On the Cahn–Hilliard equation with non–constant mobility and its asymptotic limit

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Abstract

We present an existence result for the Cahn–Hilliard equation with a concentration dependent mobility which allows the mobility to degenerate. Formal asymptotic results relate the Cahn–Hilliard equation with a degenerate mobility to motion by surface diffusion $V = -\Delta_S \kappa$. We state a local existence result for this geometric motion and show that circles are asymptotically stable.

Keywords: Cahn–Hilliard equation, degenerate parabolic equations, nonlinear diffusion, geometric motions, surface diffusion, phase transitions.

AMS subject classification: 35K55, 35K65, 73K12, 70K99, 82C26

1 Introduction

In this paper we discuss a phenomenological model for isothermal phase separation in binary alloys which was introduced by Cahn and Hilliard [6, 7]. We assume that the alloy occupies an open bounded domain $\Omega \subset \mathbb{R}^n$ with smooth boundary. If c_1 and c_2 denote the local concentrations of the two components of the alloy we can introduce $u = c_1 - c_2$ as an order parameter for the system.

Then the law of mass conservation gives

$$u_t = -\nabla \cdot \mathbf{J}$$

where the vector $\mathbf{J}$ denotes the mass flux. For $\mathbf{J}$ we assume a generalized Fick's law

$$\mathbf{J} = -B(u)\nabla w$$

with a nonnegative mobility B which can depend on the concentration. Furthermore w is the difference of the chemical potentials of the two alloy components and is given as

$$w = -\gamma\Delta u + \Psi'(u)$$

where $\gamma > 0$ is a small parameter and Ψ is the homogeneous free energy. The chemical potential w can be seen as the functional derivative of the free energy

$$\mathcal{E}(u) = \int_\Omega \left(\frac{\gamma}{2} \mid \nabla u \mid^2 + \Psi(u)\right) dx\,.$$

For the homogeneous part of the free energy we choose for the moment

$$\Psi(u) = (u^2 - \beta^2)^2 \qquad\qquad \beta \in \mathbb{R}^2\,.$$

With this choice the free energy is of the typical Ginzburg–Landau form with the gradient term representing interfacial energies and the homogeneous free energy of a double well form, where the two minima of Ψ correspond to the two different phases of the system. Similar energies arise for example in the theory of van–der–Waals fluids and in models for shape memory alloys.

We supplement the system with the no–flux boundary condition $\mathbf{n} \cdot \mathbf{J} = 0$ which ensures mass conservation and the Neumann boundary condition $\mathbf{n} \cdot \nabla u = 0$ which is the natural boundary condition for the functional $\mathcal{E}$. By $\mathbf{n}$ we denote the outer normal to Ω.

The structure of this paper is as follows. In section 2 we review some well known results for the case of a constant mobility. In particular we discuss the stationary situation. Then we consider a mobility which can depend on the order parameter u and we allow mobilities which can degenerate. An existence theorem for the resulting fourth order degenerate parabolic equation is presented in section 3. The Cahn–Hilliard equation is related to certain sharp interface models. In section 4 we discuss motion by surface diffusion which arises as the asymptotic limit in the case of a concentration dependent mobility. We give a local existence result and show that circles are asymptotically stable. Some remarks on open questions and conjectures are given in section 5.

2 Constant mobility

In the case that the mobility is constant (we choose $B \equiv 1$) the Cahn–Hilliard equation becomes

$$u_t = \Delta(-\gamma \Delta u + \Psi'(u)). \tag{2.1}$$

First we want to discuss the stationary case which is related to the question, which are the possible states as $t \to \infty$. Using the no–flux boundary condition we can lift the outer Laplacian to get

$$-\gamma \Delta u + \Psi'(u) = const$$

Solutions to this equation are critical points of the energy functional

$$\mathcal{E}(u) = \int_\Omega \left(\frac{\gamma}{2} \mid \nabla u \mid^2 + \Psi(u)\right) dx\,.$$

if we allow variations subject to a mass constraint $\fint_\Omega u = u_m$. Because we are interested in critical points which are limits as $t \to \infty$ of the Cahn–Hilliard equation the mass u_m is given by the mass of the initial values.

First of all let us investigate absolute minimizers of the functional $\mathcal{E}$ subject to the mass constraint. In the case $\gamma = 0$ all minimizers are given by the two valued functions

$$u(x) = \begin{cases} \beta & x \in \Omega_+ \, , \\ -\beta & x \in \Omega_- \end{cases} \tag{2.2}$$

where $\Omega_+ \cup \Omega_- = \Omega$ and $\beta|\Omega_+| - \beta|\Omega_-| = u_m|\Omega|$. Here we assumed $|u_m| \le \beta$. We get no restrictions on the shape of the interface and in particular interface energy is neglected. To include interfacial energy Cahn and Hilliard introduced the higher order term $\frac{\gamma}{2}|\nabla u|^2$ in the free energy. This term penalises gradients and therefore minimizers of $\mathcal{E}$ with $\gamma > 0$ have to be more regular and try to minimize interfacial regions.

There is a fundamental result by Modica [17] who proved that minimizers of the functional $\mathcal{E}$ converge as γ tends to zero (in the sense of subsequences) to a two valued function of the form (2.2) where the interface between Ω_+ and Ω_- has minimal area. This shows that the term $\frac{\gamma}{2}|\nabla u|^2$ models interface energy. In general there exist other critical points. In one space dimension Zheng Songmu [20] showed that if $|u_m| < \beta$ there exist $2N + 1$ $(N \in \mathbb{N})$ critical points, where N is related to γ and tends to infinity as γ tends to 0. In higher space dimensions the full characterization of critical points is still an open problem.

We now return to the evolutionary problem. Well posedness results such as existence and uniqueness of solutions are well established (see [14]). Furthermore in one space dimension solutions tend to the critical points as $t \to \infty$ (see [20]).

There have also been a number of numerical studies for the Cahn–Hilliard equation, which support the fact that the Cahn–Hilliard equation models phase separation. In particular they show that at early stages the solution rapidly tends to the values $\pm\beta$ and forms thin transition layers between the different phases. In material science this is known as spinodal decomposition – the solution tries to avoid the unstable concave part (the spinodal interval) of the energy. After this first stage of the evolution the solution has a fine grained separated structure and therefore the interfacial part of the energy is still large. In a second much slower part of the evolution some grains grow and other shrink in order to reduce the interfacial regions under the implicit constraint of mass conservation. This latter stage of the evolution is known as coarsening. The generic case is that the solution converges for t tending to infinity to a limit which consists of two regions (a $\{u \cong \beta\}$ and a $\{u \cong -\beta\}$ region) separated by a thin interfacial region. For numerical results and a review on results for the Cahn–Hilliard equation with constant mobility we refer to Elliott [10].

3 Non–constant mobility

The mathematical literature has so far mainly considered the case of a constant mobility. But in applications the mobility usually depends on the concentration. In the interfacial region (i.e. $|u| \cong 0$) the diffusion is much larger than in the pure components (i.e. $u \cong \pm 1$). Typically a mobility of the form

$$B(u) := \begin{cases} 1-u^2 & \text{for} \quad |u| < 1\,, \\ 0 & \text{for} \quad |u| \geq 1 \end{cases} \tag{3.3}$$

is chosen (see Hilliard [15], Langer, Bar–On and Miller [16] and deGennes [9]).
Let us now discuss some difficulties which arise in the mathematical study of the Cahn–Hilliard equation with a concentration dependent mobility of the above form.

1. Difficulty: The Cahn–Hilliard equation becomes a fourth order degenerate parabolic equation. Therefore it has similar difficulties as other degenerate parabolic equations such as the porous medium equation

$$u_t - \nabla \cdot (|u|^m \nabla u) = 0 \tag{3.4}$$

and its fourth order analogue

$$u_t + \nabla \cdot (|u|^m \nabla \Delta u) = 0\,. \tag{3.5}$$

The equation (3.5) was introduced to model the motion of viscous droplets spreading over a solid surface. Bernis and Friedman [2] studied this equation in one space dimension and proved existence of a positive weak solution and results on the behaviour of the support of the solution.
2. Difficulty: Because the equation is a fourth order parabolic equation there is no maximum principle valid. Therefore many techniques which were developed for porous medium type of equations cannot be applied.
3. Difficulty: We want to develop an existence theory which allows the homogeneous free energy to become singular when $|u| \to 1$. This is due to the fact that the homogeneous free energy in applications usually has a logarithmic form. An existence theory should include the following three cases:
i) The mean field potential

$$\Psi(u) = \frac{\theta}{2}\left((1+u)\ln(1+u) + (1-u)\ln(1-u)\right) + \Psi^2(u) \tag{3.6}$$

where Ψ^2 is a smooth function and θ is a positive temperature.
ii) The double obstacle potential

$$\Psi(u) = \begin{cases} 1-u^2 & \text{if } |u| \leq 1\,, \\ \infty & \text{otherwise.} \end{cases}$$

which is the deep quench limit $\theta \searrow 0$ of i) if one chooses $\Psi^2(u) = 1-u^2$ (see Blowey and Elliott [4]).
iii) The smooth double well potential

$$\Psi(u) = (u^2-\beta^2)^2 \quad \beta \in \mathbb{R}^+$$

which we introduced before.

Now we formulate general assumptions under which we can prove the existence of a weak solution. We define

$$\Psi(u) := \Psi^1(u) + \Psi^2(u)$$

with a smooth function Ψ^2 and a convex function $\Psi^1 : (-1,1) \to \mathbb{R}$ such that $(\Psi^1)''(u) = (1-u^2)^{-1}F(u)$ where $F \in C^1([-1,1], \mathbb{R}_0^+)$. This allows in particular the logarithmic free energy i). The mobility is chosen to be of the form (3.3). Under these assumptions the following theorem holds.

Theorem 1 (Elliott and Garcke): *Let either $\partial\Omega \in C^{1,1}$ or Ω convex and suppose $\mathcal{E}(u_0)$ is bounded and $|u_0| \leq 1$ a.e. Then there exists a pair $(u, \mathbf{J})$ such that*
a) $u \in L^2(0,T; H^2(\Omega)) \cap L^\infty(0,T; H^1(\Omega)) \cap C([0,T]; L^2(\Omega))$,
b) $u_t \in L^2(0,T; (H^1(\Omega))')$,
c) $u(0) = u_0$ *and* $\nabla u \cdot \mathbf{n} = 0$,
d) $|u| \leq 1$ *a.e in* $\Omega_T := \Omega \times (0,T)$,
e) $\mathbf{J} \in L^2(\Omega_T, \mathbb{R}^n)$,
which satisfies $u_t = -\nabla \cdot \mathbf{J}$ in $L^2(0,T; (H^1(\Omega))')$ and

$$\mathbf{J} = -B(u)\nabla\left(-\gamma\Delta u + \Psi'(u)\right)$$

in the following weak sense

$$\int_{\Omega_T} \mathbf{J} \cdot \boldsymbol{\eta} = -\int_{\Omega_T} \left[\gamma\Delta u \nabla \cdot (B(u)\boldsymbol{\eta}) + (B\Psi'')(u)\nabla u \cdot \boldsymbol{\eta}\right]$$

for all $\boldsymbol{\eta} \in L^2(0,T; H^1(\Omega, \mathbb{R}^n)) \cap L^\infty(\Omega_T, \mathbb{R}^n)$ which fulfill $\boldsymbol{\eta} \cdot \mathbf{n} = 0$ on $\partial\Omega \times (0,T)$.

The proof of this theorem is based on the following two energy estimates. We point out that the following calculations to derive the energy estimates are of formal nature, but they can be made precise for a regularized problem (see [11]). First we differentiate the free energy functional to get

$$\begin{aligned}
\frac{d}{dt}\mathcal{E}(u(t)) &= \frac{d}{dt}\int_\Omega \left(\frac{\gamma}{2}|\nabla u|^2 + \Psi(u)\right) \\
&= \int_\Omega \left(\gamma\nabla u \nabla u_t + \Psi'(u)u_t\right) \\
&= \int_\Omega \left(-\gamma\Delta u + \Psi'(u)\right) u_t \\
&= \int_\Omega w\nabla \cdot (B(u)\nabla w) = -\int_\Omega B(u)|\nabla w|^2 .
\end{aligned}$$

This gives $\mathcal{E}(u(t)) + \int_{\Omega_t} B(u)|\nabla w|^2 = \mathcal{E}(u_0)$. To derive a second estimate we introduce the function $\Phi(u) = (1+u)\ln(1+u) - (1-u)\ln(1-u)$ which is the logarithmic part of the mean field potential. We note that $\Phi''(u) = \frac{1}{1-u^2}$. Using this fact we can establish a second estimate

$$
\begin{aligned}
\frac{d}{dt}\int_\Omega \Phi(u) &= \int_\Omega \Phi'(u)u_t = \int_\Omega \Phi'(u)\nabla\cdot B(u)\nabla\left(-\gamma\Delta u + \Psi'(u)\right) \\
&= -\int_\Omega \Phi''(u)(\nabla u)B(u)\nabla\left(-\gamma\Delta u + \Psi'(u)\right) \\
&= -\int_\Omega \gamma|\Delta u|^2 - \int_\Omega \Psi_1''(u)|\nabla u|^2 - \int_\Omega \Psi_2''(u)|\nabla u|^2 .
\end{aligned}
$$

Since Ψ_2'' is bounded we can use the estimate on the gradient of u to control the last term on the right hand side. Therefore we get

$$
\int_\Omega \Phi(u(t)) + \int_{\Omega_t}\left(\gamma|\Delta u|^2 + \Psi_1''(u)|\nabla u|^2\right) \le \int_\Omega \Phi(u_0) + Ct .
$$

The idea of the proof is now to replace the degenerate mobility B by positive mobilities B_ε and the singular part Ψ_1 of Ψ by smooth Ψ_ε^1 such that B_ε and Ψ_ε^1 converge to B and Ψ^1. This modified problem has a solution for which we can show energy estimates similar to the two above. Compactness results give the existence of a converging subsequence and we can show that the limit solves the degenerate Cahn–Hilliard equation in the sense stated in Theorem 1. We want to stress that we can show that $|u|$ remains less than or equal to one without having a maximum principle.

For a complete proof of a more general version of this theorem see [11].

4 Sharp interface models as asymptotic limits

As in the stationary case one observes that the parameter γ is related to the thickness of the interface between different phases and that the thickness tends to zero as γ tends to zero. Therefore one expects to recover a sharp interface model in the limit $\gamma \searrow 0$. That means we expect to get an evolutionary problem for hypersurfaces Γ_t (t is the time parameter) which is the limit of the zero level sets of the solutions to the Cahn–Hilliard equation.

The first result in this direction for the case of constant mobility has been established by Pego [18] who used formal asymptotic results to show that Γ_t evolves according to the law

$$
\begin{aligned}
\Delta w &= 0 && \text{for } x\in\Omega\setminus\Gamma_t ,\\
w &= \kappa && \text{for } x\in\Gamma_t ,\\
V &= [\mathbf{n}\cdot\nabla w]_-^+ && \text{for } x\in\Gamma_t \\
\text{and}\quad \nabla w\cdot\mathbf{n} &= 0 && \text{on } \partial\Omega
\end{aligned}
$$

where $\mathbf{n}$ denotes the normal on Γ_t (Ω respectively), κ is the mean curvature, V is the velocity of Γ_t in normal direction and $[\,.\,]_-^+$ denotes the jump across Γ_t. To determine the evolution one has to solve Laplaces equation in the bulk with Neumann boundary condition on $\partial\Omega$ and a Dirichlet condition on Γ_t. Unless Γ_t has constant mean curvature ∇w will suffer from a jump discontinuity along Γ_t which is the driving force of the evolution. This evolutionary problem is known as the Mullins–Sekerka problem and can be seen as a quasistatic version of the Stefan–problem with a Gibbs-Thomson law on the free boundary. For rigorous results on the convergence of solutions of the Cahn–Hilliard equation to solutions of the the Mullins–Sekerka problem see the work of Stoth [19] and Alikakos, Bates and Chen [1]. All results hold after a rescaling of time ($t \longrightarrow \sqrt{\gamma}t$). This shows again that the second stage in the evolution of the Cahn–Hilliard equation is on a slow time scale.

If the mobility is $B(u) = 1 - u^2$, Cahn, Elliott and Novick–Cohen [5] used formal asymptotic techniques to show that in the limit as γ tends to zero, Γ_t evolves according to the law

$$V = -\Delta_S \kappa \tag{4.7}$$

where Δ_S is the surface Laplacian. Their result holds for the deep quench limit and after a rescaling of time ($t \longrightarrow \gamma t$). Therefore the evolution of the Cahn–Hilliard equation with the mobility $B(u) = 1 - u^2$ is on an even slower time scale.

The geometric evolution (4.7) is known as motion by surface diffusion and in contrast to the Mullins–Sekerka problem is a purely local evolution, i.e. the normal velocity is determined by local quantities. Because the mobility in the Cahn–Hilliard equation with degenerate mobility was zero in the pure phase, we do not get any bulk diffusion in the limit.

Let us now derive some simple properties of motion by surface diffusion. If one starts with a compact initial surface Γ_0 which is the boundary of an open set $\Omega(0)$ we get

$$\frac{d}{dt} Volume(\Omega(t)) = \int_{\Gamma_t} V = \int_{\Gamma_t} -\Delta_S \kappa = 0$$

where $\Omega(t)$ is the set enclosed by Γ_t. Therefore motion by surface diffsion preserves volume. For the perimeter of $\Omega(t)$ we can derive

$$\frac{d}{dt} Per(\Omega(t)) = -\int_{\Gamma_t} \kappa V = -\int_{\Gamma_t} \kappa(-\Delta_S \kappa) = -\int_{\Gamma_t} |\nabla_S \kappa|^2 \leq 0$$

which means that the perimeter decreases.

Now we want to present some results for the two dimensional case, i.e. for the evolution of curves in the plane. We have to consider $V = -\partial_{ss}\kappa$ where ∂_{ss} is the second derivative with respect to arc–length. First we state a local existence result.

Theorem 2 (Elliott and Garcke): *Assume Γ_0 is a simple connected closed curve and $\int_{\Gamma_0} (\kappa_s^0)^2$ is bounded. Then there exists a time $T > 0$ such that the motion $V = -\partial_{ss}\kappa$ possesses a strong solution on the time interval $[0, T]$ and*

$$\text{ess sup}_{0<t<T} \int_{\Gamma_t} (\kappa_s)^2 + \int_0^T \int_{\Gamma_t} V_s^2 \leq C$$

is satisfied.

This theorem is proved by writing the evolution equation as a graph over a fixed curve. The resulting equation is a nonlinear fourth order parabolic equation and can be solved via linearzation and application of Schauders fixed point theorem (see [12] for a proof).

In general we do not expect that a simple connected curve remains simple, i.e. we expect self intersections to occur. But in the case that the initial curve is close to a circle we can prove that no self intersections occur and that a global solution exists. This fact is formulated in the following theorem (see [12] for a proof).

Theorem 3 (Elliott and Garcke): *There exists a $\delta > 0$ such that :*
If the initial curve Γ_0 is given as

$$\Gamma_0 = \{d^0(\theta)(\cos\theta, \sin\theta) \mid \theta \in S^1\}$$

and satisfies

$$\|d^0(.) - 1\|_{C^1(S^1)} \leq \delta \quad \textit{and} \quad \int_{\Gamma_0} (\kappa_s^0)^2 \leq \delta$$

then the evolution problem $v = -\kappa_{ss}$ has a global strong solution with initial curve Γ_0.

Our last theorem states that solutions which exist globally in time converge to a circle. The proof of this theorem is based on energy estimates and the fact that the length decreases during the evolution (see [12]).

Theorem 4 (Elliott and Garcke): *Assume a global simple connected solution Γ of $V = -\kappa_{ss}$ exists. Then there exists a time T_0 such that Γ_t is given by a periodic function $R(.,t) : [0, 2\pi) \longrightarrow \mathbb{R}^+$ in the following way*

$$\Gamma_t = \{(x_0(t), y_0(t)) + R(\theta, t)(\cos\theta, \sin\theta) \mid \theta \in [0, 2\pi)\}$$

for all $t > T_0$. Moreover
1.) $Length(\Gamma_t) \searrow L_\infty$
and 2.) $\|R(.,t) - R_\infty\|_{L^\infty} \longrightarrow 0$
where L_∞ (R_∞) is the length (radius) of a sphere which encloses a ball with the same area as $\Omega(0)$.

The last two theorems show that circles are asymptotically stable.

5 Open questions

There remain many open questions. The most important is whether there exists a unique solution to the Cahn–Hilliard equation with degenerate mobility or not. It is known that the initial value problem for the equation $u_t = -(|u|^m u_{xxx})_x$ can have more than one solution (see Beretta, Bertsch and Dal Passo [3]). But this nonuniqueness result is for a weaker notion of solutions than ours. For our concept of solution, which is introduced in Theorem 1, we conjecture that only one solution exists.

It also would be interesting to know more about the qualitative behaviour of solutions. The dynamics will strongly depend on the choice of the homogeneous free energy. If we choose the logarithmic free energy (3.6) the minima of Ψ are strictly less than one and we expect that the set $\{|u| = 1\}$ is empty or has at least measure zero after a certain time. In the case of a constant mobility Elliott and Luckhaus [13] showed that the set $\{|u| = 1\}$ has zero measure for all $t > 0$ even if $\{|u_0| = 1\}$ has positive measure.

If we choose the double obstacle free energy, the minima of Ψ are ± 1. As in the case of constant mobility we expect that the sets $\{u = 1\}$ and $\{u = -1\}$ develop an interior and that one gets a free boundary problem for $\{u = 1\}$ and $\{u = -1\}$. Moreover there are no results known for the asymptotic behaviour as t tends to infinity.

Another important class of open questions concerns the regularity of solutions. First of all we would like to answer the questions whether the solution is continuous or not. Since the equation is of fourth order, techniques based on Moser or deGiorgi iteration techniques seem not to be applicable directly.

A rigorous result concerning the convergence of solutions of the Cahn–Hilliard equation with a degenerate mobility to solutions of motion by surface diffusion is still missing. Also there are no rigorous results on motion by surface diffusion in higher space dimensions, but numerical computations and formal analysis by Coleman, Falk and Moakher [8] show that cylinders are unstable.

Finally we want to show that self intersections of curves evolving to motion by surface diffusion are possible. If we do not understand Γ_t as a phase boundary we can continue the evolution after self intersections have occured. The question arising then is whether the nonlinear fourth order parabolic equation which governs the evolution has a global solution or if finite time singularities occur such that a solution cannot be continued.

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