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Faceted and dendritic morphology change in alloy solidification

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ABSTRACT

We present methods and results for the simulation of faceted and dendritic crystal growth. Using a thermodynamically realistic isothermal alloy model for AlSi we demonstrate, in confirmation of experimental observations, a change in morphology from perfectly faceted hexagons at smaller undercooling to dendritic growth at larger undercoolings. We also demonstrate that there exists a cut off temperature which separates the two distinct morphologies, and indeed hybrid morphologies. These results suggest that the mechanism for morphology variation observed experimentally primarily lies in anisotropic surface free energy modelling, which we adopt in preference to kinetic anisotropy.

1. Introduction

During solidification close to equilibrium crystal morphologies adopt the Wulff shape, which is itself a reflection of the underlying crystallography. However, at large departures from equilibrium alternate morphologies are often adopted. One manifestation of this is that crystals that are faceted at equilibrium become progressively less faceted and more dendritic in character with increasing growth velocity. Such a range of transitions was elegantly demonstrated by [1] in respect of Si crystals growing from an Al-Si melt solidified on a chill plate, with the transition also having been widely studied in the semiconductors Si [2,3] and Ge [4,5] together with Si-Ge [1] mixtures. Where measurements have been made of the growth velocity [2,5] the transition from faceted to dendritic growth is usually accompanied by an abrupt increase in growth velocity. In recent experimental work [6] found, in Cu_6Sn_5, there to be a change from faceted rods to non-faceted dendrites as the cooling rate was increased.

To date the modelling of two-phase crystal growth has concentrated on either continuous dendritic morphology or faceted growth. Here we present an approach to modelling facets that also allows for a transition to a continuous (dendritic) morphology under large departures from equilibrium and also accommodates the intermediate morphology of faceted dendrites. Hence, we clarify that the main mechanism for morphology change is in fact undercooling (and not cooling rate).

In [1] solidified crystals on a chill plate are seen to exhibit a range of morphologies. The observation is that of faceted crystals forming from smooth nuclei at small undercoolings, near equilibrium, giving polyhedra. At greater undercooling dendritic instability may set in before subsequently forming facets thereby giving rise to a faceted dendritic morphology. Most phase field simulations designed to simulate faceted growth only exhibit facets, [8,10–14,18,19], and, of course, the vast majority of phase-field simulations do not show facets at all.

The modelling of facets in phase field is done via a specification of the surface energy, the mobility, or both. We focus on surface energy anisotropy modelling. The link between the surface energy anisotropy and the resulting morphology is discussed in [7]. Crucially, though, the predicted morphology is only strictly valid at small undercooling. The question to be asked is therefore: what happens to the morphology of the solid as the undercooling in increased? In [9] dendritic growth is observed for a pure metal with four-fold symmetry, where, at the tip of the dendrite arms, facets are exhibited. The difference between this and the present work is that we simulate a given specific alloy (AlSi) using a full thermodynamic description, and explore the dependency of morphology on undercooling. We demonstrate that, indeed, facets occur at small undercooling and dendritic growth patterns are observed at high undercooling, with a continuous range of morphologies in between. We model AlSi in two dimensions, with the specification that hexagonal facets are to be formed at near equilibrium. A comprehensive overview of modelling approaches is given in [17], where in general anisotropy can be modelled in both the interface and/or in the mobility. If the anisotropy is in the mobility only, the effect vanishes (within phase-field at least)

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as $V \to 0$, so this does not give faceted growth during equilibrium solidification. As a minimum, using a capillary anisotropy is therefore a computational expedient to simulating the faceted to continuous growth transition.

We apply the computational techniques detailed in [20].

2. Facets and anisotropy modelling

Within the phase field model the surface features are modelled using the gradients of the phase, $\nabla\phi$ (the detailed phase model is given in Section 3).

Much of the literature concerns 4-fold symmetric growth, so we use this to illustrate the modelling. A 4-fold anisotropy used in the literature to generate 4-fold dendrites is given by

$$A = \frac{1}{2}\delta^2[1 + \epsilon\cos(4\theta)]^2\nabla\phi\cdot\nabla\phi. \tag{1}$$

which is controlled by a parameter, ϵ, which when zero give isotropic growth. This can also be written

$$A = \frac{1}{2}\delta^2\gamma^2 \tag{2}$$

where, in general, for m-fold symmetry

$$\gamma = [1 + \epsilon\cos(m\theta)]|\nabla\phi|. \tag{3}$$

γ is also a function of the surface normal in the following sense

$$\begin{bmatrix}\cos\theta\\ \sin\theta\end{bmatrix} = \frac{\nabla\phi}{|\nabla\phi|} \equiv \mathbf{n}. \tag{4}$$

and has the property $\gamma(\alpha\mathbf{n}) = \alpha\gamma(\mathbf{n})$[1] for any function, α. It is also convenient to consider the function, η, related to γ by $\gamma = \eta|\nabla\phi|$. This function has the property $\eta(\alpha\mathbf{n}) = \eta(\mathbf{n})$. Fig. 1 plots η as a function of θ in a polar plot, where we see that the circle deforms with increasing ϵ to a point where there are concave regions, here illustrated for $\epsilon = 0.1$. Setting the locus of the curve

$$\mathbf{x}(\theta) = \eta(\theta)[\cos\theta, \sin\theta]^T, \tag{5}$$

a concave region occurs if

$$\frac{\partial}{\partial\theta}|\mathbf{x}'(\theta)| = 0 \tag{6}$$

for some θ, which gives the condition to avoid concavity as $\eta''(\theta) + \eta(\theta) > 0$. For the case just considered this implies $\epsilon < 1/15$ to avoid concavity, and in general $\epsilon < 1/(m^2 - 1)$ for m-fold symmetry. Another, more direct, way of illustrating the anisotropy function, γ, is by way of a Frank diagram (or shape). Here we choose a contour of γ in the space $[\phi_x, \phi_y]$, see Fig. 2. The problem of concavity in the anisotropy function is that the resulting crystal shape can develop discontinuities in these regions, which in turn presents numerical difficulties.

By writing

$$\gamma_x \equiv \frac{\partial\gamma}{\partial\phi_x}, \quad \gamma_y \equiv \frac{\partial\gamma}{\partial\phi_y}, \tag{7}$$

both of which are functions of ϕ_x and ϕ_y, a 2D space curve, $W(t)$, can be produced by setting $[\phi_x = \cos(t), \phi_y = \sin(t)]$ in functions γ_x and γ_y to give $W(t) = [\gamma_x(t), \gamma_y(t)]$. This is shown in Fig. 3 for the four-fold symmetry case, where cusps, known as "ears", form on the $\epsilon = 0.1 > 1/15$ curve, which correspond to the concavity in the Frank diagram (Fig. 2). Formally the Wulff shape is the shape enclosed by $W(t)$ (without the ears). The 4-fold anisotropy discussed so far cannot produce facets, rather, as the parameter ϵ increases a sharp corner is produced on an otherwise rounded shape (a bulging square). So we seek other forms that, ideally, have rounded corners but perfectly flat sides. A number of approaches to modelling facets in crystal growth are found in the literature:

[1] Or $\gamma(\alpha\nabla\phi) = \alpha\gamma(\nabla\phi)$.

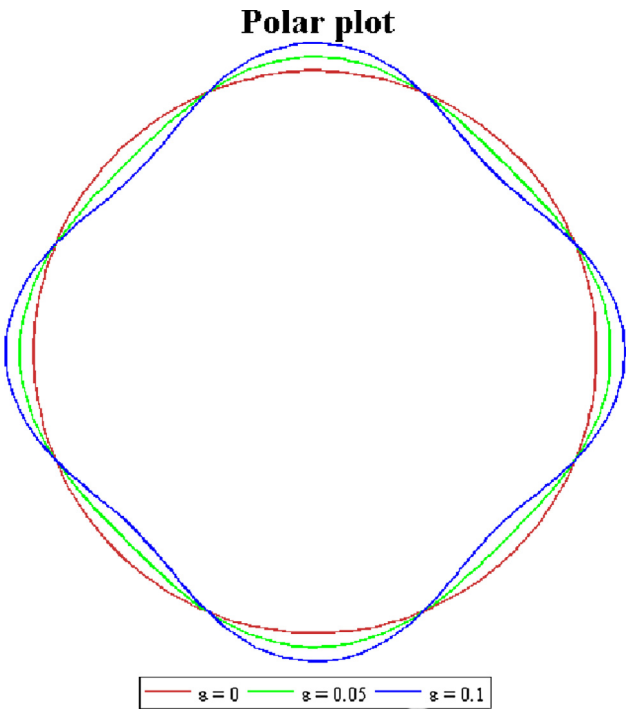

Fig. 1. Polar plot of the function, $\eta = 1 + \epsilon\cos(4\theta)$, with anisotropy varying from zero (isotropic – red) to extreme anisotropy (blue). The blue curve exhibits concave regions. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

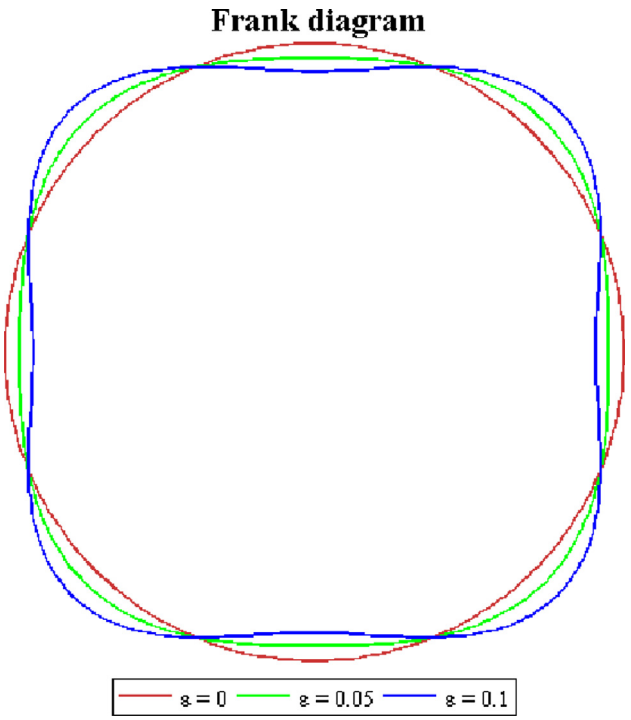

Fig. 2. The Frank diagram of the 4-fold anisotropy function is a contour of γ in the space $[\phi_x, \phi_y]$. Each contour is identical in shape and is conical for $\epsilon = 0$.