Crossover of the coarsening rates in demixing of binary viscous liquids

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Abstract

We consider a model for phase separation in binary viscous liquids that allows for material transport due to cross-diffusion of unlike particles and convection by the hydrodynamic bulk flow. Typically, during the evolution, the average size of domains of the pure phases increases with time — a phenomenon called coarsening. Siggia [24] predicts that at an initial stage, coarsening proceeds mainly by diffusion, which leads to the well-known evaporation-recondensation growth law $\ell \sim t^{1/3}$, when ℓ denotes the average domains size and t denotes time. Furthermore, he argued that at a later stage, convection by the bulk flow becomes the dominant transport mechanism, leading to a crossover in the coarsening rates to $\ell \sim t$. Siggia's predictions have been confirmed by experiments and numerical simulations.

In this work, we prove the crossover in the coarsening rates in terms of time-averaged lower bounds on the energy, which scales like an inverse length. We use a method proposed by Kohn and the first author [15], which exploits the gradient flow structure of the dynamics. Our adaption uses techniques from optimal transportation. Our main ingredient is a dissipation inequality. It measures how the optimal transportation distance changes under the effects of convective and diffusive transport.

1   Introduction

We investigate the coarsening rates in the demixing process of binary viscous liquids. Demixing in liquid mixtures typically arises when the mixture is put into a thermodynamically unstable state far from an equilibrium, for instance, after a quench from a high to a sufficiently low temperature. Thermodynamics favors the separation of the two phases. This drives the formation of microstructure: Initially, the mixture

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demixes locally by forming intertwined domains of the two pure phases. The free energy concentrates along the interface between these domains. In the subsequent evolution, the system tends to equilibrium by reducing the interfacial area. During this coarsening process the length scales describing the configuration (such as the the average domain width) grow.

In binary viscous liquids, there are two parallel transport mechanisms. Material is transported by cross-diffusion, the relative motion of the two different species through the bulk, and by convection, the material transport by the bulk flow. It turns out that each transport mechanism becomes dominant during a certain time interval in the demixing process. Initially, demixing is mediated mainly by diffusion. Later on, as the domains become large enough, viscous forces in the liquid become effective and the flow transport becomes the more efficient mechanism.

We are interested in the rate at which the coarsening of the domain morphology proceeds. Generically, the system develops a typical length scale ℓ that describes the typical width of the domains of the pure phases. Therefore, the coarsening rate is measured by the growth rate of ℓ. In 1979, in a seminal paper [24], Siggia argued that during the first, diffusion-dominated regime, the coarsening rate behaves like $\ell \sim t^{1/3}$ — the well-known coarsening law from the evaporation-recondensation process (Ostwald ripening) studied by Lifshitz, Slyozov, and Wagner [17, 31]. Siggia predicted that at a later stage, the dominance of convection leads to a crossover of the coarsening rate to $\ell \sim t$. Siggia's coarsening rates have been confirmed in numerical simulations [2, 14, 22, 27] and physical experiments [6, 32, 33, 16]; see also the discussion in Section 3.

In this paper, we rigorously establish Siggia's coarsening rates in form of weak upper bounds: We show that coarsening cannot proceed faster than $\ell \lesssim t^{1/3}$ for diffusion-mediated and subsequently $\ell \lesssim t$ for convection-mediated transport. The coarsening rates come in form of time-averaged lower bounds on the energy density — for a discussion on the energy scaling, see page 9.

Our mathematical investigation of the crossover of the coarsening rates fuses together the individual results on coarsening rates for purely diffusive transport [15] and for purely convective transport [4]. We apply the general method introduced in [15]. There are two questions that arise. One concerns the technique:

> *Can the method of [15] be adapted to capture the crossover of coarsening rates which reflect two different transport regimes?*

The second issue is more fundamentally related to the nature of the system:

> *Can the bulk transport mechanism sufficiently "help" the diffusive transport during the stage when the later is dominant, for the coarsening rate to be affected (and vice versa)?*

Below we answer the first question affirmatively and the second negatively.

Recently, Dai, Niethammer, and Pego [8] obtained rigorous results on the crossover of coarsening rates from attachment-limited to diffusion-limited dynamics as it occurs in rapid solidification of melts. They too used the framework of [15]. In addition to being a different system and a mean-field-type approximation (we work with a phase-field model), a significant difference from our work here is that in [8], the system features two limiting dissipation mechanisms rather than two transport mechanisms. The authors prove the crossover from $\ell \sim t^{1/2}$ in the regime of attachment dynamics to $\ell \sim t^{1/3}$ in the regime of diffusion dynamics. The presence of two limiting dissipation mechanisms slows down the coarsening process, $\ell \sim \min\{t^{1/2}, t^{1/3}\}$, while the presence of two transport mechanisms speeds it up, $\ell \sim \max\{t^{1/3}, t\}$.

The paper is organized as follows: In Section 2 we introduce the models, both in the diffuse-interface and the sharp-interface formulation. In Section 3 we present a heuristic argument for the rates of coarsening and determine at what length scale should the crossover occur. In Section 4 we state the main result. We present the technique used and state the propositions that combine to establish the main result. In Section 5 we present several results on how the optimal transportation distance changes under the effects of (convective and diffusive) transport. They are needed to establish the propositions of Section 4, but are of independent interest as well. In Section 6 we present the detailed proofs of the statements of Section 4. Finally in Appendix A, we prove an optimal-transportation result we need. Appendix B contains a discussion on well-posedness and regularity (in three space dimensions) of the system of PDE describing the demixing process.

2   The models

We present our results for two levels of modeling the system: for the diffuse-interface model and for the sharp-interface model. In the *diffuse-interface model*, the dynamics are described in terms of an order parameter (or phase field), which measures the local composition of the mixture. The order parameter varies smoothly between the pure phases, as it is expected in diffusive systems. The *sharp-interface model* depicts the thermodynamics on a mesoscopic level. As the name suggests, the diffuse interface is replaced by a sharp borderline between the domains of the pure phases, where the order parameter is supposed to be constant. This approximation is justifiable in many systems if the typical domain size is much larger than the interfacial width; see for example [21, 3, 5, 1]. While for the diffuse-interface model the questions of well-posedness and regularity can be answered by standard techniques (cf. Appendix B), the well-posedness theory for the sharp-interface model remains open. Nevertheless, since the sharp-interface system provides a cleaner geometric description of the dynamics, we present the results on the coarsening of that system as well. To do that, we assume that smooth enough solutions to the sharp-interface system do exist.

We consider the problem in a periodic setting. Let $\Omega \subset \mathbb{R}^d$, $d \geq 2$, be the cell of

periodicity of all functions considered. We assume that

$$|\Omega| \gg 1.$$

Since we are always working with averages, the system size, i.e., the volume of Ω, will not enter in our analysis. For any measurable set $A \subset \Omega$, we write $\fint_A = \frac{1}{|\Omega|} \int_A$ for convenience.

The mathematical quantity that models the physical configuration is the order parameter m, which describes the local composition of the mixture. It takes the values ± 1 in the stable phases. For symmetry reasons, m takes value zero where both phases are mixed in equal measure. Since we exclude chemical reactions the total mass of each phase is conserved during the evolution. We consider the critical mixture, where both phases occupy the same volume fraction so that

$$\overline{m} := \fint_\Omega m \, dx = 0.$$

The case $\overline{m} \neq 0$ can be treated analogously.

Below we introduce the models. For a more detailed introduction of the models as well as an explanation of their gradient-flow structure, we refer to [4].

2.1 Diffuse-interface model

We begin with the introduction of the model for diffuse interfaces. The free energy, averaged over cell Ω is given by the Ginzburg–Landau functional with the classical double-well potential

$$E(m) := \fint_\Omega \frac{1}{2}|\nabla m|^2 + \frac{1}{2}(1 - m^2)^2 \, dx. \tag{1}$$

It favors the values $m = 1$ and $m = -1$, representing the pure phases, and penalizes transitions between these values. It is nondimensionalized in such a way, that the typical transition layer between the domains of the pure phases is of order one, and thus much smaller than the system size. The L^2 derivative of the energy is the chemical potential

$$\mu := \frac{\partial E}{\partial m} = -\Delta m - 2(1 - m^2)m. \tag{2}$$

Since the order parameter must be conserved, the evolution equation for m comes in form of a local conservation law, $\partial_t m + \nabla \cdot J = 0$. Due to the two parallel transport mechanisms, the transporting flux combines diffusion and convection, $J = \lambda J_{\mathrm{diff}} + J_{\mathrm{conv}}$, where λ measures the relative strength of the diffusion flux compared to the convection flux. The flux due to cross-diffusion is given by $J_{\mathrm{diff}} = -\nabla \mu$, and the flux due to fluid convection is $J_{\mathrm{conv}} = mu$, where u denotes the fluid velocity. It obeys a Stokes equation:

$$\begin{aligned}
-\Delta u + \nabla p &= -m\nabla \mu, \\
\nabla \cdot u &= 0,
\end{aligned} \tag{3}$$

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in which p is the hydrodynamic pressure. The forcing term which acts on u is determined by the "principle of virtual work": It guarantees that the dissipation due to convection is given by the Stokes friction term (cf. (5)). Summing up, the demixing process is described by the equation

$$\partial_t m - \lambda \Delta \mu + \nabla \cdot (mu) = 0, \tag{4}$$

where μ is given by (2) and u solves the Stokes equation (3). This model is nondimensionalized and permits — besides the size of the system — *one dimensionless parameter*: λ. Well-posedness and regularity (for $d = 3$) of this dynamical system is established in the Appendix B.

Direct calculation shows that the energy dissipation rate is

$$\frac{dE}{dt} = -\int_\Omega \lambda |\nabla \mu|^2 + |Du|^2 \, dx. \tag{5}$$

2.2 Sharp-interface model

On the mesoscopic level, m takes only the values which characterize the pure phases, i.e., $m = \pm 1$. The energy concentrates on the interface and is proportional to its area (*length* in two space dimensions). A straightforward computation of the energy of the one-dimensional interfacial profile determines the prefactor $\frac{4}{3}$. We thus define

$$E(m) := \frac{4}{3} \times \frac{1}{2} \int_\Omega |\nabla m| \, dx = \frac{2}{3} \int_\Omega |\nabla m| \, dx. \tag{6}$$

Let Ω_+ be the subset of Ω where m takes value 1, and Ω_- the subset where $m = -1$. Let $\Gamma = \partial \Omega_+$. We denote the mean curvature of Γ by H, with the convention, that H is nonnegative along convex boundaries $\partial \Omega_+$.

The chemical potential μ is given by

$$\begin{aligned} -\Delta \mu &= 0 \qquad \text{in } \Omega \backslash \Gamma, \\ \mu &= \frac{2}{3} H \quad \text{on } \Gamma. \end{aligned} \tag{7}$$

The fluid velocity u solves the following Stokes equation in which $S = (Du + (Du)^T) - pI$ is the stress tensor, p is the hydrodynamic pressure, τ is any tangent vector to Γ, and ν is the outside (relative to Ω_+) unit normal vector to Γ:

$$\begin{aligned} -\nabla \cdot S &= 0 \qquad \text{in } \Omega \backslash \Gamma, \\ \nabla \cdot u &= 0 \qquad \text{in } \Omega, \\ \tau[S\nu] &= 0 \qquad \text{on } \Gamma, \\ \nu[S\nu] &= -\frac{4}{3} H \quad \text{on } \Gamma. \end{aligned} \tag{8}$$

Above $[A]$ denotes the jump in quantity A across Γ.

Again, the order parameter m is transported by the total flux. That is, the interface Γ_t moves with normal velocity

$$V = -\frac{1}{2}\left[\frac{\partial \mu}{\partial \nu}\right] + u \cdot \nu. \tag{9}$$

While the second term in the velocity is just the transport by the bulk flow, the first term is well-known from the Mullins–Sekerka law. The validity of the evolution (7)–(9) as a sharp-interface approximation of (3)&(4) is discussed in a generalized setting (the authors consider the full Navier–Stokes equation instead of the stationary Stokes equation) in [1, Appendix A].

Notice that this sharp interface version of (3)&(4) is — besides the system size — free of dimensionless parameters. This comes from the fact that there is no length scale that corresponds to interfacial width. Thus, in any statements in which λ appears and that apply to sharp interfaces, one should set $\lambda = 1$.

Assuming that the sharp interface system has solutions that are smooth enough, the energy dissipation can be written as:

$$\frac{dE}{dt} = \frac{4}{3}\int_{\Gamma} HV d\mathcal{H}^{d-1} = -\int_{\Omega} |\nabla \mu|^2 + |Du|^2 \, dx.$$

Finally, a direct formal computation shows that the evolution preserves the volumes of Ω_+ and Ω_-.

3 Heuristics

In this section, we present a simple heuristic argument in favor of the two coarsening rates. A fairly neutral starting point for demixing studies is an almost uniform state corresponding to full mixing, say $m \approx 0$. This configuration is unstable and linear analysis and numerical simulations (cf. [4]) indicate that a wavelength of order one grows the fastest. A natural assumption on the typical domain size ℓ at the onset of coarsening is therefore

$$\ell \gtrsim 1. \tag{10}$$

It is expected, as such behavior is ubiquitous in related energy-driven systems, that in large systems, the evolution of the characteristic length scale obeys a power law:

$$\ell(t) \sim t^{\gamma}.$$

The coarsening exponent γ is determined by the dominant material transport mechanism. We argue that $\gamma = 1/3$ for diffusion-mediated and $\gamma = 1$ for convection-mediated transport.

The heuristics are based on the assumption that the coarsening evolves statistically self-similar in the interfacial regime. This means

$$|\mathcal{F}m(t, \cdot)(k)|^2 \approx f(t^{\gamma}k), \tag{11}$$

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for some structure function f and every wave number k. Above, $\mathcal{F}m$ denotes the Fourier transform of m, i.e., $\mathcal{F}m(k) = \int_\Omega m(x)e^{ik\cdot x}\,dx$. Such a behavior is suggested by numerical simulations, but, to our knowledge, there are no rigorous results in this direction.

In order to determine the different coarsening exponents, we treat the underlying dominant transport mechanisms separately. We consider the sharp-interface models. In the purely diffusive system, the evolution of the interface Γ is the Mullins–Sekerka law

$$V = -\frac{1}{2}\left[\frac{\partial\mu}{\partial\nu}\right] \qquad \text{on } \Gamma,$$

where μ is the chemical potential defined via (7). Solutions are invariant under the scaling

$$x = \alpha\hat{x}, \quad t = \alpha^3\hat{t}, \tag{12}$$

and therefore, assuming statistical self-similarity,

$$f(t^\gamma k) \overset{(11)\&(12)}{\approx} |\mathcal{F}m(\alpha^3 t, \alpha\cdot)(k)|^2 = |\mathcal{F}m(\alpha^3 t, \cdot)(\alpha^{-1}k)|^2 \approx f((\alpha^3 t)^\gamma(\alpha^{-1}k)),$$

which yields $\gamma = 1/3$, since α is arbitrary. Thus

$$\ell(t) \sim (\lambda t)^{1/3},$$

To illustrate the λ-dependence of the coarsening rate, observe that in the purely diffusive version of (4), λ can be absorbed into the time scale via the transformation $\hat{t} = \lambda t$.

In the purely convective system, the evolution of the interface Γ is

$$V = u \cdot \nu,$$

where u solves (8). Solutions are invariant under the scaling

$$x = \alpha\hat{x}, \quad t = \alpha\hat{t}.$$

This implies, assuming statistical self-similarity, by a similar computation as above:

$$\ell(t) \sim t.$$

Comparing both coarsening rates, we expect the crossover from the diffusion-dominated to the convection-dominated regime at time resp. length

$$t_{\text{crossover}} \sim \lambda^{1/2}, \qquad \ell_{\text{crossover}} \sim \lambda^{1/2}. \tag{13}$$

In particular, in view of (10), we may think of $\lambda \geq 1$ in order to treat a crossover situation.

The heuristic argumentation presented in this section and the rigorous results in the remainder of this paper apply in any space dimension larger than or equal to two, in

particular in the physically important case of three dimensions. However, in dimension two, the validity of the convection-dominated growth law is under controversy in the physics literature. There are basically two positions. On the one hand, in [23], the authors claim that in two dimensions, the curvature-driven Stokes flow induced by (8) does not yield the appropriate evolution law of the interface, and therefore, the linear growth law $\ell \sim t$ does not hold. The argumentation is based on the investigation of Rayleigh instabilities in two dimensional hydrodynamic fluids: Linear stability analysis indicates that long cylindrical tubes of fluid surrounded by fluid of different density are *stable* under long wavelength perturbations — as opposed to the situation in three dimensions. Therefore, the authors deduce that convective coarsening is not possible. It is suggested that in two dimensions the linear coarsening rate is replaced by $\ell \sim t^{1/2}$ corresponding to droplet coalescence. This scaling law has also been numerically observed [28, 18]. On the other, the author of [11] explains the discrepancy of the coarsening rates in two and three dimensions by the occurrence of many isolated spherical domains in two dimensions, for which the hydrodynamic bulk flow is not effective, and which coarsen therefore according to the evaporation-recondensation growth law $\ell \sim t^{1/3}$. This thesis is supported by the numerical computation of different measures of length scales (different moments of the structure function). As a particular consequence, in two dimensions, the presence of a sole characteristic length scale is questionable (the largest domains, however, still show linear coarsening behavior). In three dimensions, one observes a rich connectivity among the domains, so that the accumulation of isolated droplets is less apparent. Despite this unsatisfactory ambiguity in the state of the art in 2-d coarsening of binary viscous fluids, the present work contributes into the discussion, proving that, even in two space dimensions, coarsening cannot proceed faster than $\ell \sim t$ in the convective regime (more precisely, we show that the interfacial area cannot decrease too fast).

4 Results and Method

Let us state our rigorous main results. We focus on the diffuse-interface setting, this means, we consider the energy density defined in (1), and the (gradient flow) dynamics given by (3)&(4). When it might be of interest, we also shortly comment on the corresponding sharp-interface versions of our results.

Our main result, Theorem 1 below, describes a crossover in the coarsening rates from $\ell \sim (\lambda t)^{1/3}$ in a diffusion dominated regime to $\ell \sim t$ in a convection dominated regime. The result comes in form of time-averaged lower bounds on the energy density (see discussion on page 10). Before presenting Theorem 1, we still need some preparation.

Our analysis uses two different notions of intrinsic length scales, a geometric one and a physical one. The canonical candidate for a *geometric length scale* is the inverse

energy density of the system, which, because of

$$E \approx \text{ energy of one-dimensional interfacial layer} \times \frac{\text{area of interface}}{\text{volume of system}},$$

cf. (6), indeed scales like a length (heuristically at least). The *physical length scale* is defined via a transportation distance, which we call a Monge–Kantorovich–Rubinstein distance (MKR distance) in its most abstract form, and which will be introduced in the following.

For any two measures θ, ϑ on Ω, with $\theta(\Omega) = \vartheta(\Omega)$ the MKR distance d_c with cost c is defined by

$$d_c(\theta, \vartheta) := \inf_{\pi \in \Pi(\theta, \vartheta)} \iint_{\Omega \times \Omega} c(x, y) d\pi(x, y),$$

where $\Pi(\theta, \vartheta)$ is the set off all couplings between θ and ϑ, that is the set of all measures π on $\Omega \times \Omega$ such that

$$\pi(A \times \Omega) = \theta(A) \quad \text{and} \quad \pi(\Omega \times B) = \vartheta(B) \quad \text{for all measurable sets } A, B.$$

Here, we use the notation $\iint_{A \times B} = \frac{1}{|\Omega|} \iint_{A \times B}$ for any measurable sets A and B.

With the cost function c defined by

$$c(z) := \begin{cases} \lambda^{-1/2} z & 0 \leq z \leq \lambda^{1/2}, \\ 1 + \ln z - \ln \lambda^{1/2} & \lambda^{1/2} \leq z, \end{cases} \tag{14}$$

and

$$m_+ := \max\{m, 0\} \quad \text{and} \quad m_- := \max\{-m, 0\},$$

we set

$$L := d_c(m_+, m_-). \tag{15}$$

As we shall explain later, the latter can be considered a physical length of the system. Let $\Omega = [0, \Lambda]^d$. For $z \in \mathbb{R}^d$ we set $\|z\| = \min\{|z + \Lambda(\alpha_1 e_1 + \cdots \alpha_d e_d)| : \alpha_1, \ldots, \alpha_d \in \mathbb{Z}\}$ where e_i are unit coordinate vectors. Note that if Ω is considered as a flat torus then the distance between x and y in Ω is $\|x - y\|$. We use $c(x, y) = c(\|x - y\|)$ as a symbol for both a function from $\Omega \times \Omega \to [0, \infty)$ and a function from $[0, \infty) \to [0, \infty)$ as it is clear from the argument which one is considered. Note that, since $\overline{m} = 0$, we have $\int_\Omega m_+(x, t) \, dx = \int_\Omega m_-(x, t) \, dx$ for all $t \geq 0$. We remark that for $\overline{m} \neq 0$, one would consider $(m - \overline{m})_+$ and $(m - \overline{m})_-$.

Before motivating the definition of c and commenting the interpretation of L as a physical length in our dynamical system, we present our main result and discuss the analytical method we apply.

Theorem 1. *Let $E(m(0)) \ll 1$ and $L(m(0)) \ll 1$. Then*

$$\int_0^T \max\{\lambda^{1/2} E^2, E\} \, dt \gtrsim \int_0^T \min\left\{\lambda^{1/2} \frac{1}{(\lambda t)^{2/3}}, \frac{1}{t}\right\} dt$$

for all T such that $\lambda^{-1/2} T \gg L(m(0))^3$.

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Remark 1. *The above result, with $\lambda = 1$ and without assumption on $E(m(0))$, also holds for the sharp interface system under the assumption that the sharp-interface system has smooth enough solutions.*

We use the sloppy notation "$\gtrsim$" and "$\gg$" to indicate that the inequality holds up to a generic constant which may depend on the space dimension only, provided that $E(m(0))$ and $L(m(0))$ are sufficiently small and $\frac{\lambda^{-1/2}T}{L(m(0))^3}$ is sufficiently large. In particular, the statement in Theorem 1 is uniform in $L(m(0))$ and λ, and we formally obtain the individual results from [15] (pure diffusion) and [4] (pure convection) in the asymptotic limits $\lambda \gg 1$ and $\lambda \ll 1$, respectively.

The result of Theorem 1 above is in agreement with the physical prediction in [24]. Indeed, since the normalized energy scales like an inverse length (see discussion above) and assuming that there is only one length scale present in the dynamics, a lower bound on the energy can be interpreted as an upper bound on the coarsening rate. As other results based on the technique of [15], our analysis produces only time-averaged and only lower bounds on the energy. (A counter-example for a pointwise version of the coarsening rates derived within this method is given in the original paper [15, Remark 4].) The condition on T gives a bound for the minimal size of the time interval over which the coarsening rates are averaged. Also, we have no rigorous arguments to precisely determine the crossover time in terms of λ. Our result has to be read as follows: The initial configuration has to be chosen such that its typical length scale is at least of order one, $1 \ll \frac{1}{E(m(0))}$, to ensure that we start within the interfacial regime, i.e., the mixture is separated into two domains of the two phases; but the typical length scale of the initial configuration is well below the crossover length, $L(m(0)) \ll 1$ (recall that for small length scales, L is normalized by $\lambda^{1/2}$), cf. (13). The diffusive regime, $E \gg \frac{1}{\lambda^{1/2}}$ and $t \ll \lambda^{1/2}$, is only relevant if $\lambda \geq 1$. If so, the above result is a time-averaged version of $E \gtrsim \frac{1}{(\lambda t)^{1/3}}$, since in this situation $\max\{\lambda^{1/2}E^2, E\} = \lambda^{1/2}E^2$ and $\min\left\{\frac{\lambda^{1/2}}{(\lambda t)^{2/3}}, \frac{1}{t}\right\} = \frac{\lambda^{1/2}}{(\lambda t)^{2/3}}$. Likewise, in the regime $E \ll \frac{1}{\lambda^{1/2}}$ and $t \gg \lambda^{1/2}$, our result is a time-averaged version of $E \gtrsim \frac{1}{t}$.

We want to remark that upper bounds on coarsening rates are quite different from lower bounds. There are configurations, like parallel planar layers, that do not coarsen at all, or do so exponentially slowly. Therefore, lower bounds depend strongly on the initial data and can only be generically true.

The method we apply in our analysis was first introduced in [15]. It exploits the gradient flow structure of the dynamics. In fact, the dynamical system (3)&(4) (or analogously (7)–(9)) can be interpreted as a gradient flow for the metric tensor

$$
\begin{aligned}
&g(\delta m, \delta m) \\
&= \inf_{j,u}\left\{ \fint_\Omega \frac{1}{\lambda}|j|^2\,dx + \fint_\Omega |Du|^2\,dx \,\,\Big|\,\, \delta m + \nabla\cdot(j + mu) = 0,\, \nabla\cdot u = 0 \right\}, \quad (16)
\end{aligned}
$$

cf. [4, Section 1.4]. The method of [15] now translates bounds on the energy landscape, i.e., information on how fast the can the energy decrease as a function of a

distance (originally, the distance induced by the metric tensor) to some reference configuration, to bounds on the dynamics, i.e., bounds on how fast the energy decreases as a function of time. The method consists mainly of three ingredients: an energy dissipation inequality, an interpolation (or isoperimetric) inequality, and an ODE argument.

The main contribution of this work is the dissipation inequality.

Proposition 2 (Dissipation inequality)**.** *Let m be a solution of* (4)*. Assume that $E(m(0)) \lesssim 1$. Then L is an absolutely continuous function and for a.e. $t \geq 0$,*

$$\frac{d}{dt} L(t) \lesssim \left(\fint_\Omega \lambda |\nabla \mu|^2 + |Du|^2 \, dx \right)^{1/2}.$$

The statement with $\lambda = 1$ holds for the sharp interface system as well, again provided that it has regular enough solutions. We may also drop the condition on the initial energy.

The interpolation inequality we obtain holds for a very broad range of cost functions. It represents a diffuse-interface counterpart of the one proved in [4]. One should note that the interpolation inequality for the sharp interface case does not require an assumption on energy smallness.

Proposition 3 (Interpolation inequality)**.** *Let c be monotonically increasing with $c(0) = 0$. Let $m \in L^2(\Omega)$ with $\overline{m} = 0$. Assume $E(m) \ll 1$. Then there exists constant c_0, dependent on the space dimension only, such that*

$$d_c(m_+, m_-) \gtrsim c \left(\frac{1}{c_0 E(m)} \right).$$

Finally, we state the ODE argument. The inhomogeneous form of the time averages in the statement of Proposition 4 is due to the particular feature of the argument, which only allows for L^q averages in time, with $1 < q < 3$ in the diffusive (for convenience, we choose $q = 2$), and $q = 1$ in the convective regime, in order to capture the explicit form of the cost function c from the interpolation inequality in the particular regimes. Notice that in our formulation of the ODE argument below, we have absorbed c_0 and the constants from the inequalities in Propositions 2 & 3 into E, L, and the new constant C.

Proposition 4 (ODE argument)**.** *Let $E(t)$ and $L(t)$ be two absolutely continuous, nonnegative functions satisfying*

$$\frac{dL}{dt} \leq C \left(-\frac{dE}{dt} \right)^{1/2} \qquad \text{for a.e. } t, \tag{17}$$

and

$$L \geq \begin{cases} \frac{1}{\lambda^{1/2} E} & \text{if } 1 \leq \lambda^{1/2} E, \\ 1 + \ln \frac{1}{\lambda^{1/2} E} & \text{if } 1 \geq \lambda^{1/2} E. \end{cases} \tag{18}$$

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Assume that $2L(0) \leq 1$. Then there is a constant $\tilde{C}$ such that

$$\int_0^T \max\{\lambda^{1/2} E^2, E\} \, dt \;\geq\; \frac{1}{\tilde{C}} \int_0^T \min\left\{\lambda^{1/2} \frac{1}{(\lambda t)^{2/3}}, \frac{1}{t}\right\} dt,$$

for any T such that $\lambda^{-1/2} T \geq L(0)^3$.

Now we come back to the definition of c, (14), and the interpretation of L, (15), as a *physical length scale*. A natural choice of the physical length scale is related to the gradient flow structure of the dynamics: In a gradient flow, the metric tensor encodes the limiting dissipation mechanisms. In particular, the geodesic distance induced by the metric tensor measures the minimal amount of energy that is dissipated when "moving" from one point in configuration space to another. However, in our situation, the induced distance with metric tensor defined in (16) is not know explicitly. Therefore, we construct a proxy $L(m)$ that is bounded by the induced distance. The idea of approximating induced distance functions by MKR distances has been introduced in [19, 13].

We now motivate the choice of c. The cost function has to be chosen in such a way that the MKR distance is dominated by the induced distance function. In the purely diffusive setting, the induced distance reduces to the H^{-1} norm. Though in this situation a proxy is not necessary, the authors of [15] use a slightly weaker norm, the $H^{-1,\infty}$ norm, which is equivalent to the Monge–Kantorovich distance (i.e., the MKR distance with cost function given by the Euclidean distance $c(z) = z$), in order to treat simultaneously the case of surface diffusion (diffusion along the interfacial layer in contrast to diffusion through the bulk). In the purely convective setting, the induced distance is due to the viscous dissipation, i.e., $\int_\Omega |\nabla u|^2 \, dx$. Since the dissipation mechanism only controls the *gradient* of the convecting velocity, it is not surprising that the cost functional in the MKR distance can only grow logarithmically: In the finite dimensional analogue, trajectories of a Lipschitz vector field can diverge exponentially fast. This idea is exploited in [4]. Since our goal is to prove the crossover in the coarsening rates from diffusion- to convection-mediated coarsening, we combine the cost function from [15] and [4]: We use the Euclidean distance as cost function for distances smaller (measured in terms of E) than the crossover length, $\frac{1}{E} \sim \lambda^{1/2}$, cf. (13), and the logarithmic cost function for distances larger than the crossover length.

A key insight of our work is that combining the cost appropriately for the two problems corresponding to separate transport mechanisms works for the problem in which the transport mechanisms are combined.

We 'remark that there is a subtle but nontrivial difference from [4] in the definition of L is used instead of $d_c(m+1, 1)$. The quantity above is more natural for the problem considered. Furthermore the change is necessary since there are no known L^∞ bounds on m which are uniform in time (and which one could use to find M such that $m + M \geq 0$ and then consider $d_c(m+M, M)$). However the total integrals

of m_+ and m_- change in time, which needs to be accounted for when computing $\frac{dL}{dt}$ for example.

MKR distances have been successfully introduced as proxies for the induced distance function in the framework of [15] in two further situations: In [25], the author considers a Wasserstein distance (i.e., an MKR distance with cost function given by the square of the Euclidean distance) to prove an upper bound on the coarsening rates in a non-local, degenerate Cahn–Hilliard equation modelling biological aggregation and phase segregation in binary alloys. In [20], a model for viscous thin films is under consideration, in which droplet configurations coarsen by Ostwald ripening and droplet collision. The evolution is given by a Cahn–Hilliard-type equation, the induced distance *is* the Wasserstein distance.

5 Effect of material transport on optimal transportation distances

We establish estimates on how much are the optimal transportation distances between fluid components changed if the fluid is perturbed. We consider two sources of perturbation: mass redistribution via a flux j and bulk transport via a velocity vector field u. Since this situation is of general interest, in this section j and u are not assumed to be given by a model of demixing. More precisely, we consider m to be a (distributional) solution of

$$\partial_t m + \nabla \cdot j + \nabla \cdot (mu) = 0$$

on a torus Ω, where j and u are given.

Effect of bulk transport on optimal transportation distances was considered in [4, Lemma 1]. We state the result below and refer to [4] for a proof.

Lemma 5. *Assume that $c : [0, \infty) \longrightarrow [0, \infty)$ is smooth and nondecreasing, with $c(0) = 0$. We denote the associated cost function by the same letter: $c(x, y) = c(\|x - y\|)$, where $\|x - y\|$ denotes the distance on Ω considered as a torus. Let u be a C^1 vector field with period cell Ω. Let $m_1, m_2 \in C(0, T; L^1(\Omega))$ be two distributional solutions of*

$$\partial_t m_i + \nabla \cdot (m_i u) = 0,$$

such that $m_1(0)(\Omega) = m_2(0)(\Omega)$. Then

$$\frac{\bar{d}^+}{dt} d_c(m_1(t), m_2(t))$$
$$\leq \iint_{\Omega \times \Omega} c'(\|x - y\|) \nabla_x \|x - y\| \cdot (u(x, t) - u(y, t)) \, d\pi(t),$$

where $\pi(t)$ is an optimal transportation plan with cost c between $m_1(t)$ and $m_2(t)$ and $\frac{\bar{d}^\pm}{dt}$ is the one-sided upper derivative, i.e.,

$$\frac{\bar{d}^+}{dt} f(t) := \limsup_{h \to 0+} \frac{f(t + h) - f(t)}{h}.$$

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The analogous result holds for the one-sided lower derivative.

When applying the above lemma to the demixing problems, we have in mind m being the order parameter of average 0 and set $m_1 = m_+$ and $m_2 = m_-$. The main difference between the situation considered in [4] and the one here is that the total mass of m_+, that is $m_+(t)(\Omega)$, is not constant in time in the diffuse-interface case. This is entirely due to the transport by flux j. The following lemma contains the essential estimate to handle that element. It is an integral version of the estimate on the rate of change of the transportation distance.

Lemma 6. *Let $c : [0, \infty) \longrightarrow [0, \infty)$ be smooth and nondecreasing, with $c(0) = 0$. Furthermore assume that c is subadditive and that $c'(0) \le 1$. Let m be an L^2 function on Ω and j an H^1 vector field. Let $m' = m + \nabla \cdot j$. Then*

$$d_c(m'_+, m'_-) - d_c(m_+, m_-) \ \le \ \fint_\Omega |j| \, dx.$$

Proof. Note that since c is subadditive, nondecreasing, and $c(0) = 0$, the function $c(x, y) = c(\|x - y\|)$ defines a metric on Ω. This enables is to use Lemma 10 in the Appendix to conclude

$$d_c(m'_+, m'_-) \ \le \ d_c(m'_+ + m_-, m'_- + m_+) + d_c(m_+, m_-).$$

Observe that $d_c(m_-, m_+) = d_c(m_+, m_-)$. Let d_1 be the Monge–Kantorovich distance, that is the optimal transportation distance corresponding to the Euclidean distance cost. Since c is subadditive and $c'(0) \le 1$, it holds that $c(z) \le z$ for all $z \ge 0$. Therefore

$$d_c(m'_+ + m_-, m'_- + m_+) \ \le \ d_1(m'_+ + m_-, m'_- + m_+).$$

We now recall the following representation formula for d_1 (see [29, page 38]):

$$d_1(m, m') \ = \ \inf \left\{ \fint_\Omega |\tilde{j}| \, dx \ : \ \nabla \cdot \tilde{j} = m' - m \right\}. \tag{19}$$

On the one hand, this implies that

$$d_1(m'_+ + m_-, m'_- + m_+) \ = \ d_1(m_+ - m_-, m'_+ - m'_-) \ = \ d_1(m, m').$$

On the other hand, we have the estimate

$$d_1(m, m') \ \le \ \fint_\Omega |j| \, dx.$$

Gathering the inequalities above we conclude that

$$d_c(m'_+, m'_-) \ \le \ \fint_\Omega |j| \, dx + d_c(m_+, m_-).$$

$\square$

While the above lemma applies to the sharp-interface case as well, here we state the sharp-interface result in a more geometric setting. Namely, we consider the situation when the flux is not specified, but only the evolution of interfaces is given. To state the appropriate result, consider the evolution of subsets of Ω. Let $\Omega_+(t)$ and $\Omega_-(t)$ be disjoint open subsets of Ω such that $\partial\Omega_+(t) = \partial\Omega_-(t) = \Gamma(t)$, $|\Omega_+(t)| = |\Omega_-(t)|$ for all t and $\Gamma(t)$ is a smooth manifold evolving smoothly in time. Let V be the normal velocity by which Γ is evolving.

Corollary 7. *Consider c as in Lemma 6 and $\Omega_+(t)$, $\Omega_-(t)$, $\Gamma(t)$, and V as above. Let $m_+ = \chi_{\Omega_+}$, $m_- = \chi_{\Omega_-}$, and $m = m_+ - m_-$. For $h > 0$, let Q_h be the solution of*

$$-\Delta Q_h = m(t+h) - m(t).$$

Then

$$d_c(m_+(t+h), m_-(t+h)) - d_c(m_+(t), m_-(t))$$
$$\leq \fint_\Omega |\nabla Q_h|\, dx \leq \int_t^{t+h} \fint_\Omega |\nabla q(x,s)|\, dx ds$$

where $q(\cdot, t)$ solves

$$
\begin{aligned}
-\Delta q &= 0 && in\ \Omega\backslash\Gamma(t), \\
\left[\frac{\partial q}{\partial \nu}\right] &= 2V && on\ \Gamma(t),
\end{aligned}
\tag{20}
$$

where ν is the unit outside normal vector to Ω_+ and $[\cdot]$ denotes the jump over the interface. Furthermore

$$\frac{\bar{d}^+}{dt} d_c(m_+(t), m_-(t)) \leq \left(\frac{1}{|\Omega|}\int_{\Gamma(t)} 2Vq\, d\,H^{d-1}\right)^{\frac{1}{2}}.$$

Proof. The proof of the first inequality follows immediately from the proof of Lemma 6 with $j = -\nabla Q_h$. To show the second fact note that using a smooth function ξ as a test function in (20) gives

$$
\begin{aligned}
0 &= -\int_\Omega \nabla q \cdot \nabla \xi\, dx + \int_\Gamma \left[\frac{\partial q}{\partial \nu}\right] \xi\, d\mathcal{H}^{d-1} \\
&= -\int_\Omega \nabla q \cdot \nabla \xi\, dx + \int_\Gamma 2V\, \xi\, d\mathcal{H}^{d-1} \\
&= -\int_\Omega \nabla q \cdot \nabla \xi\, dx + \frac{d}{dt}\int_\Omega m(x,t)\xi(x)\, dx.
\end{aligned}
$$

Integrating from t to $t+h$ gives that $\int_t^{t+h} q(x,s)ds$ satisfies the equation that defines Q_h. Uniqueness implies that $Q_h(x,t) = \int_t^{t+h} q(x,s)ds$, which in turn implies the

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second inequality. Dividing by h and taking $\limsup_{h \to 0+}$ implies that

$$
\frac{\bar{d}^+}{dt} d_c(m_+(t), m_-(t)) \leq \left(\fint_\Omega |\nabla q(x,t)|^2 \, dx \right)^{\frac{1}{2}}
$$
$$
= \left(\frac{1}{|\Omega|} \int_\Gamma \left[\frac{\partial q}{\partial \nu} \right] q \, d\mathcal{H}^{d-1} \right)^{\frac{1}{2}}
$$
$$
= \left(\frac{1}{|\Omega|} \int_\Gamma 2V q \, d\mathcal{H}^{d-1} \right)^{\frac{1}{2}}.
$$

$\square$

We now combine the results of Lemma 5 and Lemma 6 and generalize them to apply to less regular vector fields.

Lemma 8. *Let $c : [0, \infty) \longrightarrow [0, \infty)$ be C^1 and concave and such that $c(0) = 0$ and $c'(0) \leq 1$. Assume that u is vector field in $L^1(0, T; L^2(\Omega, \mathbb{R}^d))$ and j is a vector field in $L^1(0, T; L^1(\Omega, \mathbb{R}^d))$. Let $m \in C(0, T; L^2(\Omega))$ be a distributional solution of*

$$
\partial_t m + \nabla \cdot j + \nabla \cdot (mu) = 0
$$

with $\overline{m} = 0$. Let $L(t) = d_c(m_+(t), m_-(t))$. Then for all $t \in [0, T)$ and $h \in (0, T - t)$

$$
L(t+h) - L(t)
$$
$$
\leq \int_t^{t+h} \fint_\Omega |j(x,s)| \, dxds
$$
$$
+ \int_t^{t+h} \iint_{\Omega \times \Omega} c'(x-y) \nabla_x \|x-y\| \cdot (u(x,s) - u(y,s)) \, d\pi(s)ds, \qquad (21)
$$

where $\pi(s)$ is an optimal transportation plan, for cost c, between $m_+(s)$ and $m_-(s)$.

From (21) and assumptions on j, u, and m, it follows that L is an absolutely continuous function. That allows us to use the (a.e.) derivative of L in subsequent calculations.

Proof. Step 1. Assume that u and j are C^1 vector fields. To be able to separately account for the effects of the two transport mechanisms, we introduce for any $t \in [0, T)$ the solution ρ_t of

$$
\partial_s \rho_t + \nabla \cdot (\rho_t u) = 0 \qquad \text{on } \Omega \times [t, T],
$$
$$
\rho_t(t) = m(t).
$$

Let $\sigma_t(s) = m(s) - \rho_t(s)$. When the subscript t is clear from the context, we will omit it. To estimate the change of the transportation distance, we separate the contributions that come from the two transport mechanisms. In particular, by

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Lemma 10 in the Appendix A and $\rho_t(t) = m(t)$, we have for $0 < h < T - t$ and $\rho = \rho_t$,

$$d_c(m_+(t+h), m_-(t+h)) - d_c(m_+(t), m_-(t))$$
$$\leq \ d_c(\rho_+(t+h), \rho_-(t+h)) - d_c(\rho_+(t), \rho_-(t))$$
$$+ d_c(\rho_+(t+h) + m_-(t+h), \rho_-(t+h) + m_+(t+h)).$$

Now (21) follows from the two estimates

$$d_c(\rho_+(t+h), \rho_-(t+h)) - d_c(\rho_+(t), \rho_-(t))$$
$$\leq \ \int_t^{t+h} \iint_{\Omega \times \Omega} c'(x-y) \nabla_x \|x-y\| \cdot (u(x,s) - u(y,s))\, d\pi(s) ds \qquad (22)$$

and

$$d_c(\rho_+(t+h) + m_-(t+h), \rho_-(t+h) + m_+(t+h))$$
$$\leq \ \int_t^{t+h} \fint_\Omega |j(x,s)|\, dx ds + o(h). \qquad (23)$$

Indeed, dividing by h and taking the limit $h \to 0$ establishes the upper bound for $\frac{\bar{d}^+}{dt} L(t)$ for every $t \in [0, T)$. Integrating in time then gives (21).

Obviously, by the definition of ρ, (22) is a direct consequence of Lemma 5. We now argue in favor of (23). We estimate

$$d_c(\rho_+(t+h) + m_-(t+h), \rho_-(t+h) + m_+(t+h))$$
$$\leq \ d_1(\rho_+(t+h) + m_-(t+h), \rho_-(t+h) + m_+(t+h))$$
$$= \ d_1(\rho(t+h), m(t+h))$$
$$= \ d_1(\sigma(t+h), 0)$$
$$\leq \ \int_t^{t+h} \fint_\Omega |j(x,s)| + |\sigma(x,s) u(x,s)|\, dx ds,$$

where we used $c(z) \leq z$, the representation of the d_1 distance given in (19) and the fact that σ satisfies

$$\partial_s \sigma + \nabla \cdot j + \nabla \cdot (\sigma u) \ = \ 0,$$

with $\sigma(t) = 0$. We have to show that the second term on the r. h. s. of the above inequality is of higher order. To do so, we multiply the equation by σ; integrating by parts gives for $s \geq t$:

$$\frac{d}{ds} \frac{1}{2} \|\sigma(s)\|_{L^2(\Omega)}^2 \ \leq \ \|j(s)\|_{H^1(\Omega)} \|\sigma(s)\|_{L^2(\Omega)} + \frac{1}{2} \fint_\Omega \sigma(s)^2 |\nabla \cdot u|\, dx.$$

Above and in the rest of this proof, all the norms, $\|\cdot\|_{L^2(\Omega)}$, $\|\cdot\|_{H^1(\Omega)}$, etc., are rescaled by the size of the domain. Dividing by $\|\sigma(s)\|_{L^2(\Omega)}$ yields

$$\frac{d}{ds} \|\sigma(s)\|_{L^2(\Omega)} \ \leq \ C(j, u)(1 + \|\sigma(s)\|_{L^2(\Omega)}),$$

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where $C = C(j, u)$ is a constant involving the C^1 bounds on j and u. Via Gronwall's inequality

$$\|\sigma(s)\|_{L^2(\Omega)} \leq \exp(C(s - t)) - 1,$$

for all $s \in [t, T]$. Therefore

$$\int_t^{t+h} \fint_\Omega |\sigma(x, s)u(x, s)|\, dxds = o(h).$$

Step 2. Consider $j \in L^1(0, T; L^1(\Omega))$, with u still assumed to be C^1. Let η_ε be a standard mollifier in space and time. Consider the interval $[t, t + h] \subset (0, T)$. Convolving with η_ε, where Ω is considered as a torus, gives that $m_\varepsilon := m * \eta_\varepsilon$ is a solution of

$$\partial_t m_\varepsilon + \nabla \cdot (m_\varepsilon u) + \nabla \cdot \tilde{j}_\varepsilon = 0 \qquad \text{on } \Omega \times [t, t + h],$$

where $\tilde{j}_\varepsilon := j * \eta_\varepsilon - m_\varepsilon u + (mu) * \eta_\varepsilon$. Note that $\tilde{j}_\varepsilon$ is C^1 and thus the assumptions of Step 1 are satisfied. Let for $s \in [t, t + h]$, $L_\varepsilon(s) := d_c(m_{\varepsilon+}(s), m_{\varepsilon-}(s))$ and let $\pi_\varepsilon(s)$ be an optimal transportation plan between $m_{\varepsilon+}(s)$ and $m_{\varepsilon-}(s)$. We have

$$L_\varepsilon(t + h) - L_\varepsilon(t)$$
$$\leq \int_t^{t+h} \iint_{\Omega \times \Omega} c'(x - y)\nabla_x \|x - y\| \cdot (u(x, s) - u(y, s))\, d\pi_\varepsilon(s)ds$$
$$+ \int_t^{t+h} \fint_\Omega |\tilde{j}_\varepsilon(x, s)|\, dxds.$$

Since $m_{\varepsilon+}(s) \to m_+(s)$ and $m_{\varepsilon-}(s) \to m_-(s)$ in $L^2(\Omega)$ as $\varepsilon \to 0$, we conclude $L_\varepsilon(t) \to L(t)$ and $L_\varepsilon(t + h) \to L(t + h)$ as $\varepsilon \to 0$.

Regarding the term involving the velocity u: For any $s \in [t, t + h]$,

$$\iint_{\Omega \times \Omega} c'(x - y)\nabla_x \|x - y\| \cdot (u(x, s) - u(y, s))\, d(\pi_\varepsilon(s) - \pi(s)) \to 0 \quad \text{as } \varepsilon \to 0, \quad (24)$$

since $\pi_\varepsilon(s) - \pi(s) \to 0$ weakly in the sense of measures due to stability of optimal transportation plans (see Theorem 5.20 in [30]). Assumptions on c imply that $0 \leq c'(z) \leq 1$ for all $z \geq 0$. Thus

$$\left| \iint_{\Omega \times \Omega} c'(x - y)\nabla_x \|x - y\| \cdot (u(x, s) - u(y, s))\, d(\pi_\varepsilon(s) - \pi(s)) \right|$$
$$\leq \iint_{\Omega \times \Omega} |u(x, s)| + |u(y, s)|\, d(\pi_\varepsilon(s) + \pi(s))$$
$$= \fint_\Omega |u(x, s)|(m_{\varepsilon+}(x) + m_+(x))\, dx + \fint_\Omega |u(y, s)|(m_{\varepsilon-}(y) + m_+(y))\, dy,$$

which enables us to the dominated convergence theorem to obtain the integral-in-time form of (24).

Since $\tilde{j}_\varepsilon \to j$ in $L^1(0, T; L^1(\Omega))$ as $\varepsilon \to 0$, convergence of the term involving $\tilde{j}_\varepsilon$ follows as well. So (21) follows in the $\varepsilon \to 0$ limit.

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Step 3. Now consider $u \in L^1(0, T; L^2(\Omega))$ and $j \in L^1(0, T; L^1(\Omega))$. Again let $[t, t+h] \subset (0, T)$. Let η_ε be a mollifier in space and time and let $u_\varepsilon := u * \eta_\varepsilon$. Then $m \in C(0, T; L^2(\Omega))$ is a distributional solution of

$$\partial_t m + \nabla \cdot (m u_\varepsilon) + \nabla \cdot (j + m(u - u_\varepsilon)) = 0 \qquad \text{on } \Omega \times [t, t+h].$$

Let $j_\varepsilon := j + m(u - u_\varepsilon)$. Note that j_ε, u_ε satisfy the assumption of Step 2. To take the limit $\varepsilon \to 0$ of

$$
\begin{aligned}
L(t+h) &- L(t) \\
&\leq \int_t^{t+h} \fint_\Omega |j_\varepsilon(x, s)| \, dx \\
&\quad + \iint_{\Omega \times \Omega} c'(x - y) \nabla_x \|x - y\| \cdot (u_\varepsilon(x, s) - u_\varepsilon(y, s)) \, d\pi(s) ds,
\end{aligned}
$$

it suffices to observe that $j_\varepsilon \to j$ in $L^1(0, T; L^1(\Omega))$ as $\varepsilon \to 0$,

$$
\begin{aligned}
\bigg| \iint_{\Omega \times \Omega} & c'(x - y) \nabla_x \|x - y\| \cdot (u_\varepsilon(x, s) - u(x, s) - (u_\varepsilon(y, s) - u(y, s)) \, d\pi(s) ds \bigg| \\
&\leq \fint_\Omega |u_\varepsilon(x, s) - u(x, s)| m_+(x, s) \, dx + \fint_\Omega |u_\varepsilon(y, s) - u(y, s)| m_-(y, s) \, dy,
\end{aligned}
$$

integrate in time and use the fact that $u_\varepsilon \to u$ in $L^1(0, T; L^2(\Omega))$ and m_+, $m_- \in L^\infty(0, T; L^2(\Omega))$.

For the case $t = 0$, the result follows in the limit of the result on intervals $[\delta, \delta + h]$, due to continuity of both sides of (21) with respect to t. $\qquad\qquad\square$

Again for sharp interfaces we present the result in the geometric setting.

Corollary 9. *Consider c as in Lemma 8, and $\Omega_+(t)$, $\Omega_-(t)$, $\Gamma(t)$, m, m_+, m_- and V as in Corollary 7. Let u be a smooth vector field on $\Omega \times [0, \infty)$. Let $\tilde{V} = V - u \cdot \nu$ where ν is the unit outside normal vector to Ω_+. Then $L(t) = d_c(m_+(t), m_-(t))$ is absolutely continuous and for $q(\cdot, t)$ solving*

$$
\begin{aligned}
-\Delta q &= 0 \qquad \text{in } \Omega(t) \backslash \Gamma(t), \\
\left[\frac{\partial q}{\partial \nu} \right] &= 2\tilde{V} \qquad \text{on } \Gamma(t),
\end{aligned}
$$

we have for a.e. t

$$
\begin{aligned}
\frac{d}{dt} L(t) &\leq \iint_{\Omega \times \Omega} c'(x - y) \nabla_x \|x - y\| \cdot (u(x) - u(y)) \, d\pi + \fint_\Omega |\nabla q(x, s)| \, dx \\
&\leq \iint_{\Omega \times \Omega} c'(x - y) \nabla_x \|x - y\| \cdot (u(x) - u(y)) \, d\pi + \left(\fint_{\Gamma(t)} 2Vq \, d\mathcal{H}^{d-1} \right)^{\frac{1}{2}},
\end{aligned}
$$

where π is any optimal transportation plan between $m_+(t)$ and $m_-(t)$.

6 Proof of the upper bound on coarsening rate

6.1 Dissipation Inequality

We now turn to the proof of Proposition 2. It relies on the main result from the previous section, Lemma 8, which measures the effect of diffusive and convective material transport on transportation distances. Our proof follows closely the one of [4, Proposition 2.2], which in turn was inspired by estimates derived by Crippa & DeLellis in [7, Theorem 2.1 resp. Theorem 3.3].

Proof of Proposition 2. We apply Lemma 8 with c replaced by $\lambda^{-1/2}c$. Then $c'(0) \leq 1$ and $c'(z) \leq \frac{1}{z}$, and we have for a.e. $t \geq 0$

$$\frac{d}{dt}L(t) \; \leq \; \fint_\Omega \frac{1}{\lambda^{1/2}}|j|\,dx + \iint_{\Omega\times\Omega} \frac{|u(x)-u(y)|}{|x-y|}\,d\pi,$$

where π is any optimal transport plan with respect to cost c between $m_+(t)$ and $m_-(t)$. Recall that $j = -\lambda\nabla\mu$ in our setting. Thus

$$\fint_\Omega \frac{1}{\lambda^{1/2}}|j|\,dx \; \leq \; \left(\fint_\Omega \lambda|\nabla\mu|^2\,dx\right)^{\frac{1}{2}}.$$

The estimate for the contribution coming from convective transport is exactly the one from [4, Proposition 2]. For the convenience of the reader, we repeat the argument. The proof is based on maximal functions, $M(f)(x) = \sup_{r>0}\frac{1}{|B_r|}\int_{B_r(x)}|f|\,dx$, cf. [26]. Maximal functions have the following basic properties:

$$|f(x)-f(y)| \; \lesssim \; (M(Df)(x)+M(Df)(y))|x-y|,$$

and

$$\fint_\Omega |Mf|^2\,dx \; \lesssim \; \fint_\Omega |f|^2\,dx.$$

Using these properties of Mf, we have

$$\iint_{\Omega\times\Omega} \frac{|u(x)-u(y)|}{|x-y|}\,d\pi$$
$$\lesssim \; \iint_{\Omega\times\Omega} (M(Df)(x)+M(Df)(y))\,d\pi$$
$$\lesssim \; \fint_\Omega M(Du)(m_+ + m_-)\,dx$$
$$\lesssim \; \left(\fint_\Omega M(Du)^2\,dx\right)^{\frac{1}{2}} \left(\fint_\Omega m^2\,dx\right)^{\frac{1}{2}}$$
$$\lesssim \; \left(\fint_\Omega |Du|^2\,dx\right)^{\frac{1}{2}}.$$

Above we used that

$$
\begin{aligned}
\fint_\Omega m^2 \, dx \;=&\; \fint_{\{|m|>2\}} m^2 \, dx + \fint_{\{|m|\leq 2\}} m^2 \, dx \\
\leq&\; \fint_{\{|m|>2\}} (1-m^2)^2 \, dx + \fint_{\{|m|\leq 2\}} 2^2 \, dx \\
\leq&\; E + 4 \lesssim 5,
\end{aligned}
$$

since $E \leq E(m(0)) \lesssim 1$. Combining the estimates above and the expression for the energy dissipation, (5), gives the desired estimate. $\qquad\square$

For the sharp-interface model, we no longer assume that $E \lesssim 1$. Also recall that we take $\lambda = 1$. The proof is entirely analogous to the diffuse-interface case, so we do not present the details.

6.2 Interpolation Inequality

The interpolation inequality for sharp-interfaces is the one proved in [4][Proposition 3]. The one for diffuse interfaces can be proved very similarly. It requires the additional result that the domain is essentially divided into phases. We present a proof below for completeness. We also remark that an alternative proof of the inequality follows from the proof of Theorem 6 (and Corollary 7) in [25] (in particular Step 5 and Step 6 imply that in any coupling between m_+ and m_- a significant proportion of the mass has to be transported over distances at least of size $1/E$ which implies the inequality). Recall that the interpolation inequality was stated in Proposition 3.

Proof of Proposition 3. We introduce some notations. We denote by A the set essentially occupied by one phase, and by χ its indicator function. More precisely,

$$
A = \{x \in \Omega \; : \; m(x) \geq \frac{1}{2}\} \quad \text{and} \quad \chi = \chi_A.
$$

Furthermore, let h_W denote some constant satisfying

$$
W(m) \;\geq\; \begin{cases} 2h_W(|m|-1) & \text{for } |m| \geq \frac{3}{2}, \\ h_W & \text{for } |m| \leq \frac{1}{2}. \end{cases}
$$

Here, $W(m)$ denotes the potential energy, $W(m) := \frac{1}{2}(1-m^2)^2$. (It is easy to check that $h_W = \frac{9}{32}$ is optimal.) We first claim that

$$
\|A\| \;:=\; \frac{|A \cap \Omega|}{|\Omega|} \;\gtrsim\; 1. \tag{25}
$$

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We split the phase space according to

$$
\begin{aligned}
1 \;=\; & \fint_{\{m \le -\frac{1}{2}\}} (m+1)\,dx + \fint_{\{-\frac{1}{2} < m < \frac{1}{2}\}} (m+1)\,dx \\
& + \fint_{\{\frac{1}{2} \le m \le \frac{3}{2}\}} (m+1)\,dx + \fint_{\{\frac{3}{2} < m\}} (m+1)\,dx \\
\le\; & \frac{1}{2} + \frac{3}{2}\fint_{\{-\frac{1}{2} < m < \frac{1}{2}\}} 1\,dx + \frac{5}{2}\|A\| + \fint_{\{\frac{3}{2} < m\}} (m-1)\,dx + 2\left\|\left\{\frac{3}{2} < m\right\}\right\| \\
\le\; & \frac{1}{2} + \frac{3}{2h_W}\fint_\Omega W(m)\,dx + \frac{5}{2}\|A\| + \frac{1}{2h_W}\fint_\Omega W(m)\,dx + 2\|A\| \\
\le\; & \frac{1}{2} + \frac{2}{h_W}\fint_\Omega W(m)\,dx + \frac{9}{2}\|A\|.
\end{aligned}
$$

We can rewrite the estimate as $1 \le \frac{4}{h_W}E + 9\|A\|$. Due to the assumption $E(m) \ll 1$, this implies (25).

In the sequel, the superscript R denotes the convolution with a standard mollifier supported in the ball of radius R. From (25) and the definition of A we find

$$
1 \;\lesssim\; \fint_\Omega \chi\,dx \;\lesssim\; \fint_\Omega \chi m\,dx,
$$

which we spilt according to

$$
1 \;\lesssim\; \fint_\Omega \chi(m - m^R)\,dx + \fint_\Omega \chi m^R\,dx. \tag{26}
$$

For the first term in (26), we proceed as in [15]. Defining

$$
U(m) := \int_0^m |1 - \tilde{m}^2|\,d\tilde{m},
$$

we have $\frac{dU}{dm} = |1 - m^2|$, and thus

$$
\begin{aligned}
\fint_\Omega |\nabla(U(m))|\,dx \;=\; & \fint_\Omega |\nabla m|\frac{dU}{dm}\,dx \\
\le\; & \fint_\Omega \frac{1}{2}|\nabla m|^2 + \frac{1}{2}\left(\frac{dU}{dm}\right)\,dx \;=\; E. \tag{27}
\end{aligned}
$$

On the other hand, by the definition of $U(m)$, it holds $(m_1 - m_2)^2 \lesssim |U(m_1) - U(m_2)|$, so that

$$
\begin{aligned}
\left|\fint_\Omega \chi(m - m^R)\,dx\right| \;\lesssim\; & \frac{1}{\delta}\fint_\Omega (m - m^R)^2\,dx + \delta\fint_\Omega \chi^2\,dx \\
\le\; & \frac{1}{\delta}\sup_{|y| \le R}\fint_\Omega (m(x) - m(x+y))^2\,dx + \delta \\
\lesssim\; & \frac{1}{\delta}\sup_{|y| \le R}\fint_\Omega |U(m(x)) - U(m(x+y))|\,dx + \delta \\
\lesssim\; & \frac{R}{\delta}\fint_\Omega |\nabla(U(m))|\,dx + \delta.
\end{aligned}
$$

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Thanks to the Modica–Mortola estimate (27), we obtain

$$\left| \fint_\Omega \chi(m - m^R)\, dx \right| \lesssim \frac{R}{\delta} E + \delta. \tag{28}$$

For the second term in (26), we argue as in [4]. We have for any admissible transfer plan π

$$\begin{aligned}
\fint_\Omega \chi m^R\, dx &= \fint_\Omega \chi^R m\, dx \\
&= \fint_\Omega \chi^R m_+\, dx - \fint_\Omega \chi^R m_-\, dx \\
&= \iint_{\Omega \times \Omega} \left(\chi^R(x) - \chi^R(y) \right) d\pi(x, y).
\end{aligned}$$

To estimate the last term, we must deal separately with transport over large and small distances:

$$\begin{aligned}
\left| \fint_\Omega \chi m^R\, dx \right| &\le \iint_{\{|x-y| \le r\}} |\chi^R(x) - \chi^R(y)|\, d\pi(x, y) \\
&\quad + \iint_{\{|x-y| < r\}} |\chi^R(x) - \chi^R(y)|\, d\pi(x, y).
\end{aligned} \tag{29}$$

For the small distances, we find

$$\begin{aligned}
&\left| \iint_{\{|x-y| \le r\}} |\chi^R(x) - \chi^R(y)|\, d\pi(x, y) \right| \\
&\le \ \sup |\nabla \chi^R| \iint_{\{|x-y| \le r\}} |x - y|\, d\pi(x, y) \ \lesssim \ \frac{r}{R}.
\end{aligned} \tag{30}$$

For large distances, we use the monotonicity of c:

$$\begin{aligned}
&\left| \iint_{\{|x-y| > r\}} |\chi^R(x) - \chi^R(y)|\, d\pi(x, y) \right| \\
&\le \ 2 \sup |\chi^R| \frac{1}{c(r)} \iint_{\Omega \times \Omega} c(|x - y|)\, d\pi(x, y) \ \lesssim \ \frac{d_c(m_+, m_-)}{c(r)}.
\end{aligned} \tag{31}$$

Substituting (30) and (31) in (29) we conclude

$$\left| \fint_\Omega \chi m^R\, dx \right| \lesssim \frac{r}{R} + \frac{d_c(m_+, m_-)}{c(r)}. \tag{32}$$

In view of (28) and (32), inequality (26) turns into

$$1 \lesssim \frac{RE}{\delta} + \frac{r}{R} + \frac{d_c(m_+, m_-)}{c(r)} + \delta.$$

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Choosing δ sufficiently small and optimizing in R yields

$$2\left(\frac{1}{c_0}\right)^{1/2} \leq (rE)^{1/2} + \frac{d_c(m_+, m_-)}{c(r)},$$

when we reintroduce some constant c_0. Choosing $r = \frac{1}{c_0 E}$, this becomes

$$\left(\frac{1}{c_0}\right)^{1/2} \leq \frac{d_c(m_+, m_-)}{c\left(\frac{1}{c_0 E}\right)},$$

which is the desired estimate of Proposition 3. $\qquad\square$

6.3 ODE Argument

In this section, we provide the proof of Proposition 4.

Proof of Proposition 4. By rescaling

$$E = \lambda^{-1/2}\hat{E}, \quad L = \hat{L}, \quad t = \lambda^{1/2}\hat{t},$$

we may w. l. o. g. assume that $\lambda = 1$. We set for abbreviation:

$$c(s) := \left\{ \begin{array}{ll} s & \text{for } s \leq 1 \\ 1 + \ln s & \text{for } s \geq 1 \end{array} \right\},$$

(which is our cost function in the definition of L with $\lambda = 1$,)

$$f(e) := \left\{ \begin{array}{ll} e & \text{for } e \leq 1 \\ e^2 & \text{for } e \geq 1 \end{array} \right\} = \max\{e^2, e\},$$

and

$$g(e) := c\left(\frac{1}{e}\right) = \left\{ \begin{array}{ll} 1 + \ln\frac{1}{e} & \text{for } e \leq 1 \\ \frac{1}{e} & \text{for } e \geq 1 \end{array} \right\}.$$

Notice that g and f are related by

$$g'(e) = \left\{ \begin{array}{ll} -\frac{1}{e} & \text{for } e \leq 1 \\ -\frac{1}{e^2} & \text{for } e \geq 1 \end{array} \right\} = -\frac{1}{f(e)}.$$

Hence, as long as $L(T) \geq 2L(0)$, we have

$$L(T) \leq 2(L(T) - L(0)) = 2\int_0^T \frac{dL}{dt}\,dt,$$

and thus, applying (17), (18) and the definition of g:

$$g(E(T)) \leq \tilde{C}\int_0^T \left(-\frac{dE}{dt}\right)^{1/2}\,dt,$$

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where $\tilde{C}$ denotes a generic constant whose value may change from line to line. W. l. o. g. we may assume that $\tilde{C} \geq 1$. With the help of the Cauchy–Schwarz inequality, this estimate turns into

$$g(E(T)) \leq \tilde{C} \left(\int_0^T \frac{1}{f(E)} \left(-\frac{dE}{dt} \right) dt \int_0^T f(E) \, dt \right)^{1/2}. \qquad (33)$$

By the relation between f and g, it is

$$\frac{1}{f(E)} \left(-\frac{dE}{dt} \right) = g'(E) \frac{dE}{dt} = \frac{d}{dt} g(E),$$

and thus

$$\int_0^T \frac{1}{f(E)} \left(-\frac{dE}{dt} \right) dt = g(E(T)) - g(E(0)) \leq g(E(T)),$$

so that (33) implies

$$g(E(T)) \leq \tilde{C} \left(g(E(T)) \int_0^T f(E) \, dt \right)^{1/2},$$

or equivalently

$$g(E(T)) \leq \tilde{C} \int_0^T f(E) \, dt.$$

We rewrite this estimate as an implicit ODE for $h(T) := \int_0^T f(E) \, dt$:

$$(g \circ f^{-1})(h'(T)) \leq \tilde{C} h(T).$$

Since g is a decreasing and f^{-1} an increasing function, the above inequality is equivalent to

$$h'(T) \geq (f \circ g^{-1})(\tilde{C} h(T)).$$

Let F be the antiderivative of $\frac{1}{f \circ g^{-1}}$ with $F(0) = 0$. An easy calculation shows

$$F(z) = \left\{ \begin{array}{ll} \frac{1}{3} z^3 & \text{for } z \leq 1 \\ \exp(z-1) - \frac{2}{3} & \text{for } z \geq 1 \end{array} \right\}.$$

Then the above inequality can be rewritten as

$$\frac{d}{dT} F(\tilde{C} h(T)) \geq \tilde{C},$$

which turns into

$$F(\tilde{C} h(T)) = F(\tilde{C} h(T)) - F(\tilde{C} h(0)) \geq \tilde{C} T \stackrel{\tilde{C} \geq 1}{\geq} T$$

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after integration. Since F is an increasing function, this can be paraphrased as

$$\tilde{C}h(T) \geq F^{-1}(T)$$
$$= \left\{ \begin{array}{ll} (3T)^{1/3} & \text{for } T \leq \frac{1}{3} \\ 1 + \ln\left(T + \frac{2}{3}\right) & \text{for } T \geq \frac{1}{3} \end{array} \right\}$$
$$\sim \int_0^T \min\{\left(\frac{1}{t^{1/3}}\right)^2, \frac{1}{t}\} \, dt.$$

By the definition of h, this turns into

$$\int_0^T \max\{E^2, E\} \, dt \geq \frac{1}{\tilde{C}} \int_0^T \min\{\left(\frac{1}{t^{1/3}}\right)^2, \frac{1}{t}\} \, dt. \tag{34}$$

We now argue for the case that $L(T) \leq 2L(0)$. By (18), we have

$$c\left(\frac{1}{E(T)}\right) \leq L(T) \leq 2L(0).$$

Since $2L(0) \leq 1$, it is $c\left(\frac{1}{E(T)}\right) \leq 1$, and thus $c\left(\frac{1}{E(T)}\right) = \frac{1}{E(T)}$. Consequently, the above estimate can be rewritten as

$$\frac{1}{E(T)} \leq L(T) \leq 2L(0).$$

Since $L(0) \leq T^{1/3}$, it is

$$\frac{1}{E(T)} \leq 2T^{1/3},$$

and since $E(t)$ is an decreasing function, this implies

$$E(t) \geq E(T) \geq \frac{1}{2T^{1/3}},$$

for all $0 \leq t \leq T$. We square both sides and integrate in time:

$$\int_0^T \max\{E^2, E\} \, dt \geq \int_0^T E^2 \, dt$$
$$\geq \frac{1}{4}T^{1/3}$$
$$= \frac{1}{12} \int_0^T \left(\frac{1}{t^{1/3}}\right)^2 \, dt$$
$$\geq \frac{1}{12} \int_0^T \min\{\left(\frac{1}{t^{1/3}}\right)^2, \frac{1}{t}\} \, dt. \tag{35}$$

It remains to combine (34) and (35) to conclude the proof of Proposition 4. $\qquad \square$

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7 Appendix A

Here we establish some properties of optimal transportation distances.

Lemma 10. *Let $c : \Omega \times \Omega \longrightarrow [0, \infty)$ be a metric on Ω. Then for all positive measures $\theta_1, \theta_2, \vartheta_1,$ and ϑ_2, such that $\theta_1(\Omega) = \vartheta_1(\Omega)$ and $\theta_2(\Omega) = \vartheta_2(\Omega)$,*

$$d_c(\theta_1, \vartheta_1) \leq d_c(\theta_1 + \theta_2, \vartheta_1 + \vartheta_2) + d_c(\theta_2, \vartheta_2).$$

Proof. By Kantorovich duality, cf. [29, Theorem 1.3]

$$d_c(\theta_1, \vartheta_1) = \sup_{\{\phi, \psi \mid \phi(x) + \psi(y) \leq c(x,y)\}} \fint_\Omega \phi \, d\theta_1 + \fint_\Omega \psi \, d\vartheta_1.$$

Furthermore it is enough to consider pairs such that ϕ and ψ are c-duals of each other, for instance

$$\psi = \phi^c \text{ and } \phi = \psi^c,$$

where the c-duals are defined as

$$\phi^c(y) = \inf_{x \in \Omega} \{c(x, y) - \phi(x)\} \quad \text{and} \quad \psi^c(x) = \inf_{y \in \Omega} \{c(x, y) - \psi(y)\}.$$

We claim that if $\phi = \psi^c$ for some ψ then for all $x_1, x_2 \in \Omega$

$$|\phi(x_1) - \phi(x_2)| \leq c(x_1, x_2). \tag{36}$$

To see this, note that by the definition of ψ^c, for any $\varepsilon > 0$ and $x_1, x_2 \in \Omega$ there exists x_1' such that

$$\phi(x_1) \geq c(x_1, x_1') - \psi(x_1') - \varepsilon.$$

Therefore

$$\phi(x_2) \leq c(x_2, x_1') - \psi(x_1') \leq c(x_2, x_1') - c(x_1', x_1) + \phi(x_1) + \varepsilon \leq c(x_1, x_2) + \phi(x_1) + \varepsilon.$$

Letting $\varepsilon \to 0$ and using symmetry in x_1, x_2 allows us to conclude (36). By similar arguments, based on (36), we prove that

$$-\phi(x) - \psi(y) \leq c(x, y).$$

We deduce that

$$\fint_\Omega \phi \, d\theta_1 + \fint_\Omega \psi \, d\vartheta_1$$

$$= \fint_\Omega \phi \, d(\theta_1 + \theta_2) + \fint_\Omega \psi \, d(\vartheta_1 + \vartheta_2) - \left(\fint_\Omega \phi \, d\theta_2 + \fint_\Omega \psi \, d\vartheta_2 \right)$$

$$\leq d_c(\theta_1 + \theta_2, \vartheta_1 + \vartheta_2) + \iint_{\Omega \times \Omega} c(x, y) \, d\pi_2(x, y).$$

It remains to optimize in ϕ, ψ, and π_2. $\qquad\square$

8 Appendix B

We conclude this article with a discussion on well-posedness and regularity of our dynamical system (4), (3) in three space dimensions. For simplicity, we set $\lambda = 1$ and drop all constants. (In particular, we lower the potential term in the Ginzburg–Landau energy (1) by the factor of $1/2$.). Again, we consider the periodic setting for convenience and we denote the period cell by Ω.

Proposition 11. *Let the initial configuration m_0 be of finite energy,*

$$E(m_0) \;<\; \infty.$$

Then there there is a unique smooth periodic solution (m, u, p) (p is unique up to an additive constant) of the system

$$
\begin{aligned}
\partial_t m - \Delta \mu + u \cdot \nabla m &= 0, & (37)\\
-\Delta u + \nabla p &= -m \nabla \mu, & (38)\\
\nabla \cdot u &= 0, & (39)\\
\int_\Omega u \, dx &= 0, & (40)
\end{aligned}
$$

with $m(t = 0) = m_0$, and where $\mu = -\Delta m - m + m^3$. Moreover

$$E(m(t)) \;\leq\; E(m_0) \quad \text{for any } t \geq 0.$$

Sketch of the proof. In view of (37)&(39), it is obvious that any sufficiently strong notion of solutions is mass preserving, and so are the weak or time-discrete solutions we introduce in the following. For convenience, we may therefore w.l.o.g. assume that

$$\int_\Omega m \, dx \;=\; \int_\Omega m_0 \, dx \;=\; 0.$$

This choice (and the periodic boundary conditions) simplifies the notation slightly when we use Sobolev's inequalities, since Sobolev norms reduce to the norms of the highest derivatives thanks to the Poincaré's inequality.

Step 1. Existence of time-discrete weak solutions. For convenience, we rewrite (38) as

$$-\Delta u + \nabla q \;=\; -\nabla \cdot (\nabla m \otimes \nabla m). \tag{41}$$

Indeed, this formula can be derived by absorbing gradient terms from the r. h. s. of (38) into the hydrodynamic pressure term, producing a new conservative field ∇q with

$$q \;=\; p - m\Delta m - \frac{1}{2}|\nabla m|^2 - \frac{1}{2}m^2 + \frac{3}{4}m^4. \tag{42}$$

Let $0 < h \ll 1$ denote the time step size. Given some $m \in H^{2,2}(\Omega)$, we first consider the Stokes equation in the formulation of equation (41), this means,

$$-\Delta u + \nabla q \ = \ -\nabla \cdot (\nabla m \otimes \nabla m), \tag{43}$$

$$\nabla \cdot u \ = \ 0, \tag{44}$$

$$\int_\Omega u \, dx \ = \ 0. \tag{45}$$

Notice that the r. h. s. of (43) is in $H^{-1,2}(\Omega)$, and we can easily derive the a priori estimate

$$\int_\Omega |\nabla u|^2 \, dx \ \lesssim \ \int_\Omega |\nabla m|^4 \, dx \ \lesssim \ \left(\int_\Omega |\nabla^2 m|^2 \, dx \right)^2. \tag{46}$$

The second estimate above is due to Hölder's inequality and Sobolev embedding in three space dimensions in the sense that $L^6(\Omega) \subset L^4(\Omega)$ and $H^{1,2}(\Omega) \subset L^6(\Omega)$, respectively. From standard theory for the Stokes equation, cf. [12, Chapter IV], we deduce existence and uniqueness of a solution $u \in H^{1,2}(\Omega)$, $q \in L^2(\Omega)$ to (43)–(45). (In fact, q is unique up to an additive constant.)

On the other hand, given a divergence-free velocity field $u \in H^{1,2}(\Omega)$, we find a unique solution $m \in H^{3,2}(\Omega)$ to the elliptic equation

$$\Delta^2 m + u \cdot \nabla m \ = \ \Delta(m^3 - m) + \frac{1}{h}(m_0 - m). \tag{47}$$

The argument involves a fixed point argument in the nonlinearity such as the a priori bounds

$$\frac{1}{h} \int_\Omega m^2 \, dx + \int_\Omega |\nabla^2 m|^2 \, dx \ \lesssim \ \frac{1}{h} \int_\Omega m_0^2 \, dx, \tag{48}$$

and

$$\frac{1}{h} \int_\Omega |\nabla m|^2 \, dx + \int_\Omega |\nabla^3 m|^2 \, dx$$
$$\lesssim \ 1 + \left(\int_\Omega |\nabla u|^2 \, dx \right)^2 + \left(\frac{1}{h} \int_\Omega m_0^2 \, dx \right)^3 + \frac{1}{h} \int_\Omega |\nabla m_0|^2 \, dx. \tag{49}$$

Indeed, testing (47) with m and integrating by parts yields

$$\frac{1}{h} \int_\Omega m^2 \, dx + \int_\Omega |\nabla^2 m|^2 \, dx$$
$$= \ -3 \int_\Omega m^2 |\nabla m|^2 \, dx - \int_\Omega m \Delta m \, dx + \frac{1}{h} \int_\Omega m_0 m \, dx.$$

It remains to apply Hölder's and Young's inequality together with the fact that $h \ll 1$ to deduce (48). For (49), we test (47) with $-\Delta m$. Integration by parts gives

$$\frac{1}{h} \int_\Omega |\nabla m|^2 \, dx + \int_\Omega |\nabla^3 m|^2 \, dx$$
$$= \ -\int_\Omega \nabla \Delta m \cdot u m \, dx + 3 \int_\Omega \nabla \Delta m \cdot m^2 \nabla m \, dx$$
$$+ \int_\Omega |\nabla^2 m|^2 \, dx + \frac{1}{h} \int_\Omega \nabla m \cdot \nabla m_0 \, dx.$$

The terms coming from the convective part and the nonlinear part of the equation can both be estimated using Hölder's and Sobolev's inequality. We invoke (48) and Young's inequality to get (49).

Existence of solutions of the coupled system (43)–(45)&(47) follows again by a fixed point argument. To make it precise, we define the (nonlinear) mapping

$$A : H^{2,2}(\Omega) \to H^{3,2}(\Omega) \subset H^{2,2}(\Omega)$$

in the following way: Given $\tilde{m} \in H^{2,2}(\Omega)$, we solve the Stokes equation (43)–(45) and find $u \in H^{1,2}(\Omega)$, $q \in L^2(\Omega)$. Inserting u in (47) in turn yields a solution $m \in H^{3,2}(\Omega)$. We set $A\tilde{m} = m$. Thanks to (49), it is readily verified that this mapping is compact. Continuity of A is less obvious. In fact, we first have to argue that the solution of the Stokes equation u depends continuously on $\tilde{m}$. This property follows from the continuity of the nonlinear r. h. s. of (43) on $\tilde{m}$ via a priori estimates similar to (46). Moreover, the continuous dependence of m on $\tilde{m}$ and u uses estimates in the spirit of (49). We leave details to the interested reader. Finally, the set

$$\{m \in H^{2,2}(\Omega) \,|\, m = \sigma Am \text{ for some } 0 \leq \sigma \leq 1\}$$

is bounded. Indeed, if $m = \sigma Am$ for some $0 < \sigma \leq 1$, we may replace m in (47) by $\frac{1}{\sigma}m$ and find that (48) still holds true, independently of σ. Now, Schaefer's Fixed Point Theorem, cf. [10, p. 502], states that A has a fixed point $m_h^{(1)}$ in $H^{3,2}(\Omega)$. Here, subscript "h" emphasizes the dependence on the time step size, and superscript "(1)" marks the solution on the first time step. We may iterate this scheme for fixed h by starting with initial configuration $m_h^{(1)}$ and proving existence of solutions on the next time step. In this way, we obtain a sequence of solutions

$$m_h^{(k)} \in H^{3,2}(\Omega), \ u_h^{(k)} \in H^{1,2}(\Omega), \ \text{and} \ q_h^{(k)} \in L^2(\Omega)$$

to (43)–(45)&(47) with m_0 replaced by $m_h^{(k-1)}$ for each $k \in \mathbb{N}$. In view of the integrability of $m_h^{(k)}$ and $q_h^{(k)}$, we deduce from (42) that we can go back to the formulation (38) of the Stokes equation with $p_h^{(k)} \in L^2(\Omega)$ and $\mu_h^{(k)} = -\Delta m_h^{(k)} - m_h^{(k)} + (m_h^{(k)})^3 \in H^{1,2}(\Omega)$.

Step 2. Existence of weak solutions. In this step, we establish the existence of weak solutions to (37)–(40). For this purpose, we construct $m_h(t, \cdot)$, $u_h(t, \cdot)$, and $\mu_h(t, \cdot)$ as piecewise constant interpolations, and $\tilde{m}_h(t, \cdot)$ as a piecewise affine interpolation on the steps $((k-1)h, kh]$. For instance,

$$m_h(t, \cdot) = m_h^{(k)},$$
$$\tilde{m}_h(t, \cdot) = \frac{t - (k-1)h}{h} m_h^{(k)} + \frac{kh - t}{h} m_h^{(k-1)},$$

for any $t \in ((k-1)h, kh]$. These functions satisfy weakly the following system (the pressure drops out in the weak formulation, so that we do not need an explicit

construction):

$$
\begin{aligned}
\partial_t \tilde{m}_h - \Delta \mu_h + u_h \cdot \nabla m_h &= 0, \\
\nabla \cdot u_h &= 0, \\
-\Delta u_h + \nabla p_h &= -m_h \nabla \mu_h, \\
\int_\Omega u_h \, dx &= 0.
\end{aligned}
$$

Testing the convection-diffusion equation with m_h and μ_h, respectively, we have the new a priori estimates:

$$
\sup_{0 \le t \le T} \int_\Omega m_h^2 \, dx + \int_0^T \int_\Omega |\nabla^2 m_h|^2 \, dx dt \lesssim \int_0^T \int_\Omega m_h^2 \, dx dt + \int_\Omega m_0^2 \, dx, \qquad (50)
$$

and

$$
\sup_{0 \le t \le T} E(m_h) + \int_0^T \int_\Omega |\nabla \mu_h|^2 \, dx dt \lesssim E(m_0), \qquad (51)
$$

and thus, by convexity,

$$
\sup_{0 \le t \le T} \int_\Omega \tilde{m}_h^2 \, dx + \int_0^T \int_\Omega |\nabla^2 \tilde{m}_h|^2 \, dx dt \lesssim \int_0^T \int_\Omega m_h^2 \, dx dt + \int_\Omega m_0^2 \, dx, \qquad (52)
$$

and

$$
\sup_{0 \le t \le T} \int_\Omega |\nabla \tilde{m}_h|^2 \, dx \le E(m_0).
$$

The control of the velocity field u_h in the Stokes equation uses the Sobolev's inequality and the dissipation estimate (51):

$$
\int_0^T \int_\Omega |\nabla u_h|^2 \, dx dt \lesssim \sup_{0 \le t \le T} \left(\int_\Omega |\nabla m_h|^2 \, dx \right)^2 \int_0^T \int_\Omega |\nabla \mu_h|^2 \, dx dt \lesssim E(m_0).
$$
(53)

Finally, the time derivative of $\tilde{m}_h$ can be bounded in a similar way:

$$
\begin{aligned}
&\int_0^T \|\partial_t \tilde{m}_h\|_{H^{-1,2}}^2 \, dt \\
&\lesssim \int_0^T \int_\Omega |\nabla \mu_h|^2 \, dx dt + \sup_{0 \le t \le T} \int_\Omega |\nabla m_h|^2 \, dx \int_0^T \int_\Omega |\nabla u_h|^2 \, dx dt \\
&\lesssim 1 + E(m_0)^2.
\end{aligned}
$$

This is enough to ensure compactness. More precisely, we find for T sufficiently small:

- $m \in L^\infty(0, T; H^{1,2}(\Omega)) \cap L^2(0, T; H^{3,2}(\Omega))$,

- $\tilde{m} \in L^\infty(0, T; H^{1,2}(\Omega)) \cap L^2(0, T; H^{2,2}(\Omega))$,

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- $\mu \in L^2(0, T; H^{1,2}(\Omega))$,

- $u \in L^2(0, T; H^{1,2}(\Omega))$,

and — up to subsequences — convergence of m_h, $\tilde{m}_h$, μ_h, and u_h to m, $\tilde{m}$, μ, and u, respectively, in the corresponding weak topologies. Moreover, since $\tilde{m}_h$ is bounded in $L^2(0, T; H^{2,2}(\Omega))$ with time derivative bounded in $L^2(0, T; H^{-1,2}(\Omega))$, it follows from the Aubin–Lions Lemma that $\tilde{m}_h \to \tilde{m}$ strongly in $L^2(0, T; H^{1,2}(\Omega))$.

We have to show that

$$m = \tilde{m}.$$

This fact follows from the estimate

$$\left| \int_0^T \int_\Omega (m_h - \tilde{m}_h)\varphi \, dxdt \right| \leq C_0 h \left(\int_0^T \int_\Omega |\nabla \varphi|^2 \, dxdt \right)^{1/2}, \qquad (54)$$

where C_0 denotes a generic constant depended on the initial energy $E(m_0)$, and φ varies in the class of C^1 functions which are periodic in space. In order to derive (54), we appeal to definition of m_h and $\tilde{m}_h$, and rewrite the term on the left as

$$\int_0^T \int_\Omega (m_h - \tilde{m}_h)\varphi \, dxdt = \sum_{k=1}^K \int_\Omega \frac{1}{h} \left(m_h^{(k)} - m_h^{(k-1)} \right) \zeta_h^{(k)} \, dx,$$

where $K \in \mathbb{N}$ is such that $T = hK$ and $\zeta_h^{(k)} = \int_{(k-1)h}^{kh} (kh - t)\varphi \, dt$. Observing that $(m_h^{(k)} - m_h^{(k-1)})/h = \partial_t \tilde{m}_h = \Delta \mu_h^{(k)} - \nabla \cdot \left(m_h^{(k)} u_h^{(k)} \right)$, we obtain after integration by parts and using the Cauchy–Schwarz inequality:

$$\int_0^T \int_\Omega (m_h - \tilde{m}_h)\varphi \, dxdt$$

$$\leq \left(\sum_{k=1}^K \int_\Omega |\nabla \mu_h^{(k)}|^2 \, dx \right)^{1/2} \left(\sum_{k=1}^K \int_\Omega |\nabla \zeta_h^{(k)}|^2 \, dx \right)^{1/2}$$

$$+ \left(\sum_{k=1}^K \int_\Omega |m_h^{(k)} u_h^{(k)}|^2 \, dx \right)^{1/2} \left(\sum_{k=1}^K \int_\Omega |\nabla \zeta_h^{(k)}|^2 \, dx \right)^{1/2}.$$

Applying Hölder's and Sobolev's inequality to the convective term, expressing $m_h^{(k)}$ and $u_h^{(k)}$ by m_h and u_h, respectively, invoking the estimates (51)&(53) and using the definition of $\zeta_h^{(k)}$ we finally derive (54).

In fact, applying estimate (54) to $\varphi = m_h - \tilde{m}_h$, and using the a priori estimates (50) and (52) to bound the resulting $\nabla \varphi$-terms uniformly by the initial data, we see that $m_h - \tilde{m}_h$ converges strongly to zero, and thus, by triangle inequality, m_h converges strongly to m.

We now derive the limiting equation. Concerning the limit in the Cahn–Hilliard equation, we only have to check that

$$\mu = -\Delta m - m + m^3.$$

This means, we have to discuss convergence of the cubic term. In fact, writing $m^3 - m_h^3 = (m - m_h)(m^2 + mm_h + m_h^2)$ we see that the cubic term converges thanks to the strong convergence of m_h and since m_h^2 is uniformly bounded via Sobolev's inequality and estimate (51).

It remains to check the limit in the Stokes equation, this means, we have to prove convergence of the nonlinear r. h. s. In fact, for this purpose we invoke a standard argument, noting that $m_h \nabla \mu_h$ is a product of a strong and a weak converging sequence.

To summarize, we have shown existence of weak solutions

$$m \in L^\infty(0,T; H^{1,2}(\Omega)) \cap L^2(0,T; H^{3,2}(\Omega)), \; u \in L^2(0,T; H^{1,2}(\Omega))$$

of (37)–(40) with

$$\partial_t m \in L^2(0,T, H^{-1,2}(\Omega)) \quad \text{and} \quad m(t=0) = m_0.$$

It is easy to check that the a priori energy inequality holds for any $T > 0$:

$$E(m(T)) + \int_0^T \int_\Omega |\nabla \mu|^2 \, dxdt + \int_0^T \int_\Omega |\nabla u|^2 \, dxdt \; \leq \; E(m_0). \tag{55}$$

In particular, weak solutions exist globally in time.

Step 3: Uniqueness of weak solutions. As for the classical Cahn–Hilliard equation, we show uniqueness via a contraction principle for the $H^{-1,2}$ distance. Let (m_1, u_1, p_1) and (m_2, u_2, p_2) be two solutions of (37)–(40) with $m_1(t = 0) = m_2(t = 0) = m_0$. It suffices to prove uniqueness in m, since, given a fixed r. h. s., solutions to the Stokes problem (38)–(40) are well-known to be unique (p is unique up to an additive constant). Recall that

$$\int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx \; = \; \int_\Omega |\nabla(\varphi_1 - \varphi_2)|^2 \, dx,$$

when $\varphi_1, \; \varphi_2 : \Omega \mapsto \mathbb{R}$ are given by

$$-\Delta \varphi_i \; = \; m_i, \quad \int_\Omega \varphi_i \, dx \; = \; 0 \qquad \text{for } i = 1, 2.$$

A direct calculation based on the evolution equation (37) yields

$$\frac{d}{dt} \frac{1}{2} \int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx$$

$$= \; - \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx + \int_\Omega (m_1 - m_2)^2 \, dx$$

$$- \int_\Omega (m_1 - m_2)(m_1^3 - m_2^3) \, dx$$

$$- \int_\Omega (\varphi_1 - \varphi_2)(u_1 \cdot \nabla m_1 - u_2 \cdot \nabla m_2) \, dx. \tag{56}$$

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In the following, we argue that we can estimate the terms on the right such that

$$\frac{d}{dt} \int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx \;\lesssim\; I(t) \int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx, \qquad (57)$$

where $I(t)$ is a integrable function. Notice that this estimate immediately guarantees uniqueness. Indeed, by a Gronwall argument, we infer that

$$\int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx \;=\; 0,$$

because of $m_1(t = 0) = m_2(t = 0)$, and thus $m_1 = m_2$.

We now turn to the proof of (57). The L^2 term on the r. h. s. of identity (56) can likewise be absorbed into $H^{1,2}$ and $H^{-1,2}$ terms by duality. Furthermore, we have

$$-\int_\Omega (m_1 - m_2)(m_1^3 - m_2^3) \, dx \;=\; -\int_\Omega (m_1 - m_2)^2 (m_1^2 + m_1 m_2 + m_2^2) \, dx \;\leq\; 0.$$

Compared to the classical Cahn–Hilliard equation, it is the last term in (56), which arises due to the additional convection term in (37). It can be split into two parts according to

$$\int_\Omega (\varphi_1 - \varphi_2)(u_1 \cdot \nabla m_1 - u_2 \cdot \nabla m_2) \, dx$$
$$= \int_\Omega (\varphi_1 - \varphi_2)(u_1 - u_2) \cdot \nabla m_1 \, dx + \int_\Omega (\varphi_1 - \varphi_2)u_2 \cdot \nabla(m_1 - m_2) \, dx. \quad (58)$$

The estimate for the second term is obvious, using Hölder's, Sobolev's and Young's inequality:

$$\int_\Omega (\varphi_1 - \varphi_2)u_2 \cdot \nabla(m_1 - m_2) \, dx$$
$$\lesssim \left(\int_\Omega |\nabla u_2|^2 \, dx \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx \int_\Omega |\nabla(\varphi_1 - \varphi_2)|^2 \, dx \right)^{1/2}$$
$$\lesssim \varepsilon \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx + \int_\Omega |\nabla u_2|^2 \, dx \int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx,$$

where ε is some small number. Observe that thanks to the dissipation inequality (55), the fluid velocity term is integrable and contributes therefore to $I(t)$. In preparation of the estimate of the first term in (58), we show that

$$\int_\Omega |\nabla(u_1 - u_2)|^2 \, dx \;\lesssim\; I(t) \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx. \qquad (59)$$

Indeed, using (41) again, we have

$$\int_\Omega |\nabla(u_1 - u_2)|^2 \, dx \;\lesssim\; \int_\Omega |\nabla m_1 \otimes \nabla m_1 - \nabla m_2 \otimes \nabla m_2|^2 \, dx.$$

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Writing $\nabla m_1 \otimes \nabla m_1 - \nabla m_2 \otimes \nabla m_2 = \nabla m_1 \otimes (\nabla m_1 - \nabla m_2) + (\nabla m_1 - \nabla m_2) \otimes \nabla m_2$, we further estimate

$$\int_\Omega |\nabla(u_1 - u_2)|^2 \, dx \;\lesssim\; \left(\sup_\Omega |\nabla m_1| + \sup_\Omega |\nabla m_2| \right)^2 \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx,$$

which by the Sobolev embedding $H^{2,2}(\Omega) \subset L^\infty(\Omega)$ in three dimensions yields

$$\int_\Omega |\nabla(u_1 - u_2)|^2 \, dx \;\lesssim\; \int_\Omega \left(|\nabla^3 m_1|^2 + |\nabla^3 m_2|^2 \right) dx \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx.$$

Observe that maximal regularity for the Laplace equation $-\Delta m = \mu + m - m^3$ and Hölder's and Sobolev's inequality imply

$$\int_\Omega |\nabla^3 m|^2 \, dx \;\lesssim\; \int_\Omega |\nabla \mu|^2 \, dx + \int_\Omega |\nabla m|^2 \, dx + \int_\Omega m^4 |\nabla m|^2 \, dx$$

$$\lesssim\; \int_\Omega |\nabla \mu|^2 \, dx + \int_\Omega |\nabla m|^2 \, dx + \left(\int_\Omega |\nabla m|^2 \, dx \right)^2 \int_\Omega |\nabla^2 m|^2 \, dx,$$

and thus by interpolation and Young's inequality

$$\int_\Omega |\nabla^3 m|^2 \, dx \;\lesssim\; \int_\Omega |\nabla \mu|^2 \, dx + \left(\int_\Omega |\nabla m|^2 \, dx \right)^5 + 1.$$

In view of (55), this reads

$$\int_\Omega |\nabla^3 m|^2 \, dx \;\lesssim\; I(t),$$

and therefore (59) holds. It follows via Hölder's, Sobolev's and Young's inequality that

$$\int_\Omega (\varphi_1 - \varphi_2)(u_1 - u_2) \cdot \nabla m_1 \, dx$$

$$\lesssim\; \left(\int_\Omega |\nabla(\varphi_1 - \varphi_2)|^2 \, dx \int_\Omega |\nabla(u_1 - u_2)|^2 \, dx \int_\Omega |\nabla m_1|^2 \, dx \right)^{1/2}$$

$$\lesssim\; \varepsilon \int_\Omega |\nabla(m_1 - m_2)|^2 \, dx + I(t) \int_\Omega ||\nabla|^{-1}(m_1 - m_2)|^2 \, dx,$$

where, again, ε is some small number. Collecting the above estimates and choosing ε sufficiently small, we deduce (57).

Step 4. Regularity of solutions. We only sketch the first steps in proving regularity of weak solutions. To show that solutions are indeed smooth, the arguments can be iterated. To shorten the notation, we refrain from writing time interval and domain dependence in our notion of function spaces.

Regularity in m is based on the maximal regularity estimate

$$\int_0^T \left(\int_\Omega |\partial_t m|^r + |\nabla^4 m|^r \, dx \right)^{s/r} dt \;\lesssim\; \int_0^T \left(\int_\Omega |\partial_t m + \Delta^2 m|^r \, dx \right)^{s/r} dt, \quad (60)$$

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provided that $m(t = 0) = 0$, and where $1 < r, s < \infty$ and $T > 0$, cf. [9, p. 102, Thm 8.2]. Concerning regularity in u, we invoke the regularity estimate

$$\int_\Omega |\nabla^2 u|^r + |\nabla p|^r \, dx \lesssim \int_\Omega |-\Delta u + \nabla p|^r \, dx, \tag{61}$$

where $1 < r < \infty$, cf. [12, Chapter IV].

We start with the observation, that the r. h. s. of (38), $-m\nabla\mu$, is bounded in $L^2(L^{3/2})$, so that (61) yields $u \in L^2(H^{2,3/2}) \subset L^2(H^{1,3})$. This in turn guarantees that the convection term in the Cahn–Hilliard equation is bounded: $u \cdot \nabla m \in L^2(L^r)$ for any $r < 2$. In particular $\partial_t(tm) + \Delta^2(tm) \in L^2(L^r)$ for any $r < 2$, so that (60) implies that $m \in H^{1,2}(L^r) \cap L^2(H^{4,r})$ for any $r < 2$. We can use this fact again to show that $u \in L^2(H^{2,2})$, and the above regularity for m upgrades to $r = 2$. On the other hand, straightforward estimations show that the nonlinear terms in the Cahn–Hilliard equation are in fact in $L^{3/2}(H^{1,2})$, so that we find $m \in H^{1,3/2}(H^{1,2}) \cap L^{3/2}(H^{5,2})$ by (60). In turn, this new insight can be used to infer $u \in L^{3/2}(H^{3,2})$ from (61). The discussion has to be continued in the same spirit. Once the regularity in space is pushed far enough, regularity in time can be considered (also for u). In the end, we obtain smooth solutions in space and time:

$$m, \ u, \ p \in C^\infty((0, \infty) \times \Omega).$$

This concludes the sketch of the proof of Proposition 11. $\qquad\square$

Acknowledgments

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