Can the introduction of cross terms, from a generalised variational procedure in the phase-field modelling of alloy solidification, act as a natural anti-solute trapping current?

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Outline

We will...

- introduce a phase field model for solidification
- show the effect of changing interface widths on this model using three measures:(1) tip radius (2) tip velocity (3) solute change across interface
- introduce a bracket formulation as an alternative way of deriving the standard dynamical equations
- explore generalisations of the procedure to derive a modified model
- compare the effects of using these terms
- answer the question posed

Typical dendrite growth in 3 d

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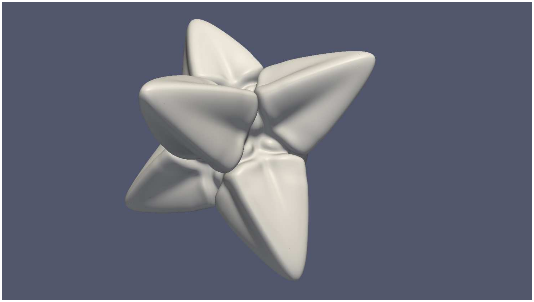


Alloy solidification model

- Phase equation for $\phi \in [0,1]$

$$\dot{\phi} = -M\frac{\delta F}{\delta \phi}, \tag{1}$$

- Conserved solute equation for $c \in [0,1]$

$$\dot{c} = \nabla \cdot D\nabla \frac{\delta F}{\delta c} \tag{2}$$

- Gibbs Free energy, $F = \int_\Omega f \, \mathrm{d}\Omega$, where the density

$$\begin{aligned} f = {} & W(\tfrac{1}{2}\delta^2 A(\nabla\phi)^2 \nabla\phi \cdot \nabla\phi + \omega(\phi)) \\ & + g(1-\phi)h_L(c,T) + g(\phi)h_S(c,T) \end{aligned} \tag{3}$$

Free energy density

- h_L and h_S are the bulk free energy densities of liquid and solid
- $\omega(\phi)$ is a double well potential
- $g(\phi) : g(0) = 0, g(1) = 1$ interpolates between h_L and h_S
- δ is a measure of the interface width
- The surface energy, W, and mobility, M, are proportional to $1/\delta$.
- Anisotropy is defined

$$A(\nabla\phi) = 1 - 3\varepsilon + 4\varepsilon(n_x^4 + n_y^4), \quad \varepsilon \sim 0.02, \quad \mathbf{n} = \frac{\nabla\phi}{|\nabla\phi|} \tag{4}$$

- Calculating the variational derivative gives the phase equation

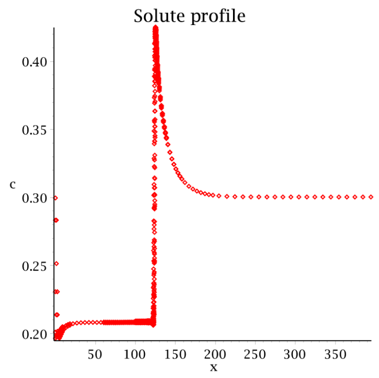
$$\frac{\delta^2}{\tilde{M}\tilde{W}}\dot{\phi} = \delta^2 K(\nabla\phi) - \frac{\partial f}{\partial \phi} - \frac{\delta}{\tilde{W}}\frac{\partial g}{\partial \phi}(h_S - h_L) \tag{5}$$

  where we define $\tilde{W} \equiv W\delta, \tilde{M} \equiv M\delta$ to see the dependence on the interface width, δ
- The contribution from anisotropy, $K(\nabla\phi)$, is given by

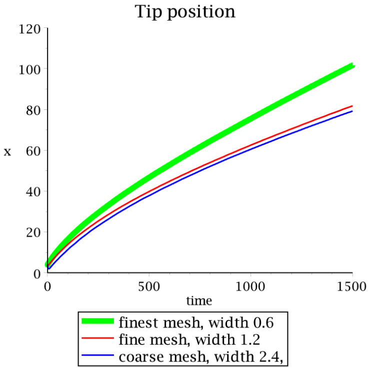
$$K(\nabla\phi) \equiv \nabla \cdot \left(A^2 \nabla\phi + A\nabla\phi \cdot \nabla\phi \left.\frac{\partial A(\mathbf{p})}{\partial \mathbf{p}}\right|_{\mathbf{p}=\nabla\phi} \right) \tag{6}$$

Effect of δ on results

- Three measures of simulation
  - tip velocity
  - tip radius
  - solute change across interface, $k_E \equiv c_{\min}/c_{\max}$



Effect of δ on tip velocity



- A marked lowering of velocity with increased interface width

Effect of δ on tip radius

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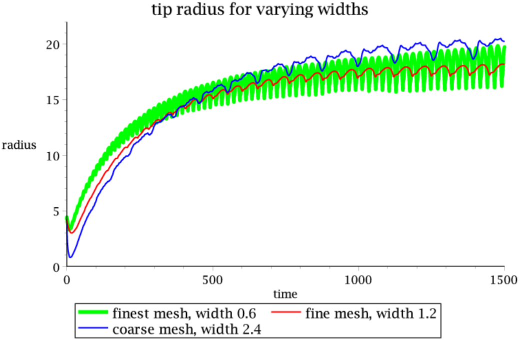

- Reasonable agreement of the tip radius across all widths

Effect of δ on solute ratio

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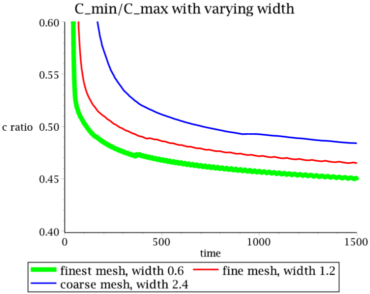

- divergence of the solute ratio $k_E = c_{\text{min}}/c_{\text{max}}$

Alloy solidification model

- One route to compensate for the solute trapping in the interfacial region is to introduce an opposing current, $\mathbf{j} \propto \dot{\phi}\mathbf{n}$ [Alain Karma, Phys. Rev. Lett. 2001]
- Phase equation for $\phi \in [0, 1]$ stays the same:

$$\dot{\phi} = -M \frac{\delta F}{\delta \phi},$$

- Conserved solute equation for $c \in [0, 1]$

$$\dot{c} = \nabla \cdot D \nabla \frac{\delta F}{\delta c} + \boxed{\nabla \cdot \mathbf{j}}$$

- We investigate an alternative approach

Alternative route

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Poisson bracket

- Hamiltonian energy:

$$H = \tfrac{1}{2}\frac{\mathbf{p}\cdot\mathbf{p}}{m} + V(\mathbf{x}), \tag{7}$$

- The Poisson bracket for arbitrary variable A:

$$\{A,B\} \equiv \sum_i^3 \left(\frac{\partial A}{\partial x_i}\frac{\partial H}{\partial p_i} - \frac{\partial A}{\partial p_i}\frac{\partial H}{\partial x_i} \right). \tag{8}$$

- Equations of motion:

$$\dot{x}_i = \{x_i, H\},$$
$$\dot{p}_i = \{p_i, H\}, \tag{9}$$

Dissipation bracket

- Gibbs free energy, $F = \int_\Omega f(\phi, \nabla\phi, c)\,\mathrm{d}^3x$
- Dissipative bracket for arbitrary functional A:

$$[A, E] \;=\; -\int_\Omega M \frac{\delta A}{\delta \phi}\frac{\delta F}{\delta \phi}\,\mathrm{d}^3x - \int_\Omega D\nabla\frac{\delta A}{\delta c}\cdot\nabla\frac{\delta F}{\delta c}\,\mathrm{d}^3x$$

- Equations of motion: $\dot{A} = [A, F]$ together with the chain rule $\dot{A} = \int_\Omega \left(\frac{\delta A}{\delta \phi}\dot{\phi} + \frac{\delta A}{\delta c}\dot{c}\right)\mathrm{d}^3x$,
- gives the dynamical equations:

$$\dot{\phi} = -M\frac{\delta F}{\delta \phi}, \quad \dot{c} = \nabla\cdot D\nabla\frac{\delta F}{\delta c} \tag{10}$$

Dissipation bracket

... with thermal/entropy field

- Gibbs free energy, $F = E - \int_\Omega sT\,\mathrm{d}^3x = \int_\Omega f(\phi, \nabla\phi, c, T)\,\mathrm{d}^3x$
- Dissipative bracket for arbitrary functional A:

$$
\begin{aligned}
[A, E] \;=\; & -\int_\Omega M \frac{\delta A}{\delta \phi}\frac{\delta E}{\delta \phi}\,\mathrm{d}^3x + \int_\Omega \frac{M}{T}\frac{\delta A}{\delta s}\frac{\delta E}{\delta \phi}\frac{\delta E}{\delta \phi}\,\mathrm{d}^3x \\
& -\int_\Omega D\nabla\frac{\delta A}{\delta c}\cdot\nabla\frac{\delta E}{\delta c}\,\mathrm{d}^3x + \int_\Omega \frac{D}{T}\frac{\delta A}{\delta s}\nabla\frac{\delta E}{\delta c}\cdot\nabla\frac{\delta E}{\delta c}\,\mathrm{d}^3x, \\
& -\int_\Omega \frac{\kappa}{T}\nabla\frac{\delta A}{\delta s}\cdot\nabla\frac{\delta E}{\delta s}\,\mathrm{d}^3x + \int_\Omega \frac{\kappa}{T^2}\frac{\delta A}{\delta s}\nabla\frac{\delta E}{\delta s}\cdot\nabla\frac{\delta E}{\delta s}\,\mathrm{d}^3x,
\end{aligned}
$$

- Equations of motion: $\dot{A} = [A, E]$

Notes: diffusion parameters, symmetric terms, no cross terms, T in “non-linear" entropy terms.

Variational relations

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- Variational chain rule

$$\dot{A} = \int_\Omega \left(\frac{\delta A}{\delta \phi}\dot{\phi} + \frac{\delta A}{\delta c}\dot{c} + \frac{\delta A}{\delta s}\dot{s} \right) \mathrm{d}^3 x, \tag{11}$$

- Generalised thermodynamics

$$\begin{aligned} \left.\frac{\delta E}{\delta \phi}\right|_{c,s} = \left.\frac{\delta F}{\delta \phi}\right|_{c,T} \quad &, \quad \left.\frac{\delta E}{\delta c}\right|_{\phi,s} = \left.\frac{\delta F}{\delta c}\right|_{\phi,T}, \\ T = \left.\frac{\delta E}{\delta s}\right|_{c,\phi} \quad &, \quad s = -\left.\frac{\delta F}{\delta T}\right|_{c,\phi}. \end{aligned} \tag{12}$$

- Derived definitions

$$C_p = T\frac{\partial s}{\partial T} \quad , \quad L = -\left.\frac{\delta E}{\delta \phi}\right|_{c,T}, K = -\left.\frac{\delta E}{\delta c}\right|_{\phi,T} \tag{13}$$

Dissipation bracket

Alloy dynamics

- This gives

$$
\begin{aligned}
\dot{\phi} &= -M\frac{\delta F}{\delta \phi}, \\
\dot{c} &= \nabla \cdot D\nabla \frac{\delta F}{\delta c}, \\
C_p\dot{T} + \nabla \cdot \mathbf{q} &= L\dot{\phi} + K\dot{c}, \\
\mathbf{q} &\equiv -\kappa\nabla T - D\frac{\delta F}{\delta c}\nabla\frac{\delta F}{\delta c} + T\frac{\partial s}{\partial \nabla \phi}\dot{\phi}.
\end{aligned}
\tag{14}
$$

- also an entropy equation.

$$\dot{s} + \nabla \cdot \mathbf{J}_s = \sigma > 0 \tag{15}$$

Dissipation bracket with cross terms

- a more general bracket (for isothermal)

$$
\begin{aligned}
[A,E] \quad = \quad & -\int_\Omega M_{\phi,\phi}\frac{\delta A}{\delta\phi}\frac{\delta F}{\delta\phi}\,\mathrm{d}^3x - \int_\Omega D_{c,c}\nabla\frac{\delta A}{\delta c}\cdot\nabla\frac{\delta F}{\delta c}\,\mathrm{d}^3x \\
& -\int_\Omega D_{c,\phi}\nabla\frac{\delta A}{\delta c}\cdot\nabla\frac{\delta F}{\delta\phi}\,\mathrm{d}^3x - \int_\Omega D_{\phi,c}\nabla\frac{\delta A}{\delta\phi}\cdot\nabla\frac{\delta F}{\delta c}\,\mathrm{d}^3x \\
& -\int_\Omega M_{\phi,c}\frac{\delta A}{\delta\phi}\frac{\delta F}{\delta c}\,\mathrm{d}^3x - \int_\Omega M_{c,\phi}\frac{\delta A}{\delta c}\frac{\delta F}{\delta\phi}\,\mathrm{d}^3x
\end{aligned}
\tag{16}
$$

- Gives dynamical equations

$$
\begin{aligned}
\dot\phi &= -M_{\phi,\phi}\frac{\delta F}{\delta\phi} - M_{\phi,c}\frac{\delta F}{\delta c} + \nabla\cdot D_{\phi,c}\nabla\frac{\delta F}{\delta c} + \nabla\cdot D_{\phi,\phi}\nabla\frac{\delta F}{\delta\phi} \\
\dot c &= -M_{c,c}\frac{\delta F}{\delta c} - M_{c,\phi}\frac{\delta F}{\delta\phi} + \nabla\cdot D_{c,\phi}\nabla\frac{\delta F}{\delta\phi} + \nabla\cdot D_{c,c}\nabla\frac{\delta F}{\delta c}
\end{aligned}
\tag{17}
$$

New terms to address k_E problem

- Considering solute conservation implies $M_{c,c} = M_{c,\phi} = M_{\phi,c} = 0$ and the symmetry of the bracket implies $D_{\phi,c} = D_{c,\phi}$
- choose: $D_{c,\phi} = \beta D, M_{\phi,\phi} = (1+\beta)M$ and $D_{c,c} = D$, to give

$$\dot{\phi} = -M(1+\beta)\frac{\delta F}{\delta \phi} + \nabla \cdot \beta D \nabla \frac{\delta F}{\delta c}, \quad (18)$$

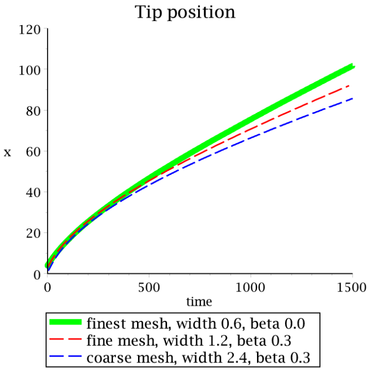
$$\dot{c} = \nabla \cdot D \nabla \frac{\delta F}{\delta c} + \nabla \cdot \beta D \nabla \frac{\delta F}{\delta \phi}, \quad (19)$$

  where $\beta > 0$ and $\beta \propto \delta$
- there is an entropy contribution of

$$\dot{s} = \cdots + \frac{M(1+\beta)}{T}\frac{\delta F}{\delta \phi}\frac{\delta F}{\delta \phi} + \frac{D}{T}\nabla \frac{\delta F}{\delta c} \cdot \nabla \frac{\delta F}{\delta c} + 2\beta \frac{D}{T}\nabla \frac{\delta F}{\delta c} \cdot \nabla \frac{\delta F}{\delta \phi} \quad (20)$$

- This gives the following results

Effect of δ on tip velocity



- Still some lowering of velocity with increased interface width, but better than without the new terms

Effect of δ on tip radius

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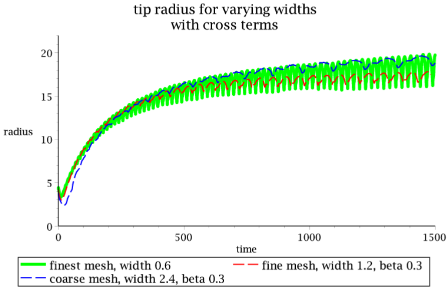
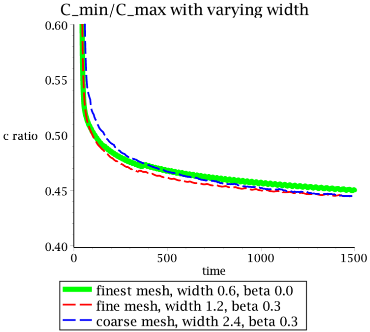
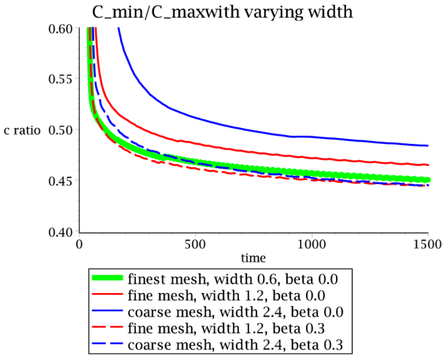


- Good agreement of the tip radius across all widths – marginally better than without the terms



- good agreement for the solute ratio $k_E = c_{\text{min}}/c_{\text{max}}$.

Effect of δ on solute ratio



- a marked improvement over the standard model

Final slide

So... yes we can

- ... by the introduction of cross terms, from a generalised variational procedure in the phase-field modelling of alloy solidification, find a natural anti-solute trapping current