Phase field modelling of solidification

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Notes

http://mme.iitm.ac.in/gphani/public/pfnotes

Outline

Premise

Microstructure is important

- Constituent Phases
- Proportions & Configuration
- Morphology
- Lengthscales

Phase field models

Introduction

- Wide range of phenomena
- Moving boundary problems
- Solidification as an example
- Why → What → Wow!

Outline

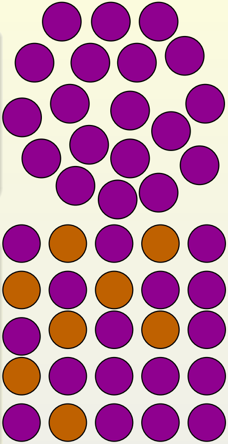
What we know, generally

Liquids

- Isotropic / No long range order
- Convection
- Relatively high solute diffusivity

Solids

- Crystalline in nature
- Chemical Order
- Stress
- Shapes / Morphology



Temperature variation in solid

Fourier Heat Conduction

$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T$$

- Easy to solve
- Analytical solutions exist for certain geometries
- H.S. Carslaw & J.C. Jaeger: Conduction of Heat in Solids

Analytical solution

Planar case

assuming,

- Steady state growth
- No anisotropy, no convection

$$\frac{T - T_\infty}{T_0 - T_\infty} = erf\left(\frac{x}{2\sqrt{\alpha t}}\right)$$

Temperature variation in liquid

Fourier Heat Conduction + Convection

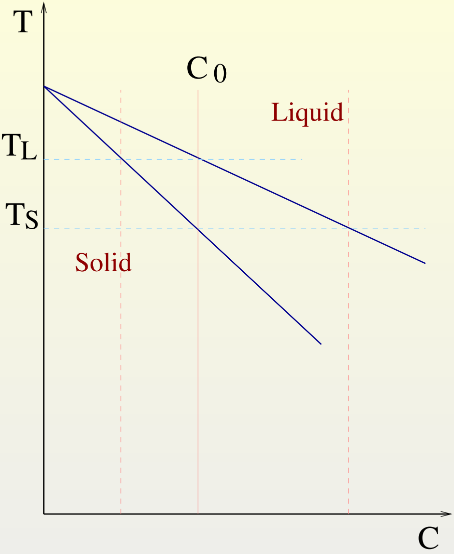
$$\frac{\partial T}{\partial t} + (\vec{u} \cdot \nabla)\, T = \alpha \nabla^2 T$$

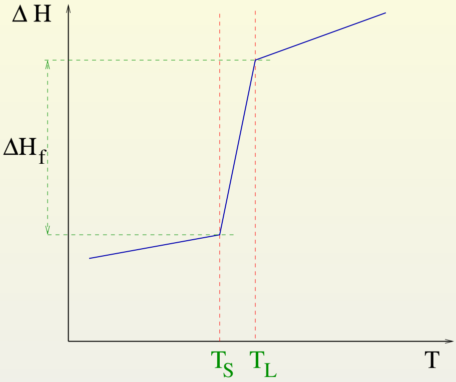
$$\frac{\partial \vec{u}}{\partial t} + (\vec{u} \cdot \nabla)\, \vec{u} = -\frac{\nabla p}{\rho} + \frac{\mu}{\rho} \nabla^2 \vec{u} + S$$

$$\nabla \cdot \vec{u} = 0$$

- Relatively difficult to solve
- Navier-Stokes equations for $\vec{u}$!
- Analytical solutions are very restricted

About liquid → solid transformation





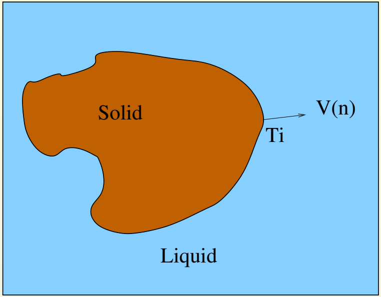
About liquid $\rightarrow$ solid transformation

- First order transformation
- Evolution of latent heat

$$(k_s \nabla T_s - k_l \nabla T_l) \cdot \hat{n} = \Delta H_f V(n)$$

- Solute partitioning

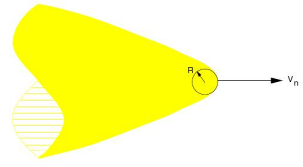
$$C_s^i = k C_l^i$$



Interface temperature

$$T_i = T_M - mC_L^* - \frac{\gamma}{\Delta S_m^v}\kappa - \frac{V}{\mu(\theta)}$$

Analytical solutions



Ivantsov's paraboloid of revolution

assuming,

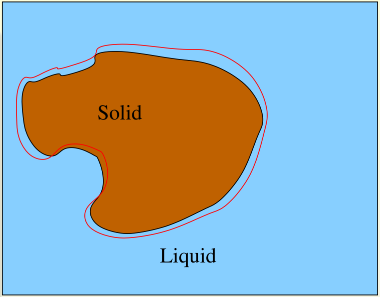
- Steady state growth
- Self-similar shape of solid
- No anisotropy, no convection

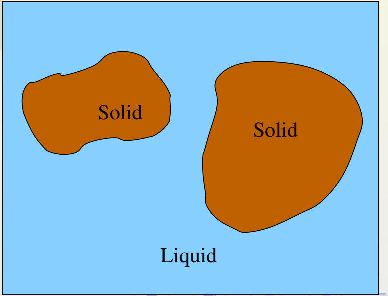
$$\frac{\Delta T}{\Delta H_f / C_p} = Pe^P E_1(P) + \Delta\theta_c$$

$$P = \frac{VR}{\alpha}$$

Trouble with Sharp Interface Method

- Solve for heat and solute flow within each domain
- Track domain boundary each time step
- Complex shapes make tracking difficult
- Bifurcation / merging of domains is horror





Single domain approach

Enthalpy Method

$$\Delta H = \rho C_p \left(T - T_{ref} \right) + \Delta H_f \phi$$

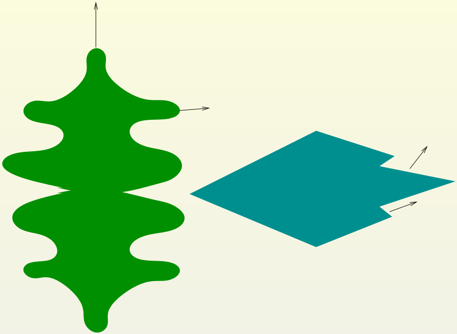
$$\frac{\partial T}{\partial t} = \alpha \nabla^2 T + \frac{\Delta H_f}{\rho C_p} \frac{\partial \phi}{\partial t}$$

- Single domain for solid and liquid
- ϕ indicates liquid fraction at any location
- Update enthalpy at each location
- Contour of ϕ gives location of interface
- Phase change assumed to take place between T_L and T_S

But we know that ...

$$T_i = T_M - mC_L^* - \frac{\gamma}{\Delta S_m^v} \kappa - \frac{V}{\mu \left(\theta \right)}$$

Interfaces: Heart of the Problem



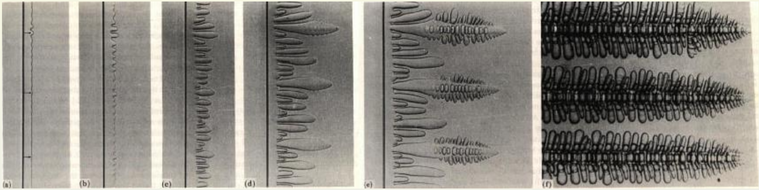
- Interface motion
- Crystallographic anisotropy of the interface
- Heat and solute flux at the interface
- Local thermodynamic equilibrium at the interface

Microstructure complexity $\Longrightarrow$ Tracking interfaces is not possible

Single phase solidification

Transparent Analogue Systems

- Planar $\rightarrow$ Cellular $\rightarrow$ Dendritic Morphologies
- Coarsening of inter-dendritic regions
- Complexity of patterns



Videos: cd_solid.mpeg kg_zu.mpg konkurre.mpg cd_dend.mpeg

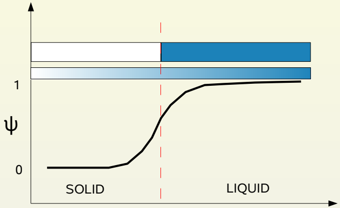
Problem Statement

Microstructure simulation

should take into account,

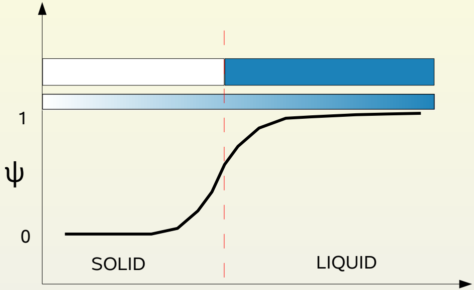
- Heat and solute transfer within domain
- appropriate boundary conditions
- possible undercooling
- realistic thermodynamic data
- effects due to crystallographic anisotropy
- convection
- nucleation of (multiple) solid phases

Sharp / Diffuse Interface



- Choose a functional F(ψ, ...)
- $\frac{\partial \psi}{\partial t} \propto -\frac{\delta F}{\delta \psi}$
- Contour at $\psi = \psi_0$ gives interface position
- Interface motion is an outcome of evolution of ψ field

Sharp / Diffuse Interface



- Interface motion
- Crystallographic anisotropy of the interface
- Heat and solute flux at the interface
- Local thermodynamic equilibrium at the interface

Outline

Diffuse Interface Approach

The first paper

The thermodynamic theory of capillarity under the hypothesis of a continuous variation of density.

- J.D. van der Waals, *Verhandl. Konink. Akad. Weten.* Amsterdam (Sect. 1) Vol. 1, No. 8 (1893)
- Translation by J.S. Rowlinson, *J. Statistical Phy.*, Vol.20, No.2, (1979)

Diffuse Interface Approach

- D.J. Korteweg, *Archives Nerlandaises de Sciences Exactes et Naturelles* pp. 1-24, (1901)
- V.L. Ginzburg and L.D. Landau, *Zh. Eksp. Teor. Fiz.* 20 (1950), p. 1064 (*Sov. Phys. JETP* 20 (1950) 1064)
- P.C. Halperin, B.I. Hohenberg, *Rev. Mod. Phys.*, Vo. 49, p.435-479 (1977)

Model C of Halperin & Hohenberg is appropriate for solidification

$$\alpha\xi^2\frac{\partial\phi}{\partial t} = \xi^2\nabla^2\phi + g(\phi) - f(T)$$

$$\frac{\partial f}{\partial t} - \lambda\frac{\partial\phi}{\partial t} = \nabla^2 f$$

Diffuse Interface Approach

Milestones

- J.W. Cahn and J.E. Hilliard, *J. Chemical Phys.*, 28:258-67 (1958)
- S.M. Allen and J.W. Cahn, *J. Phys.*, 38:C7-51 (1977)

Models

- *Cahn-Hilliard* nonlinear diffusion equation (conserved order parameter)

$$\frac{\partial C}{\partial t} = M_C \left(\nabla^2 f'(C) - \xi_C \nabla^4 C \right)$$

- *Allen-Cahn* time dependent Ginzburg-Landau equation (non-conserved order parameter)

$$\frac{\partial \eta}{\partial t} = M_\eta \left(\xi_\eta \nabla^2 \eta - f'(\eta) \right)$$

Conformance of variational approach with thermodynamics

Tasks

- Integration of kinetics into variational approach
- Guarentee thermodynamic consistency by the the variational method

Approaches

- Gradient Flow Method
  Decrease of energy at each time step
- Entropy Production
  in terms of transport variables

Thermodynamic Consistency

Correctness

Strict relaxation behavior of thermodynamic potentials during evolution is ensured.

Phase-field model is equivalent to sharp interface model

- G. Caginalp, *Phys. Rev. A.*, 39:5887 (1989)
- O. Penrose, P.C. Fife, *Physica D*, 43:44 (1990)
- S.-L. Wang et. al., *Physica D*, 69:189-200 (1993)
- E. Fried, M.E. Gurtin, *Physica D*, 91:143-181 (1996)

Outline

Phase-field model

Phase-field

- Field variables (eg., Phase-field) introduced to avoid tracking of interface
- The term Phase-field was first used for solidification of pure metal

Phase-field

What we mean

- Phase-field is a **field variable** / order parameter
- Physically (for solidification), bond angle order parameter comes close
- Density could also be thought of

What we don't mean

- A region in a phase diagram where a particular phase is stable

Applications of Phase-field models

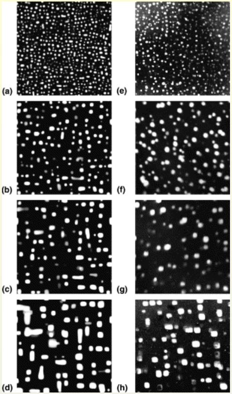
Areas

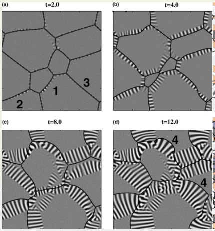
- Solidification
- Solid state phase transformations
- Coarsening and grain growth
- Other applications

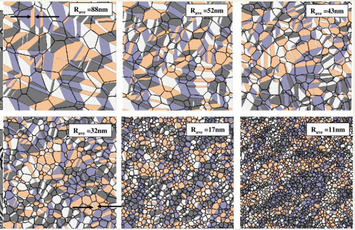
Applications of Phase-field approach

Solid state phase transformations

- Spinodal decomposition
- Precipitation of cubic ordered intermetallics from disordered matrix
- Cubic-tetragonal, Hexagonal-orthorhombic Martensitic transformations
- Ferroelectric transformations
- Phase transformation under applied stress



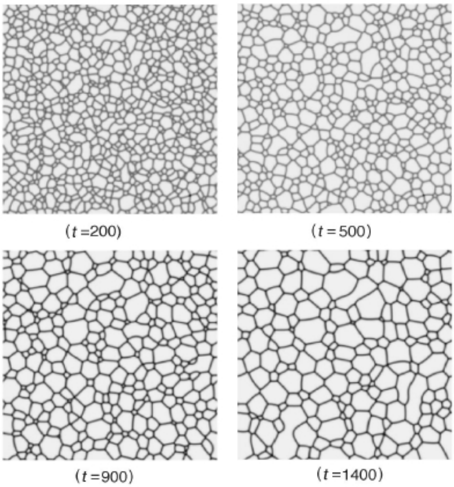




Ref:

10.1016/j.actamat.2003.10.028, 2006.09.048 & 2004.02.032

Applications of Phase-field approach



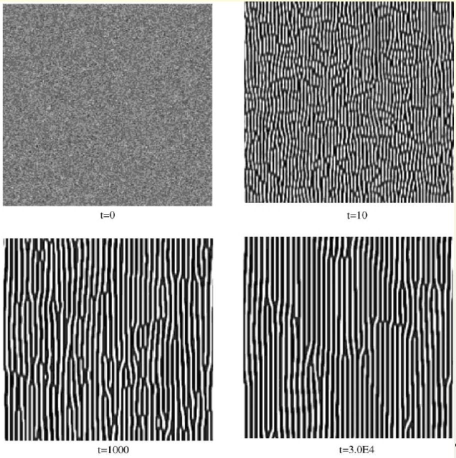
[a]

Coarsening and grain growth

- Coarsening
- Grain growth in single / two phase solid
- Anisotropic grain growth

[a] PRB, 63:184102 (2001)

Applications of Phase-field approach



[a]

Other areas

- Phase transformations in thin films
- Surface stress introduced pattern formation
- Spiral growth
- Crystal growth under stress
- Dislocation dynamics, Solute-dislocation interactions
- Crack propagation
- Electromigration

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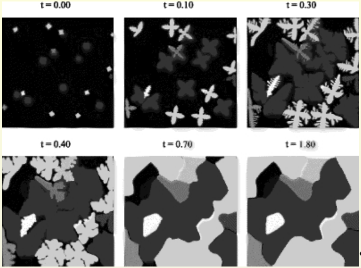
[a] JMPS 49:1937 (2001)

Outline

Applications of Phase-field approach

Solidification

- Pure Metal
- Binary alloys
- Multicomponent alloys
- Multiphase-field models for eutectic and peritectic
- Monotectic
- Non-isothermal



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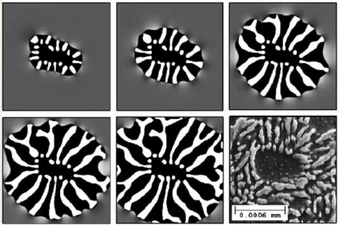


b

[a] 10.1016/S1359-6454(03)00388-4
[b] CPC 147:230-233 (2002)

Levels of complexity

Components

- Pure metal
- Binary
- Ternary, Multi-component

Phases

- Two phase
- Three phase
- Multi-phase, multi-stage

Anisotropy

- Isotropic
- Anisotropic (cubic, hexagonal, etc.,)
- Polycrystalline
- Faceted (cubic, hexagonal, etc.,)

Phenomena

- Convection
- Nucleation
- Stress

Phase-field model for solidification

First Phase-field model for solidification

J.S. Langer: Directions in Condensed Matter Physics, Models of Pattern Formation in First-order Phase Transition, 165 (1986)

First numerical solutions (2D)

- R. Kobayashi: *Bull. Jpn. Soc. Ind. Appl. Math.*, 1:22 (1991)
- R. Kobayashi: *Physica D.*, 63:410 (1993)
- A.A. Wheeler, B.T. Murray, R.J. Shaefer, *Physica D*, 66:243 (1993)

Kobayashi's model

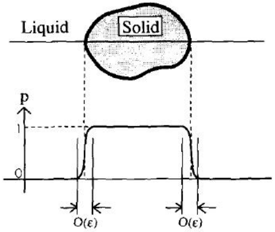


Fig. 1. Expression of the solid/liquid interface by the phase indicating function p.

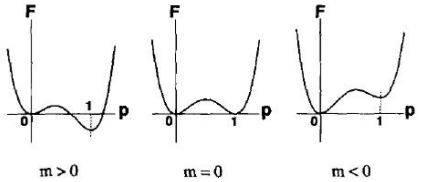


Fig. 2. Double-well potential F controlled by the parameter m. $\Delta F = F(0; m) - F(1; m) = m/6$.

Kobayashi's model

Equations

$$F = \frac{1}{4}p^4 - (\frac{1}{2} - \frac{m}{3})p^3 + (\frac{1}{4} - \frac{m}{2})p^2$$

$$\tau \frac{\partial p}{\partial t} = -\frac{\partial}{\partial x}\left(\epsilon\epsilon' \frac{\partial p}{\partial y}\right) + \frac{\partial}{\partial y}\left(\epsilon\epsilon' \frac{\partial p}{\partial x}\right) + \nabla \cdot (\epsilon^2 \nabla p) + p(1-p)(p + m - \frac{1}{2})$$

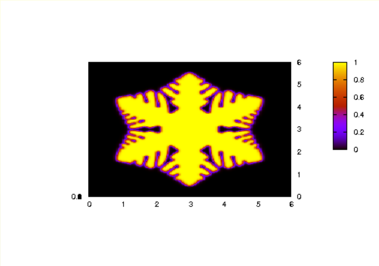
$$\epsilon = \bar{\epsilon}\sigma(\theta)$$

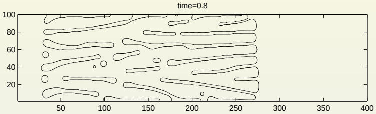
$$\sigma(\theta) = 1 + \delta \cos[j(\theta - \theta_0)]$$

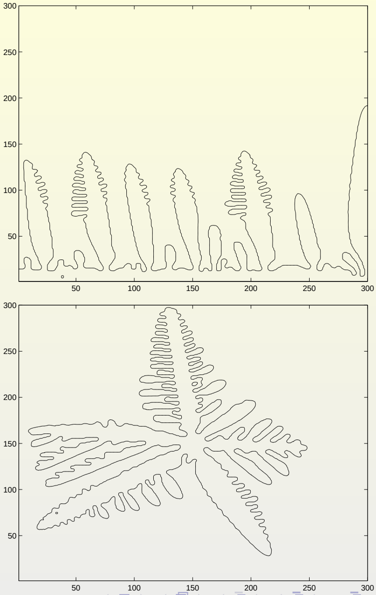
$$m(T) = (\frac{\alpha}{\pi}) \arctan[\gamma(T_e - T)]$$

$$\frac{\partial T}{\partial t} = \nabla^2 T + K \frac{\partial p}{\partial t}$$

Microstructures







Major schools of thought

- **Wheeler et al.**: Gradient energy term alone is anisotropic
- **Karma et al.**: Gradient energy term & Relaxation time are anisotropic
- **Steinbach et al.**: Coupling of Temperature to Phase-field

Parameters

Wheeler's Model

Equilibrium profile in 1D:

$$\phi(x) = \frac{1}{1 + \exp(\frac{-2x}{\delta})} = \frac{1}{2}\left[1 + \tanh\left(\frac{x}{\delta}\right)\right]$$

$$\tilde{W}_\phi = 6\sqrt{2} \cdot \frac{\tilde{\sigma_{sl}}}{\tilde{\omega_{sl}}}$$

$$\tilde{\epsilon_{sl}}^2 = 6\sqrt{2} \cdot \tilde{\sigma_{sl}} \tilde{\omega_{sl}}$$

- Fix surface tension and interface width
- Height of double well & Gradient energy term get fixed
- Choice of interface width & Grid resolution
- Time step is fixed based on numerical stability

Solution Methods

- Finite Difference
- Explicit Time Marching
- Implicit Iterative
- Finite Element

- Structured Grid
- Adaptive Grid
- Deal-II
- Multi-Domain, Multi-Grid

MICRESS software from ACCESS, Aachen, Germany
based on models by Steinbach et al.

Phase-field models

Models

Pure metal	J.S. Langer (1986)
Pure metal, thin interface	A. Karma & W-J. Rappel (1996)
Binary alloy	A.A. Wheeler et. al. (1992)
Multiphase	I. Steinbach et. al. (1996)
Multiphase	Garcke, B. Nestler & B. Stoth (1998)
Multicomponent	M. Ode et. al. (2000)
Eutectic	Wheeler, Boettinger, McFadden (1996)
Peritectic	Lo, Karma, Plapp (2001)
Eutectic + Peritectic	Nestler & Wheeler (2000)

Phase-field models for multiphase (Eutectic) solidification

Models

- A. Karma, *Phy.Rev.E*, 49:2245 (1994)
- K.R. Elder et. al., *PRL*, 72:677 (1994)
- A.A. Wheeler et. al., *Proc.R.Soc.Lond.A*, 452:495-525 (1996)
- B.Nester & A.A. Wheeler, *Physica D*, 138:114-133 (2000)
- S.G. Kim et. al., *J. Cryst. Growth*, 261:135-158 (2004)

Eutectic phase-field models

Summary

- Wheeler's second model
- Simple coupling of T with ϕ etc.,
- Flexibility of free energy functionals
- Applicability to real systems

Lamellar Microstructure

Wheeler's Model

Faceted Solidification

Motivation

- High interfacial anisotropy
- Jackson Parameter $\alpha = \frac{L_0}{k_B T_E} \frac{\eta_1}{v}$
- Sharp facets instead of smooth interfaces
- Not all orientations of the interface are allowed

Models

- J.J.Eggleston, G.B. McFadden, P.W. Voorhees, *Physica D*, 150:91-103 (2001)
- J-M. Debierre, A. Karma, F. Celestini, R. Guerin, *Phys.Rev.E*, 68:041604 (2003)

Eggleston et. al.'s model

Details

$$\theta = \arctan(\phi_y/\phi_x)$$

$$\mu = \mu_0 + \left(\gamma + \frac{\partial^2\gamma}{\partial\theta^2}\right)\kappa$$

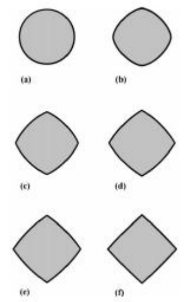
$$\epsilon = \epsilon_0\left(1 + \delta\cos 4\theta\right)$$

- Anisotropy of $\delta > \frac{1}{15}$ will not be in the usual scheme
- Concept of missing angle range

$$\bar{\epsilon} = \epsilon(\theta) \text{ for } \theta_m \leq |\theta| \leq \frac{\pi}{4}$$

$$\bar{\epsilon} = \frac{\epsilon(\theta_m)\cos(\theta)}{\cos(\theta_m)} \text{ for } |\theta| \leq |\theta_m|$$

Eggleston et. al.'s model



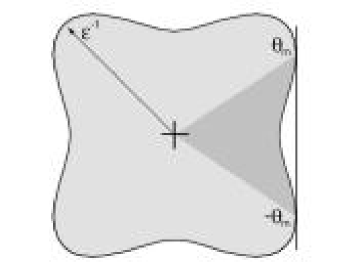


Fig. 2. Convexifying the polar plot of $1/\epsilon$ ($\epsilon_4 = 0.20$).

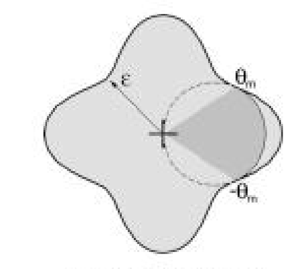


Fig. 3. Polar plot of ϵ ($\epsilon_4 = 0.20$).

Numerical Issues

- Slope of interface near the corners
- One sided differencing formula across the corner

Debierre et. al.'s model

Details

- Cusps in γ-plot are of the form

$$f(\theta) \approx 1 + \delta|\theta - \theta_c| + \ldots \text{ for } |\theta - \theta_c| << 1$$

- γ-plot is regularized using Dirac delta functions at θ_c
- E.g., $f(\theta) = 1 + \delta\left(|\sin\theta| + |\cos\theta|\right)$

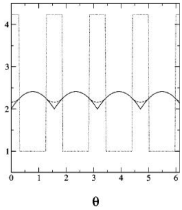


FIG. 1. Three functions of the interface orientation angle θ represented for a cusp amplitude $\delta = 1.0$. Solid line: anisotropy function with sharp cusps, $f(\theta)$. Dashed line: smoothed anisotropy function, $f_s(\theta)$. Dotted line: dimensionless stiffness, $f_s(\theta) + f_s''(\theta)$. A large smoothing angle $\theta_0 = \pi/10$ is used here to make the difference between f and f_s visible.

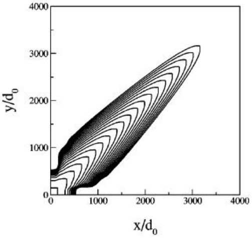


FIG. 5. Time evolution of a faceted dendrite for undercooling $\Delta = 0.55$, smoothing angle $\theta_0 = \pi/200$, and anisotropy $\delta = 1.0$. The time interval between two successive curves is $100\tau_0$.

Phasefield and faceting

Summary

- Both Eggleston et. al.'s and Debierre et. al.'s models are easily implementable
- Both are for pure metals
- Both are for 2D shapes
- Higher anisotropy requires smaller time steps

Effect of anisotropic kinetics

Faceting can also arise out of Anisotropy of kinetic coefficient !

T. Uehara & R.F. Sekerka, *J. Cryst. Growth*, 254:251-261 (2003)

$$\mu = \bar{\mu}(1 + \delta \cos 4\theta)$$

(i) n=2 (ii) n=8 (iii) n=12

Phase-field models with convection

To watch out

- Velocity - Temperature - Phase-field - (Species) - coupling
- Velocity dissipation at solid
- Stefan wind
- Consistency of transport terms with thermodynamics

Models

- R. Tönhardt & G. Amberg (1998, 2000)
- D.M. Anderson, G.B. McFadden & A.A. Wheeler (2000)
- C. Beckermann & co-workers (1999)

Tönhardt & Amberg

Model details

Single pure metal dendrite growing according to WBM model.

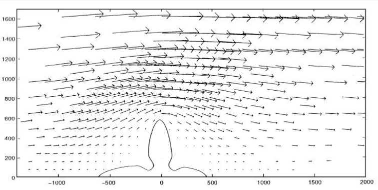
- *J. Cryst. Growth* 194:406-425 (1998): normal to shear flow
- *J. Cryst. Growth* 213:161-187 (2000): at angle to shear flow
- Velocity coupled with Temperature. Temperature with phase-field.

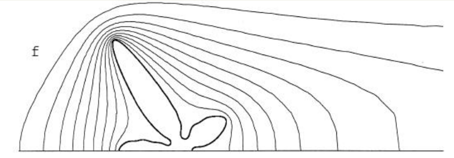
Velocity dissipation:

$$\nu = \nu_0 (1 + f_v)$$

$$f_v = 1 \text{ if } \phi < -0.6$$

$$f_v = 1 + 10(\phi + 0.6)^2 \text{ otherwise}$$





Tönhardt & Amberg

Fluid flow + Phase-field is computationally intensive !

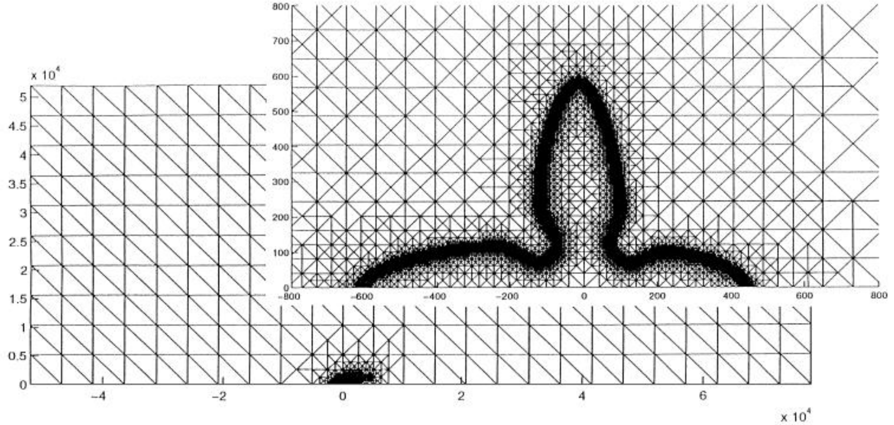


Fig. 2. The element distribution for $\varepsilon = 0.05$, $\Delta = 0.1$, $\mathrm{Pr} = 23.1$, $\mathrm{Pe} = 10^{-5}$ and $\beta_0 = 0$ at time $= 6 \times 10^5$. The corresponding solid/liquid interface to this case is shown by the dashed line in Fig. 5b. The domain size is $[-52\,000.0, 78\,000.0]W \times [0., 52\,000.0]W$ and consists of about 140 000 elements and 70 000 nodes, where the finest and the coarsest resolution are 0.6 and 5200, respectively.

Anderson et. al.

Model details

Single pure metal dendrite growing according to WBM model.

- *Physica D.* 135:175-194(2000): Parallel shear flow
- Irreversibel thermodynamics used

Velocity dissipation:

$$\mu = \mu_S \phi + \mu_L (1 - \phi)$$

Different ratios of $\frac{\mu_S}{\mu_L}$ tested.

Diepers, Beckermann & Steinbach

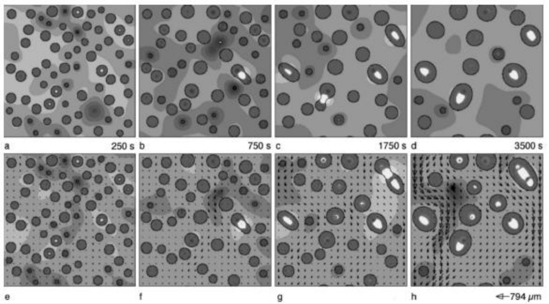
Model details

Single phase, binary alloy

- *Acta Materialia* 47:3663 (1999): Flow by constant pressure drop
- Applied to coarsening

Velocity dissipation by Darcy's Law:

$$u = \Pi \frac{\nabla p}{\nu}$$

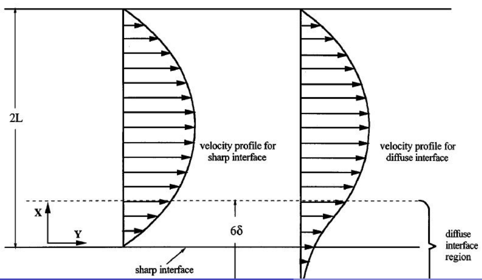


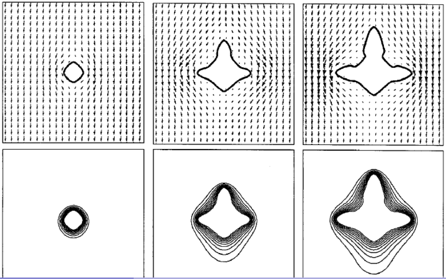
Beckermann et. al.

Model details

Pure metal, Karma-Rappel thin interface model

- *J. Comp. Phys.* 154:468-496 (1999)
- Mixture properties, Porous medium approach
- Average viscous stress is proportional to gradient of superficial liquid velocity $(1-\phi)\mathbf{v}_l$
- Mixture model for species equation, delinked from phase-field equation

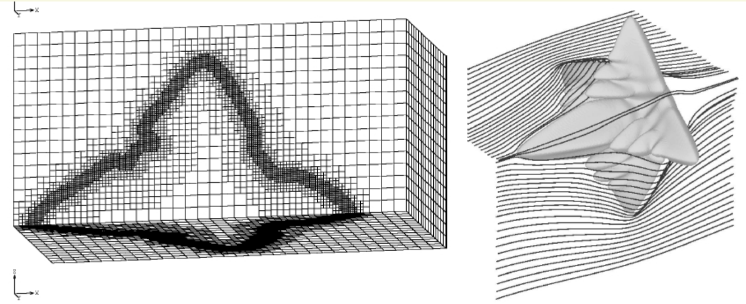




Phasefield + Convection in 3D

Model details

Beckermann's model applied to 3D by J.A. Dantzig et.al, *Phys Rev. E* 64:041602 (2001).
Computationally Intensive !



Phasefield + Convection

Summary

- Thermodynamically consistent binary phase-field model with convection ?
- Mixture models, porous medium approach, Artificial viscosity for solid etc., can give some results
- Computationally intensive

Outline

Phase-field + CALPHAD

- L.Q. Chen & co-workers: *Scripta Mat.*, 46:401-6 (2002); *Acta Mat.*, 52:833-840 (2004); *Acta Mat.*, 52:2837-2845 (2004)
- H. Harada & co-workers: *Comp. Mat. Sci.*, 39:871-879 (2007); *Intermetallics*, 16:239-245 (2008)

  Precipitation in Ni-base alloys

- G. Amberg, J.Agren & co-workers, *Acta Mat.*, 51:1327-1339 (2003); *Acta Mat.*, 52:4055-4063 (2004)

  $\gamma \rightarrow \alpha$ *in binary Fe-C*

- Jingjie Guo, Xinzhong Li, Yanqing Su, Shiping Wu, Bangsheng Li, Hengzhi Fu, *Intermetallics*, 13:275-279 (2005)

  Cellular growth in Ti-Al alloys

Phase-field + CALPHAD

- Qing Chen,
  Ning Ma, Kaisheng Wu, Yunshi Wang, *Scripta Mat.*, 50:471-476 (2004)
  Precipitate growth and dissolution in Ti-Al-V
- K. Wu, Y.A. Chang, Y. Wang, *Scripta Mat.*, 50:1145-1150 (2004)
  Ni-Al-Cr diffusion couples

R.S. Qin, E.R. Wallach, *Acta Mat.*, 51:6199-6210 (2003)
Al-Si alloy Thermodynamic database MTDATA
Mobility term discussed

Outline

Roadmap

1. General multi-phase-field model applicable to eutectic
2. Nucleation of each phase
3. Handle faceting of phases
4. Link thermodynamic data (CALPHAD) to the model
5. Effect of convection
6. Benchmark and predict experimentally observed microstructures

Thank you