The Challenge of Using Phase-Field Techniques in the Simulation of Low Anisotropy Structures

A.M. Mullis

Institute for Materials Research
University of Leeds, Leeds LS2 9JT, UK

Introduction

The dendrite is a ubiquitous feature during the solidification of metallic melts. The importance of this type of solidification morphology is reflected in the large volume of literature devoted to understanding dendritic growth. This has its origins in the observation by Papapetrou (1935) that the dendrite tip is a paraboloid of revolution and the analytical solutions of Ivantsov (1947) which showed that a parabolid of revolution was in deed a shape preserving solution to the thermal diffusion equation for an isothermal dendrite growing into its undercooled parent melt.

Introduction - Models of Dendrite Tip Radius

Analytical theories of dendritic growth generally relate the Peclet number

$$P_t = \frac{VR}{2D}$$

to undercooling ΔT, rather the velocity, V, or tip radius, R, individually.

Many models have been put forward to explain why this degeneracy is broken in nature and, for a given material, V, can always uniquely be related to ΔT. The most successful of these is the theory of microscopic solvability, the principal prediction of which is that capillary anisotropy breaks the Ivantsov degeneracy via the relationship

$$R^2 V = \frac{2Dd_o}{\sigma^*}$$

Introduction

For a sharp interface model the equations to be solved are

$$\frac{\partial T}{\partial t} = D\nabla^2 T \qquad (1)$$

$$cD[\hat{\mathbf{n}}(\nabla T)_l - \hat{\mathbf{n}}(\nabla T)_s] = -Lv_n \qquad (2)$$

$$T_i = T_m - \frac{L}{c} d(\theta)K - \beta(\theta)v_n \qquad (3)$$

Equation (2) is simply the balance of heat fluxes across the interface, while Equation (3) is the moving interface version of the Gibbs-Thomson equation with local interface temperature, T_i, with *anisotropic* capillary length and attachment kinetics

Anisotropy & Phase-Field

In phase-field modelling anisotropy can be introduced by letting the width of the diffuse interface be anisotropic. Evolution of the phase variable, ϕ, is given by (e.g Wheeler et al. (1993))

$$\frac{\tilde{\varepsilon}^2}{m}\frac{\partial\phi}{\partial\tau} = \phi(1-\phi)\left[\phi - \tfrac{1}{2} + 30\tilde{\varepsilon}\alpha\Delta u\phi(1-\phi)\right] + \nabla^2\left(\tilde{\varepsilon}^2\phi\right)$$

while evolution of the dimensionless temperature, u, $T = T_m + u\Delta T$

is given by

$$\frac{\partial u}{\partial\tau} + \frac{1}{\Delta}p'(\phi)\frac{\partial\phi}{\partial\tau} = \nabla^2 u$$

Where m, α and Δ are material dependant constants,

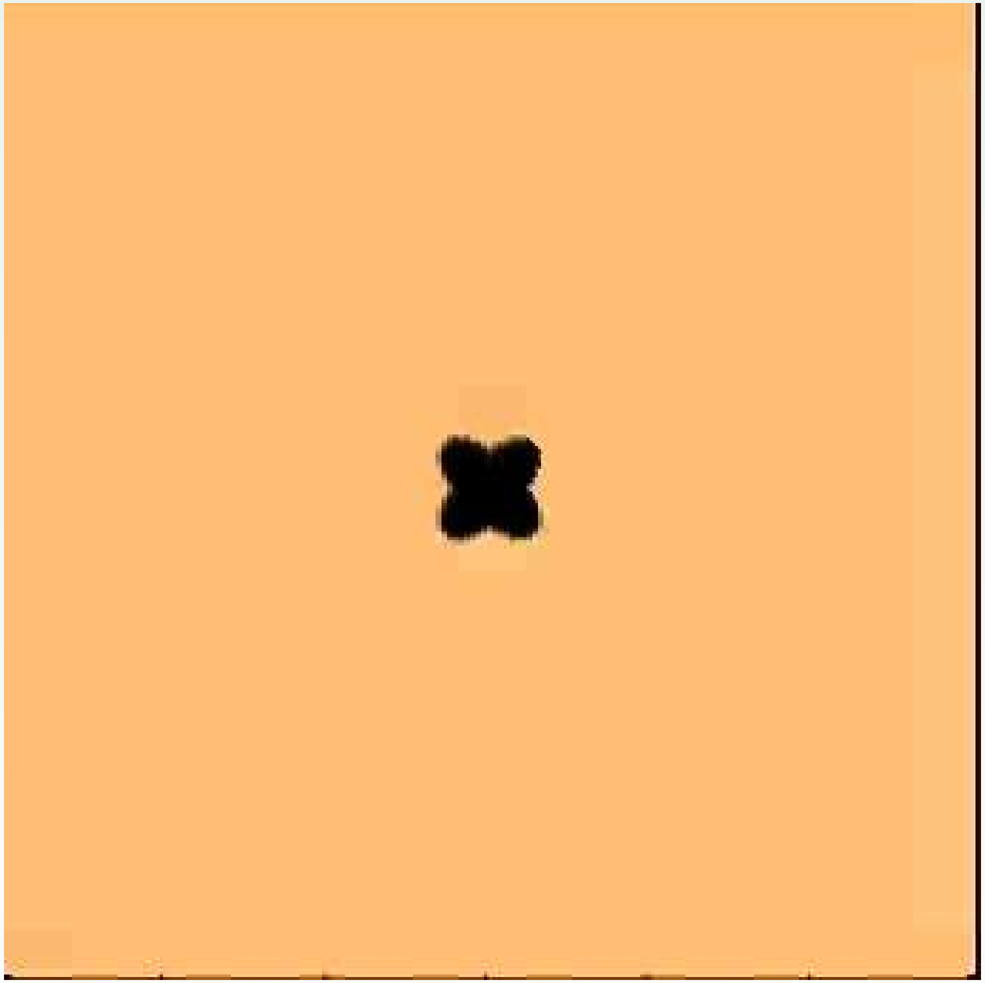
$$p(\phi) = \phi^3(10 - 15\phi + 6\phi^2)$$

and

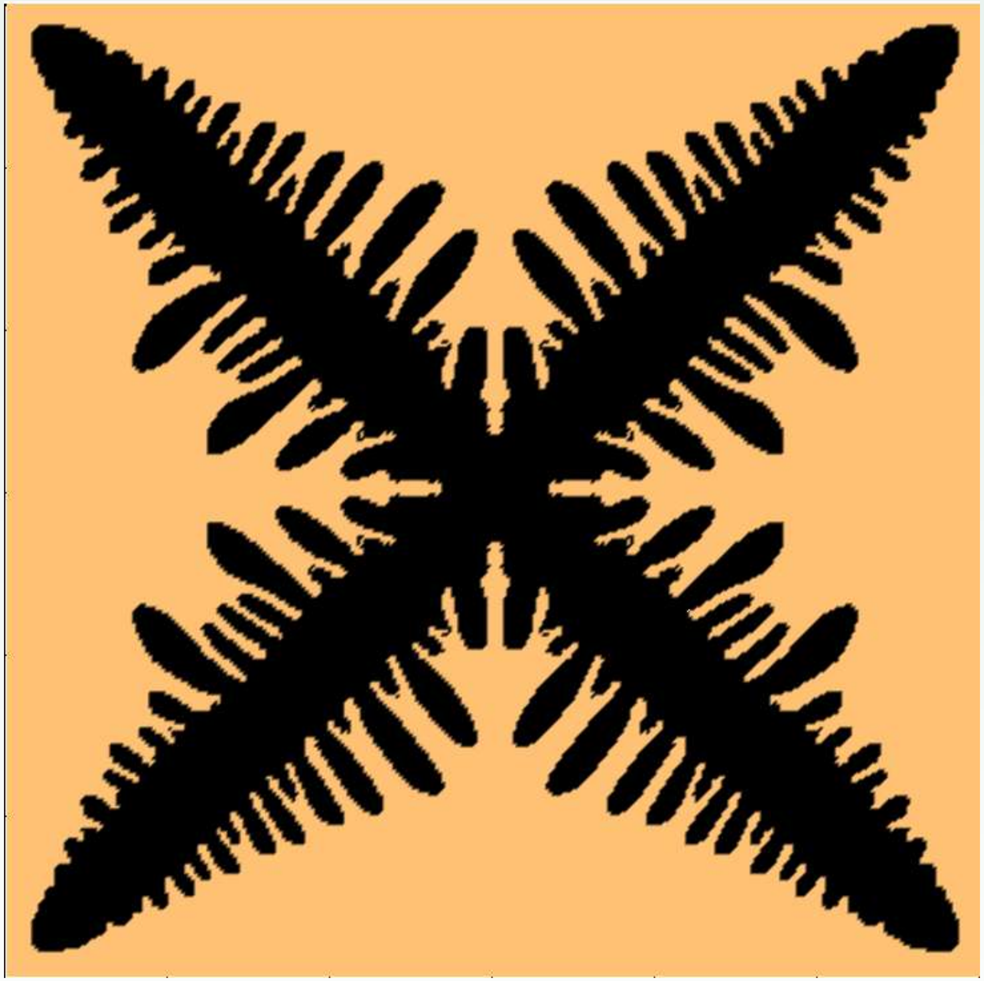
$$\tilde{\varepsilon}(\theta) = \varepsilon\ \ (\theta) = \varepsilon(1 + \gamma\cos k\theta)$$

Where γ is the anisotropy parameter and ε is a constant related to the interface thickness.

Phase-Field Simulation of Dendritic Growth



Phase-Field Simulation of Dendritic Growth

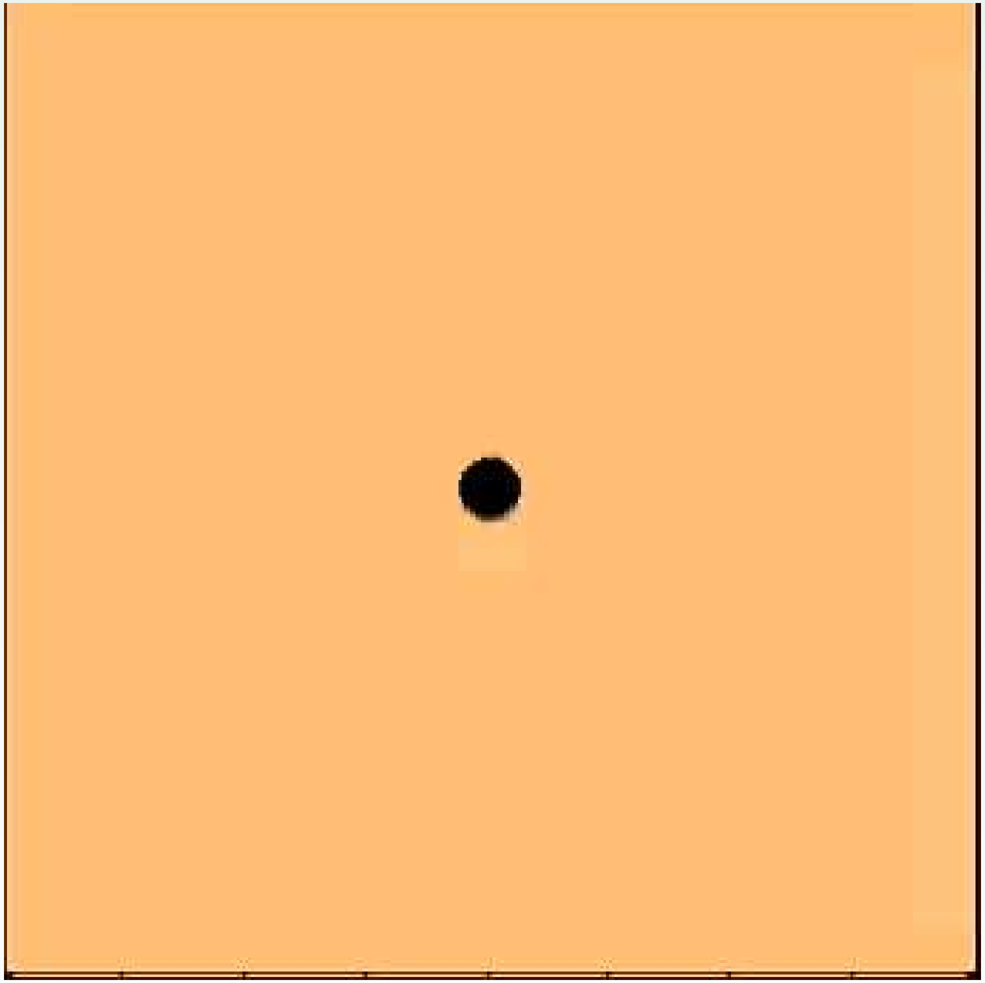


Anisotropy & Phase-Field

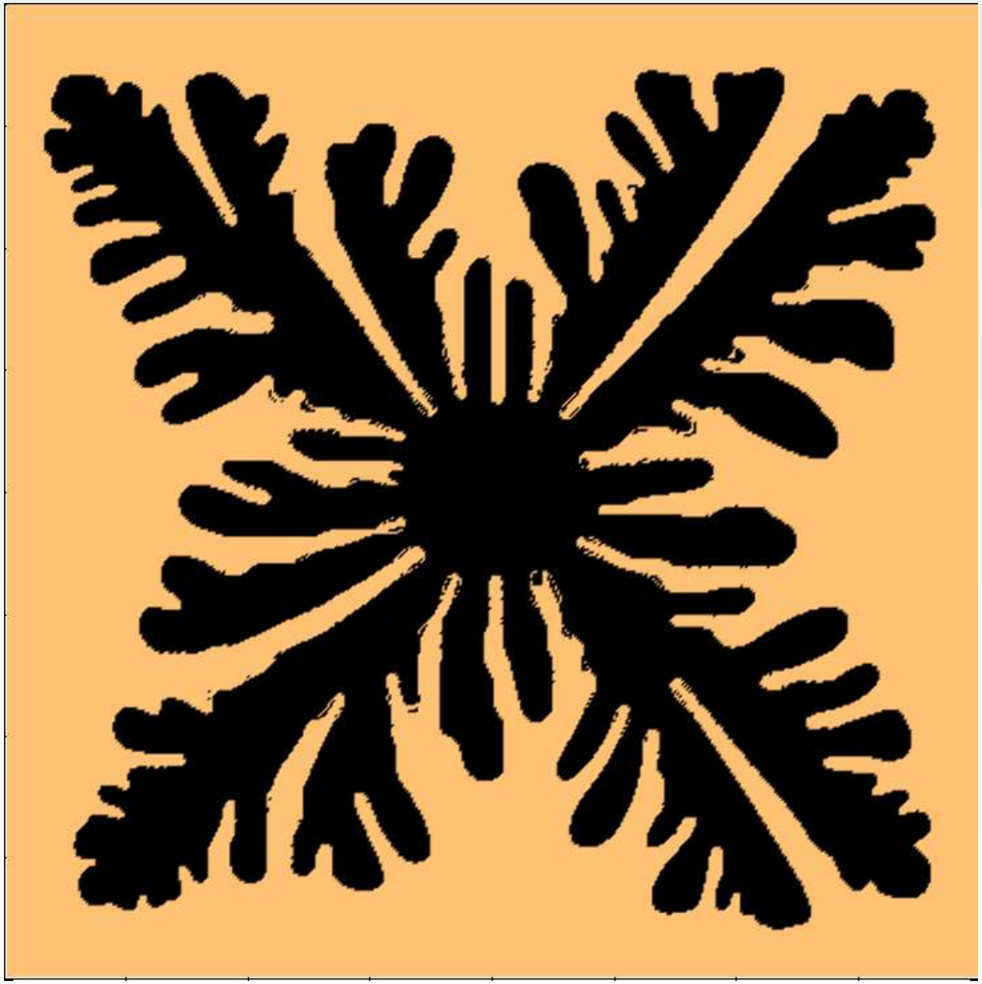
However, in virtually all cases the differential equations that arise in phase field are solved using finite difference or finite element methods utilising regular meshing. This introduces an additional implicit anisotropy due to the periodicity of the mesh. The nature of the implicit anisotropy will depend upon the mesh used, but even for a simple square grid it does not follow that the implicit anisotropy will have simple 4-fold symmetry.

This implicit anisotropy can seriously impede the study of low anisotropy features such as doublons and ‘dendritic seaweed’. Such structures play an important role in rapid solidification research.

Phase-Field Simulation of Doublon Growth



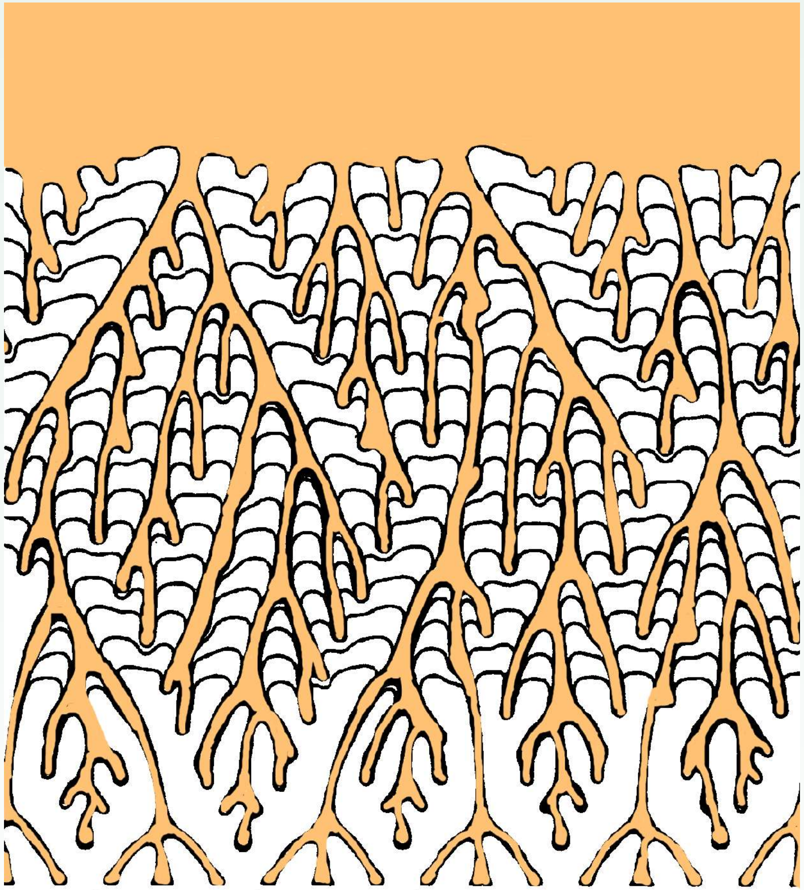
Phase-Field Simulation of Doublon Growth



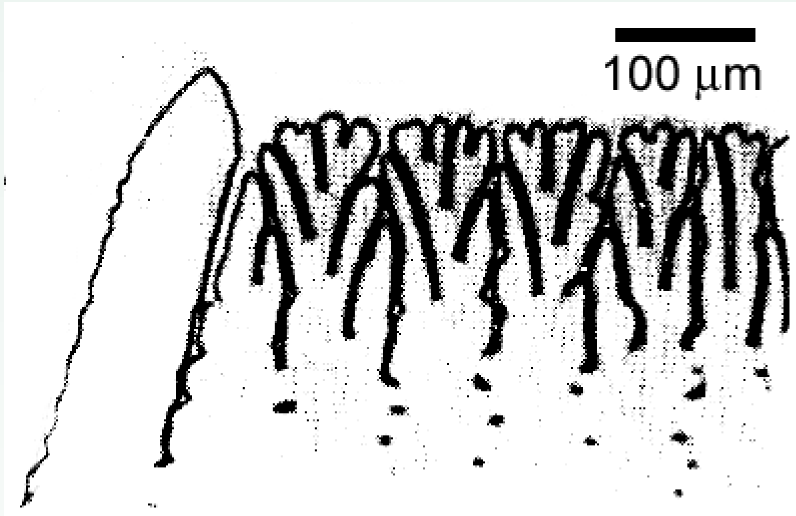
The Dense-Branching 'Seaweed' Morphology

Repeated tip-splitting or doublon formation leads to the creation of the 'seaweed' morphology

From Akamatsu, Faivre & Ihle (1995)



Simulation



Experiment in CBr_4-C_2Cl_6 analogue casting system

Experimental Solidification Research

Solidification is generally regarded as a two stage process;

- Nucleation
- Growth

In order to study the growth stage of this process it is necessary to control nucleation. This requires great care as any solid matter in contact with the melt can act as a *heterogeneous* nuclei. Such heterogeneous nuclei commonly include;

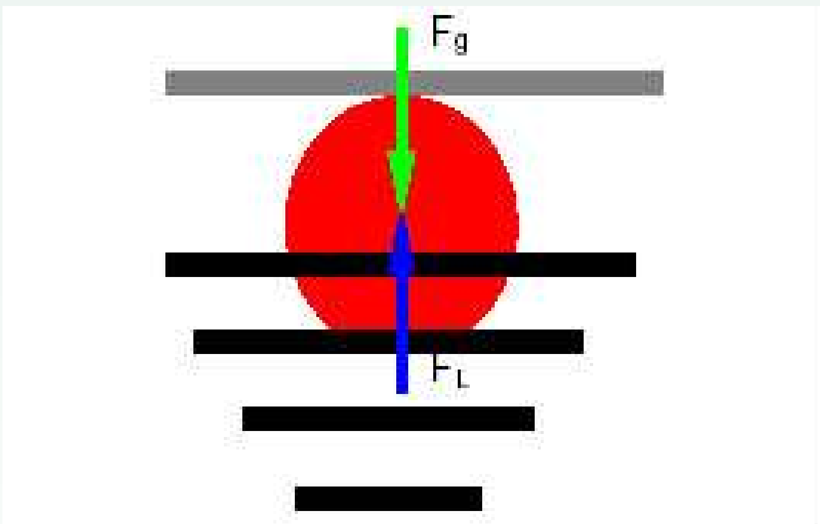
- High melting point impurities
- Oxide films
- The container !

Experimental Solidification Research

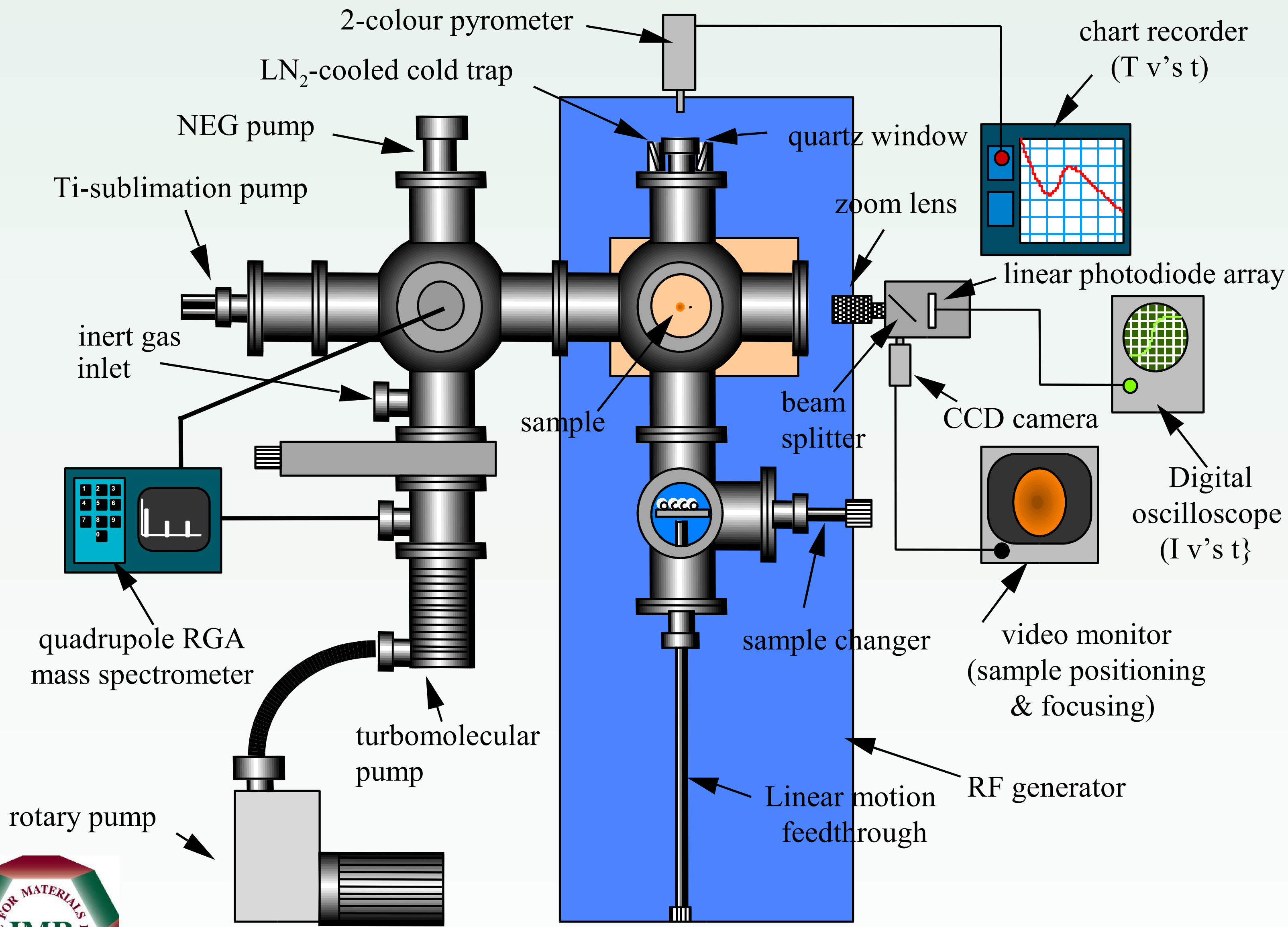
The first of these can be overcome by using high purity materials while the second can be alleviated by working under ultra-clean conditions in an inert or reducing atmosphere. However, to overcome the final condition we need to utilise containerless processing techniques. These include;

- Electromagnet, electrostatic or acoustic levitation
- Free-fall processing, e.g. drop-tubes
- μ-gravity processing such as parabolic flight or orbital experimentation (e.g. Tempus facility aboard ISS).

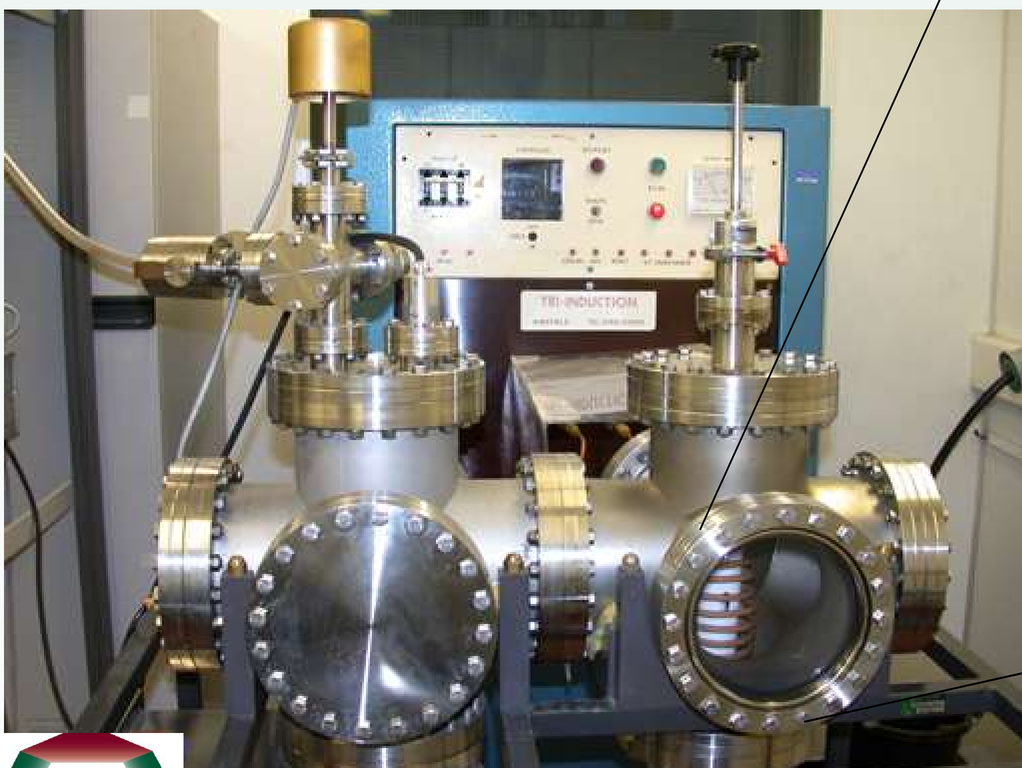
Containerless Processing via Electromagnetic Levitation



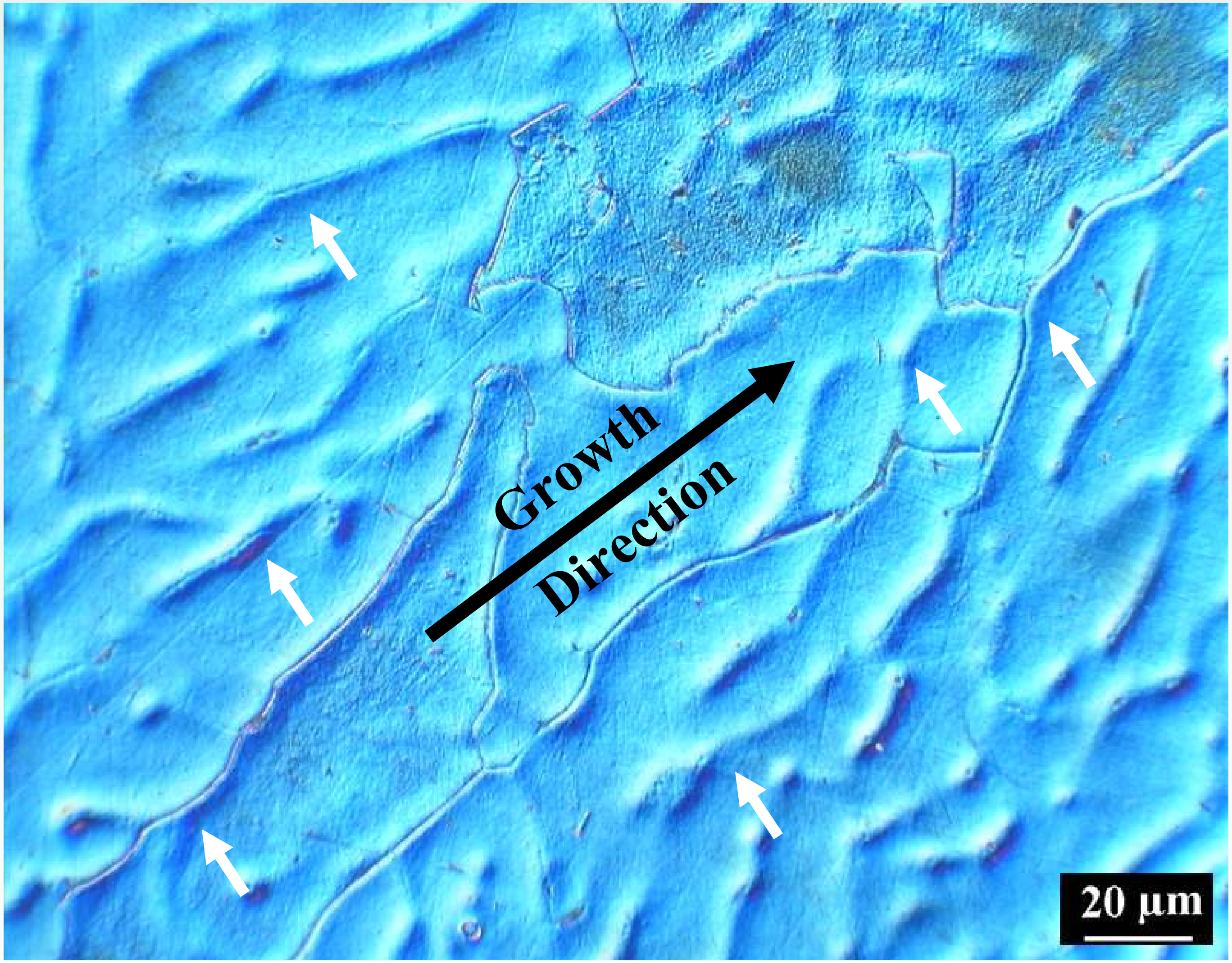
Leeds Levitation/Fluxing Apparatus



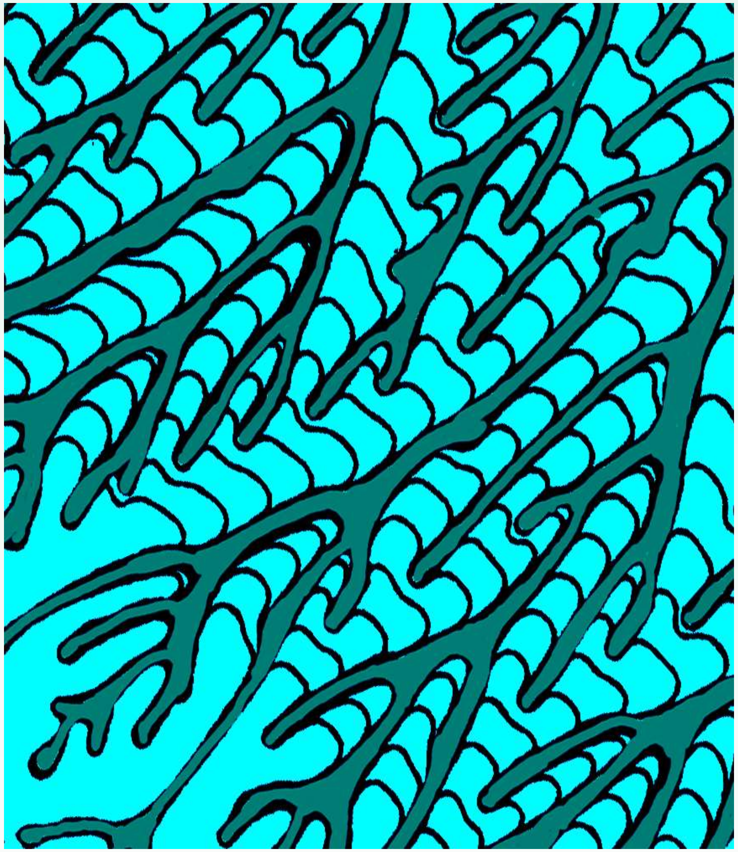
Leeds Levitation/Fluxing Apparatus

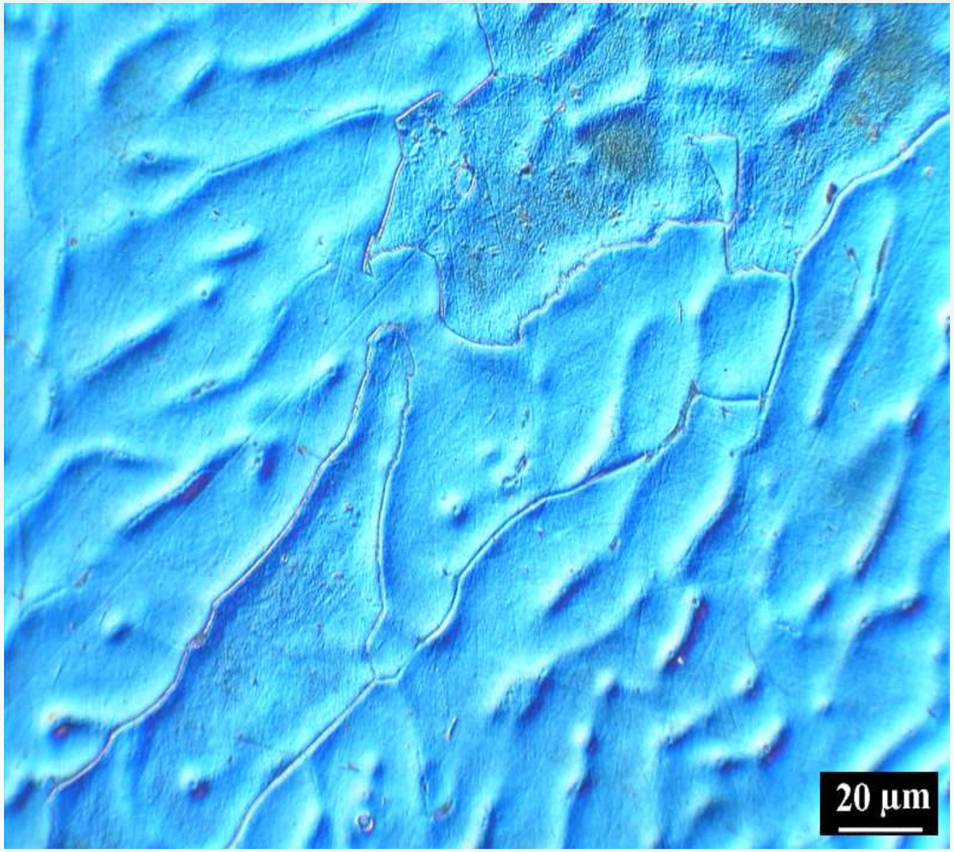


Ultra-high Purity Cu at ΔT = 280 K

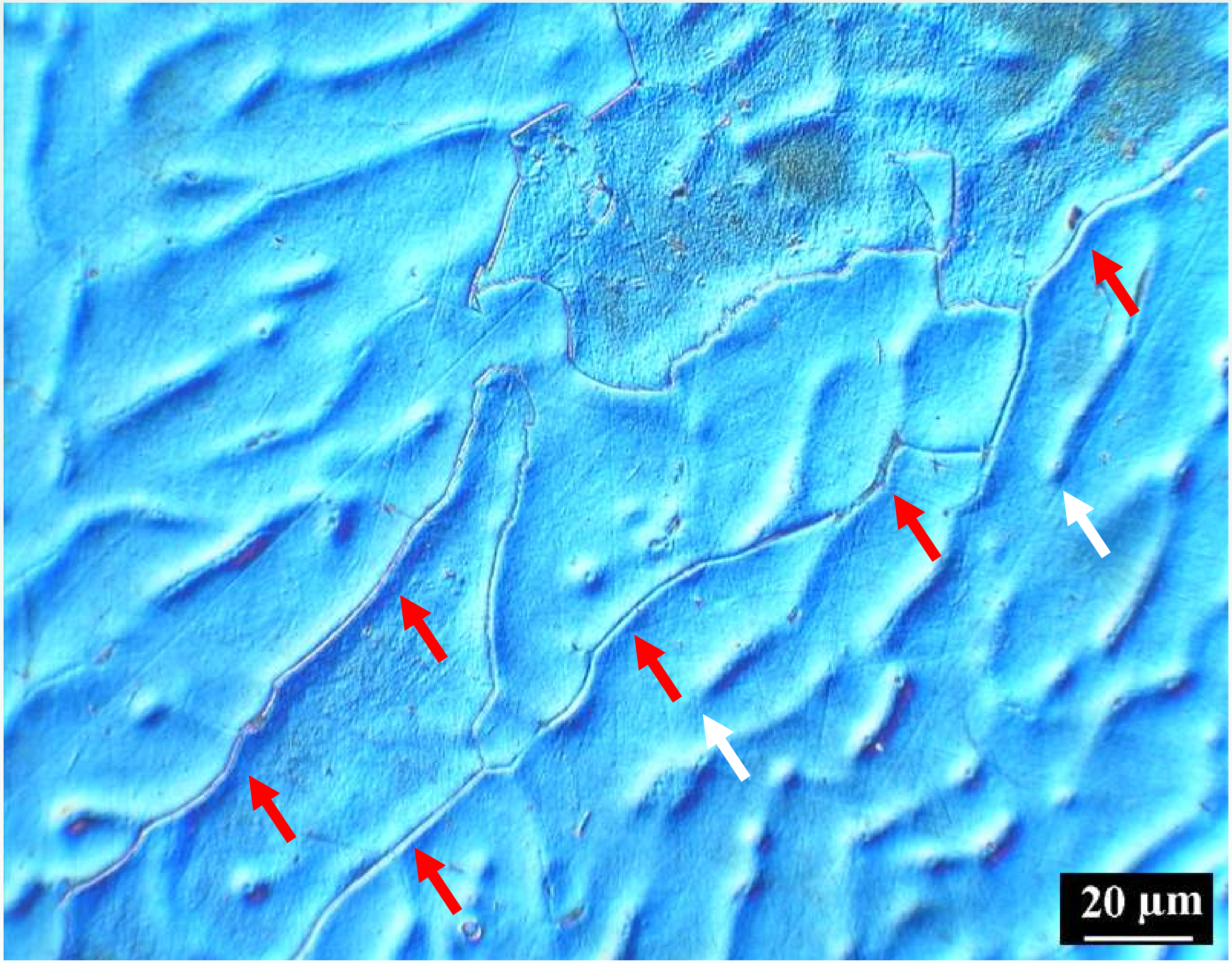


Seaweed Morphology
Comparison of Model & Experiment (?)

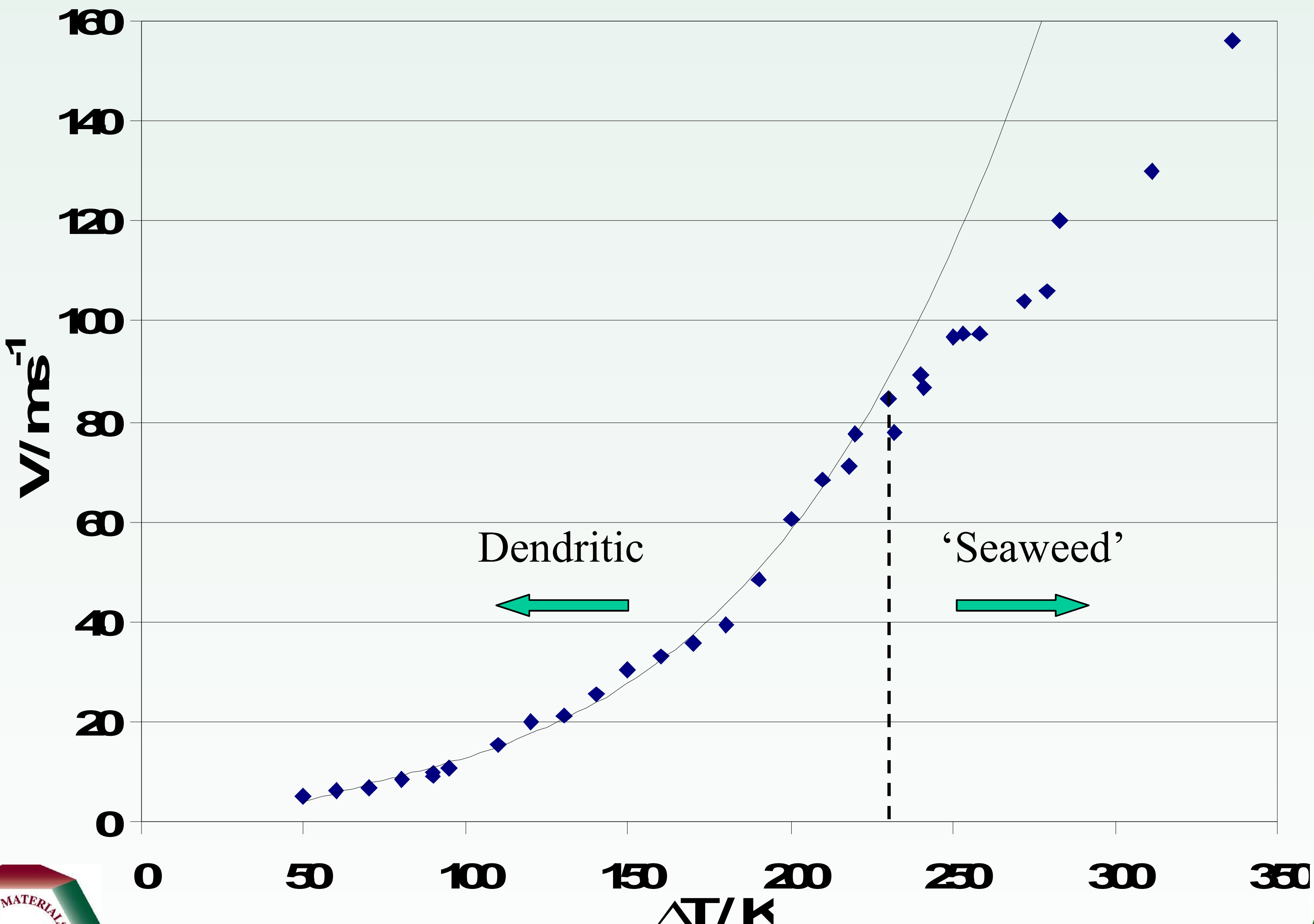




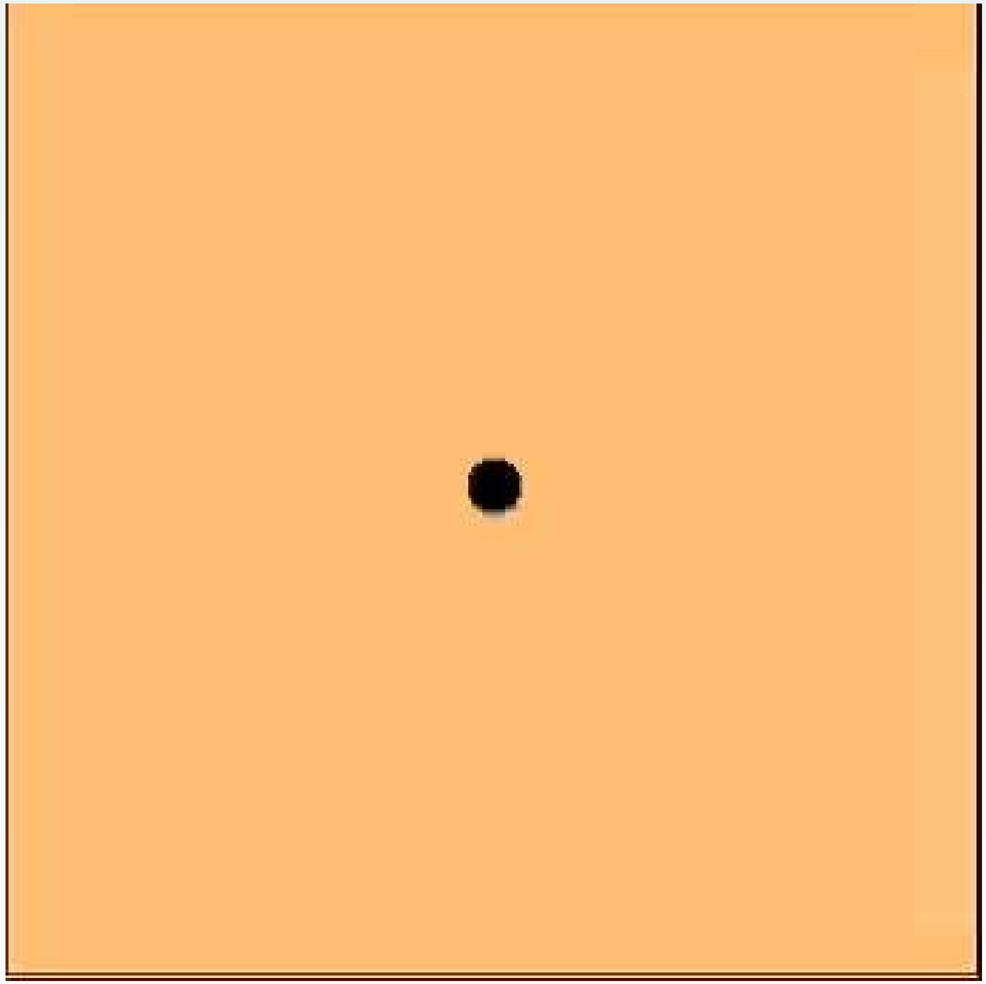
Ultra-high Purity Cu at ΔT = 280 K



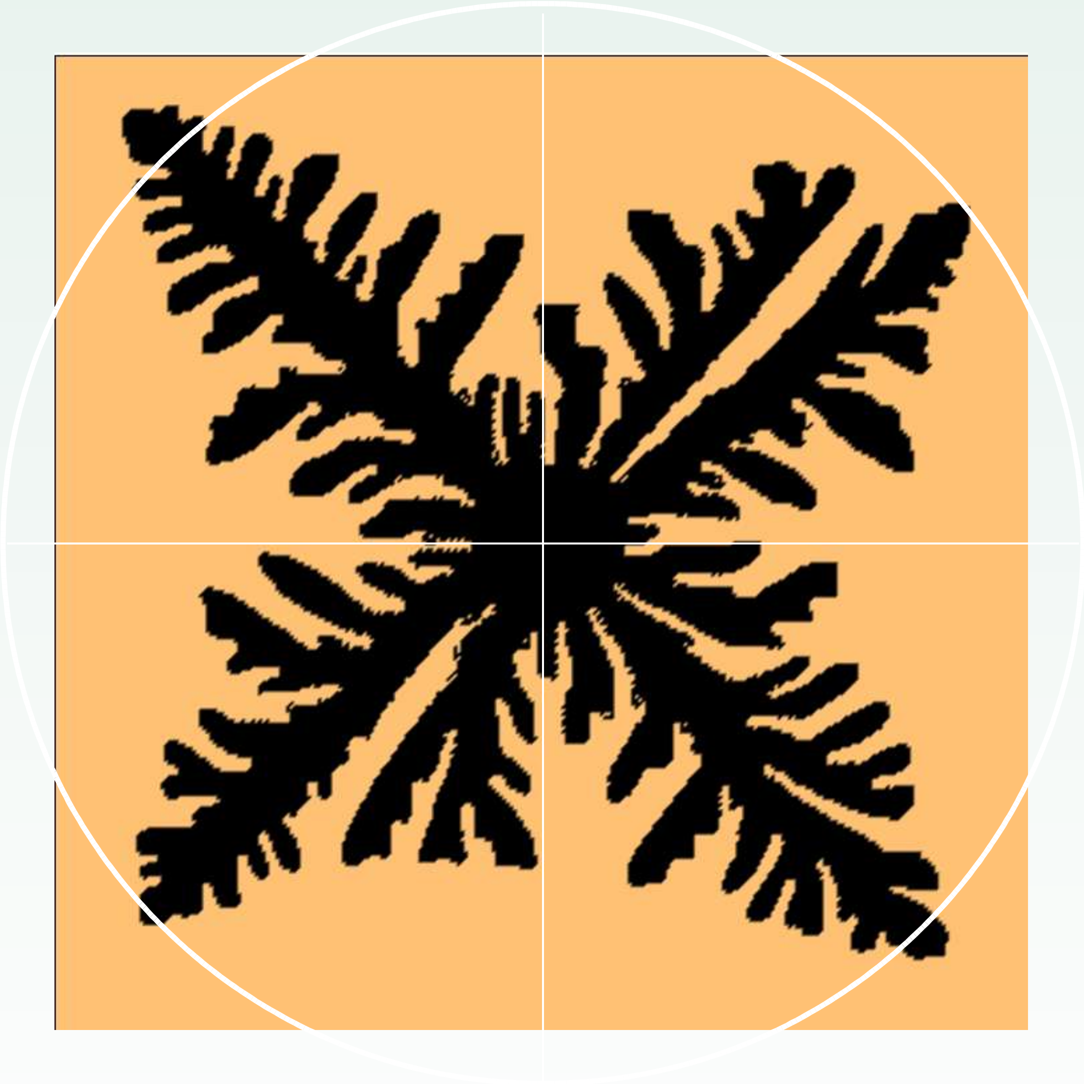
Recalescence Velocity for High-Purity Cu



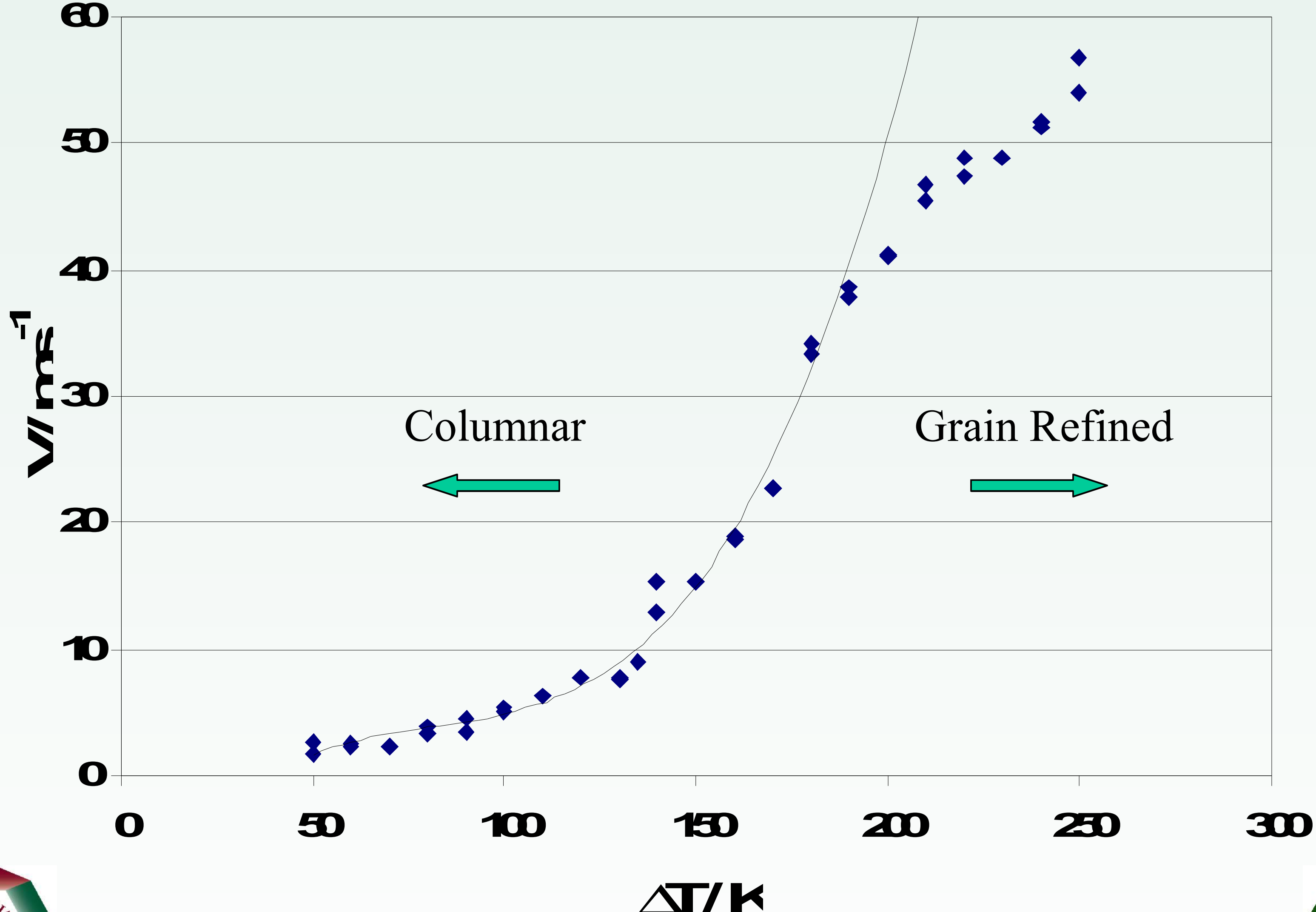
Phase-Field Simulation of Mixed Dendritic/Doublon Growth



Phase-Field Simulation of Mixed Dendritic/Doublon Growth

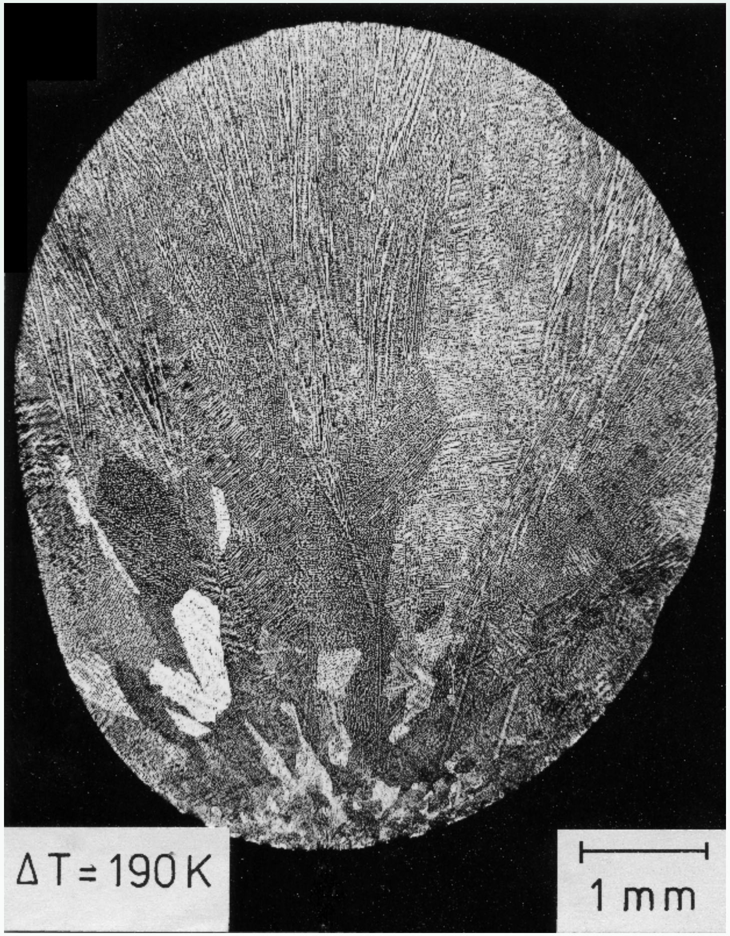


Correlation with Recalescence Velocity

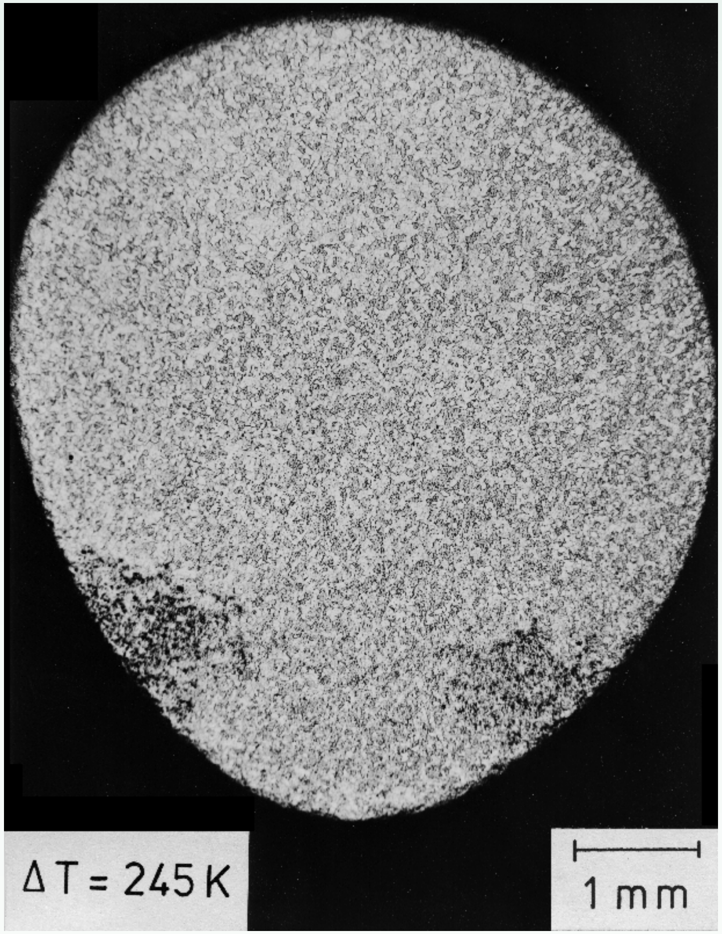


Spontaneous Grain Refinement

Images from Herlach *et. al.*

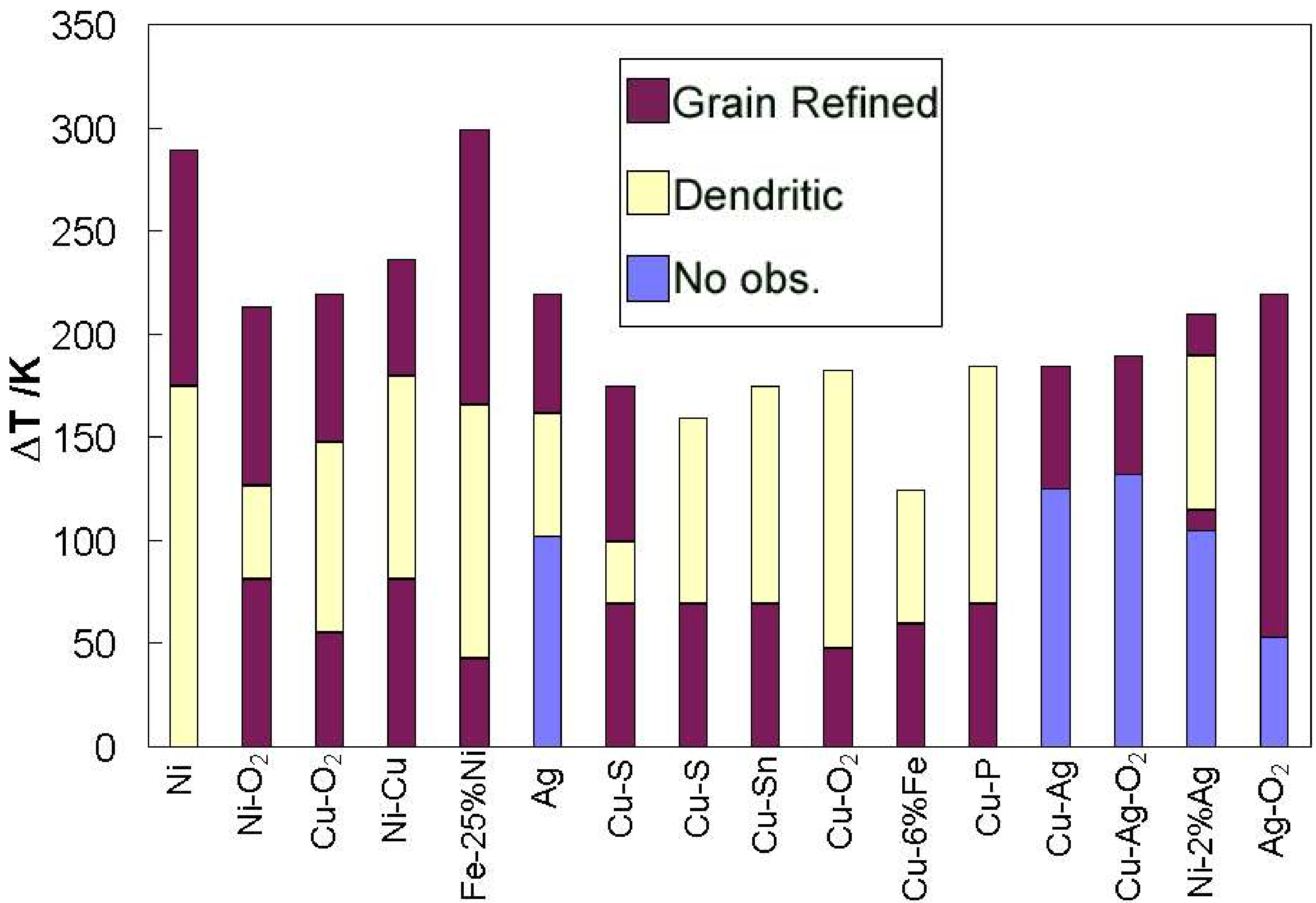


Non grain refined droplet



Grain refined droplet

Grain Refinement by Material



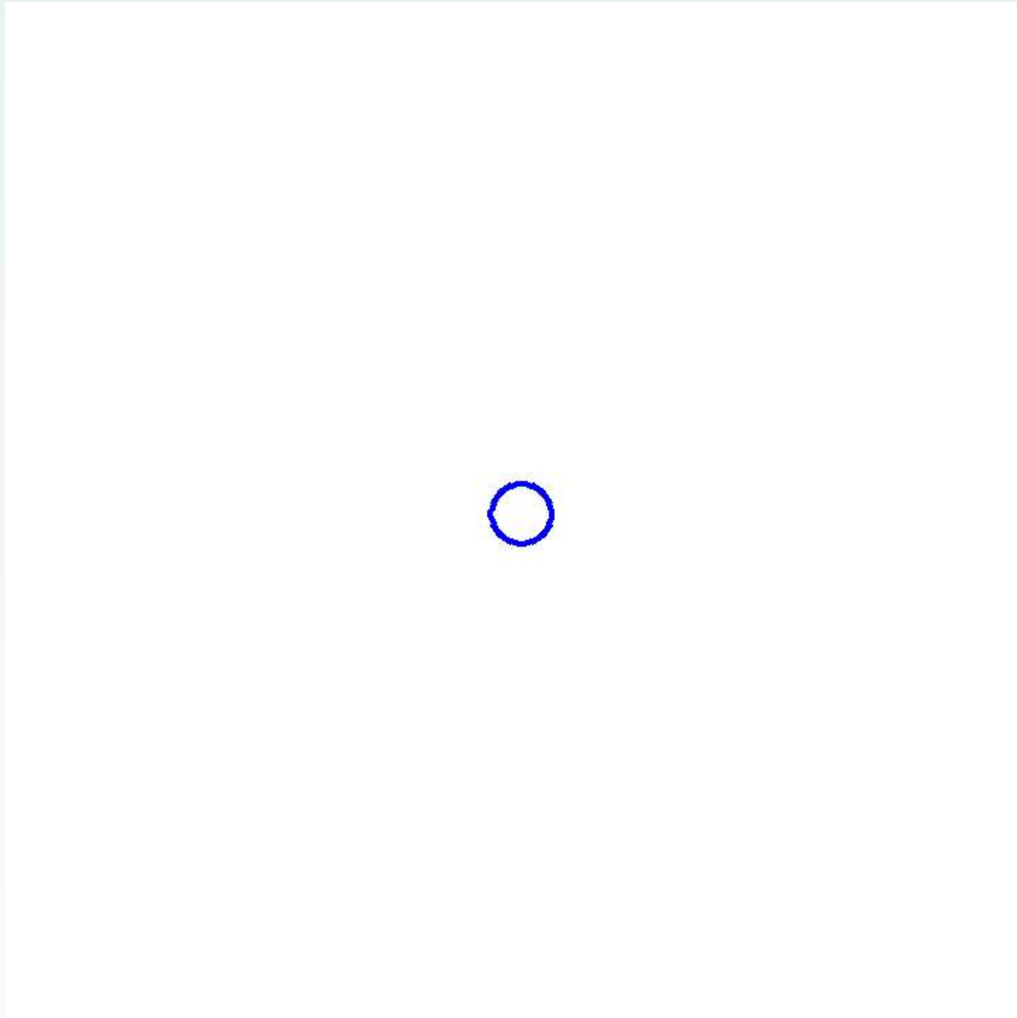
Implicit Anisotropy

Implicit anisotropy seems to be much more severe in solutal than in thermal phase-field models. Our initial suspicion was that this was due to the difference in length scale between the thermal and solutal boundary layers. Typically the thermal boundary layer, ξ, will be large relative to the dendrite tip radius, *R*, (typically $\xi > 10R$), whereas the solutal boundary layer will be small relative to *R* ($\xi > R/10$), We conjectured that because the solutal boundary layer see far fewer grid cells, the directionality introduced by the grid is greater.

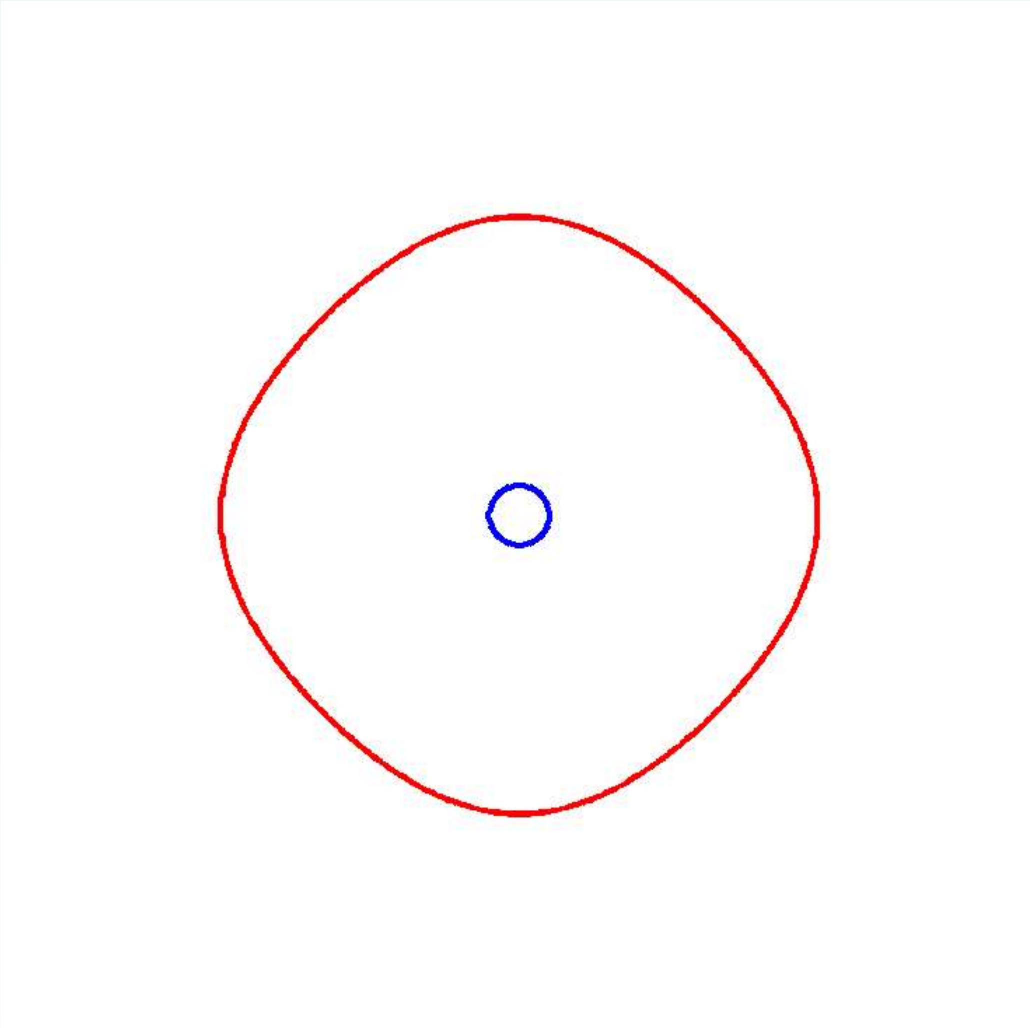
Implicit Anisotropy

To understand the origins of the implicit anisotropy we have adopted the following methodology. A phase-field model has been used to grow a small circular region of solid. The departure of the solid from a true circle has been measured and an opposing 4-fold anisotropy has been added to the model to correct the shape of the solid. When the solid grows exactly as a circle, the implicit anisotropy is taken to be equal to the introduced anisotropy *to first order*.

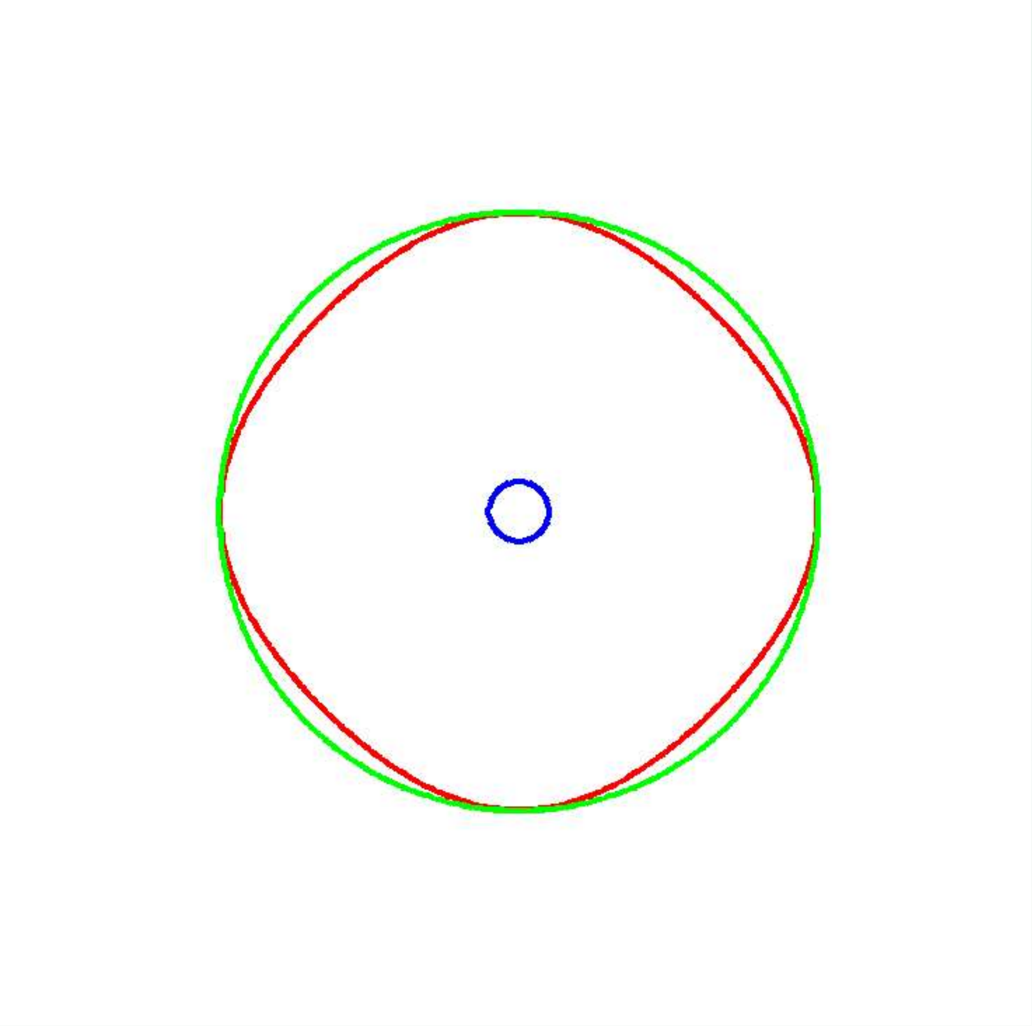
Implicit Anisotropy



Implicit Anisotropy



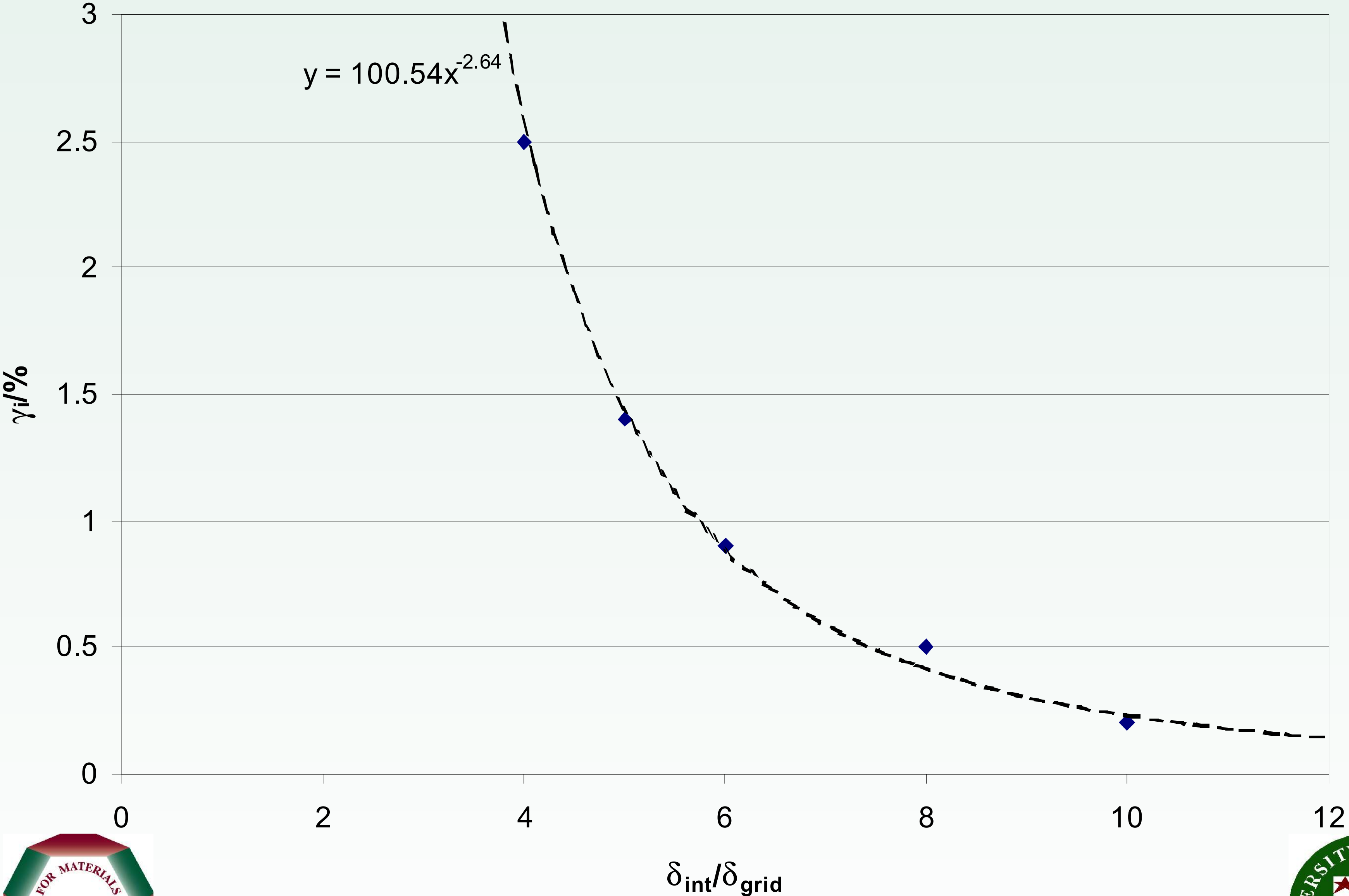
Implicit Anisotropy



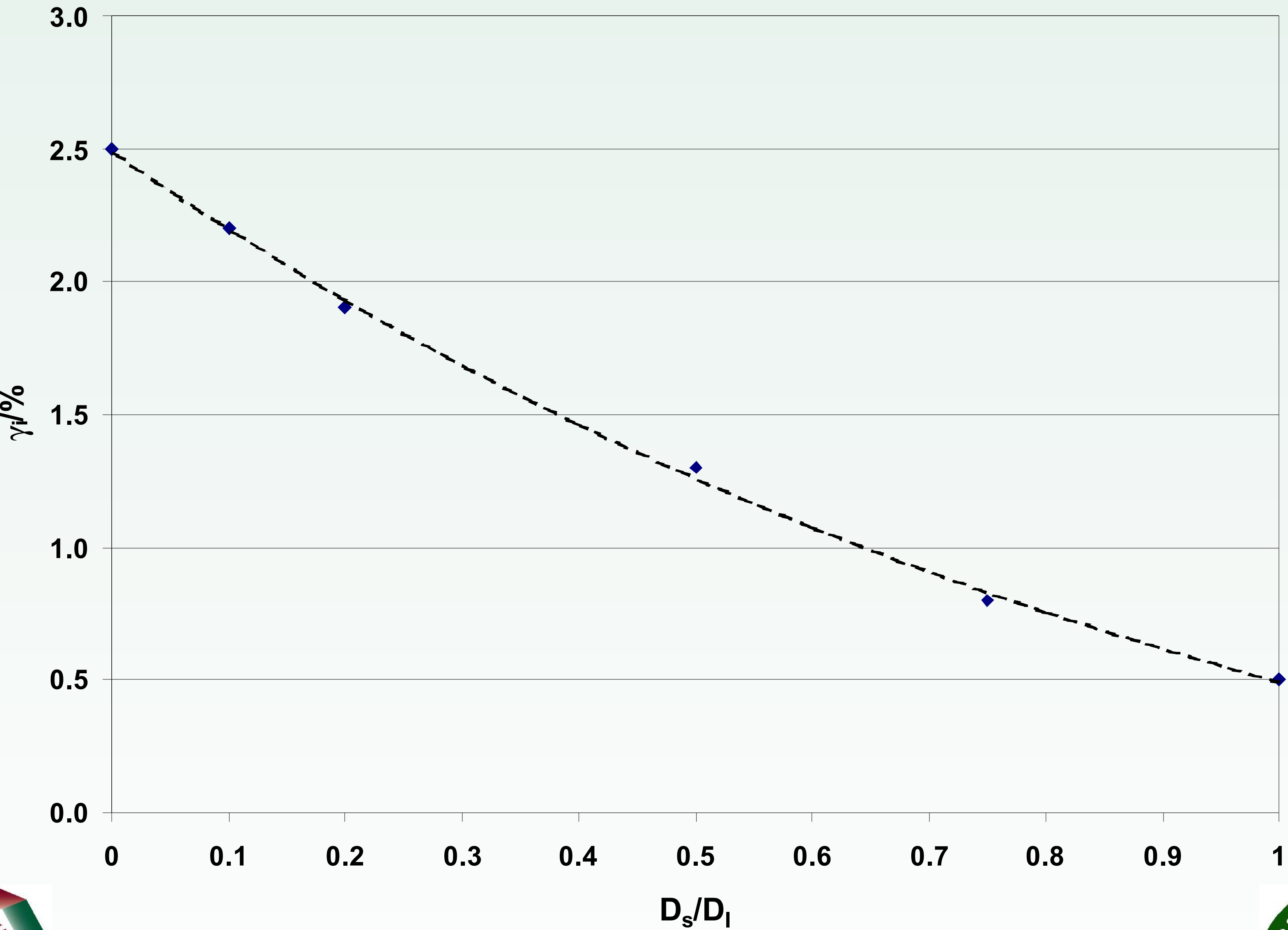
Implicit Anisotropy

On the basis that the implicit anisotropy, γ_i, is related to the width of the solute boundary layer we first investigated the dependence of γ_i on the diffusion coefficient, D_l. No dependence was found.

Implicit Anisotropy



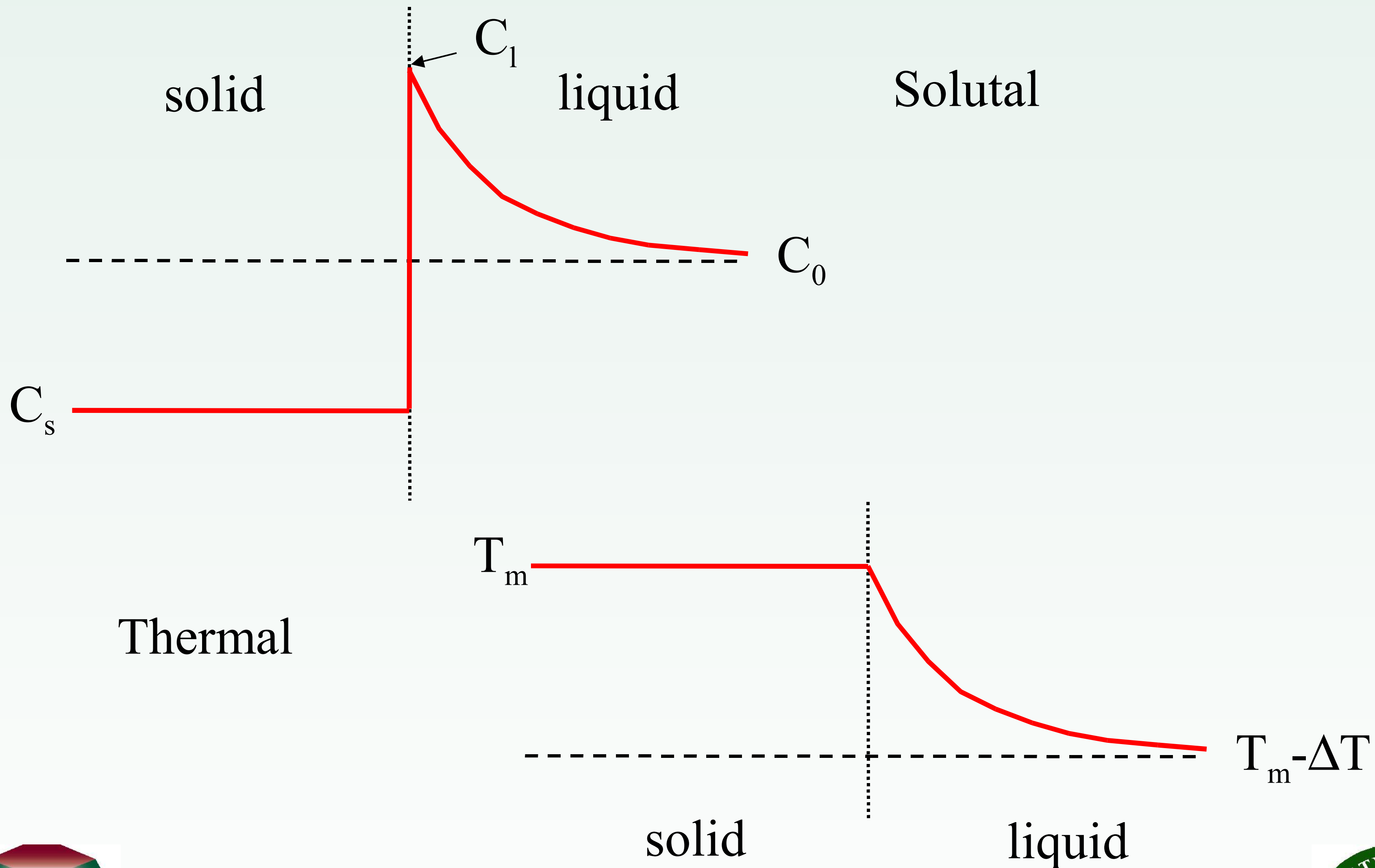
Implicit Anisotropy



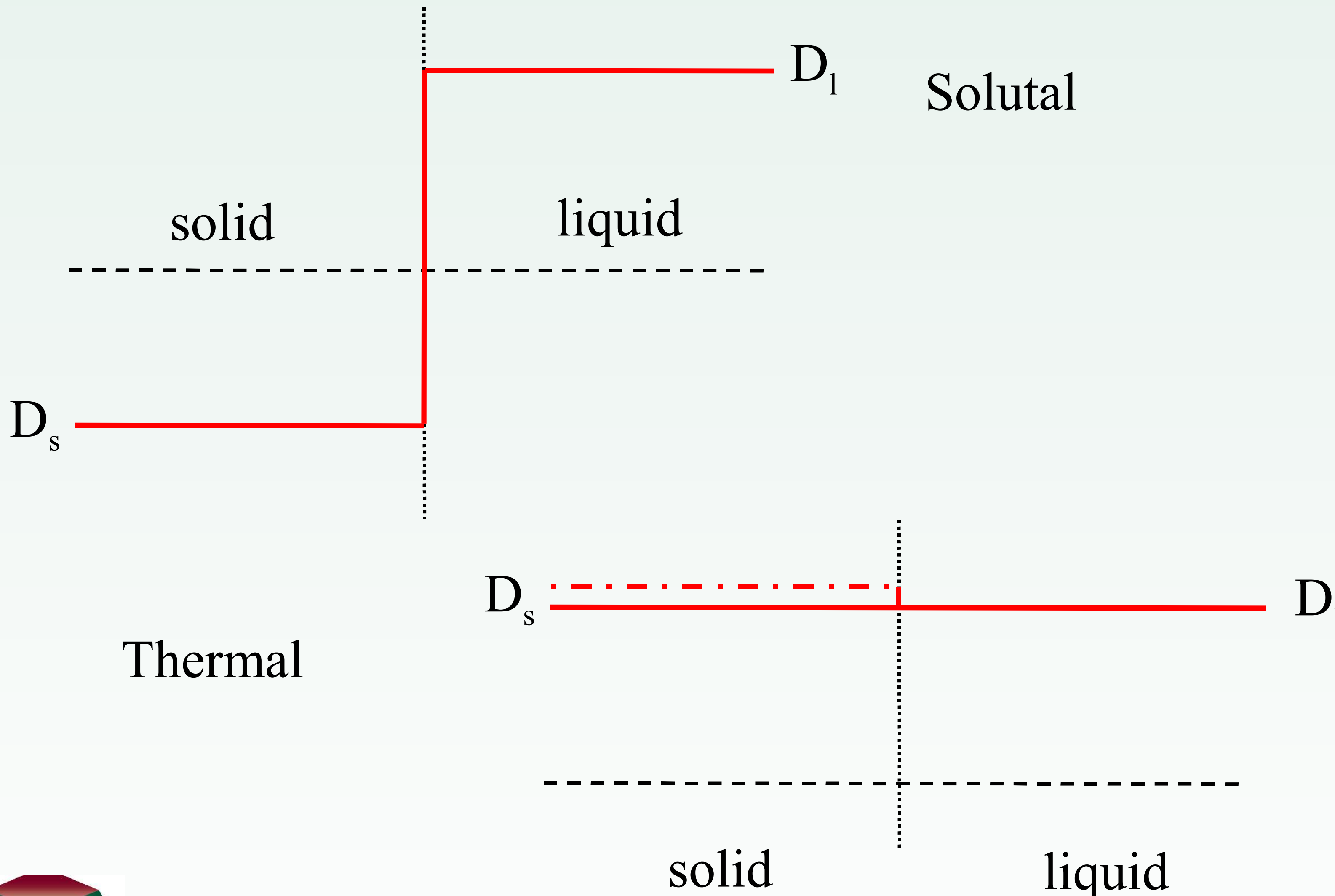
Implicit Anisotropy

O(C)	O(ϕ)	γ_i/%
2	2	2.5
2	4	2.6
4	2	0.9
4	4	1.0

Comparison of Thermal & Solutal Models



Comparison of Thermal & Solutal Models



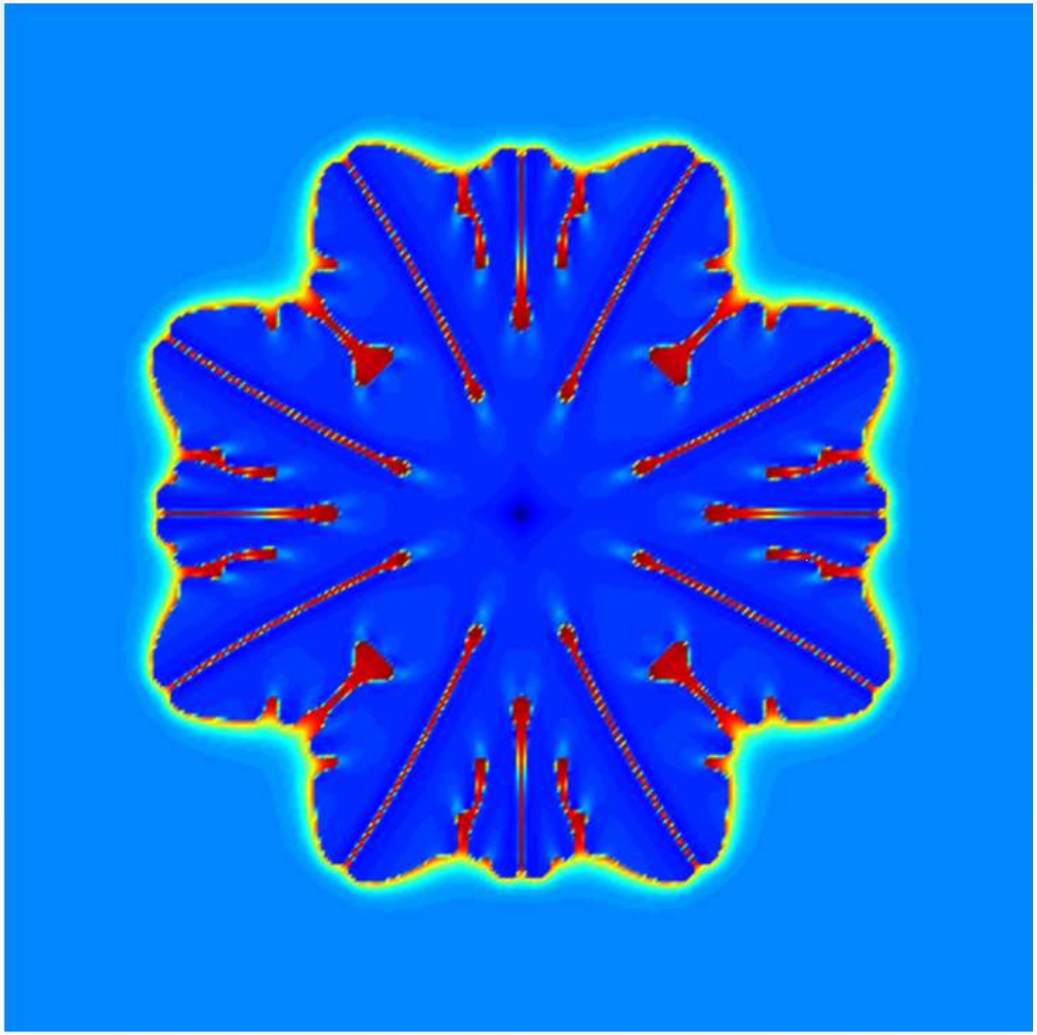
Implicit Anisotropy

Consequently we believe that the strong implicit anisotropy seen in solutal phase-field models may be due to the 'discontinuity' in both C and D. Note that the transport equation contains both ∇C and ∇D terms which are 'smeared out' over the thin interface region. Both of these terms can be potentially very large in the interface region. It appears to be that as these large derivatives sample a very small number of grid points that this is what produces the observed implicit anisotropy in the solutions.

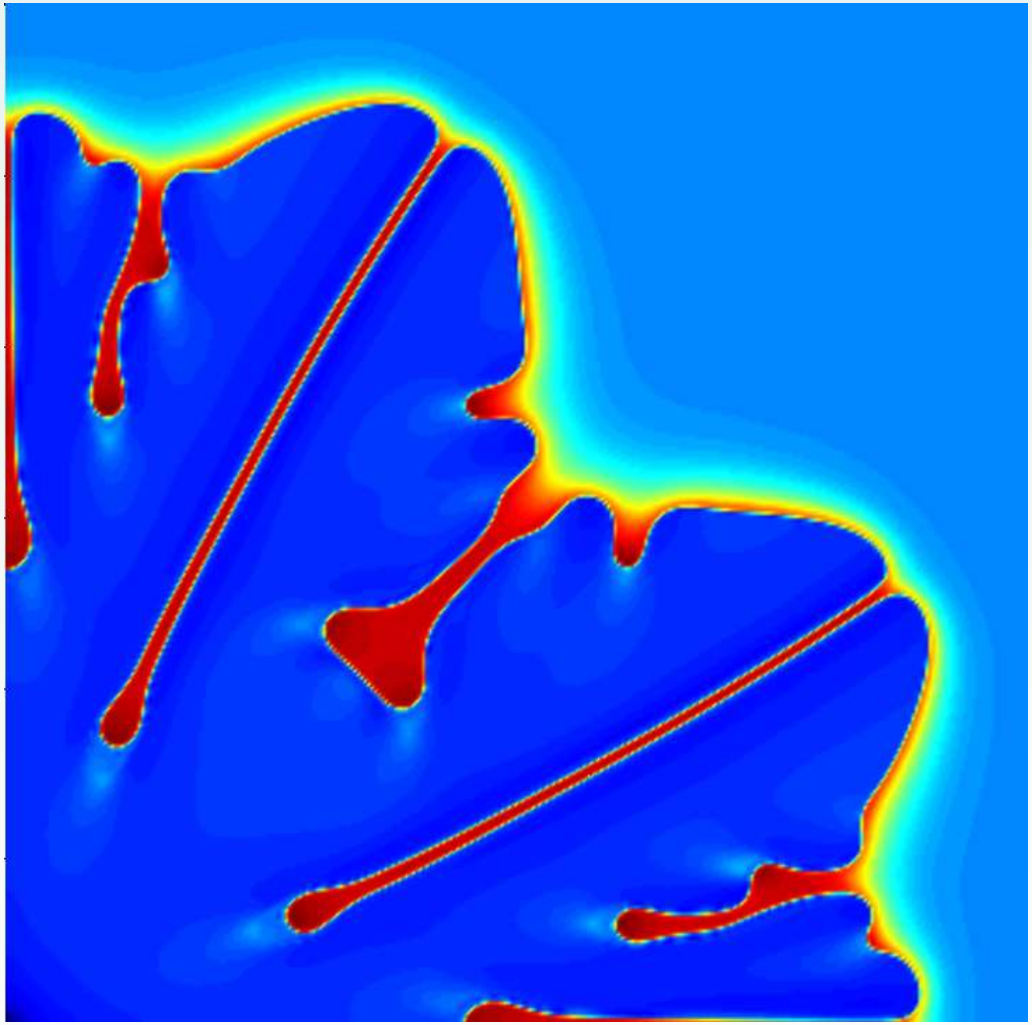
Implicit Anisotropy

The implicit anisotropy is not a simple 4-fold function, so it is not easy to simply introduce a compensating anisotropy to cancel out the implicit anisotropy introduced by the grid. When this is attempted complex periodic structures result.

Implicit Anisotropy (with cancelling) Simple Square Mesh



Implicit Anisotropy (with cancelling) Simple Square Mesh

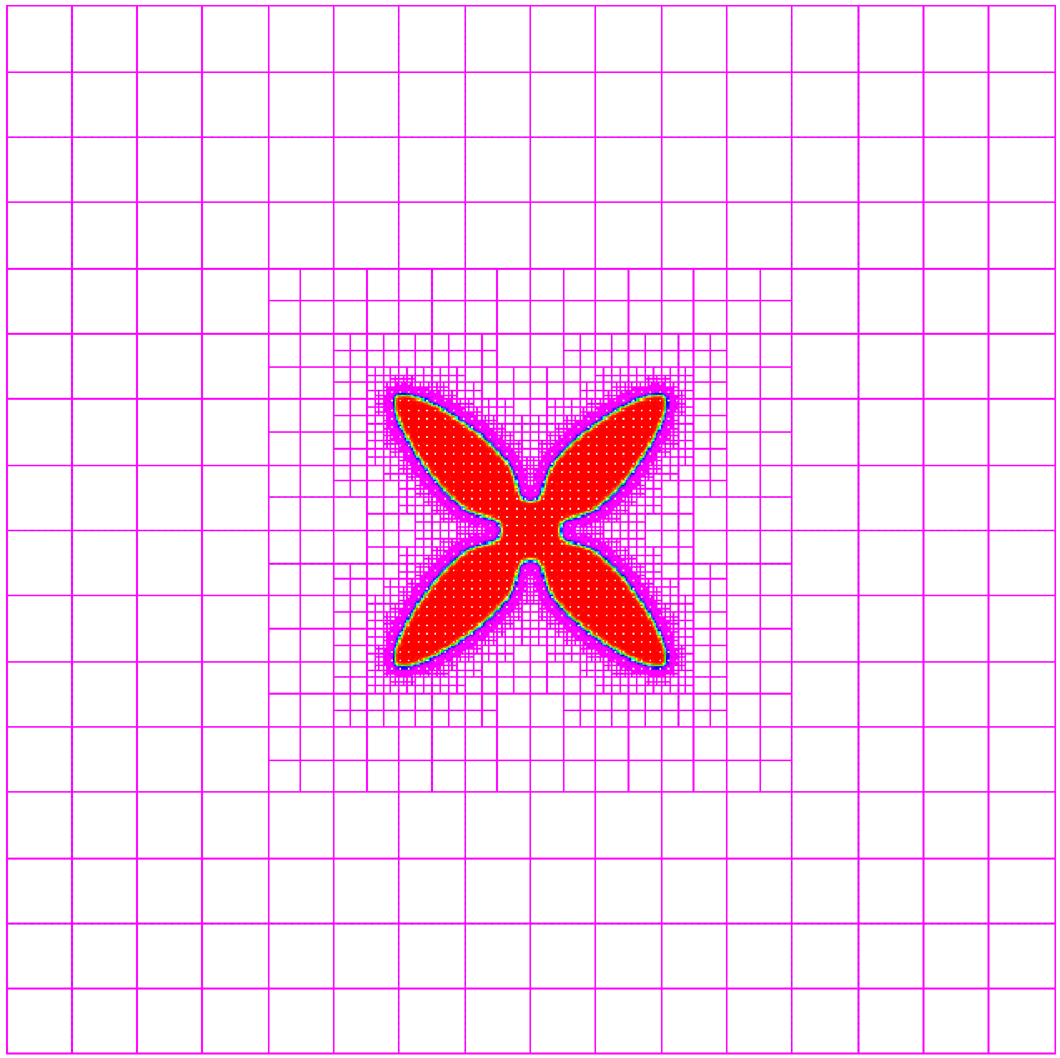


Implicit Anisotropy

Simply making the computational mesh finer is a very crude (& computationally expensive) answers to the implicit anisotropy problem. Note that as explicit solvers are still widely used in phase field the time-step often scales as $1/(\delta x)^2$, so that the actual work involved to evolve through a given time scales as $(\delta x)^4$. Mitigating techniques *could* include:

- Adaptive meshing (more grid points in the interface region)

Adaptive Meshing



Implicit Anisotropy

Simply making the computational mesh finer is a very crude (& computationally expensive) answers to the implicit anisotropy problem. Note that as explicit solvers are still widely used in phase field the time-step often scales as $1/(\delta x)^2$, so that the actual work involved to evolve through a given time scales as $(\delta x)^4$. Mitigating techniques *could* include:

- Adaptive meshing (more grid points in the interface region)
- Implicit solvers (larger time steps on fine meshes)
- Multiple rotated grids (anisotropy cancelling)
- Unstructured grids ? (no implicit anisotropy)

The Phase-Field Model

Our phase-field model is based on that of Wheeler et al. (1993). Evolution of the phase variable, ϕ, is given by

$$\frac{\tilde{\varepsilon}^2}{m}\frac{\partial \phi}{\partial \tau} = \phi(1-\phi)\left[\phi - \tfrac{1}{2} + 30\tilde{\varepsilon}\alpha\Delta u \phi(1-\phi)\right] + \tilde{\varepsilon}^2 \nabla^2 \phi$$

while evolution of the dimensionless temperature, u, $T = T_m + u\Delta T$

is given by

$$\frac{\partial u}{\partial \tau} + \frac{1}{\Delta} p'(\phi)\frac{\partial \phi}{\partial \tau} = \nabla^2 u$$

Here

$$p(\phi) = \phi^3(10 - 15\phi + 6\phi^2)$$

$$\alpha = \frac{\sqrt{2} w L^2}{12 c \sigma T_\mathrm{m}} \qquad m = \frac{\sigma T_\mathrm{m}}{\beta D L} \qquad \text{and} \qquad \tilde{\varepsilon} = \frac{\delta}{w}$$

The Phase-Field Model

As in many formulations of the phase-field method anisotropy is introduced by letting the interface width be anisotropic.

$$\tilde{\varepsilon}(\theta) = \bar{\varepsilon}\eta(\theta) = \bar{\varepsilon}(1+\gamma\cos k\theta)$$

In the asymptotic limit of a sharp interface the interface temperature, u, is given by

$$u = -\bar{d}_0\left([\eta(\theta)+\eta''(\theta)]K + \frac{\bar{v}_n}{m[\eta(\theta)]^2}\right)$$

By comparison with the standard form of the Gibbs-Thomson condition for a moving interface with capillary and kinetic anisotropies, γ_d and γ_k

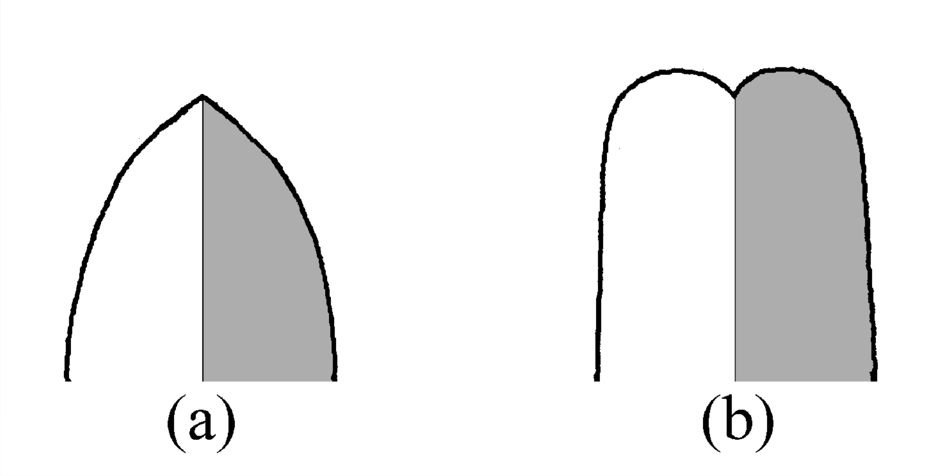
$$T_i = T_m - \frac{L}{c}d(1-\gamma_d\cos k\theta)K - \beta(1-\gamma_k\cos k\theta)v_n$$

We see that our model leads to a fixed ratio, $\gamma_d / \gamma_k = (k^2-1)/2 = 15/2$

Feather Grains & Twin Dendrites

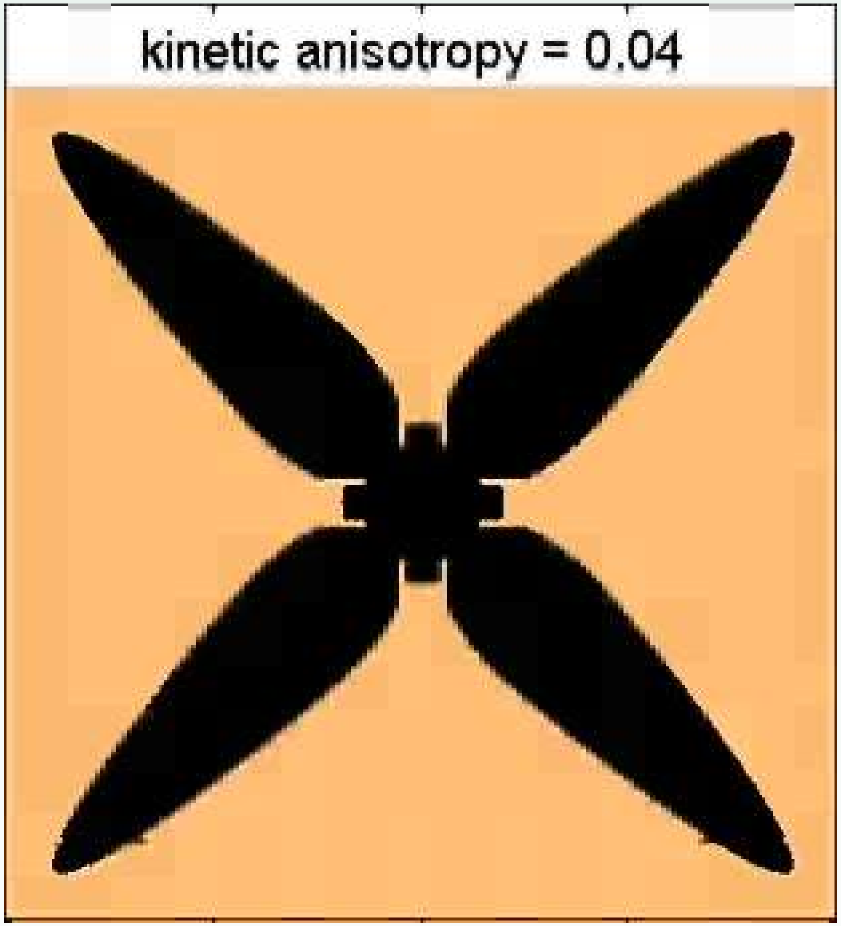
- The formation of 'feather grains' is a significant problem in the DC casting of commercial Al alloys.

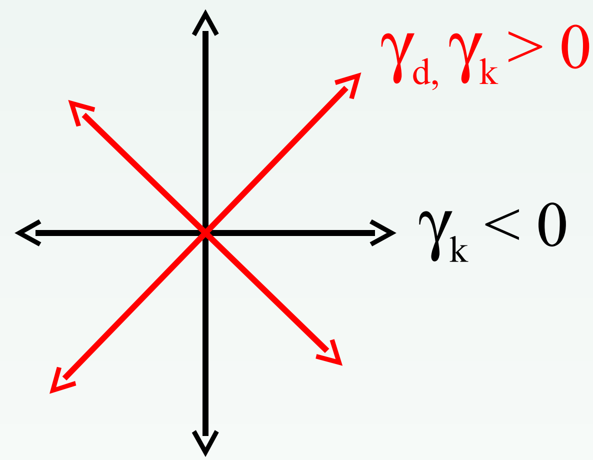
- Crystallographic investigations by Henry *et al.* have shown that feather grains in Al-alloys are twinned dendritic structures.



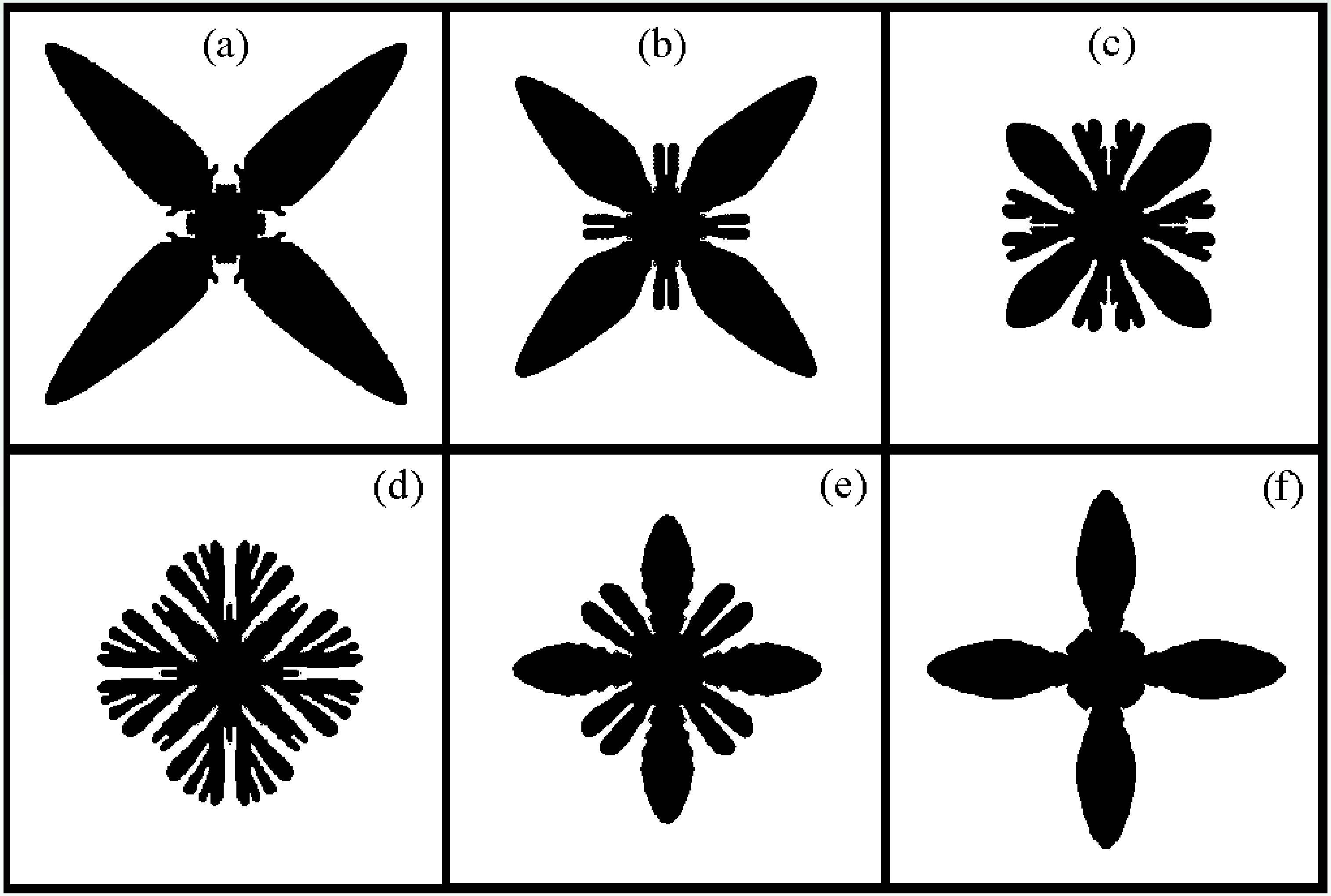
Either of these morphologies might be brought about by a situation where the capillary and kinetic anisotropies are differently directed.

Effect of Varying γ_k





An Anisotropy Competition in Thermal Growth



γ_k and Twin Formation

- Competition between oppositely directed capillary and kinetic anisotropies can result in a low effective anisotropy, giving rise to doublons, even though both γ_d and γ_k are relatively high.
- Boettinger *et al.* have speculated that doublons may play an important role in the formation of feather grains.
- Small crystallographic misorientation is present between adjacent doublon arms, as evidenced in the recent observation of a frozen-in 'seaweed' morphology in as-solidified Cu.