Abstract

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PHASE FIELD MODELING OF CRYSTAL GROWTH

Robert F. Sekerka
University Professor
Physics and Mathematics
Carnegie Mellon University
Pittsburgh, PA 15213

The phase field model is becoming the model of choice for the theoretical study of the morphologies of crystals growth from the melt. This model provides an alternative approach to the solution of the classical (sharp interface) model of solidification by introducing a new variable, the phase field, ϕ, to identify the phase. The variable ϕ takes on constant values in the bulk phases and makes a continuous transition between these values over a thin transition layer that plays the role of the classically sharp interface. This results in ϕ being governed by a new partial differential equation (in addition to the PDE's that govern the classical fields, such as temperature and composition) that guarantees (in the asymptotic limit of a suitably thin transition layer) that the appropriate boundary conditions at the crystal-melt interface are satisfied. Thus, one can proceed to solve coupled PDE's without the necessity of explicitly tracking the interface (free boundary) that would be necessary to solve the classical (sharp interface) model. Recent advances in supercomputing and algorithms now enable generation of interesting and valuable results that display most of the fundamental solidification phenomena and processes that are observed experimentally. These include morphological instability, solute trapping, cellular growth, dendritic growth (with anisotropic sidebranching, tip splitting, and coupling to periodic forcing), coarsening, recalescence, eutectic growth, faceting, and texture development. This talk will focus on the fundamental basis of the phase field model in terms of irreversible thermodynamics as well as it computational limitations and prognosis for future improvement. This work is supported by the National Science Foundation under grant DMR 9211276

PHASE FIELD MODELING OF CRYSTAL GROWTH

BY

ROBERT F. SEKERKA

UNIVERSITY PROFESSOR
PHYSICS AND MATHEMATICS
CARNEGIE MELLON UNIVERSITY

AND

SHUN-LIEN WANG
(TMA)

1.The Phase Field Model

(Langer 1978, 1986; Collins and Levine 1985)

a. Irreversible thermodynamic basis

Based on entropy functional (Penrose and Fife 1990)

$$S = \int_V [s(e,\phi) - \tfrac{1}{2}\varepsilon^2(\nabla\phi)^2]\, d^3x$$

$$\phi = 0, \text{ solid} \qquad \phi = 1, \text{ liquid}$$

$$E = \int_V e\, d^3x$$

Local positive rate of entropy production (Wang et al. Physica D, 1993) and energy conservation:

$$R_S^{prod} \equiv \dot{S} + \int_A \left(\frac{q}{T}n + \varepsilon^2 \dot{\phi} \nabla\phi \cdot n\right) d^2x \geq 0$$

$$R_E^{prod} \equiv \dot{E} + \int_A (q \cdot n)\, d^2x = 0$$

The latter yields

$$\dot{e} + \nabla \cdot q = 0.$$

Irreversible thermodynamic basis continued

The former yields (illustrated for the isotropic case)

$$\mathbf{R}_S^{prod} = \int_V \left\{ \left(\frac{\partial s}{\partial e}\right)_\phi \dot{e} + \left(\frac{\partial s}{\partial \phi}\right)_e \dot{\phi} - \varepsilon^2 (\nabla\phi)\cdot(\nabla\dot{\phi}) \right\} d^3\mathbf{x}$$

$$+ \int_A \left(\frac{\mathbf{q}}{T}\mathbf{n} + \varepsilon^2 \dot{\phi} \nabla\phi \cdot \mathbf{n}\right) d^2\mathbf{x} \geq 0.$$

After integration by parts,

$$\mathbf{R}_S^{prod} = \int_V \left\{ \mathbf{q}\cdot\nabla\left(\frac{\partial s}{\partial e}\right)_\phi + \left[\left(\frac{\partial s}{\partial \phi}\right)_e + \varepsilon^2\nabla^2\phi\right]\dot{\phi} \right\} d^3\mathbf{x} \geq 0$$

which becomes positive definite if we choose

$$\mathbf{q} = \mathbf{M}_T \nabla\left(\frac{\partial s}{\partial e}\right)_\phi = \mathbf{M}_T \nabla\left(\frac{1}{T}\right);$$

$$\tau\dot{\phi} = \left(\frac{\partial s}{\partial \phi}\right)_e + \varepsilon^2\nabla^2\phi = -\frac{1}{T}\left(\frac{\partial f}{\partial \phi}\right)_T + \varepsilon^2\nabla^2\phi.$$

For computations, choose $\mathbf{M}_T = kT^2 \Rightarrow \mathbf{q} = -k\nabla T.$

Irreversible thermodynamic basis continued

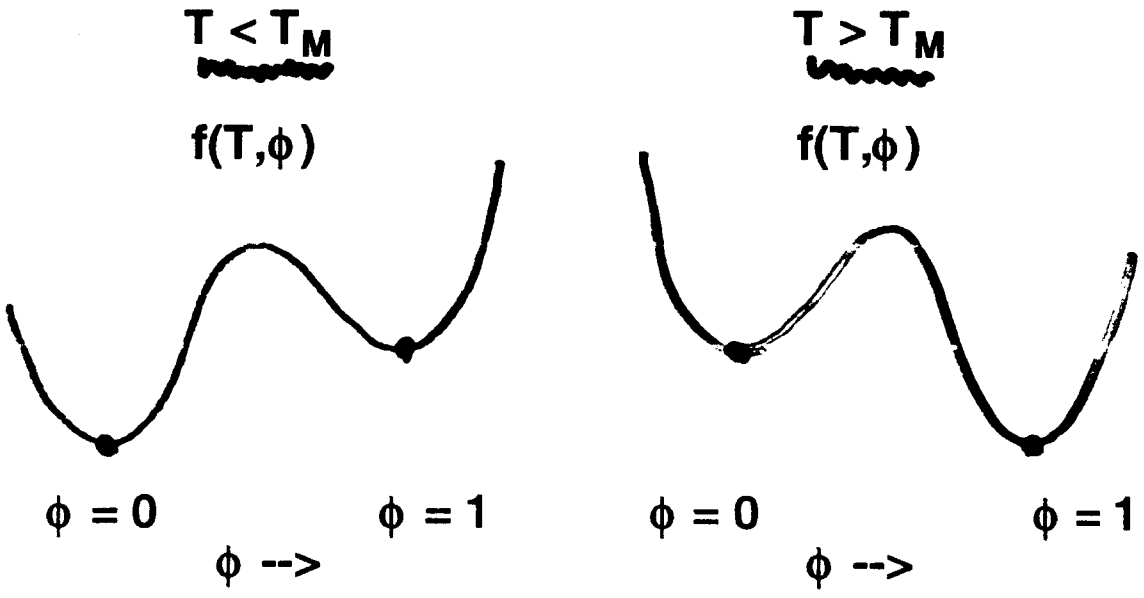
The (Helmholtz) free energy density is then

$$f(T,\phi) = e(T,\phi) - T\int_0^T \frac{1}{T'}\frac{\partial e(T',\phi)}{\partial T'}dT' + \frac{Tg(\phi)}{4a}$$

$$= f_s(T) - p(\phi)\,T\int_{T_M}^T \frac{L(T')}{(T')^2}\,dT' + \frac{Tg(\phi)}{4a}.$$

For the simple case of parallel energy curves near T_M, separated by L_0, one has simply

$$f(T,\phi) = f_s(T) + p(\phi)\frac{L_0}{T_M}(T_M - T) + \frac{Tg(\phi)}{4a}.$$



b. Anisotropy

Computations are done for anisotropic properties:

Let ε and τ depend on Θ, the angle that the vector $\nabla\phi/|\nabla\phi|$ makes with a reference direction (Kobayashi). Then asymptotics (in the manner of Caginalp except anisotropic) leads to the boundary condition

$$T_I = \frac{T_M}{1 + \left[\frac{\gamma(\theta) + \gamma(\theta)_{\theta\theta}}{L_V} K + \frac{V_n}{\mu(\theta) T_M}\right]}$$

where (McFadden et al. 1993)

$$\gamma(\theta) \propto \varepsilon(\theta); \qquad \mu(\theta) \propto \frac{\varepsilon(\theta)}{\tau(\theta)}$$

In computations, we can control separately the anisotropy of surface tension and kinetics.

This nonlinear result agrees with that for the discrete case (Gurtin 1986); the linearized version is

$$T_I = T_M - T_M \frac{\gamma(\theta) + \gamma(\theta)_{\theta\theta}}{L_V} K - \frac{V_n}{\mu(\theta)}.$$

c. Computational considerations for dendrites (Illustrated for isotropic case, dimensionless)

$$\frac{\partial u}{\partial \tilde{t}} + \frac{30 g(\phi)}{S} \frac{\partial \phi}{\partial \tilde{t}} = \tilde{\nabla}^2 u$$

$$\frac{\tilde{\varepsilon}^2}{m} \frac{\partial \phi}{\partial \tilde{t}} = \tilde{\varepsilon}^2 \tilde{\nabla}^2 \phi + 30 g(\phi) \tilde{\varepsilon} \alpha S \frac{u}{1 + S(L_V/cT_M)u} - \frac{g'(\phi)}{4}$$

where u = dimensionless temperature
(0 at melting point, –1 far field)

$m = d_0/\rho_K$ where d_0 is the capillary length
and $\rho_K = \kappa c/\mu L_0$ is the kinetic length
(m very large $\Rightarrow$ local equilibrium)

$\tilde{\varepsilon} = \delta/W$ = interface thickness parameter/system size

$\alpha = W/(6\sqrt{2}\, d_0)$

S = dimensionless supercooling

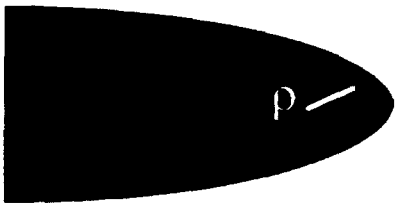
To "resolve" Gibbs-Thomson effect, need $\delta \approx d_0$

To "resolve" interface, need grid spacing $\approx \delta$

$\Rightarrow$ 8 grid points as ϕ goes from 0 to 1 (like tanh)

Computational considerations for dendrites cont.

$$8\Delta x = 8\delta \approx \eta << \rho << W$$



So practical considerations lead to

$\delta = d_0$ to resolve Gibbs-Thomson

$\eta = 8\ d_0$ to resolve interface

$\rho = 80\ d_0$ to resolve the dendrite

$W = 800$ to $3200\ d_0$ to get steady state

Since $\rho = (d_0/\sigma^*)\ P^{-1}$ and $\sigma^* \approx 0.03$, we need $P \approx 0.5$.

This requires very large supercoolings, $S \approx 1$.

4. Results

a. Morphologies at large supercoolings

$S = \Delta T/\Delta T_H = 0.8$

m = (capillary length/kinetic length) = 0.1

Fourfold sinusoidal anisotropies of capillarity and kinetics (% shown below)

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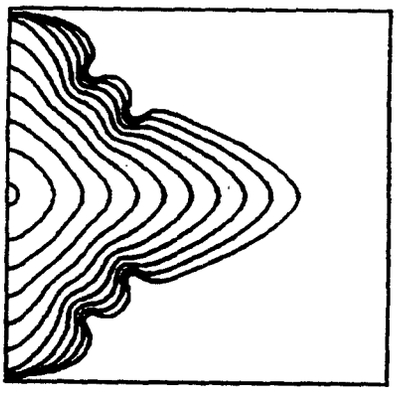

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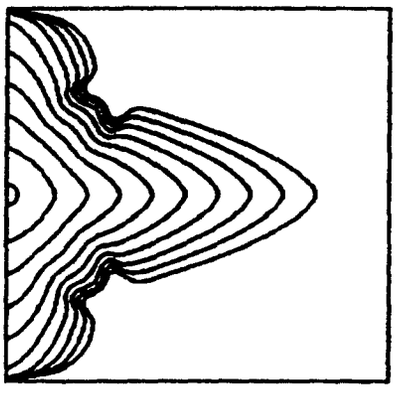

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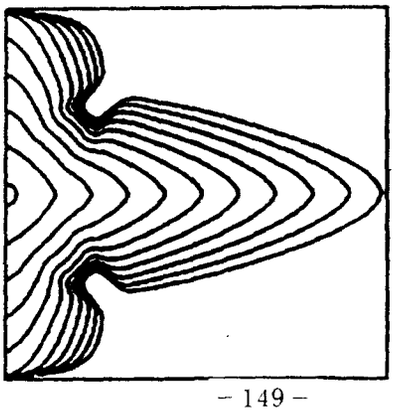
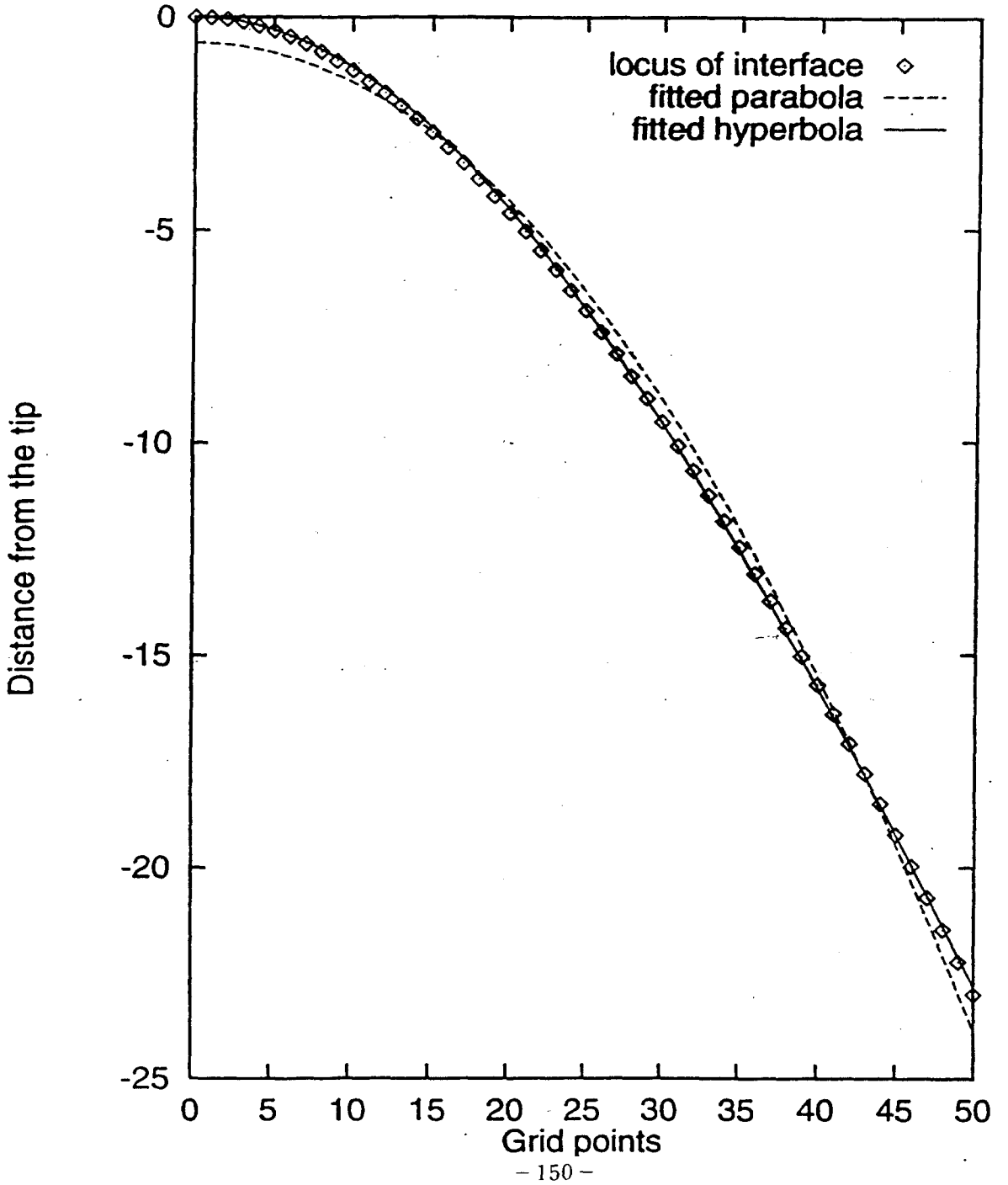


a. Morphologies at large supercoolings cont.

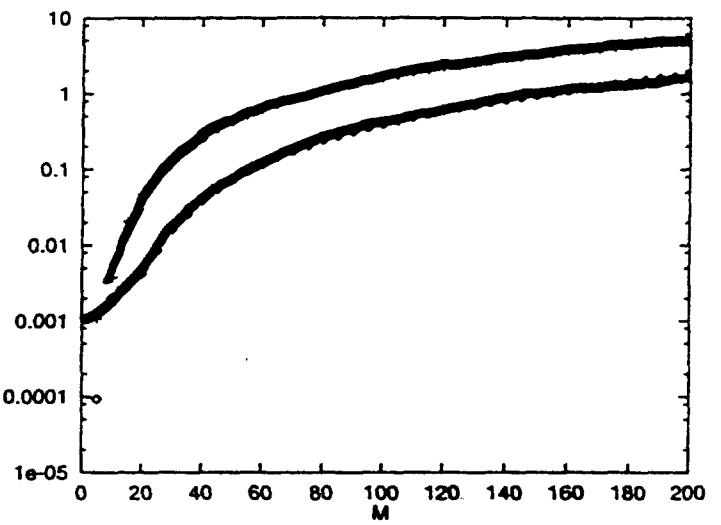
Dendrite tips are more nearly hyperbolic than parabolic (agrees with Brener & Temkin for S > 1)



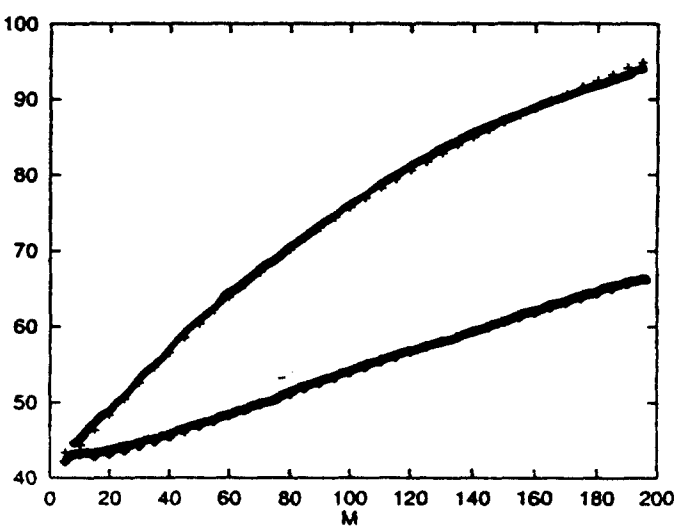
a. Morphologies at large supercoolings cont.

RMS fits for hyperbola or parabola

RMS residue



Tip radius, ρ

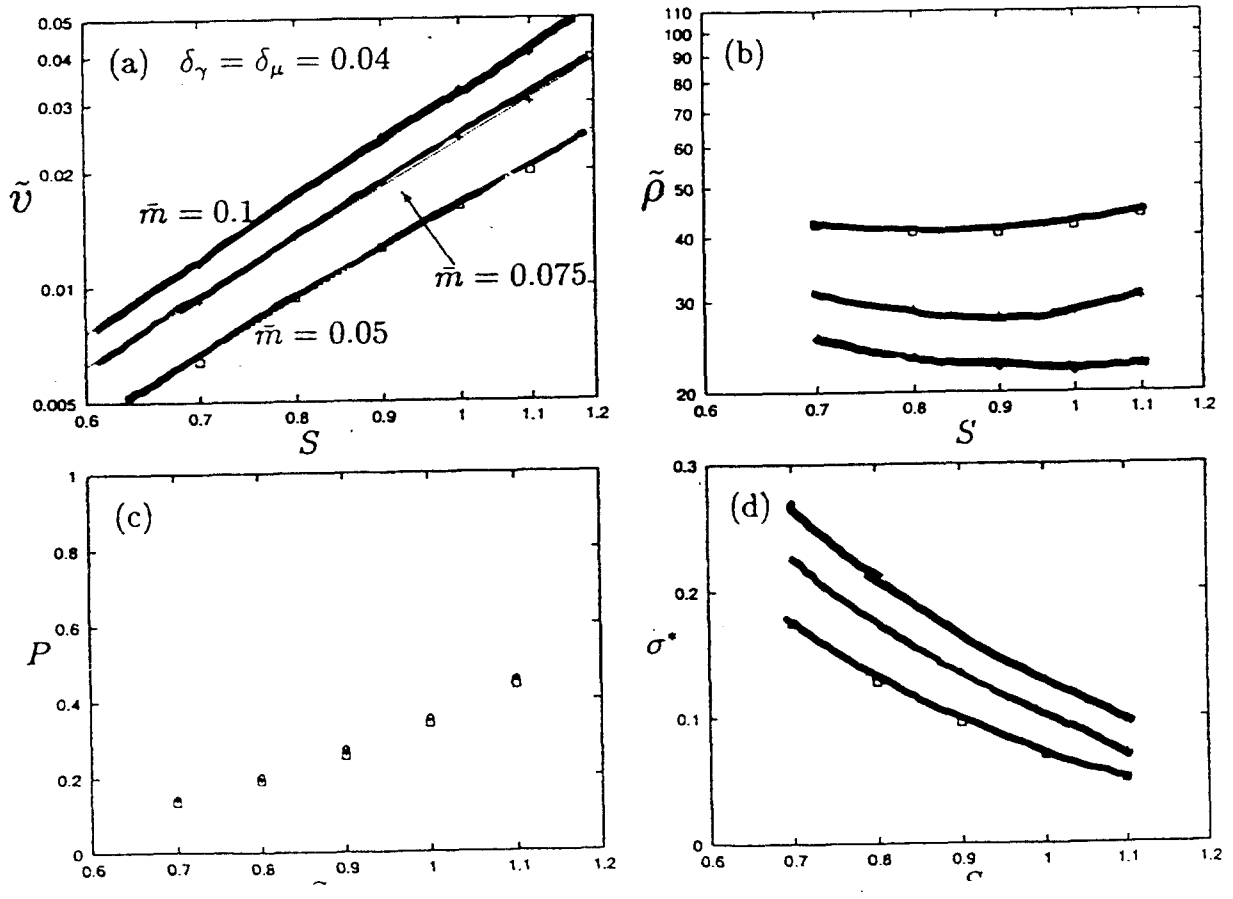


Abscissa is the number, M, of fitted points

b. Examples of power laws

Surface tension and kinetics have anisotropies of 1 + 0.04 cos(4θ)

m = 0.05 relatively strong kinetics
m = 0.075 intermediate kinetics
m = 0.10 relatively weaker kinetics
(toward local equilibrium)



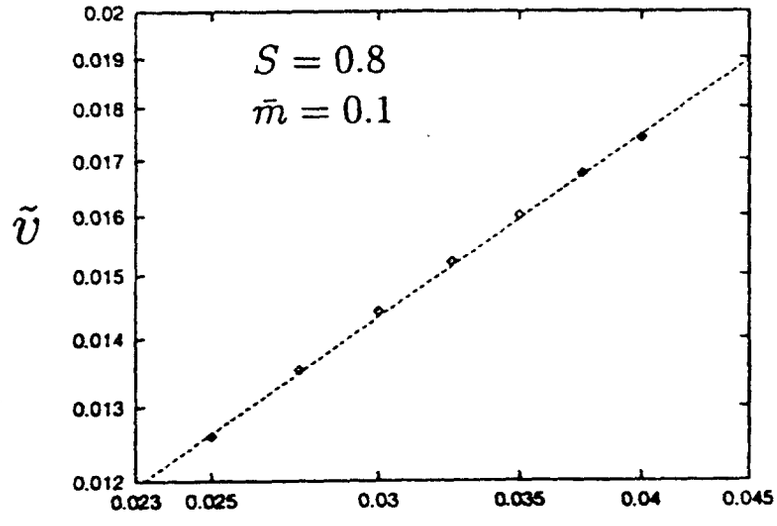
b. Examples of power laws cont.

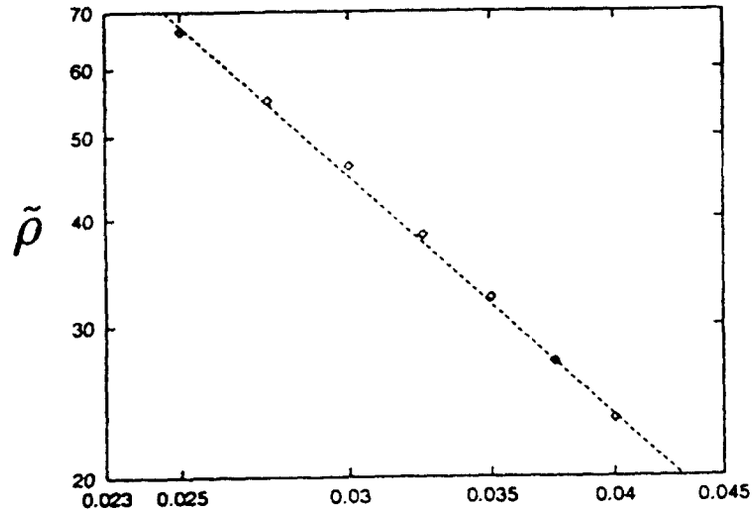
$$S = \Delta T/\Delta T_H = 0.8$$

$$m = (\text{capillary length/kinetic length}) = 0.1$$

Velocity and tip radius versus anisotropy of surface tension and kinetics, both of the form

$$1 + \delta \cos(4\theta)$$





c. Summary of power laws for large supercoolings

Case 1: Dendrites with anisotropic surface tension and isotropic kinetics: $0.03 \leq \delta_\gamma \leq 0.05$; $0.7 \leq S \leq 1.1$

$$v \approx 3.55\ (\delta_\gamma)^{0.78}\ m^{1.08}\ S^{2.58}$$

Case 2: Dendrites with isotropic surface tension and anisotropic kinetics: $0.05 \leq \delta_\mu \leq 0.075$; $0.8 \leq S \leq 1.1$

$$v \approx 0.96\ (\delta_\mu)^{0.64}\ m^{0.81}\ S^{3.46}$$

Case 3: Dendrites with anisotropic surface tension and anisotropic kinetics: $0.03 \leq \delta_\gamma \leq 0.04$; $0.01 \leq \delta_\mu \leq 0.04$; $0.7 \leq S \leq 1.1$

$$v \approx 2.04\ (\delta_\gamma)^{0.41}\ (\delta_\mu)^{0.19}\ m^{0.97}\ S^{2.71}$$

CONCLUSIONS

1. The phase field model can be used to model dendritic growth, but with today's computers and algorithms, one gets results independent of computational parameters only at very large supercoolings.

2. Growth kinetics has a significant influence on the dendrite operating state.

3. Dendrite tip shapes are more nearly hyperbolic than parabolic at large supercoolings.

4. Scaling laws are still evident, but exponents are different from those for small supercoolings.

4. σ^* is larger for increased kinetics and decreases somewhat with supercooling.

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