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Linking Anisotropic Sharp and Diffuse Surface Motion Laws via Gradient Flows

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We compare four surface motion laws for sharp surfaces with their diffuse interface counterparts by means of gradient flows on corresponding energy functionals. The energy functionals can be defined to give the same dependence on normal direction for the energy of sharp plane surfaces as for their diffuse counterparts. The anisotropy of the kinetics can be incorporated into the inner product without affecting the energy functional.

KEY WORDS: Gradient flows; inner products; morphology; motion by curvature; motion by Laplacian of curvature; parabolic partial differential equations; surface diffusion; weighted mean curvature; Cahn–Hilliard equation; Allen–Cahn equation; sharp interfaces; diffuse interfaces; anisotropy; phase field.

1. INTRODUCTION

This paper shows that two different types of equations describing motion of interfaces can be linked as gradient flows under appropriate inner products: on one hand the diffuse interfacial solutions to the partial differential equations (PDE), known as Allen–Cahn, Cahn–Hilliard, and viscous Cahn–Hilliard, and on the other hand equations for the movement of mathematical (sharp) surfaces, such as motion by mean curvature, motion by the negative Laplacian of mean curvature, a consequence of surface diffusion,[1] and a new surface motion law involving both interface kinetics and surface diffusion.[2] Various previously known pieces of these linkages are summarized here, new linkages are established, and anisotropy is introduced.

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A formulation of a gradient flow has several ingredients: a description of the free energy as a functional of the state of the system, an inner product which describes how "far apart" two states of the system are, and then an association of a gradient, a vector in function space, to the first variation of the free energy. It will be shown that prescribed kinetic factors, even anisotropic ones, can be built into the inner product, so that two states are "further apart" if they are separated by motions that are limited by slower kinetics.

Formal asymptotics have been carried out for some of the PDEs to link the flows to corresponding sharp interface motions, but actual convergence of solutions has been shown only in the simplest of the cases. Little mathematical investigation has been carried out for any of the sharp interface models except motion by mean curvature. Possible mathematical approaches to solving both sharp interface and diffuse interface formulations are discussed in ref. 2, which also contains an extensive bibliography.

2. THE FREE ENERGY FUNCTIONALS

In the sharp interface models, the state of the system is described by a surface S, and the free energy of the system is the surface energy of S, assumed to be the boundary of a region which has an exterior unit normal $\mathbf{n}_S(\mathbf{x})$ at almost every point $\mathbf{x}$ in S, and this surface energy is given by

$$E(S) = \int_{\mathbf{x} \in S} \gamma(\mathbf{n}_S(\mathbf{x}))\, dA$$

Here dA refers to integration with respect to 2-dimensional area (in a 3-dimensional system) (precisely, Hausdorff measure $\mathcal{H}^2$), and γ is a given phenomenological function of unit normal vectors, the energy per unit area of surface with that normal direction. We thus assume γ is a given positive Borel-measurable function on the unit sphere, and extend it by positive homogeneity of degree one to a function on all vectors in R^3 by defining $\gamma(r\mathbf{n}) = r\gamma(\mathbf{n})$ for all $r \geqslant 0$.

For diffuse interfaces, the state of the system is described by an order parameter or a composition u, defined in a region Ω (which might be all space R^3), and the free energy of the system is given by a functional

$$E(u) = \int_{\mathbf{x} \in \Omega} \{\tfrac{1}{2}\varepsilon^2 [\Gamma(\nabla u/|\nabla u|)]^2\, |\nabla u|^2 + F(u)\}\, dV$$

where dV denotes volume integration (i.e., integration with respect to Lebesgue 3-dimensional measure). Here Γ is a given (phenomenological) function on unit vectors $\nabla u/|\nabla u|$, and F is a given phenomenological func-

tion with two equal minima, which we take to be zero. Two examples of such functions with minima at ± 1 are $F(u) = (1-u^2)^2$ and $F(u) = 1-u^2$ for $|u| \leqslant 1$ and ∞ otherwise. Another family of such functions is given in ref. 2. If we homogeneously extend Γ to all vectors, $\Gamma(\mathbf{p}) = |\mathbf{p}|\, \Gamma(\mathbf{p}/|\mathbf{p}|)$, the energy functional of the system becomes

$$\int_\Omega \{\tfrac{1}{2}\varepsilon^2[\Gamma(\nabla u)]^2 + F(u)\}\, dV$$

When u in most of space has taken on values which are close to those locally minimizing the integral of F [that is, values close to u_a or u_b, the values for which $F(u_a) = 0 = F(u_b)$], then the "surfaces" are the regions where u changes from being close to one minimizer to being close to the other, and ε is a parameter which turns out to be related to the thickness of these transitional regions. We can identify $\varepsilon\Gamma$ as proportional to $\gamma(\mathbf{n})$ by considering a single planar "surface" in which u varies only with x, the distance in the direction of $\mathbf{n}$, so that the gradient of u is everywhere parallel to $\mathbf{n}$, and in which the energy is minimized. In this case we have $\frac{1}{2}\varepsilon^2[\Gamma(du/dx)]^2 = F(u)$. (This follows from the fact that the integrand does not depend explicitly on x, leading to the existence of a first integral of the variational derivative with a constant of integration that is zero by our assumption of zero minimum values for F.) Using the substitution $dx = \{\Gamma\in/[2F(u)]^{1/2}\}\, du$, this leads to the energy per unit area perpendicular to $\mathbf{n}$ being $\sqrt{2}\,\varepsilon[\int_{u_a}^{u_b}\sqrt{F}\, du]\,\Gamma(\mathbf{n})$. Thus if we identify $\varepsilon\Gamma$ with γ, up to a constant of proportionality determined by F only, diffuse and sharp interface models will have the same orientation dependence of surface energy. (See refs. 4–6 for a less general form of the same statement, and its use in computer simulations.) Note that if $|\Gamma(\nabla u)|^2 = \nabla u \cdot \Gamma_0 \nabla u$ for some positive-definite symmetric matrix Γ_0, then the surface energy is "ellipsoidal."[7]

A mathematical constraint which it might seem necessary to impose on Γ is that the Euler–Lagrange equation associated to the energy functional (discussed below) be elliptic. We have observed, however, that layered interfaces corresponding to diffuse interfaces with sharp corners can occur in the nonelliptic case, and in fact diffuse analogs to varifolds can also occur.[20] Thus the identification of Γ with the sharp interface γ remains valid.

3. MOTION LAWS

We consider four sharp-interface motion laws and four diffuse-interface laws; each of these decreases the total energy of the system, and the

latter three in the sharp case are volume-preserving and in the diffuse case are order-parameter-preserving. The first sharp-interface law is motion by mean curvature,

$$v(\mathbf{x}) = \kappa(\mathbf{x}) \tag{1i}$$

Its anisotropic version is motion by weighted mean curvature,

$$v(\mathbf{x}) = M(\mathbf{n}_S(\mathbf{x}))\,\kappa_\gamma(\mathbf{x}) \tag{1a}$$

Here and throughout this paper, v will represent a normal velocity defined at almost all points $\mathbf{x}$ on a surface S, M is the mobility, a given positive continuous function of normal direction, and $\kappa_\gamma(\mathbf{x})$ will denote the weighted mean curvature of S at $\mathbf{x}$ corresponding to the given surface free energy function $\gamma(\mathbf{p})$. For several equivalent definitions of κ_γ for this and other cases see ref. 3, in which κ_γ is called weighted mean curvature; see also ref. 2. When $\gamma(\mathbf{p}) = |\mathbf{p}|$ (the isotropic case, in which the energy is proportional to area), $\kappa_\gamma(\mathbf{x})$ is the mean curvature $\kappa(\mathbf{x})$ of S at $\mathbf{x}$, defined to be $\kappa_1 + \kappa_2$, where κ_1 and κ_2 are the principal curvatures of S at $\mathbf{x}$. [The sign convention is such that the surface of a positively oriented ball (with outward-pointing $\mathbf{n}$) has negative mean curvature.] When γ is an arbitrary C^2 convex function, $\kappa_\gamma(\mathbf{x}) = a_1\kappa_1 + a_2\kappa_2$, where a_i is the second partial derivative of γ in the ith principal direction, evaluated at $\mathbf{n}_S(\mathbf{x})$. Equivalently, it is the surface divergence of the vector function $\nabla\gamma(\mathbf{p})|_{\mathbf{p}=\mathbf{n}_S(\mathbf{x})}$. Note that although $M\gamma$ does not appear explicitly in Eq. (1i), it is there as an implicit constant 1 (perhaps as a result of rescaling distance or time), with units of distance^2/time. In general M is an anisotropic phenomenological kinetic parameter.

In its isotropic form the second sharp-interface law is

$$v(\mathbf{x}) = \kappa(\mathbf{x}) - [\text{Area}(S)]^{-1} \int_{\mathbf{y}\in S} \kappa(\mathbf{y})\, d\mathbf{y} \tag{2i}$$

and in its anisotropic form is

$$v(\mathbf{x}) = M(\mathbf{n}_S(\mathbf{x}))[\kappa_\gamma(\mathbf{x}) - \kappa_{av}] \tag{2a}$$

where

$$\kappa_{av} = \int_S M\kappa_\gamma\, dA \Big/ \int_S M\, dA$$

This is constrained motion by mean curvature, the constraint being that of keeping the volume enclosed by S constant.

The third sharp-interface law is, in the isotropic case,

$$v(\mathbf{x}) = -\Delta_S \kappa(\mathbf{x}) \tag{3i}$$

Here Δ_S denotes the surface Laplacian. This is the law derived by Mullins for surface diffusion. The fully anisotropic version of this law is

$$v = \nabla_S \cdot D \nabla_S (\kappa_\gamma) \tag{3a}$$

The surface diffusion coefficient D is also an anisotropic phenomenological parameter. Again, note that although neither D nor γ appears explicitly in Eq. (3i), they are there implicitly with values 1.

The fourth sharp-interface law is a new one we proposed recently. It introduces attachment and detachment kinetics (though M) into the surface diffusion law, and in its isotropic form can be written as either

$$v = \left(\frac{1}{M}\Delta_S - \frac{1}{D}\right)^{-1} \Delta_S \kappa \tag{4i}$$

or

$$v = \Delta_S \left(\frac{1}{M}\Delta_S - \frac{1}{D}\right)^{-1} \kappa \tag{4i'}$$

The anisotropic versions are

$$v = M(\nabla_S \cdot D \nabla_S - M)^{-1} \nabla_S \cdot D \nabla_S \kappa_\gamma \tag{4a}$$

and

$$v = (\nabla_S \cdot D \nabla_S)\left[(\nabla_S \cdot D \nabla_S - M)^{-1} (M \kappa_\gamma)\right] \tag{4a'}$$

These equations reduce to the second and third cases in the limit of small D and M, respectively. In the isotropic case, questions concerning the existence of the inverse operators reduce to the elementary fact that all eigenvalues of the Laplacian on a compact manifold, when restricted to the subspace of functions of average 0, are negative, just as on the circle $(d^2/d^2\theta - \lambda)u = 0$, only for nonpositive λ, and only for negative λ when the average of u is zero. Thus the operator $\Delta_S - M/D$ is invertible, at least on a suitable space of functions. In the anisotropic case, given that M must be positive, the operator $(\nabla_S \cdot D \nabla_S - M)$ should likewise be invertible on a suitable space of functions, at least under some set of assumptions on D and M; these hypotheses have not yet been sought.

For each of these four sharp-interface motion laws there are corresponding diffuse interface laws. We consider the isotropic laws first. Corresponding to motion by mean curvature (1i), we have

$$\frac{\partial u}{\partial t} = \varepsilon\, \Delta u - \frac{F'(u)}{\varepsilon} \tag{5i}$$

which is the Landau–Ginzburg or Allen–Cahn equation. A similar equation that conserves the integral of u and corresponds to constrained motion by mean curvature (2i) is

$$\frac{\partial u}{\partial t} = \varepsilon \Delta u - \frac{F'(u) - \text{const}}{\varepsilon} \tag{6i}$$

where the constant is $\int_\Omega F'(u)\, dV / \int_\Omega 1\, dV$. In a real physical situation there may be a kinetic coefficient which could have a dependence on u in these equations. Suitably normalized it would result in replacing $\partial u/\partial t$ with $[1/\mathcal{M}(u)]\, \partial u/\partial t$. This change has little effect on the results according to ref. 8. Corresponding to surface diffusion without attachment kinetics (3i), we have

$$\frac{\partial u}{\partial t} = \nabla \cdot B(u)\, \nabla \left(-\varepsilon\, \Delta u + \frac{F'(u)}{\varepsilon} \right) \tag{7i}$$

which is the Cahn–Hilliard equation with a u-dependent diffusional mobility B. Corresponding to our new law (4i), we have

$$\frac{\partial u}{\partial t} = \mathcal{M} \left[(\nabla \cdot B \nabla - \mathcal{M})^{-1}\, \nabla \cdot B \nabla \left(\varepsilon\, \Delta u - \frac{F'(u)}{\varepsilon} \right) \right] \tag{8i}$$

which is the viscous Cahn–Hilliard equation derived in ref. 9 but with $\varepsilon = 0$, or its equivalent form

$$\frac{\partial u}{\partial t} = \nabla \cdot B \nabla \left\{ (\nabla \cdot B \nabla - \mathcal{M})^{-1} \left[\mathcal{M} \left(\varepsilon\, \Delta u - \frac{F'(u)}{\varepsilon} \right) \right] \right\} \tag{8i'}$$

If we introduce a potential $-\theta$ and rewrite the equation as the system of equations

$$u_t = -\nabla \cdot B \nabla \theta, \qquad \theta = -u_t/\mathcal{M} + \varepsilon\, \Delta u - F'(u)/\varepsilon$$

we see that the chemical potential $-\theta$ has in it a time derivative of u—a sensitivity to what happened in the recent past. As in the sharp-interface case, we are guaranteed the existence of the inverse operators above.

One essential criterion in order that Eq. (7i) approximate surface diffusion is that $B=0$ at points outside of a relatively narrow interfacial band; one then sees that the diffusion inherent in the Cahn–Hilliard equation happens solely within that band. Two simple forms of B to accomplish this are (1) to let $B=\text{const}$ for $|u|<1$ and $B=0$ otherwise, or (2) to let $B=\mathscr{D}(1-u^2)/2kT$ for some constant $\mathscr{D}$. The latter is not only simple, but behaves near $u=1$ and $u=-1$ in a way that is defensible on thermodynamic grounds. The physical factors that appear in B for dilute solutions show that B is a product of the diffusion coefficient and the concentration of the minor diffusing species;[10,11] specifically $B=\mathscr{D}c/kT$ when c is small, and $B=\mathscr{D}(1-c)/kT$ when c is near 1, where $\mathscr{D}$ is the self-diffusion coefficient as measured with isotopes, and T is the temperature and k is Boltzmann's constant. To ensure that the results be relatively insensitive to ε, $B(u)$ could be taken to be $B(u)=\beta(u)/\varepsilon$ for $|u|<1$. This is consistent with experience; experimental surface diffusion coefficients have dimensions of m^3/sec; they are usually reported as an ordinary diffusion coefficient (m^2/sec) with an arbitrarily chosen thickness (usually 1 nm) factored out.[12]

With anisotropic surface energy, $\varepsilon^2|\nabla u|^2$ is replaced in the energy functional by $\varepsilon^2[\Gamma(\nabla u)]^2$. This leads to Δu being replaced by $\nabla\cdot[\Gamma(\mathbf{p})\,\nabla\Gamma(\mathbf{p})|_{\mathbf{p}=\nabla u}]$, which we abbreviate by $\Delta_\Gamma u$. This can be written as $\sum_{ij}(\Gamma_i\Gamma_j+\Gamma\Gamma_{ij})\,\partial^2u/\partial x_i\,\partial x_j$, in which $\Gamma_i=\partial\Gamma/\partial p_i|_{(\mathbf{p}=\nabla u)}$, etc. If Γ is not smooth or Δ_Γ is not an elliptic operator, then the appropriate weak form of the PDE is derived from the variational (gradient) versions described below. As was pointed out to us by Mete Soner, dependence on ∇u in Γ automatically leads to the same dependence on ∇u for the mobility of the interfacial region. Soner's argument (in arbitrary dimensions) is reproduced in the appendix. A formal asymptotic analysis on the phase field equations in R^2 which produces the same automatic factor of γ in the mobility [but which does not use a u-dependent diffusion coefficient and hence reduces to a (2a)-type equation], first appeared in ref. 5. For more arbitrary kinetics (as pointed out in ref. 5), additional directional anisotropy has to be incorporated through the diffusional mobilities B (which becomes a matrix rather than a scalar) and $\mathscr{M}$. Both of these functions might have to show a dependence on $\nabla u/|\nabla u|$ as well as u.

It is not clear whether making these assumptions of the dependence of various functions on $\nabla u/|\nabla u|$ creates insurmountable mathematical problems when ∇u is zero, since Δ_Γ, $\mathscr{M}$, and B would not be defined there. However, in fact, the dependence of these functions on ∇u should probably be modified near $\nabla u=0$ in any case, since near $\mathbf{p}=0$, $(\Gamma(\mathbf{p}))^2$ ought (for physical reasons) to reduce to $\mathbf{p}\cdot\Gamma_0\mathbf{p}$ for some matrix Γ_0.

We obtain four anisotropic diffuse interface models:

$$\frac{\partial u}{\partial t} = \mathcal{M}\left(u, \frac{\nabla u}{|\nabla u|}\right)\left(\varepsilon \Delta_\Gamma u - \frac{F'(u)}{\varepsilon}\right) \tag{5a}$$

$$\frac{\partial u}{\partial t} = \mathcal{M}\left(\varepsilon \Delta_\Gamma u - \frac{F'(u)}{\varepsilon} - \frac{\int_\Omega F'(u)\, dV}{\varepsilon \int_\Omega 1\, dV}\right) \tag{6a}$$

$$\frac{\partial u}{\partial t} = \nabla \cdot B(u) \nabla\left(-\varepsilon \Delta_\Gamma u + \frac{F'(u)}{\varepsilon}\right) \tag{7a}$$

$$\frac{\partial u}{\partial t} = \mathcal{M}\left[(\nabla \cdot B\nabla - \mathcal{M})^{-1} \nabla \cdot B\nabla\left(\varepsilon \Delta_\Gamma u - \frac{F'(u)}{\varepsilon}\right)\right] \tag{8a}$$

or equivalently

$$\frac{\partial u}{\partial t} = \nabla \cdot B\nabla\left\{(\nabla \cdot B\nabla - \mathcal{M})^{-1}\left[\mathcal{M}\left(\varepsilon \Delta_\Gamma u - \frac{F'(u)}{\varepsilon}\right)\right]\right\} \tag{8a'}$$

4. GRADIENT FLOWS

Suppose e is a differentiable function on R^2. Then

$$\frac{\partial}{\partial t} e(\mathbf{x}_0 + t\mathbf{v})|_{t=0} = v_1 \frac{\partial e}{\partial x_1} + v_2 \frac{\partial e}{\partial x_2}$$

is the rate of change of e when $\mathbf{x}$ moves with velocity vector $\mathbf{v}$. We observe that if $\cdot$ is the usual inner product on R^2 so that $\mathbf{w} \cdot \mathbf{v} = v_1 w_1 + v_2 w_2$, then the vector $\mathbf{w} = (\partial e/\partial x_1, \partial e/\partial x_2)|_{\mathbf{x}=\mathbf{x}_0}$ satisfies $\mathbf{v} \cdot \mathbf{w} = (\partial/\partial t)\, e(\mathbf{x}_0 + t\mathbf{v})|_{t=0}$. Indeed $\mathbf{w}$ is what we usually call the gradient of e, because we are accustomed to the usual inner product. Motion by the gradient $\mathbf{w}$ increases e fastest (and by $-\mathbf{w}$ decreases e fastest) compared to all other vectors of the same length as $\mathbf{w}$ (Schwarz's inequality) when length is measured in the usual way $[|\mathbf{v}| = (\mathbf{v} \cdot \mathbf{v})^{1/2}]$.

Similarly, given a functional E defined on some space of functions, one can define the first variation of E as a linear functional; given an inner product on that space of functions, one can define the operator that acts on functionals that is the gradient of E in that inner product space. The usual inner product for two functions u and v on a surface S without boundary is the L^2 inner product, $\int_S uv\, dA$. Related inner products are of the form $\int_S uv/M\, dA$ for some given positive function M on S. As we will see, such a function M allows us to incorporate kinetic anisotropies into a gradient formulation while leaving E unchanged to serve as a free energy Lyapunov functional.

Among the many other inner products is the H^{-1} inner product, which for two functions w and v on S, each with integral 0, is often

given in the symmetric form as a product of two (standard) gradients, $\int_S \nabla\phi_w \cdot \nabla\phi_v \, dA$. Here ϕ_v must satisfy $\Delta_S \phi_v = v$ and similarly for ϕ_w; since S is a surface without boundary, there is a unique solution to this equation up to a constant (which will not affect the inner product), so we can write $\phi_v = \Delta_S^{-1} v$. Integration by parts leads to the equivalent definitions of the H^{-1} inner product $-\int_S w\phi_v \, dA$ and $-\int_S v\phi_w \, dA$, also written as $-\int_S w \, \Delta_S^{-1} v \, dA$ and $-\int_S v \, \Delta_S^{-1} w \, dA$. The last form is directly usable in finding a suitable gradient. Related inner products are of the form $\int_S \nabla\psi_w^G \cdot G \nabla\psi_v^G \, dA$ (equivalently $-\int_S v\psi_w^G \, dA$) for some fixed positive-definite-matrix-valued function G on S, where for any w (or v), ψ_w^G satisfies $\nabla \cdot G \nabla\psi_w^G = w$. Again, there is a unique solution (up to constants) so that one can write $\psi_w^G = (\nabla \cdot G\nabla)^{-1} w$.

For functions v, w defined on some region Ω in R^3, one has the L^2 inner product $\int_\Omega vw \, dV$ and the related inner product (which depends on $\mathcal{M}$ and which in turn may depend on the current values of the gradient of u) $\int_\Omega vw/\mathcal{M} \, dV$. To define H^{-1}-type inner products, one has to require these functions to have integral 0. When B is constant, one obtains unique (up to a constant) ψ_w satisfying $\Delta\psi_w = w$ by solving the Neumann problem of having the normal derivative of ψ_w be zero on the boundary of Ω. For a u-dependent diffusional mobility B, whether it is a matrix or a scalar, one instead solves $\nabla \cdot B(u) \nabla\phi_w = w$. One then has the inner product (depending on the current value of u) $v \cdot w = \int_\Omega \nabla\phi_w \cdot B(u) \nabla\phi_v \, dV = -\int \phi_w v \, dV$.

Note that the L^2 inner product is naturally defined on a different space of functions than H^{-1}, most noticeably in that functions in the latter space must have integral 0 whereas functions in the former need not. However, some physical applications are limited to functions with integral 0, so even when we use the L^2 inner product we will be restricting the space of functions to the subspace of functions satisfying that constraint (either explicitly or by modifying the function E by adding a term which is a Lagrange multiplier times the integral of the function). In fact, the space of functions on which the inner product is naturally defined and which one usually uses is the completion in the metric induced by $\bullet$ of the set of smooth functions v (with integral 0 if necessary) with finite $v \bullet v$.

5. EQUATIONS (1)–(4) AS GRADIENT FLOWS

For sharp interface formulations with anisotropic surface energies, the weighted mean curvature κ_γ is defined, as stated in earlier papers,[2,3] to be the function on the surface S such that the first variation operator δ_γ acting on S satisfies $\delta_\gamma S(v) = (d/dt) \, E(S_t)|_{t=0} = \int_S (-\kappa_\gamma) v \, dA$, where $\{S_t\}$ is a family of surfaces moving with initial velocity v. This integral is precisely in the form of the L^2 inner product of v with the function $w = -\kappa_\gamma$. Thus with

the L^2 inner product, the gradient is the function $w = -\kappa_\gamma$. Motion by mean curvature with isotropic interface mobility [Eq. (1i)] is gradient flow for E with the L^2 inner product, and decreases energy fastest compared to all other velocities of the same "length" (measured in the L^2 sense). But motion by mean curvature does not in general conserve volume. Motion by the difference between mean curvature and the average mean curvature is a gradient flow for the same Lyapunov functional E with respect to the same L^2 inner product, but restricted to the subset of velocities that conserve volume (i.e., have integral 0); it will in general not decrease energy as fast as the unconstrained gradient flow. An alternative way of viewing this motion is to regard it as gradient flow for a different Lyapunov functional that includes a Lagrange multiplier (which turns out to be the average mean curvature) times the volume enclosed; this has the apparent advantage of making the flow be an unconstrained gradient flow, but the disadvantage of introducing a physically less relevant energy.

Using the same first variation operator, relating $(d/dt)\, E(S_t)|_{t=0}$ to $\int_S (-\kappa_\gamma) v\, dA$, with the H^{-1} inner product, the defining condition for the gradient becomes $-\phi_w = -\kappa_\gamma$, and therefore $w = \Delta_S \kappa_\gamma$. Hence surface motion by the Laplacian of weighted mean curvature is gradient flow for E in the H^{-1} inner product, and decreases energy fastest compared to all other velocities of the same "length" (measured in the H^{-1} sense).

Anisotropy is introduced into the energy via the function γ, and appears in the growth law through κ_γ, the first variation of the surface energy. Anisotropy in the kinetics is introduced into the inner products, not into the energy. Thus the L^2 inner product is modified to $\int_S uv/M\, dA$ (so that the gradient is $M\kappa_\gamma$) and the H^{-1} inner product is modified to $\int_S v\psi_w^D\, dA$ (so that the gradient satisfies $\psi_w^D = -\kappa_\gamma$, or equivalently is $w = \nabla \cdot D\nabla\kappa_\gamma$). Again, by Schwarz's inequality, gradient flow has the property of decreasing energy fastest subject to a length constraint (length being measured using the appropriate inner product).

To obtain the velocity law (4i), as a gradient flow, one would like to define an inner product $\bullet$ on functions w, v such that $w \bullet v = -\int_S \psi_w v\, dA$, where setting $\psi_w = \kappa_\gamma$ should imply $w = -[(1/M)\Delta_S - (1/D)]^{-1} \Delta_S \kappa_\gamma$. Thus, given any w, we want to define ψ_w so that $\psi_w = -\Delta_S^{-1}[(1/M)\Delta_S - (1/D)]v$. Again, this equation can be solved uniquely (up to a constant which affects nothing). We compute

$$-\int_S \psi_w v\, dA = \int v\Delta_S^{-1}\left(\frac{1}{M}\Delta_S - \frac{1}{D}\right) w\, dA = \frac{1}{M}\int_S vw\, dA + \frac{1}{D}\int_S -v\,\Delta_S^{-1} w\, dA$$

Therefore the desired inner product $\bullet$ is in fact a positive linear combination of inner products and is itself an inner product automatically. Further-

more, it is the most natural possible inner product, in that it is just the appropriate positive linear combination of the inner products for the extreme cases.

Finally, we do the same for the general anisotropic case. In order to have Eq. (4a) be a gradient flow, one would like to define an inner product $\bullet$ on functions w, v such that $w \bullet v = -\int_S \psi_w v \, dA$, where $\psi_w = \kappa_\gamma$ should imply $w = -M(\nabla \cdot D\nabla - M)^{-1} \nabla \cdot D\nabla_{\kappa_\gamma}$. Again, the resulting equation can be solved uniquely (up to a constant) so that for any w, $\psi_w = -(\nabla \cdot D\nabla)^{-1}(\nabla \cdot D\nabla - M)(w/M)$. [We could also equivalently use the other formula (4a′) for the velocity, and achieve the same result.] We compute

$$-\int_S \psi_w v \, dA = \int_S v(\nabla \cdot D\nabla)^{-1} [(\nabla \cdot D\nabla - M)(w/M)] \, dA$$

$$= \int_S \frac{vw}{M} dA + \int_S -v(\nabla \cdot D\nabla)^{-1} w \, dA$$

Again, the desired inner product is the most natural possible one, the sum of the inner products for the extreme cases.

It should be noted that a great many growth laws that one might write down can also be realized as gradient flows for some inner product. For example, the volume-conserving, energy-decreasing flow $v = M(\kappa_\gamma - \kappa_{\text{av}}) - \nabla D\nabla\kappa_\gamma$ is gradient flow for the inner product $w \bullet v = \int_S v L^{-1} w \, dA$, where L^{-1} is the inverse of the linear operator L defined by $L(u) = Mu - \nabla \cdot D\nabla u$.

6. EQUATIONS (5)–(8) AS GRADIENT FLOWS

For the diffuse interfaces, we begin with the energy functionals described earlier,

$$E(u) = \int_{\mathbf{x} \in \Omega} [\varepsilon^2 |\nabla u|^2 + F(u)] \, dV$$

in the isotropic case and

$$E(u) = \int_{\mathbf{x} \in \Omega} \{\varepsilon^2 [\Gamma(\nabla u)]^2 + F(u)\} \, dV$$

in the anisotropic surface free energy case. The first variation of the former is

$$(d/ds) \, E(u + sv)|_{s=0} = \int [F'(u) - \varepsilon^2 \Delta u] \, v \, dV$$

(there has been an integration by parts to write it in this form). The first variation of the latter is

$$(d/ds)\, E(u+sv)|_{s=0} = \int [F'(u) - \varepsilon^2 \Delta_\Gamma u]\, v\, dV$$

As was shown by Fife[13, 14] (and in fact first told to us by Robert Kohn), the Allen–Cahn equation is gradient flow for $E(u)$ in the L^2 inner product and the usual, constant-B, Cahn–Hilliard equation is gradient flow for $E(u)$ in the H^{-1} inner product for variational functions with integral 0 on Ω. The L^2 statement should be fairly clear from the first variation formula and the foregoing description. In order to have the gradient w of E be $-\Delta[F'(u) - \varepsilon^2 \Delta u]$ (so that Cahn–Hilliard will be gradient flow), we need to have $F'(u) - \varepsilon^2 \Delta u = \Delta^{-1} w$; since the inner product of w and v in the H^{-1} sense is $\int -v\, \Delta^{-1} w\, dV$, such a w is precisely the gradient of E with regard to the H^{-1} inner product.) To obtain Eq. (7i) or (7a), the Cahn–Hilliard equation with the u-dependent diffusional mobility B, one uses the inner product $\int \nabla \phi_w \cdot B(u)\, \nabla \phi_v\, dV = -\int \phi_w v\, dV$, where $\phi_w = [\nabla \cdot B(u) \nabla]^{-1} w$. Note that $(d/dt) \int u\, dV = 0$ is enforced automatically when $\partial u/\partial t = \nabla \cdot B \nabla[-F'(u) + \varepsilon^2 \Delta u]$, by the divergence theorem; thus the gradient in the H^{-1} inner product automatically satisfies volume conservation.

All the forms of Eq. (8) are similarly gradient flow for the inner product

$$v \bullet w = \int_\Omega \nabla \phi_w \cdot B(u)\, \nabla \phi_v\, dV + \int_\Omega vw/\mathscr{M}\, dV$$

7. DISCUSSION

We have shown that sets of sharp-interface and diffuse-interface motion laws can be described as gradient flows in appropriate inner products, and have proposed methods for introducing anisotropy into all of these laws. Even though the energy functionals are in quite different spaces, we have found that corresponding sharp-interface and diffuse-interface motion laws are linked by being gradient flows for analogous inner products.

The relationship between Eqs. (1i) and (5i) is well established. Not only has the formal asymptotics been done, but also convergence of solutions for (5i) to those for (1i) as ε approaches 0 has been shown.[15]

Much remains to be done for the other relationships (though very recent unpublished work of Elliott and Garke[16] begins to answer many of the fundamental questions). Rubenstein and Sternberg[17] use formal

asymptotics to show that the asymptotic limit of Eq. (6i) is (2i) (though the fact that the second term is average mean curvature is somewhat obscure). They compare the behavior of some approximate solutions to this equation to those of the Cahn–Hilliard equation. But nothing has been proved about the convergence of solutions to this equation to solutions to the corresponding sharp-interface equation. For Eqs. (3i) and (7i) (Mullins' surface diffusion and Cahn–Hilliard), but not for Eqs. (4i) and (8i), recent results[18] have established formal asymptotic convergence. The short-time existence of solutions to (3i) and (4i) has been shown with smooth initial data. Note that there is no maximal principle. There is no proof of convergence of solutions to either (7i) or (8i) to solutions to (3i) and (4i).

Incorporation of kinetics is accomplished by modifying the inner products, leaving the free energy (Lyapunov) functionals unchanged. Gradient flows from such modified inner products are then always consistent with both the thermodynamic and kinetic formulations; the free energy always goes down fastest with the given kinetics.

Conservation (mass or volume) laws are also automatically introduced in the H^{-1} inner product.

The new combined inner product leads to equations that conserve mass or volume and in the sharp-interface case are intermediate between motion by curvature minus average mean curvature and minus the Laplacian of curvature, and in the diffuse-interface case between a mass-conserving Cahn–Allen and the Cahn–Hilliard equations. The isotropic version of the diffuse-case intermediate equation contains what has been called a viscous term.

It is possible to make the anisotropy in the energy of the diffuse surfaces identical to that of the sharp planar interfaces by setting Γ proportional to γ. This proportionality remains when both functions are extended to all vectors. But for small values of ∇u, $[\Gamma(\nabla u)]^2$ should reduce to a tensor or matrix with the symmetry of the medium, and should always be convex.[10, 19] This anisotropy in general is that of an ellipsoid, but is that of a spheroid for tetragonal and hexagonal crystals, and a sphere for cubic crystals and fluids. The anisotropy of γ, while consistent with the symmetry of the medium, is not restricted to that of a tensor, and quite commonly is not convex. Thus the proportionality between Γ and γ must break down for small gradients. For calculations of energy and motion of diffuse interfaces this is probably not serious, since the contribution from the small-gradient regions should be small. It is, however, serious in spinodal decomposition, where one usually has random initial data that differ little from a constant and have small gradients. There is a need to construct a Γ that is given by a matrix at small gradients and reproduces the anisotropy of γ at larger gradients.

APPENDIX[3]

Equation (5a), with $M = 1/\varepsilon$ and with u denoted by u^ε to make explicit its dependence on ε, is

$$\varepsilon u_t^\varepsilon = -\frac{1}{\varepsilon} F'(u^\varepsilon) + \varepsilon \Delta_\Gamma u^\varepsilon$$

Introduce the standing wave $q'' = F'(q)$, which implies $q' = [2F(q)]^{1/2}$. [$q(s)$ is $\tanh(\sqrt{2}\, s)$ for $F(u) = (1-u^2)^2$ and is $\sin(\sqrt{2}\, s)$ for $F = 1 - u^2$ when $|u| \leqslant 1$]. Rescale u^ε as

$$z^\varepsilon(tx) = \varepsilon q^{-1}(u^\varepsilon(t, x))$$

so that

$$u^\varepsilon = q\left(\frac{z^\varepsilon}{\varepsilon}\right)$$

and

$$u_t^\varepsilon = \frac{1}{\varepsilon} q'\left(\frac{z^\varepsilon}{\varepsilon}\right) z_t^\varepsilon$$

Now

$$\nabla\Gamma(\nabla u^\varepsilon) = \nabla\Gamma(\nabla z^\varepsilon)$$

[because $\nabla\Gamma(p) = \nabla\Gamma(p/|p|)$ for all $p \neq 0$]. So

$$\Delta_\Gamma u^\varepsilon = \frac{1}{\varepsilon} q'\left(\frac{z^\varepsilon}{\varepsilon}\right) \Delta_\Gamma z^\varepsilon + \frac{1}{\varepsilon^2} q''\left(\frac{z^\varepsilon}{\varepsilon}\right) [\nabla\Gamma(\nabla z^\varepsilon) \cdot \nabla z^\varepsilon)]^2$$

Therefore (5a) implies

$$z_t^\varepsilon - \Delta_\Gamma z^\varepsilon + \frac{q''}{\varepsilon q'} [|\nabla\Gamma(\nabla z^\varepsilon) \cdot \nabla z^\varepsilon)|^2 - 1] = 0$$

Formally, in the limit as $z^\varepsilon \to z$, $|\nabla\Gamma(\nabla z^\varepsilon) \cdot \nabla z^\varepsilon)|^2$ goes to 1, which says that z is the signed distance to the interface in some metric that is related to Γ, and $\{z = 0\}$ is the limit interface, with normal velocity equal to $z_t/|\nabla z|$ and weighted mean curvature $\kappa_\gamma = [1/\Gamma(\nabla z)]\, \Delta_\Gamma z$. So $z_t^\varepsilon - \Delta_\Gamma z^\varepsilon = 0$ implies that the normal velocity v is given by

$$v = \frac{z_t}{|\nabla z|} = \frac{1}{|\nabla z|} \Gamma(\nabla z)\kappa_\gamma = \Gamma(n)\kappa_\gamma$$

[3] Due to Mete Stoner.

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