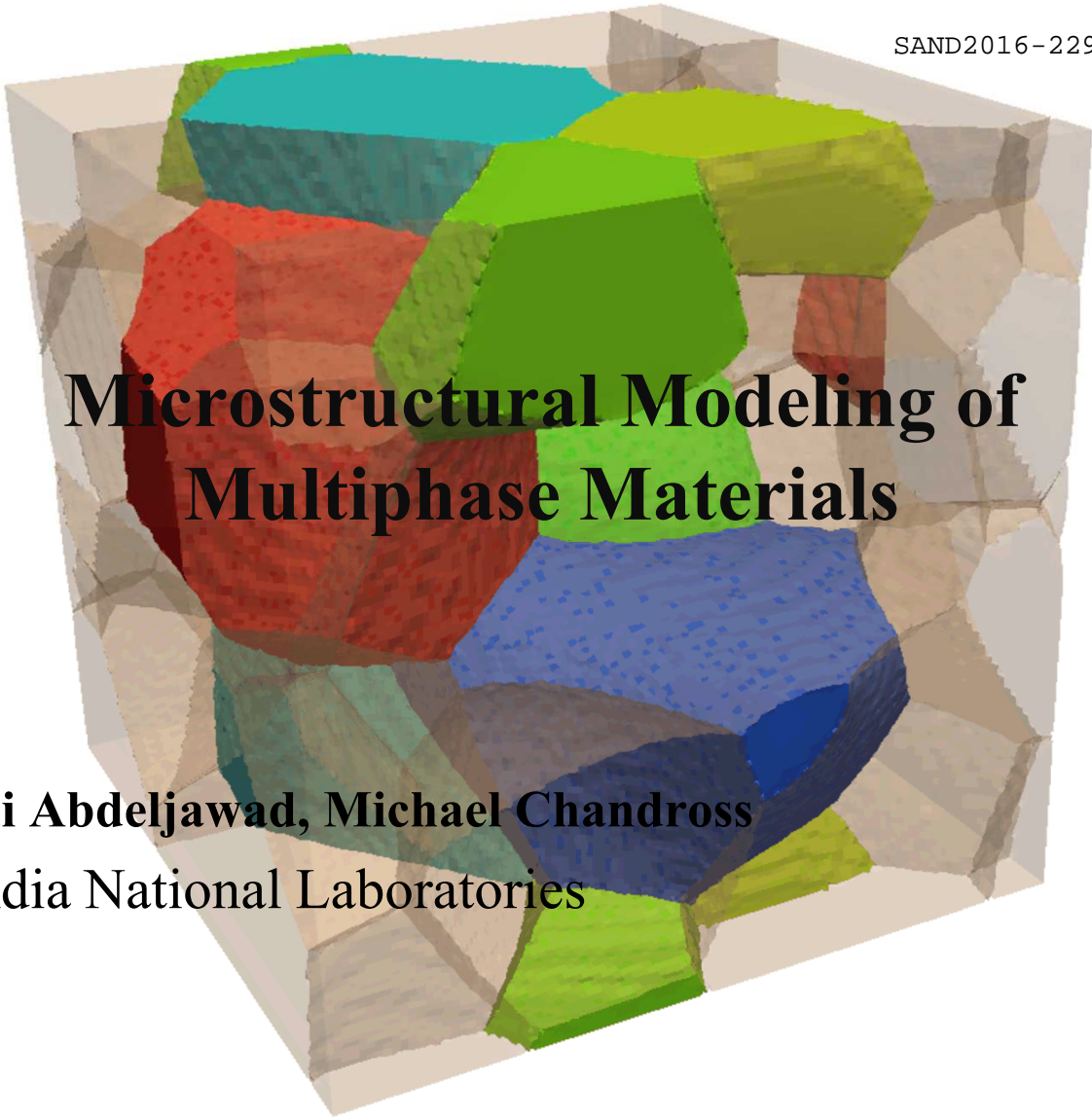
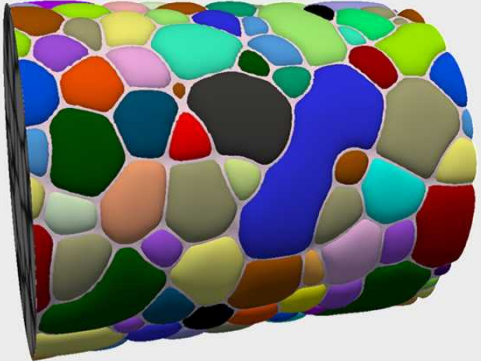


$$v_n = M\gamma H \quad \gamma = \Delta F - \Gamma \left. \frac{\partial f_{mix}}{\partial c} \right|_{eq}$$

$$\frac{\partial c}{\partial t} = \nabla \cdot \left[M_c \nabla \left(\frac{\delta \mathcal{F}_{tot}}{\delta c} \right) \right]$$



Microstructural Modeling of Multiphase Materials

Fadi Abdeljawad, Michael Chandross
 Sandia National Laboratories



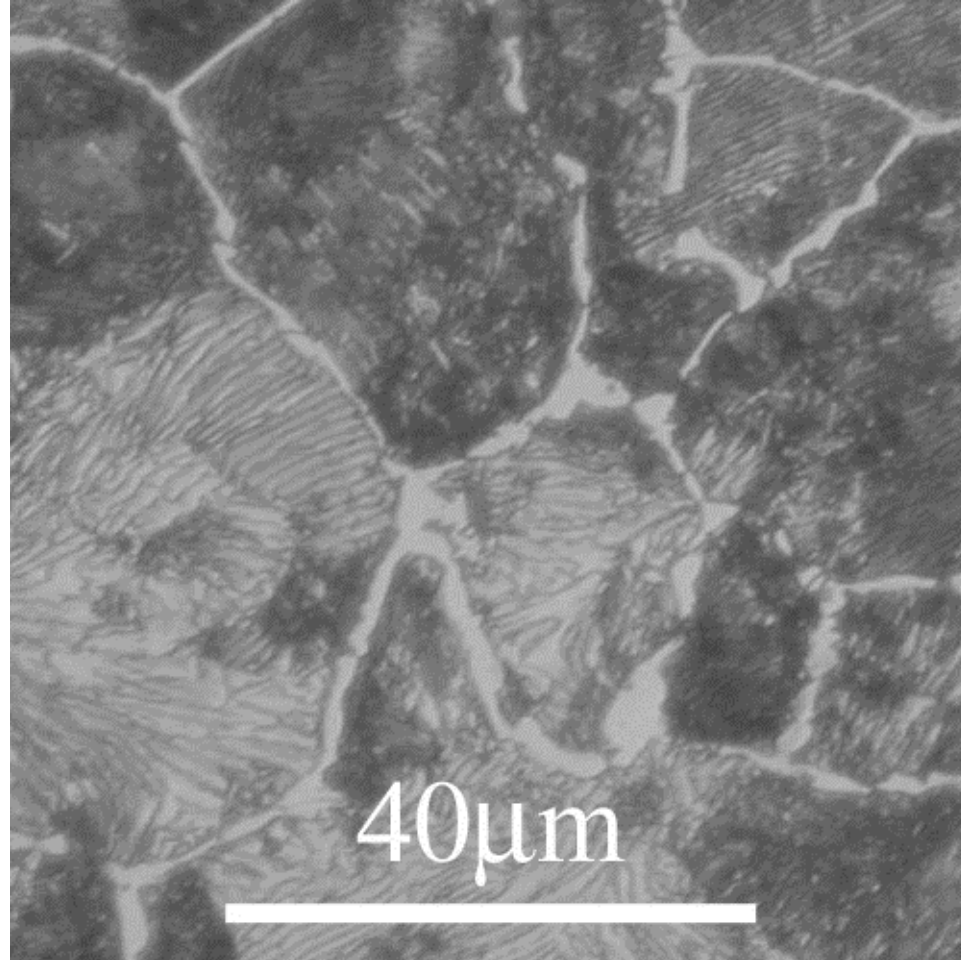
*Exceptional
 service
 in the
 national
 interest*



Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND NO. 2011-XXXXP

A Zoo of Features

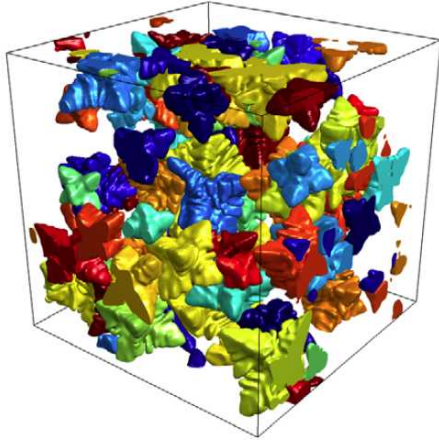
Fe, C 1.0 (wt%)
Hypereutectoid alloy
Widmanstätten structure



Univ. Cambridge: <http://www.doitpoms.ac.uk>

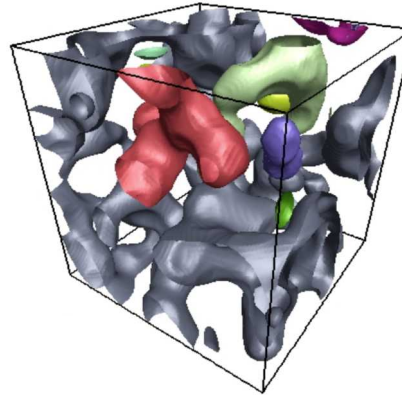
Crystals are like people, it is the defects in them which tend to make them interesting

Dendritic growth



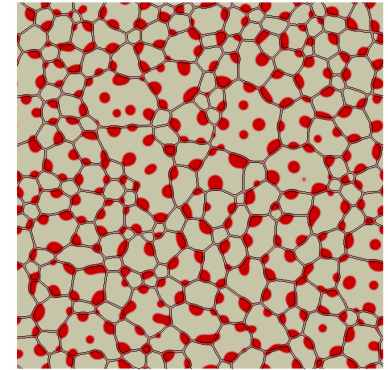
J. Phys: Cond. Matter. **20** (2008)

Coarsening



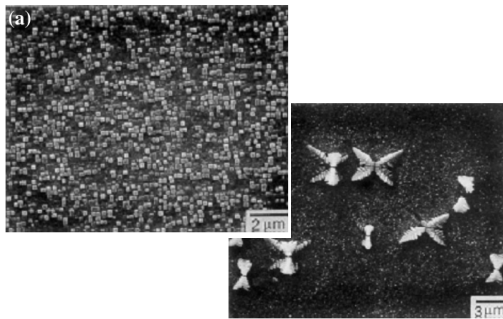
J. Power Sources. **250** (2014)

Interface segregation



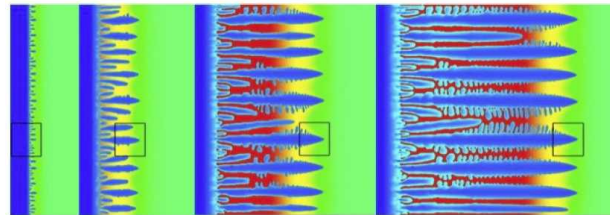
In preparation

Phase transitions



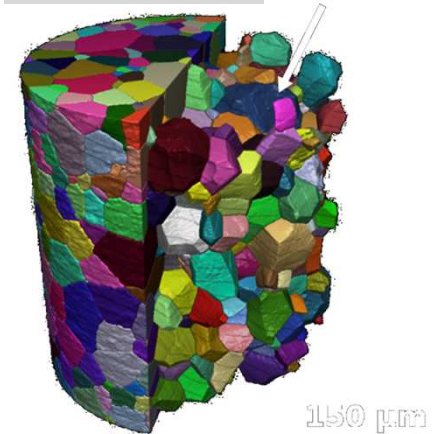
MSMSE. **13** (2005)

Solidification



N. Provatas, (2006)

Grain growth



150 μm
Acta Mater. **78** (2014)

Key Questions

- Given an additive manufacturing (AM) process, can we:

I An AM technique

- Power bed, sintering, ...
- **Temperature, particle size and distribution, compositions, ...**
- Geometry, constraints, ...

Translate

II

Materials processes

- Phase transitions (**solidification**, structural, ...)
- Phase diagrams (multi-phase/multi-component)
- **Defects** (interfaces, dislocations, voids, ...)
- Mass **transport** mechanisms
- **Elasticity**

Relate

Predict

III

Microstructural Modeling

- Framework(s) for relevant physics
- Track **microstructure evolution** subject to external stimuli (temperature, stress, ...)
- Relevant **metallurgical features** (grain size, second phase, interfaces, ...)
- Evaluate **properties** (mechanical, transport, ...)

■ Solidification

- Non-isothermal to reflect AM process
- Multi-grain (grains with different crystallographic orientations)
- Alloying
- Elasticity (thermal “eigen” strains)

■ Sintering/Wetting phenomena

- Multi-phase and/or multi-component
- Diffusive pathways
- Interface energies and wetting

Machinery: Phase Field

■ Order parameters

- c : Concentration (alloying elements)
- ϕ_i : Crystallographic orientation of grains (internal interfaces)
- η : Mass density (solid, vapor, liquid)
- etc ...

■ Coarse grained free energy

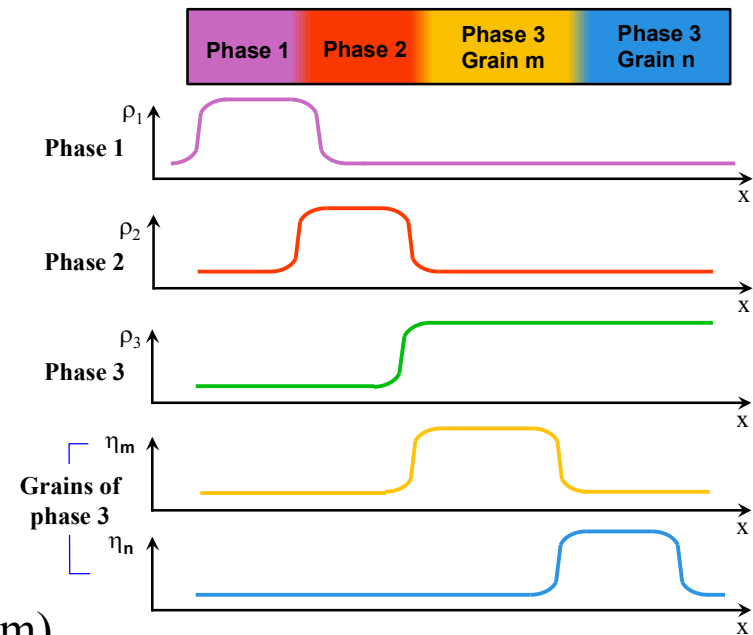
○ Bulk thermodynamics

- Phase transitions (Nucleation, solidification, etc...)
- Multicomponent

○ Interfacial energies and thermodynamics

- Gibbs-Thomson boundary condition (Stefan problem)
- Anisotropy

○ Dynamics driven by minimization of energy



Phase Field Formalism

- Total free energy

$$\mathcal{F}_{tot} = \int d\mathbf{r} \left[\underbrace{f_{bulk}(c, \phi_i, \eta, T)}_{\text{Bulk thermodynamics: chemical, elastic, magnetic, etc...}} + \underbrace{f_{int}(|\nabla c|, |\nabla \phi_i|, |\nabla \eta|)}_{\text{Interfacial energy: free surfaces, GBs, phase boundaries, etc...}} \right]$$

Bulk thermodynamics: chemical, elastic, magnetic, etc...

Interfacial energy: free surfaces, GBs, phase boundaries, etc...

- Dynamics

Non-conserved quantities (grain orientation, solidification, phase transitions)

- **Model A:** $\frac{\partial \phi_i}{\partial t} = -L_i \left(\frac{\delta \mathcal{F}_{tot}}{\delta \phi_i} \right) + \xi(\mathbf{r}, t)$ Allen-Cahn Eq. (Gradient flow of \mathcal{F}_{tot})

Conserved quantities (concentration, amount of phase(s))

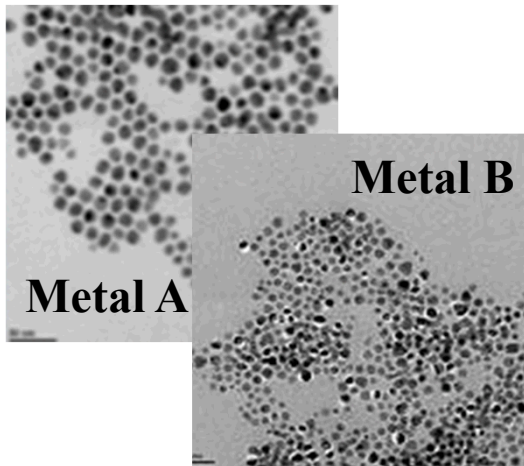
- **Model B:** $\frac{\partial c}{\partial t} = \nabla \cdot \left[M \nabla \left(\frac{\delta \mathcal{F}_{tot}}{\delta \phi_i} \right) \right] + \lambda(\mathbf{r}, t)$ Cahn-Hilliard Eq. (conservation of mass)

ξ and λ are Gaussian (thermal) noise [in accordance with Fluctuation dissipation theorem]

Example 1: Sintering/Wetting

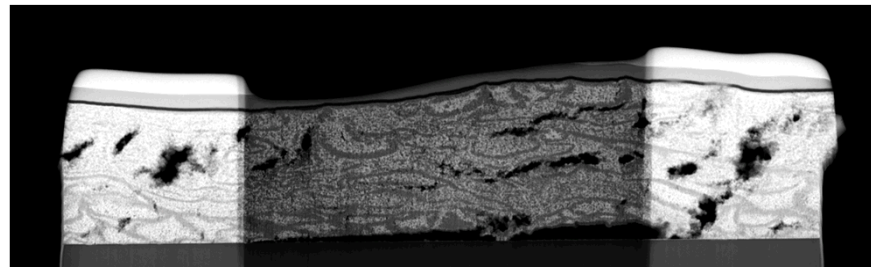
- Evolution of multi-phase materials systems
 - Powder with two material types (metals A and B)
 - **Surface energies:** γ_A, γ_B and **diffusivities:** Bulk and surface
 - Thermodynamics: **Phase diagram** of A-B alloy
 - Relevant features: domain/**pore size** and distribution, tortuosity (contiguity)

Initial



Adam Cook, SNL (2015)

Coarsened



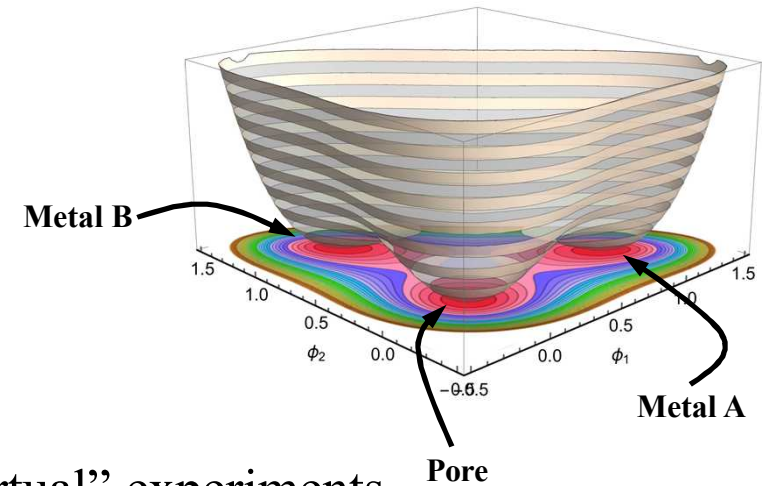
Adam Cook, SNL (2015)

Observations

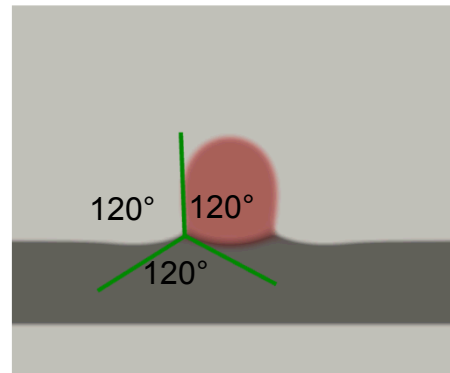
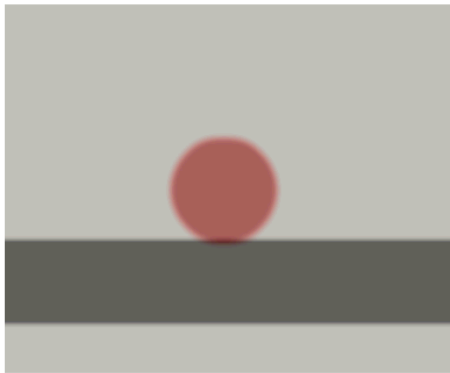
- Phase separation (no mixing)
- Swirls of A in B matrix
- Solid volume fraction: (A, B) = (40%, 60%)

Example 1: Sintering/Wetting

- 3-phase system (metals A and B and pore)
- 2 OPs needed: $(\phi_A, \phi_B) = (\text{metal A}, \text{metal B})$
- Pore space = $1 - \phi_A - \phi_B$
- Evolution via: $\frac{\partial \phi_i}{\partial t} = \nabla \cdot M_i \nabla \left(\frac{\delta \mathcal{F}_{tot}}{\delta \phi_i} \right), \quad i = A, B$



- Surface energies: Calibrate via sessile drop “virtual” experiments



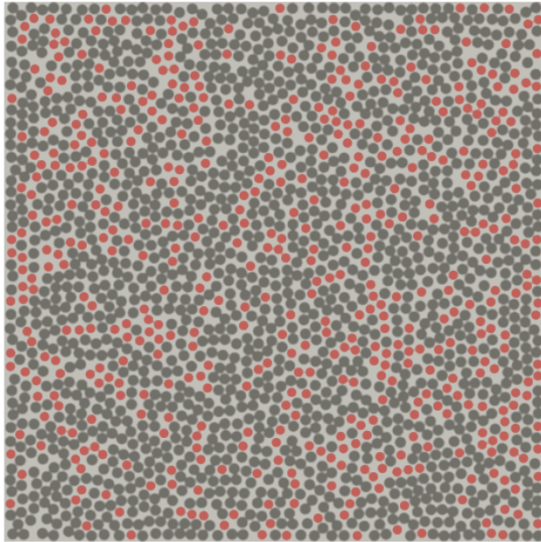
Results: 2D systems

- Vary volume fraction of two phases (metal A and B)

Volume fraction

A: 25%

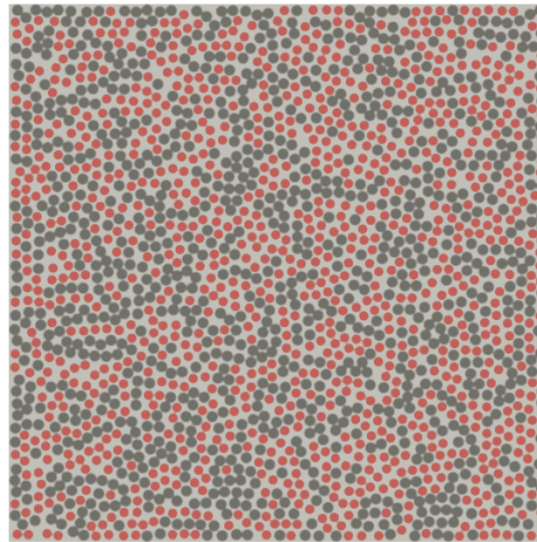
B: 75%



Volume fraction

A: 50%

B: 50%



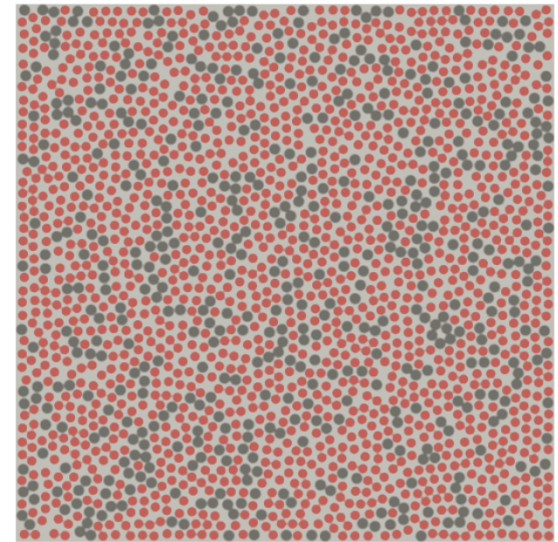
Volume fraction

A: 75%

B: 25%

■ Mat. A

■ Mat. B



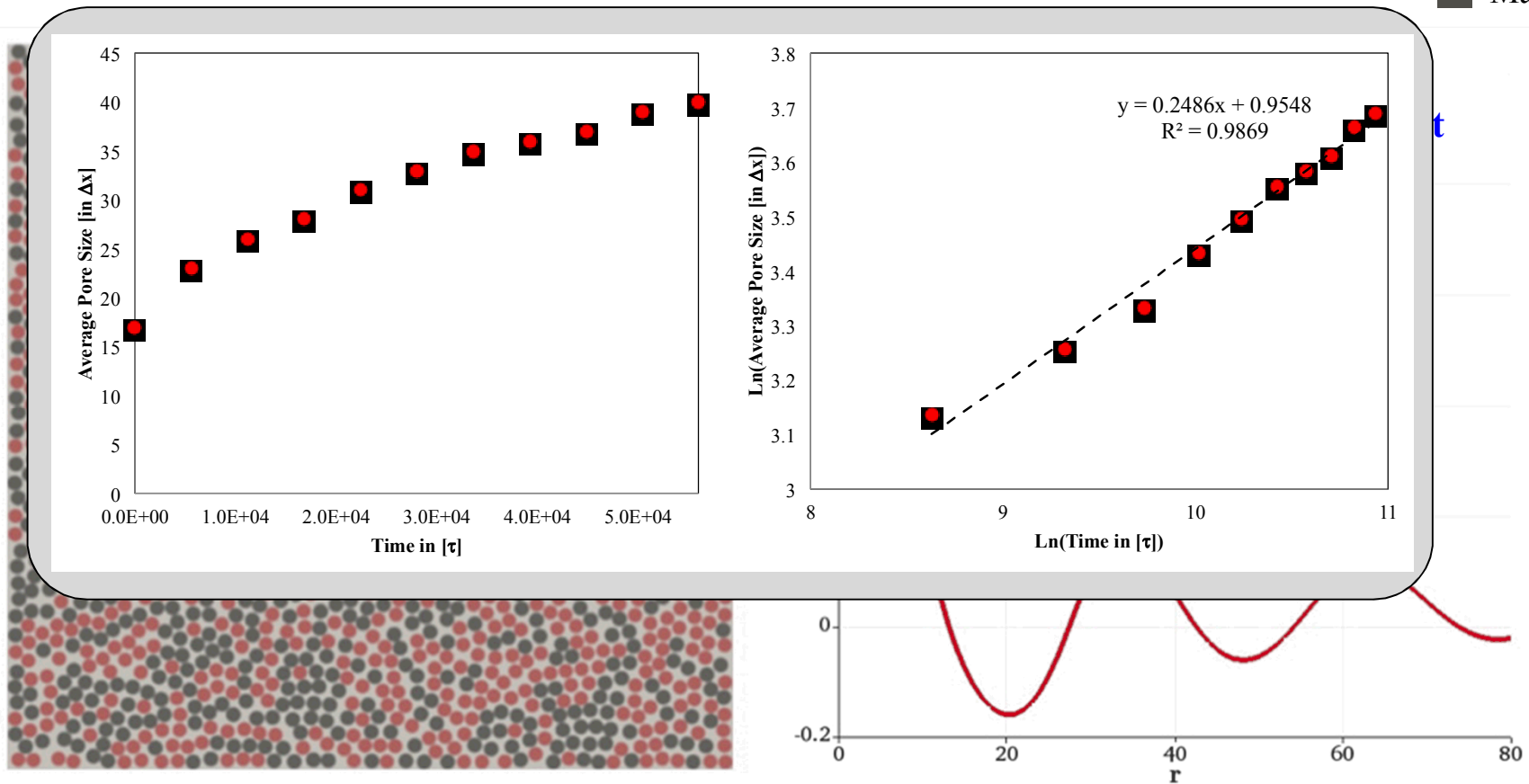
- Spatio-temporal evolution of pore phase and its statistics

- Two-point correlation function: $g_2(\mathbf{r}, t) = \left\langle \left[(\Gamma(\mathbf{r}, t) - \langle \Gamma(\mathbf{r}, t) \rangle) \right] \left[\Gamma(\mathbf{r} + \Delta\mathbf{r}, t) - \langle \Gamma(\mathbf{r}, t) \rangle \right] \right\rangle / \mu^2$
- $\Gamma = 1$ in pore and 0 elsewhere

Results: 2D systems

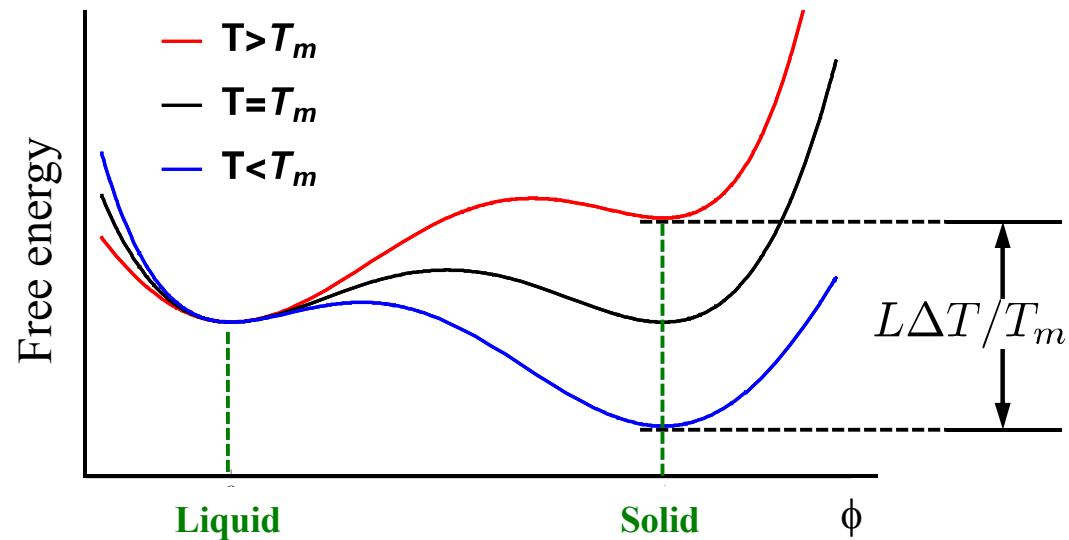
- Solid volume fraction: (A,B) = (50%, 50%)
- All interfacial properties are isotropic and equal
- Surface diffusion is the active mode and equal in A and B

■ Mat. A
■ Mat. B



Example 2: Solidification

- First order phase transition
- Exothermic reaction
- Driven by undercooling and latent heat of fusion

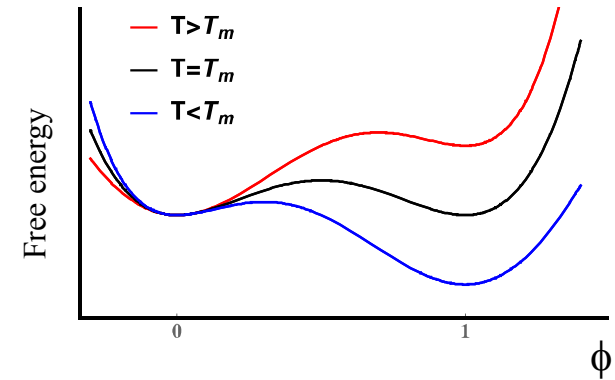


$$\mathcal{F}_{tot} = f_{bulk} + f_{trans} + f_{int}$$

- f_{bulk} : Free energy of bulk states/alloy (solid, liquid, ...)
- f_{trans} : Energy associated with solidification (latent heat, undercooling)
- f_{int} : Solid-liquid interface energy and anisotropy

Example 2: Solidification

- Introduce OP, ϕ , for the solid ($\phi=1$) and liquid ($\phi=0$)
- Free energies in terms of ϕ
- Non-dimensional temperature: $u = \frac{\Delta T}{(L/C_p)}$



$$\mathcal{F}_{tot} = \int dr \left[\underbrace{H f(\phi)}_{\text{Bulk}} + \underbrace{\frac{L\Delta T}{T_m} P(\phi)}_{\text{Phase transition}} + \underbrace{\frac{1}{2} |\epsilon(\phi) \nabla \phi|^2}_{\text{Anisotropic Surface energy}} \right]$$

Bulk


**Phase
transition**

**Anisotropic
Surface
energy**

■ Dynamics

$$\frac{1}{M(\phi)} \frac{\partial \phi}{\partial t} = - \frac{\delta \mathcal{F}_{tot}}{\delta \phi}$$

$$\frac{\partial u}{\partial t} = \nabla \cdot \alpha(\phi) \nabla u + \frac{dP(\phi)}{d\phi} \frac{\partial \phi}{\partial t}$$


 Source term due to
latent heat generation

L: Latent heat

T_m : Melting temperature

$\Delta T = T - T_m$: Undercooling

M: Anisotropic interface mobility

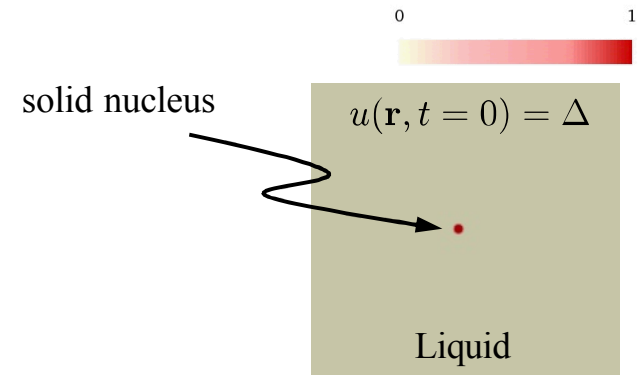
$\alpha = k/C_p \rho$ thermal diffusivity

C_p : Specific heat at constant pressure

ρ : Mass density

Growth Rates vs. Undercooling

- Fix material type \rightarrow Fix $(L, T_m, \alpha, C_p, \rho)$
- Vary initial temperature field $u(\mathbf{r}, t = 0) \rightarrow$ Driven by AM process
- Examine:
 - Solid growth rates and tip speeds
 - anisotropy type and strength
 - Evolution of temperature field

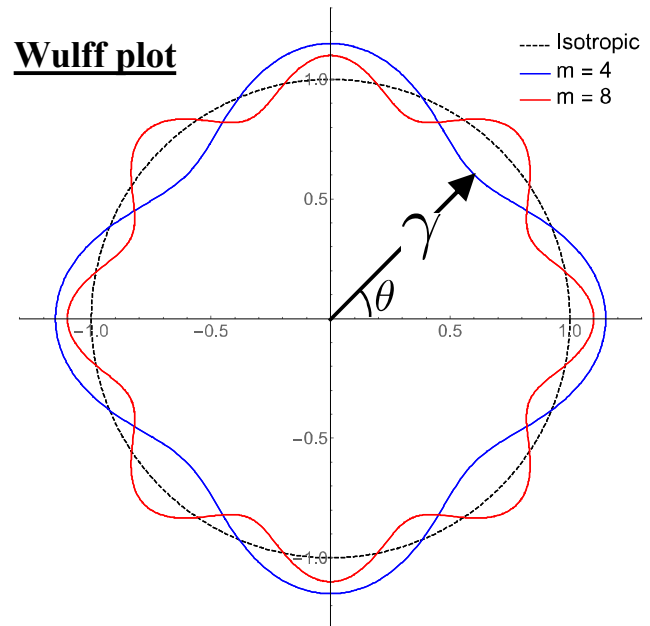


Example:

1. A solid nucleus in super-cooled melt
2. Initially: $u(\mathbf{r}, t = 0) = \Delta$
3. Examine growth rates vs. $\Delta = -0.25, -0.50, -0.75$
4. Examine growth rates vs. anisotropy mode $m = 4, 8$

$$\epsilon(\theta) = \epsilon_o [1 + \delta \cos(m\theta)]$$

Wulff plot



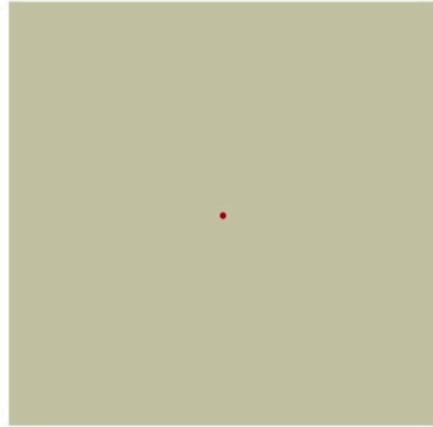
Growth Rates vs. Undercooling

Solid-liquid phase field

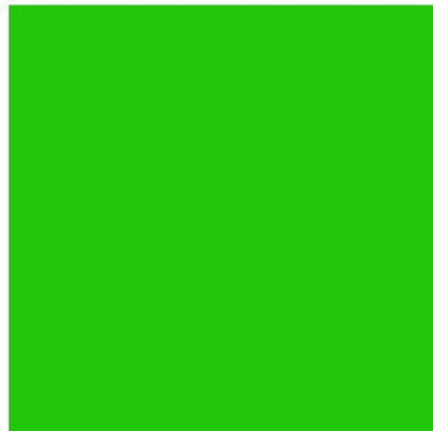
Temperature field

$$\Delta = -0.25$$

Solid/Liquid OP

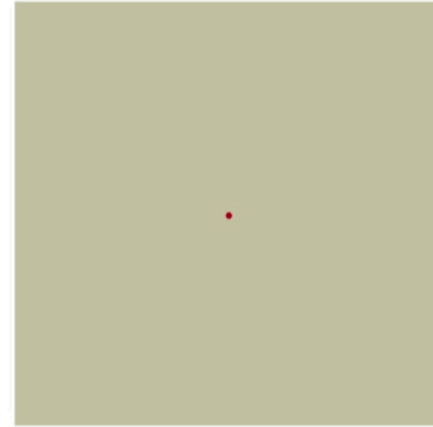


Temperature Field



$$\Delta = -0.50$$

0 1

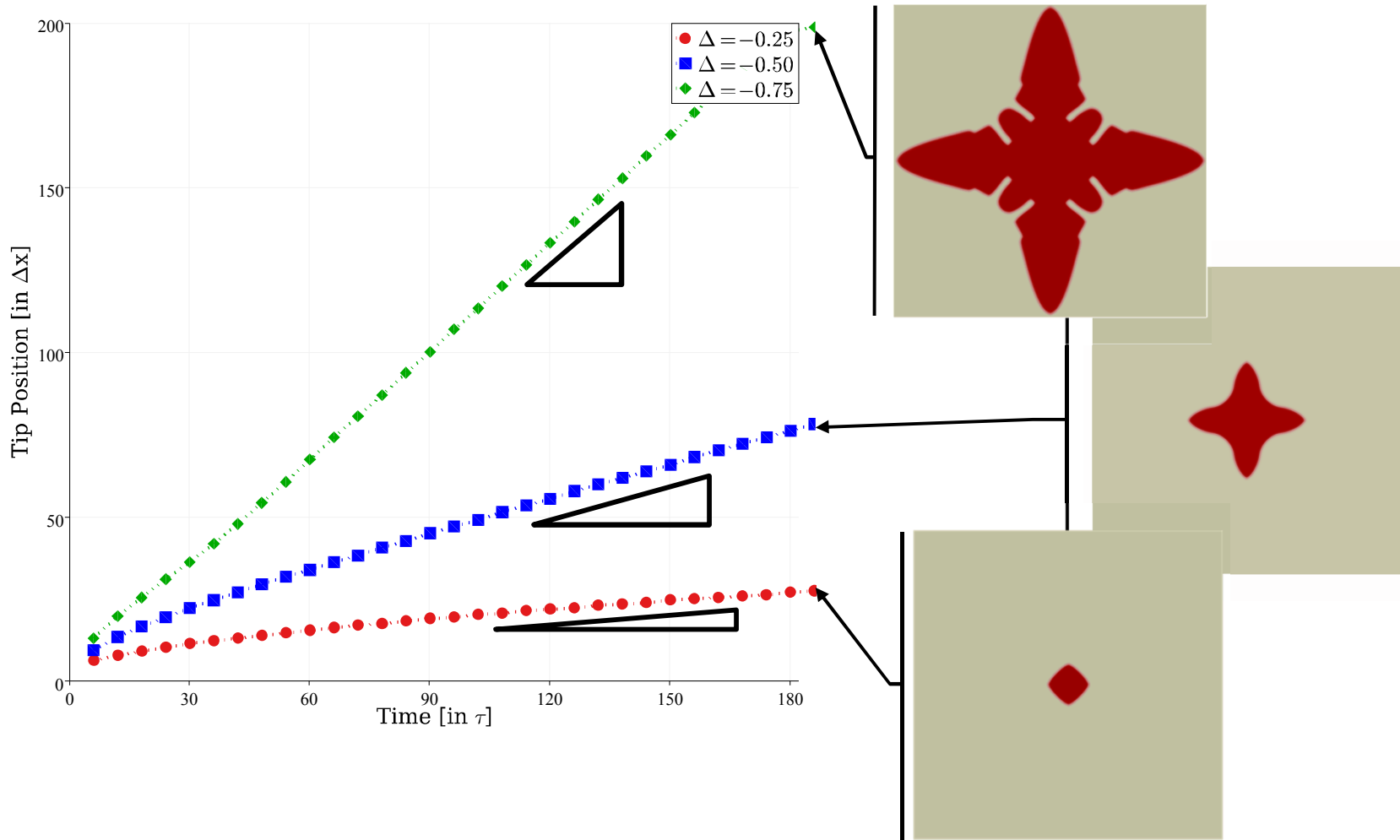


-0.5 -0.25 0



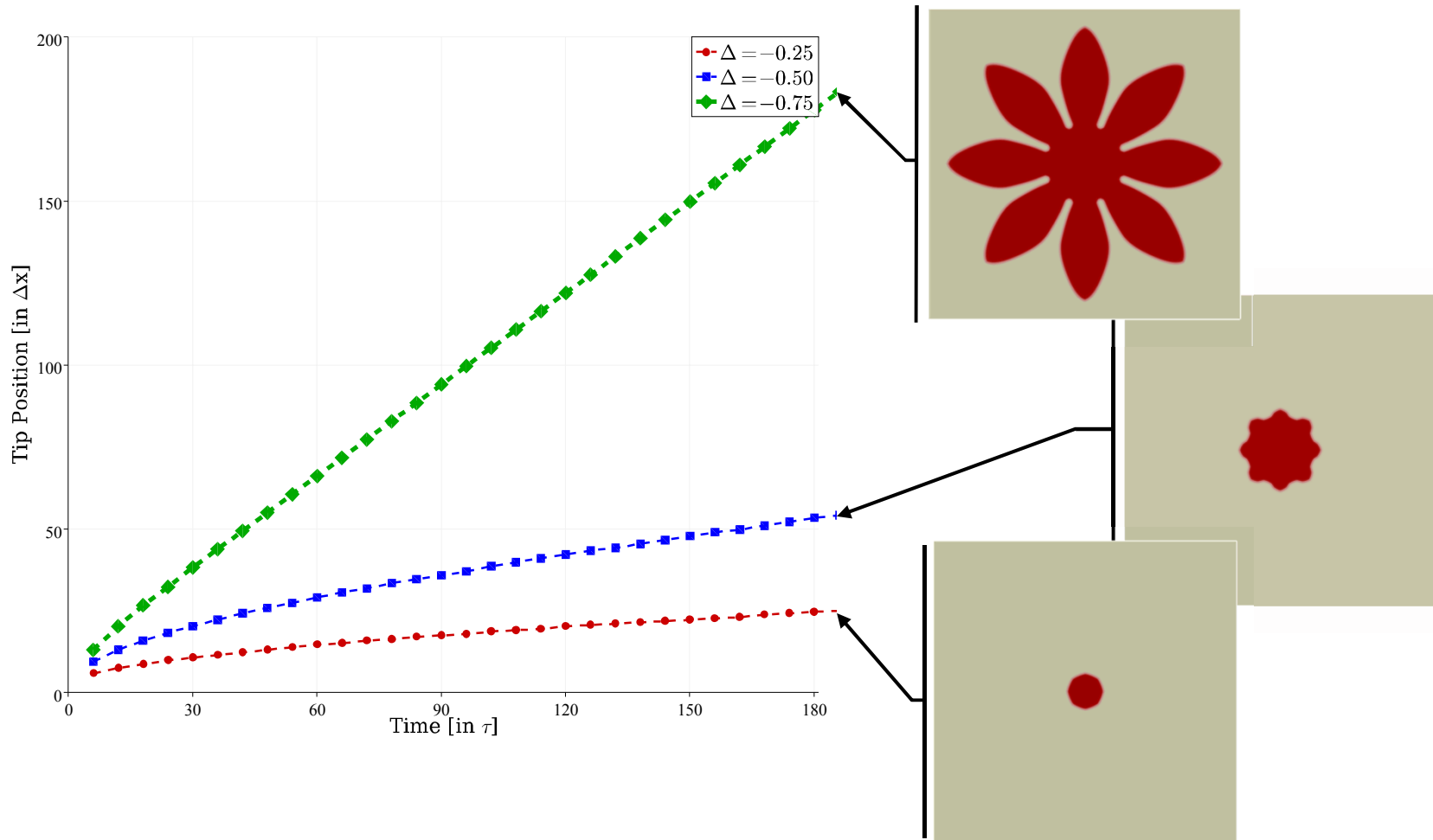
Growth Rates vs. Undercooling

- Dendrite tip speed



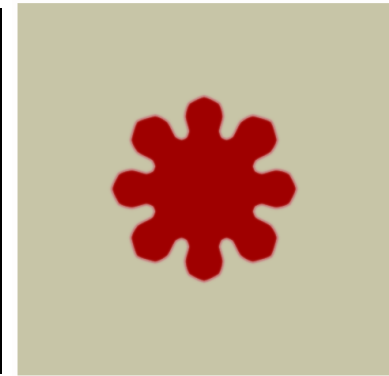
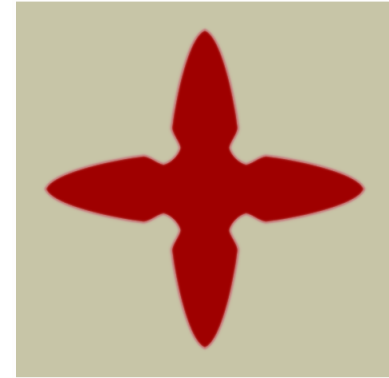
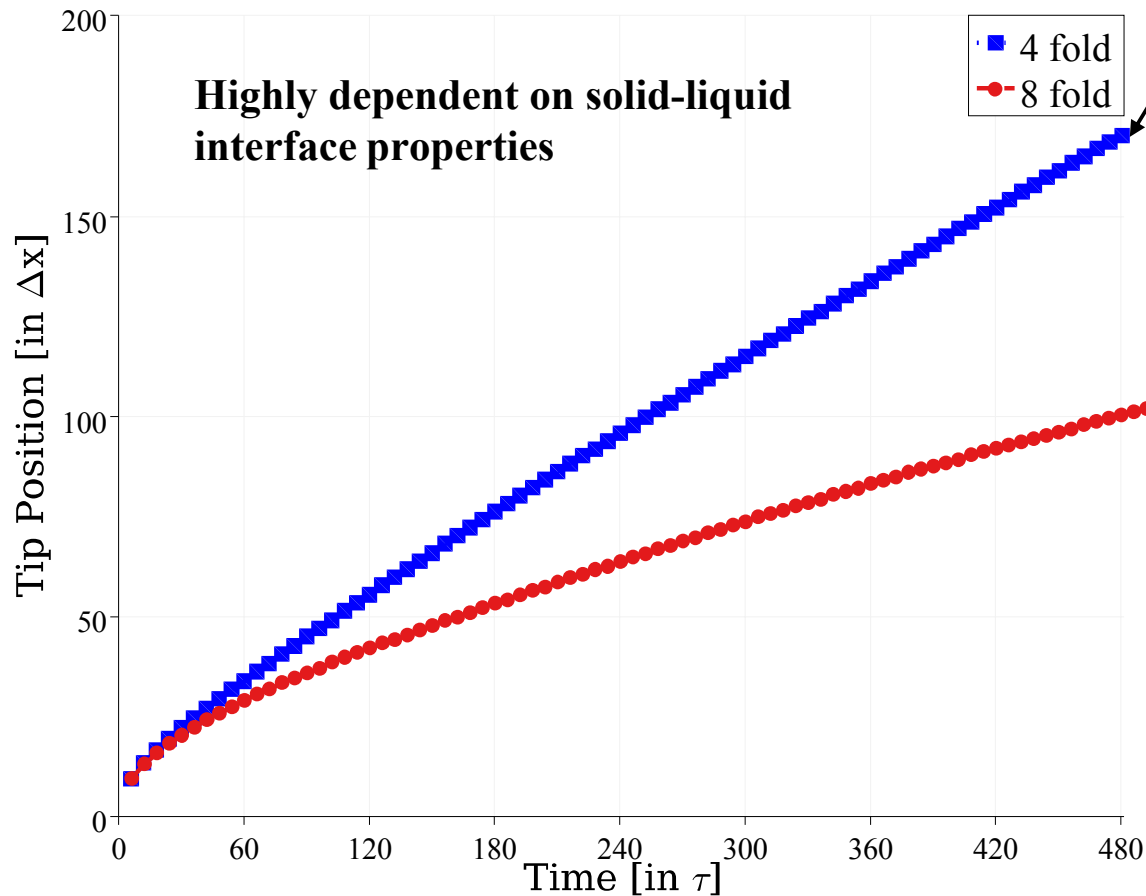
Growth Rates vs. Anisotropy

- Dendrite tip speed



Growth Rates vs. Anisotropy

- Undercooling: $\Delta = -0.50$
- Vary interface anisotropy strength/type $m = 4, 8$



Directional Solidification

