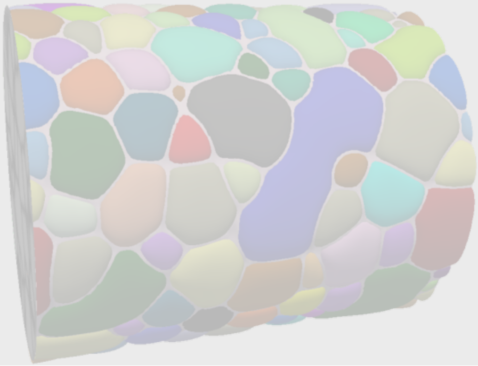
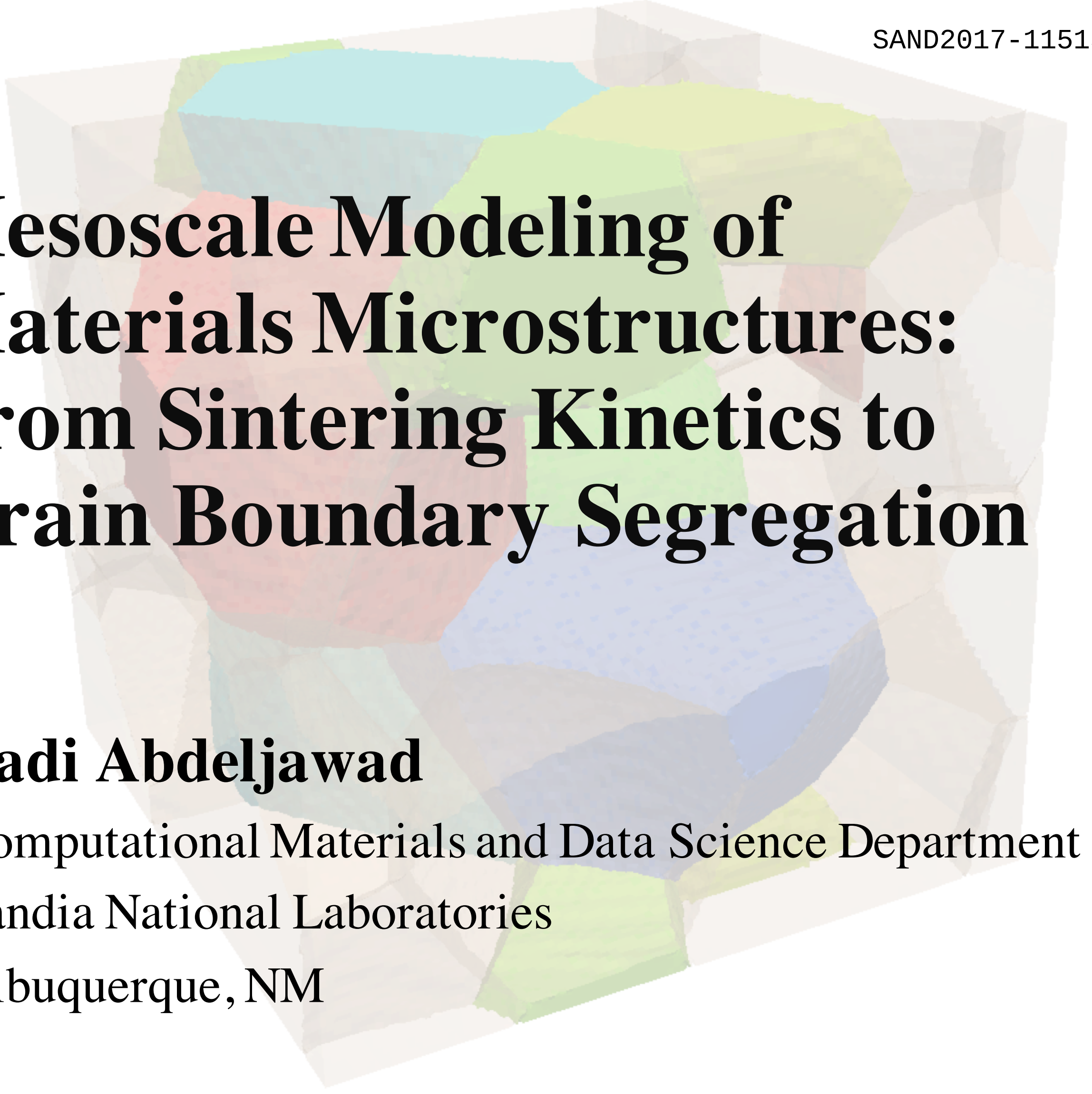
SAND2017-11515PE

$$v_n = M\gamma H \qquad \gamma = \Delta F - \Gamma \frac{\partial f_{mix}}{\partial c}\bigg|_{eq}$$

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





Mesoscale Modeling of Materials Microstructures: From Sintering Kinetics to Grain Boundary Segregation

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Albuquerque, NM

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Exceptional

service

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

U.S. DEPARTMENT OF ENERGY | Office of Science

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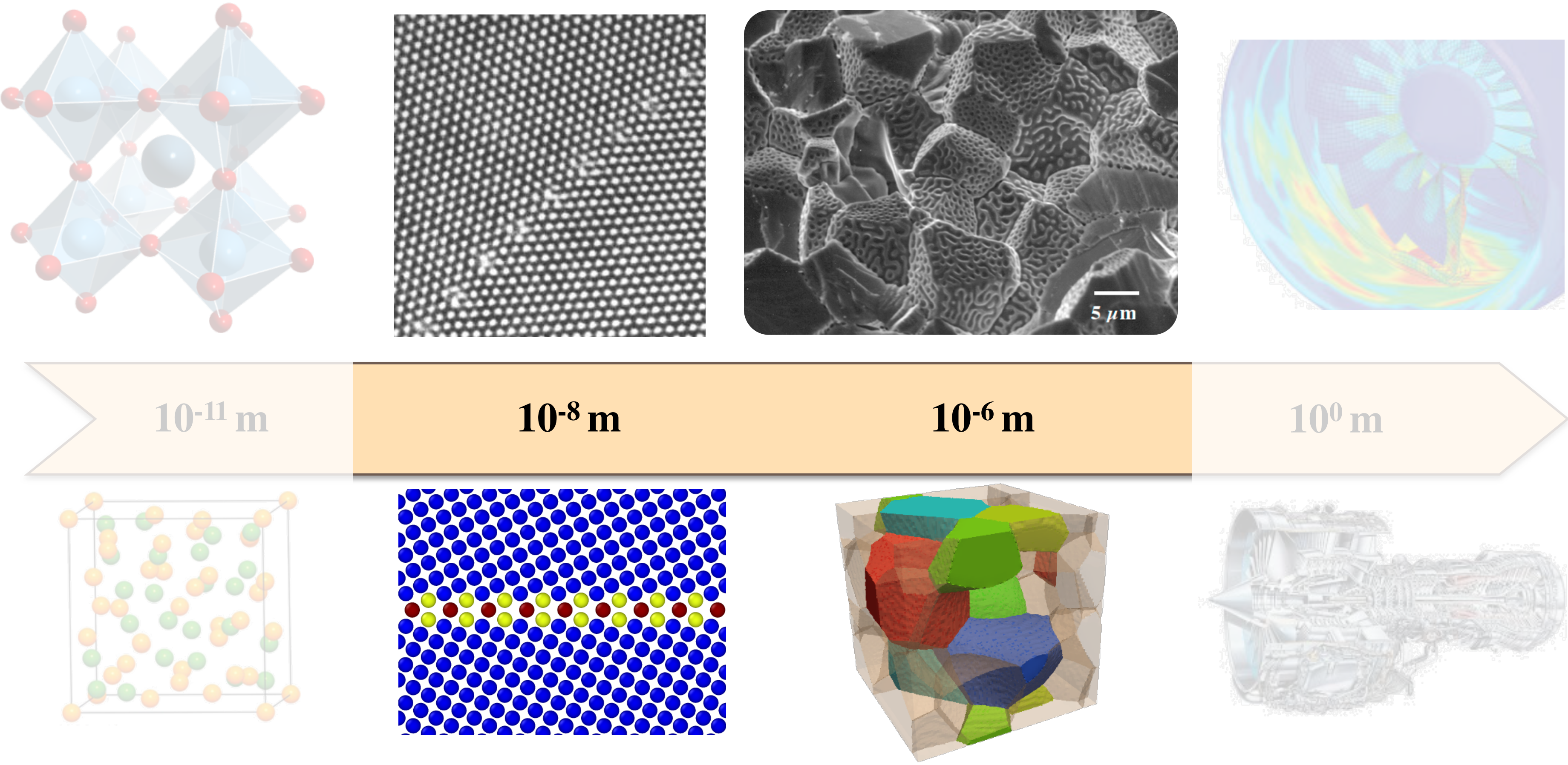
Materials Modeling

- **Meso:** (Etymology) Greek “mésos” middle



10^{-11} m

10^{-8} m

10^{-6} m

10^{0} m

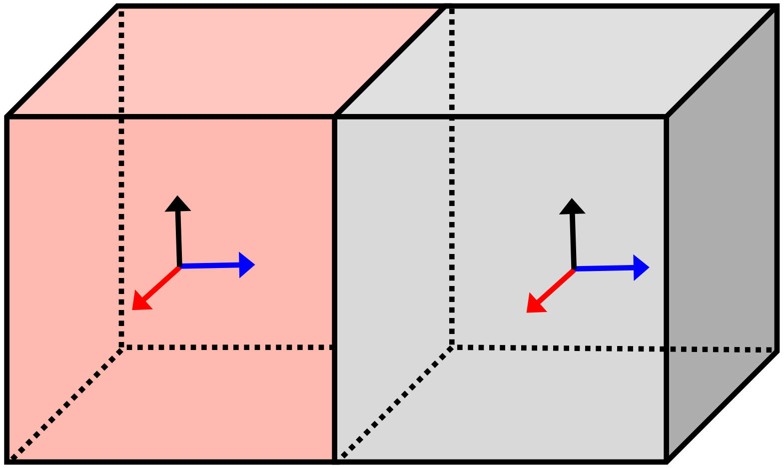
Outline

- **Preliminaries:** Interfaces at the meso-scale
  - Grain boundaries (GBs)
- **Part I:** Sintering kinetics: Application to 3D printing
  - Mesoscale model of coarsening and densification
  - Pore space evolution and shrinkage
  - GB anisotropy
- **Part II:** GB solute segregation in nanocrystalline metals
  - The Gibbs adsorption equation and GB segregation isotherms
  - Enhanced thermal stability
  - Competing processes: Phase transitions, grain growth and GB segregation
- Concluding remarks and future work

Preliminaries

Grain Boundaries (GBs)

- Start with a perfect crystal
- Cut into halves



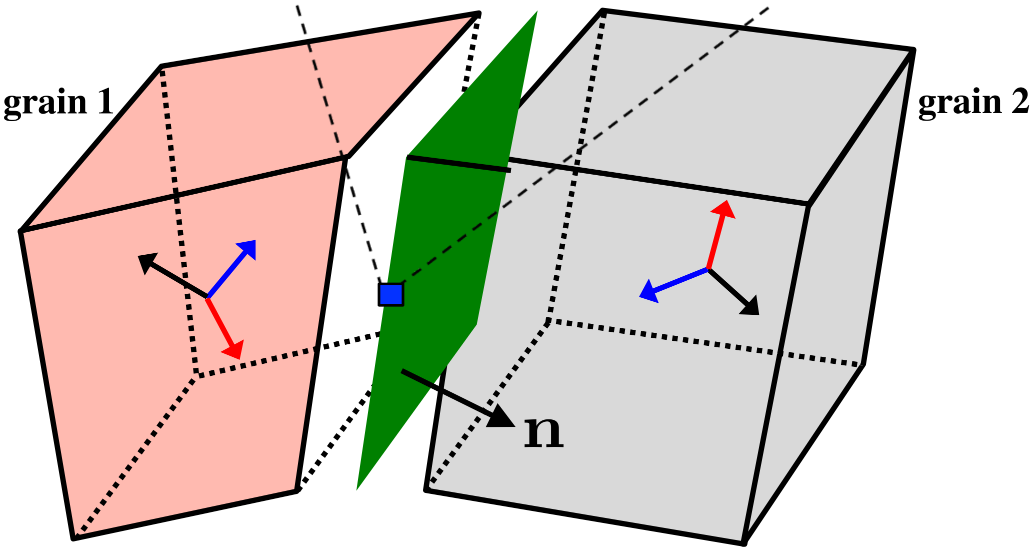
Grain Boundaries (GBs)

- Start with a perfect crystal
- Cut into halves
- Rotate each half independently
- Glue back the two halves

GB geometry described by five "macroscopic" degrees of freedom

- **Misorientation:** (3 DOF)
Angles {$\boldsymbol{\phi}_1$ $\boldsymbol{\Phi}$ $\boldsymbol{\phi}_2$} to rotate grain 2 into 1

- **Inclination:** GB plane normal (2 DOF)
n unit vector. {$\boldsymbol{\theta}_1$ $\boldsymbol{\theta}_2$} polar angles



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

C. Humphreys (some argue it is by C. Frank)

Grain Boundaries (GBs)

Free energy: $\gamma_{gb} = \underbrace{\gamma_o}_{\text{High school definition}} + f\underbrace{(\{\theta_1, \theta_2\}, T, \mu_i, \{\phi_1, \Phi, \phi_2\})}_{\text{The real deal}}$

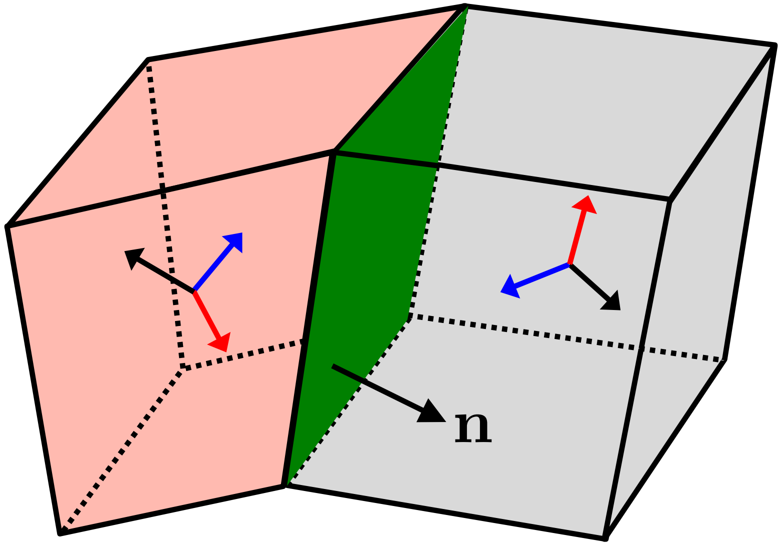
6+i dimensional space

Cahn, J. Phys. (1982)

GB geometry described by five "macroscopic" degrees of freedom

- **Misorientation:** (3 DOF)
Angles {$\boldsymbol{\phi}_1$ $\boldsymbol{\Phi}$ $\boldsymbol{\phi}_2$} to rotate grain 2 into 1

- **Inclination:** GB plane normal (2 DOF)
n unit vector. {$\boldsymbol{\Theta}_1$ $\boldsymbol{\Theta}_2$} polar angles



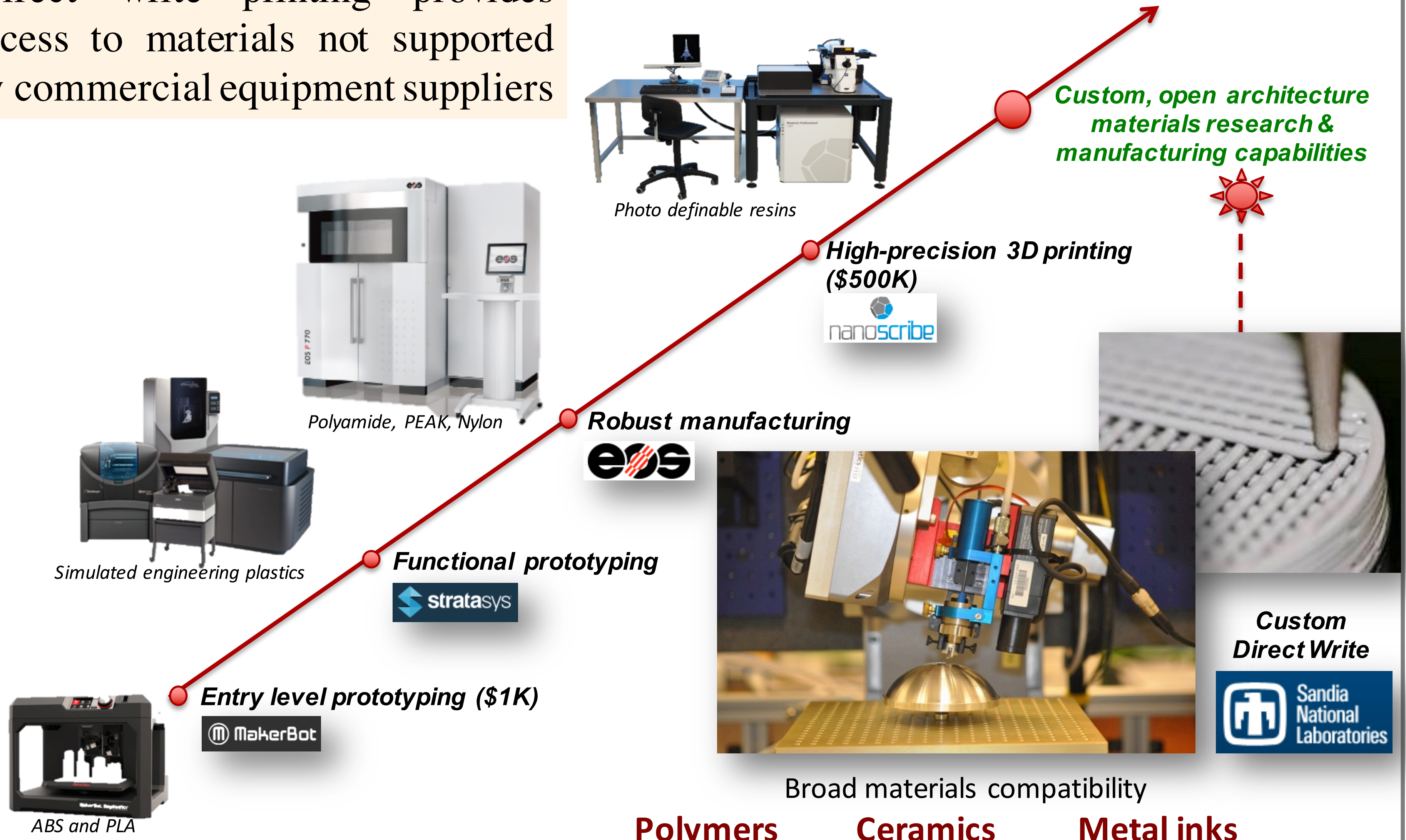
Crystals are like people, it is the defects in them which tend to make them interesting

C. Humphreys (some argue it is by C. Frank)

Sintering Kinetics:
Application to Direct Ink Write

Additive Manufacturing of Ceramics

Direct write printing provides access to materials not supported by commercial equipment suppliers



ABS and PLA

Entry level prototyping ($1K)

MakerBot

Simulated engineering plastics

Functional prototyping

stratasys

Polyamide, PEAK, Nylon

Robust manufacturing

eos

Photo definable resins

High-precision 3D printing ($500K)

nanoscribe

Custom, open architecture materials research & manufacturing capabilities

Custom Direct Write

Sandia National Laboratories

Broad materials compatibility

Polymers **Ceramics** **Metal inks**

Ceramic AM: Composite Processing

Powder
(particle size, distribution)

Mix with vehicle

Direct Write

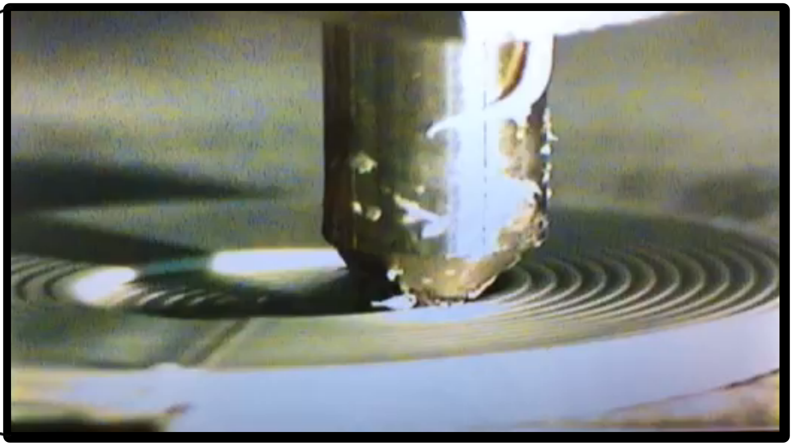
De-bind
(binder burn out)

Shaped powder form
(green body)

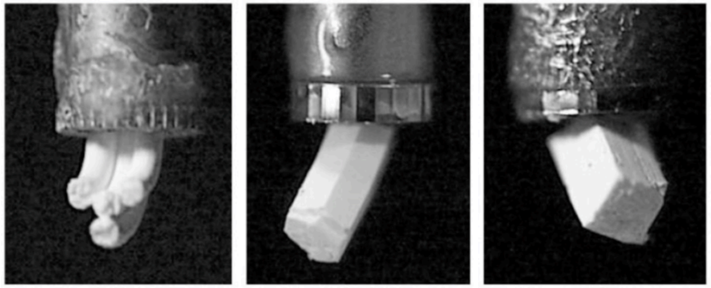
Sintering (~ 0.5 – 0.7 T_m)

Dense polycrystalline product

Direct Write printing of composite alumina (video)



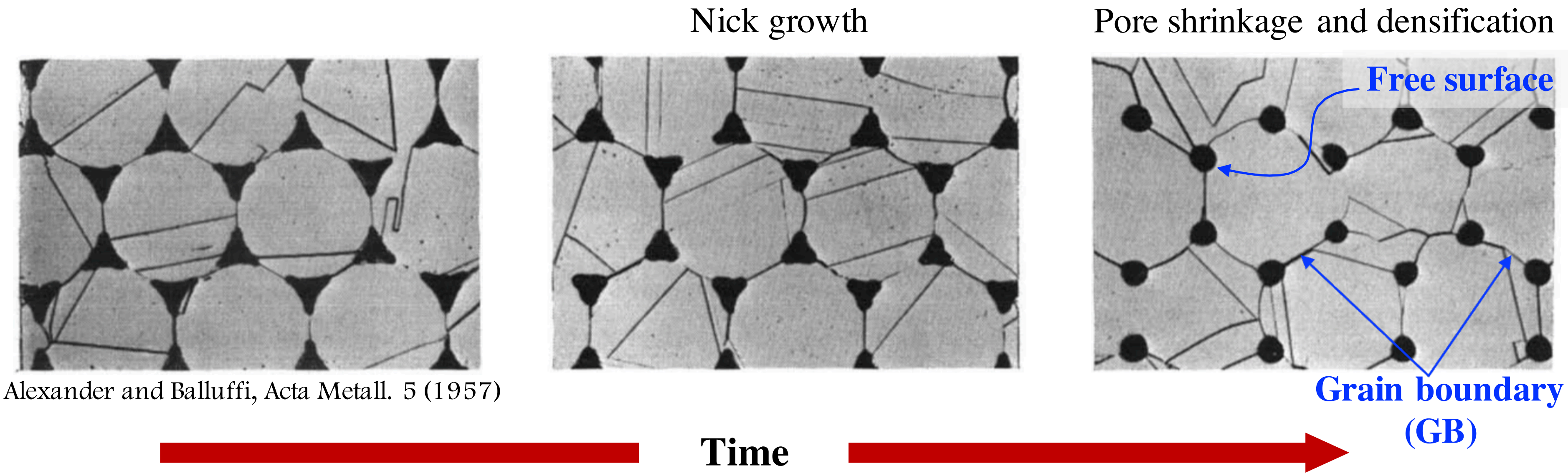
- ○ Computer-controlled translation stage
- ○ Moves a heated ink-deposition nozzle
- ○ Patterns materials with complex architectures



Lewis, Curr. Opin. Solid State Mater. Sci. **6** (2002)

Sintering: Basic Concepts

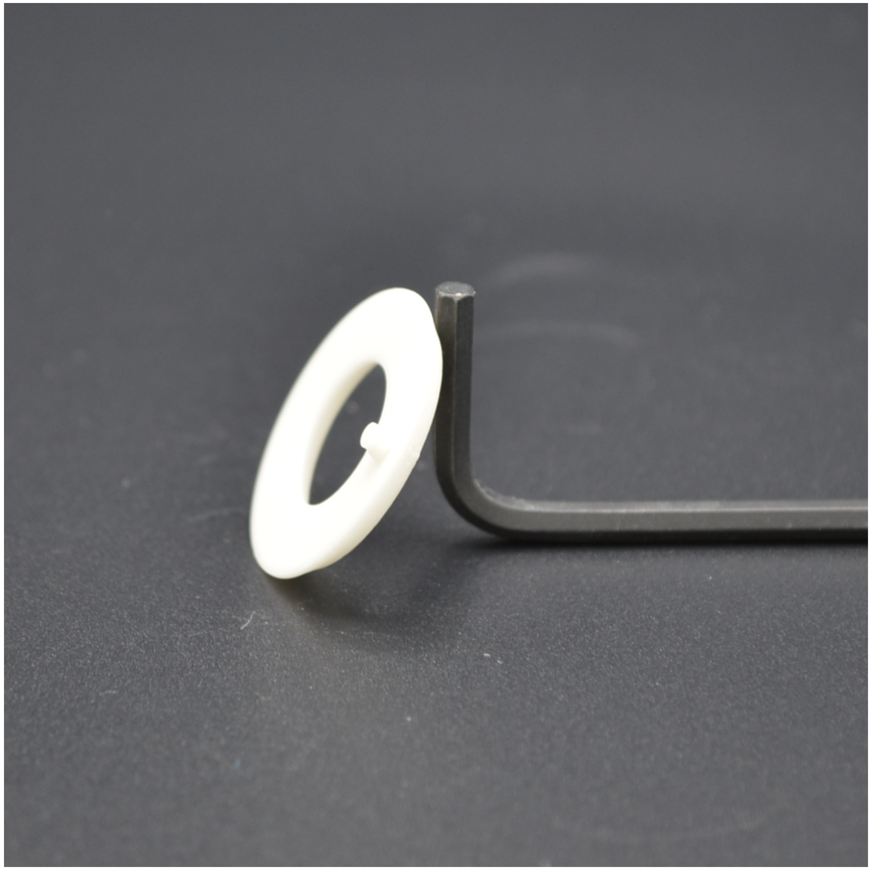
A heat treatment that leads to the consolidation of a powder compact by reduction of its surface area



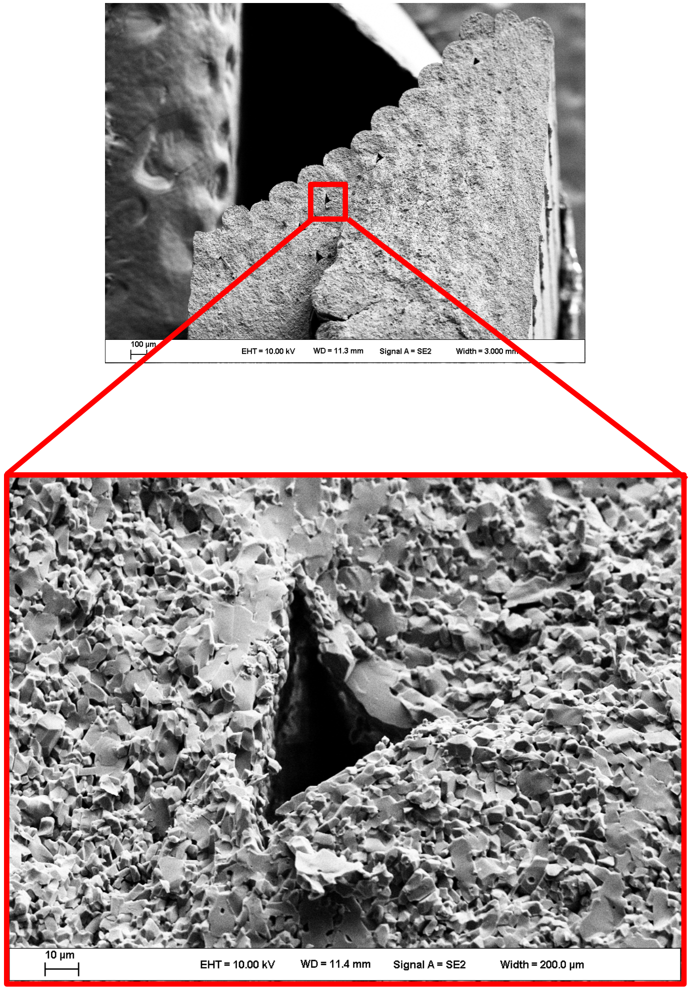
Alexander and Balluffi, Acta Metall. 5 (1957)

- Diffusional transport of matter (surface, GB, lattice, etc.)
- Moving boundary problem (Stefan conditions)
- Two competing processes:
    - **Densification**: reduction in porosity (shrinkage of body)
    - **Coarsening**: free surface evolution and grain growth processes

Inter- vs. intra-filament porosity



Bimodal distribution of pores



Adam Cook, Sandia National Labs (2017)

Key Questions

Key physical processes (mother nature)

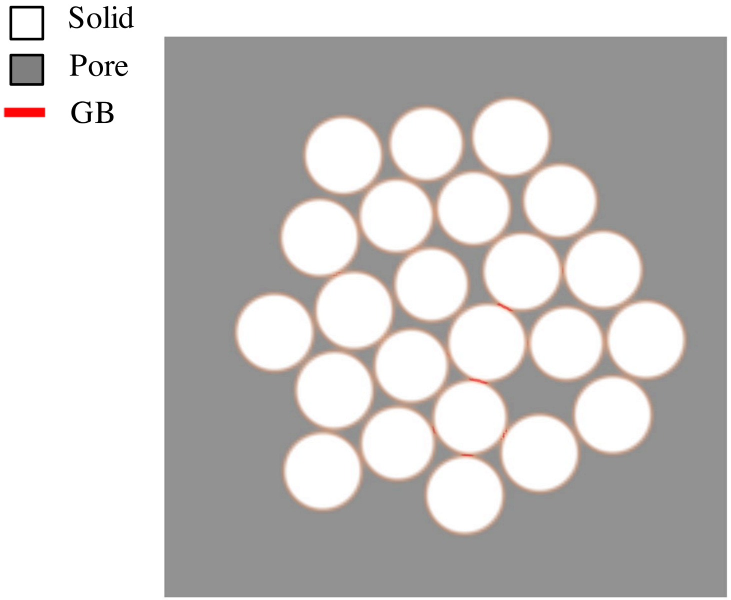
- Coarsening and surface smoothing
- Densification and pore shrinkage
- Mass transport mechanisms
- Interface properties and anisotropies

Processing parameters (our bag of tricks)

- Particle size and/or distribution
- Volume fractions
- Additives
- Thermal profile/history

Mesoscale phase field model

- Captures key processes
- Evolution at diffusive scales
- Examines the role of microstructure
- Scaling laws



Mesoscale: Phase Field Formalism

Sandia National Laboratories

- Free energy
  - $\phi(\mathbf{r},t)$: Conserved field (solid-vapor field)
  - $\eta_i(\mathbf{r},t)$, $\{i = 1, \ldots, n_\eta\}$: grain structural order parameters (OPs)

$$\mathcal{F}_{tot} = \int d\mathbf{r} \left[\underset{\swarrow}{f_{loc}(\phi,\vec{\eta})} + \underbrace{\frac{\kappa^2}{2}|\nabla\phi|^2 + \sum_i^{n_\eta} \frac{\epsilon_i^2}{2}|\nabla\eta_i|^2}_{\text{Gradient terms for GBs and free surfaces}} \right]$$

Local energy density

$$f_{loc}(\phi,\vec{\eta}) = m_H\phi^2(1-\phi)^2 + W\left[\phi^2 + 6(1-\phi)\sum_i^{n_\eta}\eta_i^2 - 4(2-\phi)\sum_i^{n_\eta}\eta_i^3 + 3\left(\sum_i^{n_\eta}\eta_i^2\right)^2\right]$$

Abdeljawad *et al.* Acta Mater. 126 (2017)
Wang, Acta Mater. 54 (2006)

- Dynamics

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

Conservation of mass

Diffusivity: $M_c = M_{vol}f_1(\phi) + M_{sur}f_2(\phi) + M_{gb}f_3(\vec{\eta})$

$$\frac{\partial\eta_i}{\partial t} + \nabla\cdot(\eta_i\mathcal{V}_{adv}) = -L_i\frac{\delta\mathcal{F}_{tot}}{\delta\eta_i}$$

Gradient flow (interface migration)

L_i: GB mobility

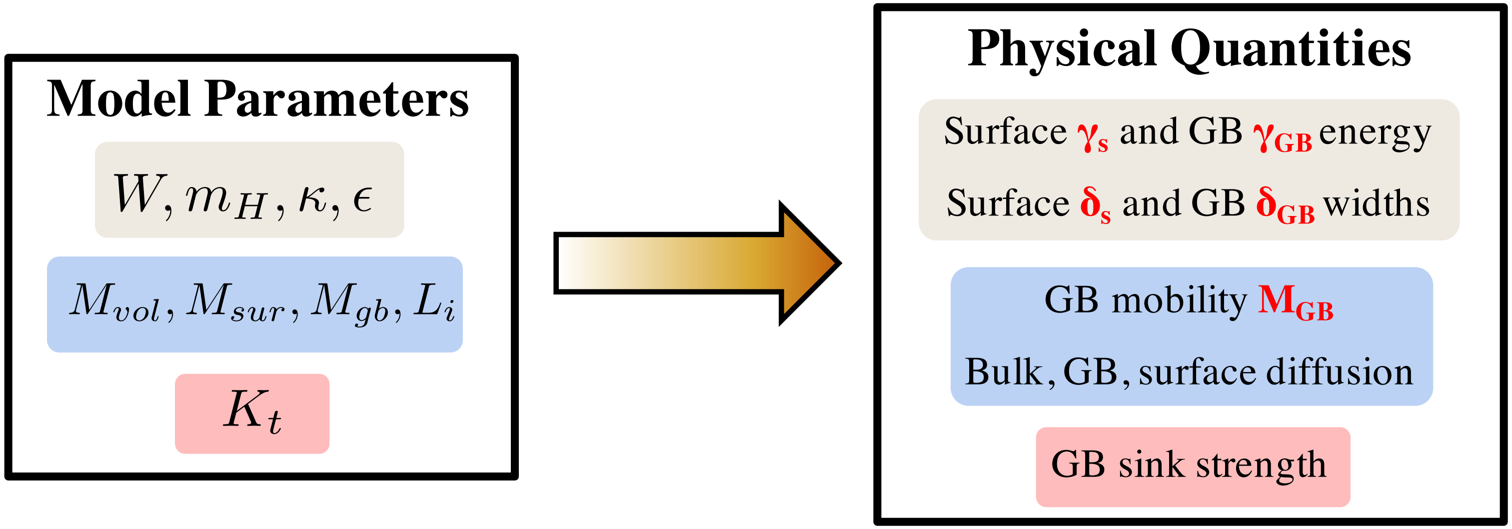
- Advection fluxes: $\mathbf{j}_{\eta_i} = \eta_i\ \mathcal{V}_{adv}$

  $\mathbf{j}_\phi = \phi\ \mathcal{V}_{adv}$

$$\mathcal{V}_{adv} = K_t\sum_i^{\alpha}\frac{\eta_i}{V_i}\int d\mathbf{r}\left[\sum_{j\neq i}^{\alpha}(\phi-\phi_o)\eta_i\eta_j\left(\nabla\eta_i - \nabla\eta_j\right)\right]$$

Abdeljawad *et al.* (In preparation)

Model Parameters



Model Parameters

W, m_H, κ, ϵ

$M_{vol}, M_{sur}, M_{gb}, L_i$

K_t

Physical Quantities

Surface γ_s and GB γ_{GB} energy

Surface δ_s and GB δ_{GB} widths

GB mobility M_{GB}

Bulk, GB, surface diffusion

GB sink strength

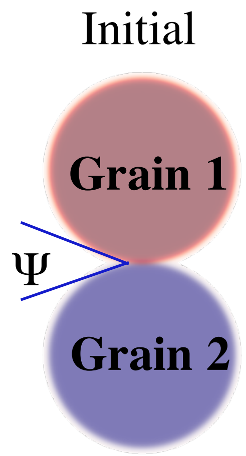
With four parameters I can fit an elephant, and with five I can make him wiggle his trunk

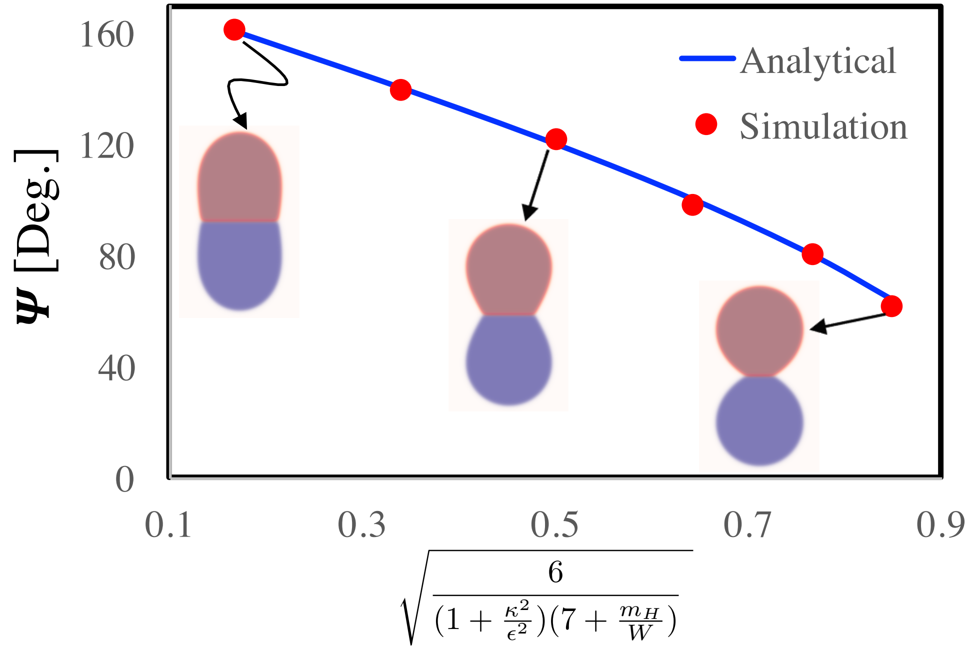
J. von Neumann

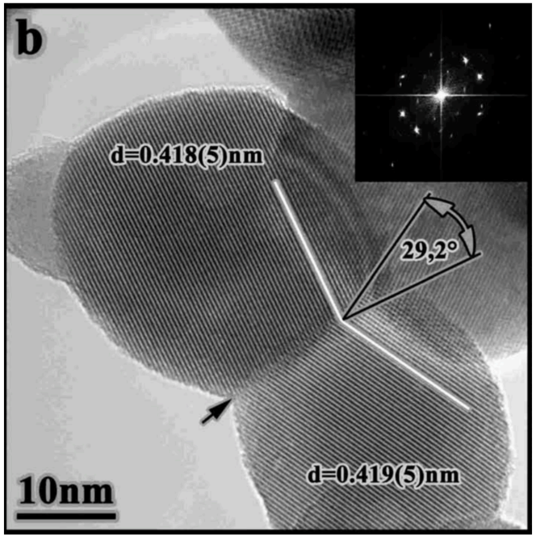
Model Parametrization

- A two particle geometry: The dihedral angle Ψ

$$\Psi = 2\cos^{-1}\left(\frac{\gamma_{gb}}{2\gamma_s}\right) = 2\cos^{-1}\left[\sqrt{\frac{6}{(1+\kappa^2/\epsilon^2)(7+m_H/W)}}\right]$$



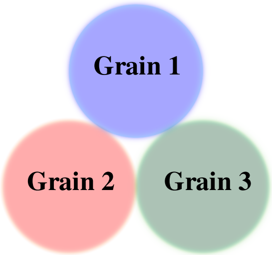


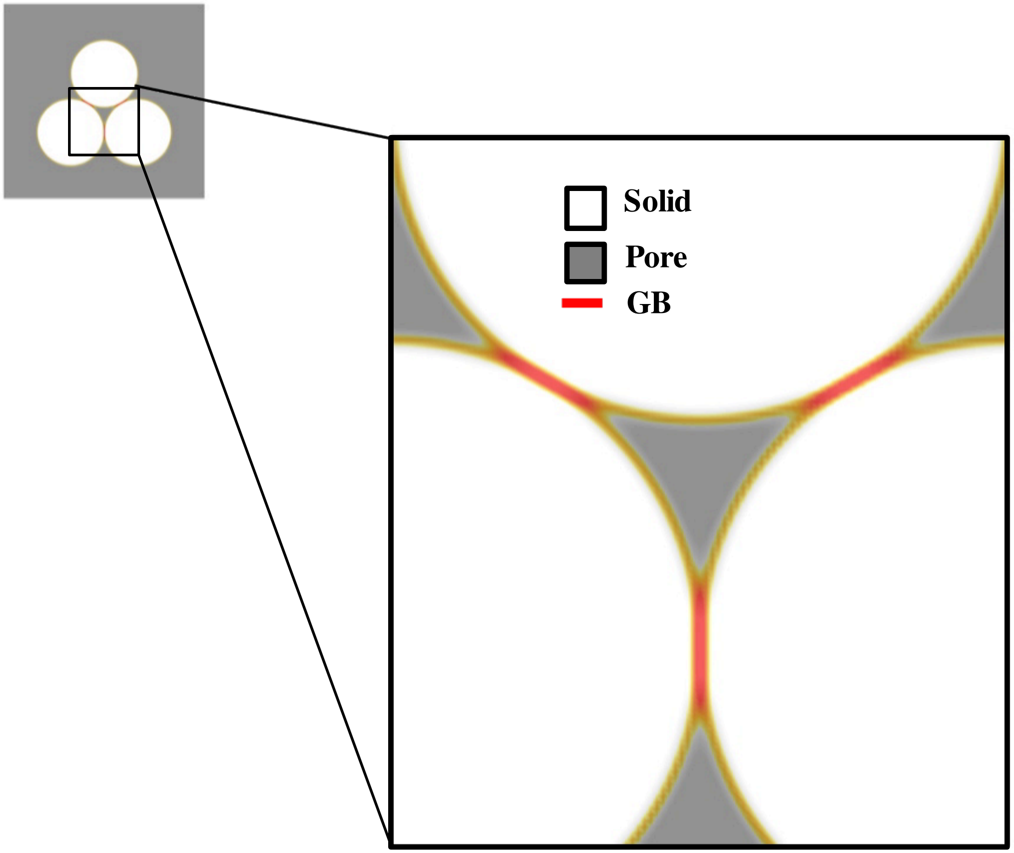


Theissmann et al., Phys. Rev. B **78** (2008)

Three Particle Geometry

- Pore shrinkage

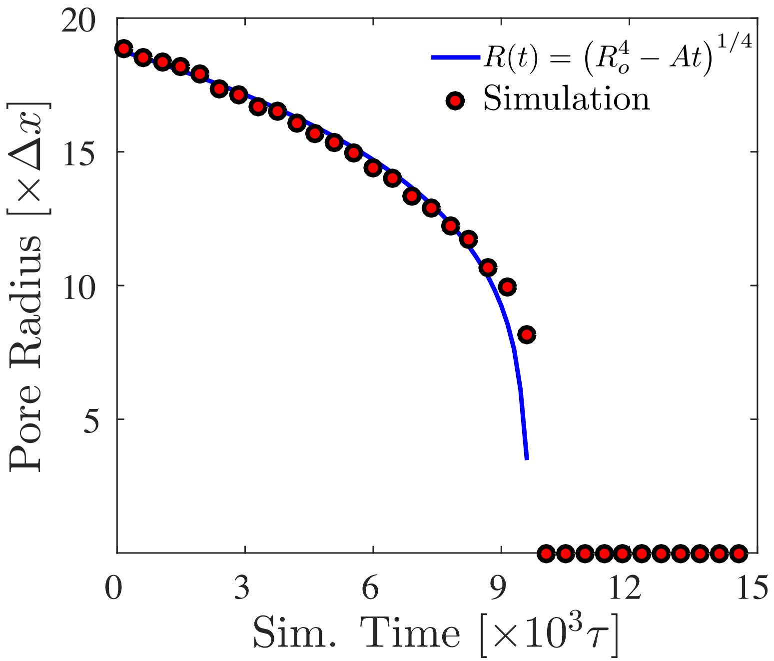




Dynamical scaling[1,2,3]

$$R^4(t) = R_o^4 - At$$

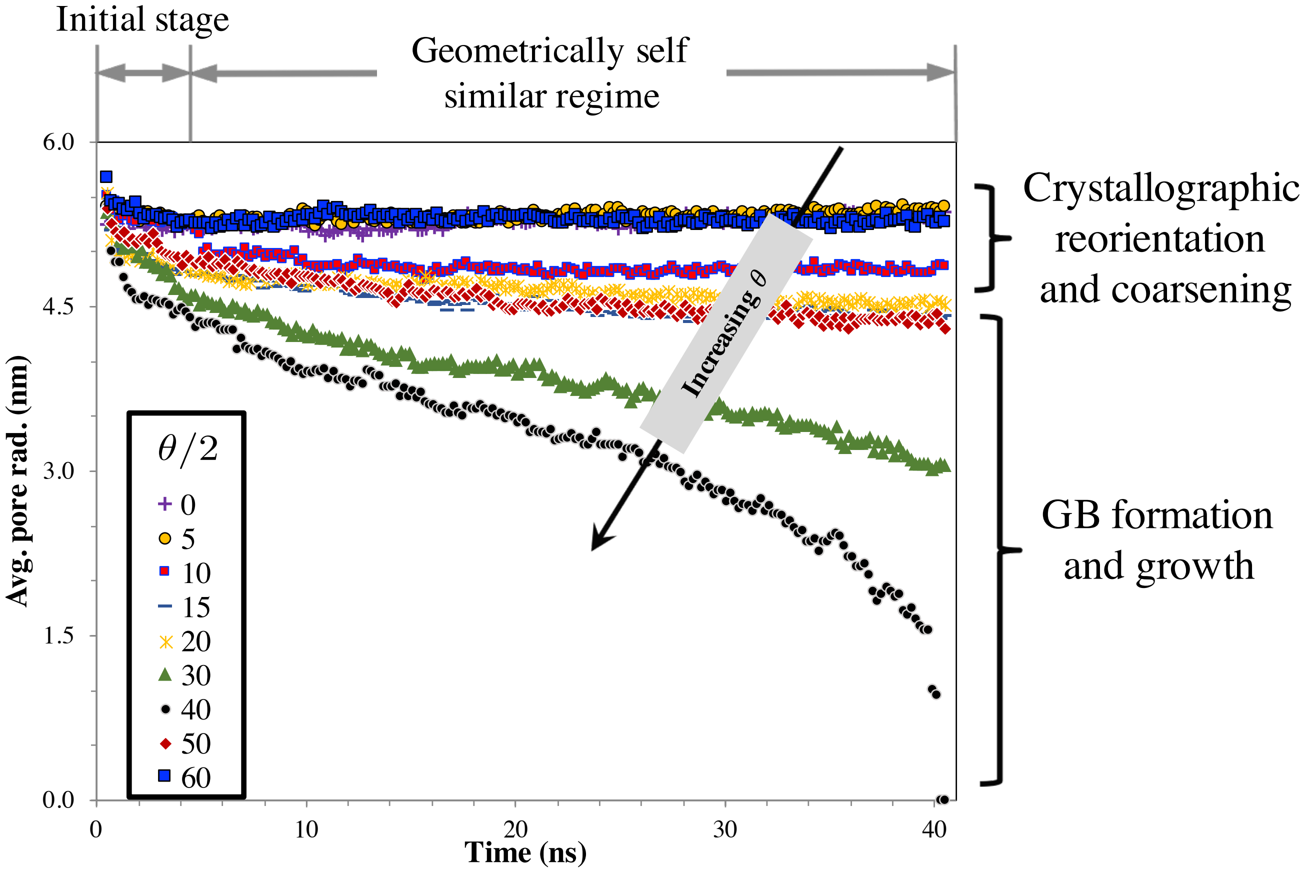
$R_o = 18.8$
$A = 13.1343$



1 Herring, App. Phys. **21** (1950)
2 Johnson, J. App. Phys. **40** (1969)
3 Tikare and Braginsky **86** (2003)

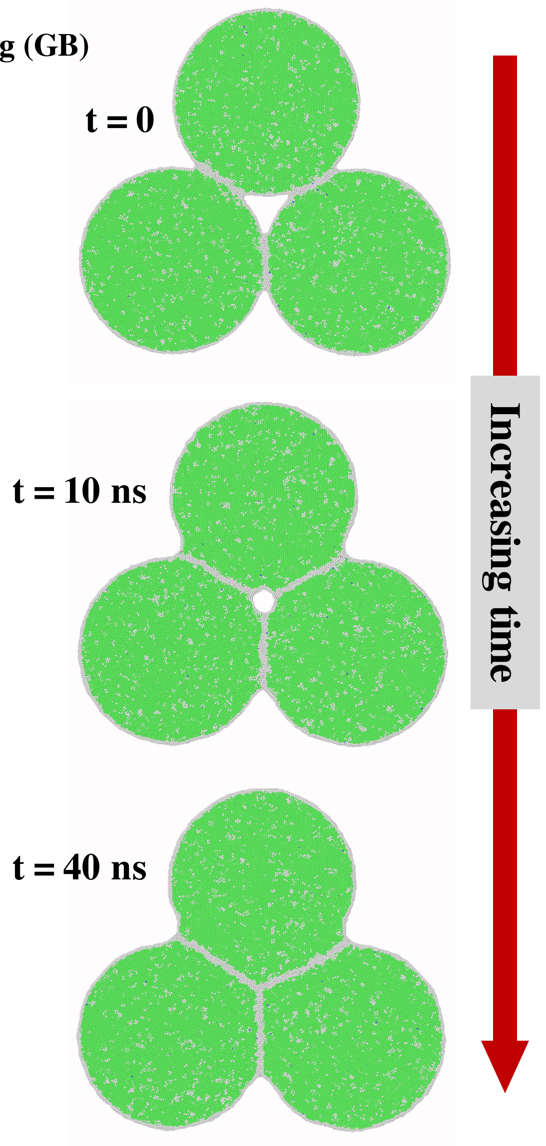
Atomistic Insight: GB Anisotropy

- Pore shrinkage vs. GB type



🟩 **FCC atoms**

☐ **No ordering (GB)**



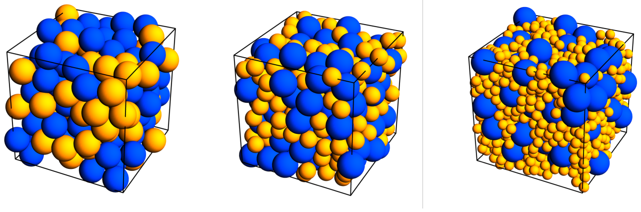
Simulation set-up

- Rotate particles about [111]; (-θ/2, + θ/2, 0) for particles (1, 2, 3)
- One GB of θ°/<111> and two GBs of θ/2°/<111>

“Virtual” Design Loop

I

Numerical generation of green microstructures



← Vary microstructural features →

II

Mesoscale model of Sintering

- Polycrystalline nature of ceramic phase
- Dominant mass transport mechanisms
- Interface thermodynamics
- Coarsening and densification

III

Track microstructural features

- Grain size
- Pore space statistics (size, shape, contiguity)
- Curvature tensor (and distribution)
- Total interfacial areas

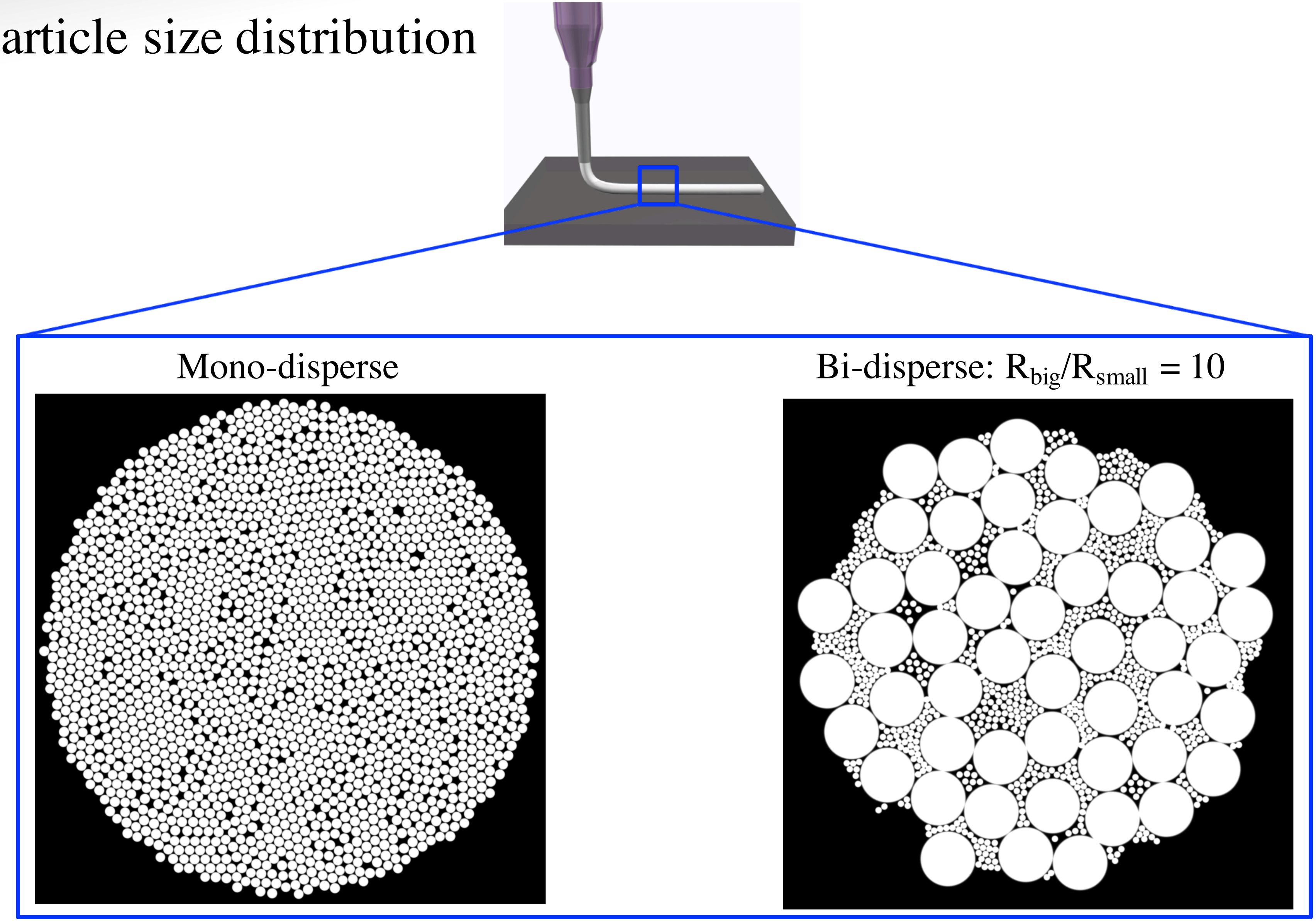
IV

Mechanical Performance

- Mechanical and structural response
- Stress risers and potential sites for failure

Filament Geometry

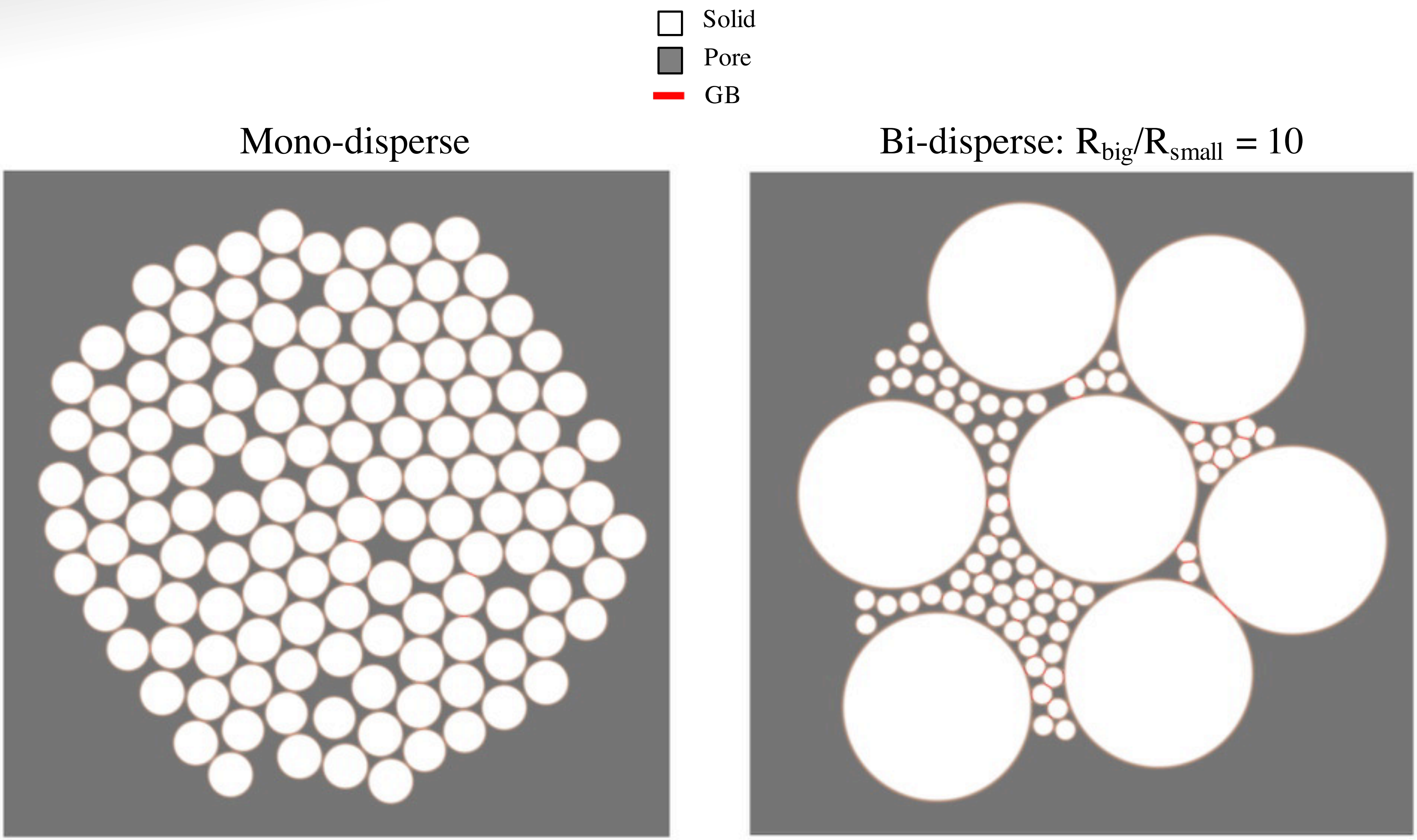
- Particle size distribution



Mono-disperse

Bi-disperse: $R_{big}/R_{small} = 10$

Results: Evolution



☐ Solid

▨ Pore

— GB

Mono-disperse

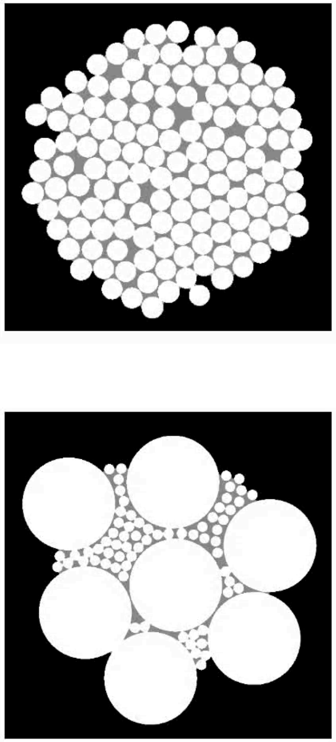
Bi-disperse: $R_{big}/R_{small} = 10$

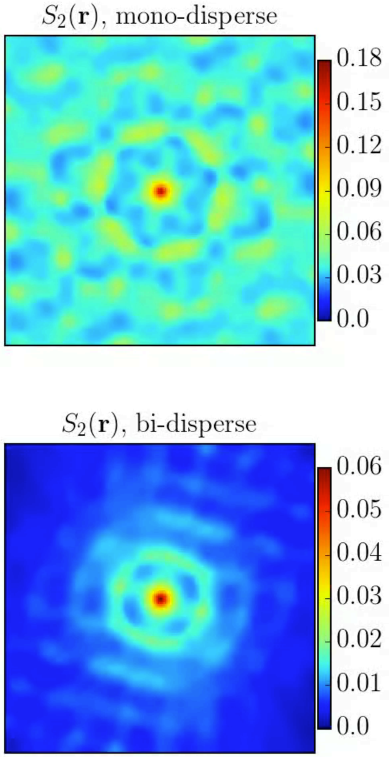
Observation: Mono-dispersity maintains GBs for longer times

Results: Quantitative

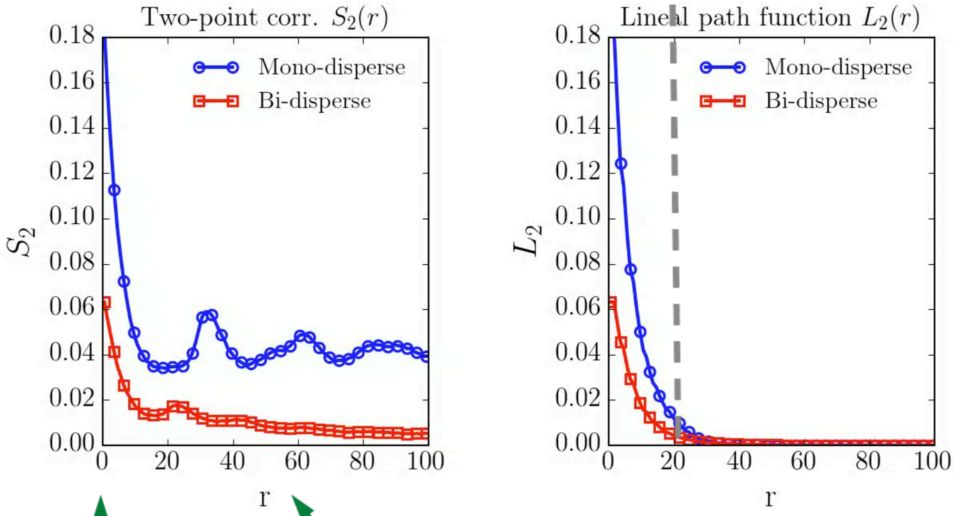
- Radial distribution and lineal path functions

☐ **Solid**
▨ **Internal pores**





Probability of finding a pore of size r = 20

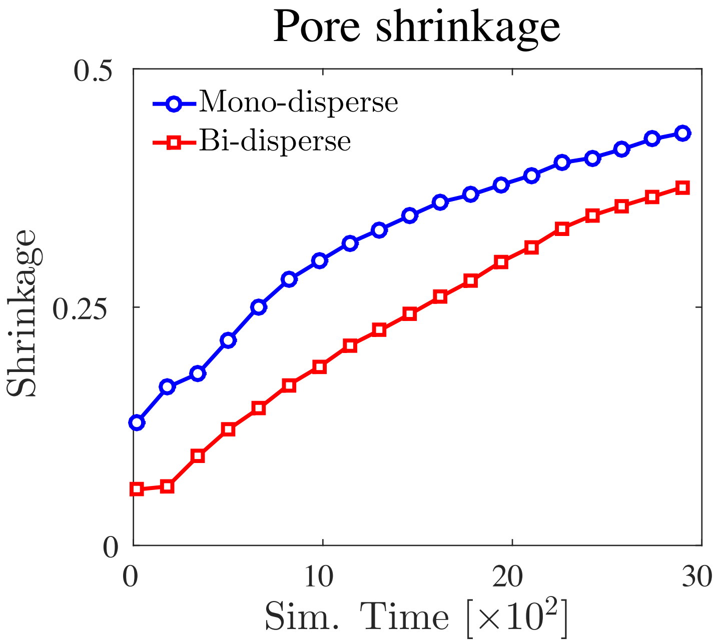


Shift in Y-intercept of S_2(r): **densification**

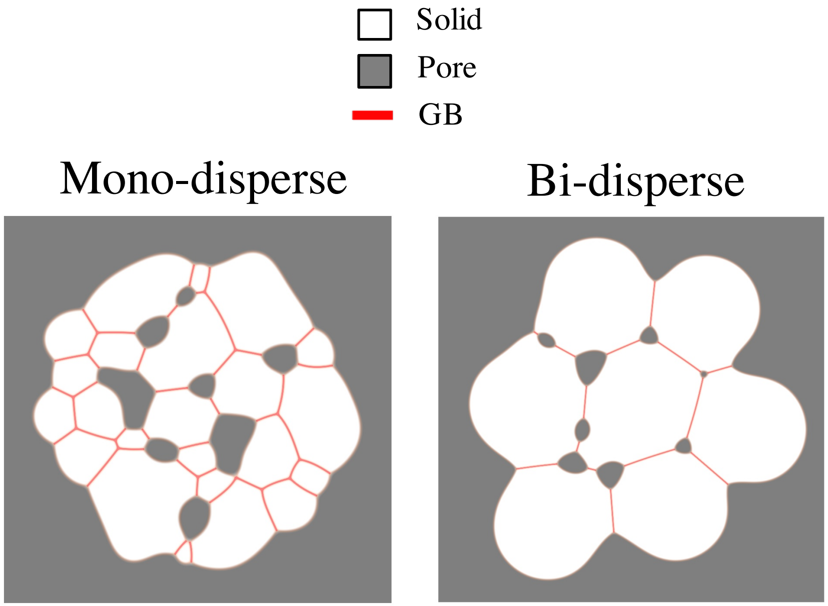
Shift in 1^{st} minima of S_2(r): **Pore growth**

Results: Pore Shrinkage

$$\text{Shrinkage} = \frac{A_{pore}(0) - A_{pore}(t)}{A_{pore}(0)}$$



Pore shrinkage



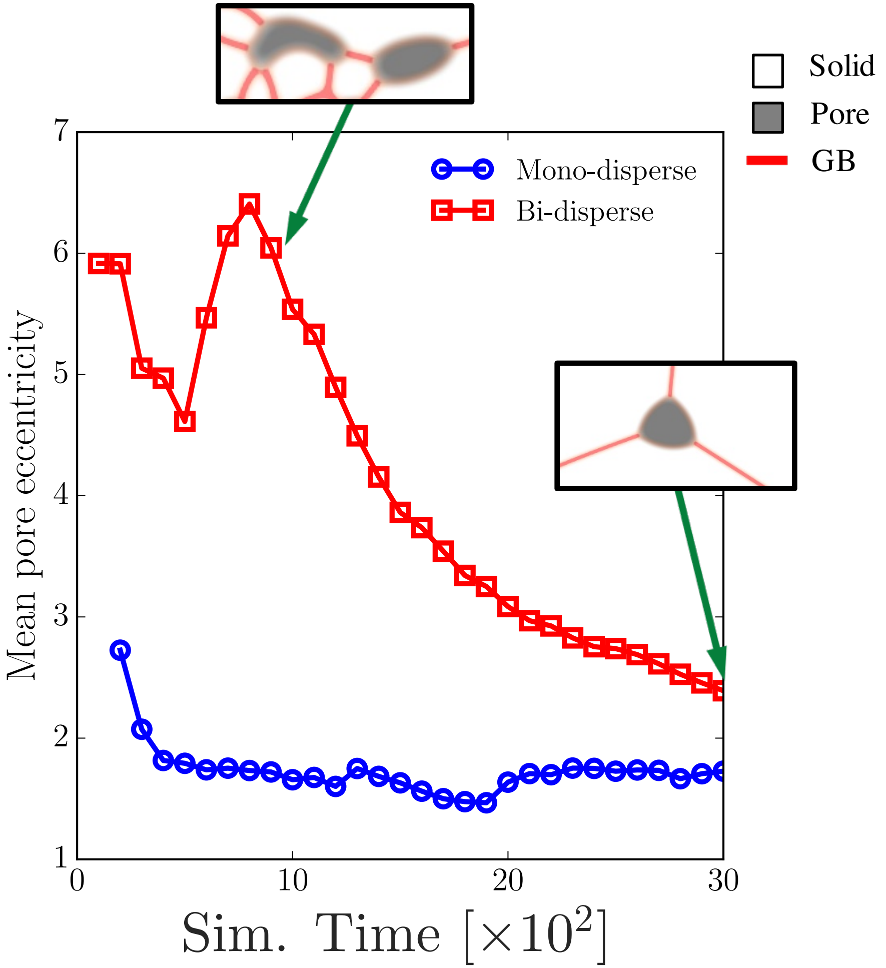
Mono-disperse

Bi-disperse

Observation: Mono-dispersity leads to faster shrinkage

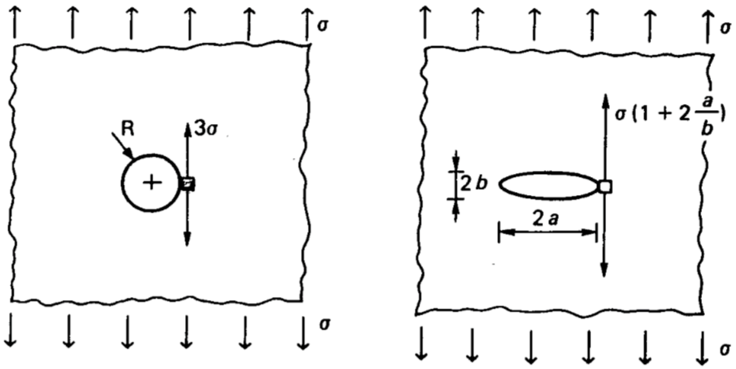
Results: Pore Eccentricity

- Eccentricity: Describes how non-spherical a domain is



Large eccentricity is bad in terms of stress amplification

$$\sigma_{max} = \sigma\left(1 + 2\frac{a}{b}\right)$$



Large eccentricity ➔ Large a/b

Analytical solutions by Kirsh, G. (1898) and Inglis, C. (1913)

Observation: Bi-dispersity may lead to more "bad" flaws

Results: Summary

Mono-disperse

- Poor initial packing density ✗
- Faster shrinkage kinetics ✓
- Lower eccentricity (less "bad" stress concentration sites, i.e. a/b ratio) ✓
- Pore volume fraction always higher (never catches up to mono-disperse systems) ✗

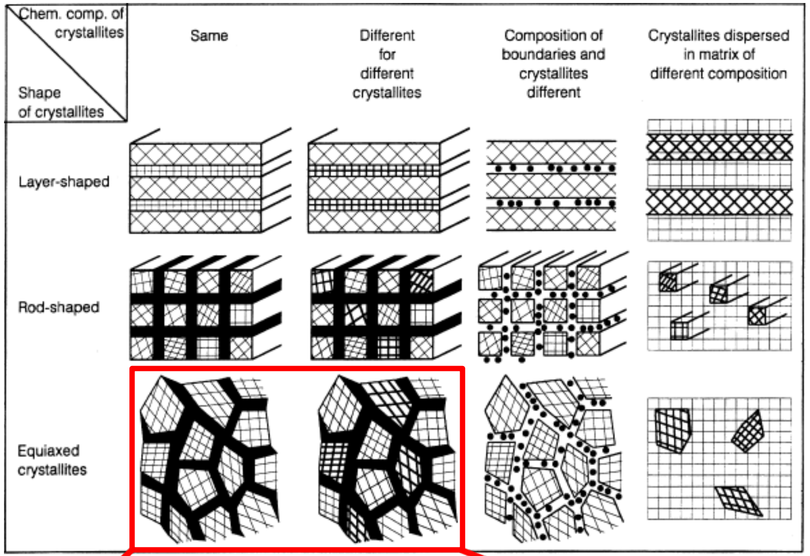
Bi-disperse

- Higher initial packing density (lower initial porosity) ✓
- Slower shrinkage kinetics ✗
- Pore density always lower than mono-disperse systems ✓
- Probability of finding critical flaw always lower ✓
- Higher eccentricity ✗

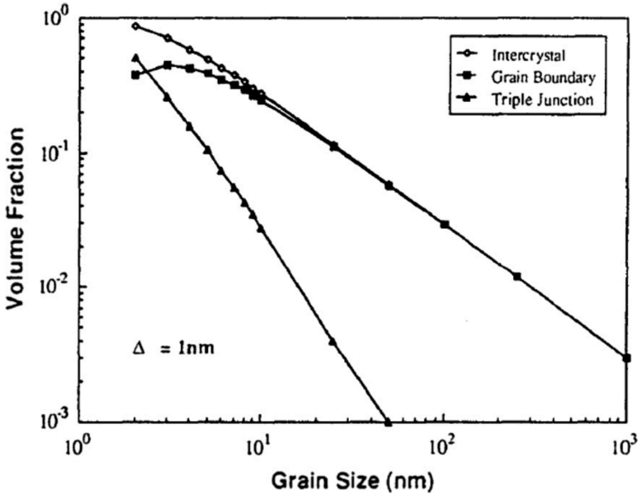
GB Solute Segregation in Nanocrystalline Materials

Nanocrystalline (NC) Metals

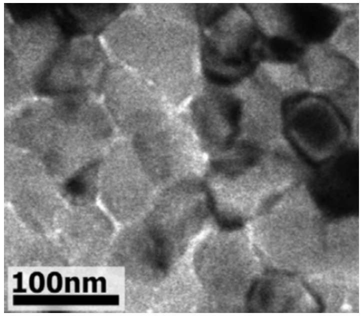
- Nanometric feature size (grain size)



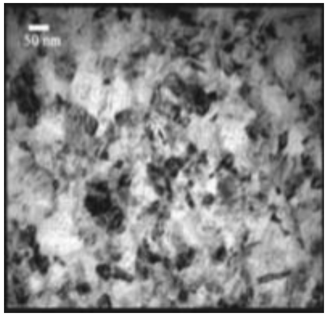
Gleiter, Acta Mater. (2000)



Palumbo *et al.*, Scr. Metall. Mater. (1990)



Vetterick *et al.*, JNM (2016)

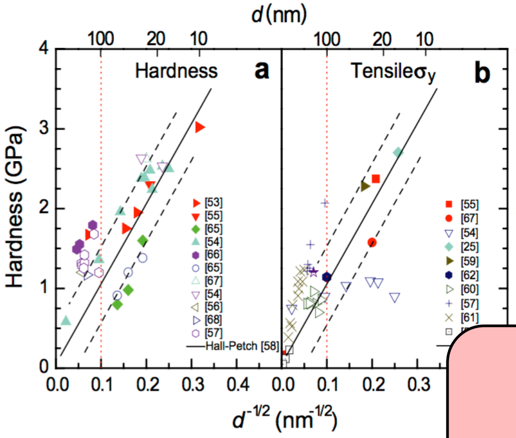


Kaoumi *et al.*, J. ASTM Intl. (2006)

Interface dominated physics

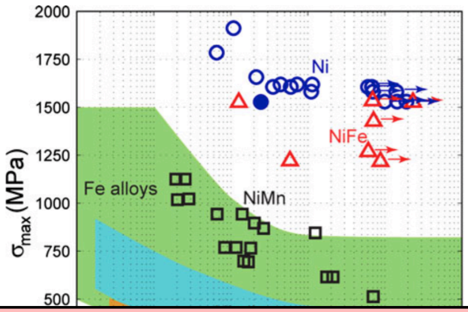
Properties and Applications

Hardness and Yield

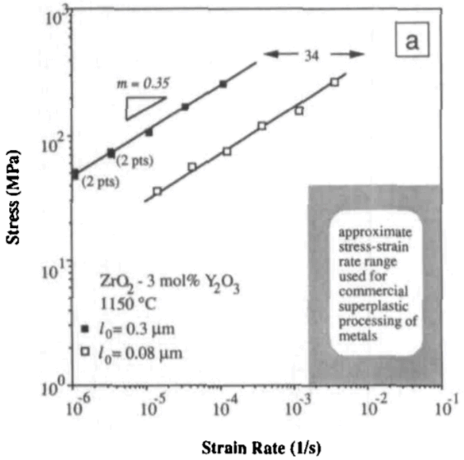


Dao *et al.*, Acta Mater. (2007)

Fatigue



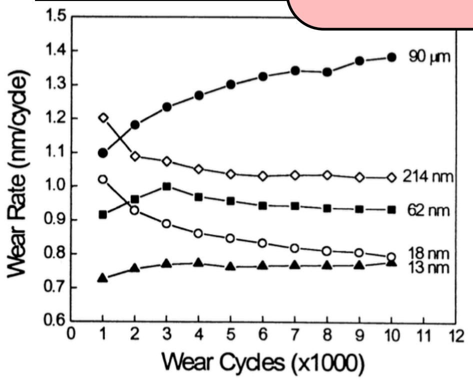
Strain Rate



Weertman *et al.*, MRS Bull. (1999)

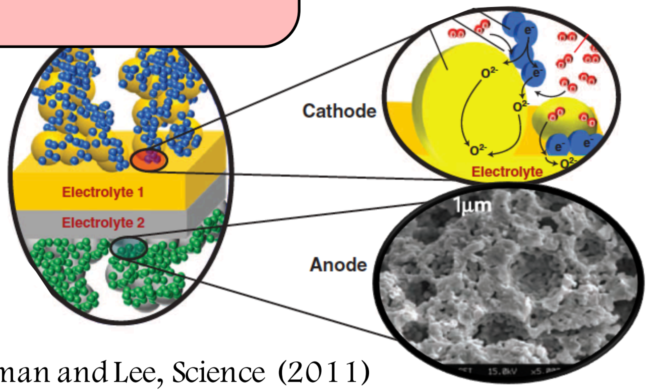
But …

Wear (electrica



Jeong *et al.*, Scripta Mat. (2001)

olid oxide fuel cells)



Wachsman and Lee, Science (2011)

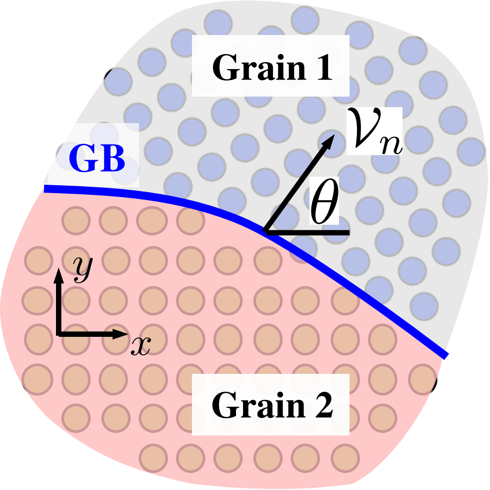
Suzuki *et al.*, Science (2009)

The Case for NC Metals

- **Problem:** High density of grain boundaries (GBs)
  - Excess free energy: $\Delta \mathcal{F} = \Delta\left(\gamma A\right)$
  - Grain-growth and homogenization processes

$$\mathcal{V}_n = M_{gb}\gamma_{gb}\mathcal{K}$$

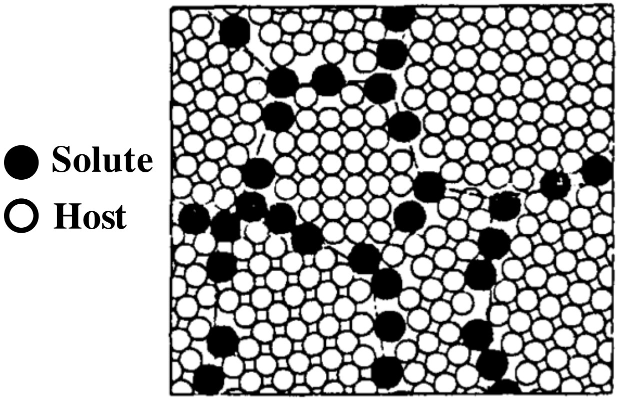
Herring, Phys. Powd. Metall. (1951)
Burke and Turnbull, Prog. Metal. Phys. (1952)
Mullins, J. App. Phys. (1956)



- **Possible solution:** Solute segregation to GBs
  - Solute atoms preferentially occupy GB sites (diffusive transport)
  - **Kinetic** and/or **thermodynamic** stabilization

GB mobility
Solute drag

GB energy



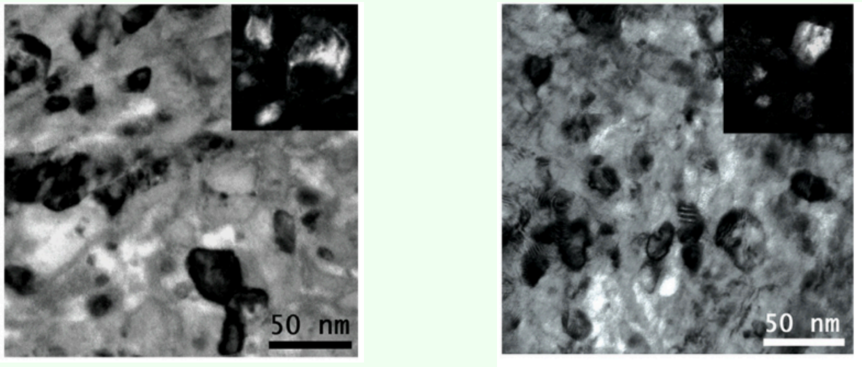
Weissmüller, Nanostruct. Mater. (1993)

An Active Area of Research

Sluggish growth dynamics
W-20 at.% Ti

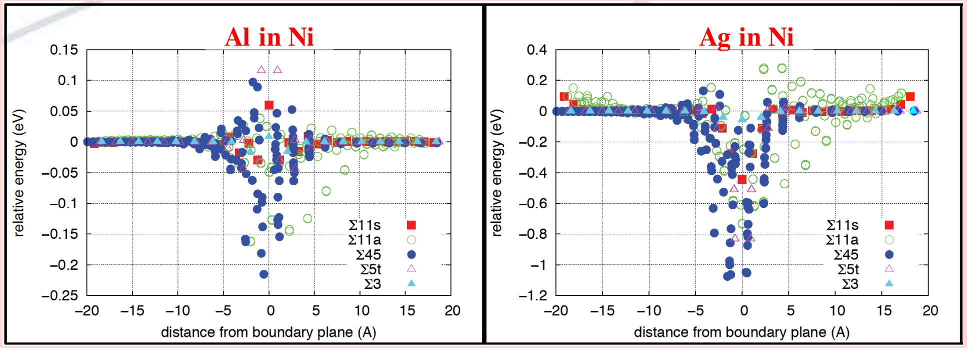
As milled

Annealed
1 week at 1100°C



Chookajorn *et al.*, Science. (2012)
Chookajorn *et al.*, Acta Mater. (2014)

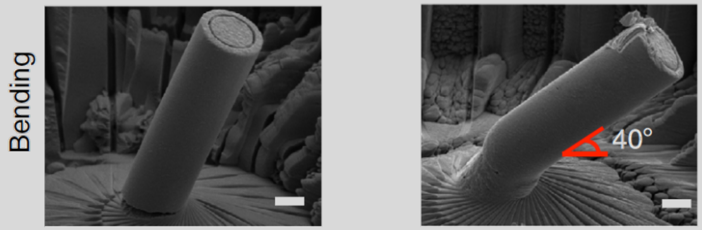
Dependency on solute and GB types



B. Uberuaga, E. Martinez *et al.*, LANL (2016)

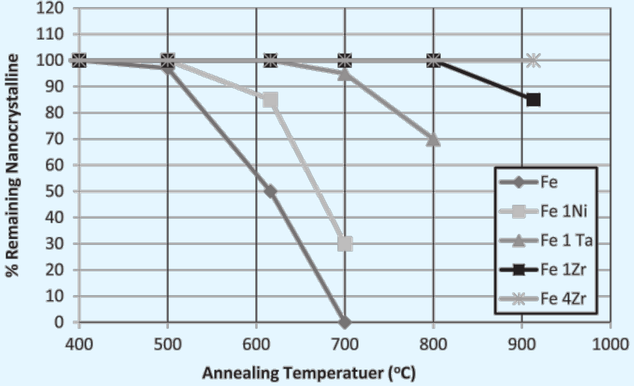
Pure Cu   Cu-Zr (d = 45nm)

$\varepsilon_{\text{failure}} = 4.4 \pm 0.5\%$ $\varepsilon_{\text{failure}} = 56 \pm 2.5\%$



Khalajhedayati *et al.*, Nature Comm. (2016)

NC alloy design

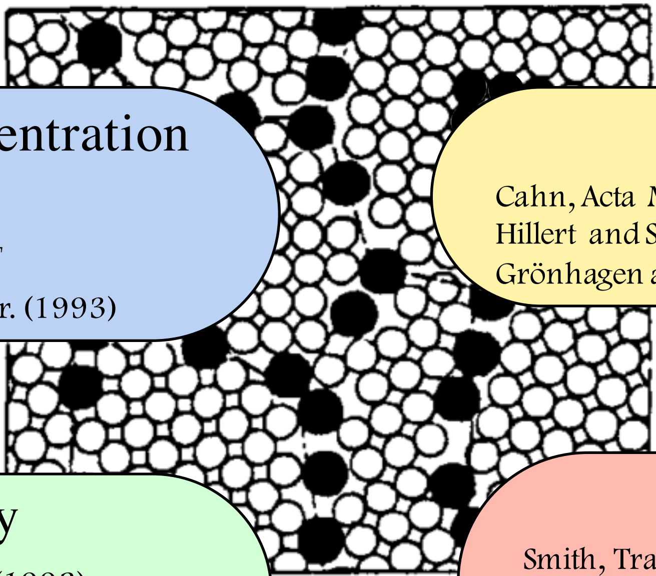


Darling *et al.*, Mat. Sci. Eng. A (2011)

Key Questions

- Predict role of GB solute segregation on **microstructural evolution**
- **Detangle** thermodynamic aspects from kinetic ones
- Use the framework in the **design** of NC metallic alloys

Phase field framework



GB energy vs. concentration

$$\Gamma = -\left(\frac{\partial \gamma_{gb}}{\partial \mu}\right)_{P,T}$$

Weissmüller, Nanostruct. Mater. (1993)

Solute drag

Cahn, Acta Metall. (1962)
Hillert and Sundman, Acta Metall. (1976)
Grönhagen and Ågren, Acta Mater. (2007)

GB anisotropy

Udler and D. Seidman, Phys. Stat. Sol. (1992)
Seki *et al.*, Acta Metall. Mater. (1991)
Hashimoto *et al.*, Acta Metall. (1984)

Zener pinning

Smith, Trans. AIME **175** (1948)
Hellman and M. Hillert, Scand. J. Met. (1975)
Chen *et al.*, J. Am. Ceram. Soc. (1998)
Moelens *et al.*, Acta Mater. (2005)

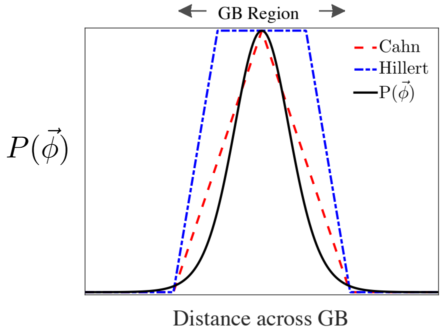
Phase Field Formalism

- Free energy
  - c(**r**,t): alloy concentration
  - $\boldsymbol{\phi}_i(\mathbf{r},t)$, {i = 1, …, $n_{\boldsymbol{\phi}}$}: grain order parameters

$$\mathcal{F}_{tot}[c,T] = \int d\mathbf{r} \left[\underset{\text{Local energy density}}{f_{loc}(c,T)} + \underbrace{\sum_i^{n_\phi} \frac{\epsilon^2}{2} |\nabla \phi_i|^2 + \frac{\kappa^2}{2} |\nabla c|^2}_{\text{Gradient terms for GBs and phase boundaries}} \right]$$

$$f_{loc}(c,T) = P(\vec{\phi}) f_{gb}(c,T) + \left[1 - P(\vec{\phi})\right] f_b(c,T)$$

$P(\vec{\phi})$: interpolates free energy between bulk and GB



- Dynamics

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

Mass conservation

Cahn-Hilliard Eq.

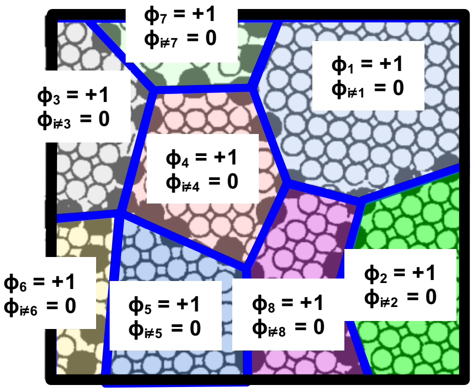
$$\frac{\partial \phi_i}{\partial t} = -L_i \left(\frac{\delta \mathcal{F}_{tot}}{\delta \phi_i} \right)$$

Gradient flow

Allen-Cahn Eq.

● **Solute**

○ **Host**

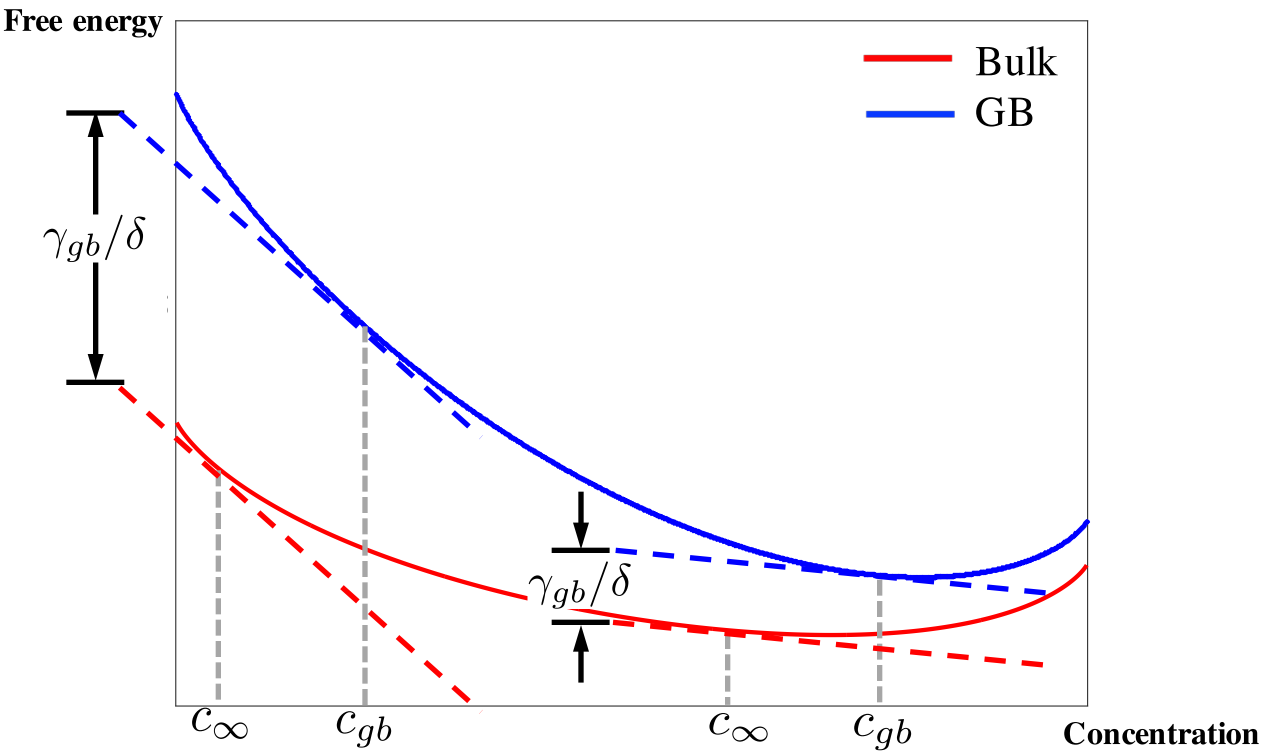


Abdeljawad and Foiles, Acta Mater. (2015)

Free Energies: Regular Solution

- Parallel tangent construction

$$f_i^{mix}(c,T) = \mathbf{G_i^B} c + \mathbf{G_i^A}(1-c) + \frac{RT}{V_m}\Big[c\ln c + (1-c)\ln(1-c)\Big] + \mathbf{\Omega_i} c(1-c), \quad i = gb, bulk$$



A: Solvent (matrix phase)

B: Solute atoms

Bulk: G_b^A, G_b^B, Ω_b

GB: $G_{gb}^A, G_{gb}^B, \Omega_{gb}$

c_∞ : Alloy concentration

δ : GB width

M. Hillert (2007)

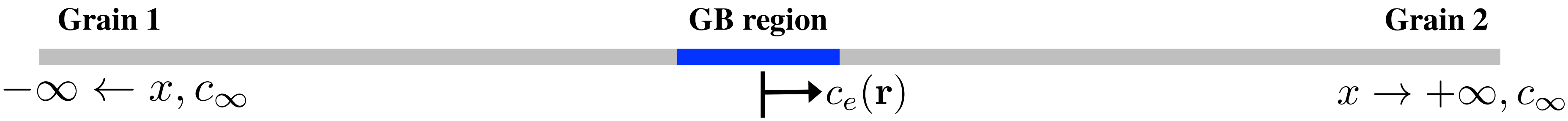
P. Lejcek (2010)

Equilibrium Properties

- Regular solution model:

Bulk: G_b^A, G_b^B, Ω_b GB: $G_{gb}^A, G_{gb}^B, \Omega_{gb}$

$$W = (G_{gb}^A - G_b^A),$$
$$\xi_1 = (G_{gb}^A - G_b^A) - (G_{gb}^B - G_b^B) - (\Omega_{gb} - \Omega_b),$$
$$\xi_2 = (\Omega_{gb} - \Omega_b)$$



Grain 1

GB region

Grain 2

$-\infty \leftarrow x, c_\infty$

$\mapsto c_e(\mathbf{r})$

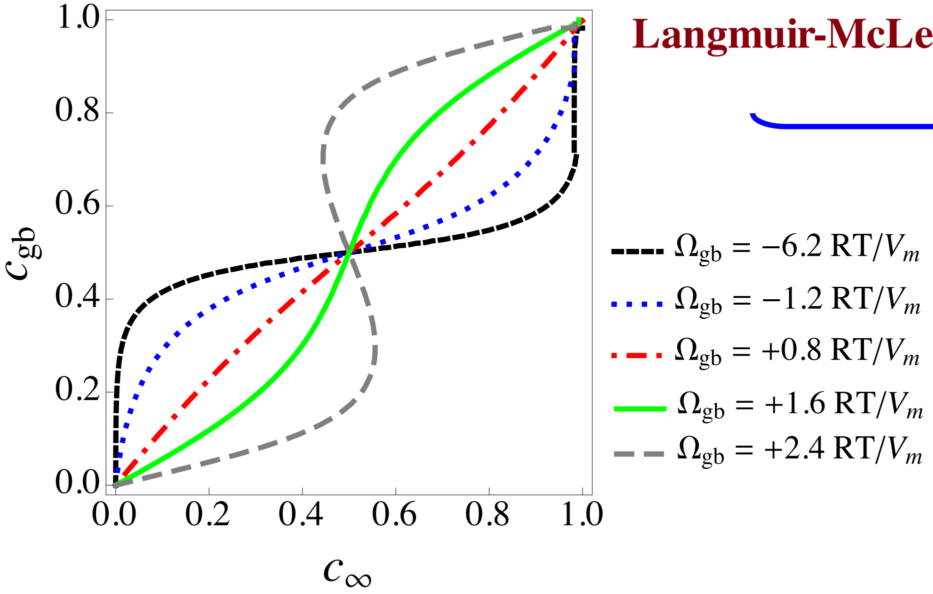
$x \rightarrow +\infty, c_\infty$

- Segregation isotherm

$$\frac{c_e(\mathbf{r})}{1 - c_e(\mathbf{r})} = \frac{c_\infty}{1 - c_\infty} \exp\left[\frac{\underbrace{16\xi_1\phi_e^2(1-\phi_e)^2}_{\text{Langmuir-McLean isotherm*}} + 32\xi_2\phi_e^2(1-\phi_e)^2 c_e + 2\Omega^{bulk}(c_e - c_\infty)}{(RT/V_m)}\right]$$

Langmuir-McLean isotherm*

Fowler-Guggenheim isotherm**

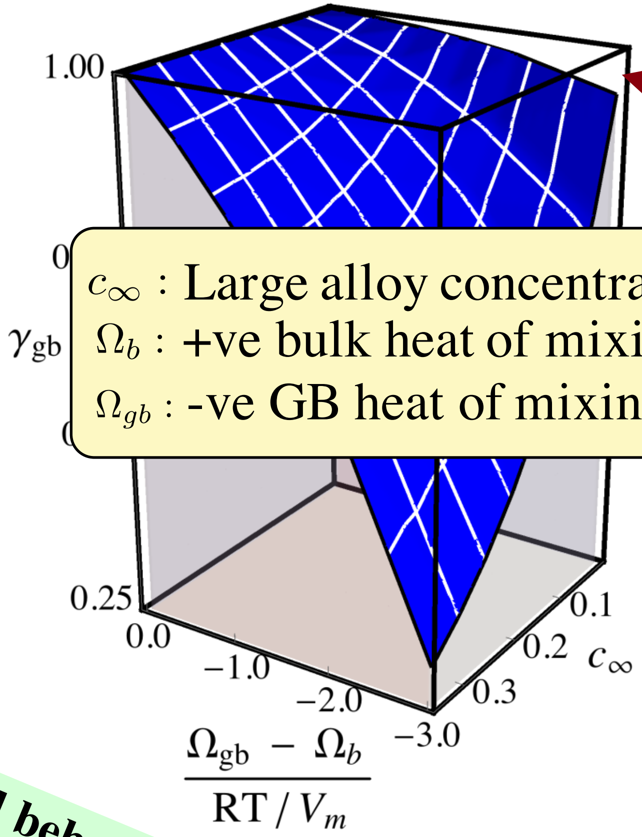


* McLean (1957)

** Fowler and Guggenheim(1939)

GB Energy

- Gibbs adsorption eq.: $\gamma = \Delta F - \Gamma \frac{\partial f_{mix}}{\partial c}\Big|_{eq}$



c_∞ : Large alloy concentration
Ω_b : +ve bulk heat of mixing
Ω_{gb} : -ve GB heat of mixing

Dilute limit

c_∞ : Alloy concentration
Ω_b : Bulk heat of mixing
Ω_{gb} : GB heat of mixing

Non-ideal behavior

Concentration away from dilute limit

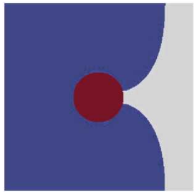
Considerable reduction in GB energy

Abdeljawad *et al.*, Acta Mater. (2017)

Competing Processes

- Grain growth, GB segregation and phase separation
  - ○ Large concentration needed for stability ➔ inside miscibility gap
  - ○ Solute-rich precipitated phase ➔ Zener pinning particles

Zener pinning: GB drag by second phase particles



Computational approach

○ **Fix bulk thermodynamics**

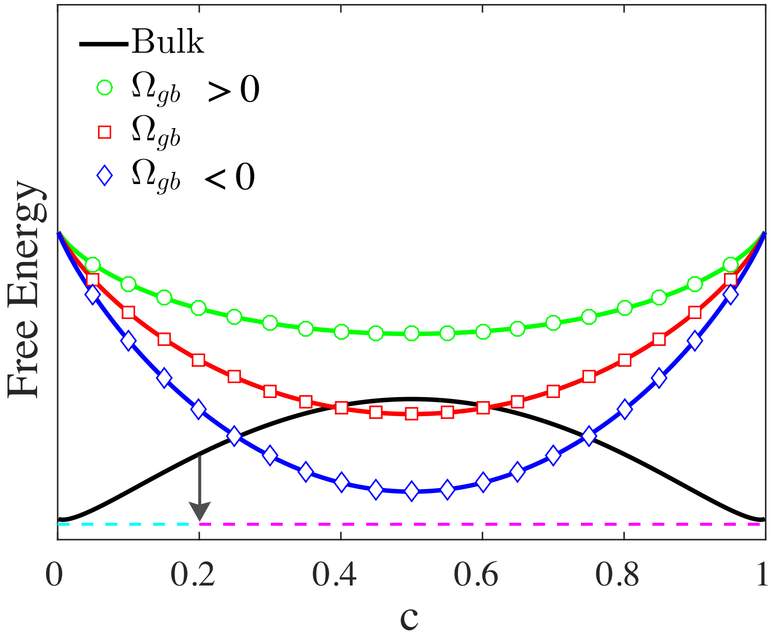
- Non-convex free energy
- Concentration within spinodal line

○ **Vary GB heat of mixing** Ω_{gb}

c_∞ : Large alloy concentration

Ω_b : +ve bulk heat of mixing

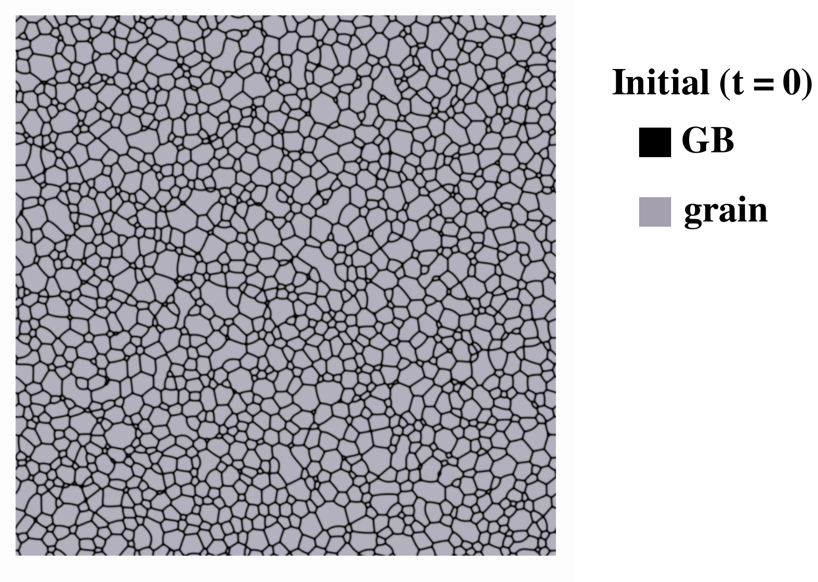
Ω_{gb} : -ve GB heat of mixing



GB free energies

Competing Processes

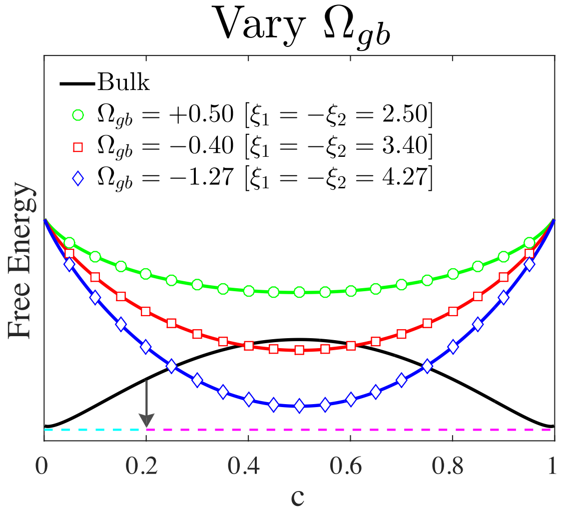
- Grain growth, GB segregation and phase separation
  - Initially (t=0): **1560** grains
  - $c(\mathbf{r}, t=0) = \langle c \rangle = 0.2$ (unstable alloy)
  - Vary GB heat of mixing: Ω_{gb}



- Metrics
  - Average grain dimeter $\langle D \rangle$
  - Define solute partitioning factor

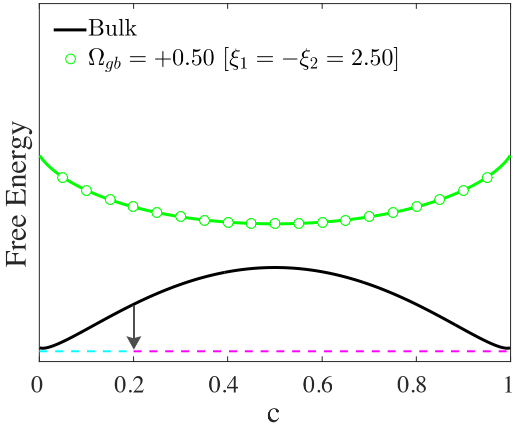
$$\Upsilon(t) = \frac{\int d\mathbf{r}\ c(\mathbf{r},t) P(\vec{\phi})}{\int d\mathbf{r}\ c(\mathbf{r},t)}$$

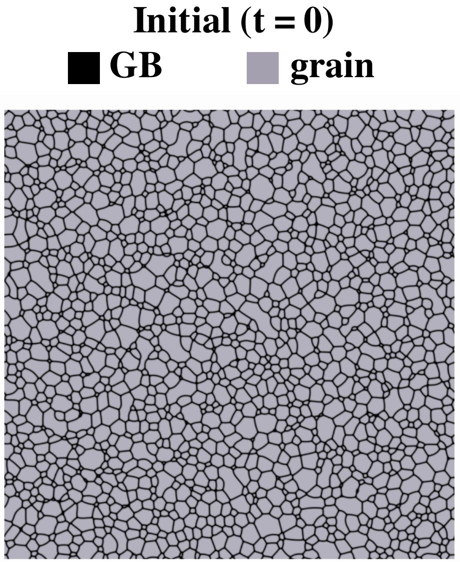
$$\Upsilon = \begin{cases} 0, & \text{No solute at GBs} \\ 1, & \text{GBs saturated with solute} \end{cases}$$


Vary Ω_{gb}

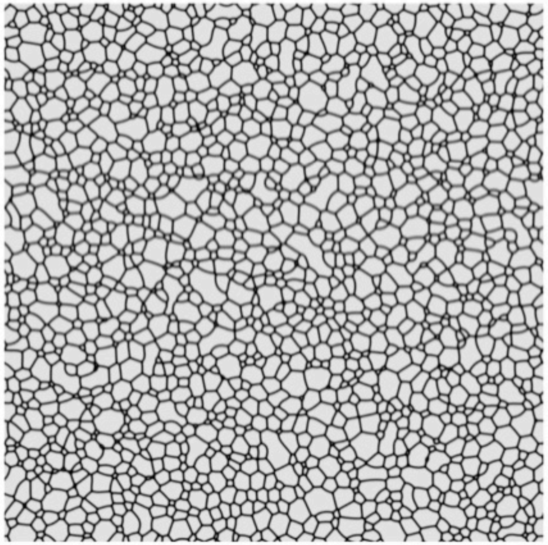
Competing Processes

- Polycrystalline systems
  - Initially (t=0): **1560** grains
  - $c(\mathbf{r}, t = 0) = \langle c \rangle = 0.2$

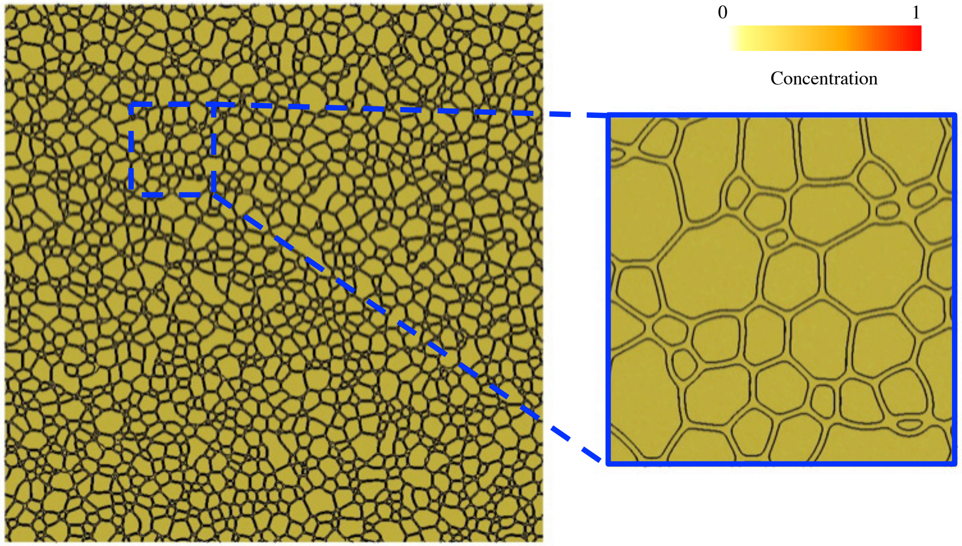




Normal grain growth
No segregation



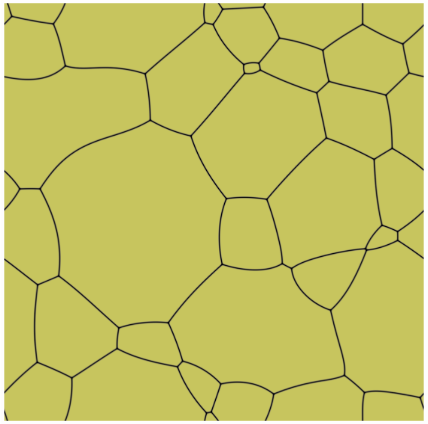
With GB segregation and
phase separation

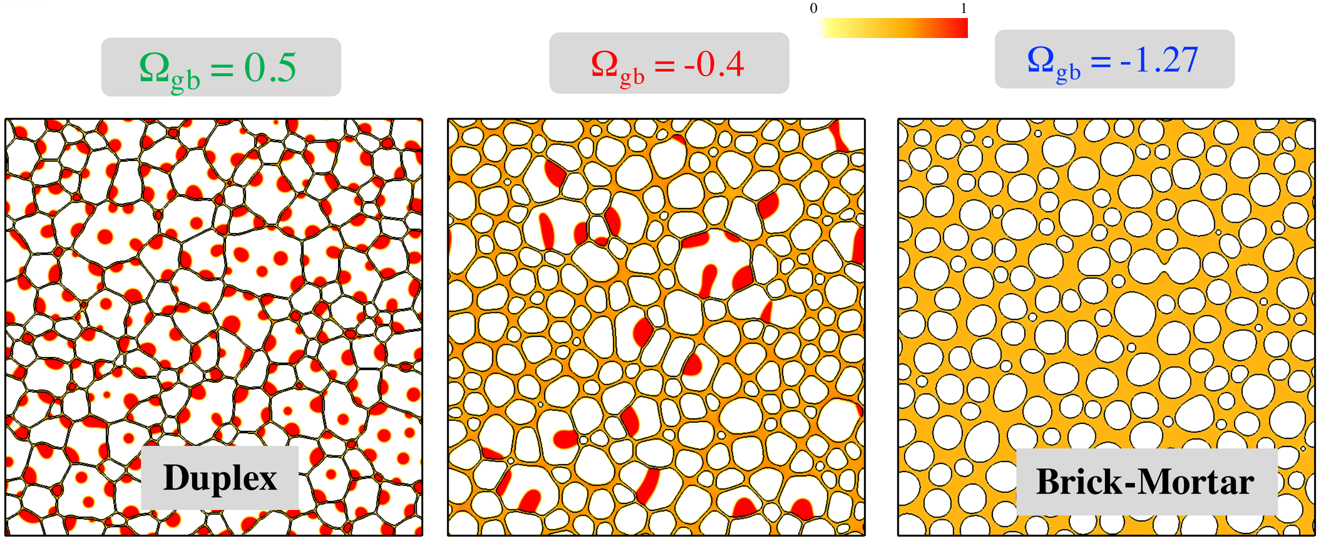


Abdeljawad *et al.*, Acta Mater. (2017)

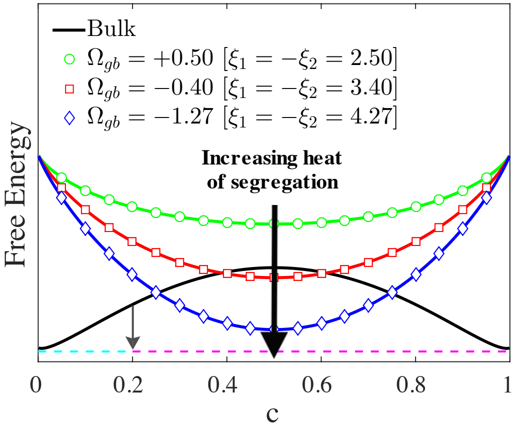
Competing Processes

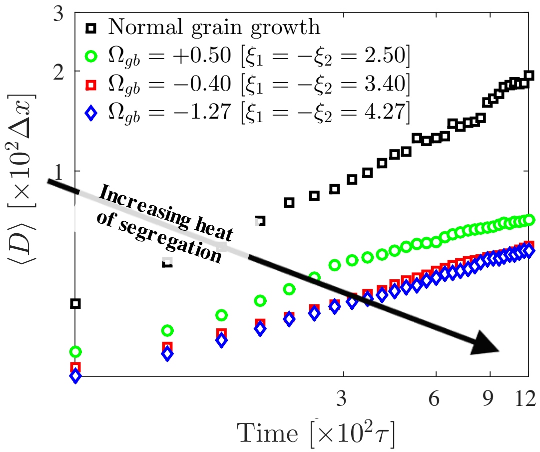
Normal grain growth
(No segregation)



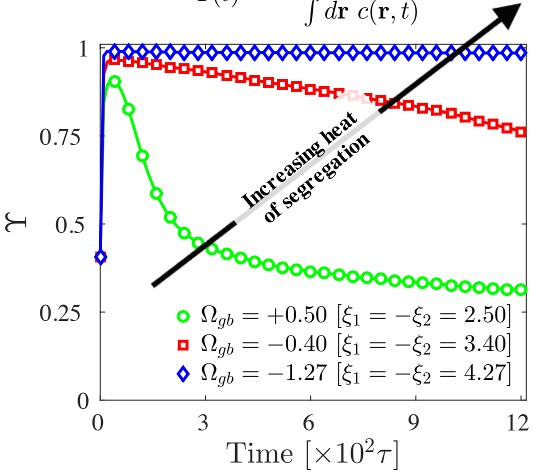


Increasing heat of segregation





$$\Upsilon(t) = \frac{\int d\mathbf{r}\ c(\mathbf{r},t) P(\vec{\phi})}{\int d\mathbf{r}\ c(\mathbf{r},t)}$$



Fix Ω_{gb} and Vary G^B_{gb} (Tilting)

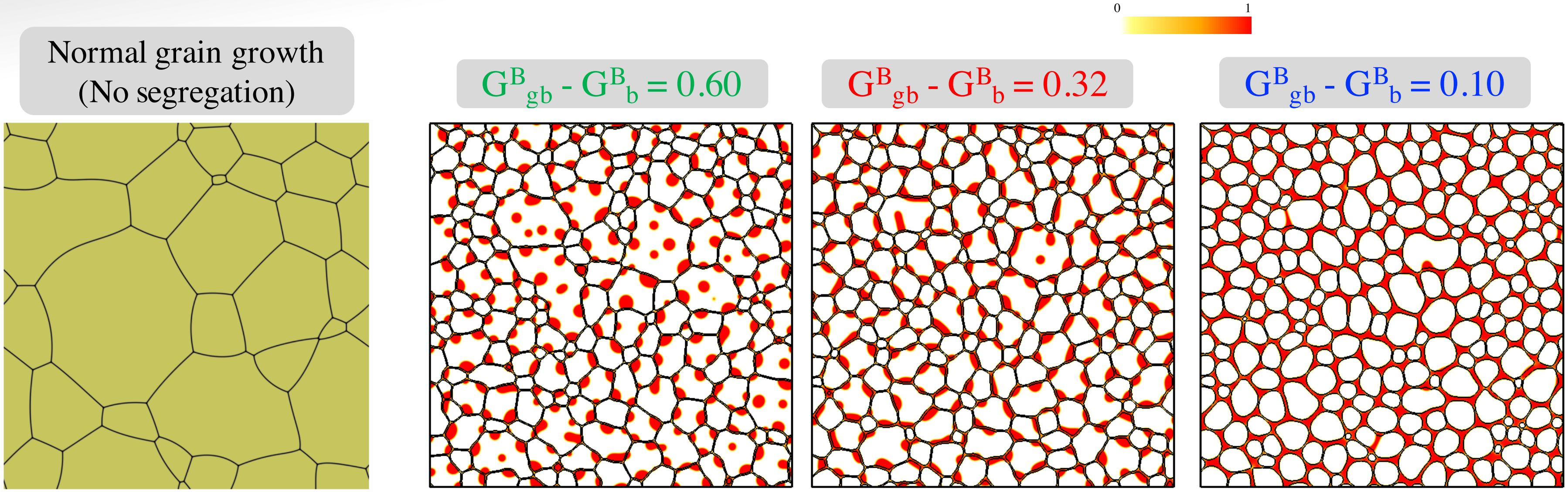
Sandia National Laboratories

Normal grain growth
(No segregation)

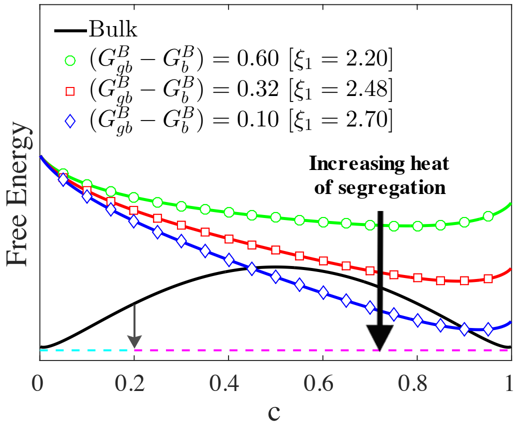
$G^B_{gb} - G^B_b = 0.60$

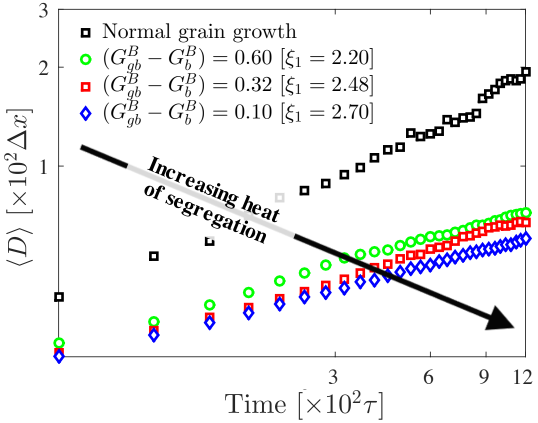
$G^B_{gb} - G^B_b = 0.32$

$G^B_{gb} - G^B_b = 0.10$

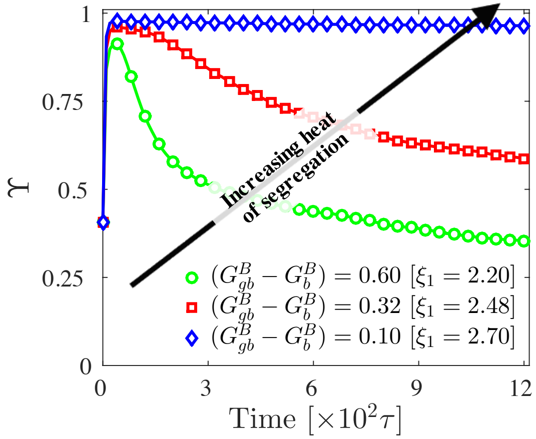


Increasing heat of segregation





$$\Upsilon(t) = \frac{\int d\mathbf{r}\ c(\mathbf{r},t) P(\vec{\phi})}{\int d\mathbf{r}\ c(\mathbf{r},t)}$$

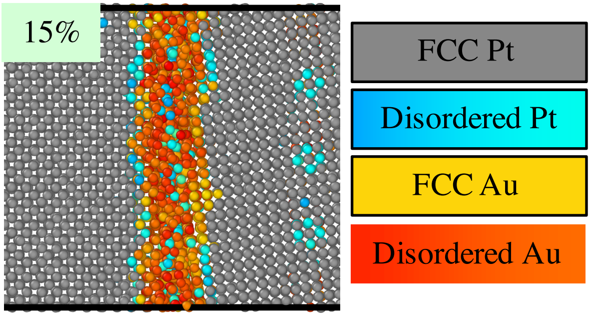


Observations

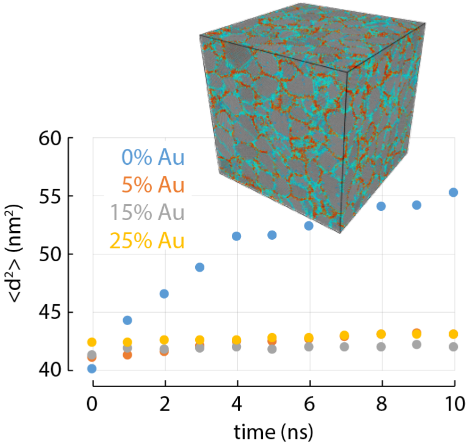
Atomistics: Pt-Au

- $\Sigma 5$ GB
- Au segregates to Pt GBs

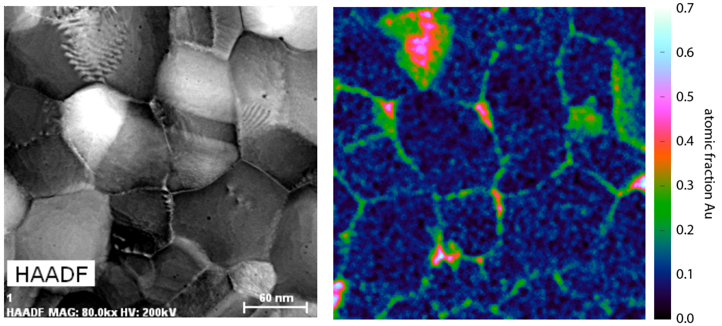
- Atomistic Monte Carlo
- T ~ 500K and $c_\infty \sim 0.15$



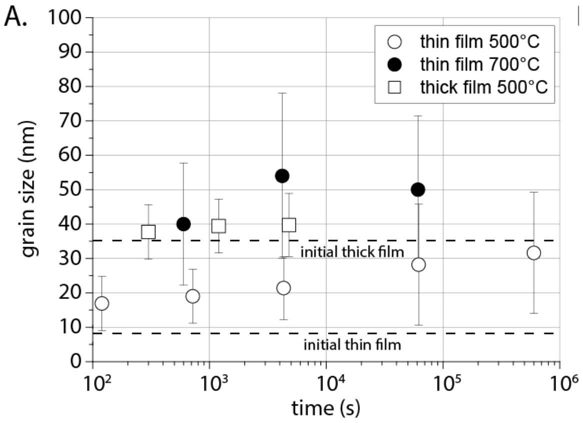
Foiles *et al.* (Manuscript in preparation)



Experimental: Pt-Au

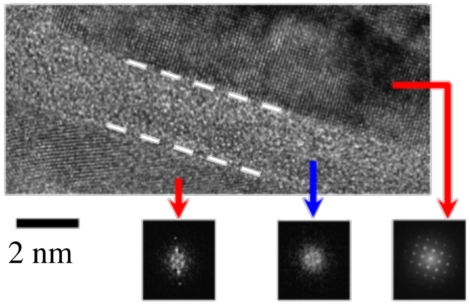


Abdeljawad *et al.*, Acta Mater. (2017)



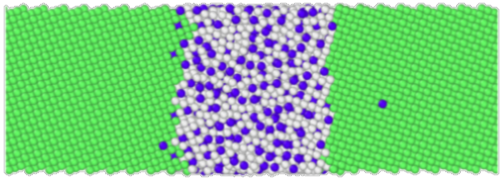
Argibay *et al.* (Manuscript in preparation)

Other immiscible alloys: Cu-Zr



Khalajhedayati *et al.*, Nature Comm. (2016)

$\Sigma 5$ (013)



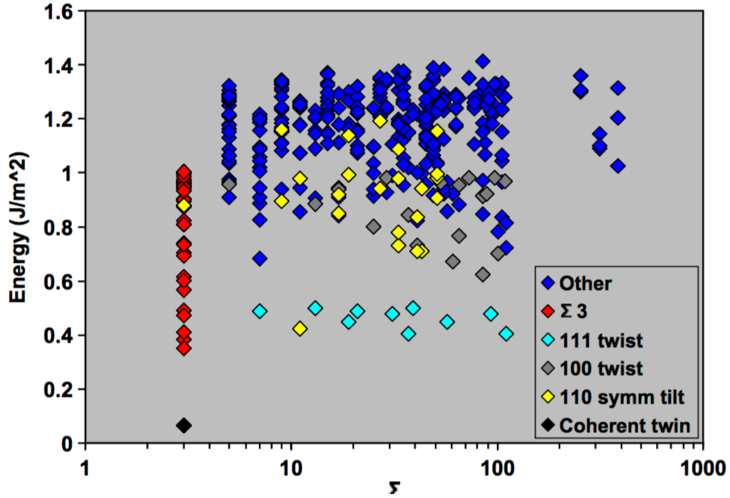
Pan *et al.*, Phys. Rev. B (2016)

What’s Next?

- Interface anisotropy

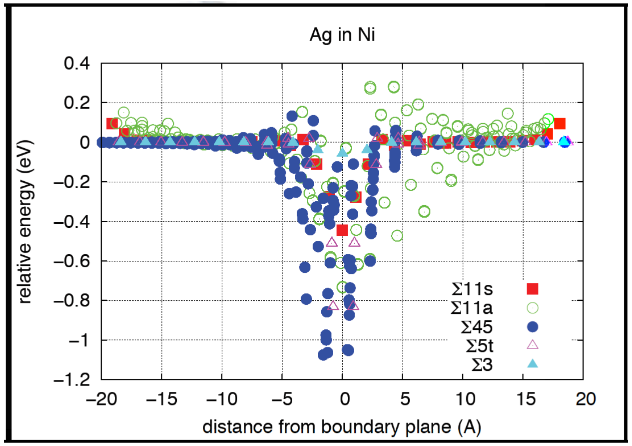
$$\gamma_{gb} = \gamma_o + f(\{\theta_1, \theta_2\}, T, \mu_i, \{\phi_1, \Phi, \phi_2\})$$

Anisotropy in GB energy



Olmsted *et al.*, Acta Mater. (2009)

Solute segregation anisotropy



B. Uberuaga, E. Martinez *et al.*, LANL (2016)

Thank you

fabdelj@sandia.gov

Published work relevant to this presentation

Abdeljawad, Lu, Argibay, Clark, Boyce and Foiles, Acta Mater. **127** (2017)

Medlin, Hattar, Zimmerman, *Abdeljawad* *et al.*, Acta Mater. **124** (2017)

Abdeljawad, Medlin, Foiles, Hattar and Zimmerman, J. App. Phys. **119** (2016)

Lim, *Abdeljawad*, Owen *et al.*, MSMSE **24** (2016)

Abdeljawad and Foiles, Acta Mater. **101** (2015)

Bufford, *Abdeljawad* and Hattar, App. Phys. Lett. **107** (2015)

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1) Department of Energy (DOE) Basic Energy Sciences (BES)
2) Sandia National Laboratories, Laboratory Directed Research and Development program (LDRD)
3) Sandia National Laboratories, Advanced Scientific Computing (ASC)

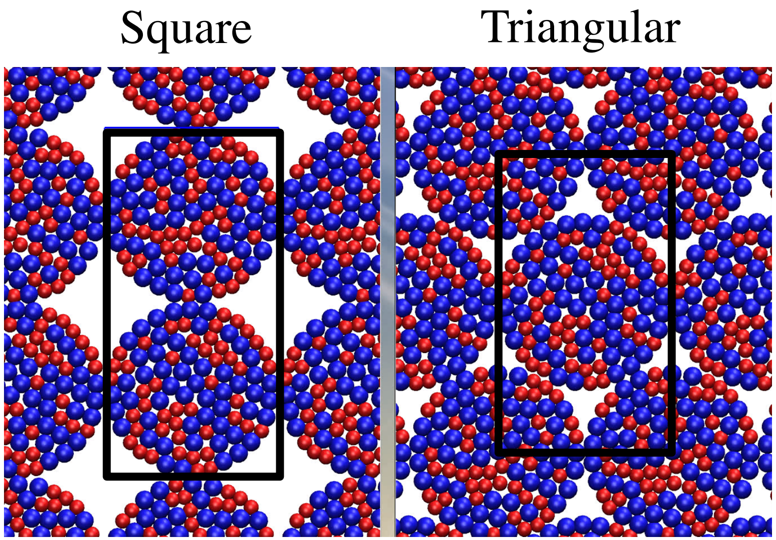
Collaboration:

1) Sandia National Laboratories: S. M. Foiles, N. Argibay, D. Medlin, H. Lim, B. L. Boyce, P. Lu, D. Bufford and K. Hattar
2) Los Alamos National Laboratory (LANL): B. Uberuaga and E. Martinez
3) Jason Trelewicz (Stony Brook Univ.), Heather Murdoch (Army Research Lab), Garritt Tucker (Drexel Univ.)

Backup Slides

Inter- vs. intra-filament porosity

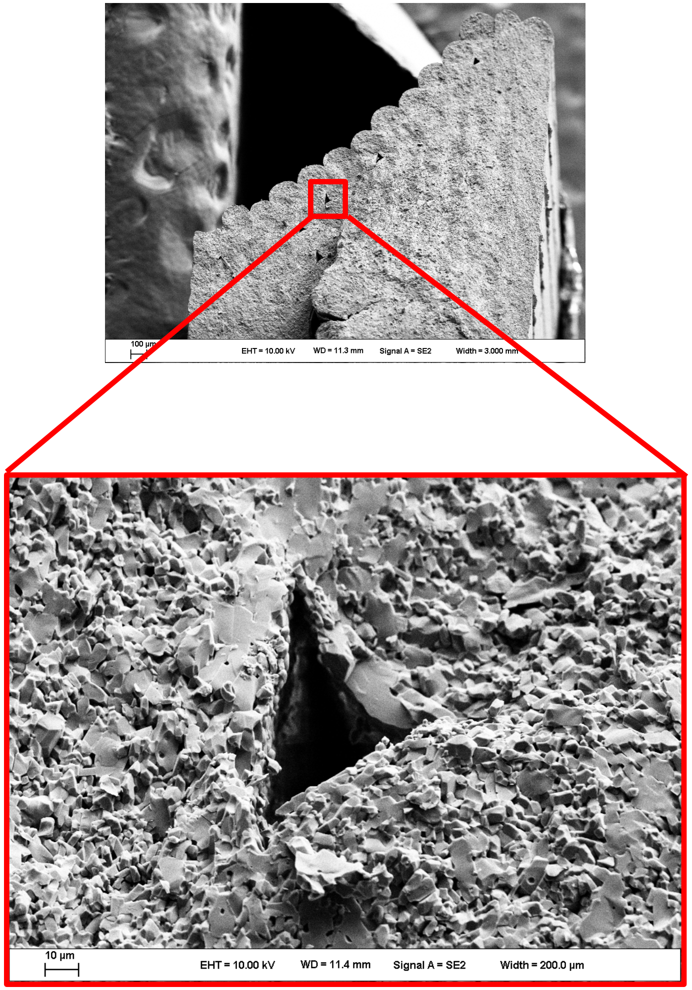
Filament arrangement



Square

Triangular

Bimodal distribution of pores



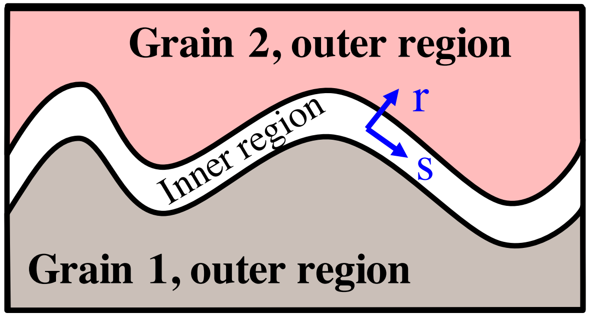
Adam Cook, Sandia National Labs (2017)

Relation to Solute Drag

- Sharp interface asymptotics (underway)

$$\mathcal{F}_{tot} = \int d\mathbf{r} \left[W f_{loc}(c, \phi, T) + \frac{\epsilon^2}{2} |\nabla \phi|^2 \right]$$

$$\frac{\partial \phi}{\partial t} = -L \left(\frac{\delta \mathcal{F}_{tot}}{\partial \phi} \right)$$



$$\phi = \phi_0 + \omega \phi_1 + \omega^2 \phi_2 + \cdots$$

$$c = c_0 + \omega c_1 + \omega^2 c_2 + \cdots$$

Provatas and Elder (2009)

Karma and Rappel, Phys. Rev. E **57** (1998)

Normal grain growth

$$\mathcal{V}_n = M_{gb} \gamma_{gb} \mathcal{K} = L \epsilon^2 \mathcal{K}$$

With GB solute segregation

$$\mathcal{V}_n = \frac{L\epsilon^2}{1 + \left(\frac{LW}{D}\right) \frac{K_I}{K_{II}}} \mathcal{K} \simeq L\epsilon^2 \left(1 - \frac{LW}{D} \frac{K_I}{K_{II}} \right) \mathcal{K}$$

$$K_I = \int_{-\infty}^{+\infty} \frac{\partial c_o}{\partial x} \int_0^x (c_o - c_\infty) \, dx' dx$$

$$K_{II} = \int_{-\infty}^{+\infty} \left(\frac{d\phi_0}{d\xi} \right)^2 d\xi$$

D: solute diffusivity

Young Man, in mathematics you don't understand things. You just get used to them

J. Von Neumann

Abdeljawad *et al.*, In preparation (2017)

Relation to Solute Drag

- Phase field sharp interface asymptotics (underway)

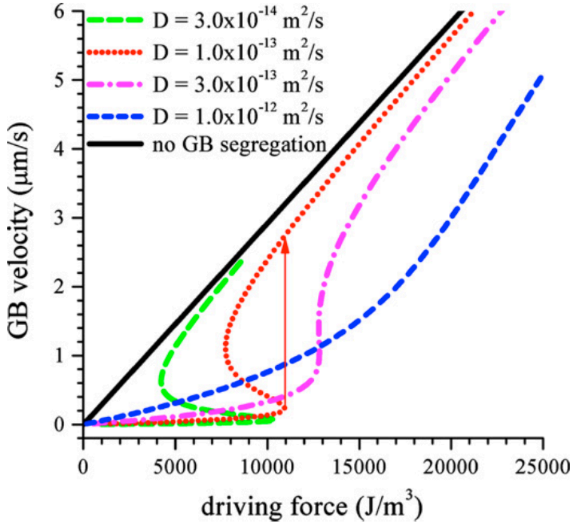
$$\mathcal{V}_n = \frac{L\epsilon^2}{1+\left(\frac{LW}{D}\right)\frac{K_I}{K_{II}}}\mathcal{K} \simeq L\epsilon^2\left(1-\frac{LW}{D}\frac{K_I}{K_{II}}\right)\mathcal{K}$$

- Solute drag model [ideal, dilute alloys]

$$\mathcal{V}_n = M_{gb}\Big(P - P^*(\mathcal{V}_n)\Big)$$

$P = \gamma_{gb}\mathcal{K}$: Curvature driven flow

$P^*(\mathcal{V}_n)$: Drag pressure



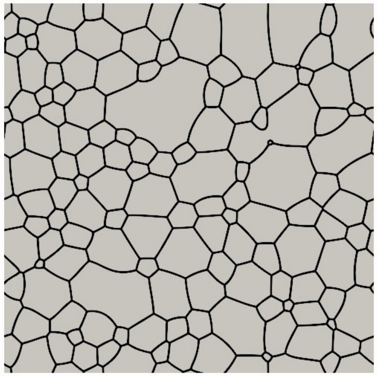
Kim and Park, Acta Mater. (2008)

Cahn, Acta Metall. (1962)
Hillert and Sundman, Acta Metall. (1976)
Gottstein and Shvindlerman (1999)

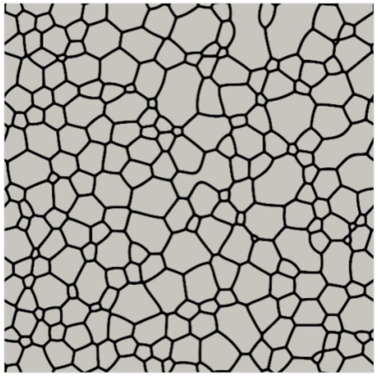
GB Widening?

- Fix free energy model parameters
- Vary the initial alloy concentration

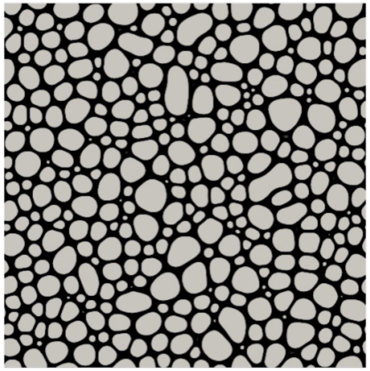
Normal grain growth



$\langle c \rangle = 0.2$



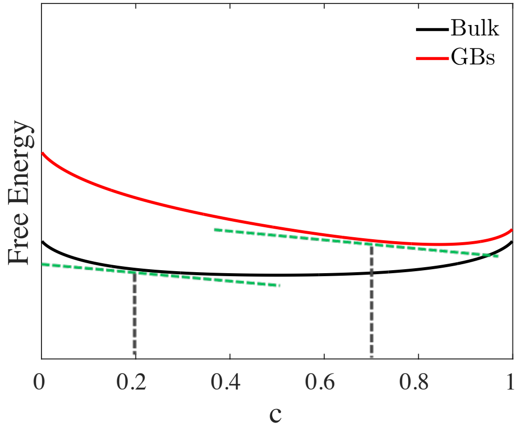
$\langle c \rangle = 0.35$



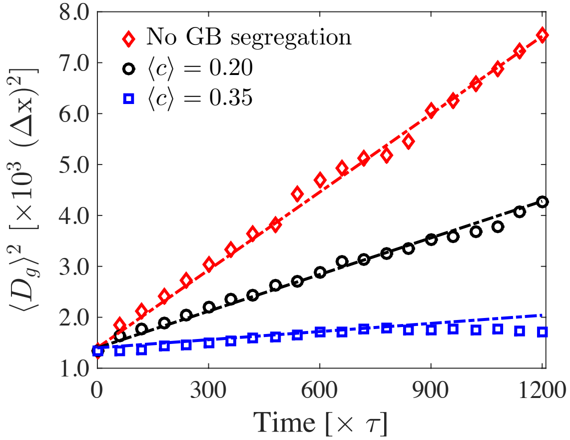
■ GB

▨ grain

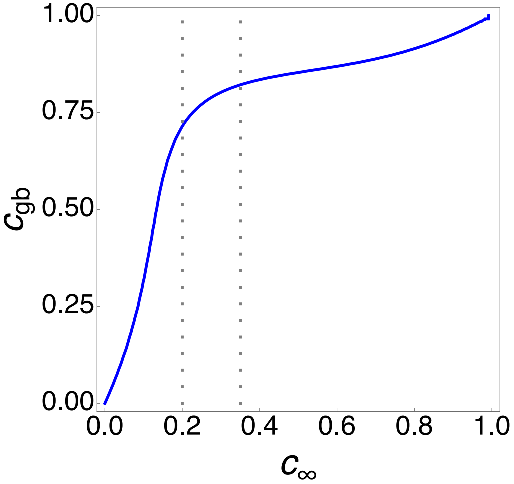
Free energies



Avg. grain area



GB segregation isotherm



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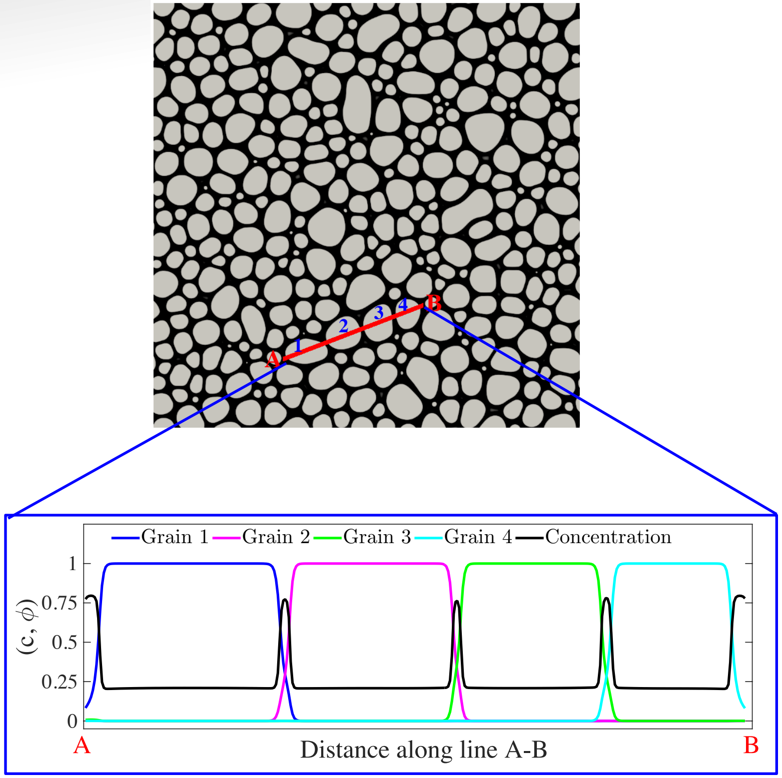
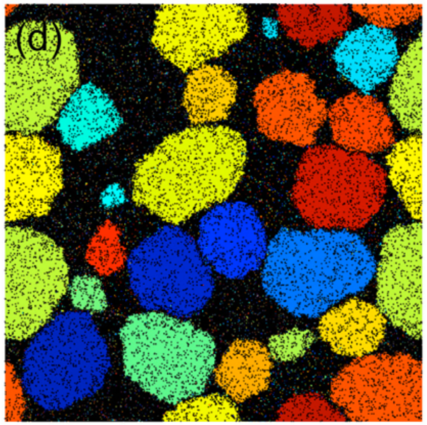
GB Widening?



$$\mathcal{F}_{tot} = \int d\mathbf{r} \left[W f_{loc}(c, \phi, T) + \frac{\epsilon^2}{2} |\nabla \phi|^2 \right]$$

- Concentration-dependent energy barrier: $W = W(c)$
- Control GB width: $\epsilon \overset{?}{=} \epsilon(c)$



Monte Carlo
alloy

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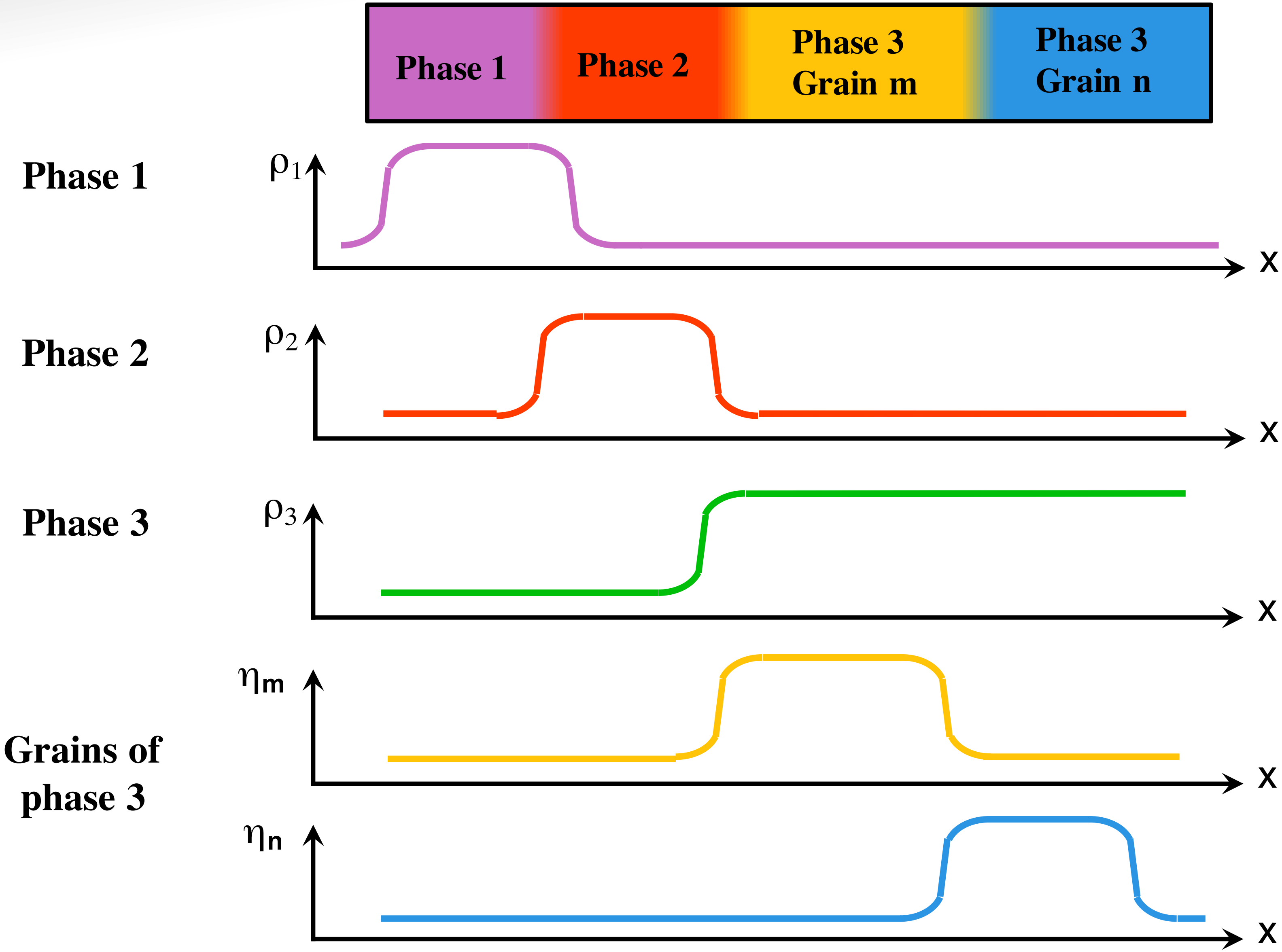
Energy: $\gamma_{gb} \sim \epsilon \sqrt{W}$

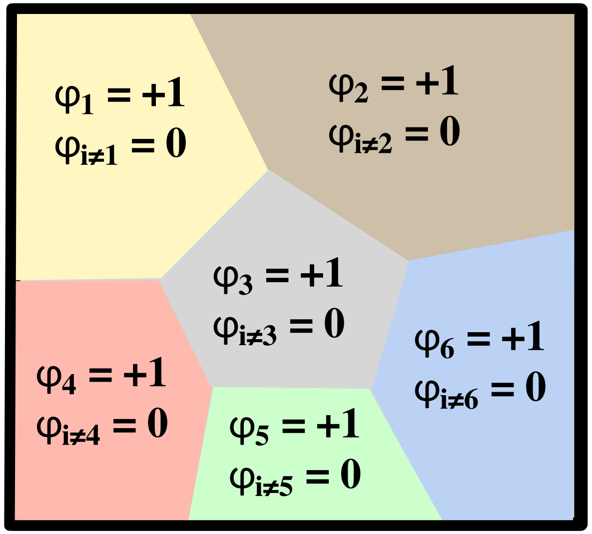
Width: $\delta_{gb} \sim \dfrac{\epsilon}{\sqrt{W}}$

Machinery: Phase Field



Machinery: Phase Field (cont.)

- Order parameters “phase fields”
  - Concentration (alloying elements)
  - Crystallographic orientation of grains (internal interfaces)
  - Mass density (solid, vapor, liquid)
  - Displacement fields

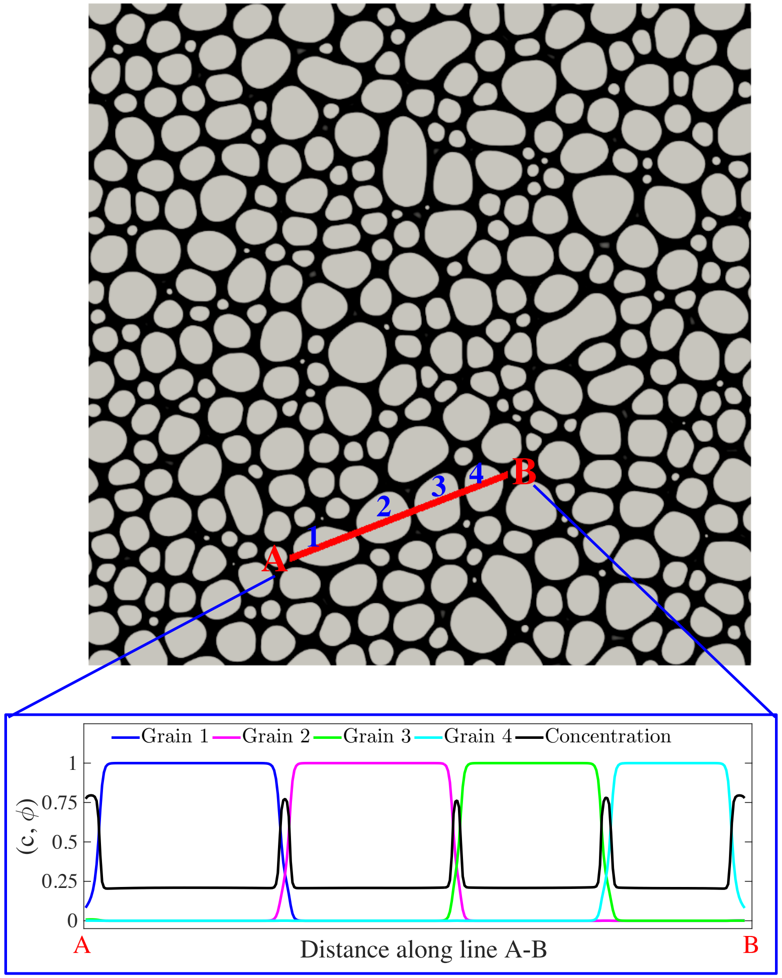


- Coarse grained free energy
  - Bulk thermodynamics
    - Phase transitions (solidification, martensitic, etc…)
    - Multicomponent
  - Interfacial energies and thermodynamics
    - Gibbs-Thomson boundary condition (Stefan problem)
    - Anisotropy

- Dynamics driven by minimization of energy

Brick-Mortar Microstructure

Our treatment



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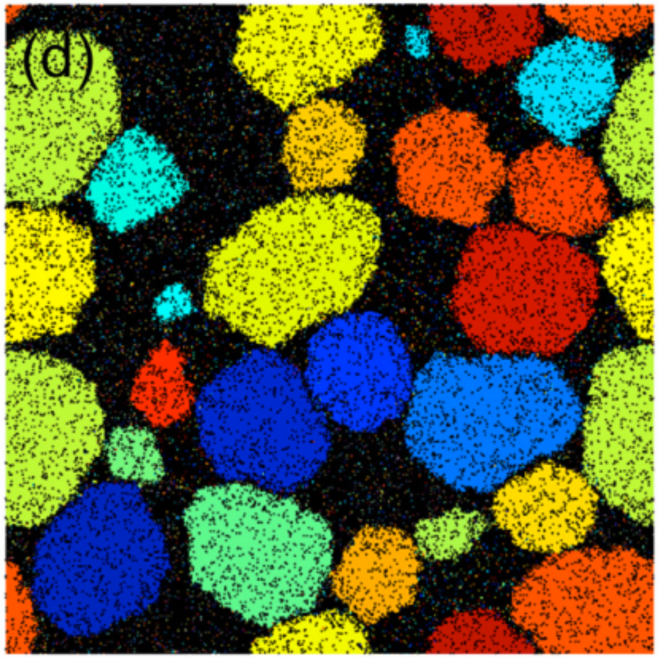
Monte Carlo Alloy



Chookajorn and Schuh, Acta Mater. 73 (2014)