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# A phase field model with non-local and anisotropic potential

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## Abstract

We present an alternative formulation of the phase field method for modeling interfaces. In this approach one works directly with integral equations instead of differential equations. The technique allows one to derive, from microscopic potentials, the surface tension and macroscopic conditions at the interface. In the last section a numerical example is given to illustrate the use of the technique. For a given form of interaction potential, the surface tension and its anisotropy are calculated from which we construct the equilibrium shapes.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

Surface tension anisotropy has a pivotal role in the solidification of materials [9]. Not only is it instrumental in deciding the equilibrium shapes [11], but is also a key component of the dynamics of growth, e.g. in the formation of dendrites [9]. One of the fundamental issues involving anisotropy is the process whereby anisotropy at the molecular level is transformed into macroscopic shapes and structures. This is a basic theoretical problem since it demands an understanding of the mechanism whereby the molecular structure retains the symmetries as it forms a shape whose volume is  $10^{23}$  larger. Another, and more practical, aspect of anisotropy in materials concerns the computation and modeling of interfaces during solidification in complex industrial processes.

At the macroscopic level, anisotropy is manifested largely through surface tension. From a mechanical perspective, surface tension can be viewed as force per unit area opposing the formation of ‘new’ surface area. It arises from unbalanced forces on the surface. In general, these pulling forces do not lie on a plane but exist on curved surfaces resulting in interface conditions that must be satisfied. Originally formulated for pressure differences, this basic equilibrium condition is known as the Gibbs–Thomson relation

$$u[s]_E = -\sigma\kappa \quad (1.1)$$

where  $\sigma$  is the surface tension,  $[s]_E$  is the entropy difference between the phases per unit volume,  $\kappa$  is the sum of principal curvatures and  $u := (T - T_m)$  is the reduced temperature ( $T$  being the temperature at a point on the interface, and  $T_m$  is the equilibrium melting temperature). When there is anisotropy, surface tension can be written as a function of surface normal, i.e.  $\sigma(\hat{n})$ . In this case, the equation above needs to be modified. Thermodynamic and geometric ideas were put forward in the past to find the appropriate extension of the above result for anisotropic surface tension [10]. In two dimensions the result is

$$u[s]_E = -(\sigma + \sigma'')\kappa. \quad (1.2)$$

On the other hand, a mesoscopic approach that has been instrumental in developing both the fundamental and pragmatic aspects of anisotropy, surface tension and evolution of a solidification interface has been the phase field approach [1]. This approach can be used to derive the macroscopic interface relations such as (1.1) and their dynamic generalization. Furthermore, the method allowed for modeling of anisotropy at the microscopic level. In this context, anisotropic microscopic interactions with a lattice-spin Hamiltonian were first taken into account in [2] for two-fold anisotropy. Later the approach was refined by adding the contribution of higher order correlation terms to the Hamiltonian [3–5]. These were followed by the research aimed towards finding the equilibrium shapes [7] and obtaining macroscopic equations as probabilistic limits of discrete spin dynamics [12, 14, 15].

The generalization of the results on anisotropy in three dimensions is not immediate, as the polar coordinate approach is inadequate. Beyond the geometric issues, it is also desirable to seek a formulation in which one retains all of the detailed anisotropy. We resolve these problems by presenting results on a general set of interactions that contains both local and non-local interactions in arbitrary dimension. Instead of approximating Ising-type interactions with mean field and solving the corresponding phase field equation, a direct integral equation form is formulated, accomplishing the following objectives simultaneously:

- (i) the formulation is physically exact (as a continuum model);
- (ii) the formulation is valid for an arbitrary dimension;
- (iii) the existence of solutions to the integral equations is easily established using the current theory [6];
- (iv) there is an asymptotically stable numerical scheme for the solutions of the integral equations.

## 2. Anisotropic relations in arbitrary dimension with non-local interactions

In this section we present the idea of working directly on the integral form (see also [13]) for non-local interactions. This has a number of advantages, as summarized above, particularly in terms of avoiding arbitrarily large order differential equations. We shall derive a phase field model from a free energy functional that includes a non-local interaction term of the form  $\iint J_\varepsilon(x - y)[\phi(x) - \phi(y)]^2 dx dy$ .

In order to motivate this inclusion, we consider a microscopic lattice system involving a set of ‘spins’, denoted by a real value  $\phi_k$  for each lattice point  $k$ , and interactions of strength  $J_{kl}$  between these spins. In statistical mechanics, the (reduced) Hamiltonian of this physical system is described by

$$H_{\text{interaction}}[\phi] = \sum_{k,l} \frac{1}{4}(\phi_k - \phi_l)^2 J_{kl}.$$

The entropic part of the free energy, which is the temperature times the logarithm of number of accessible states i.e.,  $\{-\phi_k \ln \phi_k + (1 - \phi_k) \ln(1 - \phi_k)\}$ , is often approximated in applications

by a smooth double well potential, denoted by  $W(\phi_k)$ , which takes its minimum values on the bulk (i.e. single phase) material. In particular,  $\phi_k \simeq 1$  denotes the higher energy phase (liquid), while  $\phi_k \simeq -1$  denotes the lower energy (solid). Moreover, in undercooled melts, the free energy is further reduced by an amount proportional to latent heat. These ideas lead to the free energy that can be written as

$$F[\phi] = \sum_{k,l} \frac{1}{4} J_{kl} (\phi_k - \phi_l)^2 + \sum_k W(\phi_k) + \sum_k uG(\phi_k). \quad (2.1)$$

When passing to the continuum limit, the interaction strength must be scaled appropriately [2]. In the continuum limit we replace the summation by integrals (and the free energy by its calligraphic counterpart). Hence, we arrive at the functional form of the interfacial excess free energy:

$$\mathcal{F}[\phi] = \frac{1}{4} \int J_\varepsilon(x-y)(\phi(x) - \phi(y))^2 dx dy + \int W(\phi(x)) dx + \int uG(\phi(x)) dx \quad (2.2)$$

where  $J_\varepsilon(z) = \varepsilon^{-N} J(\varepsilon^{-1}z)$ ,  $\varepsilon$  is an atomic length scale and  $G(\phi)$  represents the entropy difference per unit volume. For simplicity we assume  $\Omega = \mathbb{R}^N$  with  $N \geq 2$ .

The phase field equation can be obtained under the assumption that the variational derivative of the free energy should be proportional to the time derivative of the phase field, i.e.

$$\alpha \varepsilon^2 \phi_t = J_\varepsilon * \phi - \phi - W'(\phi) + \varepsilon u G'(\phi). \quad (2.3)$$

Equations (2.2) and (2.3) are the two main equations from which the key results are derived. As a consequence of this approach, one obtains several mathematical and practical benefits that will be discussed in the following sections.

### 3. The Gibbs–Thomson–Herring condition with linear kinetics

#### 3.1. Stationary solutions with planar interfaces

Given a point  $x_0 \in \mathbb{R}^N$  and a direction  $\zeta \in \mathbb{R}^N \setminus \{0\}$ , we seek solutions of (2.3) such that  $\phi(x_0, t) = 0$  and the level sets of the phase field are the hyperplanes perpendicular to a given direction  $\zeta$ , i.e.

$$\phi(x) = Q(\zeta, z), \quad z = \frac{(x - x_0) \cdot \zeta}{\varepsilon}. \quad (3.1)$$

Making use of this special form, we shall rewrite the integral term in (2.3). By a change of variables,  $y = z\zeta + \nu$  such that  $\nu \perp \zeta$ , the term  $J_\varepsilon * \phi$  in (2.3) becomes

$$\begin{aligned} J_\varepsilon * \phi(x) &= \int_{\mathbb{R}^N} J(y) \phi(x - \varepsilon y) dy = \int_{\mathbb{R}^N} J(y) Q(\zeta, z - y \cdot \zeta) dy \\ &= \int Q(\zeta, z - \bar{z})(|\zeta| \int J(\bar{z}\zeta + \nu) d\nu) = (j * Q)(\zeta, z) \end{aligned} \quad (3.2)$$

where  $j := j(\zeta, z)$  is defined by

$$j(\zeta, z) := |\zeta| \int_{\nu \perp \zeta} J(\bar{z}\zeta + \nu) d\nu. \quad (3.3)$$

Thus, for fixed  $\zeta$ , and a given function  $W$ , the stationary solutions of the phase field equation with zero undercooling are obtained by solving the integro-differential equation:

$$\begin{aligned} 0 &= [(j * Q)(\zeta, z) - Q(\zeta, z)] - W'(Q(\zeta, z)) \\ \pm 1 &= \lim_{z \rightarrow \pm\infty} Q(\zeta, z), \quad Q(\zeta, 0) = 0 \end{aligned} \quad (3.4)$$

whose existence and uniqueness of solutions have already been proved in [6]. To determine the solutions of (3.4) we perform the following iteration:

$$\begin{aligned}\phi_0(z) &= \tanh z \\ \phi_{k+1}(z) &= \phi_k(z) + \Delta t \{ (j(\zeta) * \phi_k)(z) - \phi_k(z) - W'(\phi_k(z)) \} \\ Q(\zeta, z) &= \lim_{k \rightarrow \infty} \phi_k(z).\end{aligned}\tag{3.5}$$

The convergence of this scheme to the stationary solution  $Q$  is also guaranteed by the existing theory [6].

### 3.2. Surface tension

In materials science, surface tension is defined as the difference per unit area between the free energy with the interface and without, i.e.

$$\sigma = \frac{\mathcal{F}[\text{liquid} + \text{solid}] - (\mathcal{F}[\text{liquid}] + \mathcal{F}[\text{solid}])/2}{\text{Area}}.$$

To compute the ‘area’ one considers a sufficiently small cross section that the free energy does not vary substantially as one moves parallel to the cross section.

In the framework of the phase field models [1], this can be written as

$$\sigma = \frac{\mathcal{F}[\phi] - (\mathcal{F}[-1] + \mathcal{F}[1])/2}{\text{Area}}\tag{3.6}$$

where  $\phi$  is the solution of the phase field equation that makes a transition from solid to liquid, while  $-1$  and  $1$  represent the solid and liquid phases, respectively. In order to make this concept more precise we use the stationary solutions (solutions of equation (3.4)). Once this is obtained for a fixed direction  $\zeta$ , it can be used in (3.6) to calculate the free energy density. The planar symmetry eliminates the transverse integral together with the division by area so that one has a precise mathematical definition in terms of a one-dimensional integral. Indeed, a brief calculation using (2.2), (3.3) and (3.6) yields

$$\begin{aligned}\sigma(\zeta) &:= \int_{\mathbb{R}} \{ W(Q(\zeta, z)) + \frac{1}{2} Q(\zeta, z)(Q(\zeta, z) - (j * Q)(\zeta, z)) \} dz \\ &= \int_{\mathbb{R}} (W(Q(\zeta, z)) - \frac{1}{2} Q(\zeta, z)W'(Q(\zeta, z))) dz,\end{aligned}\tag{3.7}$$

where the second line is obtained by making use of equation (3.4). When  $\zeta = \hat{n}$ ,  $\sigma(\zeta) = \sigma(\hat{n})$  corresponds to the surface tension of the interface.

### 3.3. Main result

In order to give the statement of the main theorem we introduce the basic notation for the theorem with  $N$  as the spatial dimension using the convention that vectors such as  $x$  or  $\hat{n}$  are represented by  $1 \times N$  row vectors and operators such as the gradient  $\vec{\nabla}$  by  $N \times 1$  column vectors. Let  $h(x, t)$  be the signed distance (positive in the liquid) from the point  $x$  to  $\Gamma_t$ , the limit interface between liquid and solid at time  $t$ . Then, in the local coordinate system  $(s', h) := (s^1, \dots, s^{N-1}, h) \in \mathbb{R}^N$ , one has

$$x = x_0(s') + h\hat{n}(s'),\tag{3.8}$$

where  $\hat{n}$  is the unit normal. Thus, with these definitions,  $D^2h$  and  $\vec{\nabla}\hat{n}$  are  $N \times N$  matrices with components  $(D^2h)_{ij} = \partial h_i / \partial x_j$  and  $(\vec{\nabla}\hat{n})_{ij} = \partial \hat{n}_i / \partial x_j$  and are related to each other in the following way:

$$\vec{\nabla}h = \hat{n}^T \quad \text{and} \quad D^2h = \vec{\nabla}\hat{n}.$$

Also, let the velocity of the interface at an arbitrary point  $x_0 \in \Gamma_t$  be expressed as

$$v(x_0, t) = \frac{\partial h(x_0, t)}{\partial t}.$$

Elsewhere we give the proof of our main theorem, that our phase field model provides the following condition.

**Theorem.** *The solution  $\phi$  of (2.3) admits a formal asymptotic expansion only if the Gibbs–Thomson–Herring condition (with linear kinetics) is satisfied, i.e.*

$$u + \alpha(\hat{n})v + \text{Trace}\left(\vec{\nabla}\hat{n} D^2\sigma(\hat{n})\right) = 0. \tag{3.9}$$

Using tensor notation this can also be expressed as

$$u + \alpha(\hat{n})v + \vec{\nabla}\hat{n} : D^2\sigma(\hat{n}) = 0. \tag{3.10}$$

#### 4. The Wulff shape—a numerical example

We consider a two-dimensional example with anisotropic interactions using  $J(x, y) =: \bar{J}(r, \theta)$ . We will compute the Wulff shape in three steps by (i) utilizing the planar solutions of (3.4), (ii) calculating the corresponding surface free energy density  $\bar{\sigma}(\theta) =: \sigma(\hat{n})$  and (iii) solving the differential  $\bar{\sigma}(\theta) + \bar{\sigma}''(\theta) = -u[s]_E/\kappa(\theta)$  by use of arc-length parametrization.

Let  $\bar{J}(r, \theta)$  be given by

$$\bar{J}(r, \theta) = f_0(r) + \delta \cos(n\theta) f_1(r), \quad r = \sqrt{x^2 + y^2}, \quad \tan \theta = \frac{y}{x}.$$

As an illustration, we choose the following:

$$n = 6, \quad f_0(r) = \frac{e^{-r^2}}{\pi}, \quad f_1(r) = -\frac{r^6 e^{3-2r^2}}{27\pi}.$$

For  $\hat{n} = (\cos \theta, \sin \theta)$  we have

$$\begin{aligned} \bar{j}(\theta, z) &:= \int_{-\infty}^{\infty} \bar{J}\left(\sqrt{z^2 + \ell^2}, \theta + \arctan \frac{\ell}{z}\right) d\ell \\ &= \hat{j}(\delta \cos n\theta, z) \end{aligned}$$

where

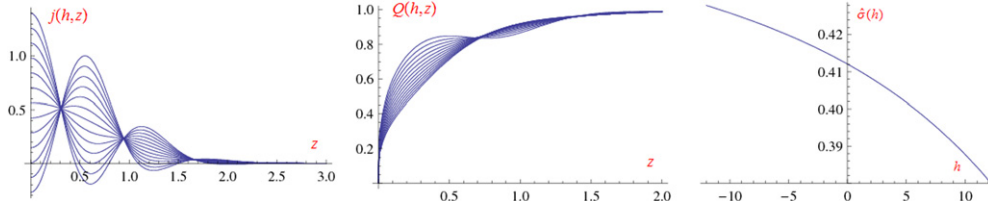
$$\begin{aligned} \hat{j}(h, z) &= j_0(z) + h j_n(z) \\ j_0(z) &:= 2 \int_0^{\infty} f_0(\sqrt{z^2 + \ell^2}) d\ell \\ j_n(z) &:= 2 \int_0^{\infty} f_1(\sqrt{z^2 + \ell^2}) \cos\left(n \arctan \frac{\ell}{z}\right) d\ell. \end{aligned}$$

In our example, we have

$$j_0(z) = \frac{e^{-z^2}}{\sqrt{\pi}}, \quad j_6(z) = \frac{e^{3-2z^2}(15 - 180z^2 + 240z^4 - 64z^6)}{1728\sqrt{2}\pi}.$$

The function  $\hat{j}(h, z) := j_0(z) + h j_6(z)$  is shown in figure 1(a). It is easy to verify that

$$\begin{aligned} J(x, y) \geq 0 \quad \forall (x, y) \in \mathbb{R}^2 &\iff |\delta| \leq 1 \\ \bar{j}(\theta, z) = \hat{j}(\delta \cos n\theta, z) \geq 0 \quad \forall \theta \in [0, 2\pi], \quad z \in \mathbb{R} &\iff |\delta| \leq 6.15285\dots \end{aligned}$$



**Figure 1.** (a) The function  $\hat{j}(h, z)$ ;  $h = -12, 10, \dots, 12$ ; (b) the odd function  $\hat{Q}(h, z)$ ; (c) the function  $\hat{\sigma}(h)$ .

For each  $h \in [-12, 12]$ , we denote by  $\hat{Q}(h, z)$  the solution  $Q$  of (3.4) with

$$j(\zeta, z) = \hat{j}(h, z), \quad W(q) = \frac{(1 - q^2)^2}{4}.$$

Numerically, we compute the solution by the iteration scheme

$$Q_0(z) = \tanh(z), \quad Q_{k+1}(z) = \sqrt[3]{\hat{j} * Q_k(z)}, \quad \hat{Q}(h, z) = \lim_{k \rightarrow \infty} Q_k(z).$$

The solution  $\hat{Q}(h, z)$  is shown in figure 1(b). For  $h \in [11, 12]$ , we find that  $\hat{Q}(h, z)$  is not monotonic; this is caused in part by the fact that  $\hat{j}(h, z)$  is not positive when  $h \geq 6.15 \dots$ . Making use of equation (3.4) the corresponding surface energy density, plotted in figure 1(c), is calculated via

$$\hat{\sigma}(h) = \int_{-\infty}^{\infty} \left( W(\hat{Q}(h, z)) - \frac{1}{2} \hat{Q}(h, z) W'(\hat{Q}(h, z)) \right) dz = \frac{1}{4} \int_{-\infty}^{\infty} (1 - \hat{Q}^4(h, z)) dz.$$

Now for fixed  $\delta$ , denoting  $\hat{n} = (\cos \theta, \sin \theta)$  we have

$$Q(\hat{n}, z) = \hat{Q}(\delta \cos(6\theta), z), \quad \sigma(\hat{n}) = \bar{\sigma}(\theta) = \hat{\sigma}(\delta \cos(6\theta)).$$

From the plot of  $\hat{\sigma}(\cdot)$  in figure 1(c), we see that

$$\hat{\sigma}(h) \approx 0.412062 - 0.00177h - 0.0000500h^2.$$

For  $\delta = 1, 8$  and  $12$ , the functions  $\bar{\sigma}(\theta) = \hat{\sigma}(\delta \cos(6\theta))$  and  $\bar{\sigma}(\theta) + \bar{\sigma}''(\theta)$  are plotted in figure 2. Numerically, we find that  $\bar{\sigma}(\theta) + \bar{\sigma}''(\theta) > 0$  for all  $\theta \in [0, 2\pi]$  when  $|\delta| < 9.78 \dots$

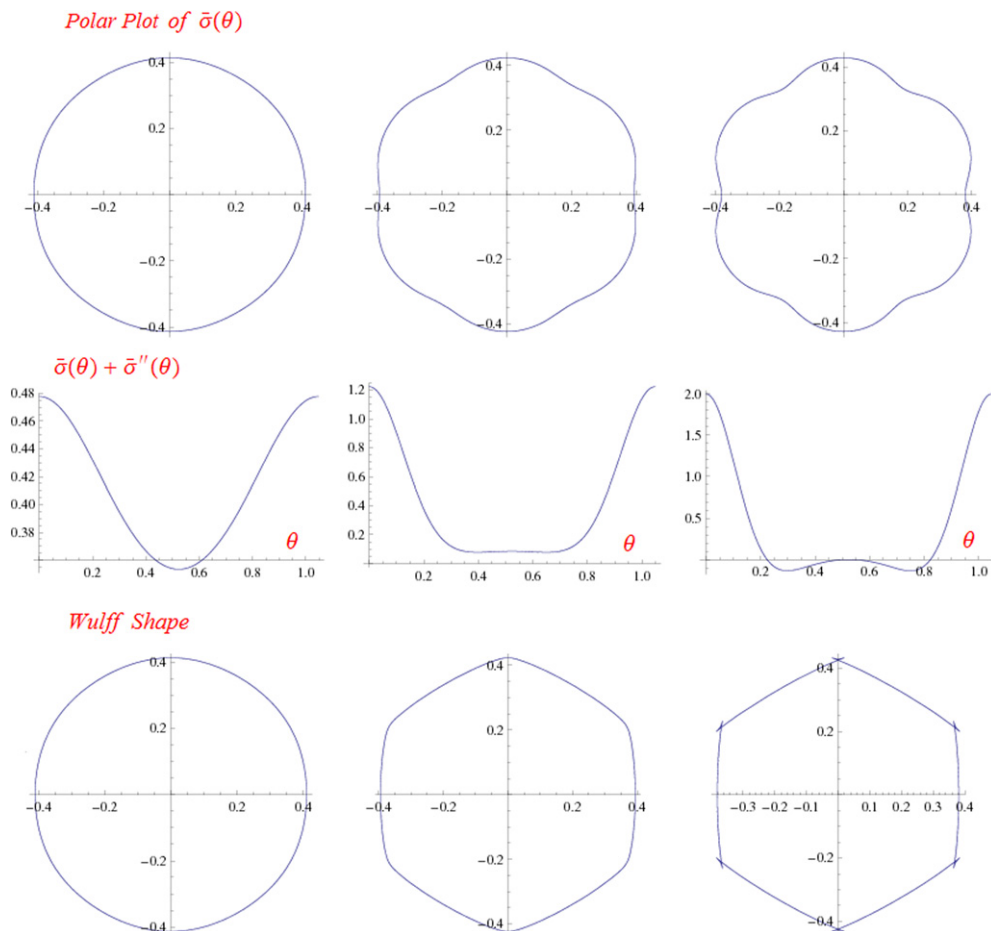
The *Wulff shape* [11] is the shape of a solid at the undercooling temperature  $u \equiv -1$ . Instead of finding the shape by computing the minimizers of (2.2) with a prescribed volume condition, we construct the Wulff shape directly from the surface tension computed from infinite planar interfaces, i.e.

$$X(\theta) = \bar{\sigma}(\theta) \langle \cos \theta, \sin \theta \rangle + \bar{\sigma}'(\theta) \langle -\sin \theta, \cos \theta \rangle, \quad \theta \in [0, 2\pi].$$

For  $\delta = 1, 8, 12$ , the Wulff shapes are given in the third row in figure 2. When  $\delta \in [9.79, 12]$ , the function  $\bar{\sigma} + \bar{\sigma}''$  is not positive; the corresponding Wulff shape is close to a hexagon with ‘ears’ at the vertices.

## 5. Conclusion

In this paper, we have addressed the question of computing anisotropy from microscopic physics. A physically satisfactory model should answer the following: given a set of microscopic interactions, namely the  $J_{kl}$ , what is the resulting interface condition (e.g. analogous to the Gibbs–Thomson–Herring condition)? Moreover, what is the resulting shape (e.g. the Wulff shape)? This involves the understanding of how the microscopic interactions are communicated to the macroscopic shape. We have presented a method that considers microscopic interactions and utilizes the integral equation form and differential geometry in order to derive the interface relation in arbitrary dimension and construct the Wulff shape.



**Figure 2.** First row: polar plot of surface tension, i.e. the function  $\bar{\sigma}(\theta)$  with  $\theta$  axis represented by unit circle and  $\bar{\sigma}(\theta)$  the distance to the origin; second row: the function  $\bar{\sigma}(\theta) + \bar{\sigma}''(\theta)$  in one period  $([0, \pi/3])$ ; last row: Wulff shapes.

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