U.S. DEPARTMENT OF ENERGY | Office of Science

SAND2013-3081P

Nanomechanics and Nanometallurgy of Boundaries

April 23-24, 2013

Brad L. Boyce

Requested Funding				
	FY13	FY14	FY15	Total
Operating	$736K	$736K	$736K	$2,208K
Capital	$455K	--	--	$455K
Total	**$1191K**	**$736K**	**$736K**	**$2,663K**

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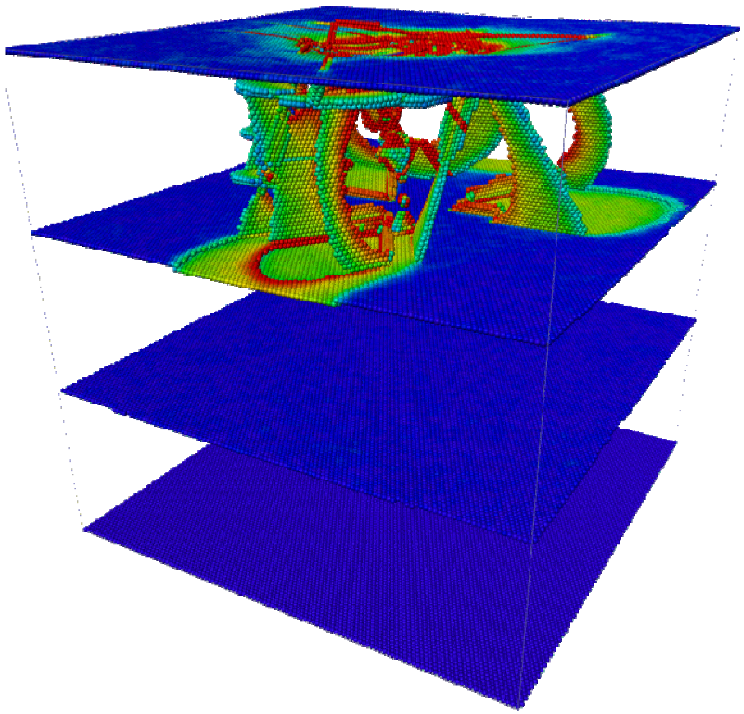
Outline and Agenda

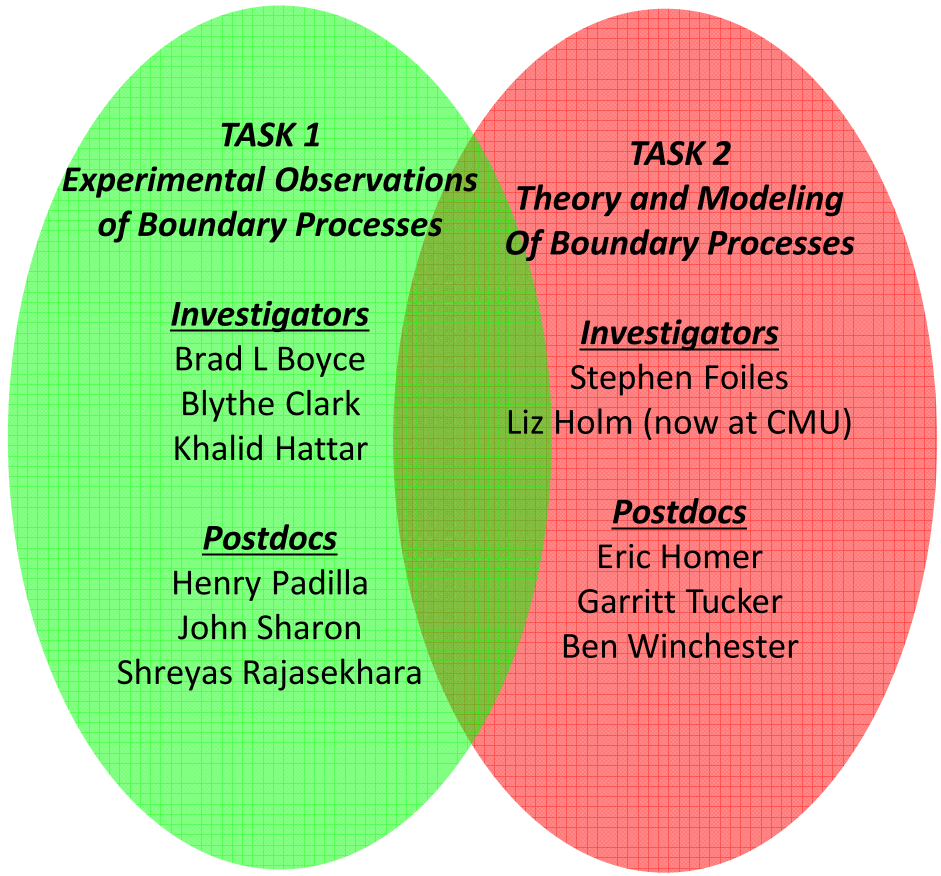
- **Project Description**
  - Project Goal and Objectives
  - Project Focus & Impact
- **Key Expertise and Facilities**
  - Investigator / Institution / Role
  - Facilities and Resources
- **Project Summary**
  - Science Driver
  - Project Highlights
- **Future work**
  - Inspiration from Literature
  - Facets of proposed work



Project Description

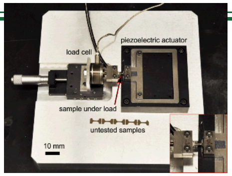
- **Background and Significance**
  - Whether by tension, indentation, fatigue, or wear, nanostructured alloys are prone to exhibit mechanically-induced grain growth.
- **Hypothesis**
  - Grain boundary instability can dissipate mechanical energy as a means to enhance the ductility, toughness, fatigue, and wear in nanocrystalline metals.
- **Motivating Questions**
  - *Is mechanically-driven grain growth a diffusional process controlled by Arrhenius kinetics?*
  - *What effect do impurities, defects, and local composition variations have on GB stability?*
  - *How do polycrystalline grain boundary networks serve to assist or impede grain boundary motion?*

Key Expertise and Facilities

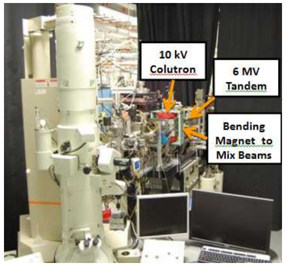


External Collaborators, Co-conspirators, & Overall Decent People (mostly): Ian Robertson (UIUC), Tony Rollett & Greg Rohrer (CMU), Greg Thompson (Alabama), Paulo Ferreira (UT), Amit Misra (LANL), Mitra Taheri (Drexel), Daniel Keiner (Leoben, Austria), Chris Schuh (MIT), Douglas Spearot (Arkansas), Xinghang Zhang (Texas A&M), Julia Greer (Cal Tech), Dan Gianola (U Penn), Andy Minor & Peter Hosseman (Berkeley), Kevin Hemker (Johns Hopkins), Apurva Mehta (Stanford Synchrotron), O. El-Atwani (Purdue), Tom LaGrange (LLNL), Emmanuelle Marquis & Sam Daly (Michigan)

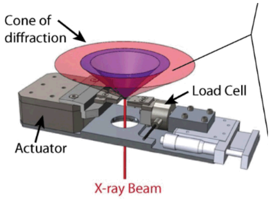
Key Expertise and Facilities



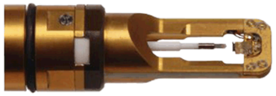
Thin film tensile



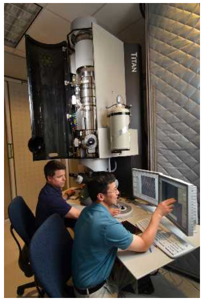
i3 TEM



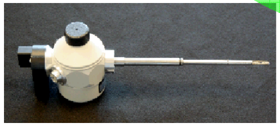
In-situ synchrotron XRD thin film tension



TEM in-situ nanomechanics



AC-STEM



TEM Cryoindentation (on loan from LANL)

TASK 1
Experimental Observations of Boundary Processes

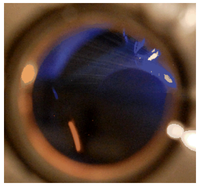
Investigators
Brad L Boyce
Blythe Clark
Khalid Hattar

Postdocs
Henry Padilla
John Sharon
Shreyas Rajasekhara

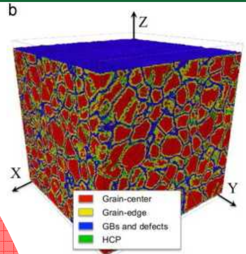
TASK 2
Theory and Modeling Of Boundary Processes

Investigators
Stephen Foiles
Liz Holm (now at CMU)

Postdocs
Eric Homer
Garritt Tucker
Ben Winchester



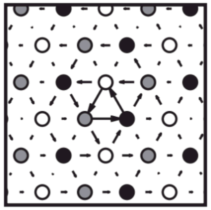
Pulsed Laser Deposition



LLAMPS EAM Molecular Dynamics

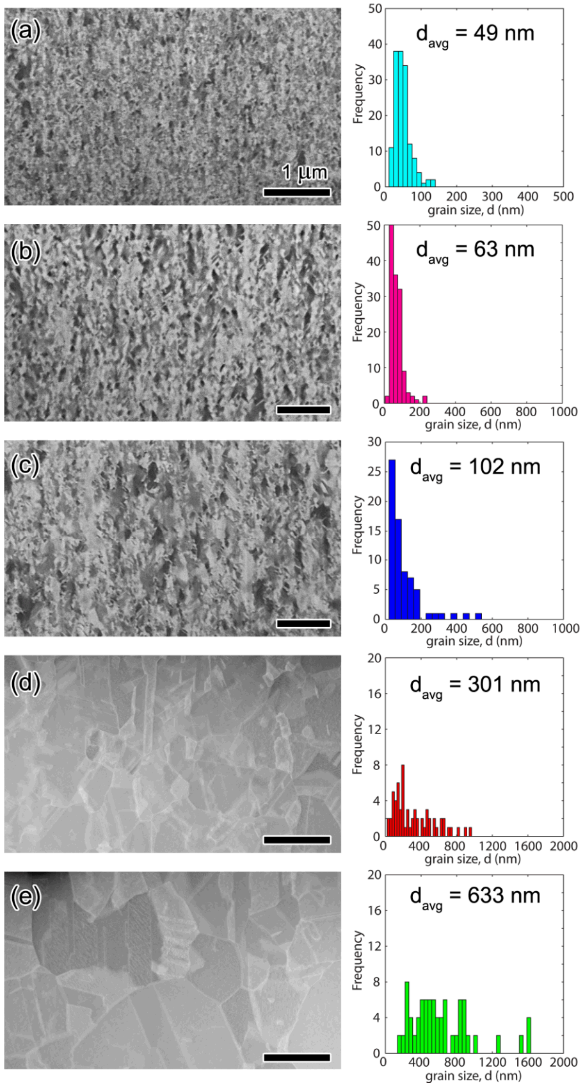
Phase Field Modeling

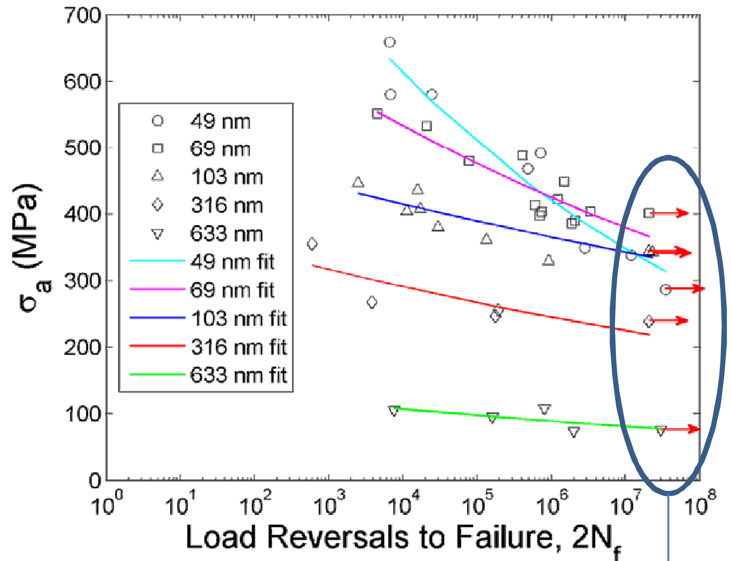
Thunderbird Cluster

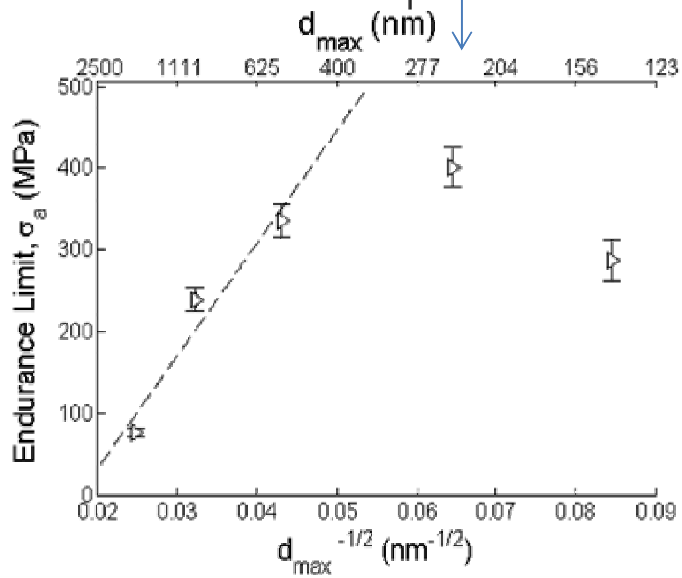


Density Functional Theory

Fatigue-induced grain growth

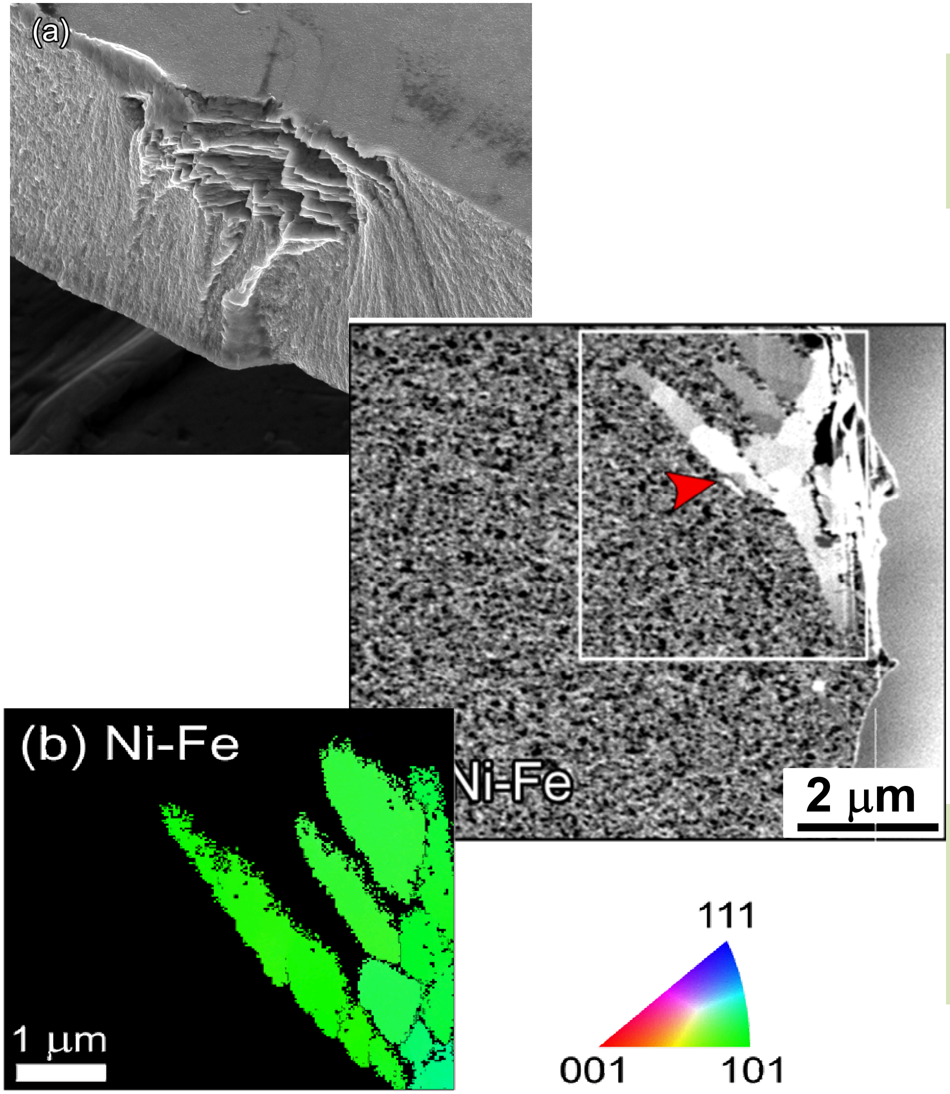




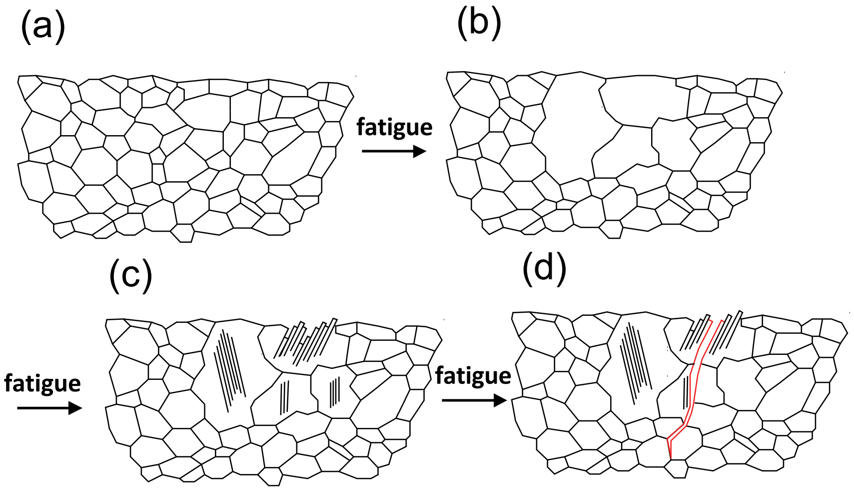


While there is a general trend of increasing fatigue resistance with decreasing grain size, there is evidence of a mechanistic transition when the average grain size is ~70 nm and the maximum grain size is ~300 nm.

Fatigue-induced grain growth



At average grain sizes of ~70 nm and below, fatigue crack initiation is preceded by fatigue-induced grain growth.

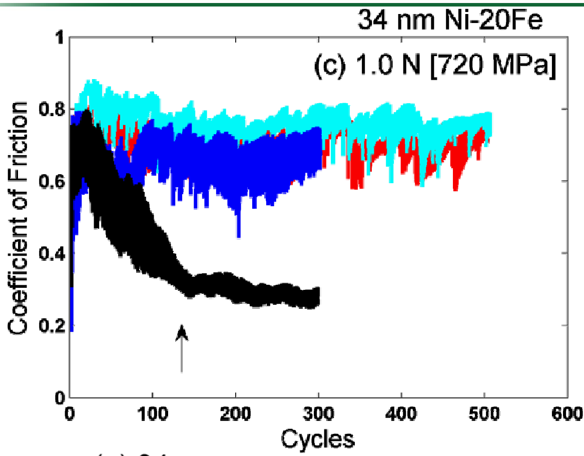


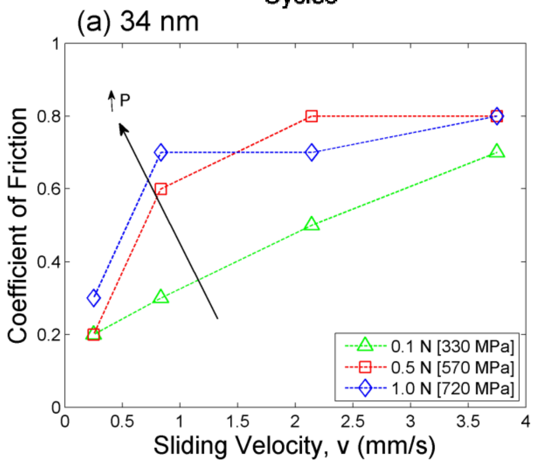
We have never actually observed fatigue crack initiation in a nanocrystalline alloy. Rather, we observe grain growth followed by initiation in the now larger grains.

B.L. Boyce & H.A. Padilla II, *Metall. Mater. Trans. A*, 2011 {Recipient of ASM's Marcus A. Grossman Award}

RESEARCH HIGHLIGHT 2

Wear-induced grain growth

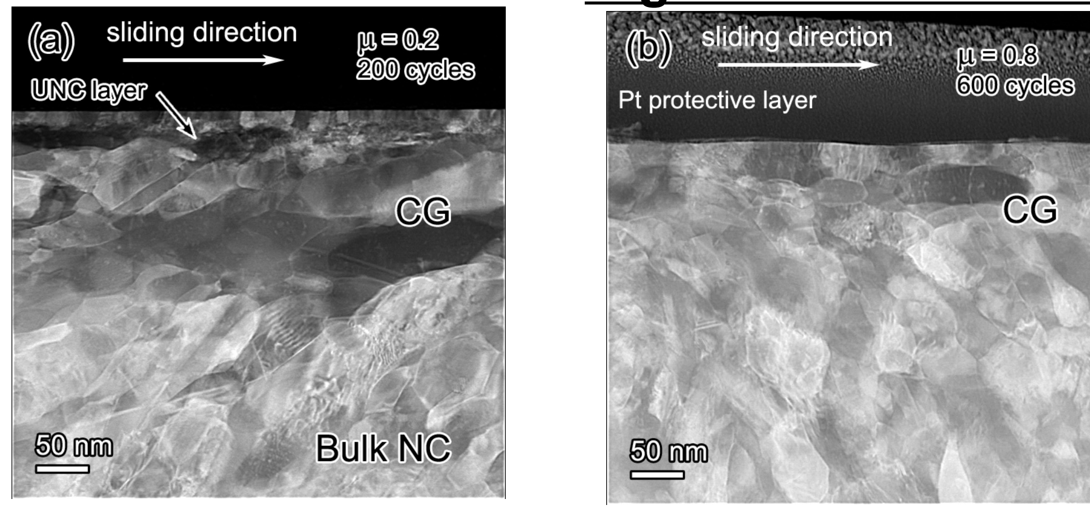


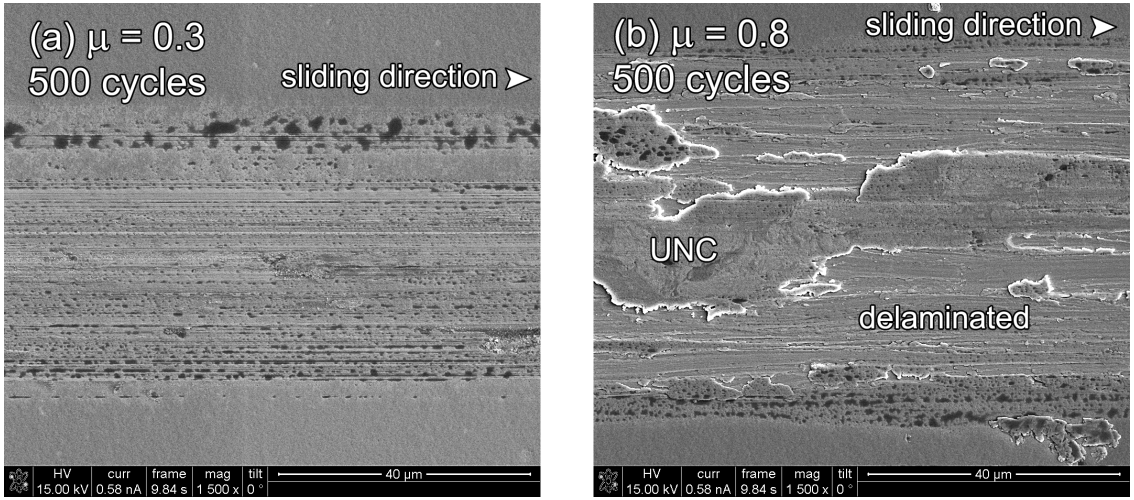


Nanocrystalline metals can exhibit either typical high friction or unexpected low friction.

Low friction behavior

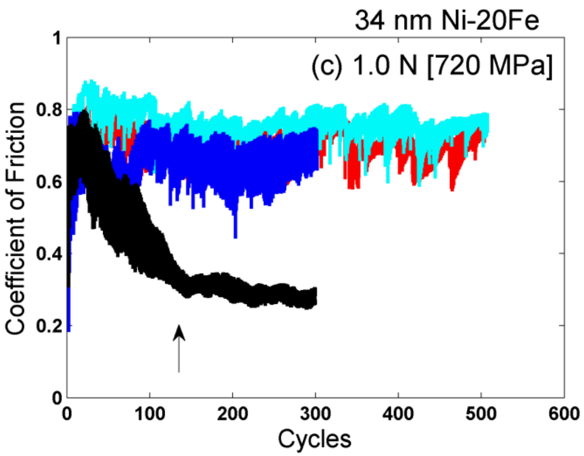
High friction behavior

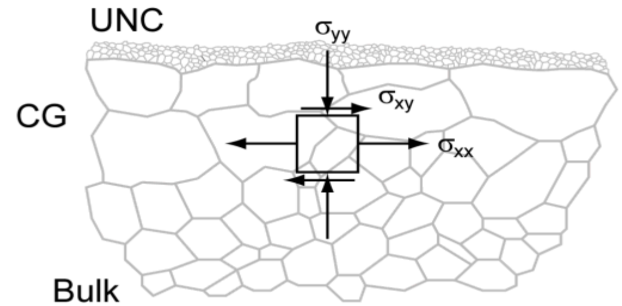


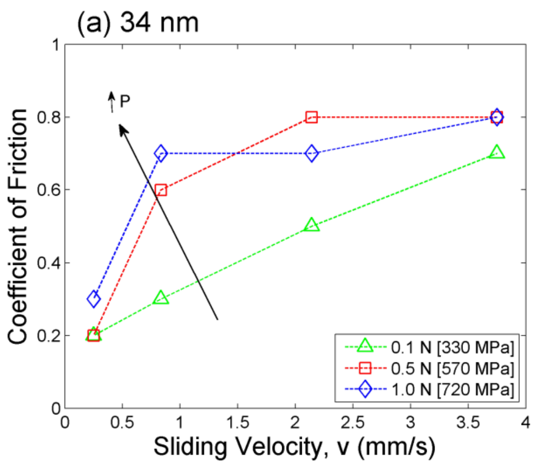


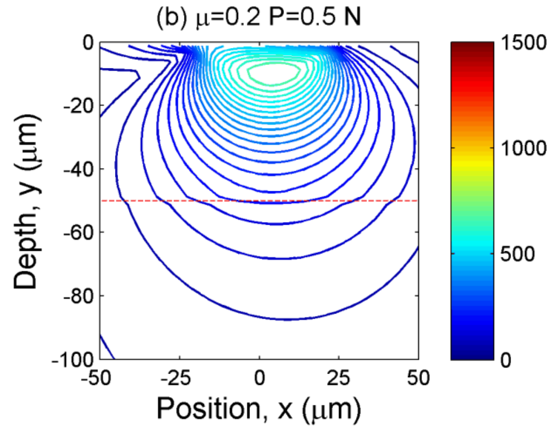
H.A. Padilla II, B.L. Boyce, C.C. Battaile, and S.V. Prasad, *Wear*, 2013.

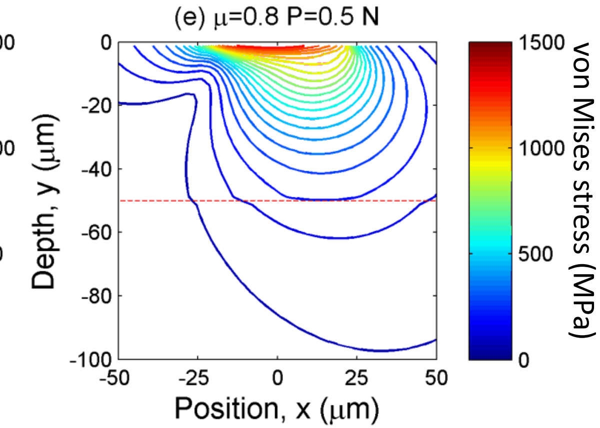
Wear-induced grain growth











Low friction behavior is associated with weaker peak stresses, far below the UNC-CG interface.

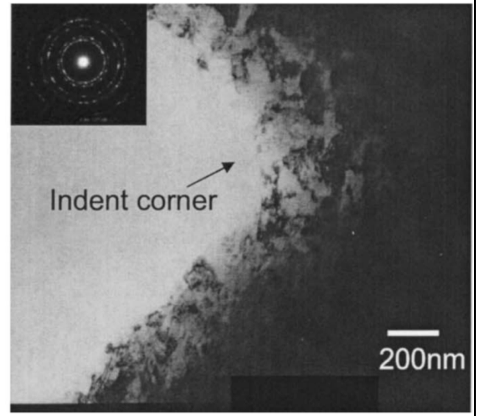
High friction behavior is associated with elevated peak stresses immediately at the UNC-CG interface

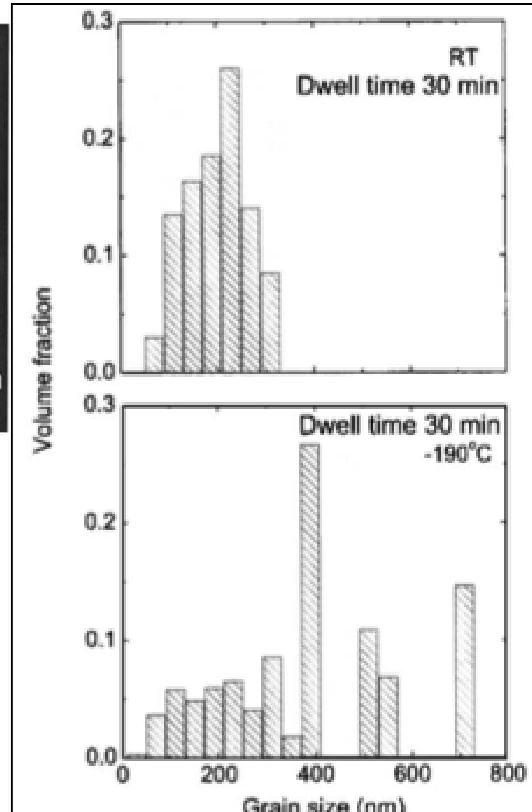
H.A. Padilla II, B.L. Boyce, C.C. Battaile, and S.V. Prasad, *Wear*, 2013.

Cryogenic indentation-induced grain growth

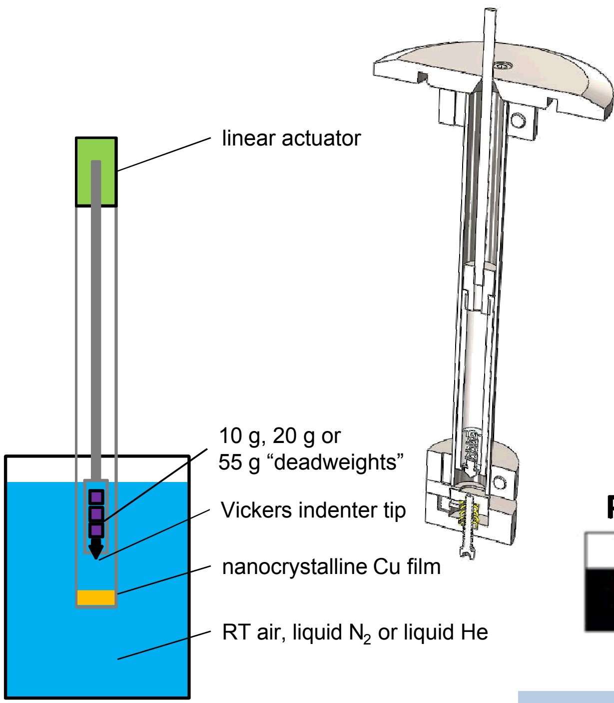
Is mechanically-induced grain growth still a thermally-activated process?

Weertman and coworkers observed more indentation-induced grain growth at 77K compared to 298K in high purity Cu indented!

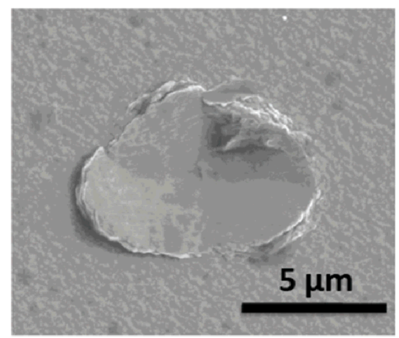




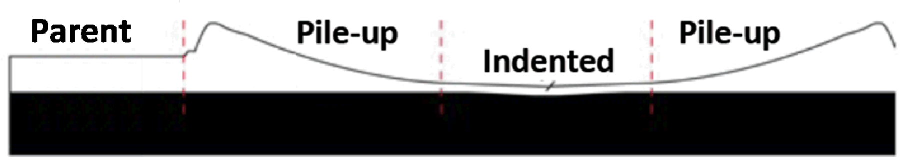
Zhang, Weertman, and Eastman, *Appl. Phys. Lett.* (2005)



(a)

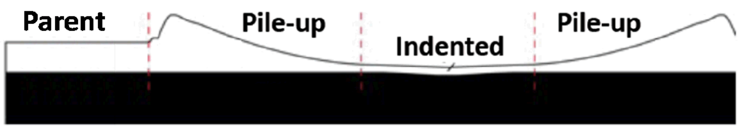


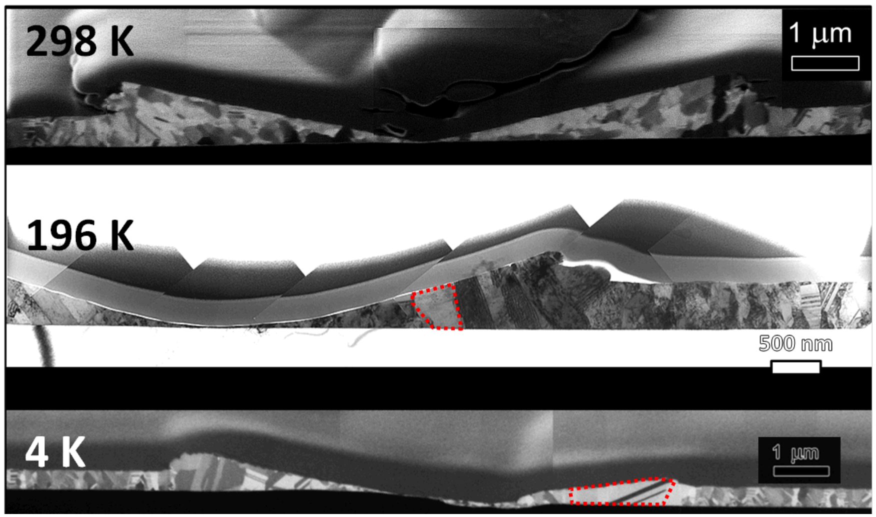
(b)

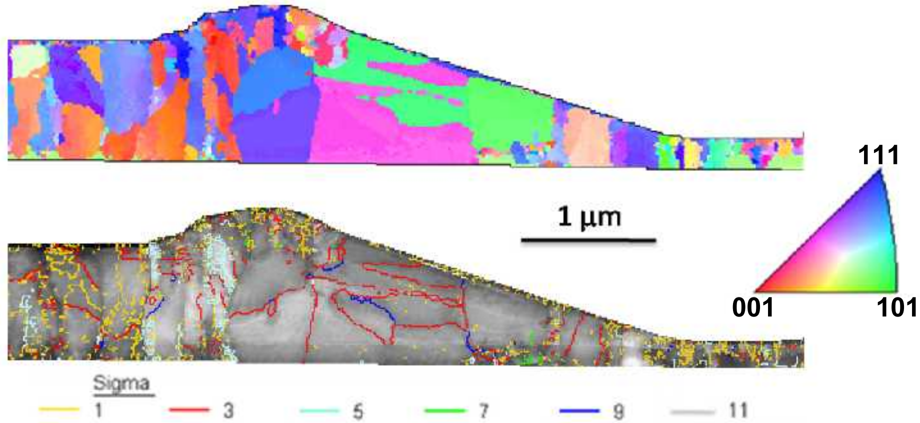


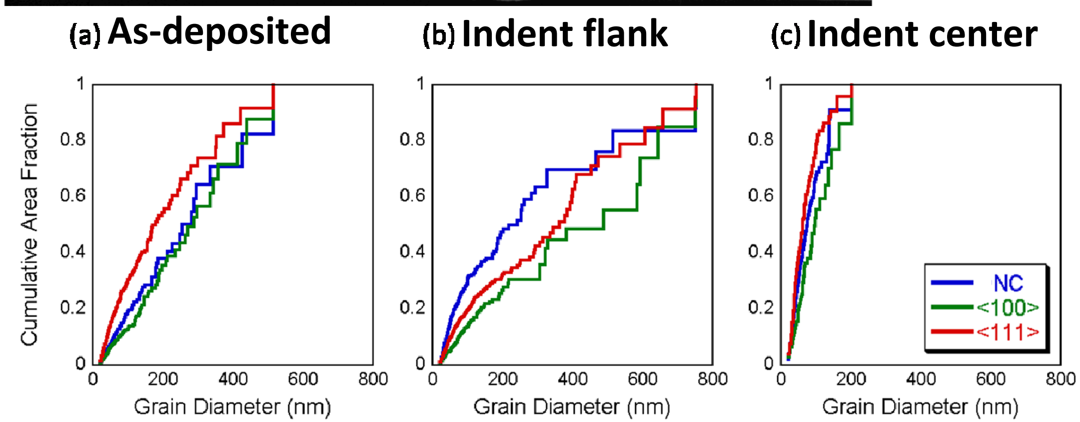
J.G. Brons, H.A. Padilla II, G.B. Thompson, and B.L. Boyce, *Scripta Mater*, 2013.

Cryogenic indentation-induced grain growth







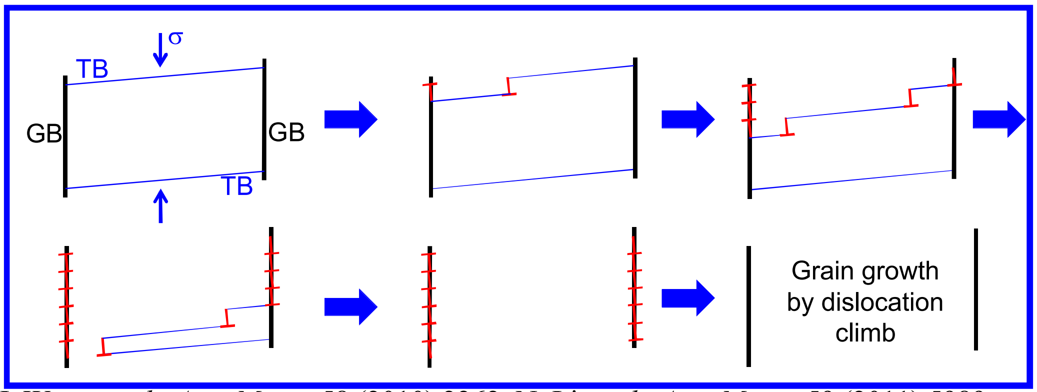


Grain growth was preferred at low temperatures (even down to 4K). TEM precession suggests the important role of special boundaries (Σ3 or Σ7) as the only boundaries left after indentation. They are either the fast moving sweepers, or stubborn remnant boundaries.

J.G. Brons, H.A. Padilla II, G.B. Thompson, and B.L. Boyce, *Scripta Mater*, 2013.

MD Simulation: Role of Twins

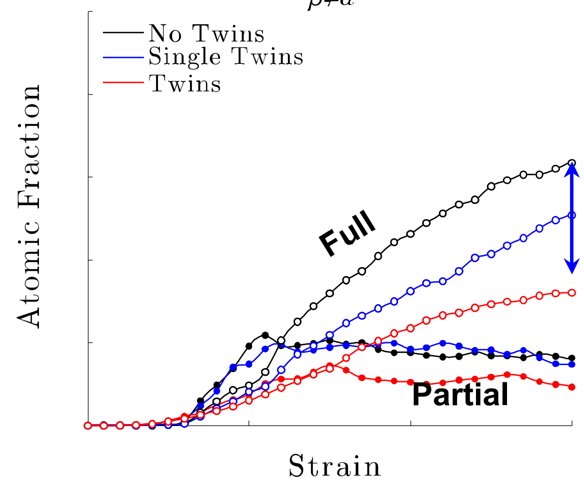
Coherent Σ3 twin boundaries are known to promote grain growth. What is the mechanistic role that these boundaries play?



J. Wang *et al.*, *Acta Mater.* 58 (2010) 2262; N. Li *et al.*, *Acta Mater.* 59 (2011) 5989;
D. Jang, C. Cai, J. Greer, *Nano Lett.* (2011)

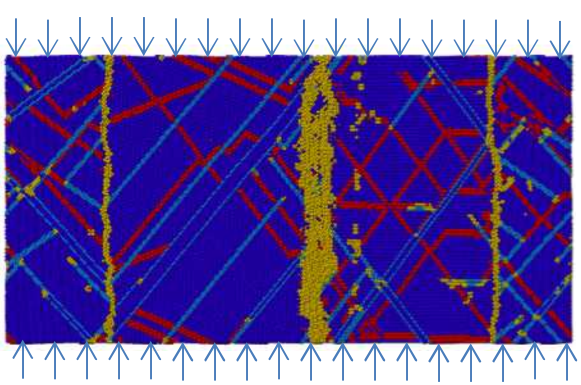
Slip vector for each atom α

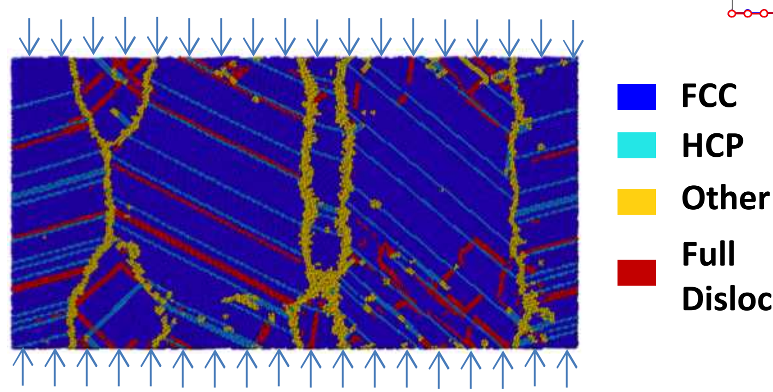
$$s^{\alpha} = -\frac{1}{n_s}\sum_{\beta\neq\alpha}^{n}\left(x^{\alpha\beta} - X^{\alpha\beta}\right)$$



Untwinned columnar crystals

Heavily twinned columnar crystals

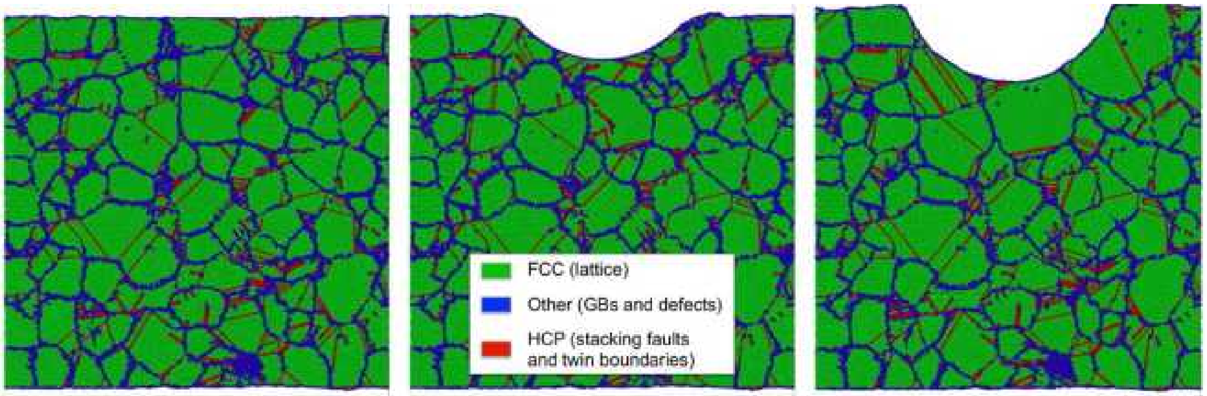


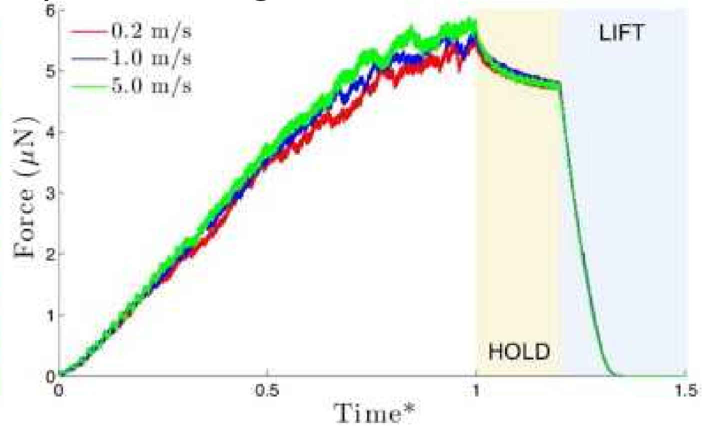


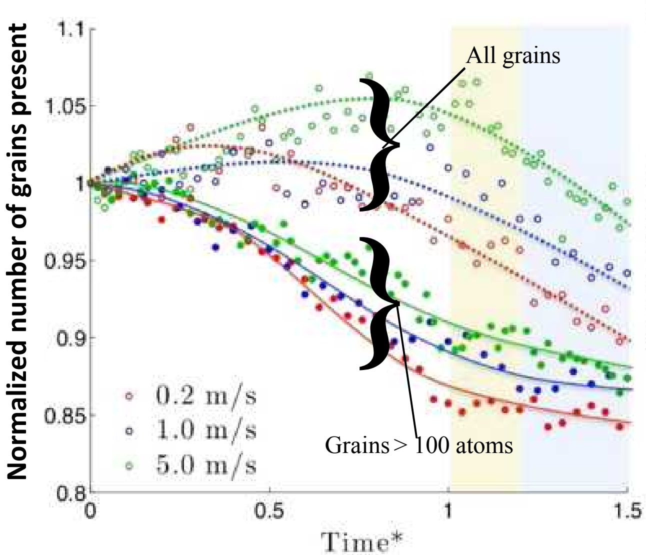
Twins channel dislocation activity to the grain boundary and suppress both full and partial slip in the parent lattice.

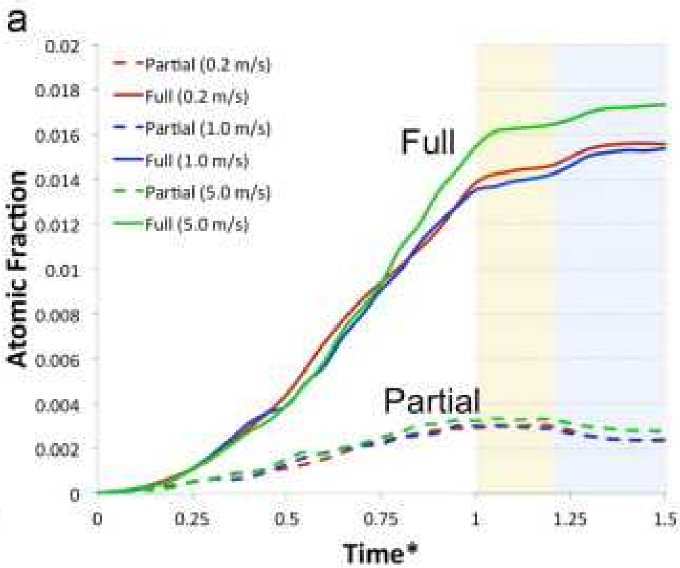
MD: Grain evolution kinetics and defect structure

3D MD simulation of Ni with 700 grains (grain size ~ 4 nm, 1.3M atoms) at 3 loading rates









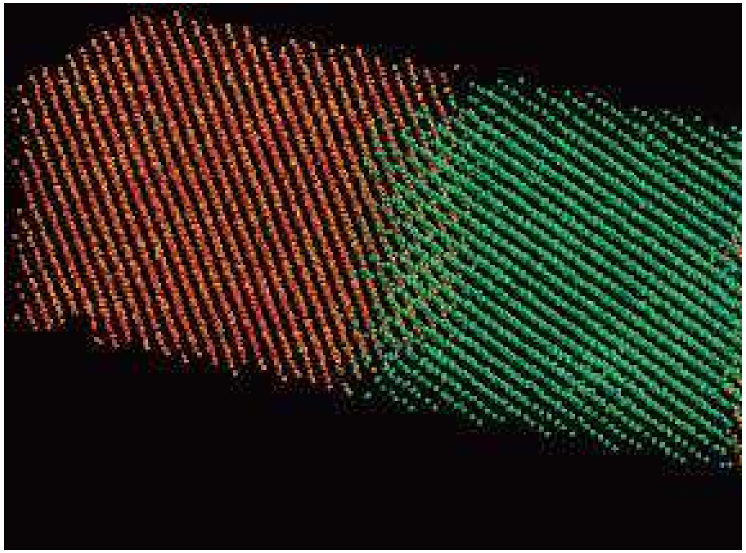
While full dislocations, partials, twinning and detwinning all take place, full dislocation density continues to increase on hold and unloading whereas partials are pulled back into the boundaries. As the partials collapse back into the boundaries, the boundaries continue to migrate.

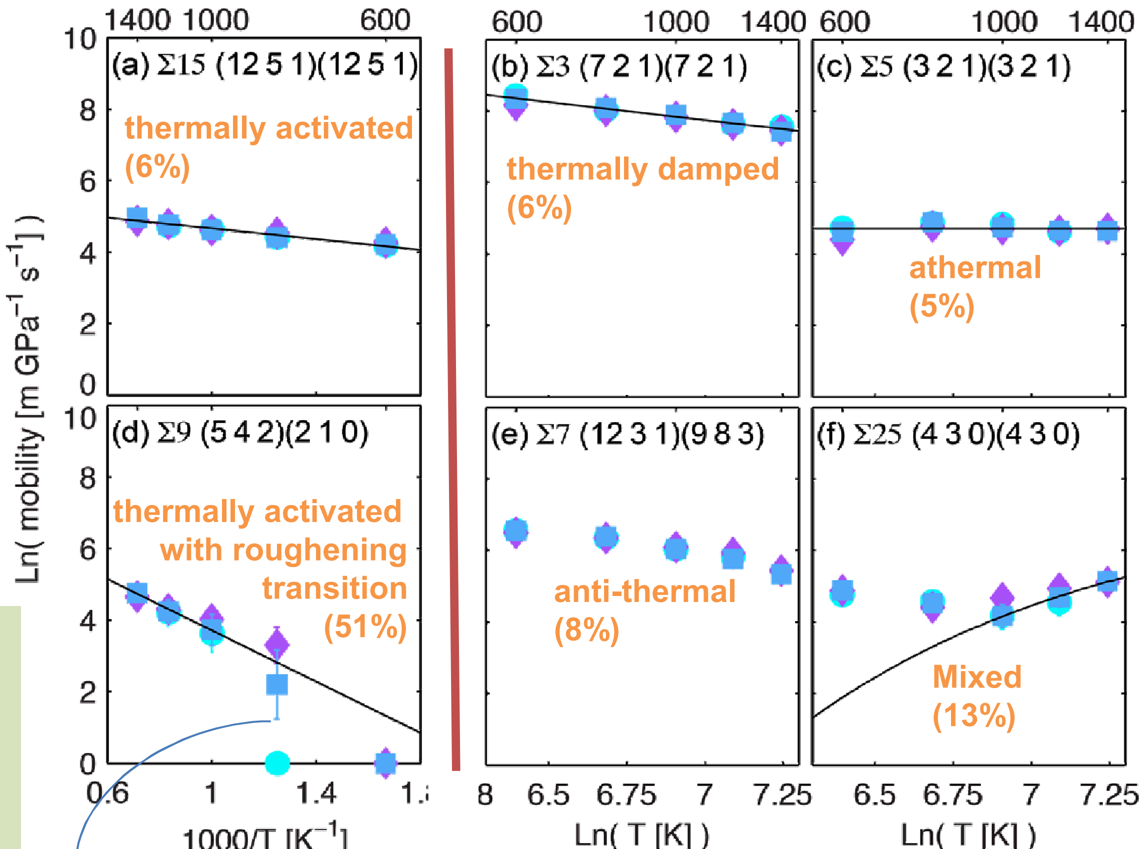
Growth = f($\dot{\varepsilon}$); growth continues during hold and unload

G.J. Tucker and S.M. Foiles, *Mater Sci and Eng A*, DOI:10.1016/j.msea.2012.08.045 , 2013.

MD: Boundary mobility in bi-crystals

388 Grain Boundary configurations in 4 FCC metals





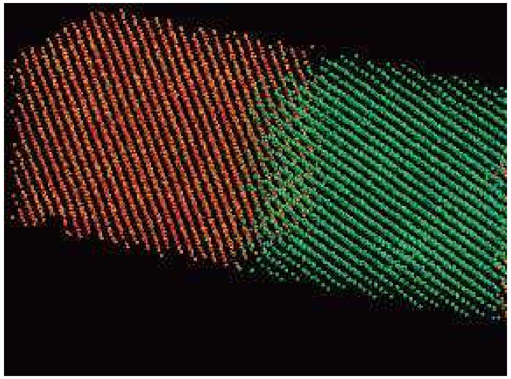
The mobility of a majority of grain boundary types are thermally activated, as expected

A surprising number fraction of boundaries (up to 30%) behave unexpectedly: thermally damped, anti-thermal, athermal, or mixed.

E.A. Holm, S.M. Foiles, *Science*, 2010

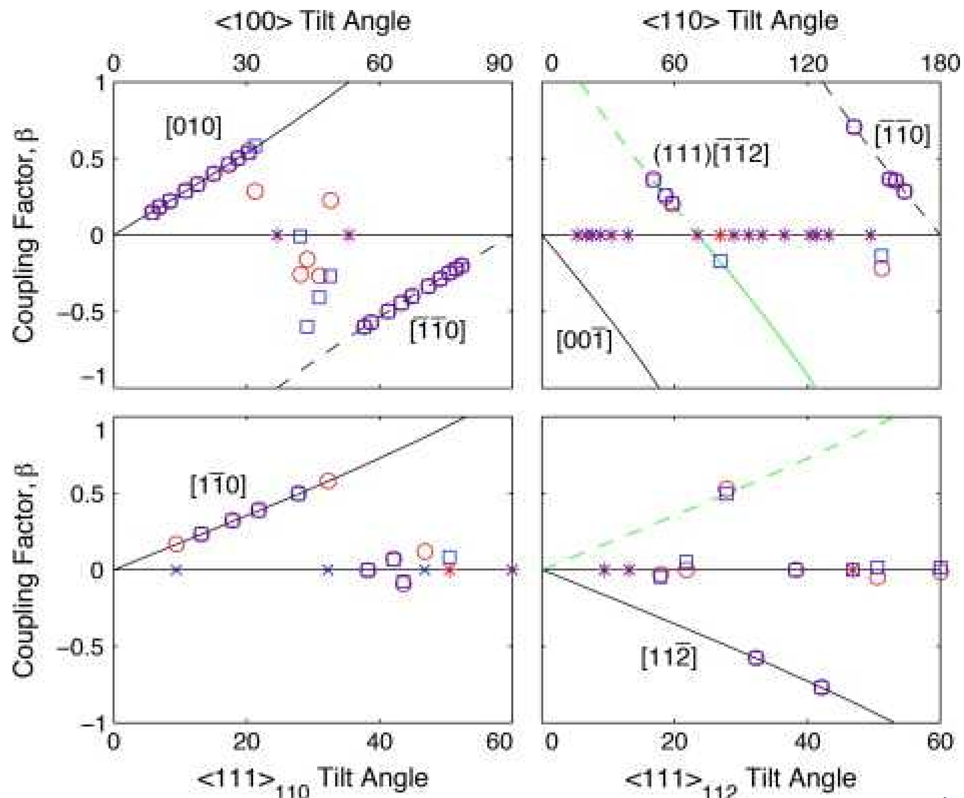
E.R. Homer, E.A. Holm, S.M. Foiles, and D.L. Olmsted, *in preparation*

MD: Shear-coupling in bicrystals



Coupling factor, β

$$\beta = \frac{v_{||}}{v_n} = \frac{v_x}{v_z}$$



Symmetrical tilt boundaries exhibit shear coupling similar to that predicted from Frank-Bilby analysis in Cahn & Mishin. We also find that some general boundaries exhibit coupling, and many of the shear coupled boundaries behaved athermally or anti-thermally.

SEE POSTER BY STEPHEN FOILES

E.R. Homer, E.A. Holm, S.M. Foiles, and D.L. Olmsted, *Acta Materialia*, 2013

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Sandia National Laboratories

Additional highlights…

Simulations of grain boundary energy, mobility, and defect interactions

1. E. A. Holm, S. M. Foiles, E. R. Homer, D. L. Olmsted, "Comment on 'Toward realistic molecular dynamics simulations of grain boundary mobility,' by Zhou and Mohles," *Scripta Mater.*, 66(9), p. 714-716, 2012.
2. E.A. Holm, G.S. Rohrer, S.M. Foiles, A.D. Rollott, H.M. Miller and D.L. Olmsted, "Validating computed grain boundary energies in fcc metals using the grain boundary character distribution", Acta Materialia 59, 5250-5256 (2011).
3. D.L. Olmsted, D. Buta , A. Adland, S.M. Foiles, M. Asta, A. Karma, "Dislocation-pairing transitions in hot grain boundaries", Physical Review Letters, 106(4), pp. 046101, 2011.
4. E.A. Holm, D.L. Olmsted, S.M. Foiles, "Comparing grain boundary energies in face-centered cubic metals: Al, Au, Cu and Ni", Scripta Materialia, 63(9), p. 905-908, 2010.
5. M. Chandross, E.A. Holm, "Measuring Grain Junction Angles in Discretized Microstructures", Metallurgical and Materials Transactions A, 41A, p. 3018-3025, 2010.
6. E. A. Holm, T. D. Hoffmann, A. D. Rollett, C. G. Roberts, "Particle-Assisted Abnormal Grain Growth," Recrystallization and Grain Growth, E. J. Palmiere et al. (eds.) (Trans Tech Publications, Zurich, 2010).
7. E. Fjeldberg, E. Holm, A. D. Rollett, and K. Marthinsen, "Mobility driven abnormal grain growth in the presence of particles," *Recrystallization and Grain Growth,* E. J. Palmiere *et al.* (eds.) (Trans Tech Publications, Zurich, 2010).
8. D. L. Olmsted, E. A. Holm, S. M. Foiles, "Survey of grain boundary energies in four elemental metals," (Extended Abstract) *Recrystallization and Grain Growth,* E. J. Palmiere *et al.* (eds.) (Trans Tech Publications, Zurich, 2010).
9. G. S. Rohrer, E. A. Holm, A. D. Rollett, S. M. Foiles, J. Li, and D. L. Olmsted, "Comparing calculated and measured grain boundary energies in nickel," *Acta Mater.* 58, p. 5063-5069 (2010). doi:10.1016/j.actamat.2010.05.042.

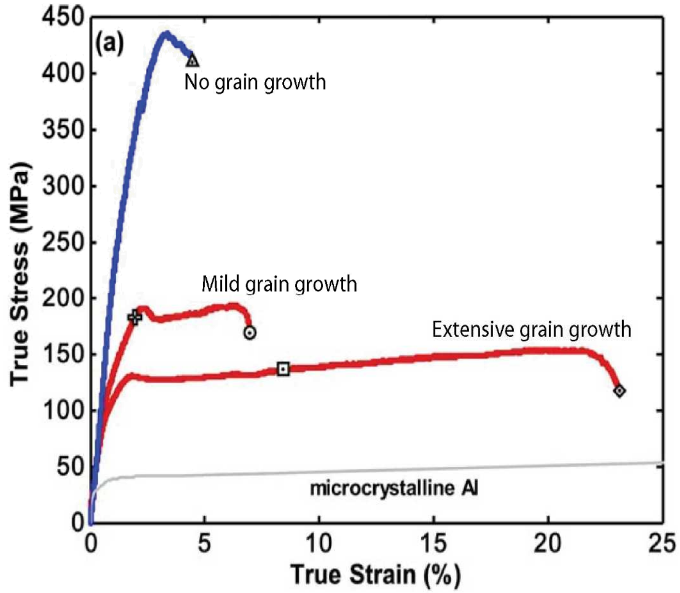
Deformation behavior of nanostructured metals and nanoscale experimental techniques

1. C.C. Battaile, B.L. Boyce, C.R. Weinberger, S.V. Prasad, J.R. Michael, B.G. Clark, "The hardness and strength of metal tribofilms: an apparent contradiction between nanoindentation and pillar compression", *Acta Materialia*, 60(4), p. 1712-1720, 2011.
2. K. Hattar; "Chapter 8: Deformation structures including twins in nanograined pure metals." (213-242) In S. H. Whang (Ed.), Nanostructured metals and alloys: processing, microstructure, mechanical properties and applications Oxford, UK, Woodhead Publishing (2011)
3. (invited review article) H.A. Padilla II, and B.L. Boyce, "A review of fatigue behavior in nanocrystalline metals", Experimental Mechanics, vol. 50 (1), p. 5-23, 2010.
4. B.L. Boyce, J.Y. Huang, D.C. Miller, and M. Kennedy, "Deformation and Failure of Small Scale Structures", *JOM*, 62(4), 62-63, 2010.
5. M.S. Steighner, L.P. Snedeker, B.L. Boyce, K. Gall, D.C. Miller, C.L. Muhlstein, "Dependence on diameter and growth direction of apparent strain to failure of Si nanowires", Journal of Applied Physics, 109(3), 033503, 2011.
6. K. Hattar, A. Misra, M. R. F. Dosanjh-, P. Dickerson, I. M. Robertson, R. G. Hoagland, "Direct observation of crack propagation in copper-niobium multilayers", *Journal of Engineering Materials and Technology*, 134(2), 2012.
7. J.R. Greer, Q. Guo, K. Hattar, P. Landau; The Effect of He Implantation on the Tensile Properties and Microstructure of Cu/Fe Nano-Bicrystals; Advanced Functional Materials; DOI 10.1002/adfm.201201776E.

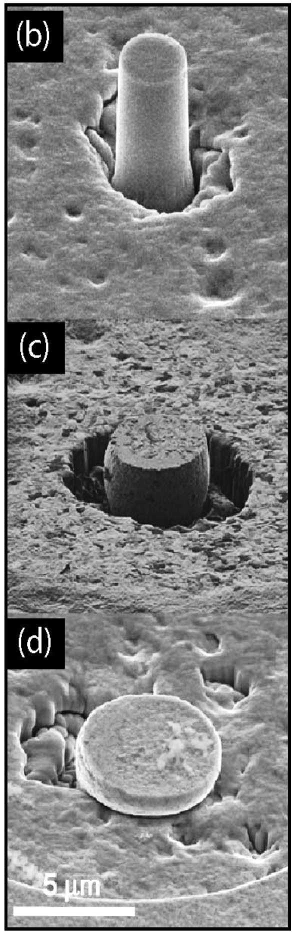
Thermal stability of nanostructured metals

1. S. Rajasekhara, K. Hattar, K. J. Ganesh, J. A. Knapp, P. J. Ferreira, "Evidence of Metastable hcp phase grains in As-deposited Nanocrystalline Nickel", *Scripta Materialia*, 67, p. 189-192, 2012.
2. J. Kacher, I.M. Robertson, M. Nowell, J. Knapp, K. Hattar, "Study of rapid grain boundary migration in a nanocrystalline Ni thin film", *Materials Science and Engineering A*, 528(3), p. 1628-1635, 2011.
3. J. Kacher, P. Elizaga, S.D. House, K. Hattar, M. Nowell, I.M. Robertson, "Thermal stability of Ni/NiO multilayer" *Mat. Sci. & Eng. A*, 568 (2013).

Inspiration…



From Gianola (*Acta Mater*, 2006), different deposition parameters for nanocrystalline Al affect both the susceptibility to grain growth and the ductility.



From Pan, Nano Letters, 2007. High compressive plasticity attributed to mechanical grain growth.

Hypothesis

Grain boundary instability can dissipate mechanical energy as a means to enhance the ductility, toughness, fatigue, and wear in nanostructured metals.

Alloys with modified stability

$$v = Mp$$

Boundary velocity, *v*

- The speed at which a boundary moves in response to a driving force.

Boundary mobility, *M*

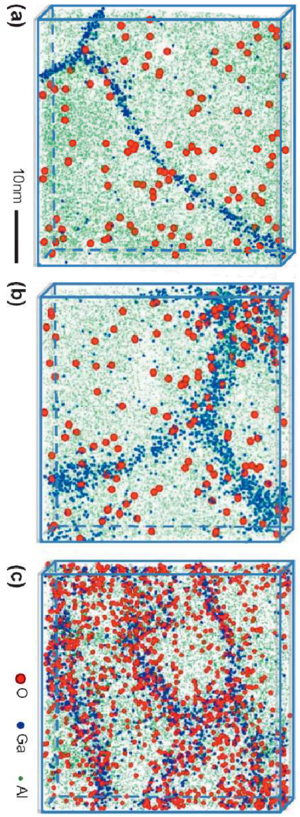
- Dependent on:
  - Boundary structure
  - Temperature
  - Magnitude of the driving force
- Independent of the nature of the driving force

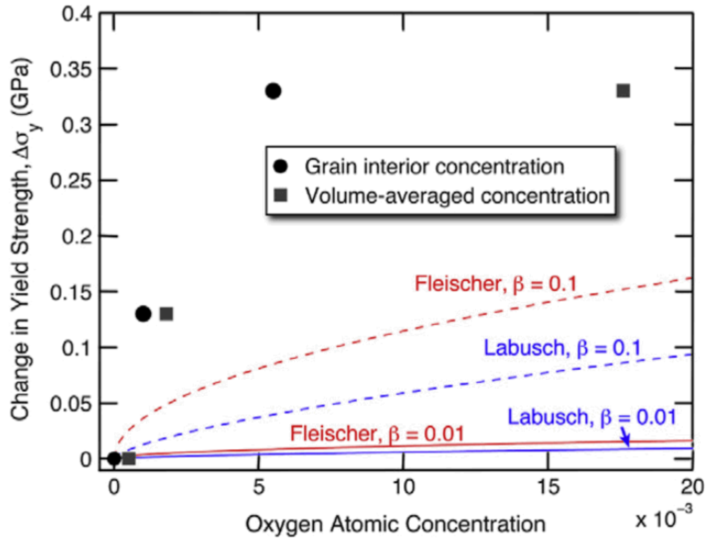
Driving force, *P*

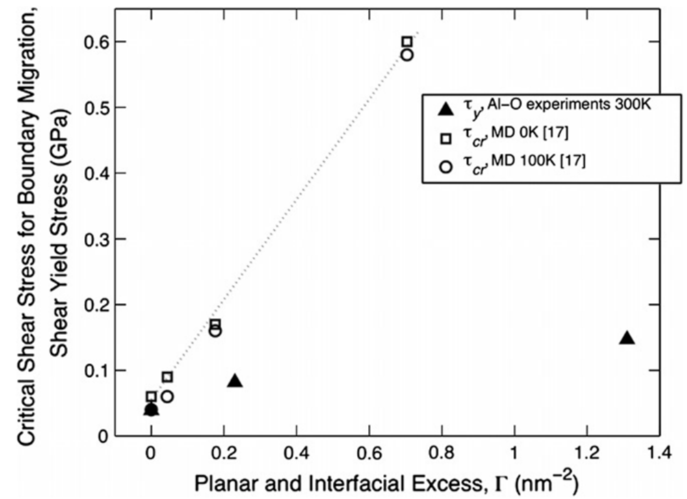
- Caused by:
  - Boundary energy
  - Elastic energy
  - Stored strain (plastic) energy
  - Magnetic energy
  - Chemical energy
  - Etc.

Drag on the grain boundary

$$v = M\,p$$



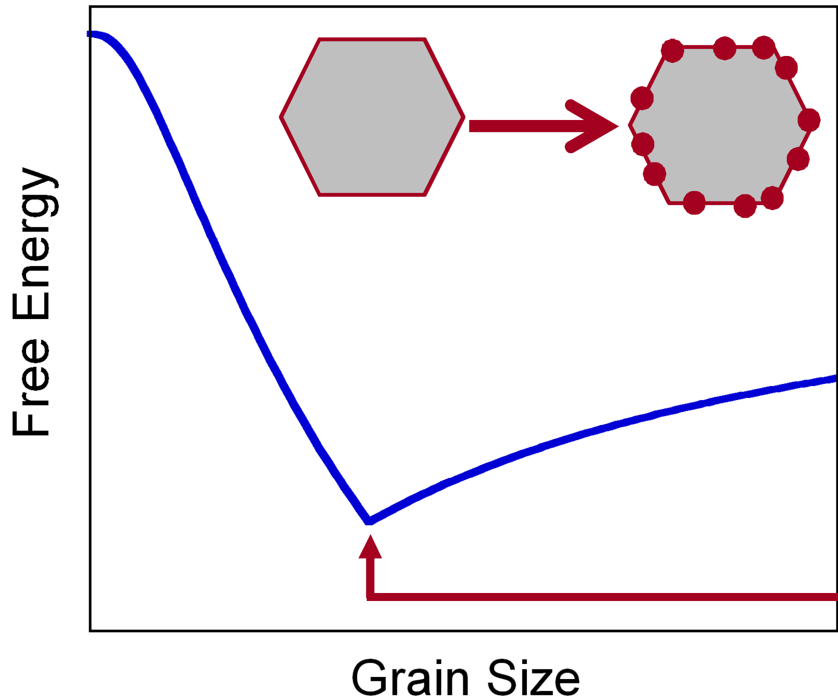




Al-O
Tang, Gianola, Moody, Hemker, and Cairney, *Acta Mater.* (2012)

- ***Can controlling oxygen level allow one to 'dial in' the mobility to tailor the susceptibility to mechanically-induced grain growth?***
- ***Can in-situ irradiation be used to dynamically modify the mobility of a moving grain boundary?***

Modified driving force for grain growth



$$dG = \gamma dA$$

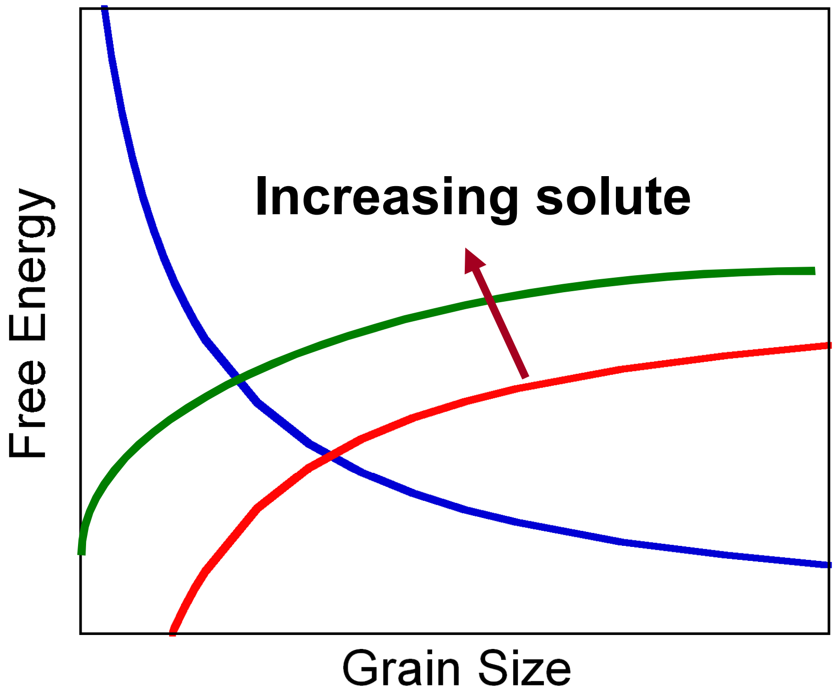
$$dG = \left[\gamma - \frac{N_\beta}{A}\Delta G_{seg}\right] dA$$

The *thermodynamically preferred* grain size

Weissmuller: Journal of Materials Research 1994
Kirchheim: Acta Materialia 2002

Chris Schuh & Heather Murdoch, Fall MRS 2012

Modified driving force for grain growth



$$dG = \gamma dA$$

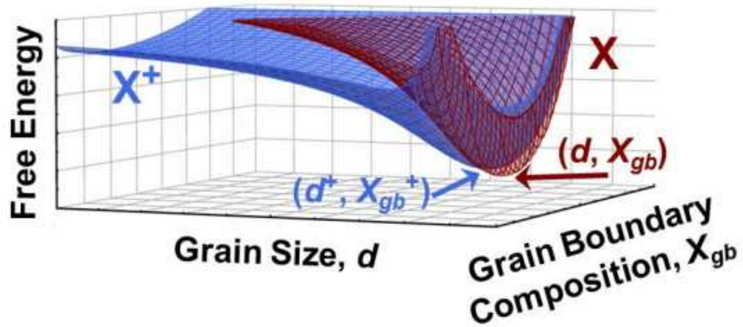
$$dG = \left[\gamma - \frac{N_\beta}{A}\Delta G_{seg}\right]dA$$

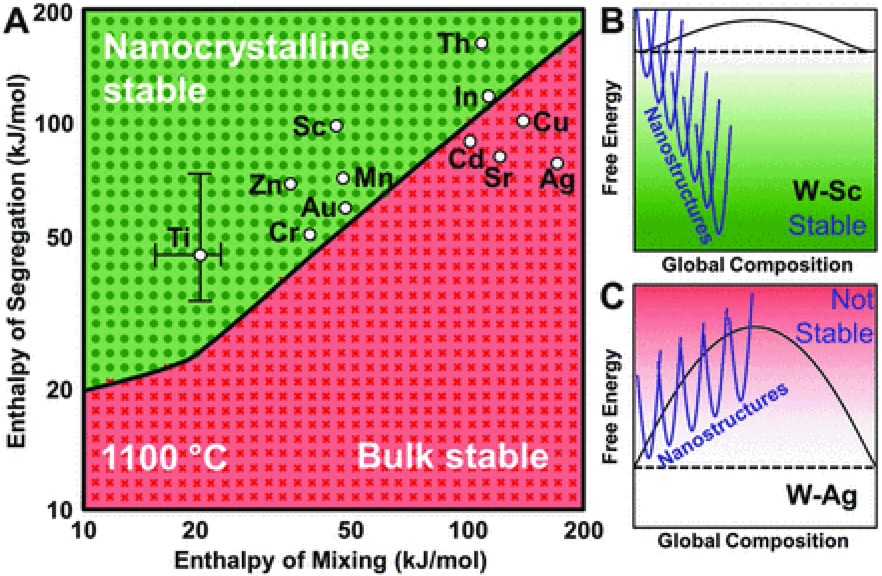
$$\Delta G^{\mathrm{mix}} = (1 - f_{\mathrm{gb}})\Delta G_{\mathrm{c}}^{\mathrm{mix}} + f_{\mathrm{gb}}\Delta G_{\mathrm{gb}}^{\mathrm{mix}} + z\nu f_{\mathrm{gb}}(X_{\mathrm{gb}} - X_{\mathrm{c}})\left[(2X_{\mathrm{gb}} - 1)\omega_{\mathrm{gb}} - \frac{1}{zt}(\Omega^{\mathrm{B}}\gamma^{\mathrm{B}} - \Omega^{\mathrm{A}}\gamma^{\mathrm{A}})\right] \quad (2)$$

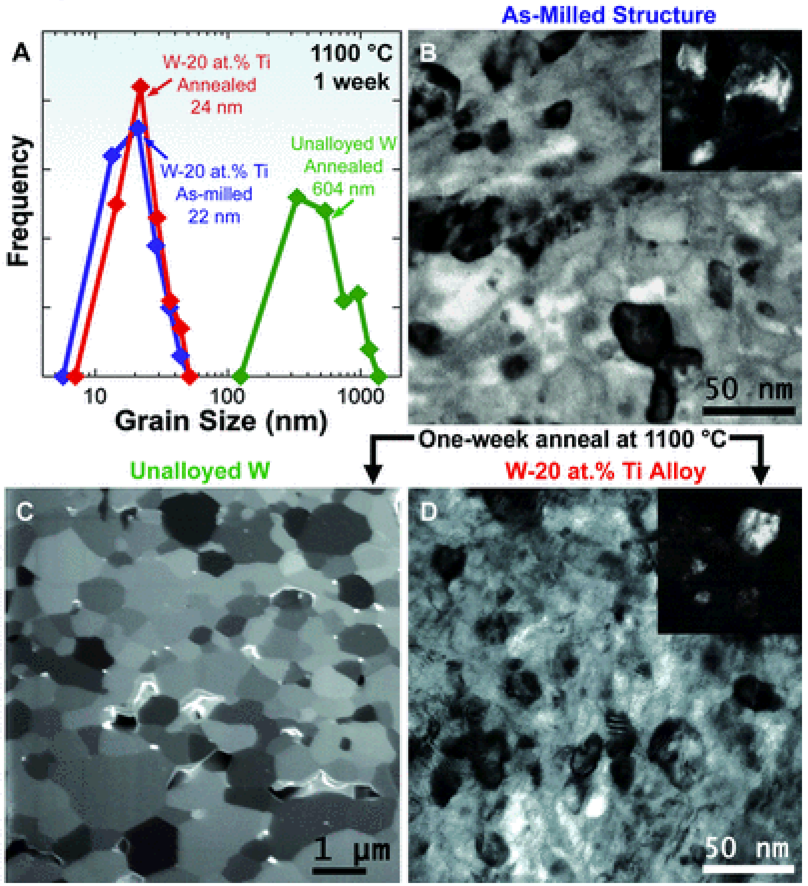
Chris Schuh & Heather Murdoch, Fall MRS 2012

Modified driving force for grain growth

Tongjai Chookajorn, Heather A. Murdoch, Christopher A. Schuh*, *Science*, 2012



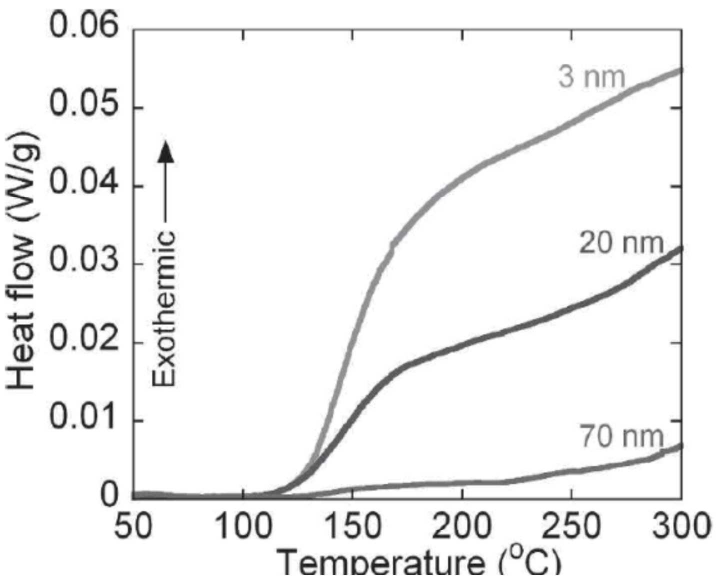




Does thermodynamic stability bear any relevance to stability against mechanically-induced grain growth?

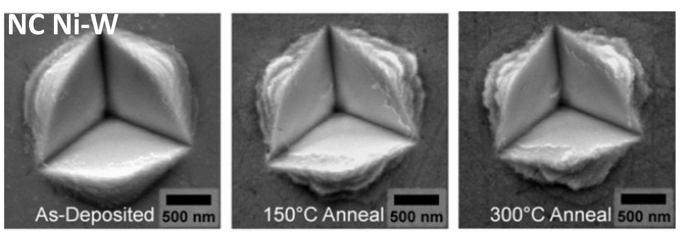
Does the boundary segregation also lower the toughness of the boundary?

Effect of Excess Boundary Free Volume



Irreversible exotherm prior to grain growth during annealing

Detor & Schuh, *JMR*, 2007



Continuous Flow in Unrelaxed State

Shear steps in relaxed boundary

Rupert et al., *JMR*, 2012

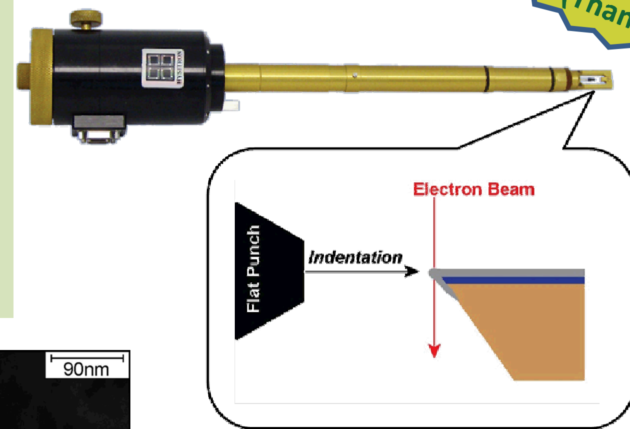
As-deposited grain boundaries are often far from their ideal equilibrium configuration. How does the GB excess entropy affect susceptibility to mechanically-induced grain growth?

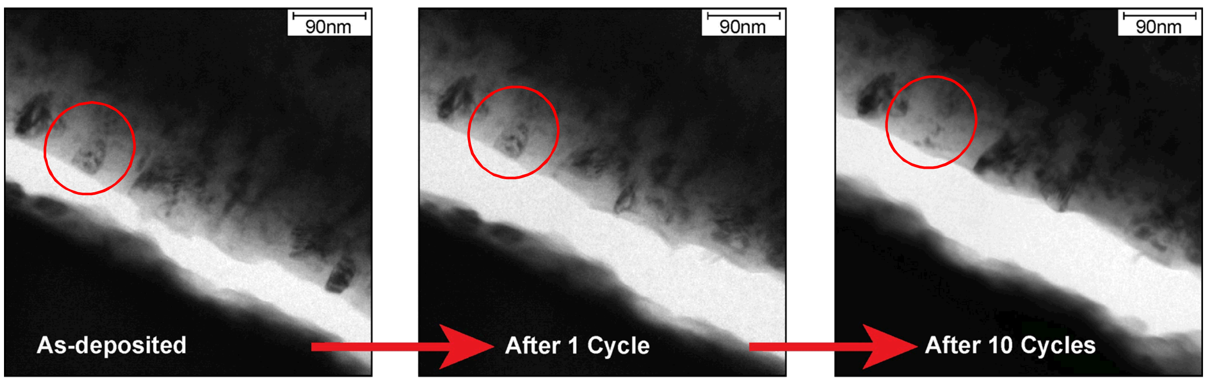
- Direct experimental evidence of the boundary configuration will rely on AC-STEM imaging and inferrence from DSC exotherms during thermal and mechanical testing.
- This question is well-suited to MD simulation.

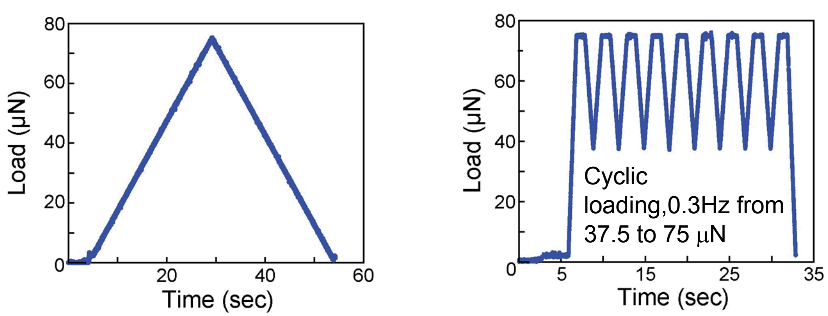
In-situ observation of boundary evolution

Instrument acquired through BES (Thanks!)

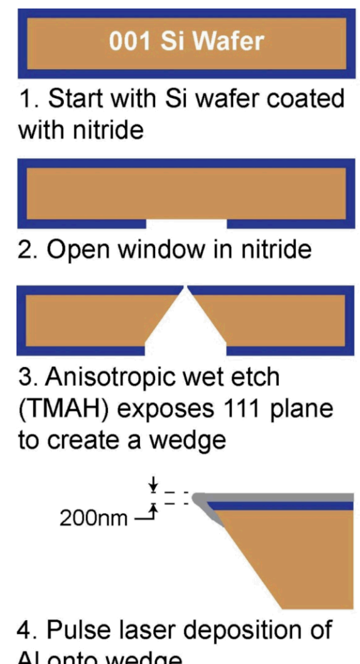
How does cycle-driven grain boundary motion occur? Are there dislocations or other defects that impinge on the boundary to cause its motion? What are the remnant defects left behind?







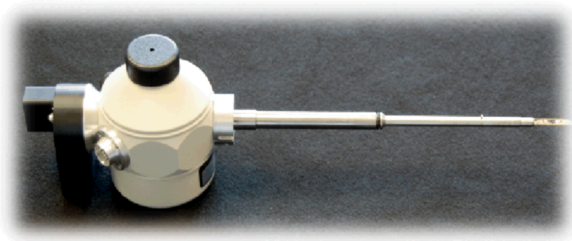
375 MPa nominal contact stress

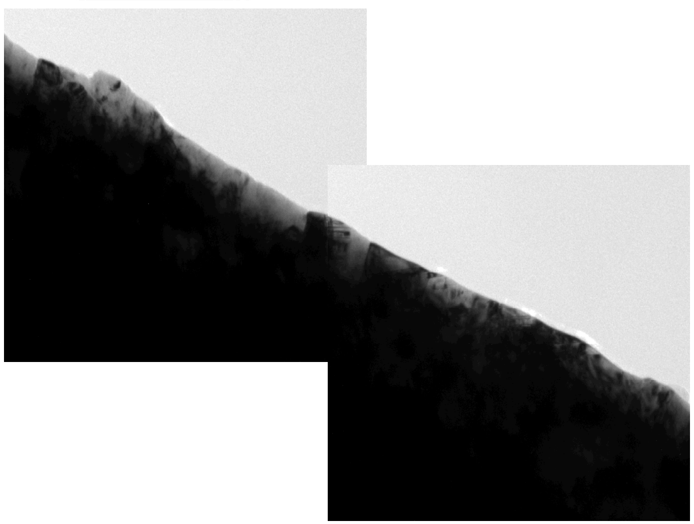


Thermal dependency of mechanically-induced grain growth

Our initial cryogenic indentation studies appear to support the notion that mechanically-induced grain growth occurs more readily at lower temperatures.

In-situ TEM cryoindenter

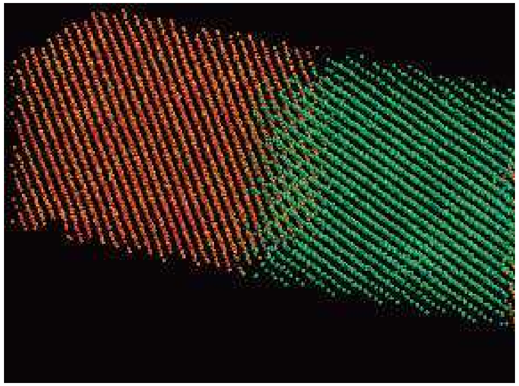


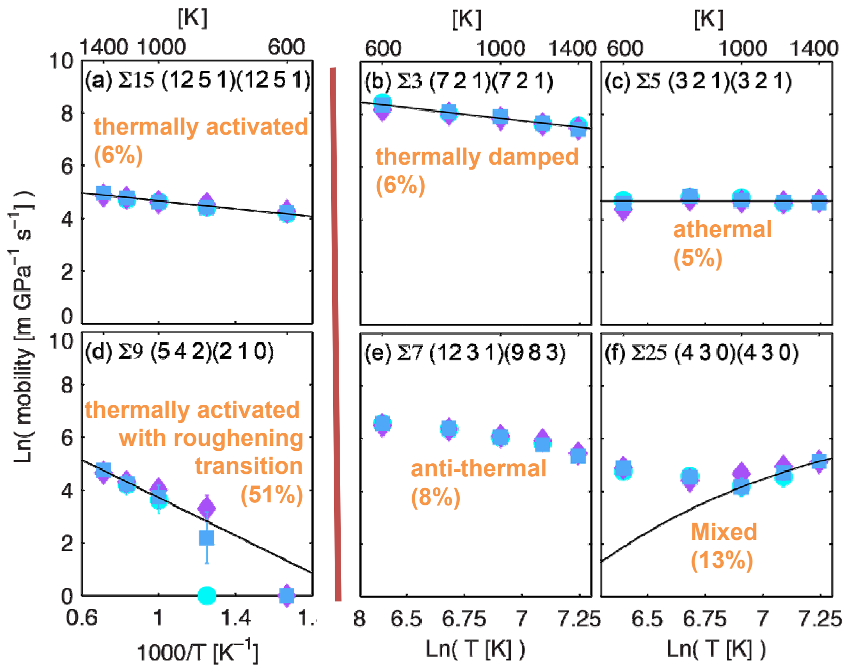


- Does cryogenic grain growth occur during loading, unloading, or return to room temperature?
- Does grain-growth occur during constant sustained monotonic stress? If so, what are the kinetics?

Tools for this study:
TEM cryoindenter (on loan from LANL)
TEM picoindenter with hot stage
Molecular dynamics vs molecular statics

Motion of Planar Boundaries

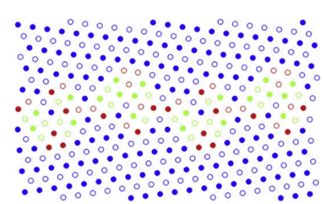




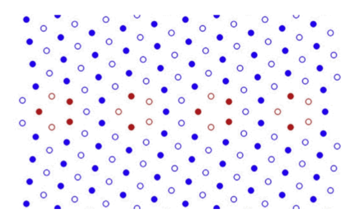
Building off past success, Expand our high-throughput MD capability to address:

- Slow moving boundaries
- Low temperature behavior
- More modest driving forces
- Effects of impurities and grain boundary segregation

Shift focus from surveying trends to identification of atomic mechanisms of boundary motion

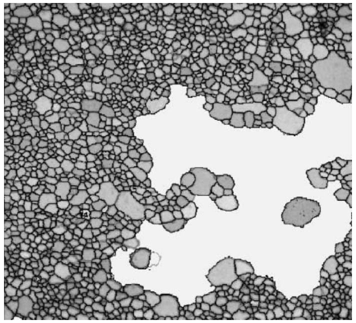


Σ51 16° <1 1 0> tilt

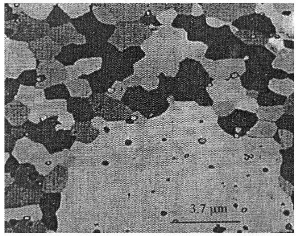


Σ51 164° <1 1 0> tilt

Polycrystalline Boundary Networks: Effect of a network on mobility



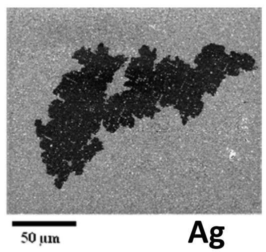
Fe-Si
Etter, *et al.*, *Scripta Mater.* (2002)



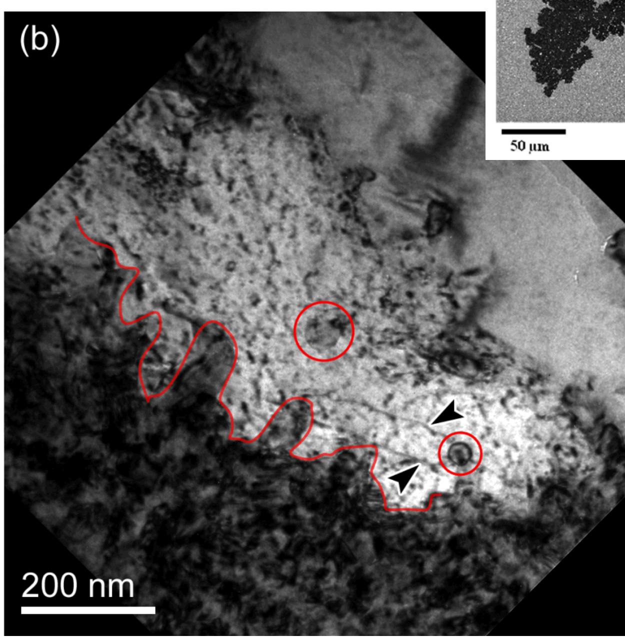
Al-Cu
Calvet, Renon, *Mem. Sci. Rev. Met.* (1960)

How does the microstructural motif and the constraint of interconnected boundaries influence the susceptibility of grain boundaries to mechanically-induced motion?

- In addition to MD modeling, phase field modeling provides insight into these questions.



Ag
Grieser, Mullner and Arzt, *Acta Mater* (2001)



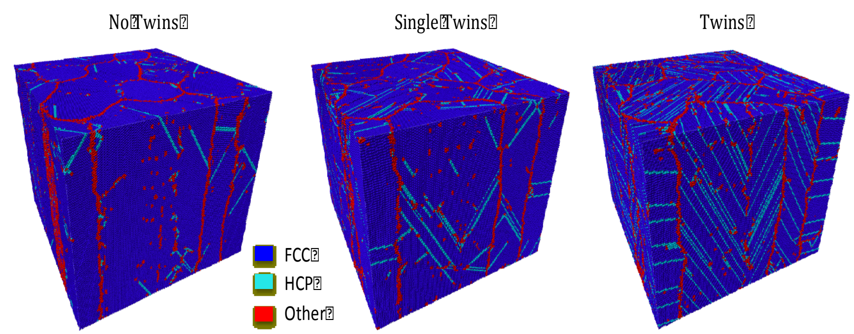


Figure : Three molecular dynamics samples of columnar microstructures created as described in the text that contain varying concentrations of twin boundaries.

Nanocrystalline Ni
B.L. Boyce & H.A. Padilla II, *Metall. Mater. Trans. A* (2011)

Summary: Program Impact

- **Impact: Advancement of Science**
  - 28 journal articles published in the past 3 years (5 more in review)
  - Publications in *Science* (1X), *Adv. Funct. Mater.* (1X), *Acta Mater.* (4X)
- **Impact: Development of People**
  - Postdoc Garritt Tucker accepted professorship at **Drexel**
  - Postdoc Eric Homer is now a professor at **Brigham Young Univ.**
  - Postdoc Henry Padilla is now staff at **Sandia** (batteries)
  - Task lead Liz Holm accepted professorship at **Carnegie Mellon**
- **Impact: Advocacy for the Profession**
  - Liz Holm served as TMS vice president and is now TMS president.
  - Brad Boyce served as chair of the TMS Mechanical Behavior committee and now serves as Structural Materials Division rep.
  - Several (6) international symposia on nanostructured metals organized or co-organized by our team (MRS, TMS, Plasticity).

More Information on the Posters…

MD Simulation of Grain Boundary Motion Identifying the Complexity

Stephen Foiles, Elizabeth Holm, Eric Homer, David Olmsted, Garritt Tucker, Brad Boyce

Fatigue and Wear of Nanocrystalline Ni

Brad L. Boyce, Henry A. Padilla II, John A. Sharon, Khalid Hattar, Blythe G. Clark

Investigating Thermal Stability of Tailored Nanocrystalline Ni Thin Films

K. Hattar, S. Rajasekhara, B.G. Clark, J. Kacher, and I.M Robertson