering the equation for the chemical potential differences (see (3)), is singular if c_k tends to zero and that $W_{,\mathbf{c}}$ grows quadratically in $\nabla \mathbf{u}$. We remark that the boundary condition (8) for the elasticity system only specifies the displacement \mathbf{u} up to infinitesimal rigid displacements (i.e. translations and infinitesimal rotations). We note that the strain $\mathcal{E}(\mathbf{u})$ is uniquely determined and hence in the evolution equation for \mathbf{c} the non-uniqueness of \mathbf{u} will not play any role. To fix the freedom for the displacement we introduce the space

$$X_2 := \{\mathbf{u} \in H^1(\Omega, \mathbb{R}^n) \mid (\mathbf{u}, \mathbf{v})_{H^1} = 0 \text{ for all } \mathbf{v} \in X_{\text{ird}}\} = X_{\text{ird}}^\perp,$$

where X_{ird} is the space of all infinitesimal rigid displacements (see e.g. [3]).

Let us state the definition of weak solutions we will use.

Definition 3.1. (Weak solution) *A triple*

$$(\mathbf{c}, \mathbf{w}, \mathbf{u}) \in L^2(0, T; H^1(\Omega, \mathbb{R}^N)) \times L^2(0, T; H^1(\Omega, \mathbb{R}^N)) \times L^2(0, T; X_2)$$

with $\mathbf{P}\Psi_{,\mathbf{c}}(\mathbf{c}) \in L^1(\Omega_T)$ is called a weak solution of (2)–(8) if and only if

(i)

$$-\int_{\Omega_T} \partial_t \boldsymbol{\xi} \cdot (\mathbf{c} - \mathbf{c}^0) + \int_{\Omega_T} \nabla \mathbf{w} : \nabla \boldsymbol{\xi} = 0 \quad (11)$$

for all $\boldsymbol{\xi} \in L^2(0, T; H^1(\Omega, \mathbb{R}^N))$ with $\partial_t \boldsymbol{\xi} \in L^2(\Omega_T)$ and $\boldsymbol{\xi}(T) = 0$,

(ii)

$$\int_{\Omega_T} \mathbf{w} \cdot \boldsymbol{\zeta} = \int_{\Omega_T} \{\nabla \mathbf{c} : \nabla \boldsymbol{\zeta} + \mathbf{P}\Psi_{,\mathbf{c}}(\mathbf{c}) \cdot \boldsymbol{\zeta} + \mathbf{P}W_{,\mathbf{c}}(\mathbf{c}, \mathcal{E}(\mathbf{u})) \cdot \boldsymbol{\zeta}\} \quad (12)$$

for all $\boldsymbol{\zeta} \in L^2(0, T; H^1(\Omega, \mathbb{R}^N)) \cap L^\infty(\Omega_T, \mathbb{R}^N)$, and

(iii)

$$\int_{\Omega_T} W_{,\mathcal{E}}(\mathbf{c}, \mathcal{E}(\mathbf{u})) : \nabla \boldsymbol{\eta} = 0 \quad (13)$$

for all $\boldsymbol{\eta} \in L^2(0, T; H^1(\Omega, \mathbb{R}^n))$.

In the following theorem the existence result is stated.

Theorem 3.2. *Let Ψ and W be of the form (9) and (10) respectively, with smooth \mathcal{C} and \mathcal{E}^* . In addition, assume that $\mathbf{c}^0 \in H^1(\Omega, \Sigma)$ fulfill $c_k^0 \geq 0$ and $\int_{\Omega} c_k^0 > 0$ for $k = 1, \dots, N$.*

Then there exists a weak solution of the elastic Cahn-Hilliard system which has the following properties:

- (i) $\mathbf{c} \in C^{0, \frac{1}{4}}([0, T]; L^2(\Omega))$,
- (ii) $\partial_t \mathbf{c} \in L^2\left(0, T; (H^1(\Omega))^*\right)$,
- (iii) *there exists a $p > 2$ such that $\mathbf{u} \in L^\infty(0, T; W^{1,p}(\Omega, \mathbb{R}^n))$,*
- (iv) *there exists a $q > 1$ such that for $k \in \{1, \dots, N\}$*

$$\ln c_k \in L^q(\Omega_T).$$

In particular, $c_k > 0$ almost everywhere.

We remark that the assumptions on \mathbf{c}^0 are physical. As \mathbf{c}^0 represents concentrations only non-negative values for c_k^0 are physically meaningful. Furthermore, if $\int_{\Omega} c_k^0 = 0$, the component k would not be present at all and the system could be reduced to $N - 1$ components.

Idea of the proof. The proof is rather technical and we only sketch the methods. A full proof can be found in [5]. First of all one has to regularize the logarithmic part of Ψ by approximating smooth functions. Then one obtains time discrete solutions to the regularized problems by the method of implicit time discretisation. To solve the time discrete problems, one can use that the system is the H^{-1} -gradient flow with respect to the variable \mathbf{c} . This observation allows it to use variational methods to show existence of time discrete solutions. The free energy decay property stated in section 2 has a discrete counterpart that gives first important a priori estimates. Anyhow, these are not enough to use compactness arguments to pass to the limit. To proceed, one has to show a higher integrability property for $\nabla \mathbf{u}$ (see (iii) in theorem 3.2). This is necessary since $\nabla \mathbf{u}$ appears quadratically on the right hand side in the equation for the chemical potential (see equation (3)). This can be achieved by using a perturbation technique introduced by Giaquinta and Modica (see [7]). Finally, one has to use that the non-smooth term appearing in Ψ is convex to obtain higher integrability of $\ln c_k$ (see (iv) in theorem 3.2). The properties (iii) and (iv) together with more or less standard compactness arguments are then enough to pass to the limit in the time discrete version of the regularized problem with both the time step and the regularizing parameter.

Remark 3.3. a) *A uniqueness result can be shown by an energy method in the case that \mathcal{C} does not depend on \mathbf{c} (see [5]).*
 b) *If $N = 2$, if \mathcal{C} does not depend on \mathbf{c} and if Ψ is assumed to have polynomial growth with a strictly positive leading term, an existence and uniqueness result was shown independently by Carrive, Miranville and Piétras [2]. However, the main difficulties in our study arise due to the facts that \mathcal{C}*

is concentration dependent and that the chemical energy Ψ contains non-smooth logarithmic terms and these cases are not covered by the analysis of [2].

4. The Sharp Interface Limit

In this section we study solutions of the variational problems.

(\mathbf{P}^ε) Find a minimizer $(\mathbf{c}, \mathbf{u}) \in X_1 \times X_2$ of

$$E^\varepsilon(\mathbf{c}, \mathbf{u}) := \int_{\Omega} \left(\varepsilon |\nabla \mathbf{c}|^2 + \frac{1}{\varepsilon} \Psi(\mathbf{c}) + W(\mathbf{c}, \mathcal{E}(\mathbf{u})) \right), \quad \varepsilon > 0,$$

subject to the constraint $\int_{\Omega} \mathbf{c} = \mathbf{m}$.

Here $\mathbf{m} \in \Sigma$ is a fixed constant. Under some natural assumptions it turns out that minimizers of the above variational problem are, roughly speaking, of the following form. In most of Ω the solution is close to values that minimize Ψ and the regions where the solution is close to minimizers of Ψ are separated by transition layers which are of a thickness proportional to ε . It is the goal of this section to study the limiting behaviour of E^ε and its minimizers as ε tends to zero. The scaling in ε (i.e. $\gamma \rightarrow \varepsilon^2$, $W \rightarrow \varepsilon W$, $E \rightarrow \frac{1}{\varepsilon} E$) is motivated by former studies for the case when no elastic contributions are present and by formally matched asymptotic expansions by Leo, Lowengrub and Jou [10].

For the rest of this section we assume that the chemical energy Ψ is such that

$$\Psi \geq 0 \quad \text{and} \quad \Psi(\mathbf{c}') = 0 \quad \Leftrightarrow \quad \mathbf{c}' \in \{\mathbf{p}_1, \dots, \mathbf{p}_M\}, \quad (14)$$

where $\mathbf{p}_1, \dots, \mathbf{p}_M \in \Sigma$ are mutually different and $M \geq 2$. Under the assumption (14) it is energetically favourable for \mathbf{c} to attain the values $\mathbf{p}_1, \dots, \mathbf{p}_M$. And in fact minimizers of E^ε in most of the domain Ω have values close to $\mathbf{p}_1, \dots, \mathbf{p}_M$. The values $\mathbf{p}_1, \dots, \mathbf{p}_M$ correspond to M different phases and if \mathbf{c} is close to a \mathbf{p}_k we say that \mathbf{c} is in phase k . It turns out that minimizers of E^ε converge (along subsequences) to minimizers of the functional

$$E^0: L^1(\Omega, \Sigma) \times X_2 \rightarrow \mathbb{R} \cup \{\infty\}$$

with

$$E^0(\mathbf{c}, \mathbf{u}) = \begin{cases} \sum_{\substack{k,l=1 \\ k < l}}^M \sigma_{kl} \mathcal{H}^{n-1}(\partial^* \{\mathbf{c} = \mathbf{p}_k\} \cap \partial^* \{\mathbf{c} = \mathbf{p}_l\}) + \int_{\Omega} W(\mathbf{c}, \mathcal{E}(\mathbf{u})) \\ \quad \text{if } \mathbf{c} \in BV(\Omega), \Psi(\mathbf{c}) = 0 \text{ a.e., } \int_{\Omega} \mathbf{c} = \mathbf{m}, \\ \infty \quad \text{otherwise.} \end{cases}$$

Here, $\{\mathbf{c} = \mathbf{p}_k\} := \{\mathbf{x} \in \Omega \mid \mathbf{c}(\mathbf{x}) = \mathbf{p}_k\}$, $BV(\Omega)$ is the space of functions with bounded variation and $\partial^* D$ is the reduced boundary of a set D (for more details on these concepts we refer to Giusti [8]). The real numbers σ_{kl} are the surface

tensions of the interfaces between phases k and l and they are defined via the chemical energy Ψ (see [5] for details).

Theorem 4.1. *Let W be of the form (10) with smooth \mathcal{C} and \mathcal{E}^* and assume that $\Psi \in C^1(\mathbb{R}^N, \mathbb{R})$ grows at least quadratically for large \mathbf{c} . Then the variational problems (\mathbf{P}^ε) possess minimizers $(\mathbf{c}^\varepsilon, \mathbf{u}^\varepsilon) \in H^1(\Omega, \Sigma) \times X_2$ provided that ε is small enough. Furthermore, there exists a sequence $\{\varepsilon^\kappa\}_{\kappa \in \mathbb{N}} \subset \mathbb{R}^+$ with $\lim_{\kappa \rightarrow \infty} \varepsilon^\kappa = 0$ and a $(\mathbf{c}, \mathbf{u}) \in L^2(\Omega, \Sigma) \times H^1(\Omega, \mathbb{R}^n)$ such that*

i)

$$\begin{aligned} \mathbf{c}^{\varepsilon^\kappa} &\rightarrow \mathbf{c} \quad \text{in } L^2(\Omega, \Sigma), \\ \mathbf{u}^{\varepsilon^\kappa} &\rightarrow \mathbf{u} \quad \text{in } H^1(\Omega, \mathbb{R}^n); \end{aligned}$$

ii) (\mathbf{c}, \mathbf{u}) is a global minimizer of E^0 .

Remark 4.2. i) *To obtain the convergence result above, one shows that the energy E^ε converges in the sense of Γ -limits to the energy E^0 . In a second step one has to show compactness for the sequence of minimizers $(\mathbf{c}^\varepsilon, \mathbf{u}^\varepsilon)_{\varepsilon > 0}$. With respect to the concentration \mathbf{c} one uses the L^2 -topology for the Γ -limit and for the compactness argument.*

ii) *In the binary case, i.e. $N = 2$, it is also possible to analyze the limiting behaviour of the Euler-Lagrange equation for minimizers of the elastic Ginzburg-Landau energies as the interfacial thickness tends to zero. In [5] it is shown that the Lagrange multiplier entering the Euler-Lagrange equations converge and in the sharp interface limit we obtain a weak formulation of the Gibbs-Thomson relation, which is one of the conditions that has to hold on the free boundary.*

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Institut für Angewandte Mathematik,
Universität Bonn,
Wegelerstraße 6,
D-53115 Bonn, Germany
E-mail address: harald@iam.uni-bonn.de