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Microstructure Evolution in Heterogeneous Systems: from Tin Whiskers to Anisotropic Grain Growth

Carol Handwerker

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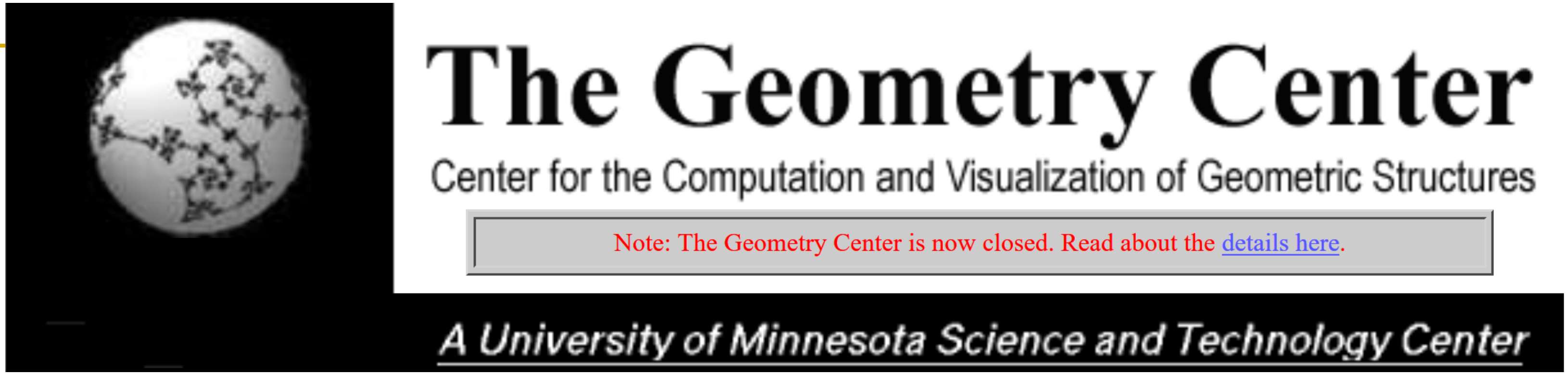
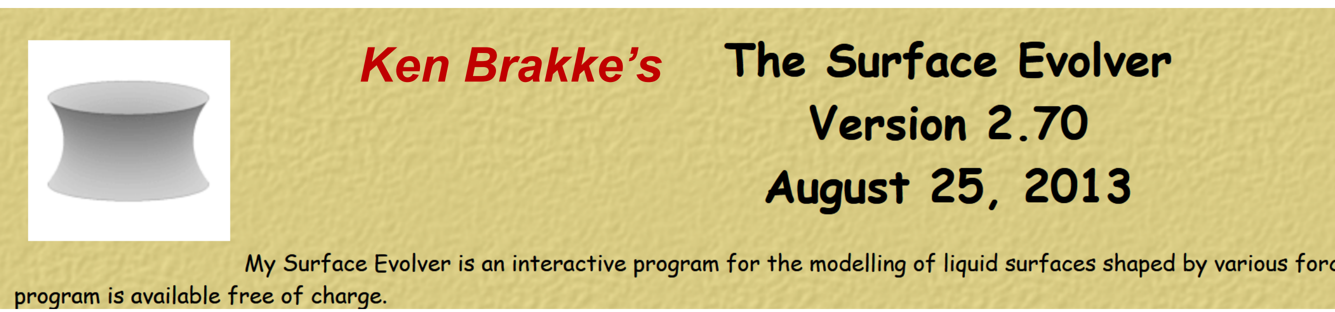
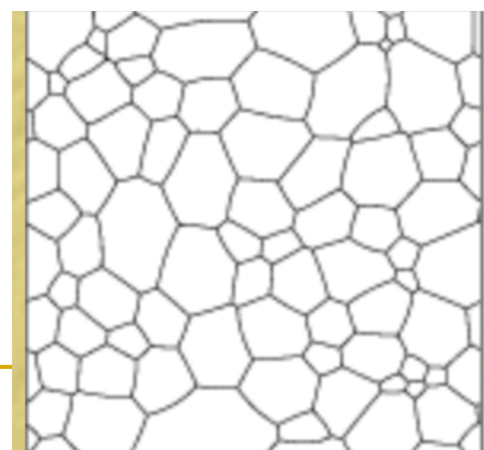
2018 SIAM Conference on Mathematical Aspects of Materials Science - Portland OR



Bridges from Mathematics & Geometry to Materials Science:
Jean Taylor, Fred Almgren, John Cahn, Ken Brakke,
Robert Kohn, Frank Morgan



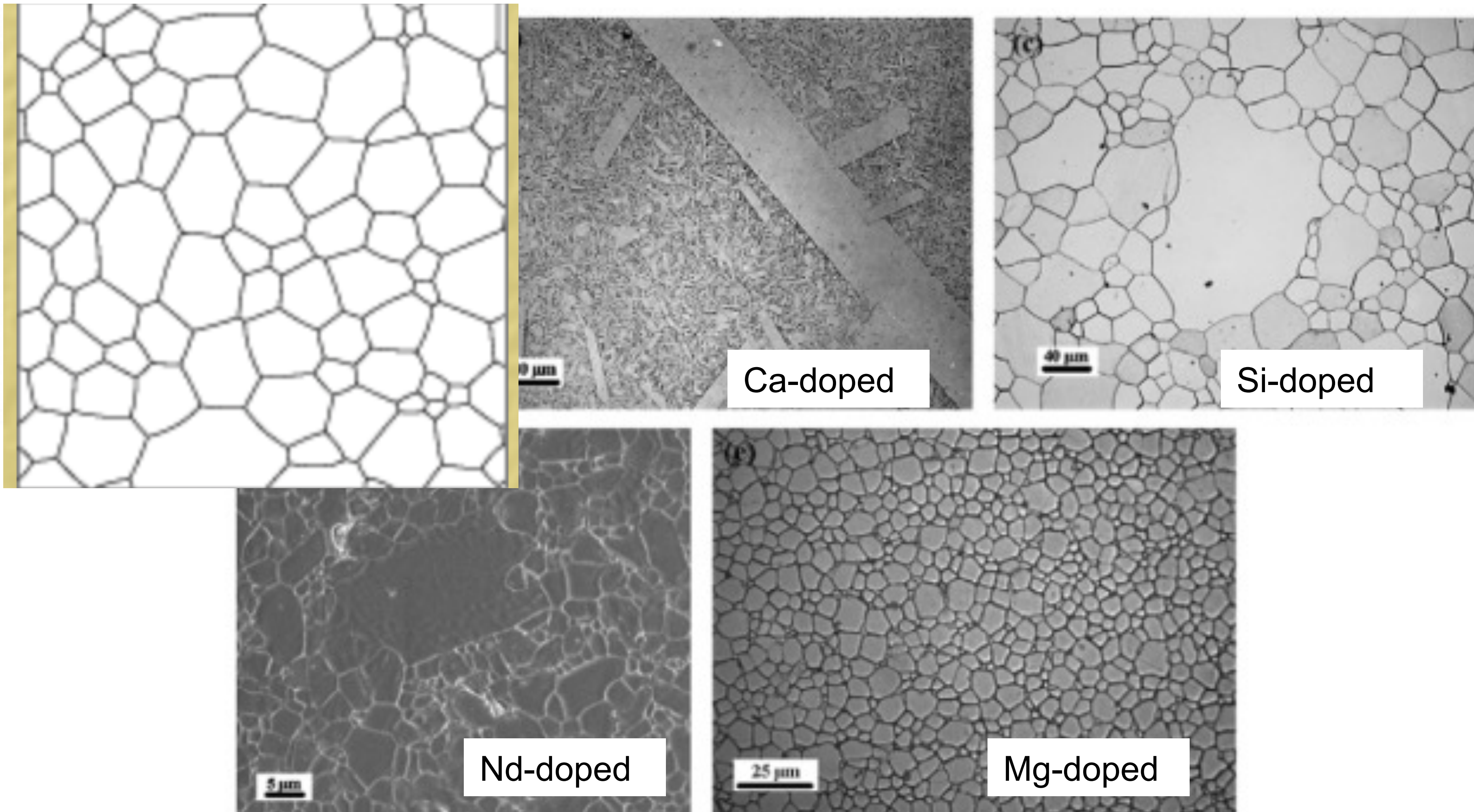
Giphy



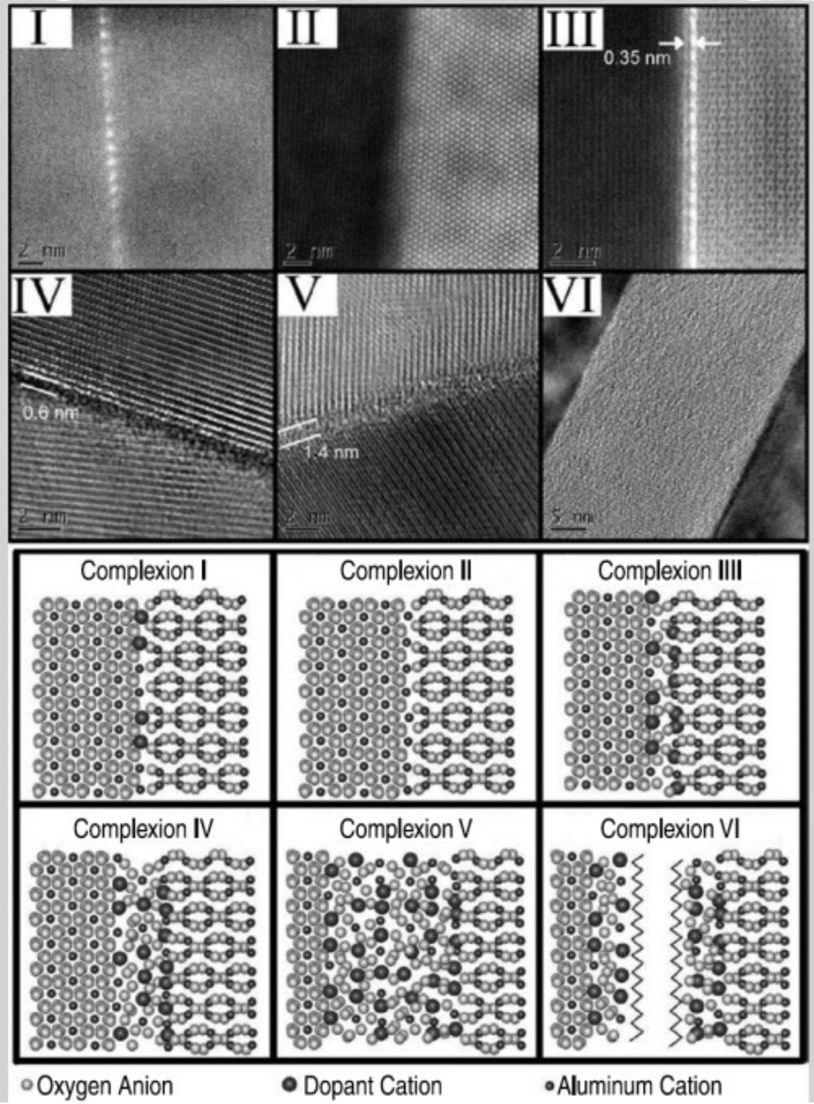
Complexion: A new concept for kinetic engineering in materials science

Shen J. Dillon [a,*], Ming Tang [b], W. Craig Carter [b], Martin P. Harmer [a]

Acta Materialia 55 (2007) 6208–6218

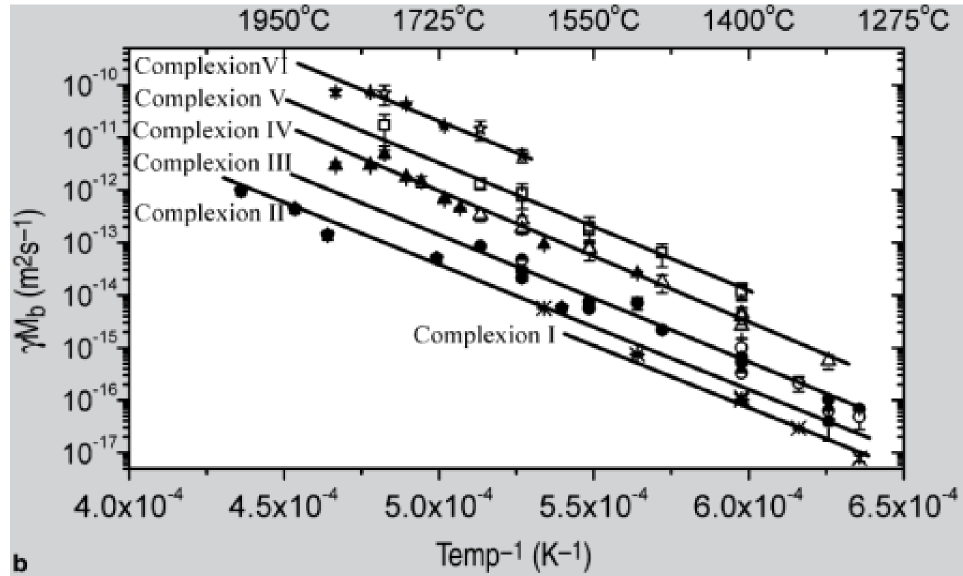


Grain Boundary Complexions in Ceramics and Metals: An Overview



Shen J. Dillon, Martin P. Harmer, and Jian Luo

Vol. 61 No. 12 • JOM



Focused Ion Beam Milling (FIB)
Local composition & structure

- HRTEM & HAADF STEM
- EBSD

- Understand and separate factors controlling normal/abnormal grain growth

Microstructure Evolution in Heterogeneous Systems

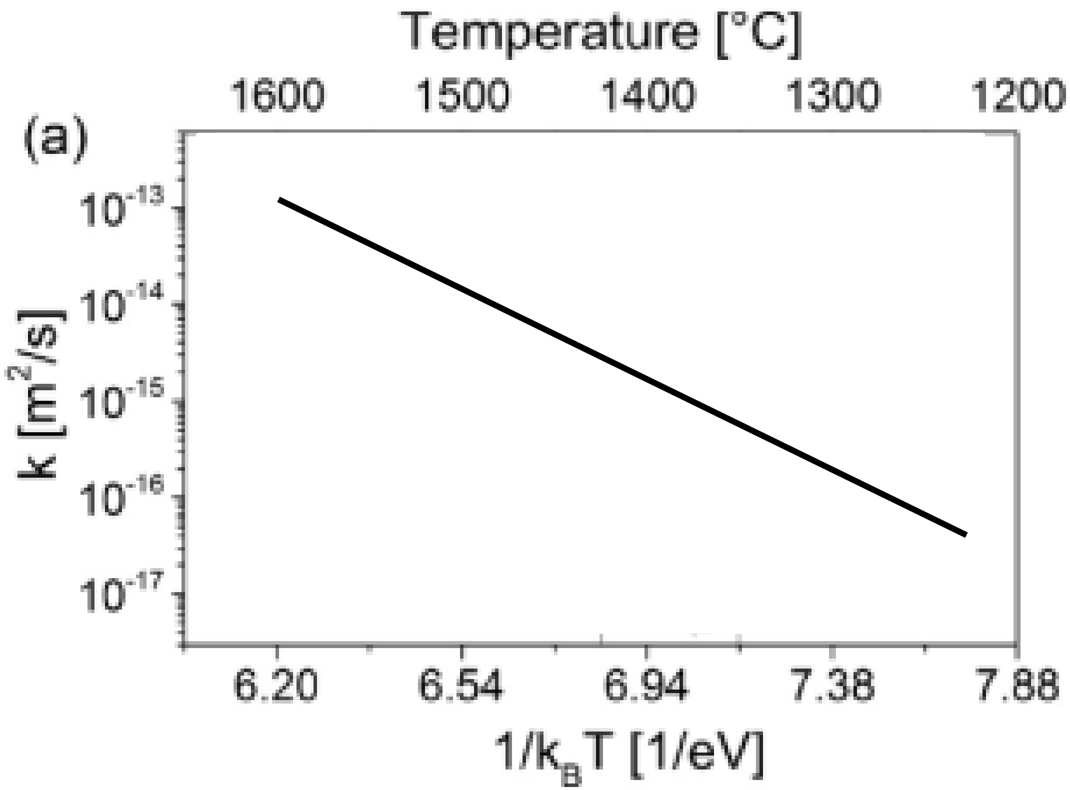
- Anisotropic Grain in Strontium Titanate
  - Anti-thermal
  - “Normal” grain growth is not normal
  - Abnormal grain growth in the absence of a liquid phase
  - Grain growth stagnation

- Tin Whisker Formation in Sn Thin FIlms
  - Stress-driven crystal growth out of the film surface from a grain embedded in the film
  - 1 grain out of 10,000 or 100,000 grains forms a whisker
  - Sources and sinks of atoms
  - Role of grain boundary geometry and crystallographic orientation
  - Evidence of grain boundary sliding and coupling

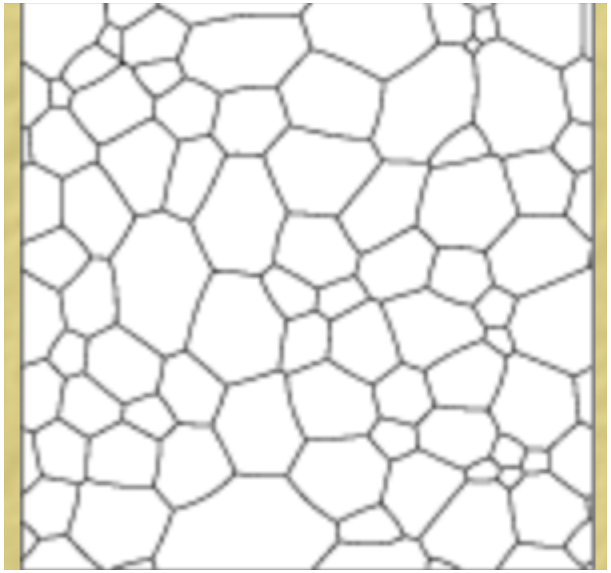
Grain growth in perovskites: What is the impact of boundary transitions?

Wolfgang Rheinheimer *, Michael J. Hoffmann

Current Opinion in Solid State and Materials Science 20 (2016) 286–298



"Arrhenius" behavior



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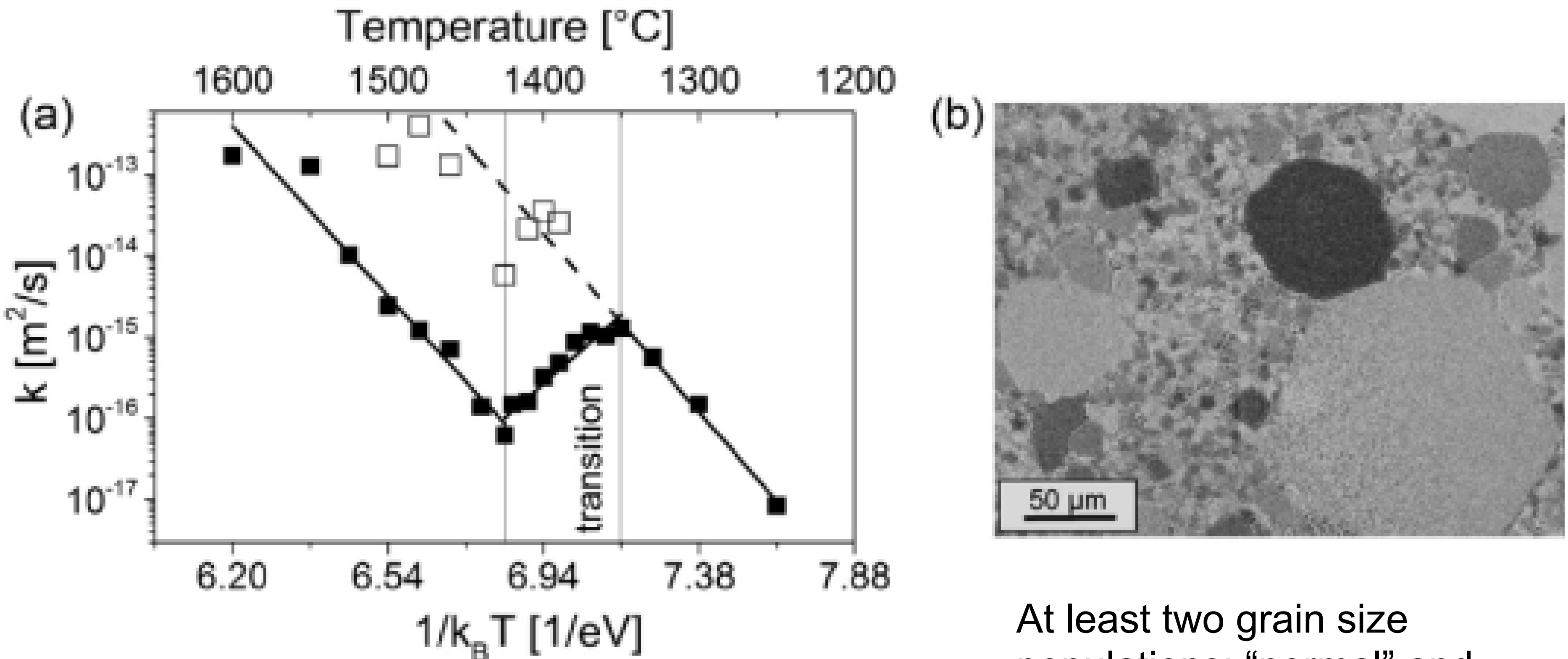

At least two grain size populations: “normal” and “abnormal”

No liquid films present

Grain growth in perovskites: What is the impact of boundary transitions?

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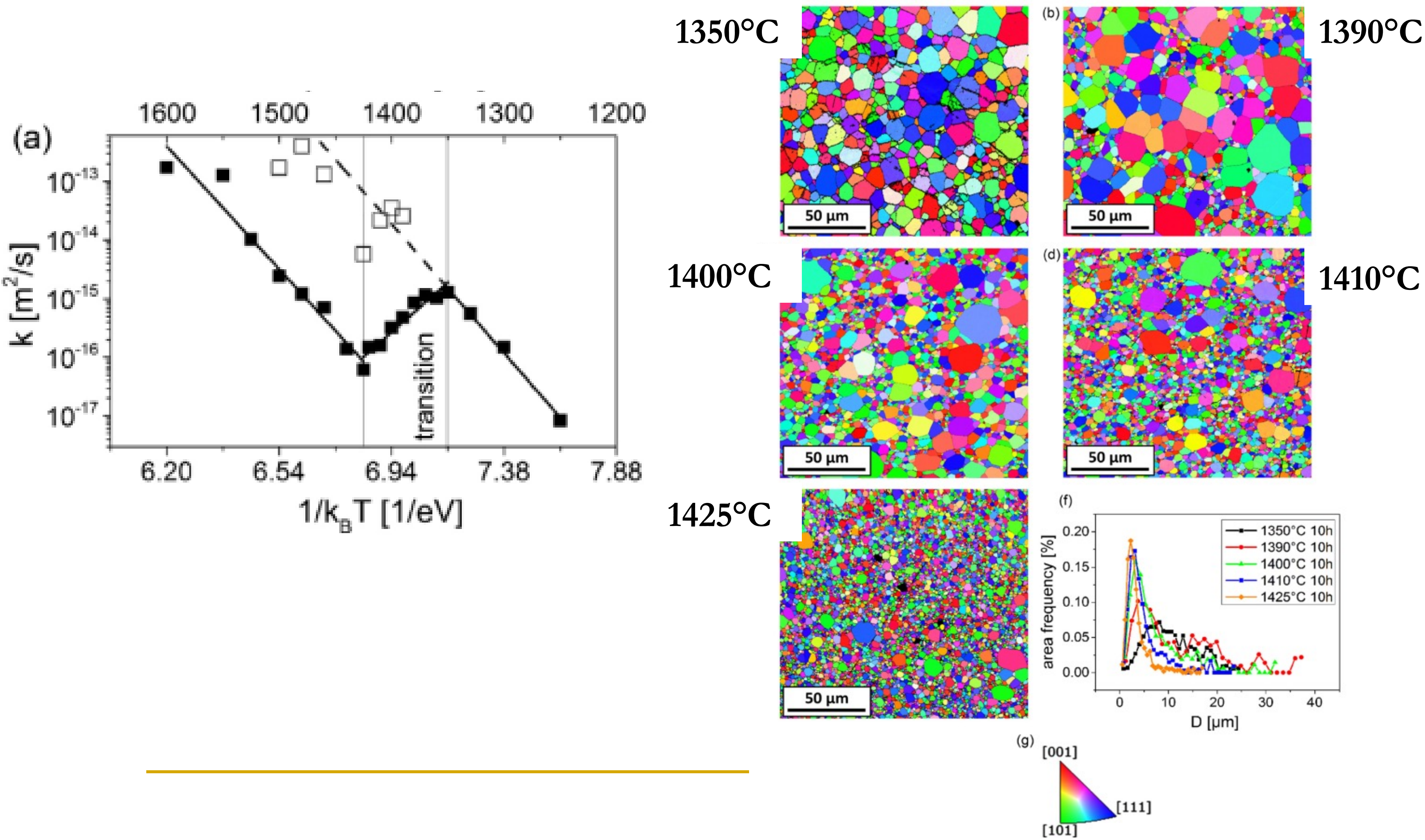
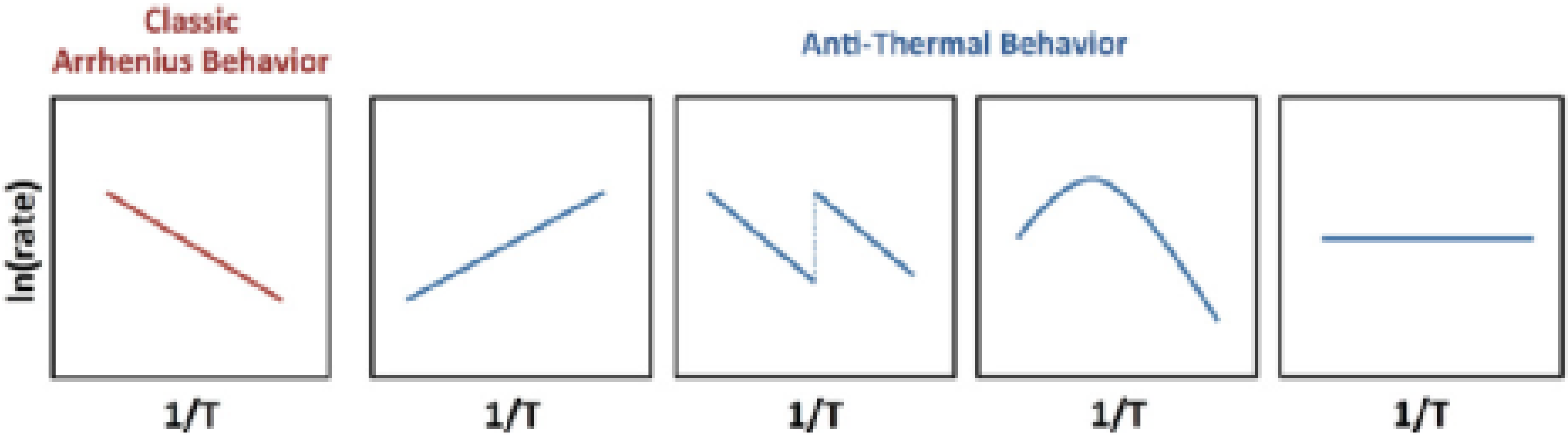


Viewpoint Paper

Anti-thermal behavior of materials

Patrick R. Cantwell,[a] Elizabeth A. Holm,[b] Martin P. Harmer[c,*] and Michael J. Hoffmann[d]

P.R. Cantwell et al. / Scripta Materialia 103 (2015) 1–5



JOM, Vol. 66, No. 1, 2014

Trends in Grain Boundary Mobility: Survey of Motion Mechanisms

ERIC R. HOMER,[1,2,5] ELIZABETH A. HOLM,[1,3] STEPHEN M. FOILES,[1] and DAVID L. OLMSTED[4]

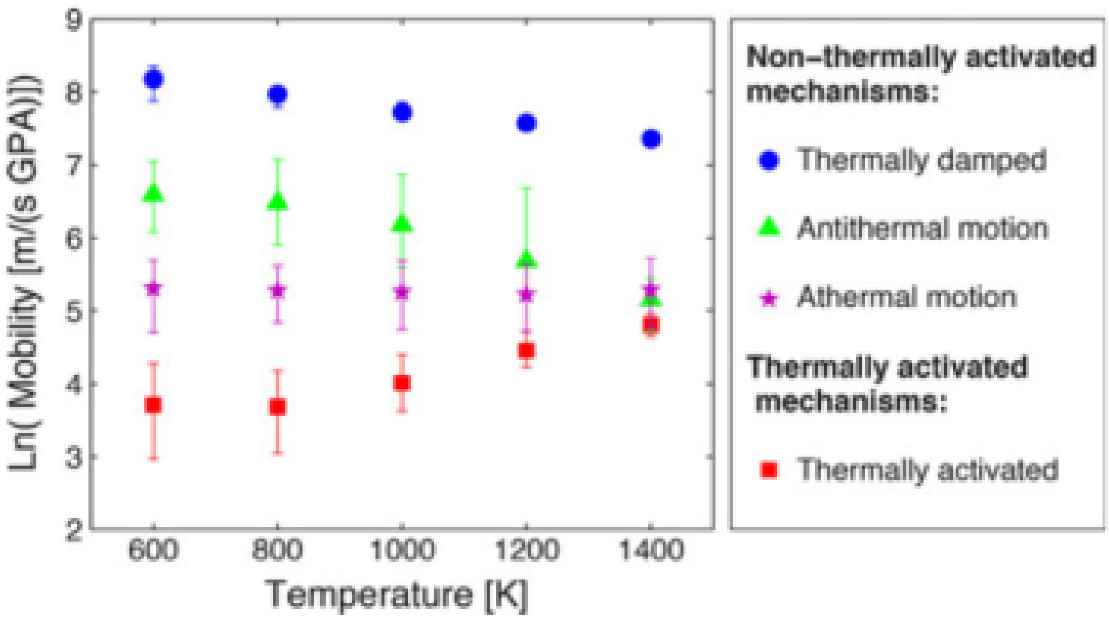


Fig. 4. Plot of median mobility for thermally activated and three non-thermally activated motion trends: thermally damped, antithermal, and athermal motion. Interestingly, although they have different temperature-dependent trends, the antithermal, athermal, and thermally activated motion trends converge at high temperatures near the melting point. *Error bars* indicate the first and third quartiles of the mobility distribution for each trend and temperature.

Synthetic driving force molecular dynamics model

Mobilities of 388 grain boundaries in Ni

- 57% thermal activated mobility
- 20% athermal or decreasing with increasing temperature
- 14% mixed modes
- 9% unclassifiable or immobile

Microstructure Evolution in $SrTiO_3$

Growth of single crystalline seeds into polycrystalline strontium titanate: Anisotropy of the mobility, intrinsic drag effects and kinetic shape of grain boundaries
W. Rheinheimer, M. Baeurer, C. Handwerker, J. Blendell, M. Hoffmann
Acta Materialia, 95 (2015) 111-123.

The equilibrium crystal shape of strontium titanate and its relationship to the grain boundary plane distribution,
W. Rheinheimer, M. Baeurer, H. Chien, G. Rohrer, C. Handwerker, J. Blendell, M. Hoffmann Acta Materialia, 82 (2015) 32-40

Decomposing the effects of

- Surface energy anisotropy as a function of T
- Grain boundary energy anisotropy as a function of T, t
- Anisotropic grain boundary motion – single xtl/poly & AGG
- Assumptions of limiting crystal growth shapes from a uniformly supersaturated medium – kinetic growth shape
- Possible causes of stagnation

Grain Boundary Plane Distributions in Polycrystals

- Relationship between surface energy, grain boundary energy (γ_{GB}), and the population of grain boundary planes found in a polycrystal based on the work of Rohrer, Saylor, Sano, El Dasher, …

$$\gamma_{GB}(g, \boldsymbol{n}) = \gamma_{s1} + \gamma_{s2} - E_{binding}$$

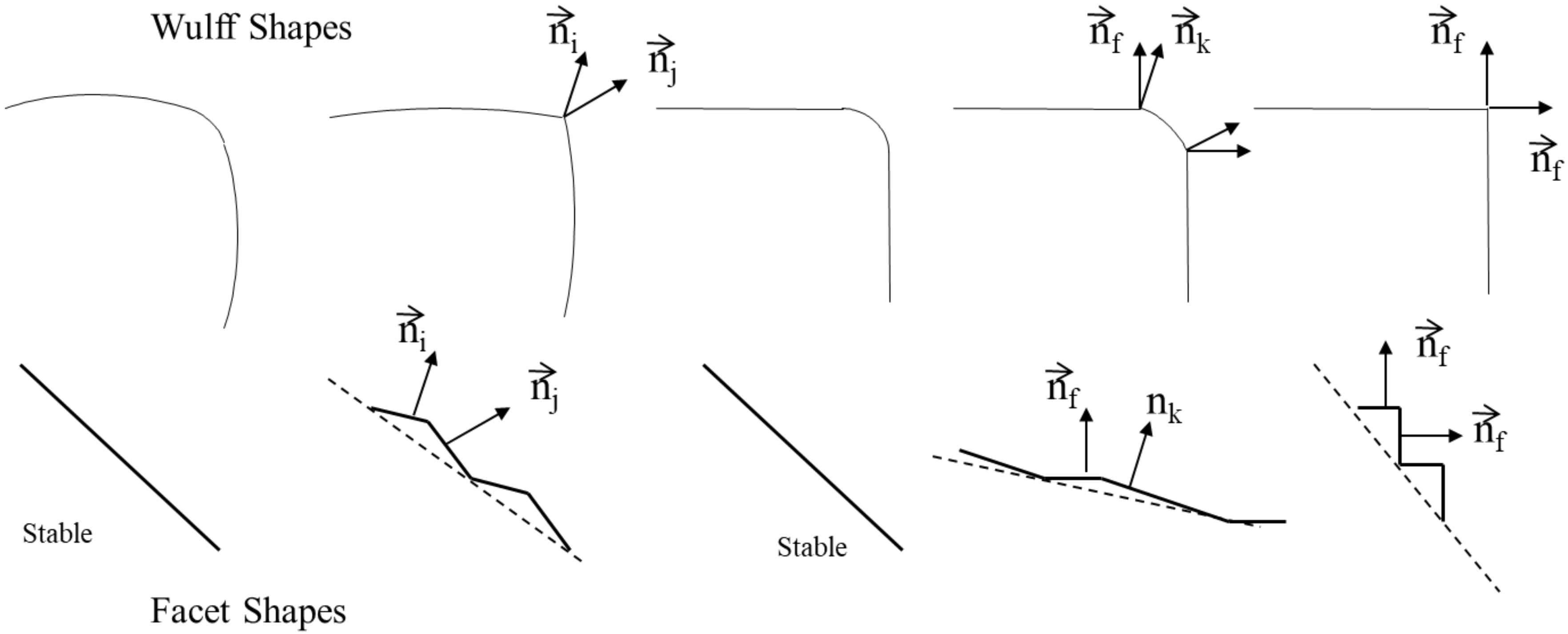
$E_{binding} \approx const.$ for all misorientations

Distribution of grain boundaries in magnesia as a function of five macroscopic parameters - David M. Saylor, Adam Morawiec, Gregory S. Rohrer Acta Materialia 2003

From G. Rohrer, Annual Reviews (2005)

1. distribution of grain boundary planes is anisotropic
2. preferred habit planes for grains within polycrystals correspond to the same low-energy, low-index planes that dominate the **external growth forms** and **equilibrium shapes** of isolated crystals of the same phase with correlation identified between the sum of the surface free energies that make up the boundaries and the grain boundary energy, γ (g, **n**)
3. grain boundary character distribution is correlated to grain boundary energies, γ (g, **n**)

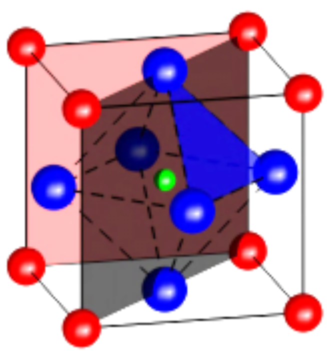
Interface Shapes

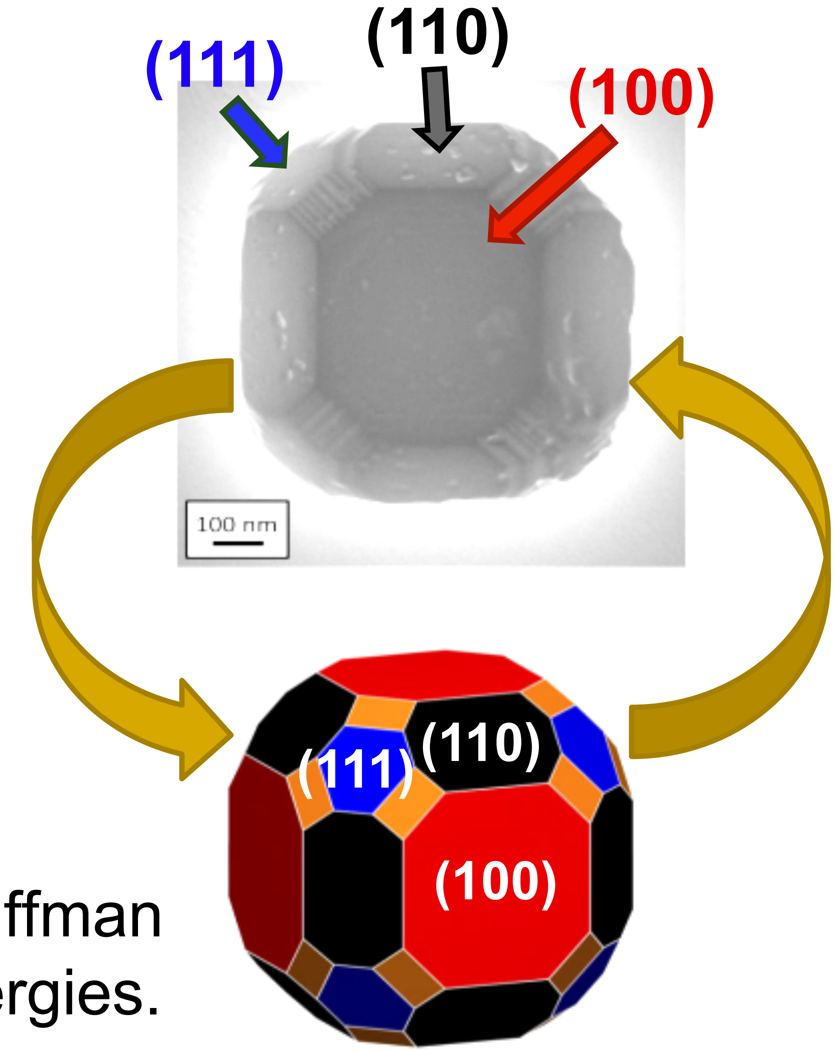


- As T changes, previously stable orientations become unstable but retain previous average orientation.
- Follow the local changes in the structures and grain boundary plane orientations as a function of T, time, and misorientation

Reconstruction of the Wulff shape from shapes of internal pores

- 2D SEM images
- Indexing crystallographic planes
  - Symmetry of perovskite

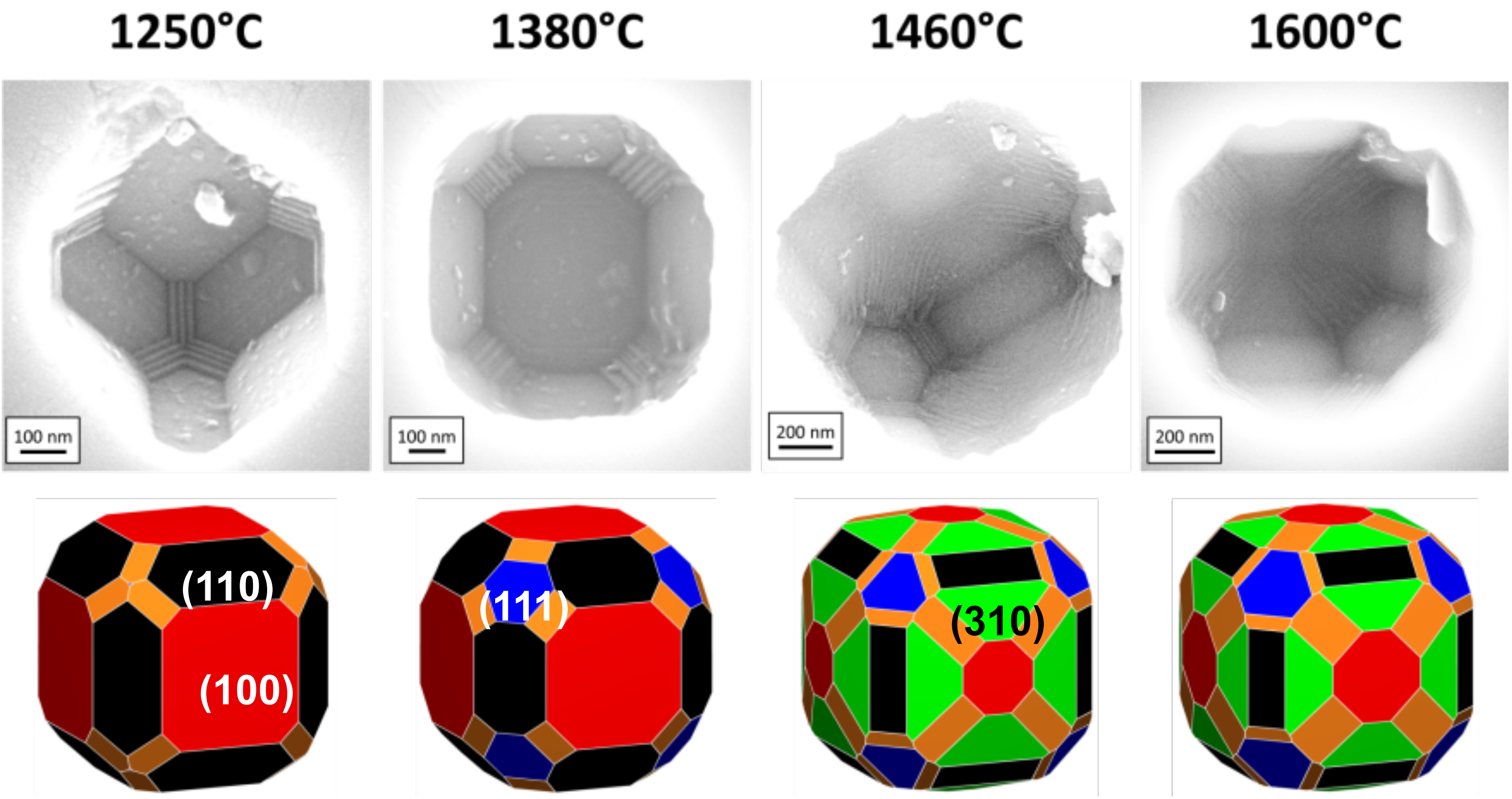




- Fitting the pore shape using Wulffman by changing relative surface energies.

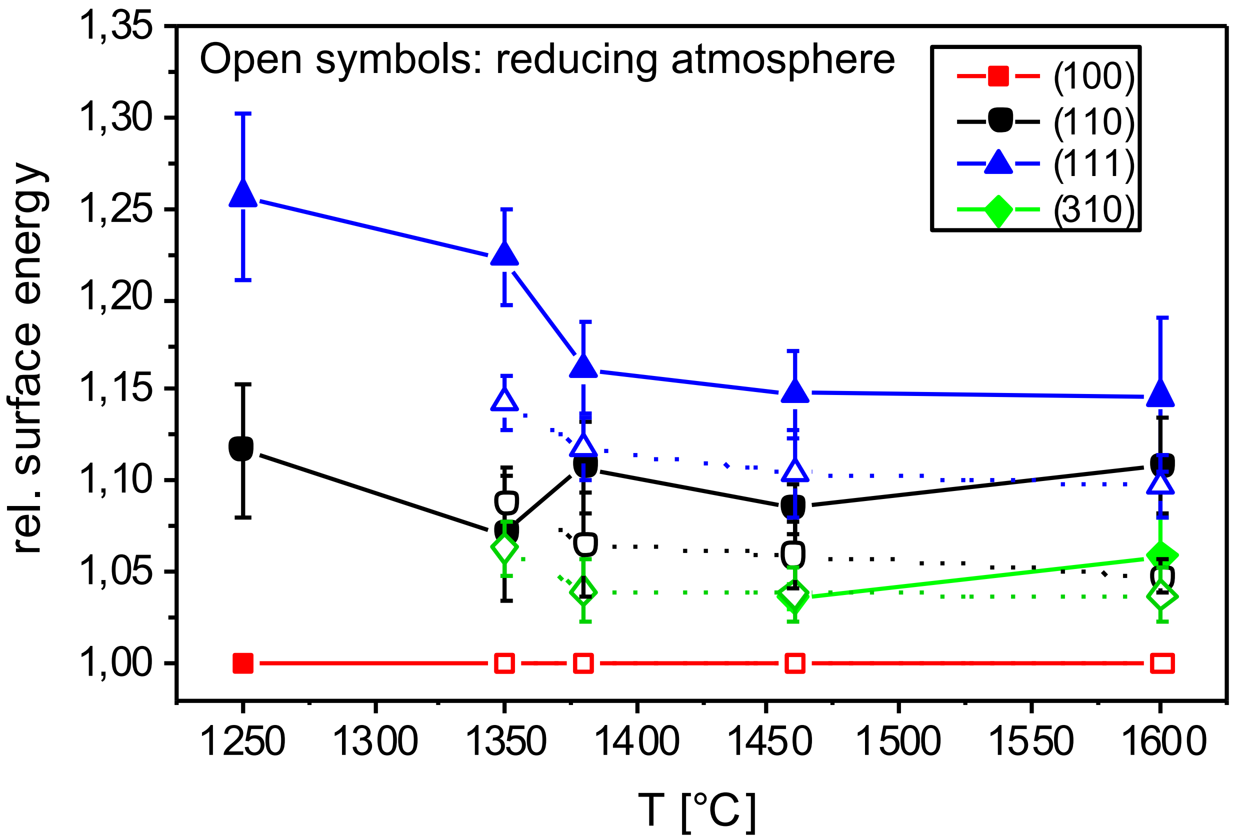
W. Rheinheimer, M. Baeurer, H. Chien, G. Rohrer, C. Handwerker, J. Blendell, M. Hoffmann
Acta Materialia, 82 (2015) 32-40

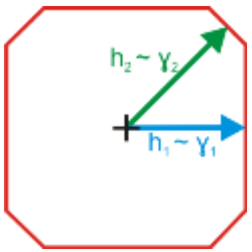
The Wulff shape of Strontium Titanate



- 4 low energy planes: (100), (110), (111), (310)
- Pore shape becomes more isotropic with increasing temperatures
- Microfacetted areas, not equilibrium

Relative surface energy of strontium titanate

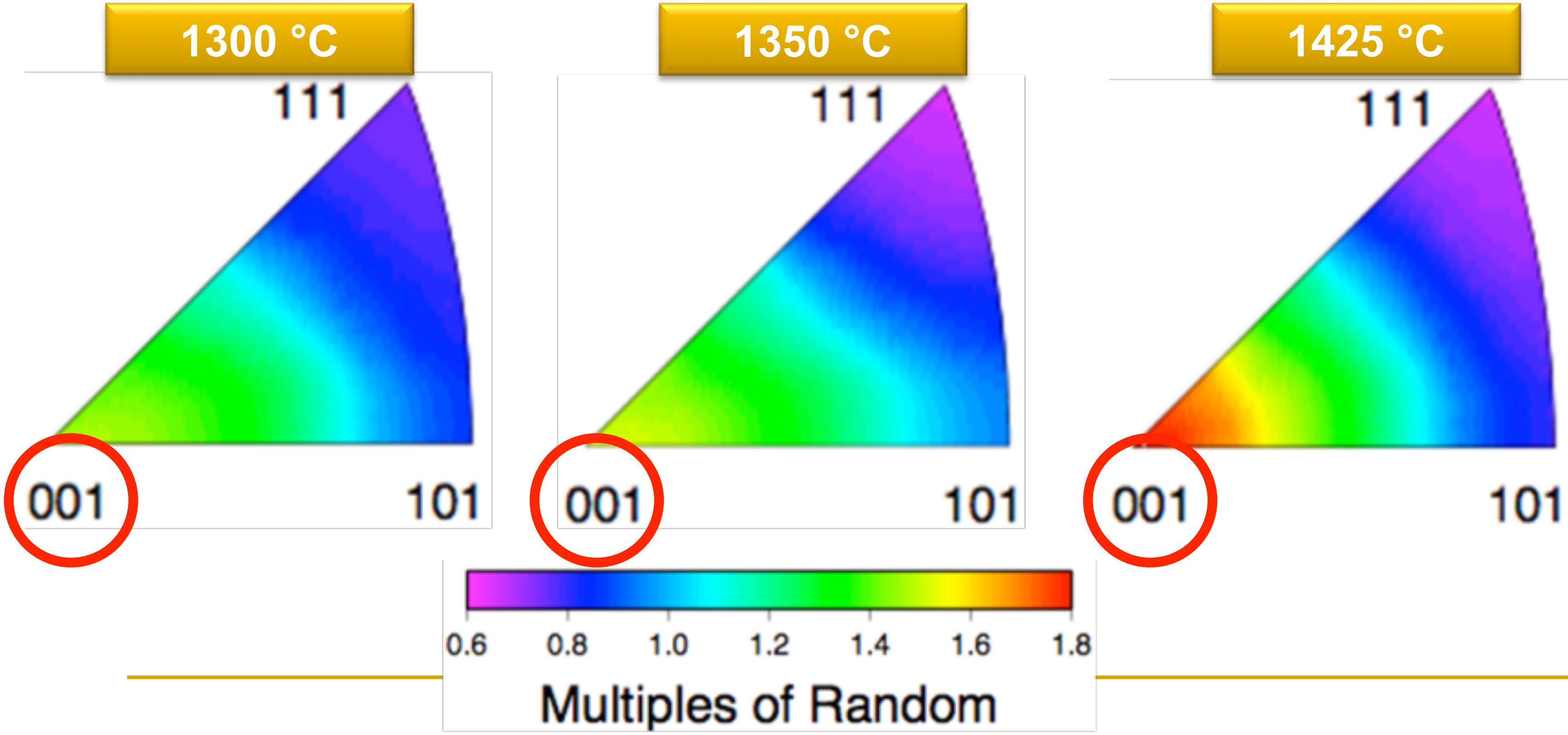




- Anisotropy decreases with increasing temperature
- No evidence to explain the grain growth anomaly

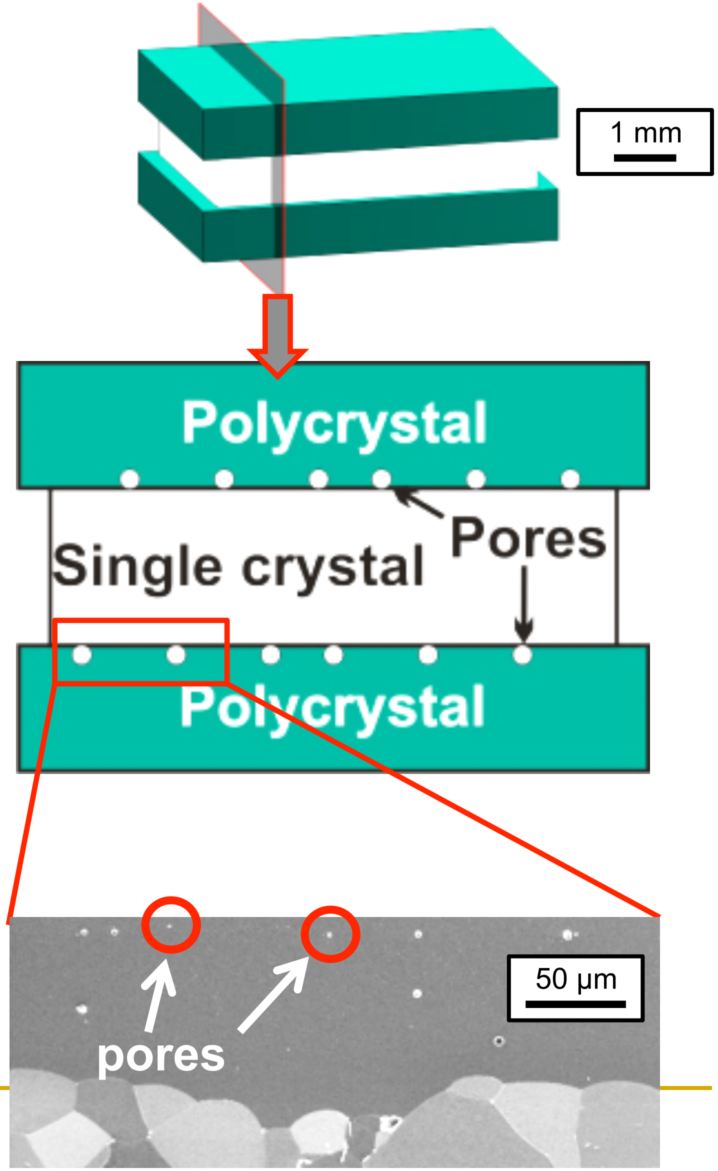
Grain boundary plane distribution in strontium titanate

- Frequency of grain boundaries facetted in (100) expected to decrease with increasing temperature and (111) expected to increase
  - Grain boundary plane distribution for same average grain size
  - Frequency of (100) increases and (111) decreases with temperature

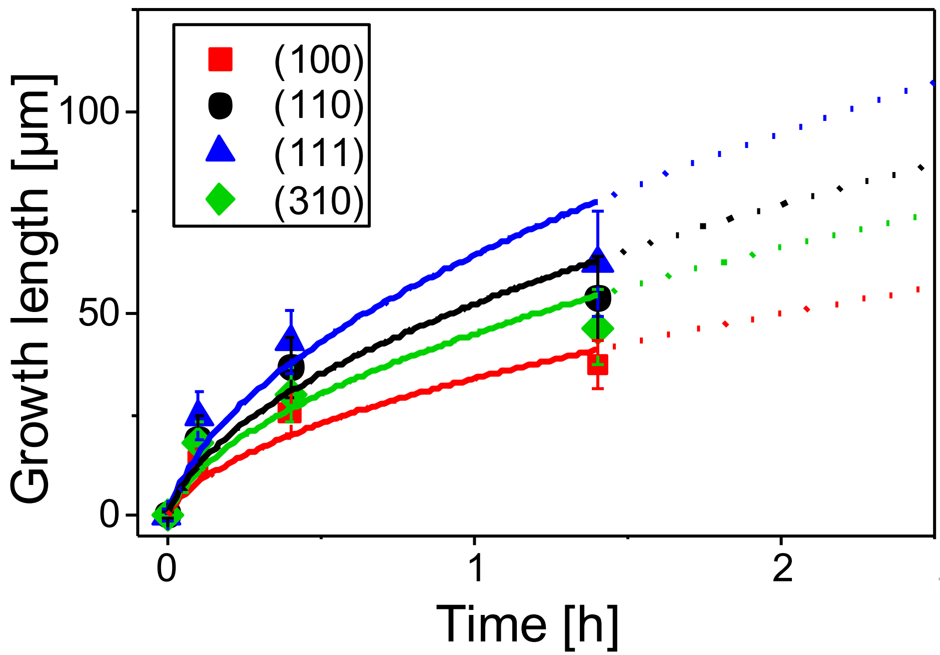


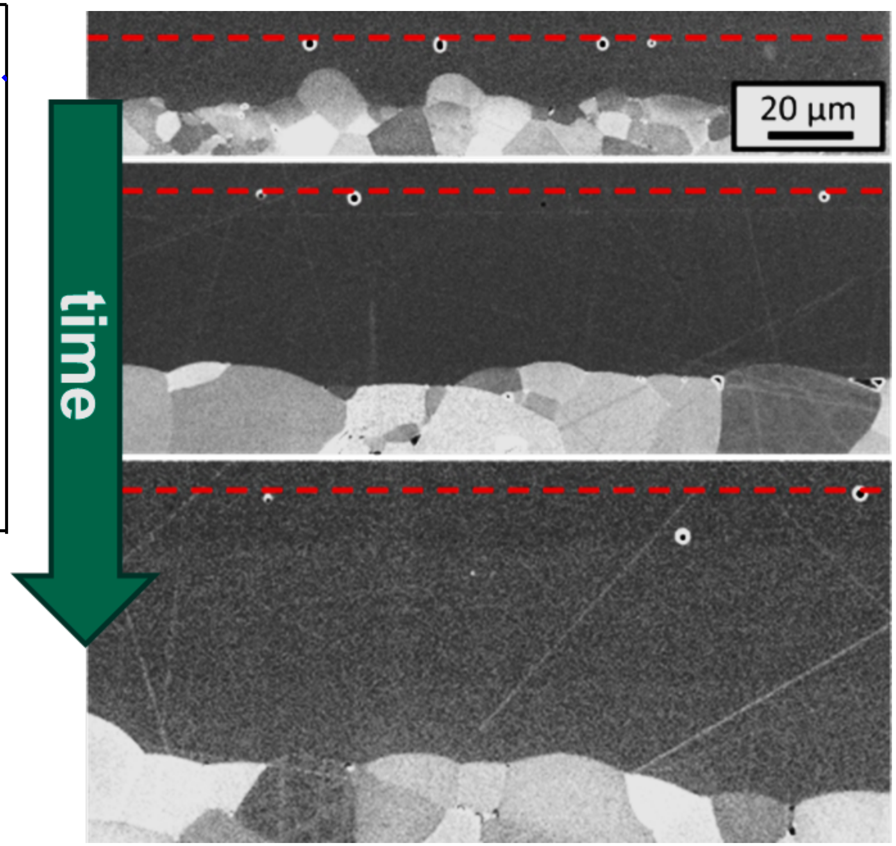
Model experiments: single crystal cut at specific orientations migrating into polycrystals

- Diffusion bonding of single crystals and polycrystals
  - Small scratches on the surface result in pores on the interface (d ≈ 1 µm)
  - Pores have to be small for depinning



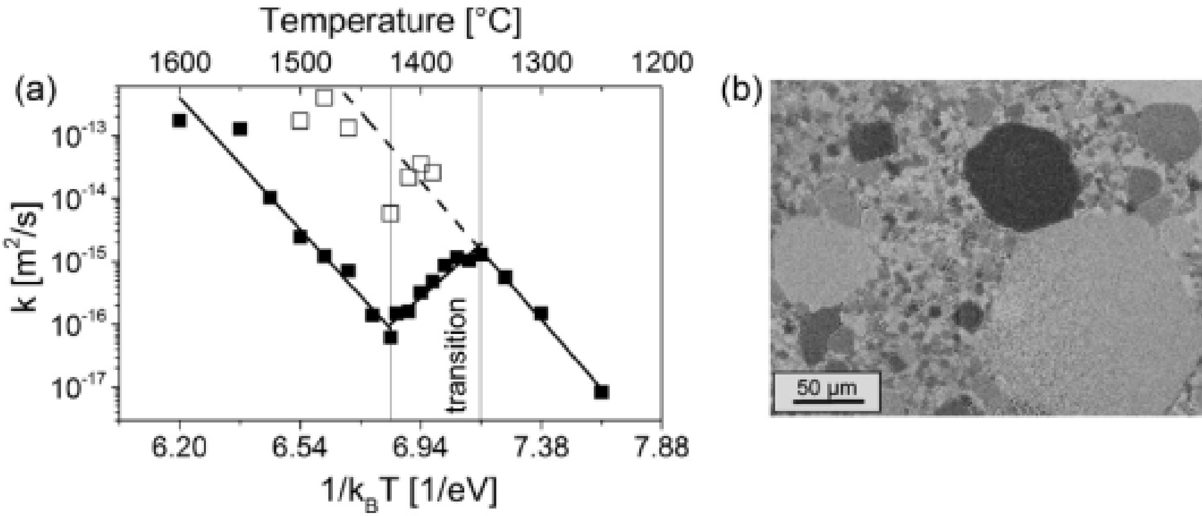
Growth of the single crystals





- Growth of the single crystals
  - 4 different orientations
  - As a function ot temperature
    - example here: 1550°C in oxygen

Rheinheimer, Bäurer, Handwerker, Blendell & Hoffmann, *Acta Materialia,* 2015



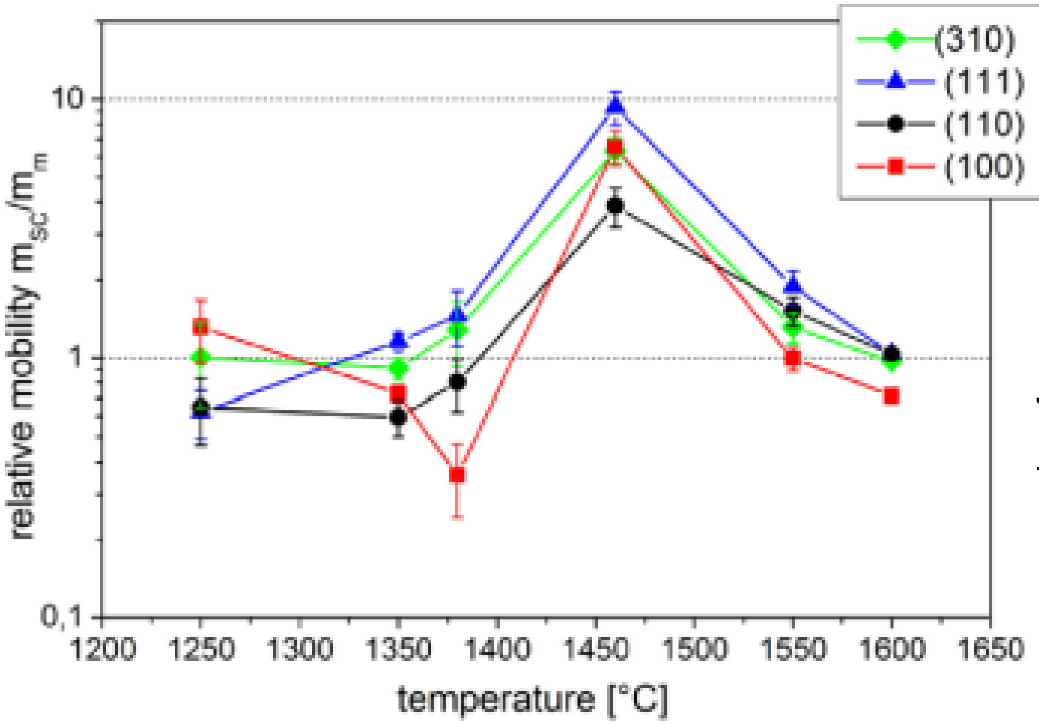
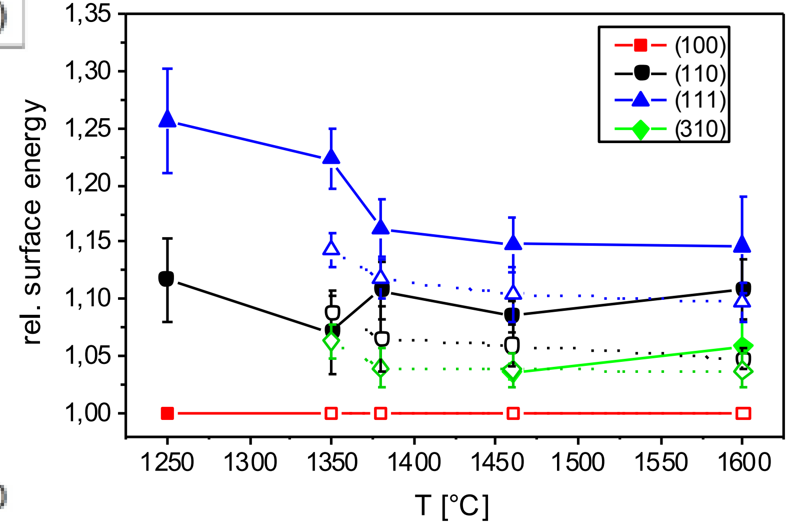
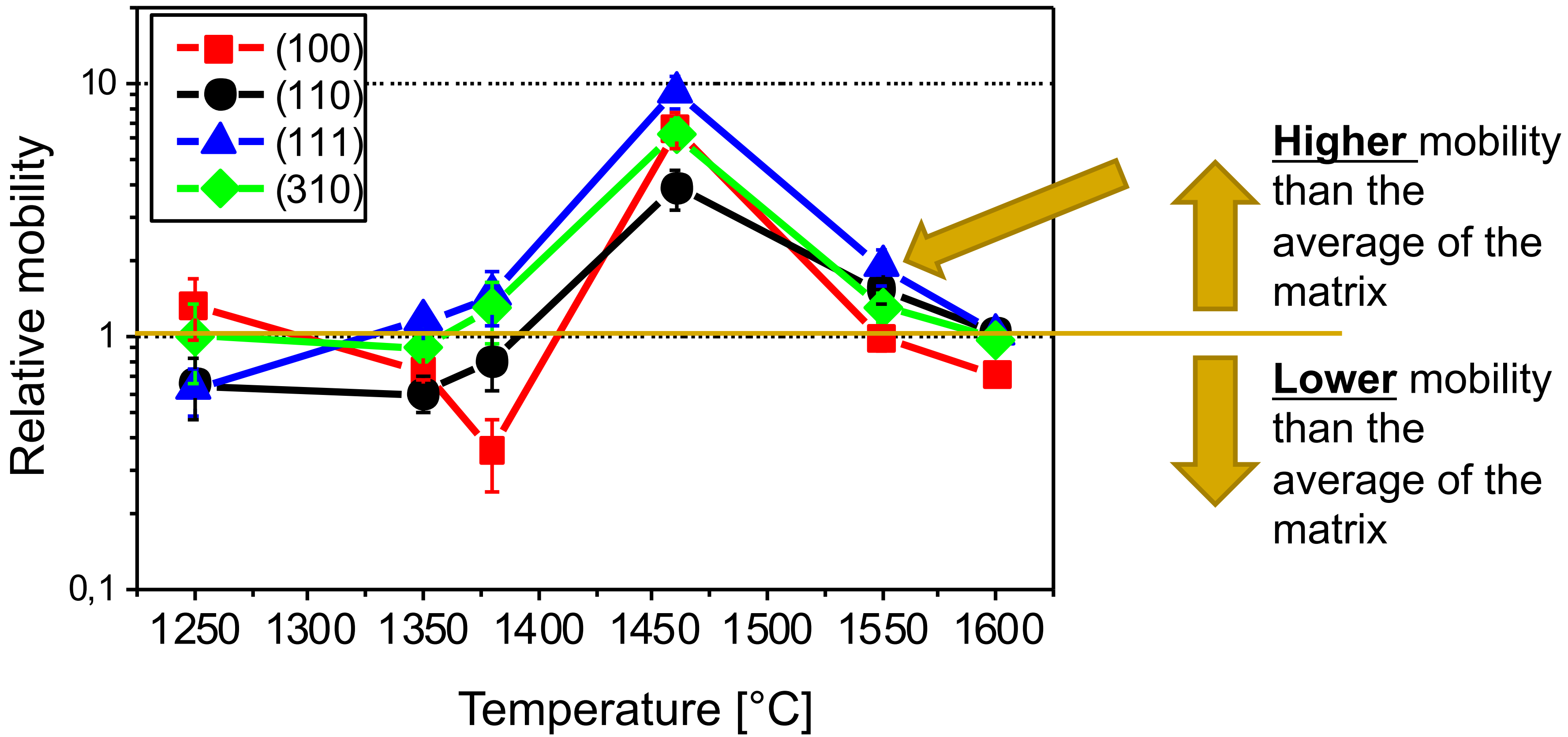


Fig. 11. Relative mobility of different single crystal orientations. The lines are drawn to guide the eye.

Open symbols: reducing atmosphere



Relative mobility and the grain growth anomaly



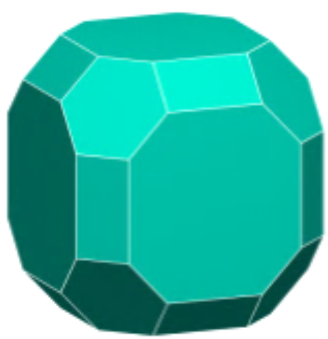
- Mobility of the single crystals relative to the matrix
- Below the line => matrix grains are catching up

Growth shape versus equilibrium shape

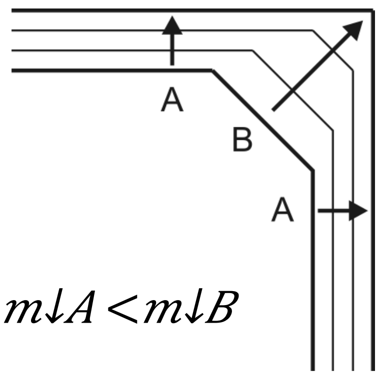
Which planes dominate the boundaries of large abnormal grains in the matrix?

Which orientations dominate the boundary planes?

Equilibrium crystal shape (Wulff shape)



Kinetic crystal shape



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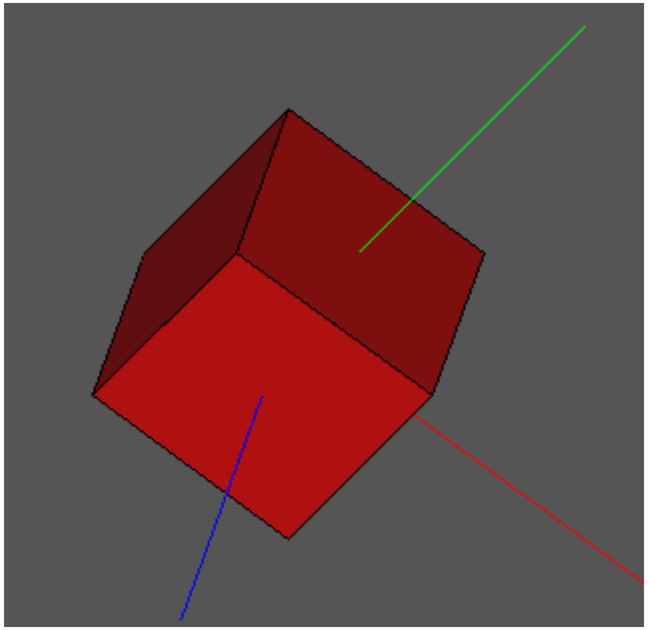
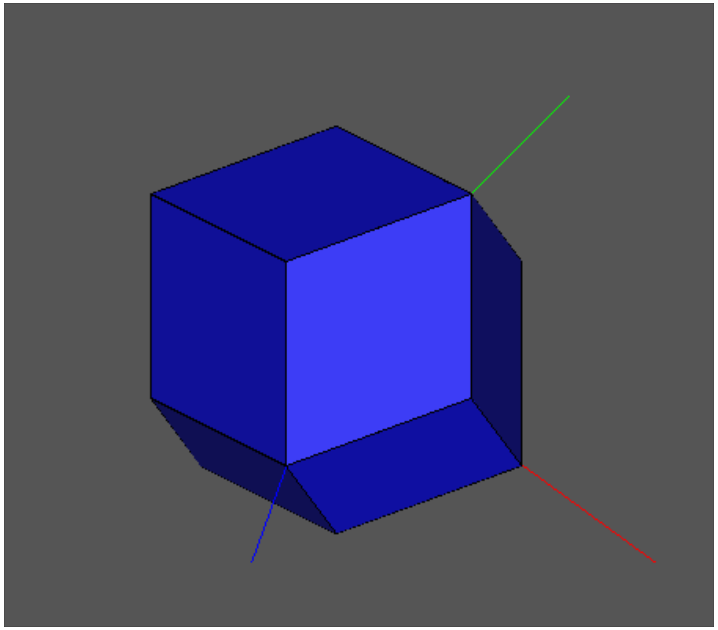
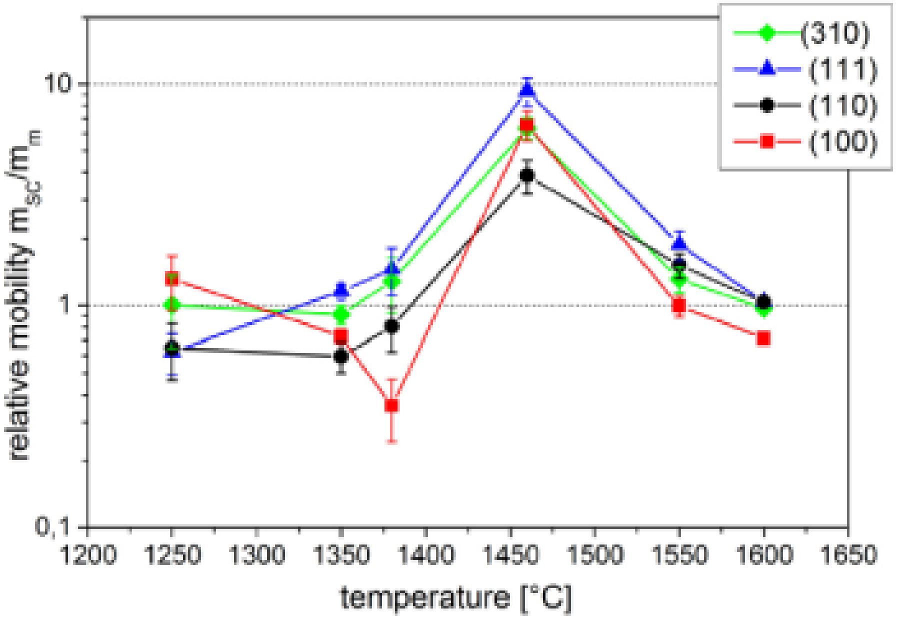
1380 °C

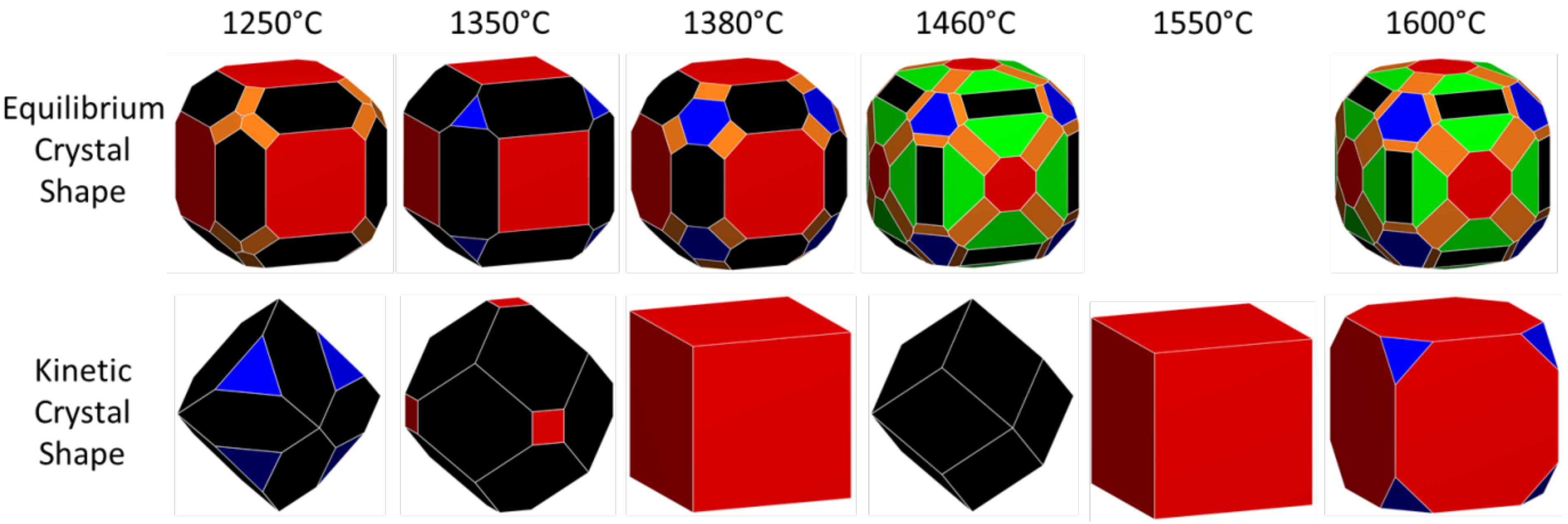


1460 °C



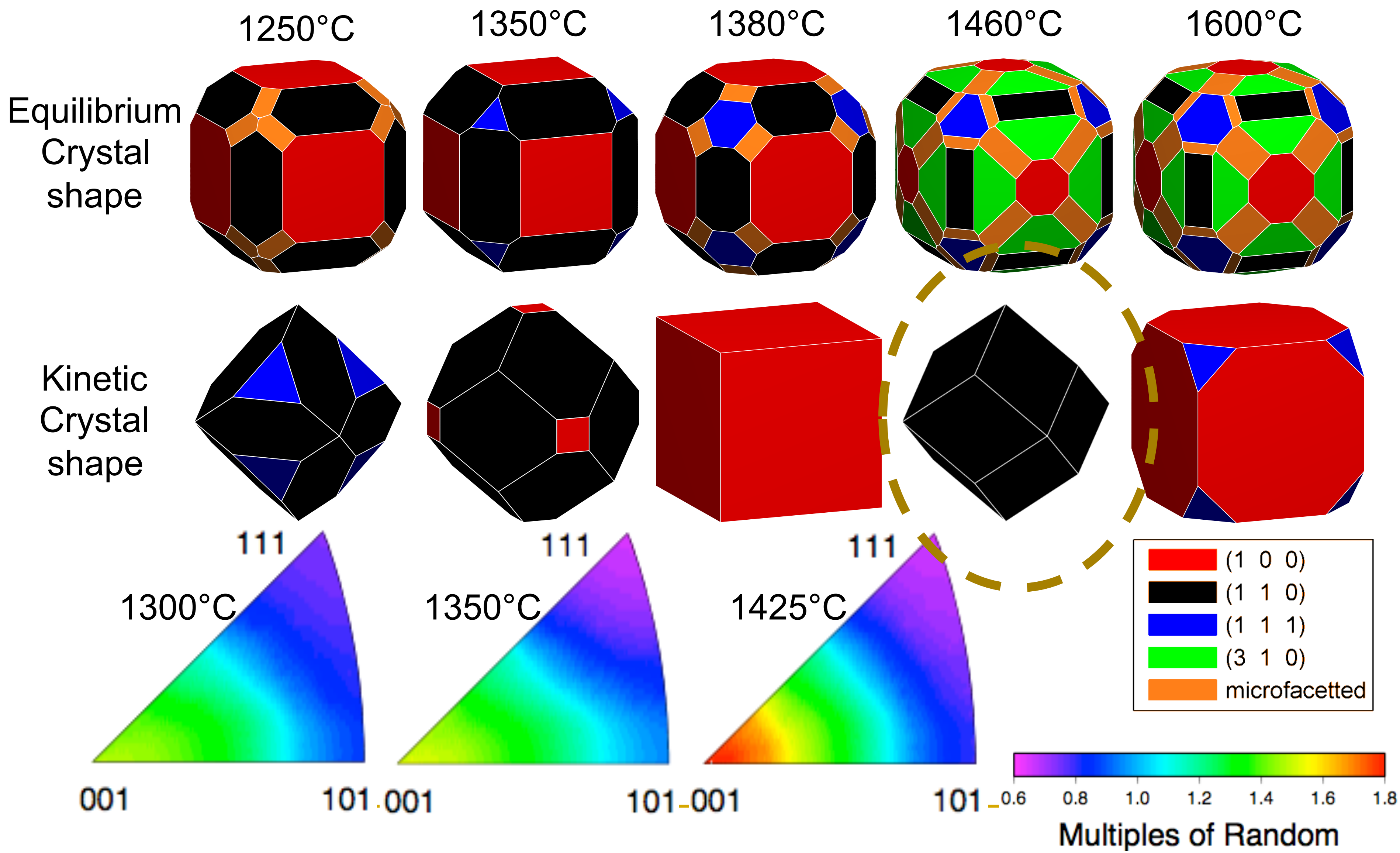


Equilibrium and kinetic crystal shape



- Leads to specific predictions for shapes of AGGs
- Explains the changes in the GBPD with temperature

Equilibrium and kinetic crystal shape



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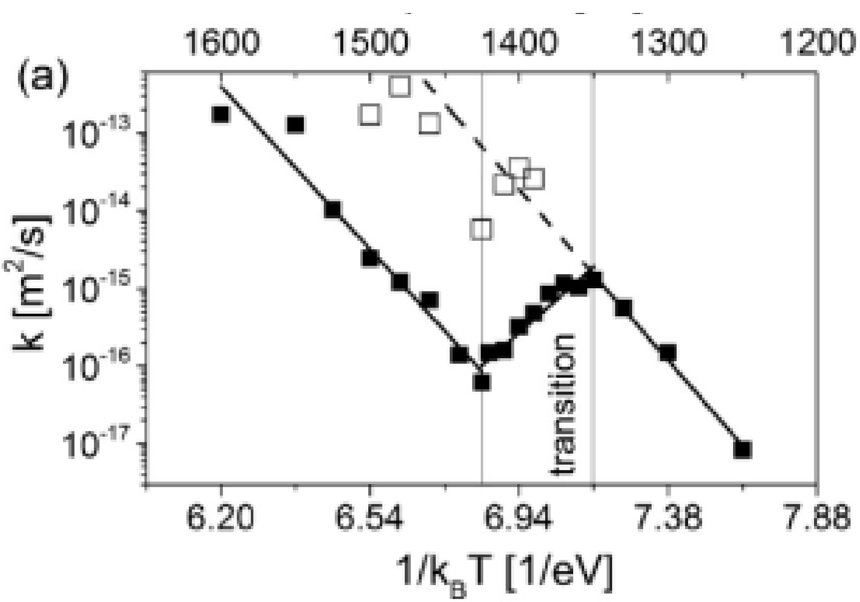

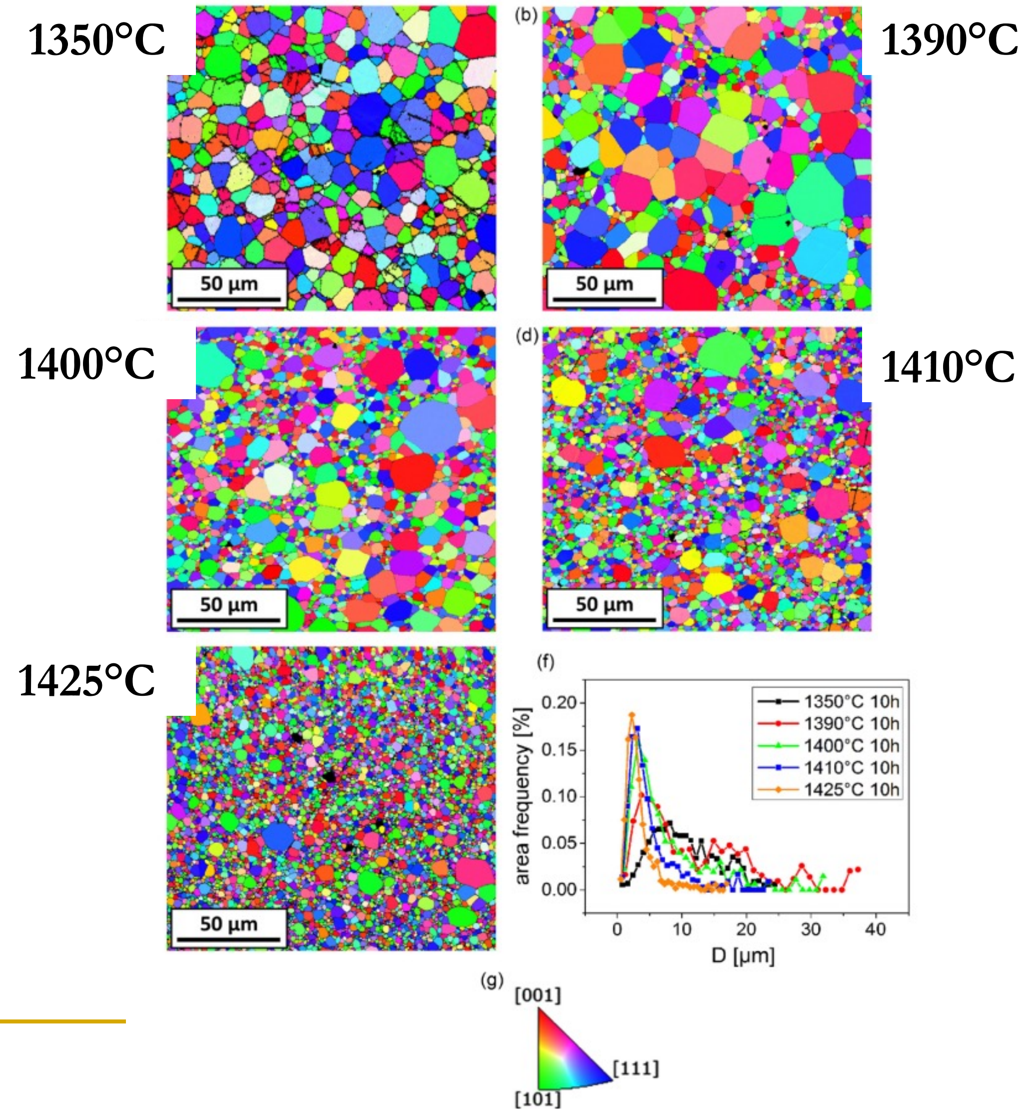

Superposition of Grain Growth and Single Crystal Growth Rate Data

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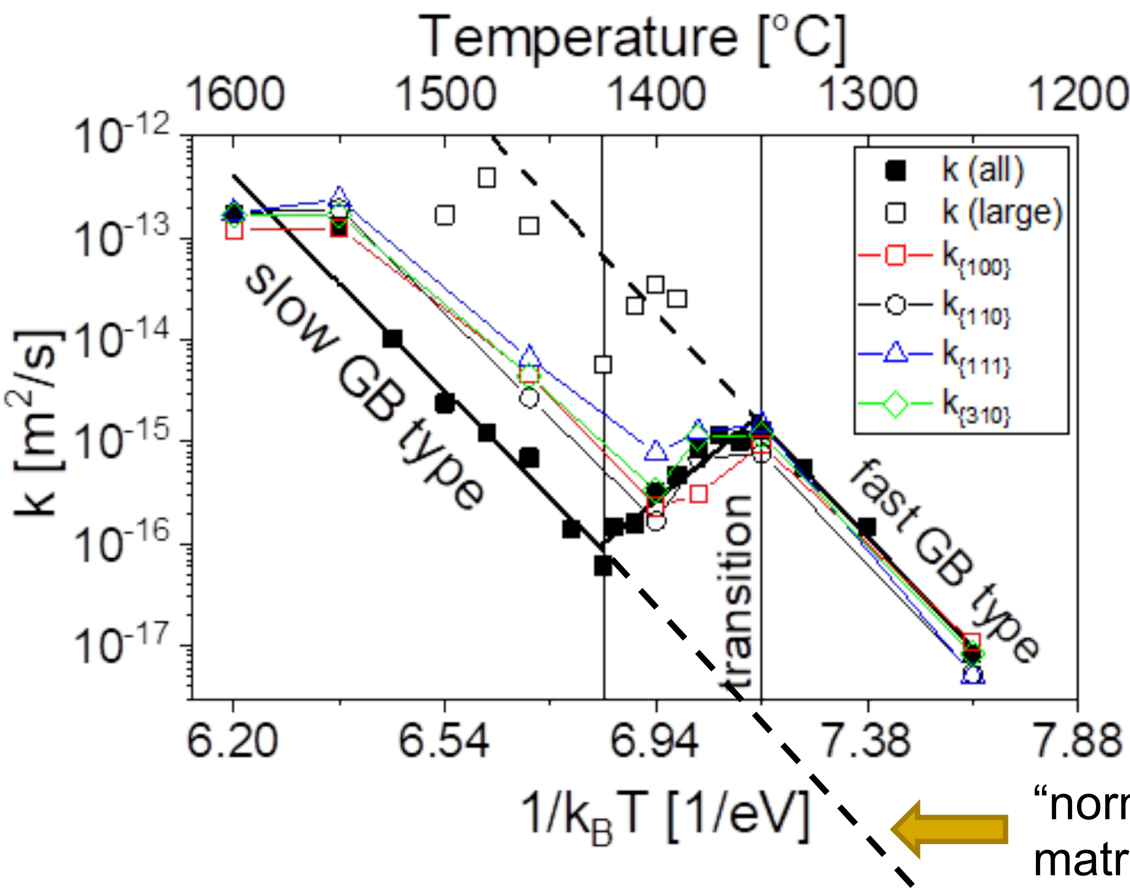
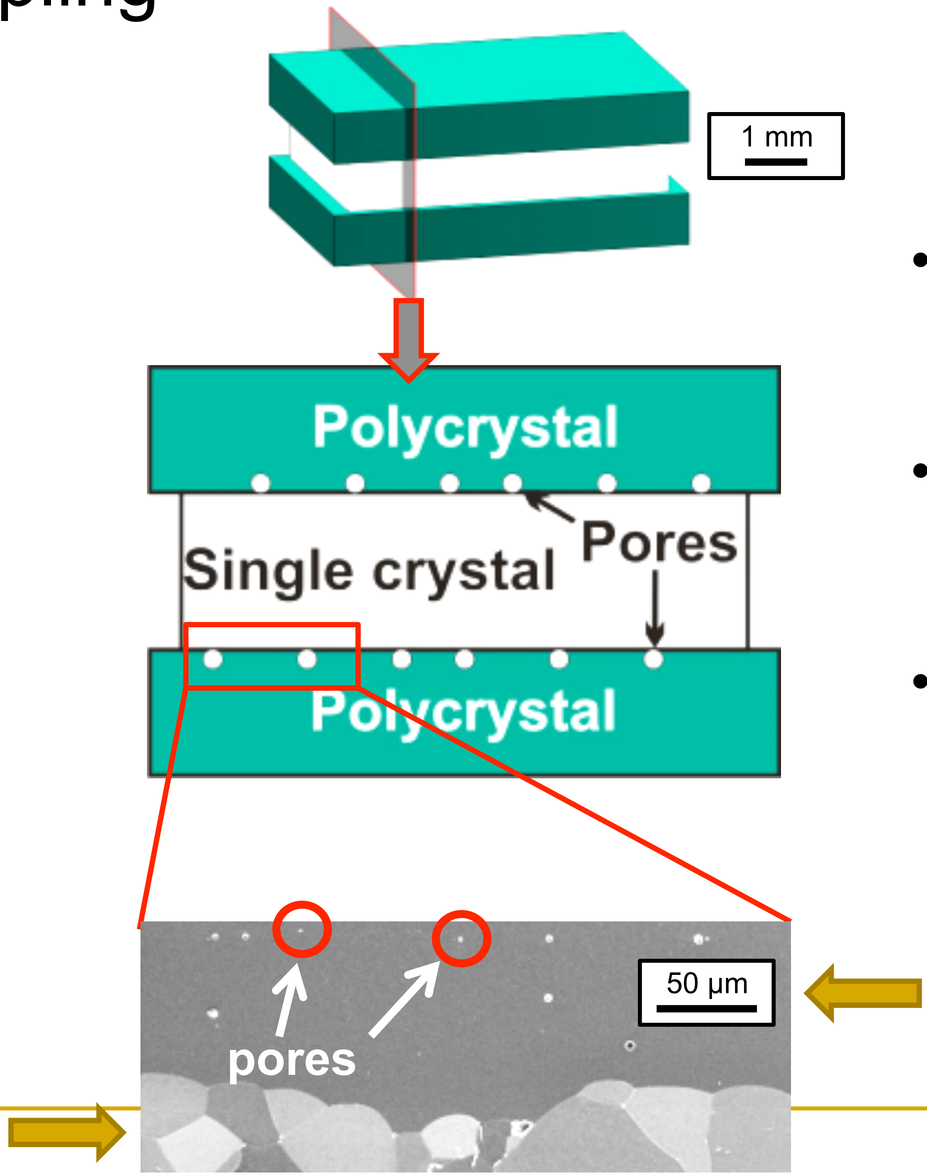


- Number of fast GB type grains decreases with increasing T
- Depends on number of abnormal grains and when they impinge

"normal" slow matrix grains

Cause for Stagnation and Possible Coupling



- No sign of impurity segregation
- Stagnation due to coupling?
- Apply shear along single crystal-polycrystal boundary

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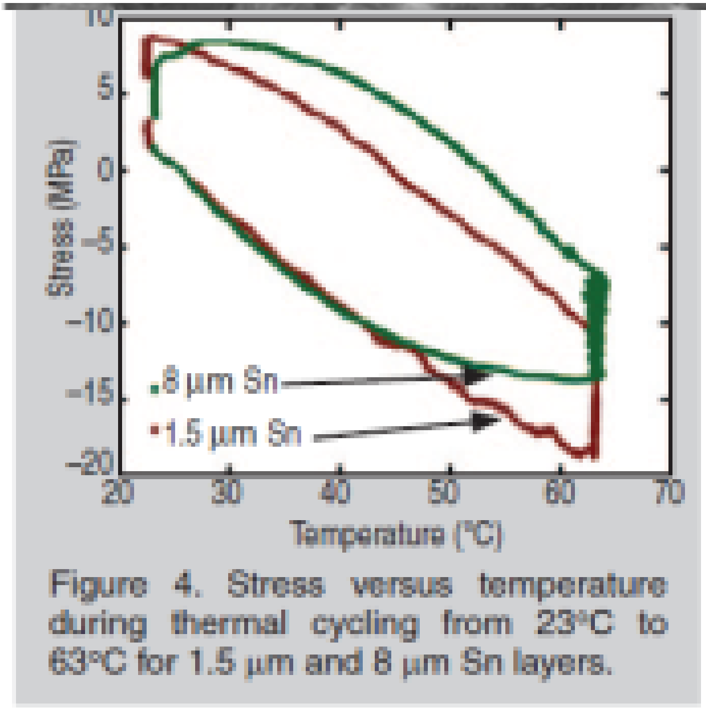
Heterogeneous Stress Relaxation in Tin Films: Whiskers, Hillocks, and Beyond

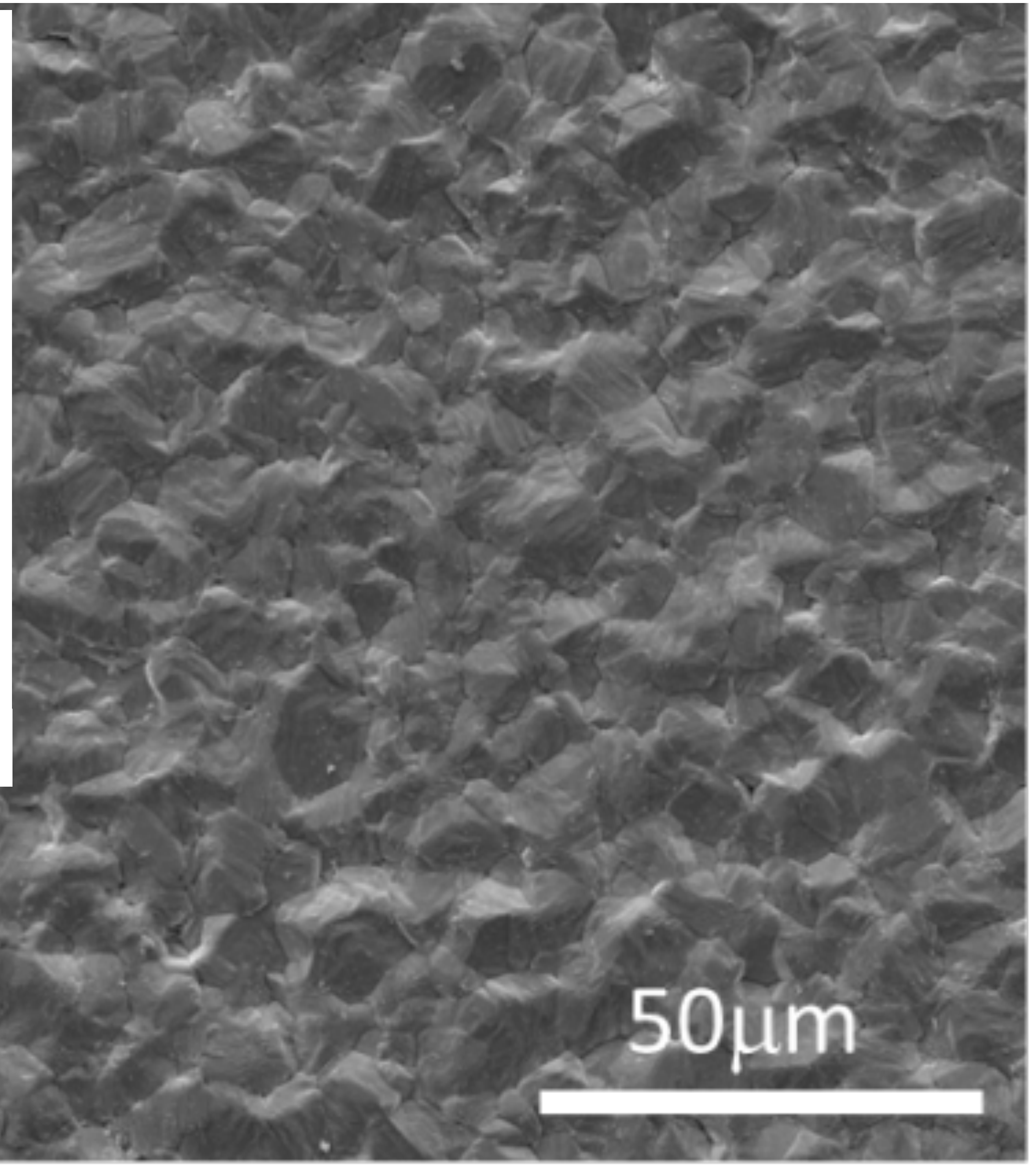
Carol Handwerker, John Blendell, Ying Wang,
Wei-Hsun Chen, Pylin Sarobol, John Koppes,
Aaron Pedigo, Congying Wang, Xi Chen,
Byung-Gil Yoo[1], Bastian Phillipi[2],
Oliver Kraft[1], Gerhard Dehm[2], Maureen Williams[3]
Dominique Chatain[4], Stephano Curriotto[4]

Purdue University,
[1] Karlsruhe Institute of Technology,
[2] Max Planck Institut für Eisenforschung,
[3] National Institute of Standards and Technology
[4]CNRS/CiNAM – Marseille

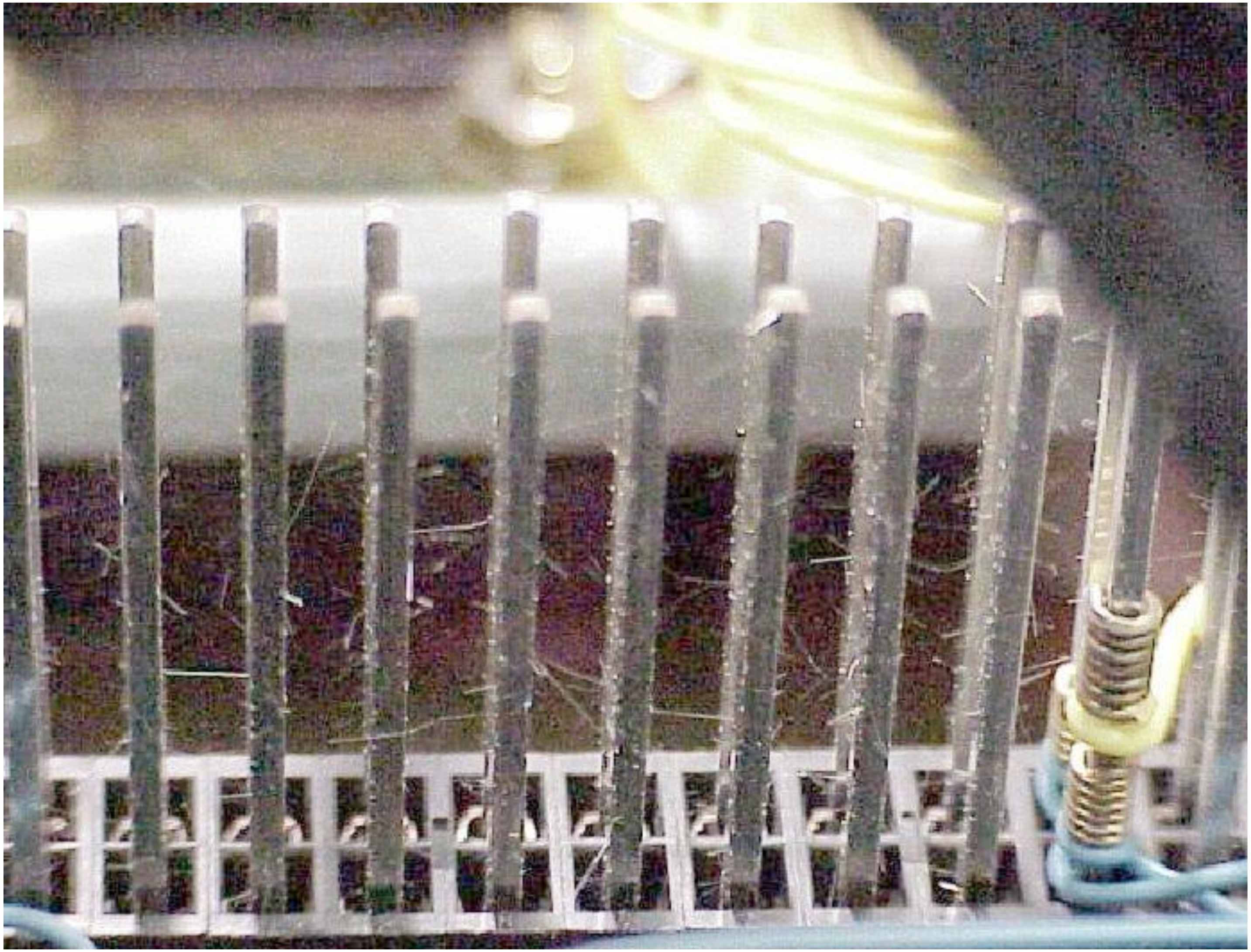
Tin Film – approx. 6 μm thick



Chason, Jadhav, Pei, JOM, 2011

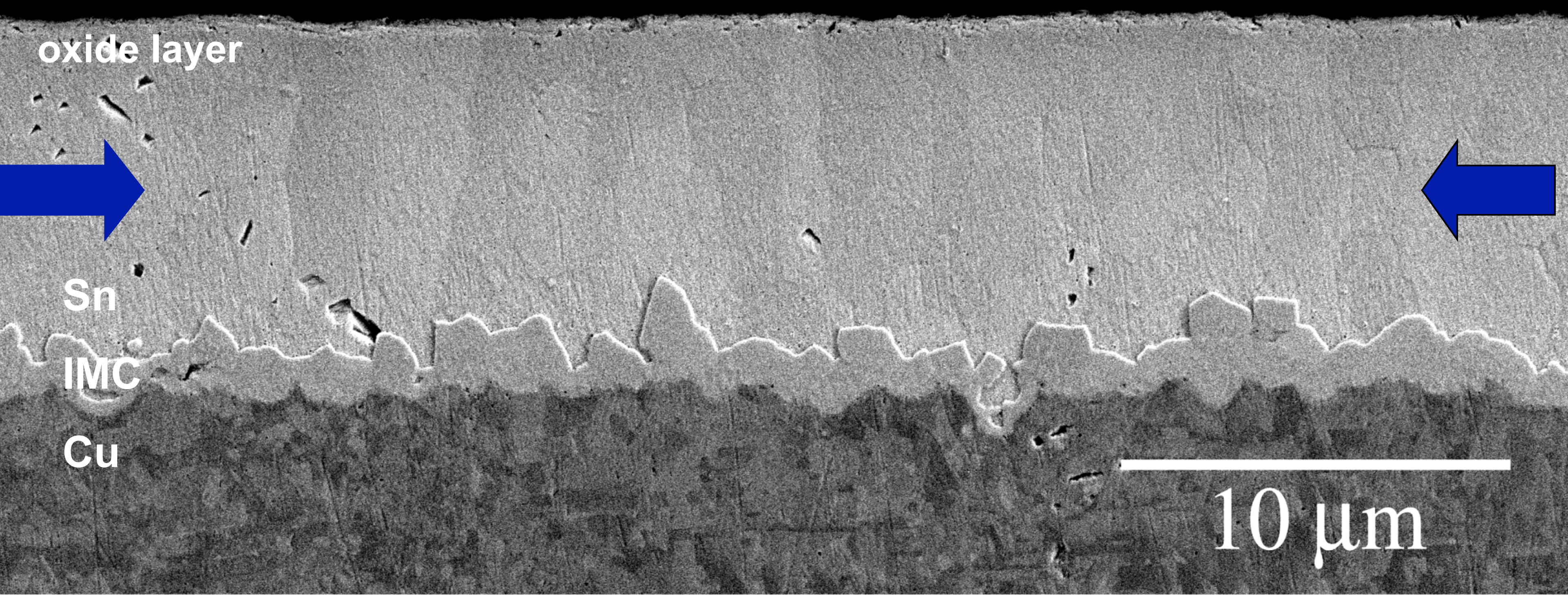


Tin Whisker Formation in Electronic Circuits

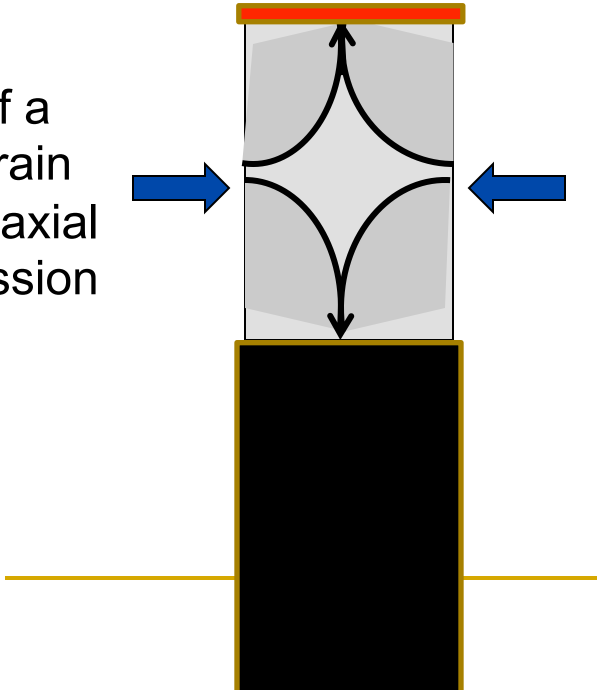


Tin-Plated Connector Pins after 10 years
Courtesy of NASA - Goddard Space Flight Center

Photo from: http://nepp.nasa.gov/whisker/index.html



Creep of a single grain under biaxial compression



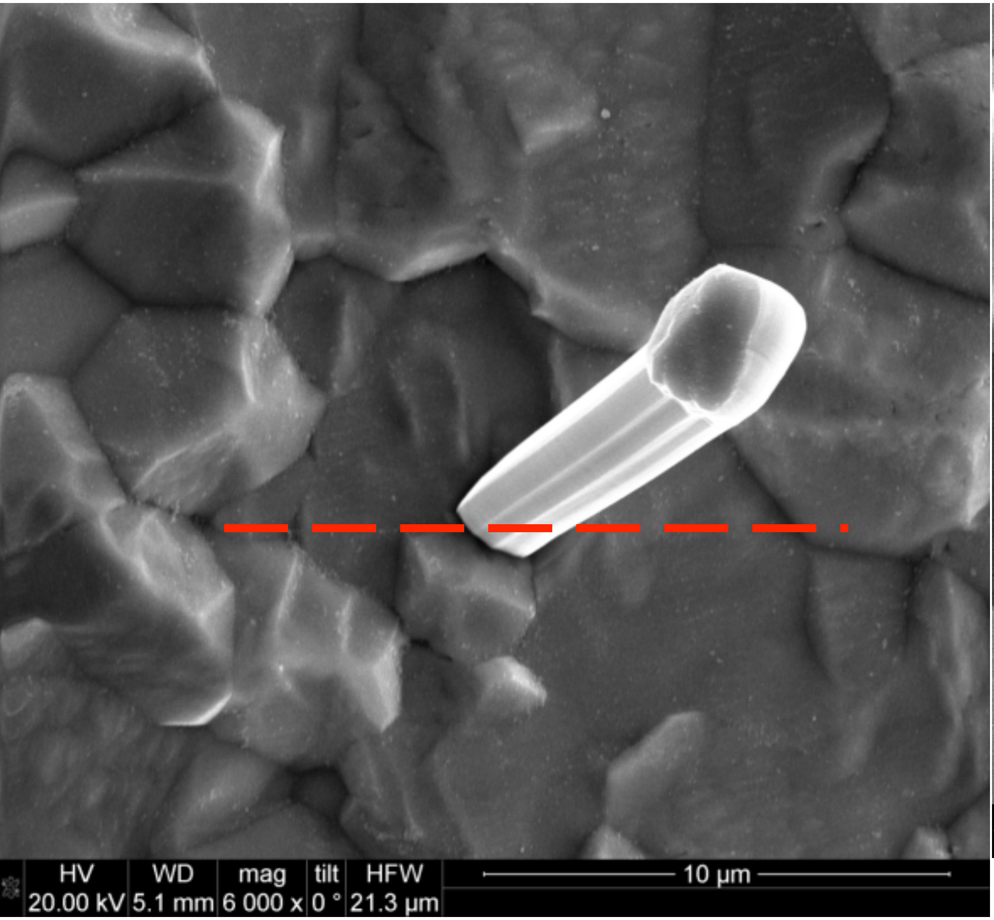
Intermetallic growth or thermal expansion mismatch creates compressive stress in film – 10 MPa (critical stress)

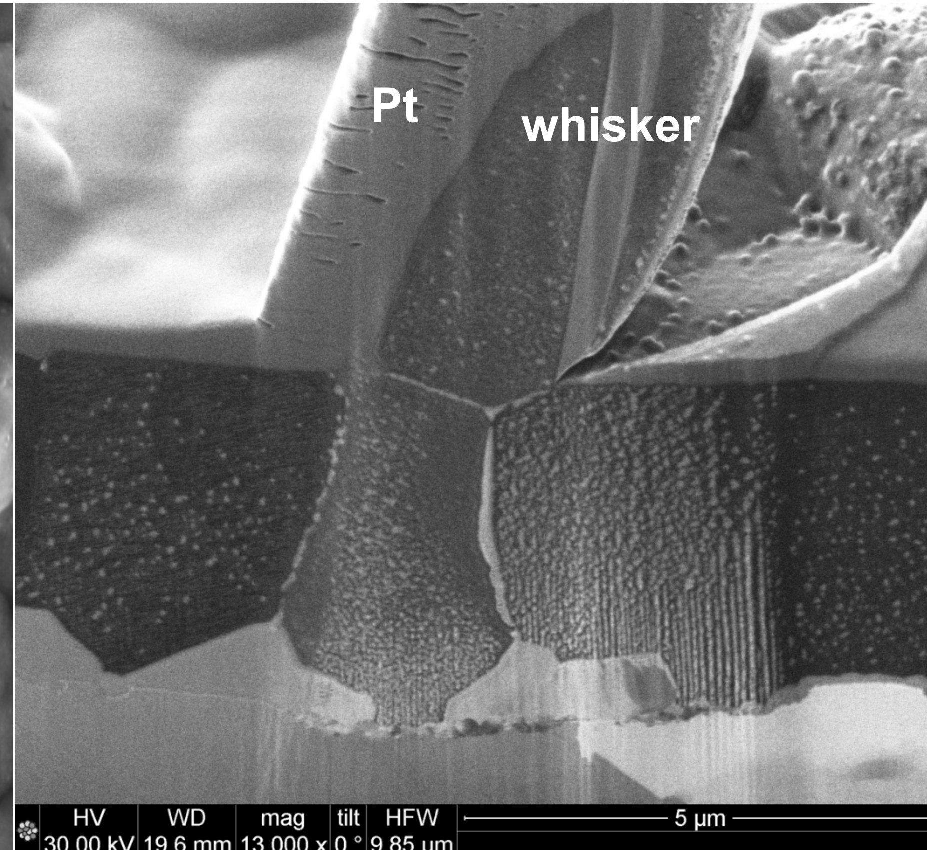
Sn @ room temperature = 60% $T_{melting}$

Creep
Coble – gb diffusion controlled
Nabarro Herring – lattice diffusion controlled

But oxide and substrate suppress diffusional creep

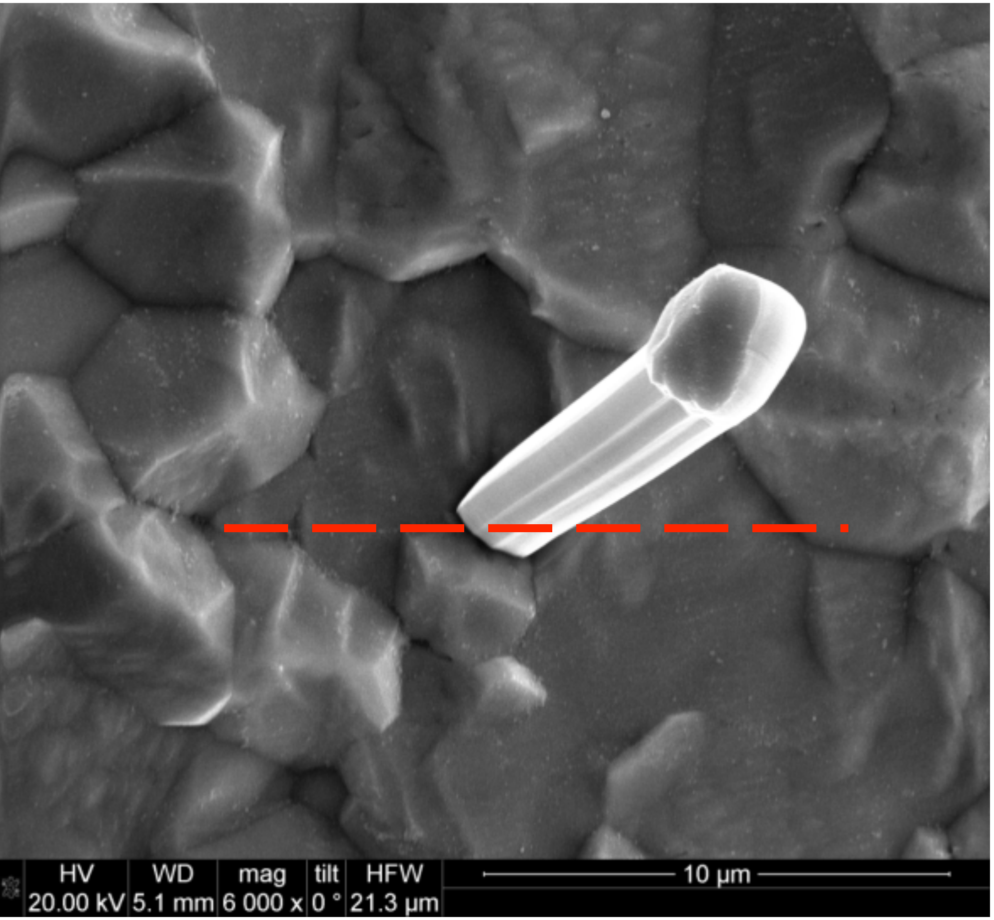
Tin Whisker Morphology – Importance of Shallow Surface Grains



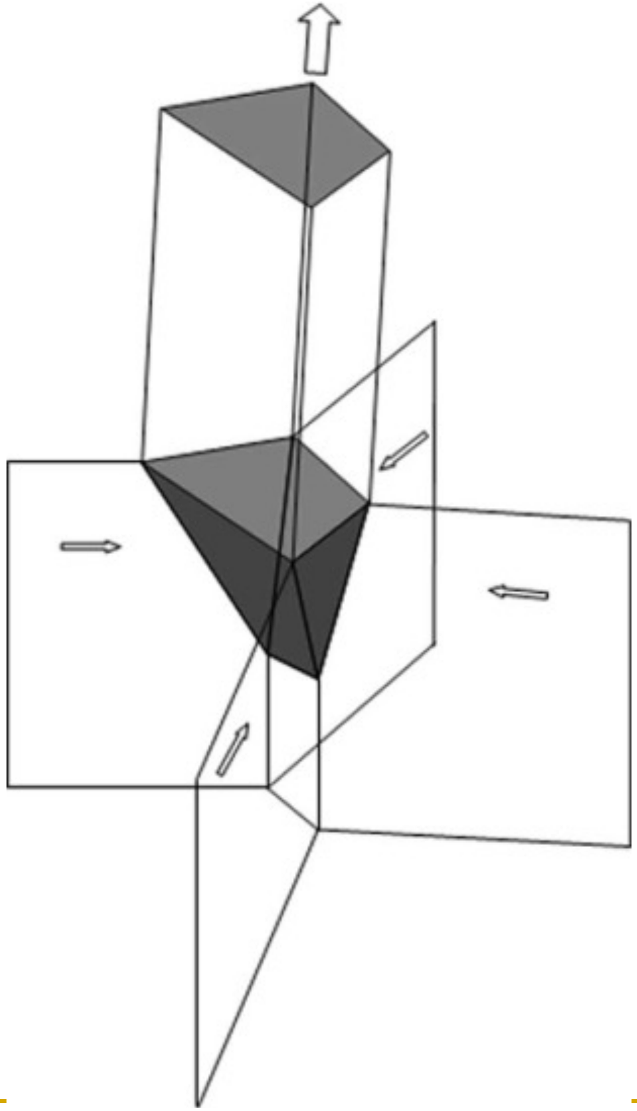


Ga FIB cut through whisker and film with protective Pt overlayer

Tin Whisker Morphology – Importance of Shallow Surface Grains

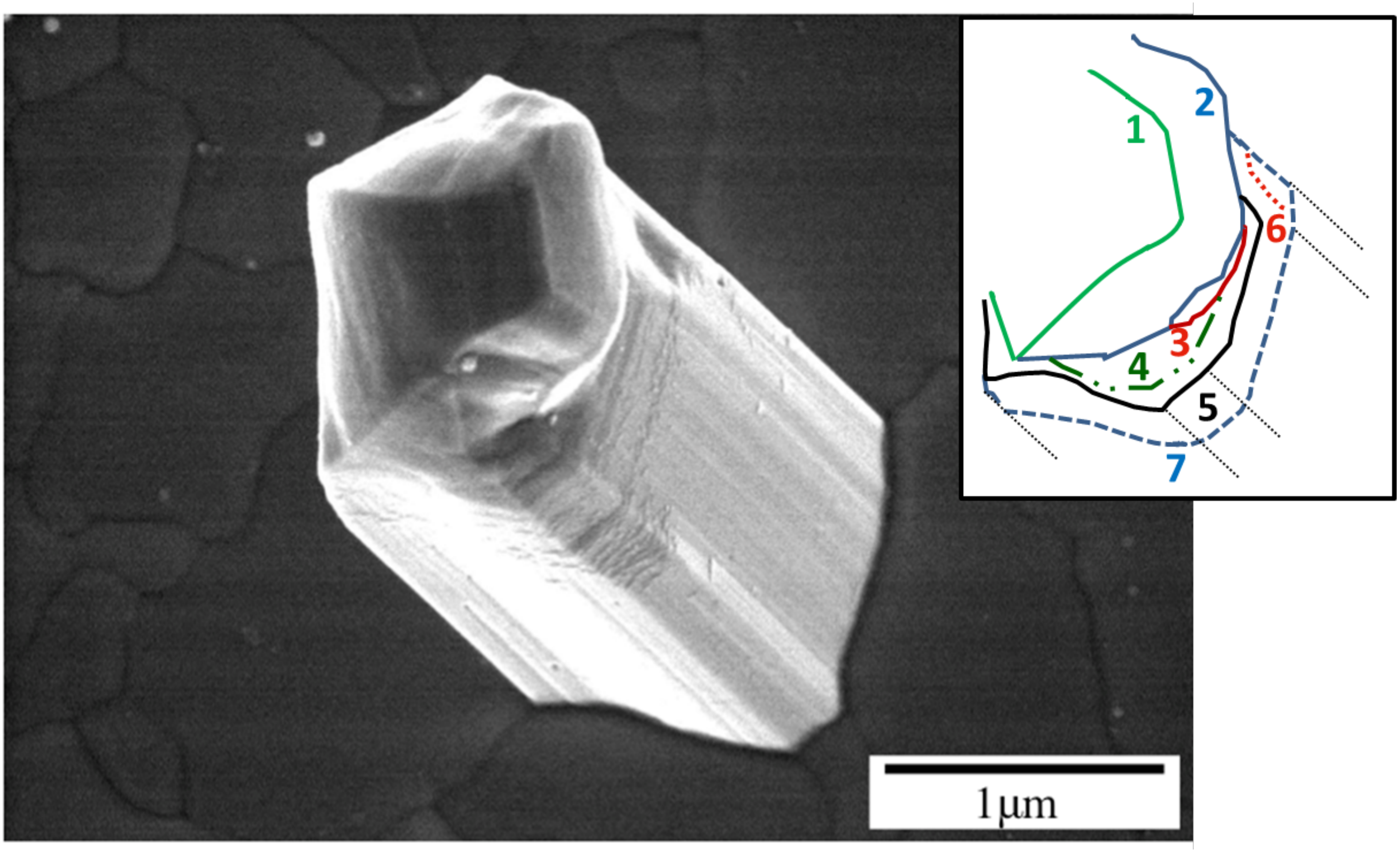


Acta Materialia 53 (2005) 5033–5050



Boettinger, et al. Acta Materialia 53 (2005) 5033

Whisker Morphology – Growth Processes



P. Sarobol, Y. Wang, W.H. Chen, A.E. Pedigo, J.P. Koppes, J.E. Blendell, and C.A. Handwerker, “A Predictive Model for Whisker Formation Based on Local Microstructure and Grain Boundary Properties,” JOM, 2013;65:1350-1361.

Hillock Morphology – Growth Processes

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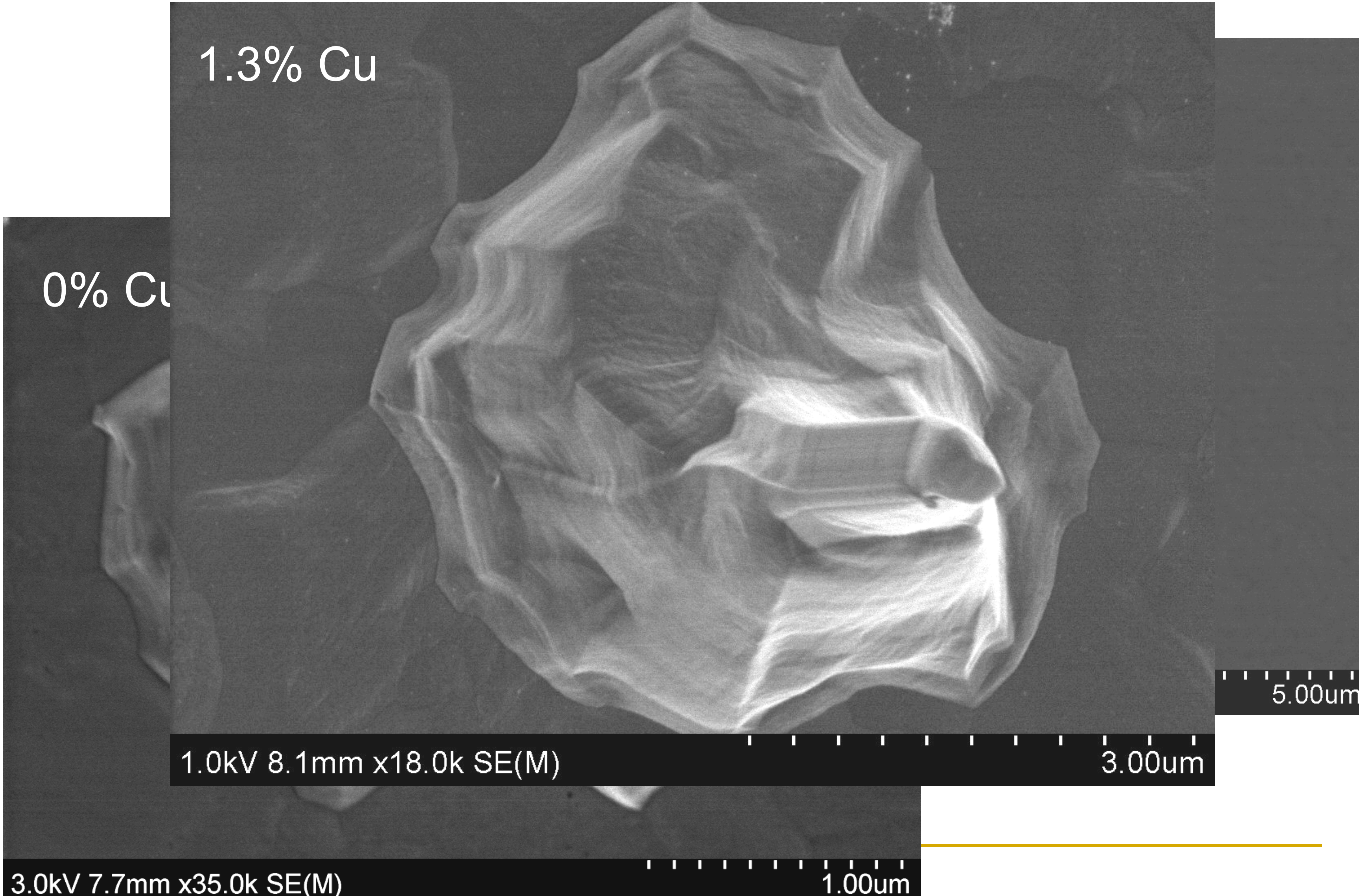
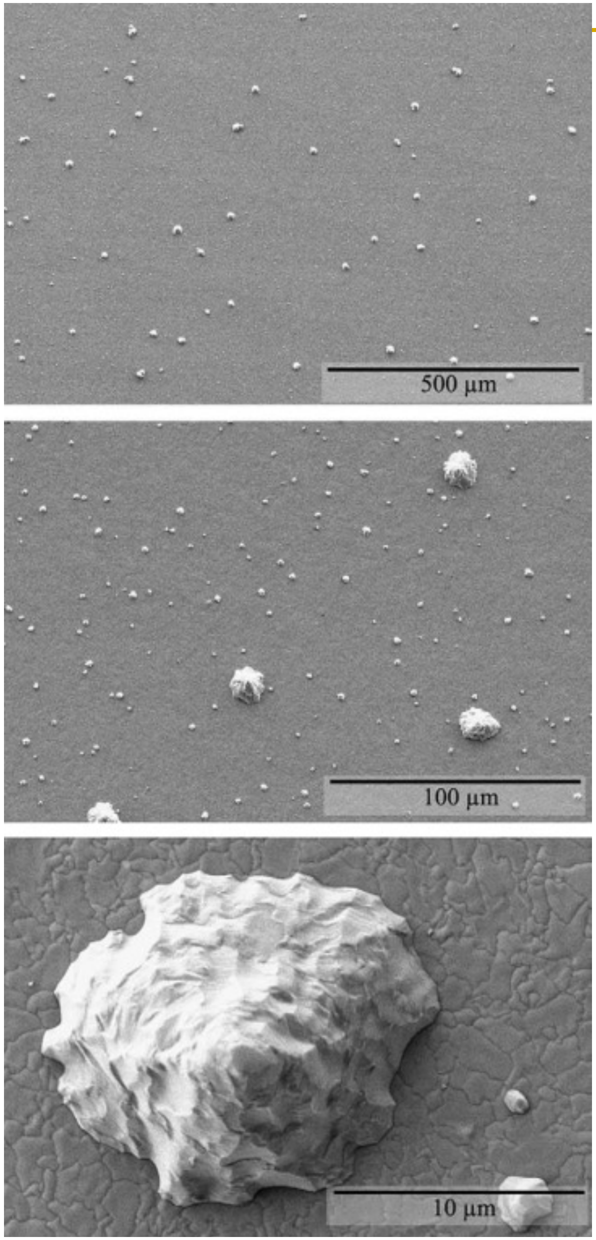
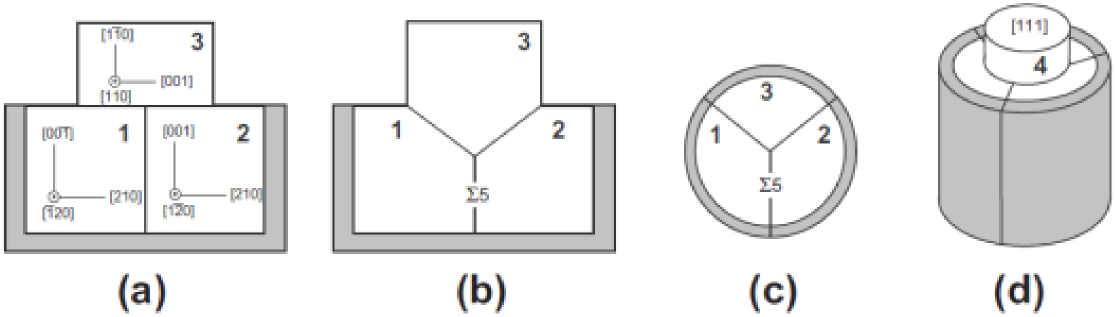


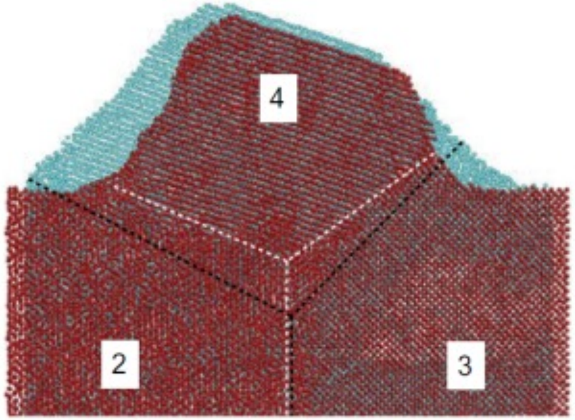


Atomistic simulation of hillock growth

T. Frolov [a,*], W.J. Boettinger [b], Y. Mishin [a]

Acta Materialia 58 (2010) 5471–5480



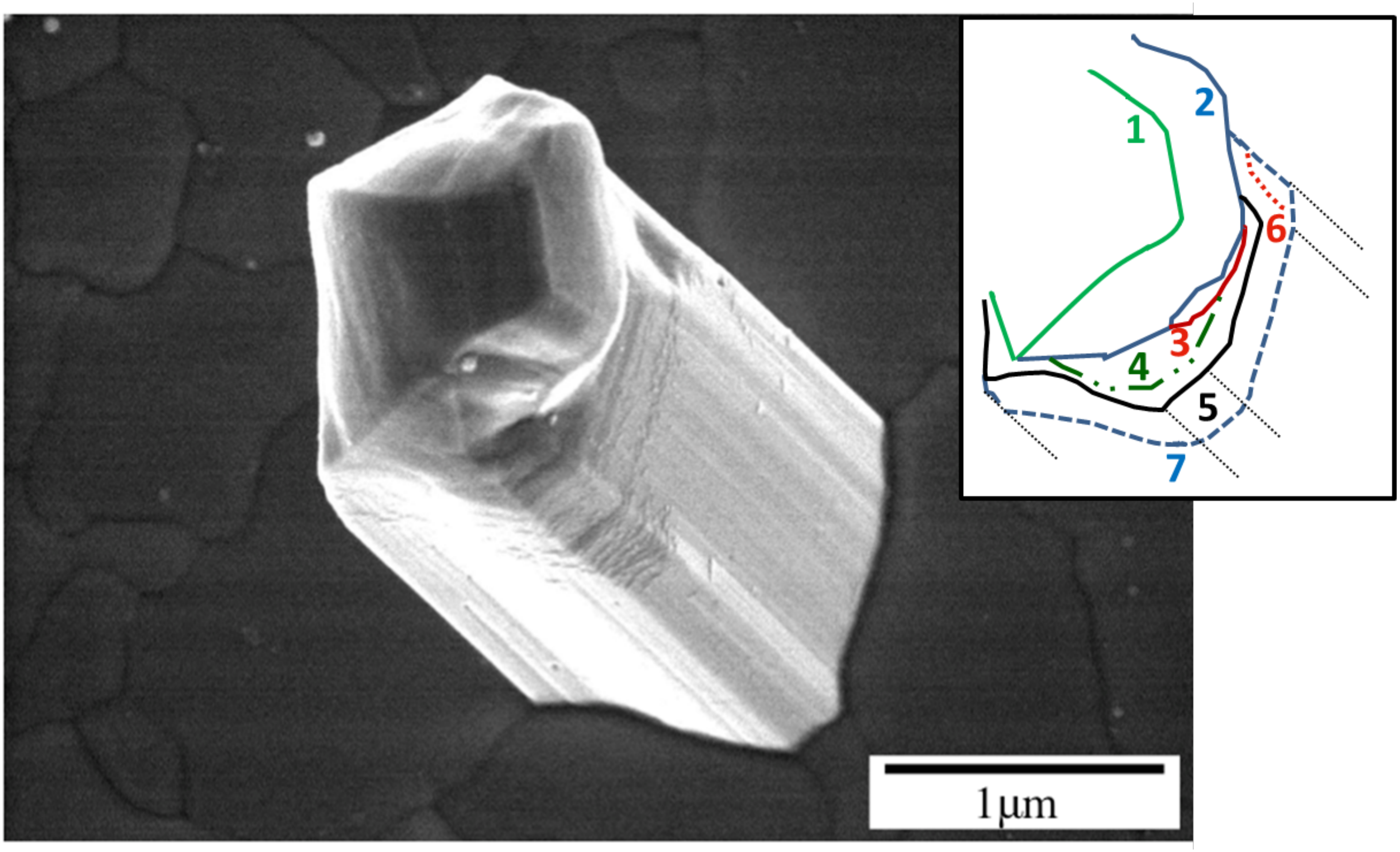


MD – One quad junction configuration

- Upward motion
- Migration
- Grain rotation
- Twinning
- Shear coupling
- Formation of special boundary
- Stacking faults

Boettinger, et al. Acta Materialia 53 (2005) 5033

Whisker Morphology – Growth Processes



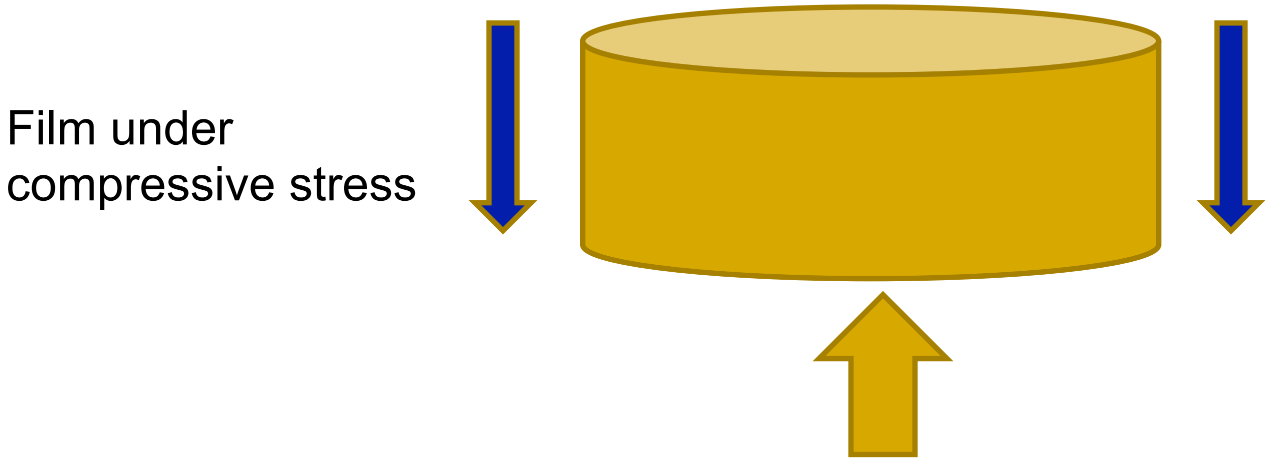
P. Sarobol, Y. Wang, W.H. Chen, A.E. Pedigo, J.P. Koppes, J.E. Blendell, and C.A. Handwerker, "A Predictive Model for Whisker Formation Based on Local Microstructure and Grain Boundary Properties," JOM, 2013;65:1350-1361.

Cylindrical Grain in a Thin Film

Accretion stress = σ   Sliding friction = β

For cylindrical grain of radius r and height h
in a film of thickness t

$$\sigma \pi r^2 > 2\pi r h \beta$$



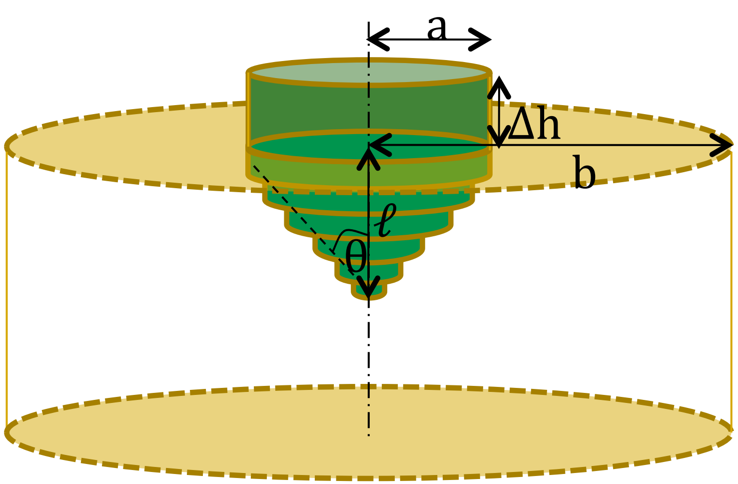
For cylindrical grains of radius r and height = film thickness t

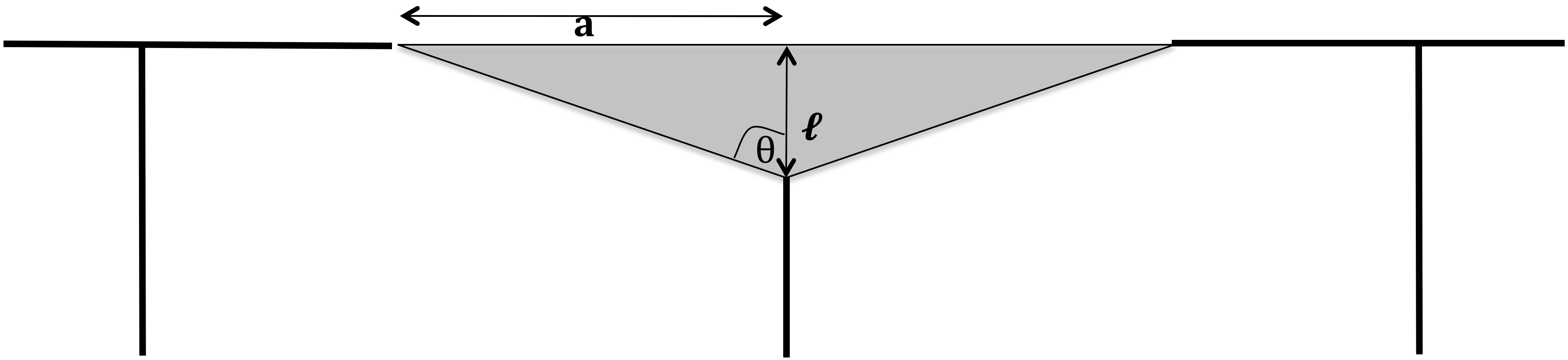
$$\sigma \pi r^2_{critical} > 2\pi r t \beta$$

$$r_{critical} > 2 \beta t / \sigma$$

P. Sarobol, J.E. Blendell and C.A. Handwerker, Acta Materialia, 2013;61:1991-2003.

Whisker Growth: GB Sliding



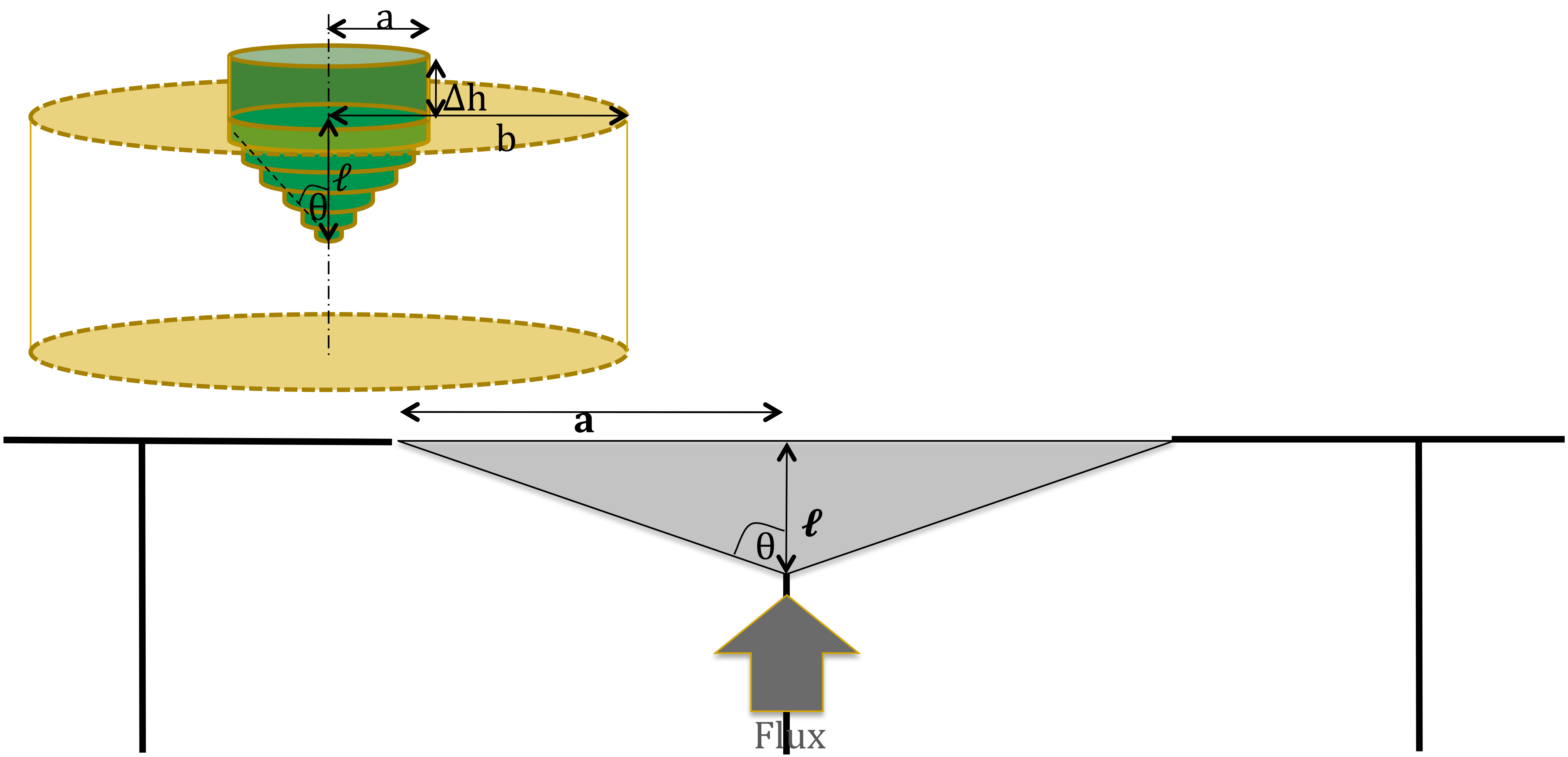


Straight Whisker Growing normal to film surface

P. Sarobol, J.E. Blendell and C.A. Handwerker, Acta Materialia, 2013;61:1991-2003.

Whisker Growth: GB Sliding

- Localized Coble creep → atomic flux to surface grain

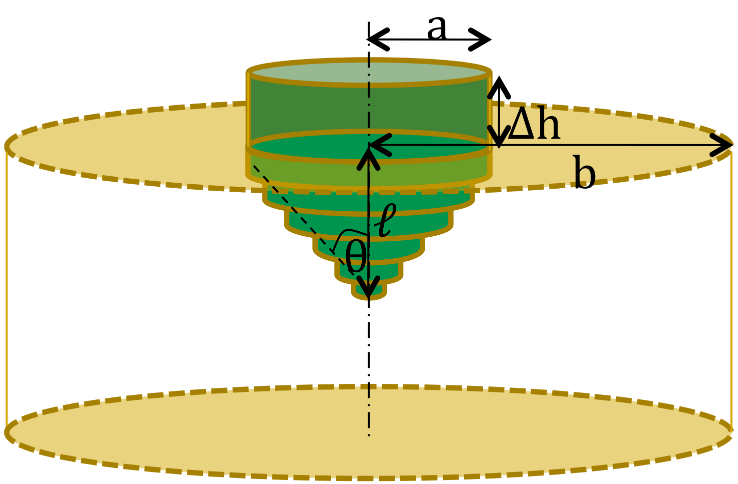


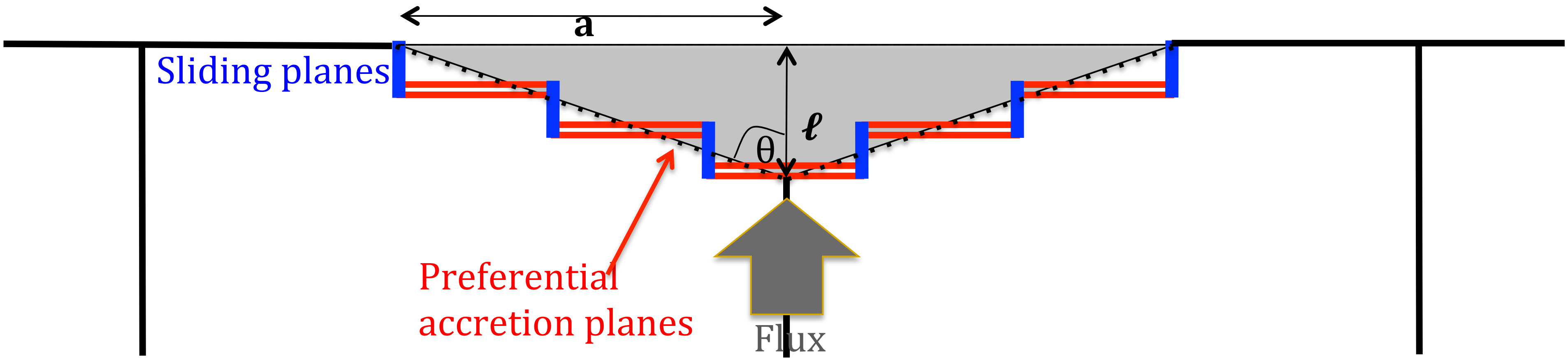
Straight Whisker Growing normal to film surface

P. Sarobol, J.E. Blendell and C.A. Handwerker, Acta Materialia, 2013;61:1991-2003.

Whisker Growth: GB Sliding

- GB are faceted (accretion and sliding components)
- Flux at accretion planes





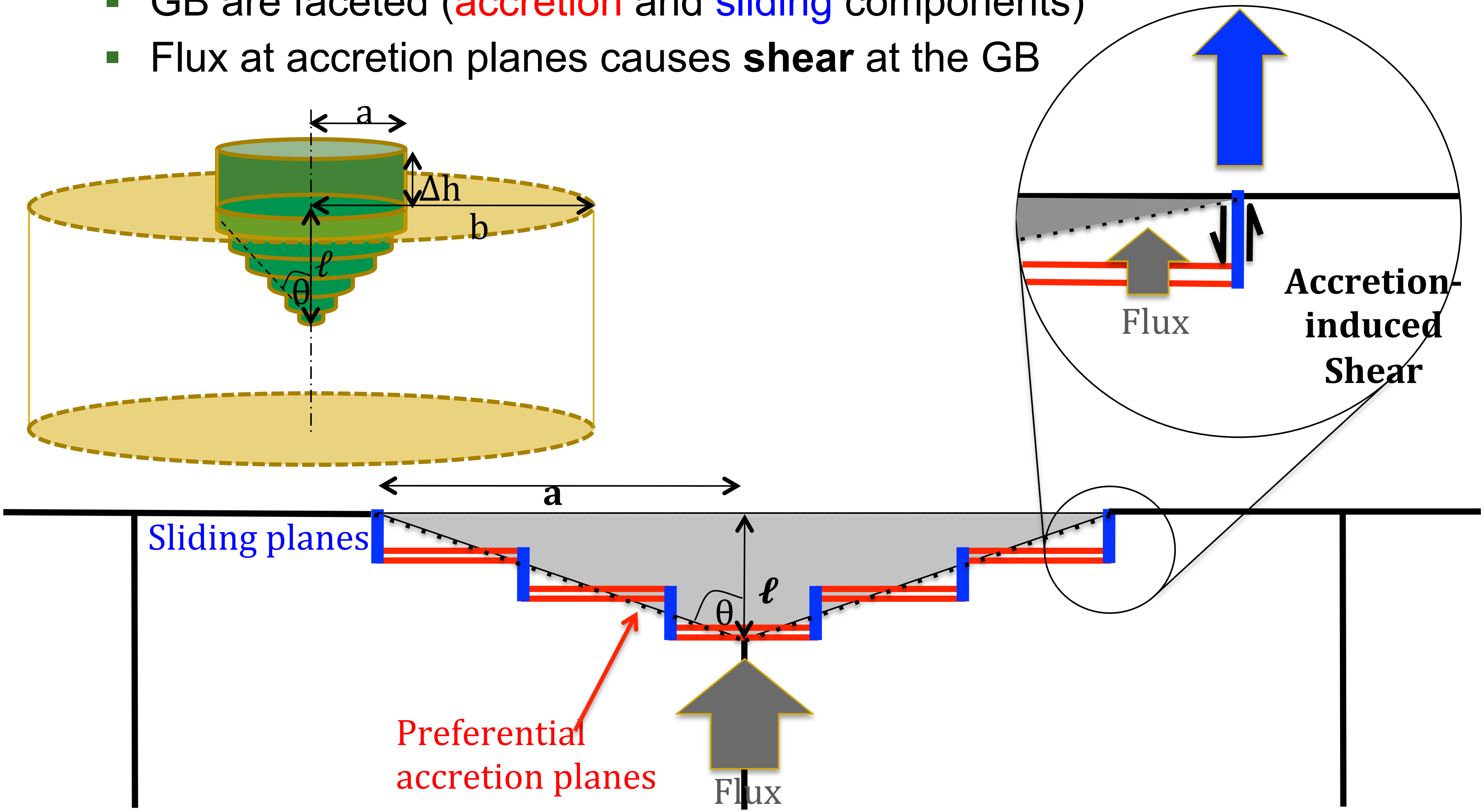
Straight Whisker Growing normal to film surface

P. Sarobol, J.E. Blendell and C.A. Handwerker, Acta Materialia, 2013;61:1991-2003.

Whisker Growth: GB Sliding

PURDUE UNIVERSITY

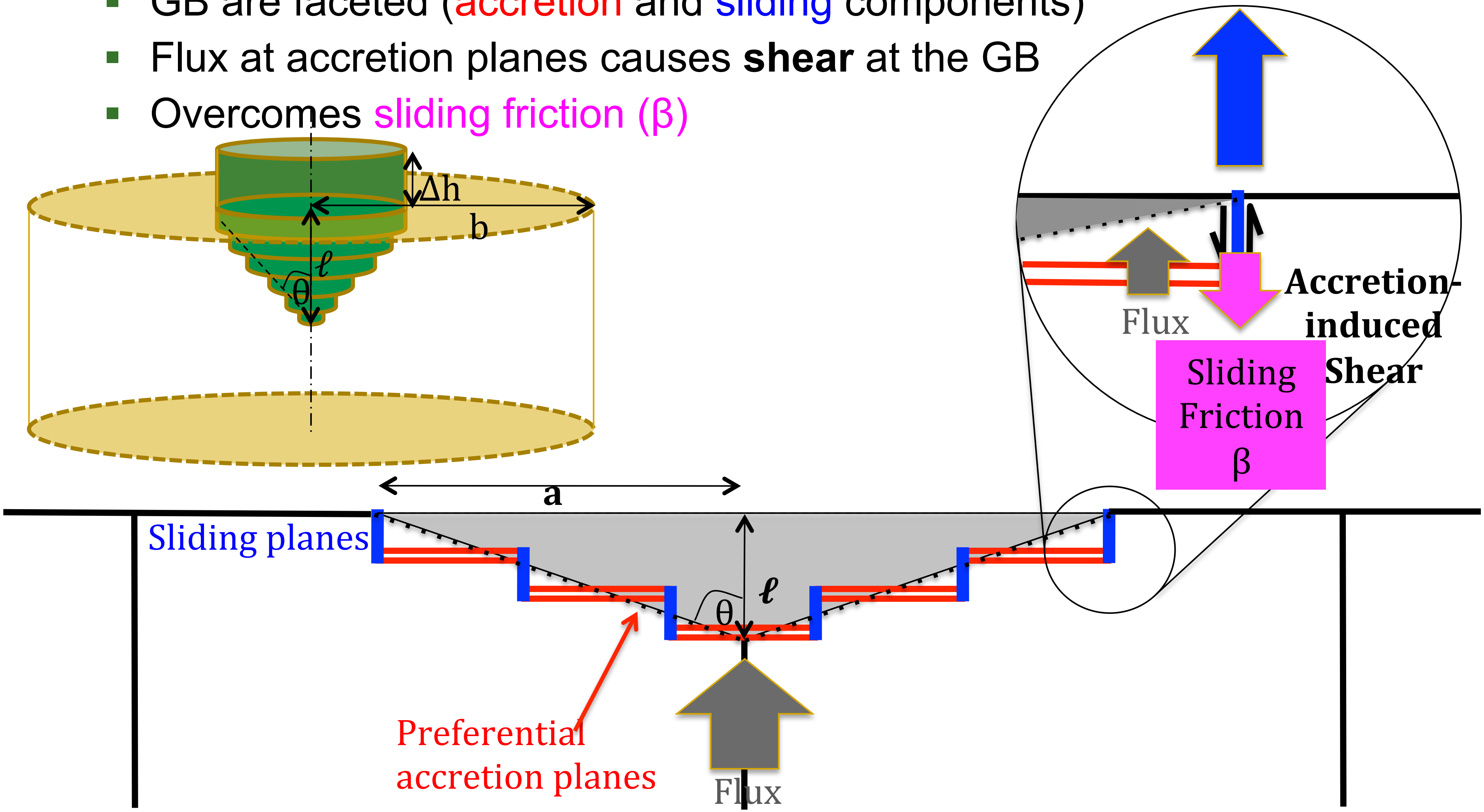
- GB are faceted (accretion and sliding components)
- Flux at accretion planes causes **shear** at the GB



Straight Whisker Growing normal to film surface

Whisker Growth: GB Sliding

- GB are faceted (accretion and sliding components)
- Flux at accretion planes causes **shear** at the GB
- Overcomes sliding friction (β)



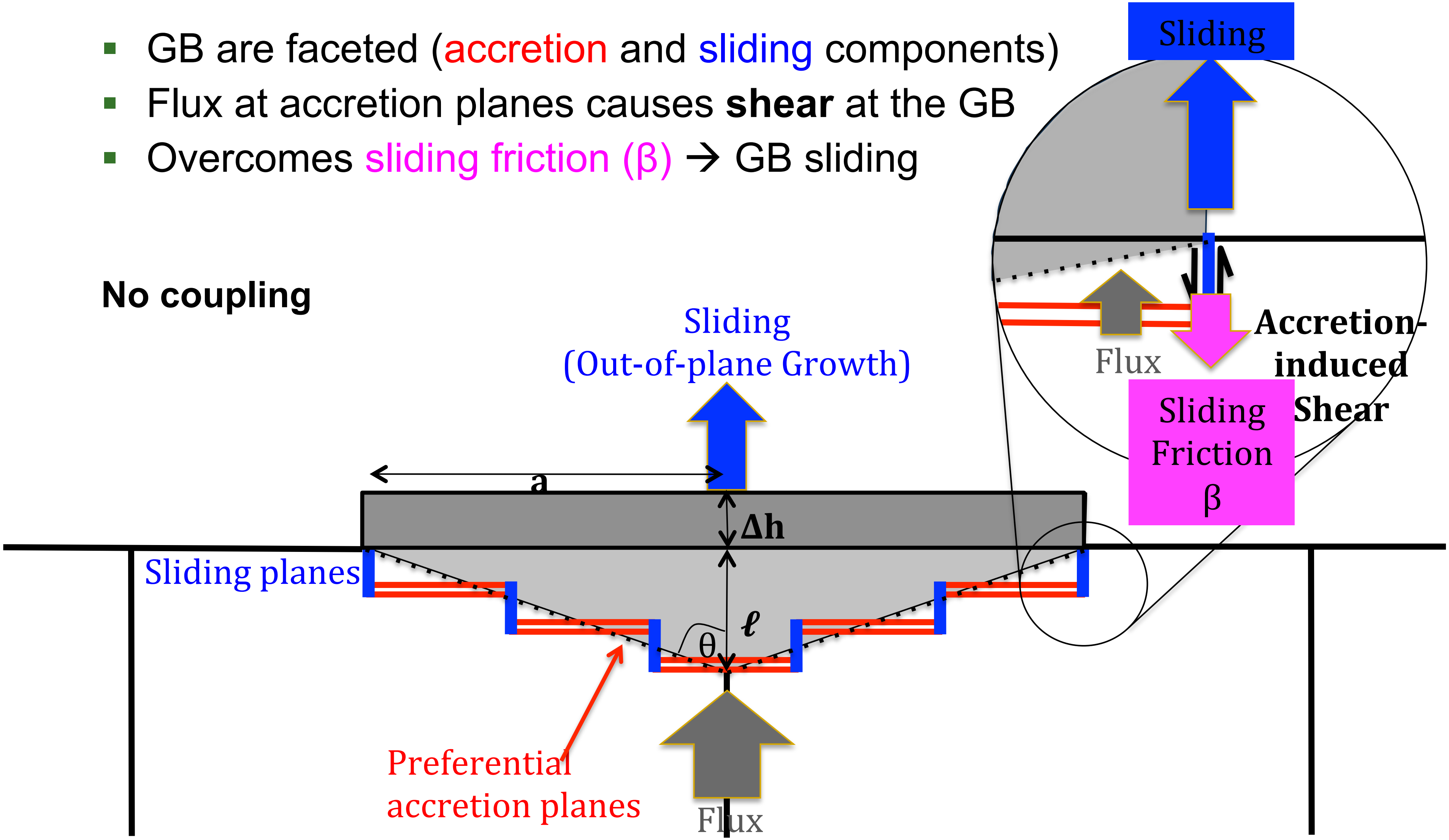
Straight Whisker Growing normal to film surface

P. Sarobol, J.E. Blendell and C.A. Handwerker, Acta Materialia, 2013;61:1991-2003.

Whisker Growth: GB Sliding

- GB are faceted (accretion and sliding components)
- Flux at accretion planes causes **shear** at the GB
- Overcomes sliding friction (β) → GB sliding

No coupling



Straight Whisker Growing normal to film surface

Whisker Growth: GB Sliding

- GB are faceted (accretion and sliding components)
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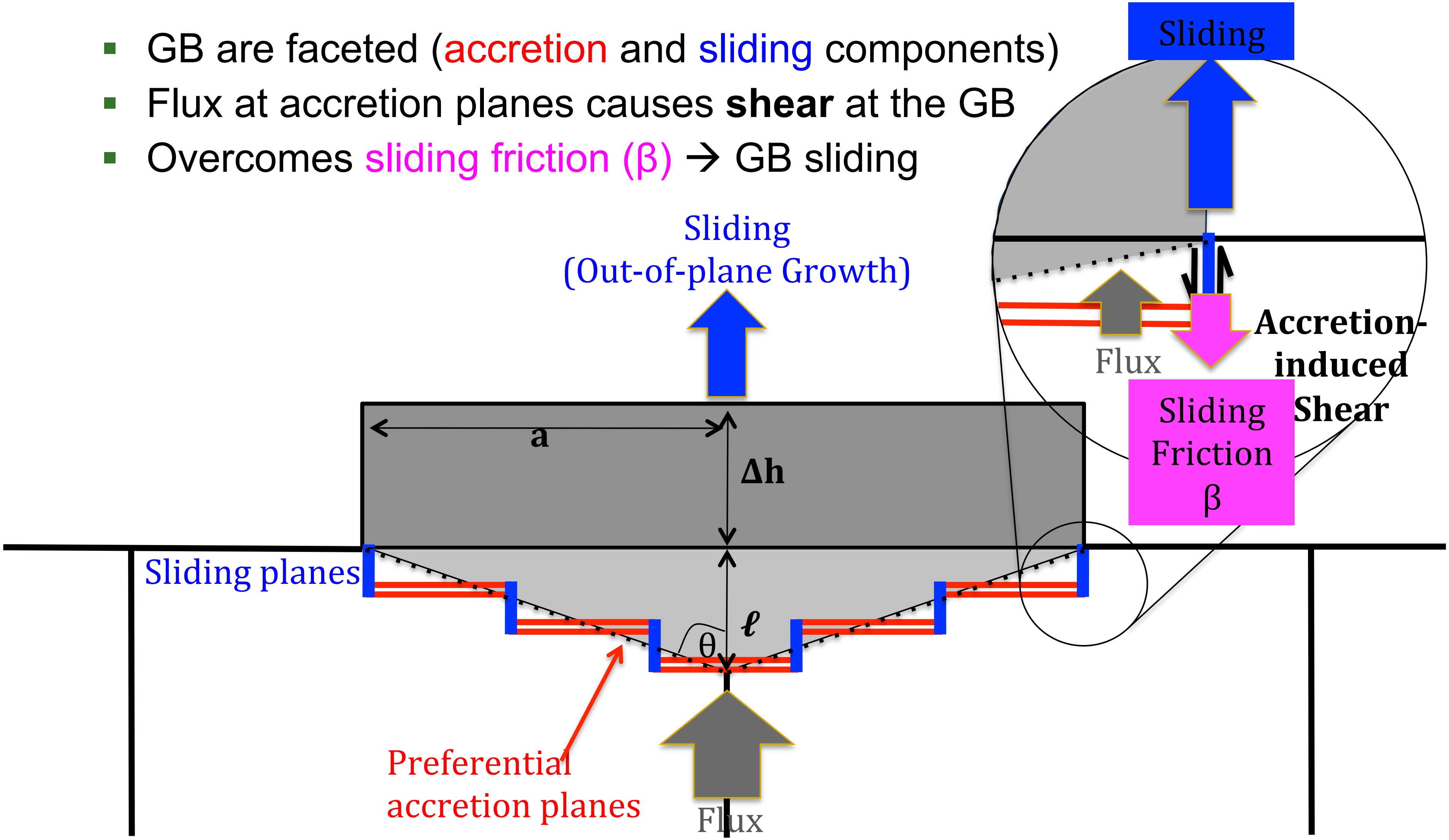


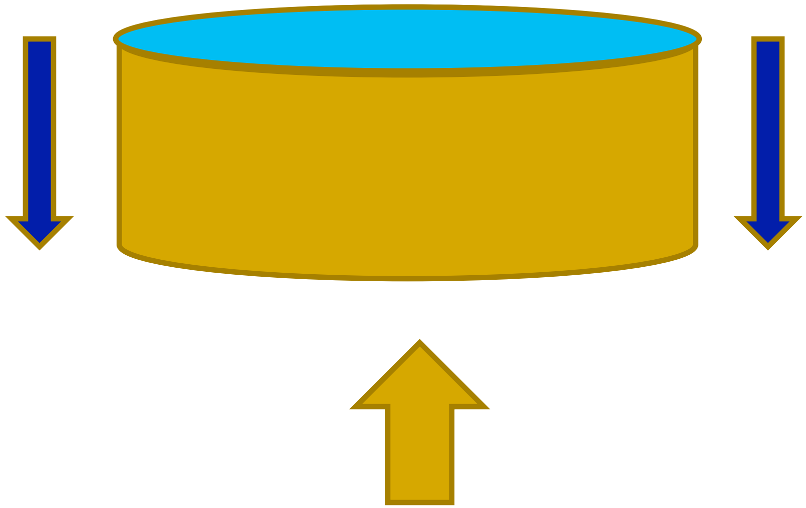
Straight Whisker Growing normal to film surface

Cylindrical Grain in a Thin Film with a Capping Oxide Layer

Accretion stress = σ Sliding friction = β Oxide fracture stress = σ_{oxide}

For cylinder of radius r and height h in a film of thickness t
capped by an **oxide of thickness x**

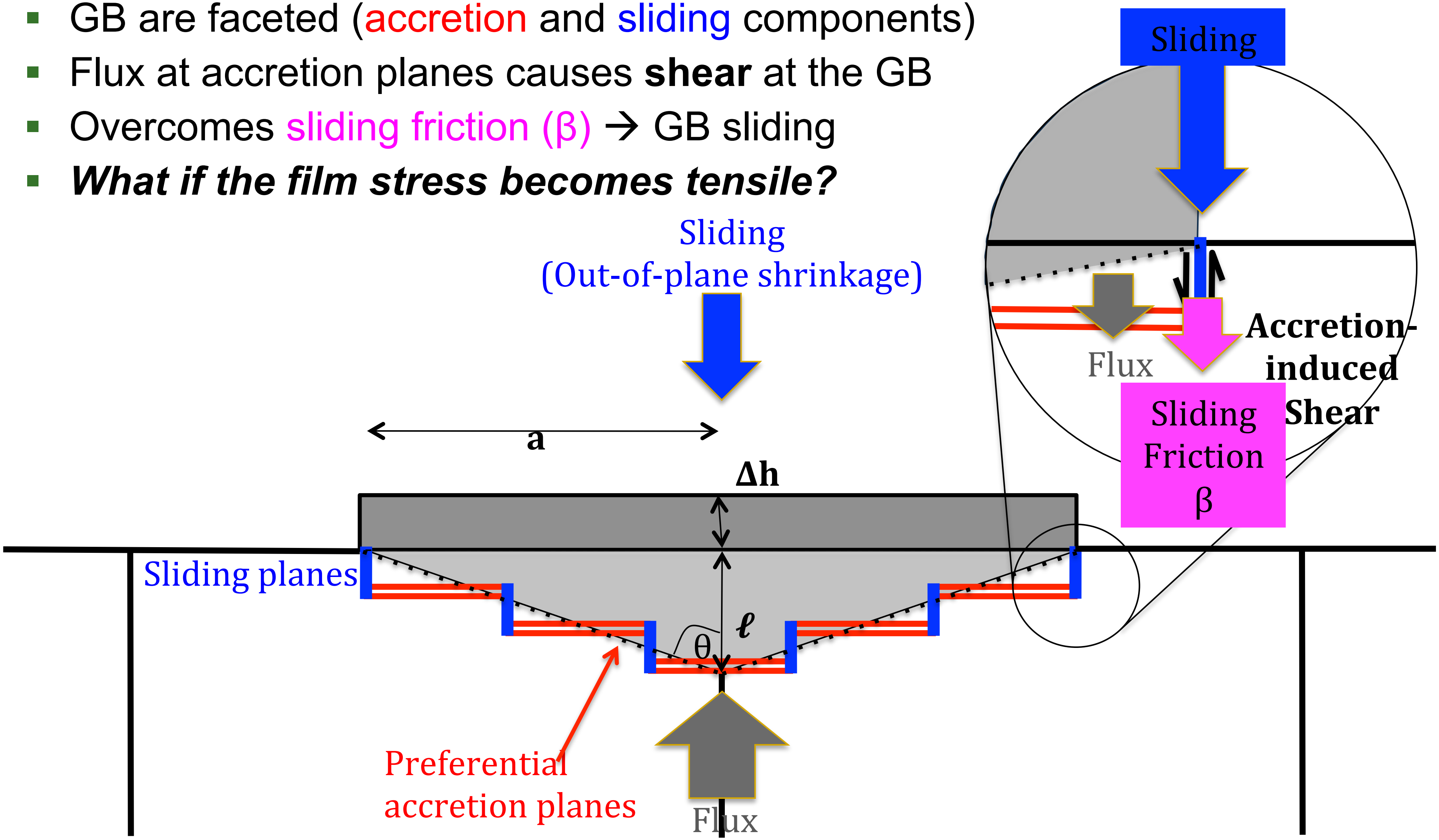
$$\sigma \pi r^2 > 2\pi r h \beta + 2\pi r \mathbf{x} \sigma_{oxide}$$



Whisker Growth: GB Sliding

PURDUE UNIVERSITY

- GB are faceted (accretion and sliding components)
- Flux at accretion planes causes **shear** at the GB
- Overcomes sliding friction (β) → GB sliding
- ***What if the film stress becomes tensile?***



Straight Whisker Growing normal to film surface

Whisker Growth: GB Sliding

- GB are faceted (accretion and sliding components)
- Flux at accretion planes causes **shear** at the GB
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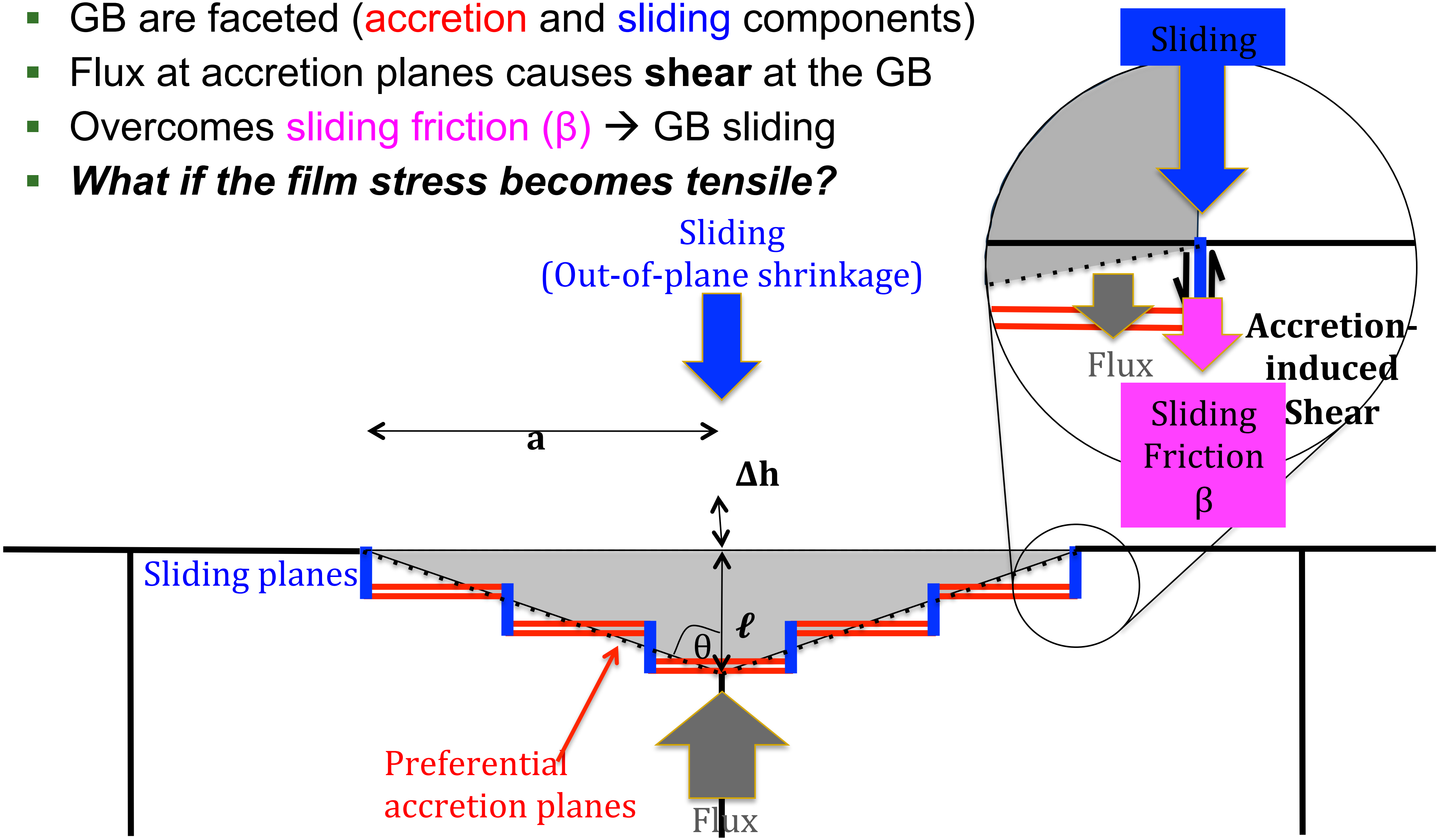
- ***What if the film stress becomes tensile?***



Straight Whisker Growing normal to film surface

E. Chason, F. Pei, C.L. Briant, H. Kesari, and A.F. Bower, *J. Electron. Mater.* 43, 4435 (2014).

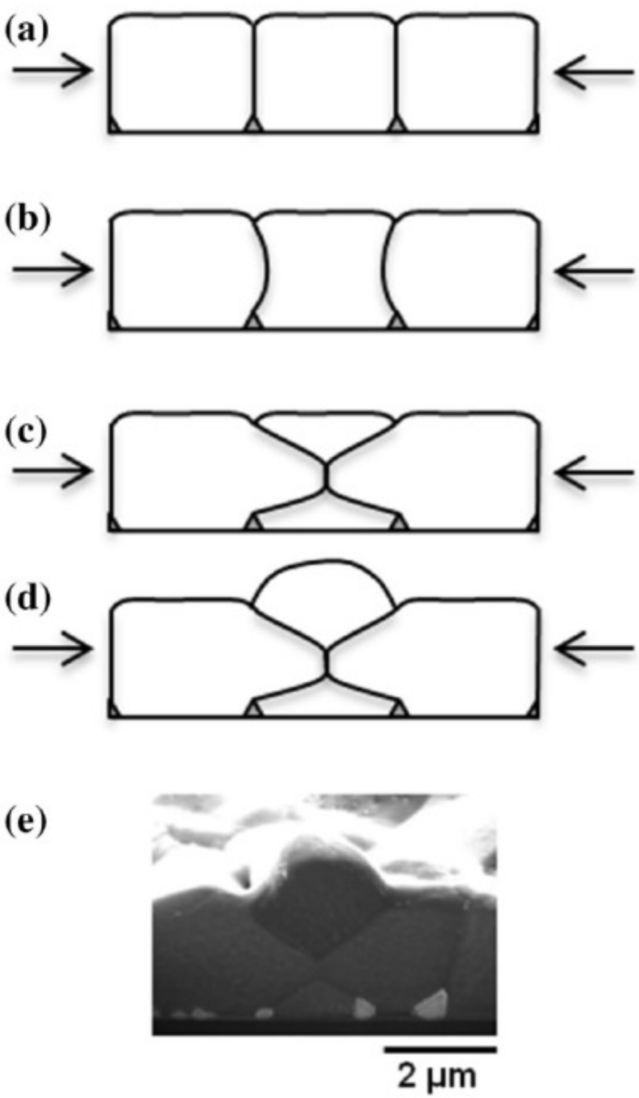
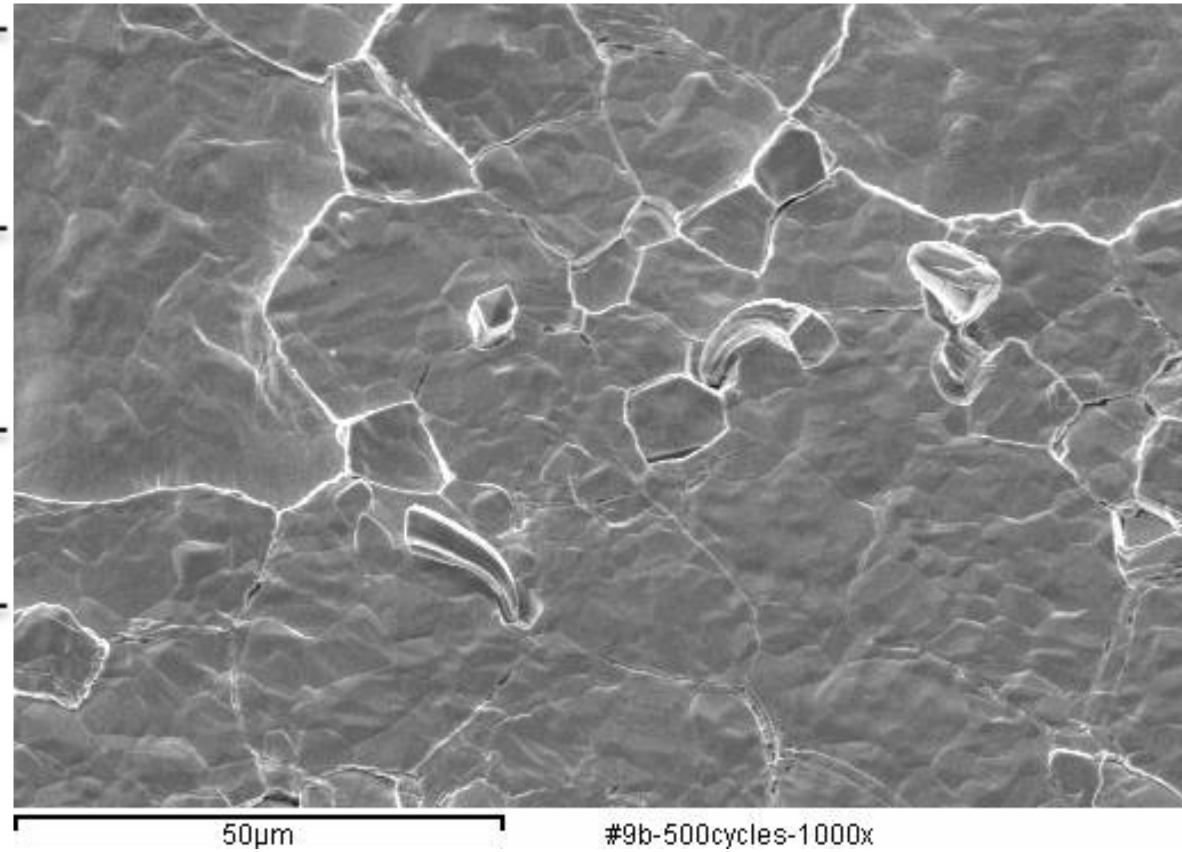


Fig. 4. (a–d) Schematic of proposed whisker nucleation mechanism described in text. (e) Cross-section of nucleus showing microstruc-

R. Parker, unpublished work



- **Grain growth and topological changes occur with larger CTE mismatch**

Microstructure Evolution in Hete
Systems

- Anisotropic Grain in Strontium Tita
  - Anti-thermal
  - “Normal” grain growth is not norma

Important role for theory, modeling, and simulation in understanding the sources of the responses we observe

  - Stress driven crystal growth out of
    embedded in the film
  - 1 grain out of 10,000 or 100,000 gr
  - Sources and sinks of atoms
  - Role of grain boundary geometry a
  - Evidence of coupled grain boundary sliding and migration

Giphy

Microstructure Evolution in Heterogeneous Systems

PURDUE UNIVERSITY

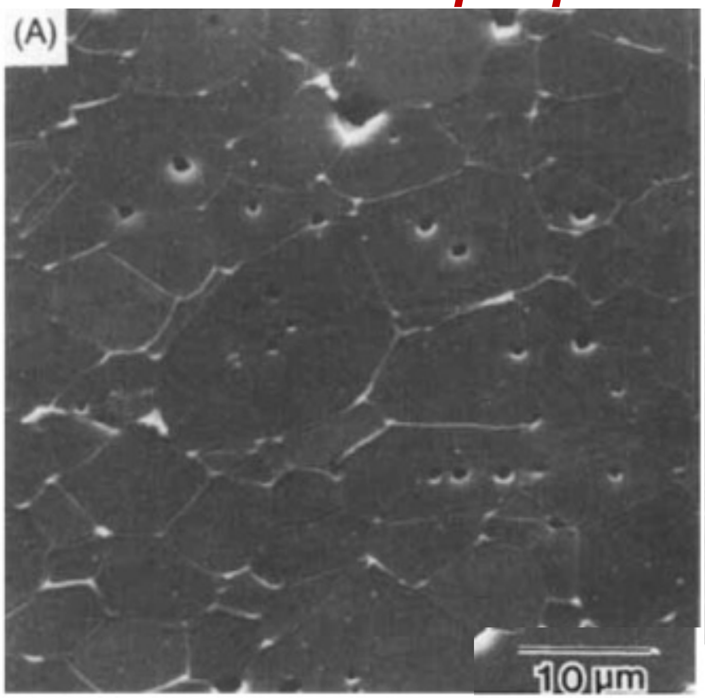
- Anisotropic Grain in Strontium Titanate
  - Anti-thermal
  - "Normal" grain growth is not normal
  - Abnormal grain growth in the absence of a liquid phase
  - Grain growth stagnation

- Tin Whisker Formation in Sn Thin FIlms
  - Stress-driven crystal growth out of the film surface from a grain embedded in the film
  - 1 grain out of 10,000 or 100,000 grains forms a whisker
  - Sources and sinks of atoms
  - Role of grain boundary geometry and crystallographic orientation
  - Evidence of coupled grain boundary sliding and migration

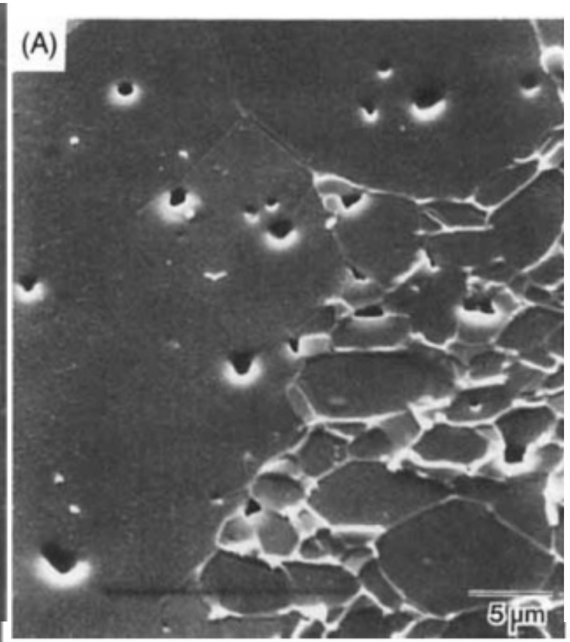
Effects of Chemical Inhomogeneities on Grain Growth and Microstructure in Aluminum Oxide

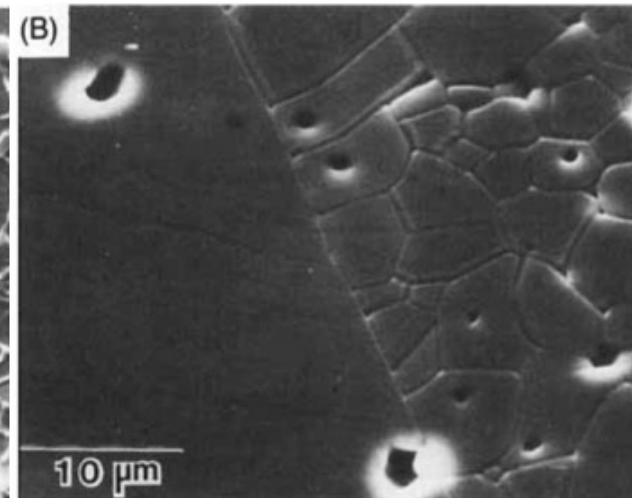
Handwerker, Morris, Coble
J. Am. Ceram. Soc., 72 [1] 130-36 (1989)

Normal GG – updoped



Transitions to Abnormal GG - undoped





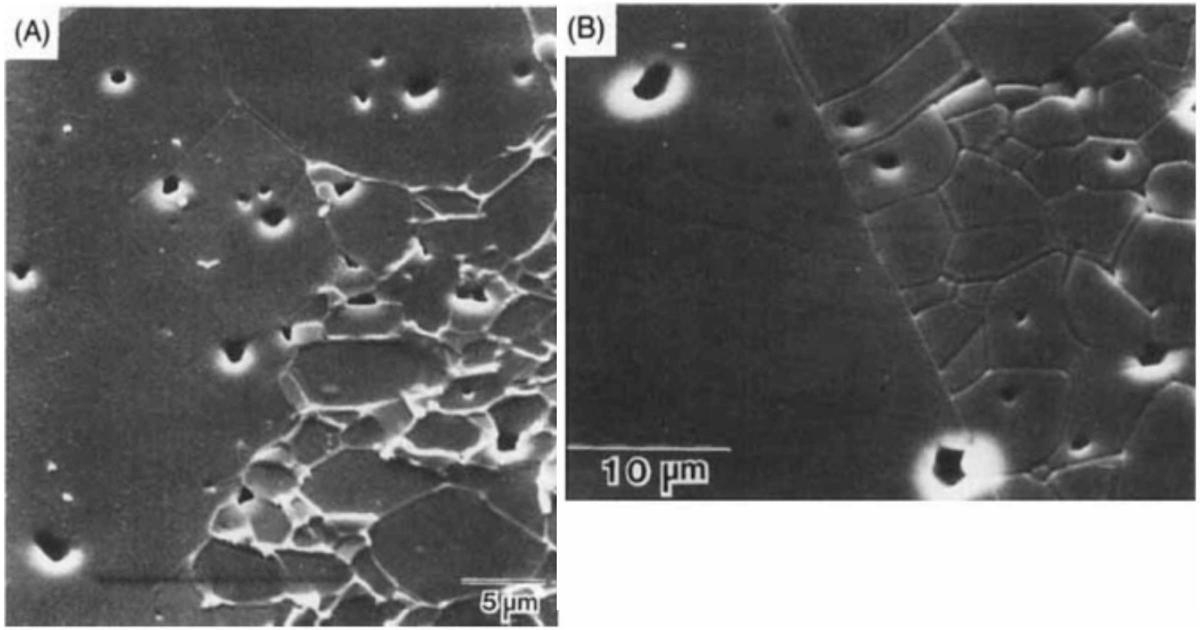
Effect of a wetting liquid phase along the grain boundaries on grain growth –

- Faster moving grain boundaries accumulate solute
- Reaches solubility limit and forms a liquid
- Faster mobility with liquid phase
- Liquid layer has maximum thickness due to liquid pinch-off
- Some grains accumulate more liquid => abnormal grain growth
- => Stop the liquid from forming

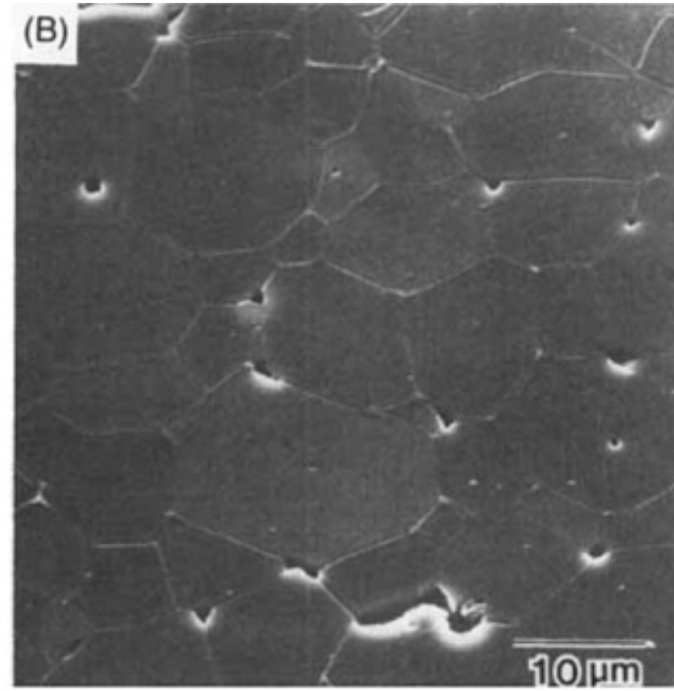
Grain Growth in Alumina

Handwerker, Morris, Coble
J. Am. Ceram. Soc., 72 [1] 130-36 (1989)

Transitions to Abnormal- undoped



Normal – MgO-doped

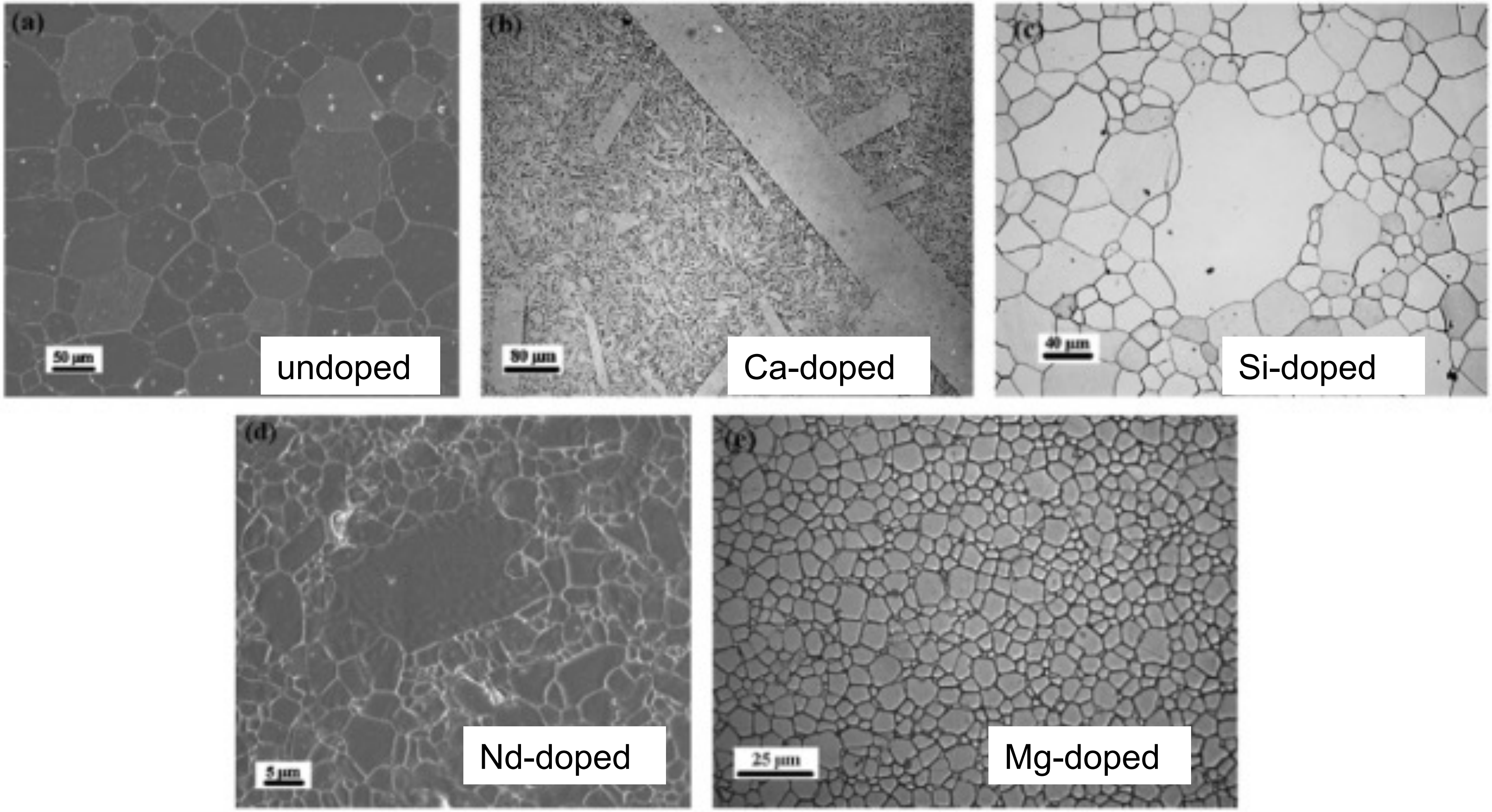


- **Holm simulations**: with isotropic grain boundary energies and mobilities – abnormal grain growth only happens for grains with boundaries that are always "fast"
- **Heuristic:** AGG is controlled by liquid wetting and/or roughening transitions with curvature driving interface motion
- How and why interfaces move – crystal growth vs. grain growth
- Understand and separate factors controlling normal/abnormal grain growth

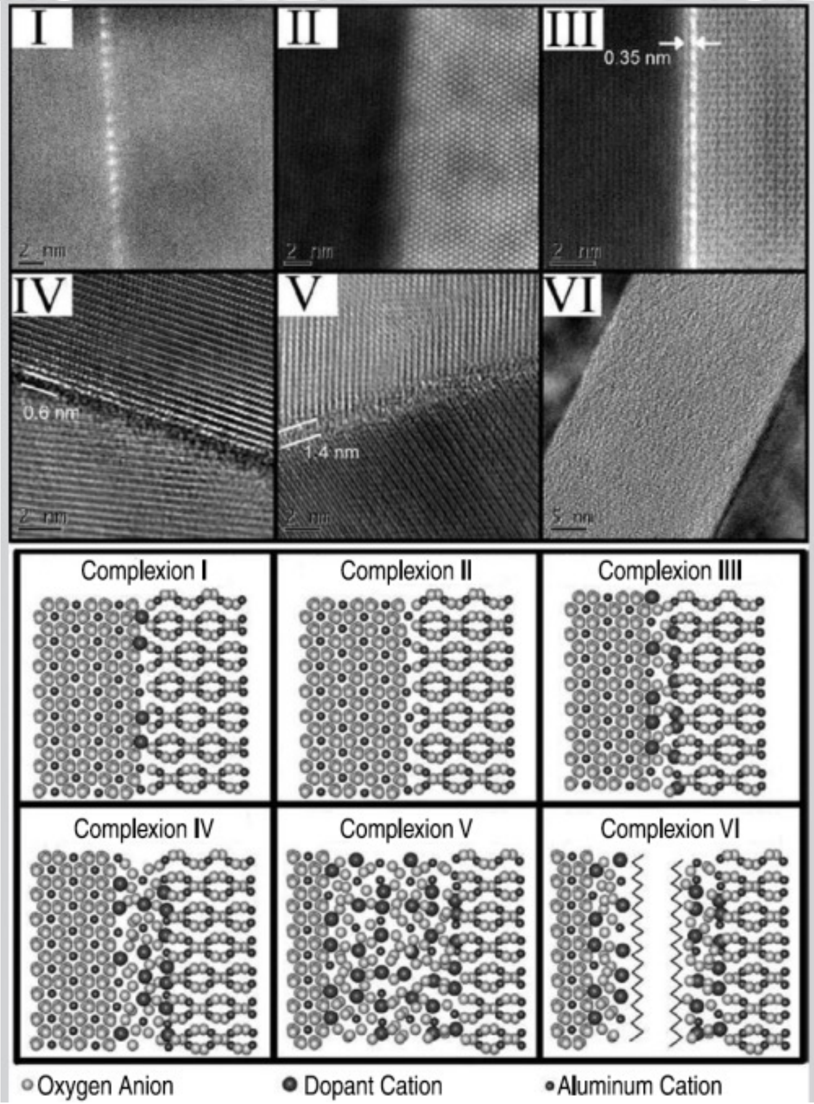
Complexion: A new concept for kinetic engineering in materials science

Shen J. Dillon [a,*], Ming Tang [b], W. Craig Carter [b], Martin P. Harmer [a]

Acta Materialia 55 (2007) 6208–6218

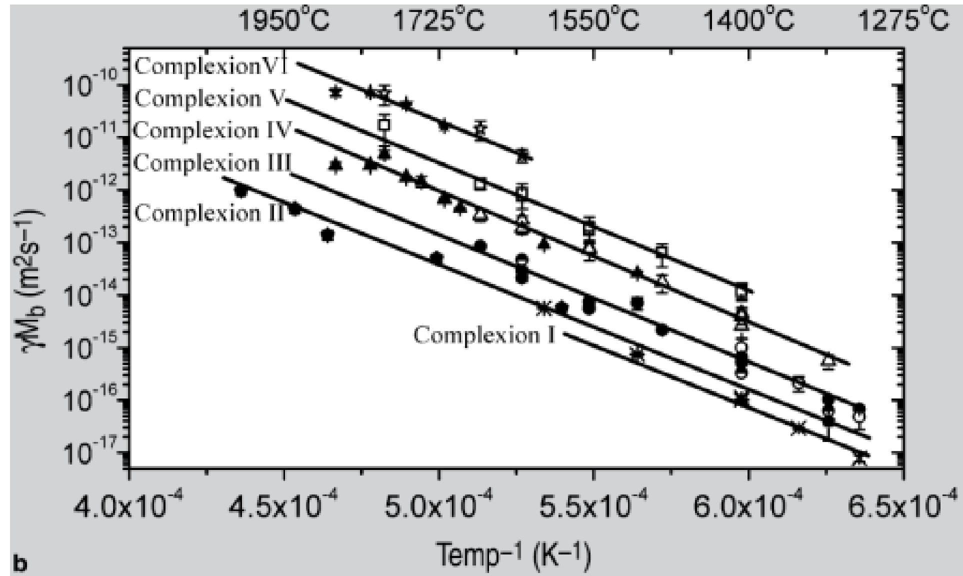


Grain Boundary Complexions in Ceramics and Metals: An Overview



Shen J. Dillon, Martin P. Harmer, and Jian Luo

Vol. 61 No. 12 • JOM



Focused Ion Beam Milling (FIB)
Local composition & structure

- HRTEM & HAADF STEM
- EBSD

- Understand and separate factors controlling normal/abnormal grain growth