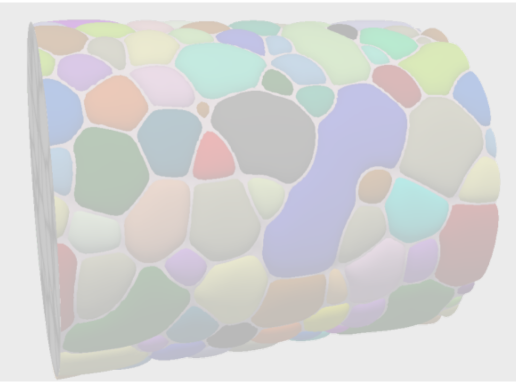
SAND2017-3807C

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

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



A Mesoscale Model of Grain Boundary Faceting: The Role of Facet Junctions

Fadi Abdeljawad, Douglas Medlin, Jonathan Zimmerman, Khalid Hattar, Stephen M. Foiles

Sandia National Laboratories

Sandia National Laboratories

Exceptional service in the national interest

U.S. DEPARTMENT OF ENERGY

National Nuclear Security Administration

U.S. DEPARTMENT OF ENERGY | Office of Science

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.

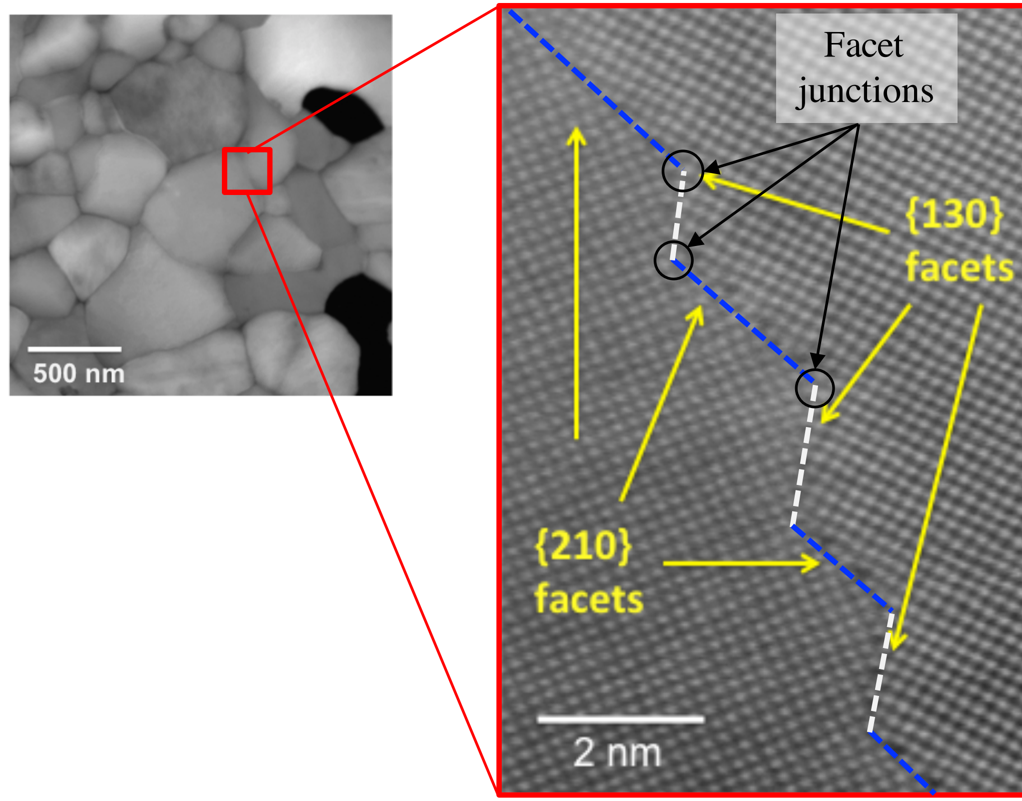
Outline

- **Introduction**
  - Observations: Interface faceting
- **Modeling framework**
  - Motivation
  - Analytical treatments: Vector thermodynamics, free body diagram
  - Modeling framework
- **Results**
  - Critical quench and spontaneous faceting
  - Facet junctions and interactions
- **Concluding remarks and future work**

Interface Faceting

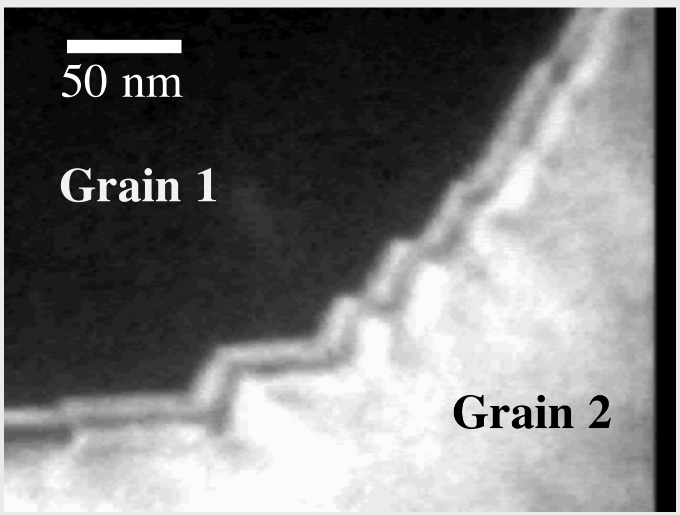
- Σ5 <001> tilt GB in BCC Fe
- GB breaks into {130}/{120} facets
- Thin film (36nm) annealed at 657°C

HAADF-STEM



D. Medlin, Sandia National Labs

Facet coarsening

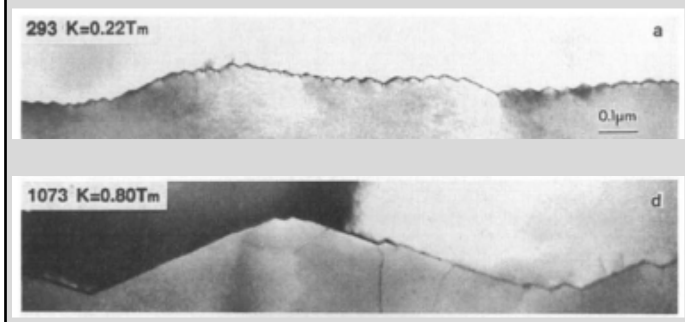


Medlin and Lucadamo (2000)

Σ3 GB in Au

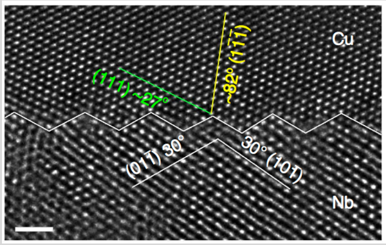
$\{11\bar{2}\}$ facets

Interface Faceting



Facet coarsening

Hsieh and Balluffi, Acta Metall. **37** (1989)



Faceting in phase boundaries

Zheng et al., Nature Comm. **7** (2013)

Key Questions

- Energetics associated with GB faceting
- Mesoscale model beyond vector thermodynamics: Facet coarsening
- Use the framework in models of microstructural evolution

Grain Boundaries (GBs)

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

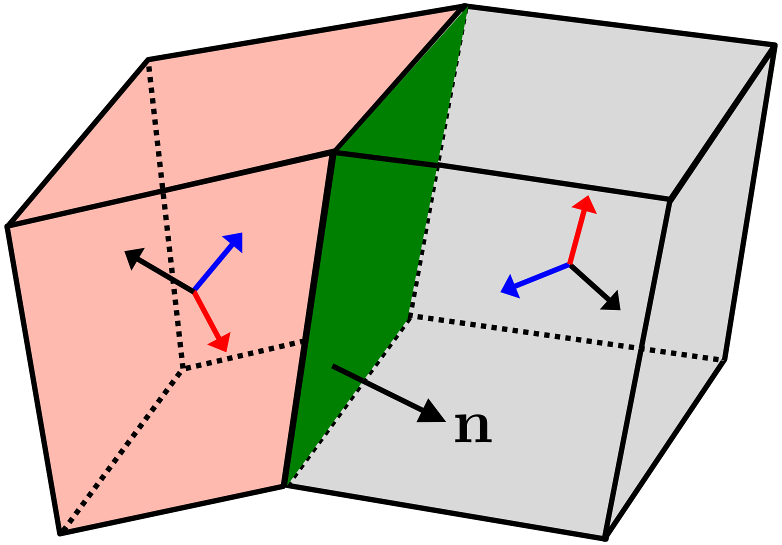
6+i dimensional space

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)

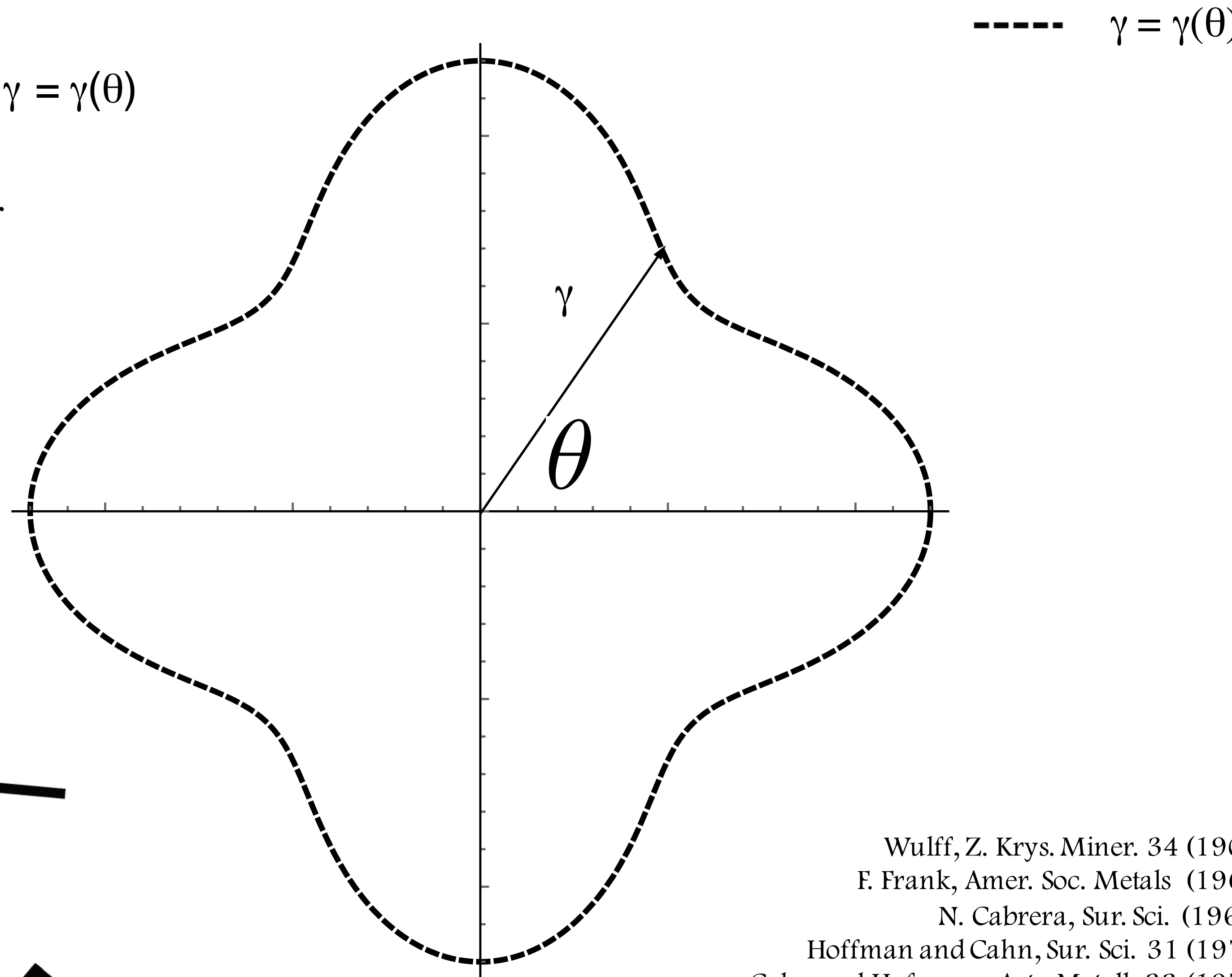
Faceting: Graphically

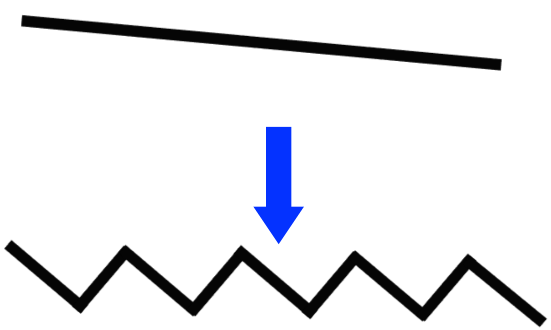
Sandia National Laboratories

- Wulff (polar) plot: $\gamma = \gamma(\theta)$
- Plot of $1/\gamma$
- ξ-capillarity vector

$$\xi = \gamma \mathbf{e_r} + \gamma_\theta \mathbf{e}_\theta$$

An ear in ξ-vector



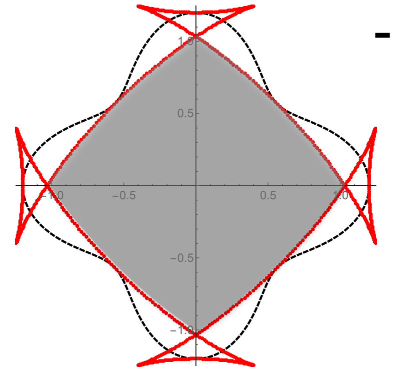


Wulff, Z. Krys. Miner. 34 (1901)
F. Frank, Amer. Soc. Metals (1963)
N. Cabrera, Sur. Sci. (1964)
Hoffman and Cahn, Sur. Sci. 31 (1972)
Cahn and Hofmann, Acta Metall. 22 (1974)
Herring, Phys. Rev. 82 (1951)

Faceting: Mathematically

- Interface anisotropy
  - ξ- vector: $\xi = \gamma \mathbf{e_r} + \gamma_\theta \mathbf{e}_\theta$

An ear in ξ-vector

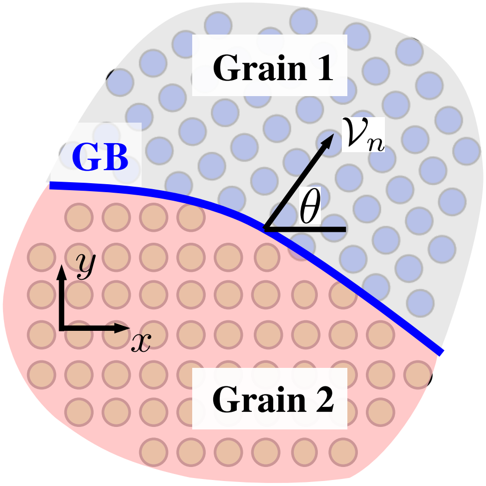


- Sharp interface law

$$\mathcal{V}_n = M_{gb}\left(\gamma_{gb} + \frac{\partial^2 \gamma_{gb}}{\partial \theta^2}\right)\mathcal{K}$$

Linearization: $$\frac{\partial y}{\partial t} = M_{gb}\underbrace{\left(\gamma_{gb} + \frac{\partial^2 \gamma_{gb}}{\partial \theta^2}\right)_o}_{\text{When } \gamma_{gb} + \partial^2\gamma_{gb}/\partial\theta^2 < 0}\frac{\partial^2 y}{\partial x^2}$$

Locally backward parabolic PDE

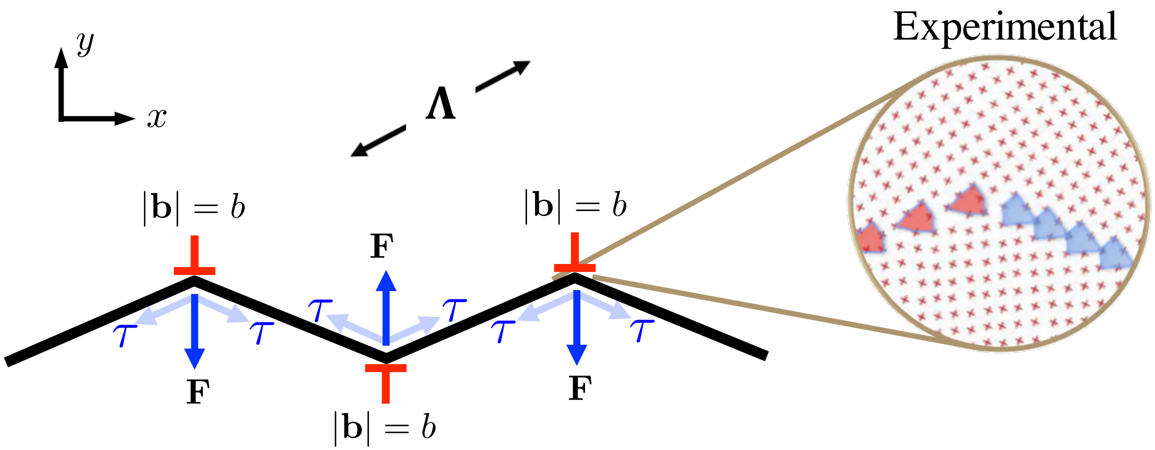


Thermodynamic instability of an initially planar surface with negative surface stiffness leads to faceting (hill and valley) structure

W. Mullins, Amer. Soc. Metals (1963)

Energetics of a Faceted GB

- Fix misorientation and vary inclination



Λ :Average facet length scale

:Dislocation with Burgers vector **b** due to mismatch in translation vectors of interfaces

Dimitrakopulos *et al.*, Inter. Sci. (1996)

$\mathcal{T}$:Interface stress. Discontinuity across junction leads to line forces

Sutton and Balluffi (1994)

- Total energy

$$\mathcal{F}_{tot} = f_{inter} + f_{local} + f_{nonlocal}$$

Anisotropic interface

Junction energy due to dislocation and interface stress

Non-local interactions between dislocations and point forces

$$\mathcal{F}_{tot} = \frac{a_1}{\Lambda} \ln \Lambda + \frac{a_2}{\Lambda}$$

Λ: facet length

a_1, a_2: material constants

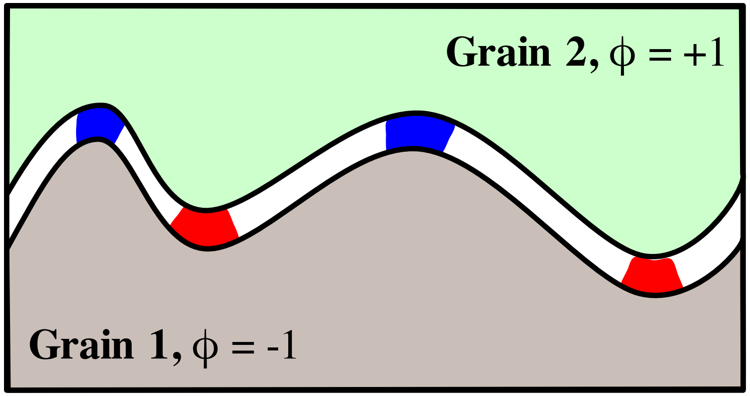
Hamilton *et al.*, Phys. Rev. Lett. (2003)

Phase Field Framework

- Structural order parameter
  - Mean curvature: $\mathcal{K} = \nabla \cdot \left(\frac{\nabla\phi}{|\nabla\phi|}\right) \sim \nabla^2\phi$
  - Higher order expansion:

$$\gamma = a_o(\theta) + a_2(\theta)\mathcal{K}^2 + a_4(\theta)\mathcal{K}^4 + \cdots$$

Liu and Metiu, Phys. Rev. B **48** (1993)



- Total energy

$$\mathcal{F}_{tot} = \int d\mathbf{r} \left[A_H f(\phi) + \frac{\epsilon^2(\theta)}{2}|\nabla\phi|^2 + \Gamma_o(\nabla^2\phi)^2 + \underbrace{\Gamma_1 \int_{|\mathbf{r}-\mathbf{r}'|>R_c} d\mathbf{r}' \frac{\nabla^2\phi(\mathbf{r})\ \nabla^2\phi(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}}_{\text{facet junction-junction interactions}} \right]$$

Anisotropic GB
(fit via atomistics)

Local facet junction energy
(Willmore regularization)

Wise *et al.*, App. Phys. Lett. (2005)

facet junction-junction interactions

Stewart and Goldenfeld, Phys. Rev. A (1992)

- Dynamics

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

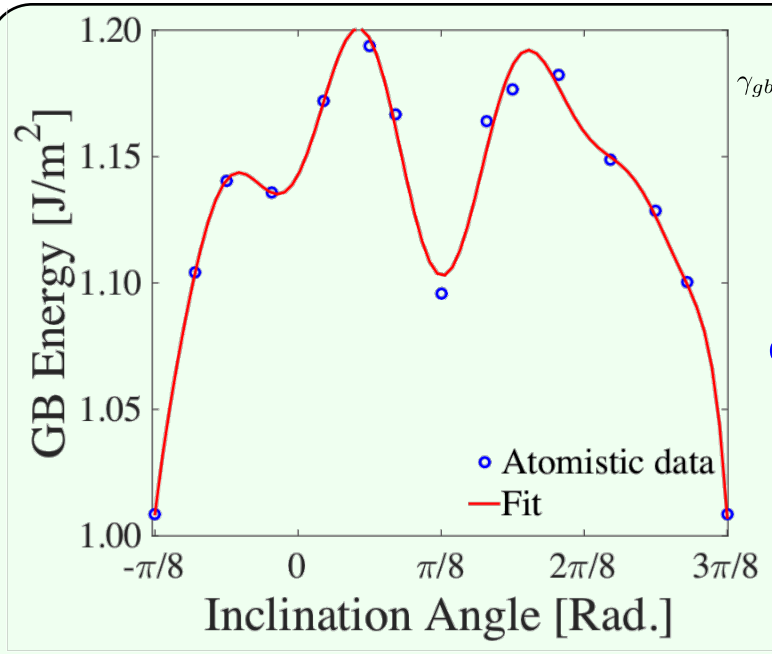
<u>Model parameters:</u> $\varepsilon(\theta)$, $\mathbf{\Gamma_o}$, $\mathbf{\Gamma_1}$

Abdeljawad *et al.*, J. App. Phys. **119** (2016)

Model Parameters: Atomistics

- The case for Σ5 <001> tilt GB in BCC Fe

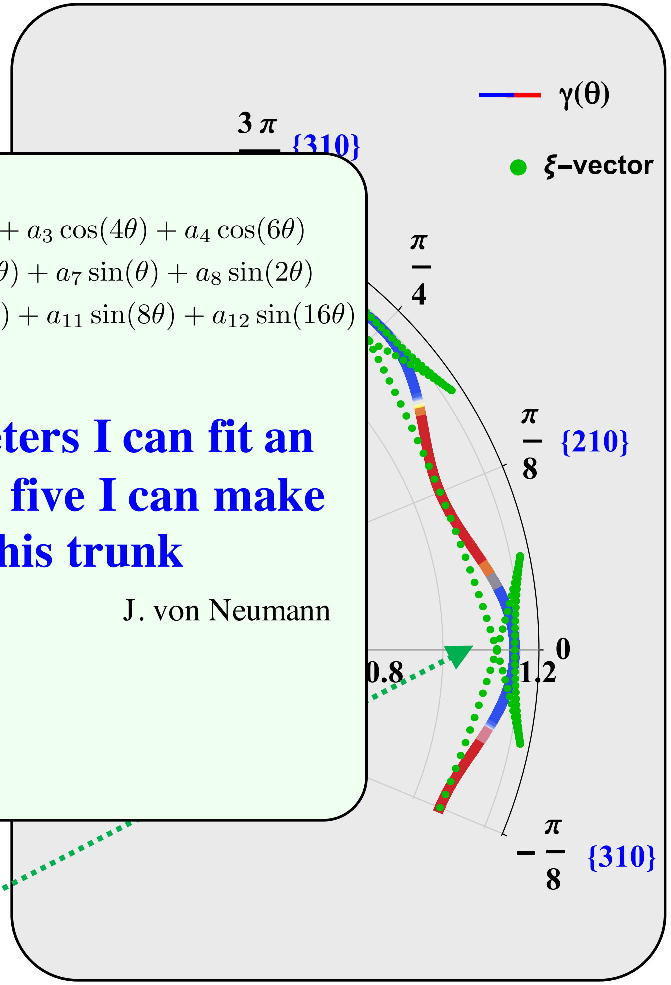
Model parameters: $\varepsilon(\theta)$, $\boldsymbol{\Gamma}_o$, $\boldsymbol{\Gamma}_1$



$$\gamma_{gb} = a_0 + a_1\cos(\theta) + a_2\cos(2\theta) + a_3\cos(4\theta) + a_4\cos(6\theta) + a_5\cos(8\theta) + a_6\cos(16\theta) + a_7\sin(\theta) + a_8\sin(2\theta) + a_9\sin(4\theta) + a_{10}\sin(6\theta) + a_{11}\sin(8\theta) + a_{12}\sin(16\theta)$$

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

J. von Neumann



Notice the presence of ears in **ξ**-vector

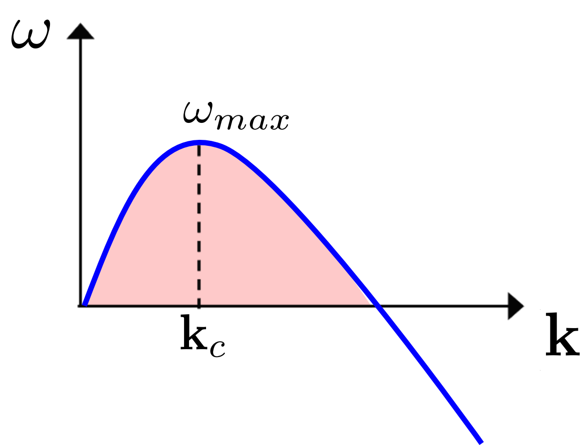
Linear Stability

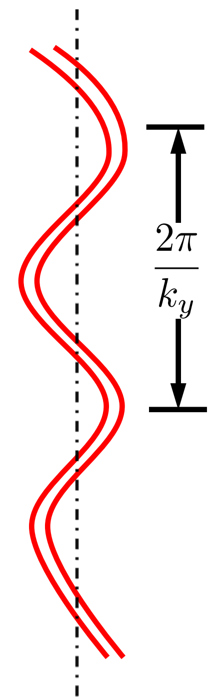
- Assume Fourier modes: $\phi = \hat{\delta}_\phi \exp\left[i\mathbf{k}\cdot\mathbf{x} + \omega(\mathbf{k})t\right]$
- Dispersion relation: $\omega(\mathbf{k}, \Gamma_o, \Gamma_1)$
- Max growth rate: $\omega_{max}(\mathbf{k}_c, \Gamma_o, \Gamma_1)$

<u>Model parameters:</u> $\varepsilon(\theta)$, $\mathbf{\Gamma}_o$, $\mathbf{\Gamma}_1$

Γ_o: Junction energy

Γ_1: Junction interactions





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

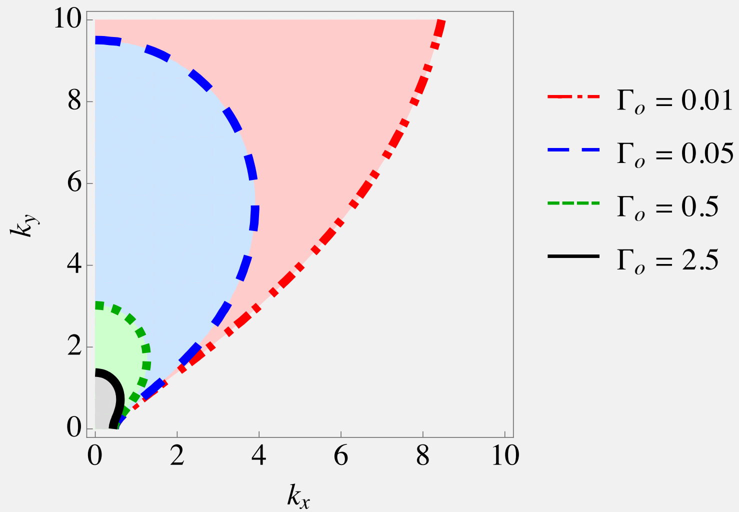
J. von Neumann

Linear Stability (cont.)

Shaded regions correspond to unstable perturbations

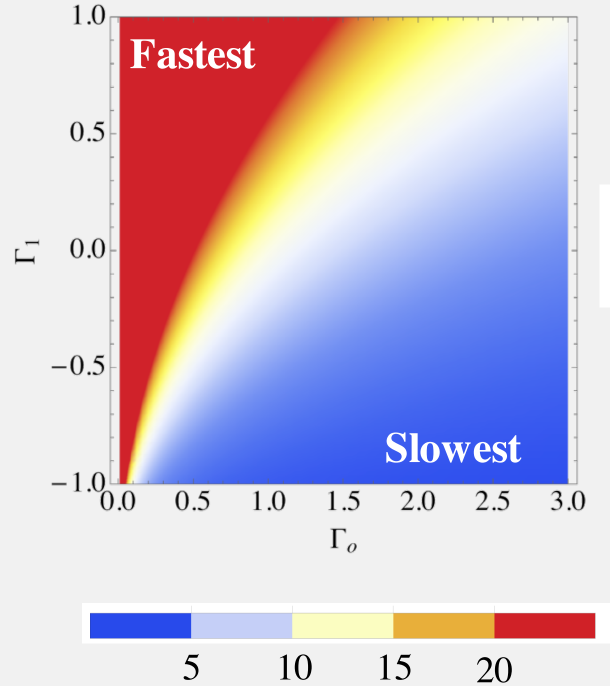
Γ_o: Junction energy
Γ_1: Junction interactions

Facet junction energy with no interactions ($\Gamma_1 = 0$)



- Regions of instability decrease in size as facet junction energy increases
- Faceted GBs with large spacing between junctions

Maximum growth rate $\omega_{max}(\mathbf{k}_c, \Gamma_o, \Gamma_1)$

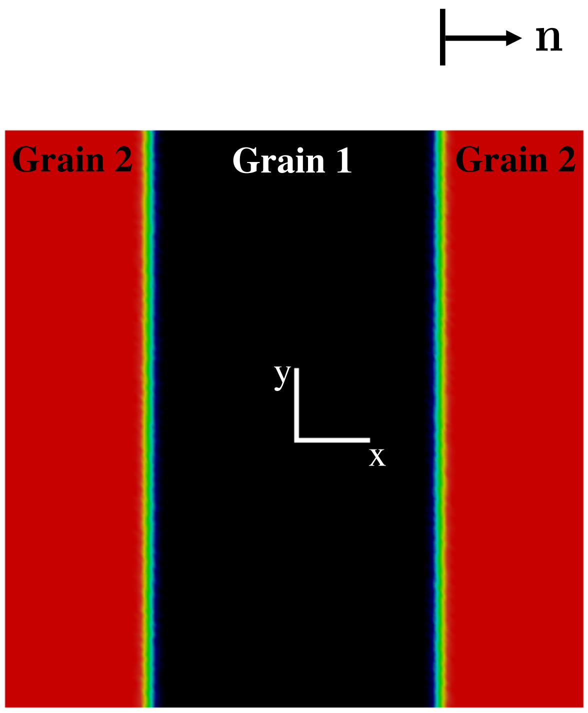


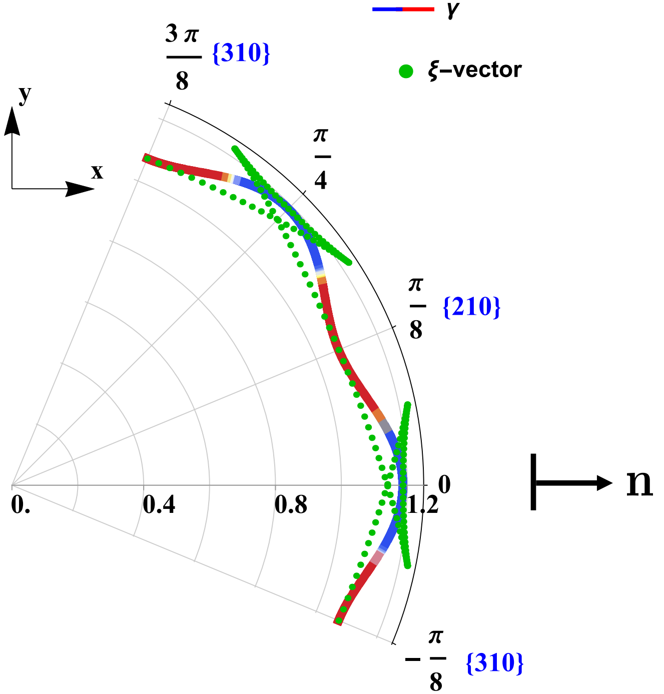
Stabilizing effect

Abdeljawad et al. J. App. Phys. **119** (2016)

Results: A Two Grain Structure

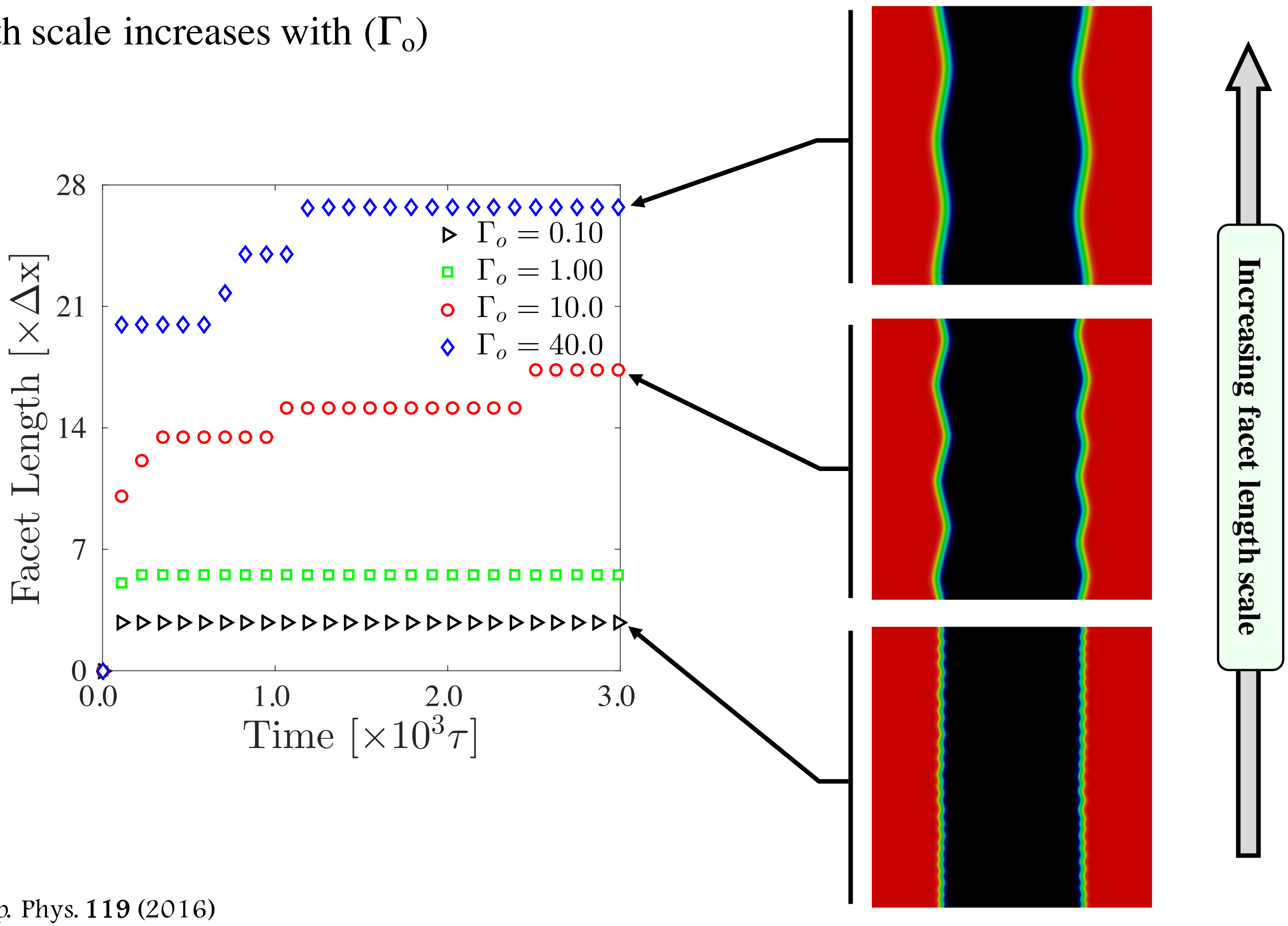
- Vary local junction energy (Γ_o) and junction interactions (Γ_1) independently





Results: Facet Junction Energy Γ_o

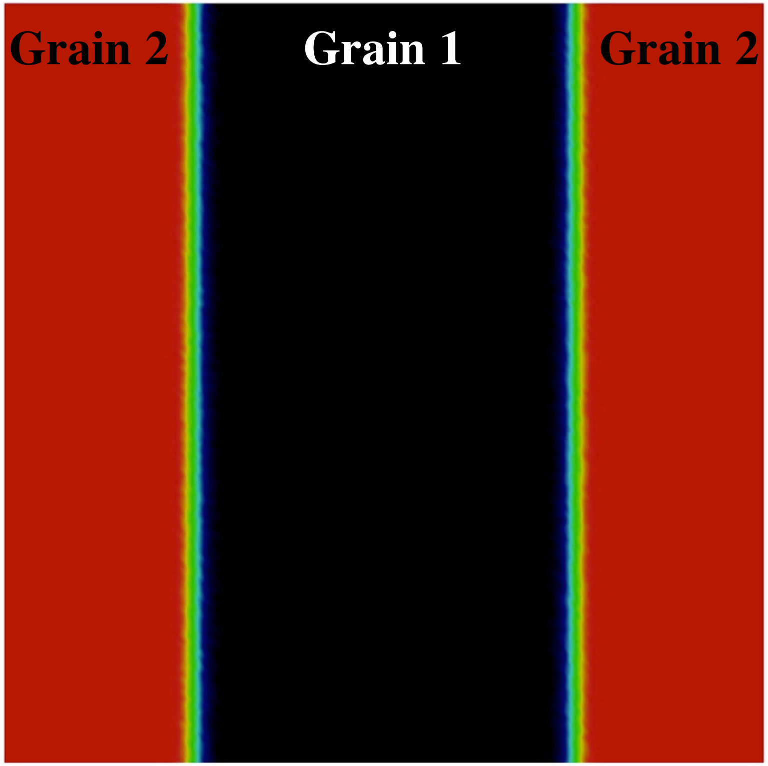
- A two-grain slab geometry
  - Facet length scale increases with (Γ_o)

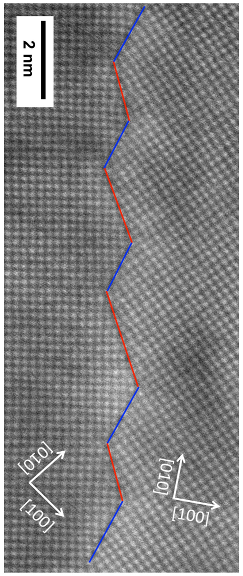


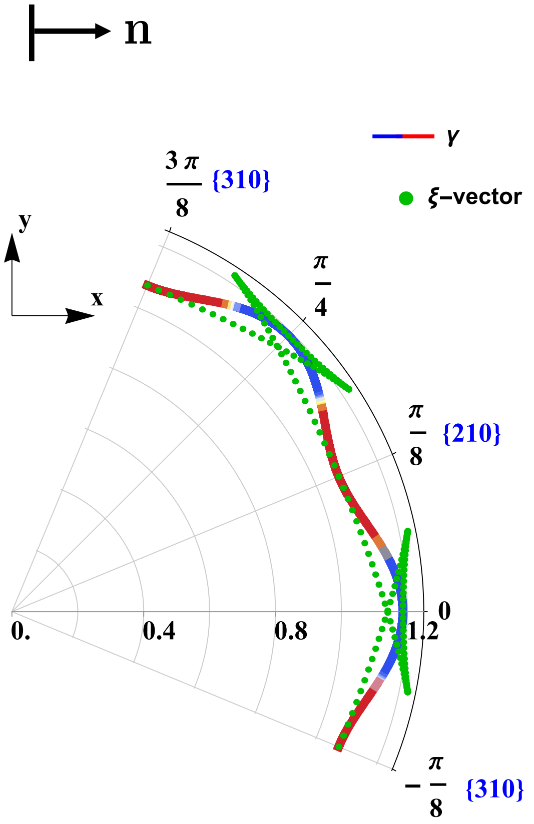
Abdeljawad et al. J. App. Phys. **119** (2016)

Results: Junction Interactions Γ_1

- A two-grain slab geometry
  - $\Gamma_1 > 0$

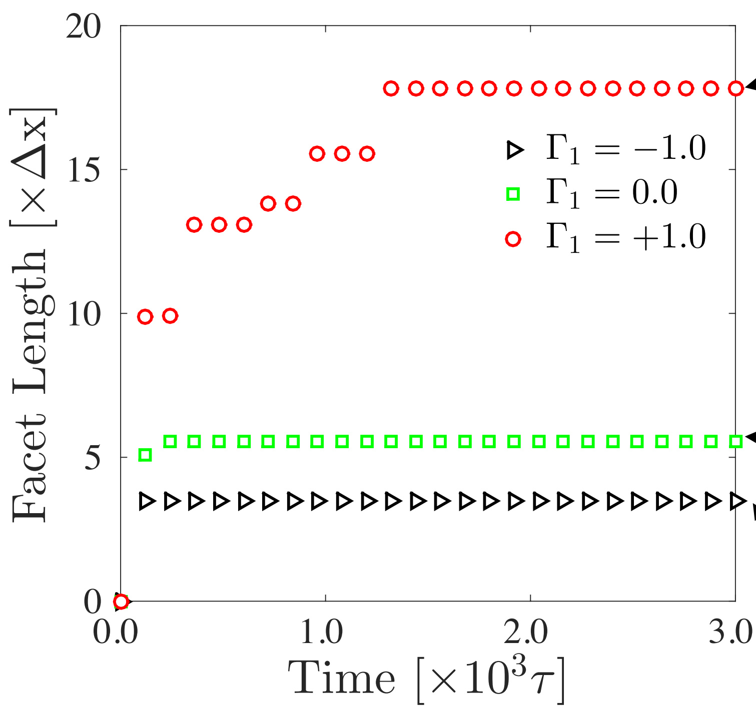


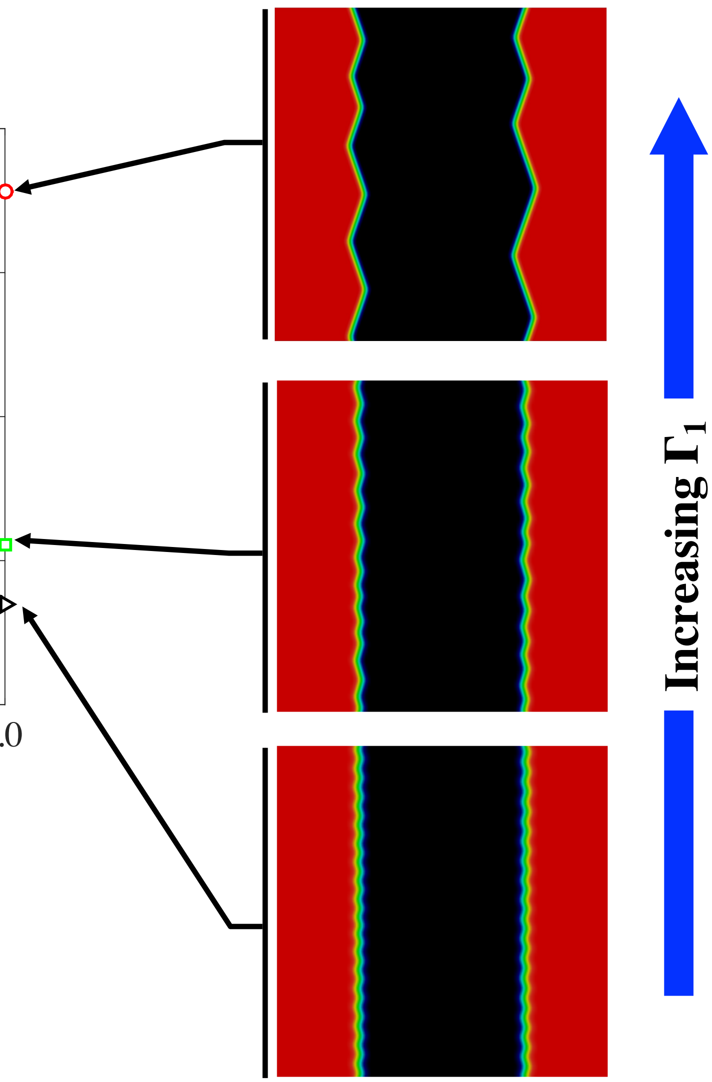


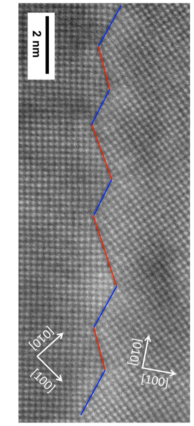


Results: Junction Interactions Γ_1

- A two-grain slab geometry
  - Negative (Γ_1) plays a stabilizing role







Dynamical scaling

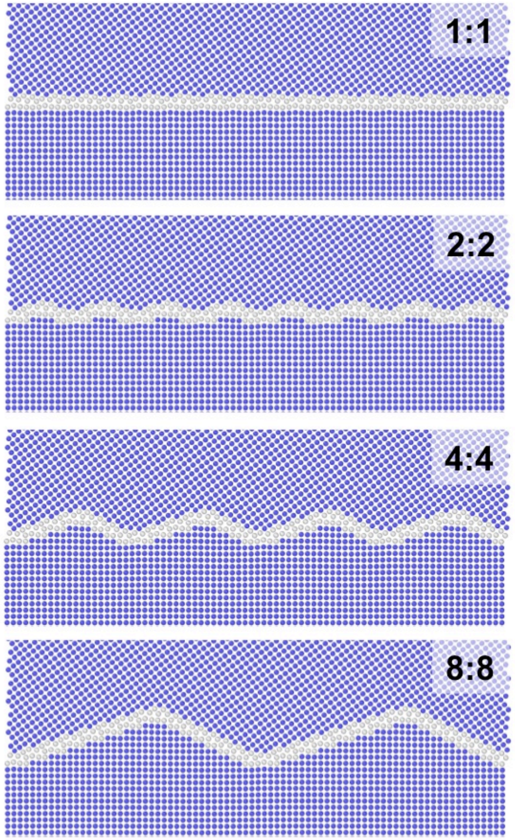
Facet growth: $\Lambda \propto t^{1/4}$

Normal grain growth: $\langle D \rangle \propto t^{1/2}$

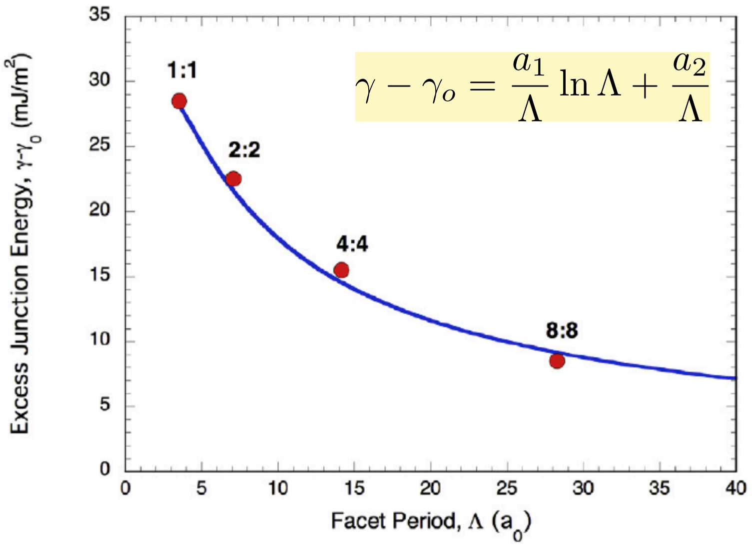
Abdeljawad *et al.*, J. App. Phys. **119** (2016)

Results: Atomistics

- Atomistic simulations
  - Fix misorientation ($\Sigma 5$) and inclination (26.565° from {310} plane)
  - Vary number of facet junctions for a given system size



Facet coarsening is favored



Medlin, Hattar, Zimmerman, Abdeljawad and Foiles, Acta Mater. (2017)

Future Work

- Faceting in 3D (underway)
  - Dependence on both polar angles: $\gamma_{gb} = \gamma_{gb}(\theta_1, \theta_2)$
  - Anisotropic GB mobility: $M_{gb} = M_{gb}(\theta_1, \theta_2)$

Homer, Patala, Priedeman, Scientific Reports (2015)

Thank you

fabdelj@sandia.gov

Medlin, Hattar, Zimmerman, *Abdeljawad* and Foiles, Acta Mater. **124** (2017)

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

Acknowledgments

Funding:

Sandia National Laboratories, Advanced Scientific Computing (ASC)

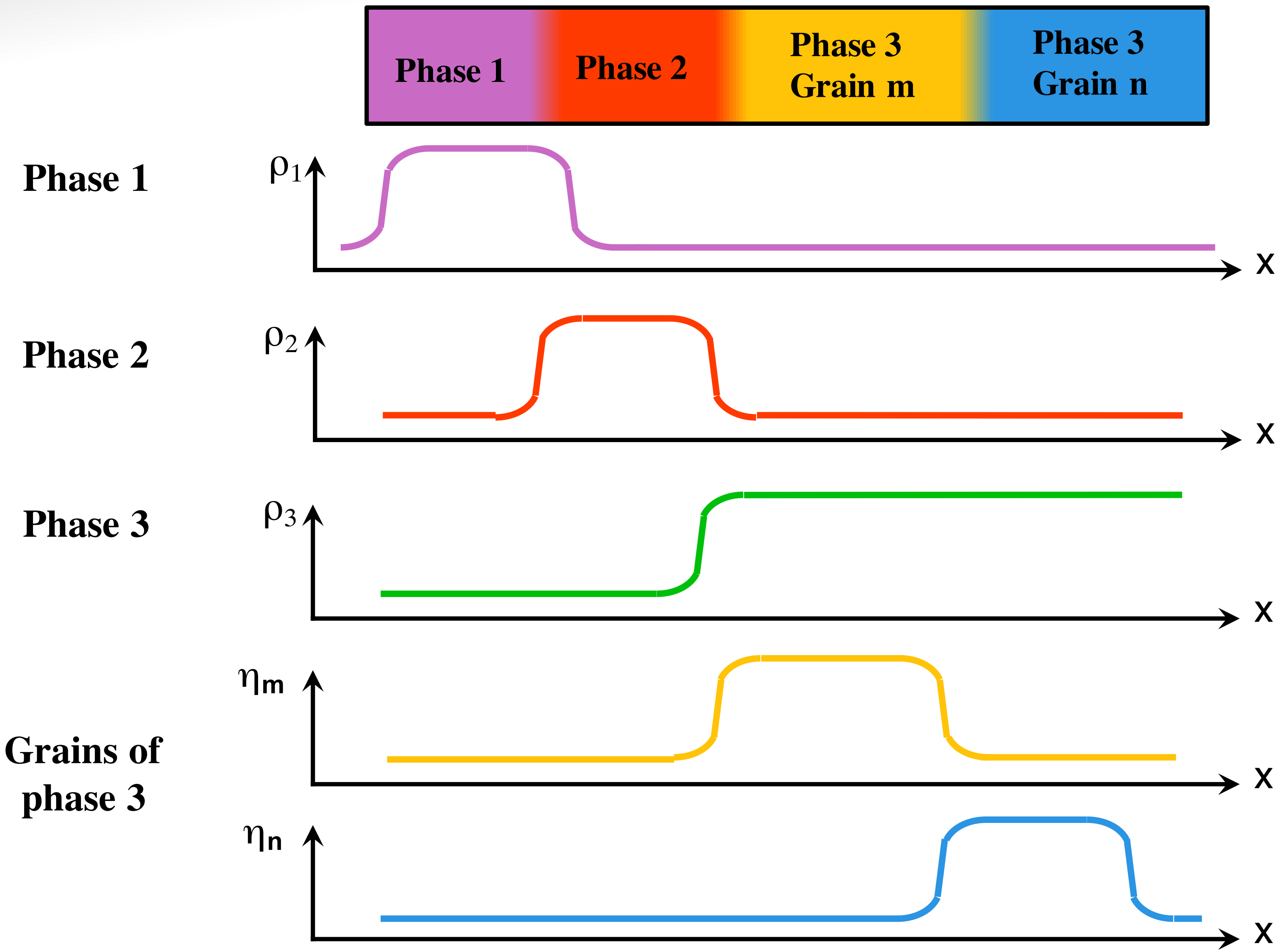
Collaboration:

1) Sandia National Laboratories: S. M. Foiles, N. Argibay, D. Medlin, H. Lim, B. L. Boyce, P. Lu, D. Bufford and K. Hattar
2) Srikanth Patala (NC State Univ.), Eric Homer (BYU)

Backup Slides

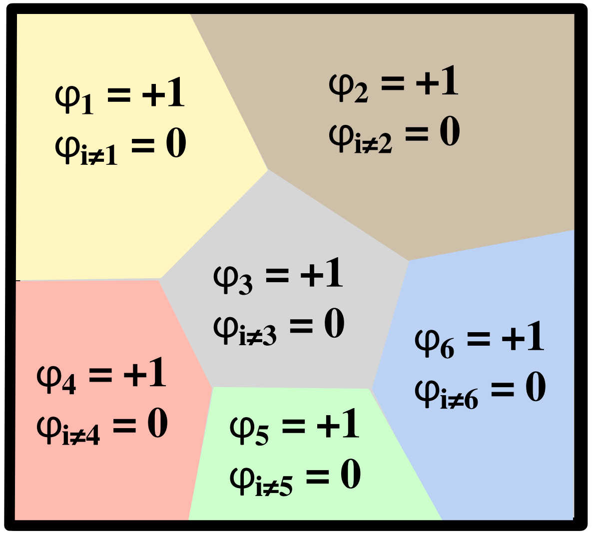
Machinery: Phase Field

Sandia National Laboratories



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