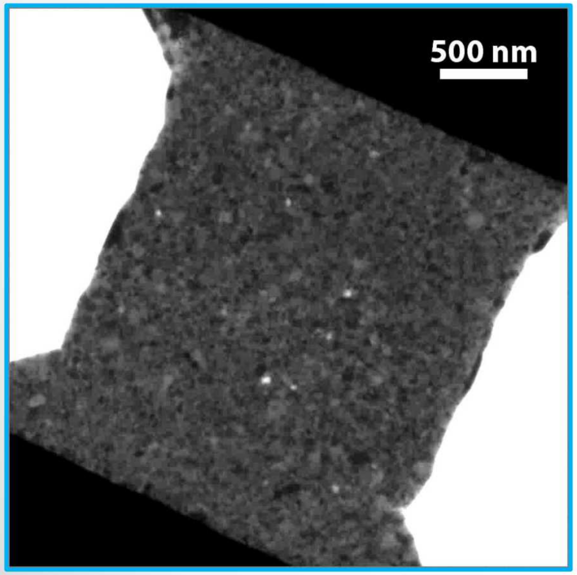
SAND2018-2851PE

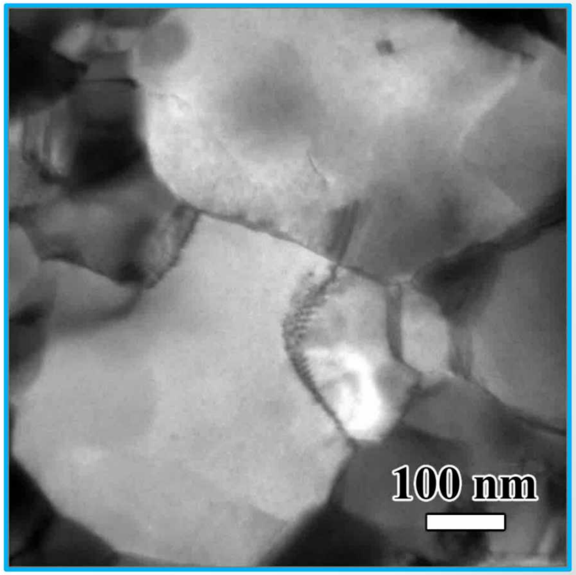
Sandia National Laboratories

Structural evolution in extreme environments: real-time nanoscale observations of radiation damage, deformation, and fatigue



Daniel Bufford*
Sandia National Laboratories
Albuquerque, NM, USA

*and many collaborators!



U.S. DEPARTMENT OF ENERGY NNSA National Nuclear Security Administration

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. SAND2016-10557 PE

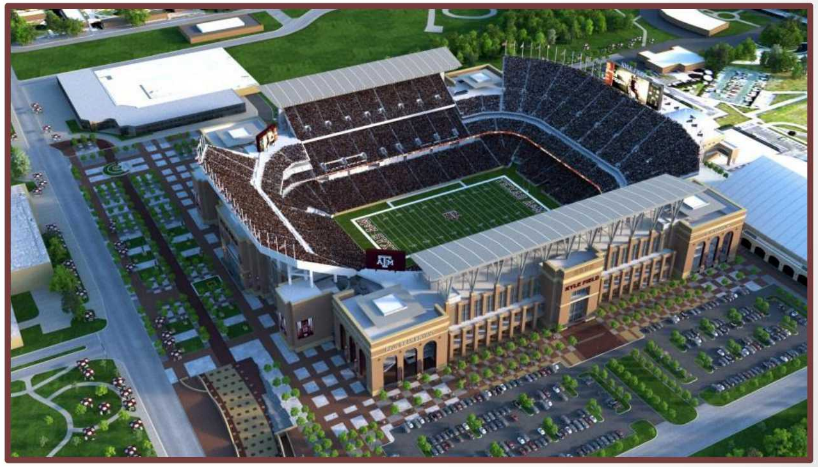
6 March, 2017

My Background

Sandia National Laboratories

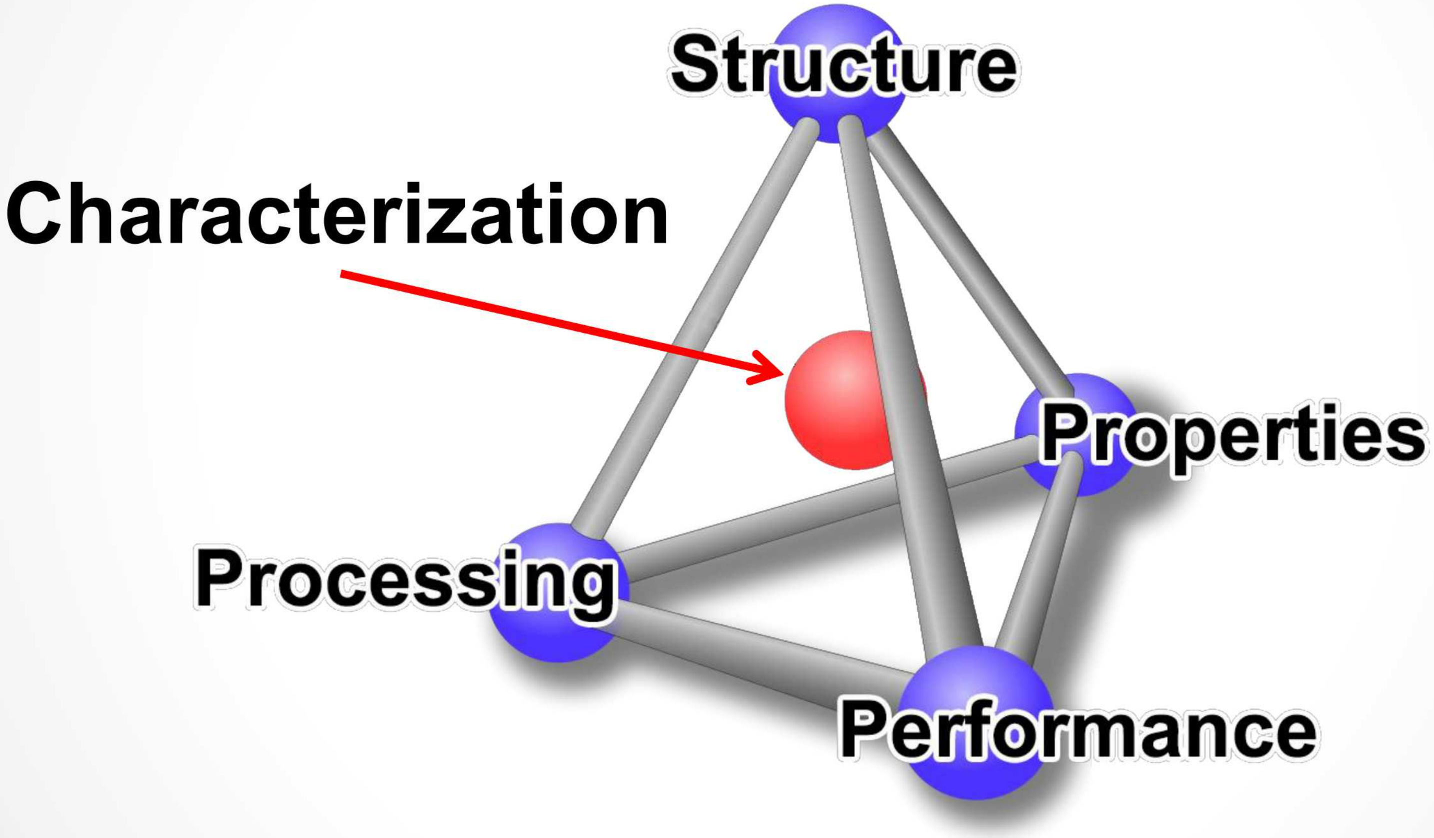
Olin College of Engineering

TEXAS A&M UNIVERSITY



Sandia National Laboratories

The Heart of Materials Science and Engineering



Microstructure-processing-property relationships define the field!

Microstructures

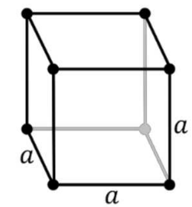
1 nm

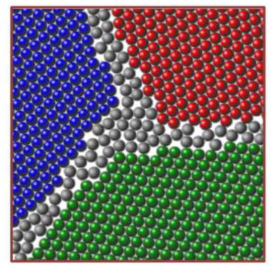
10 nm

100 nm

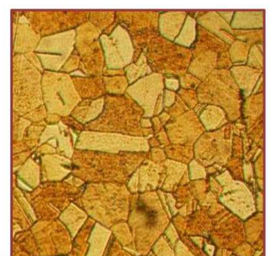
1000 nm

Stannered, 2007, *via* Wikimedia Commons.

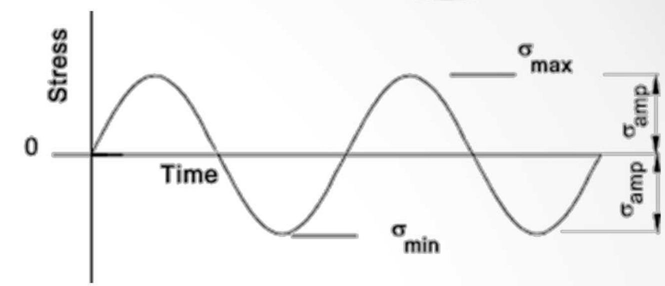


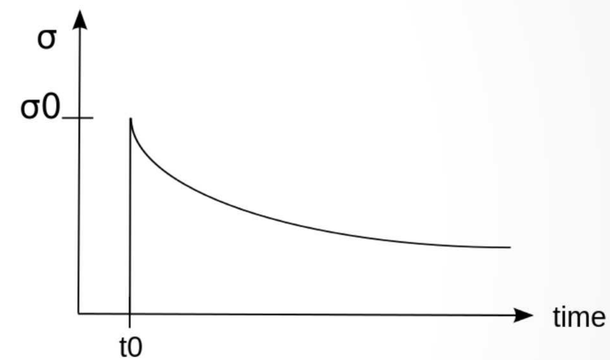


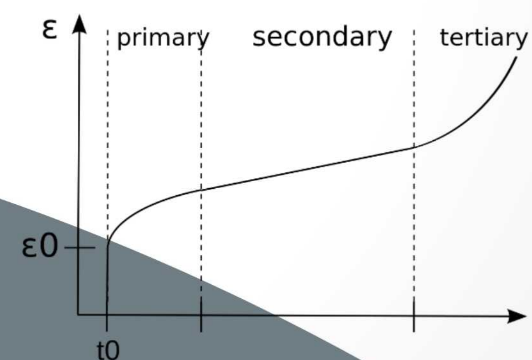
Strangerhahaha, 2008, *via* Wikimedia Commons.



Vassil, 2008, *via* Wikimedia Commons.



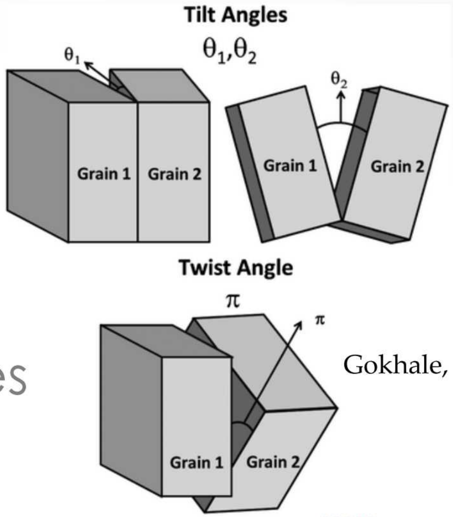




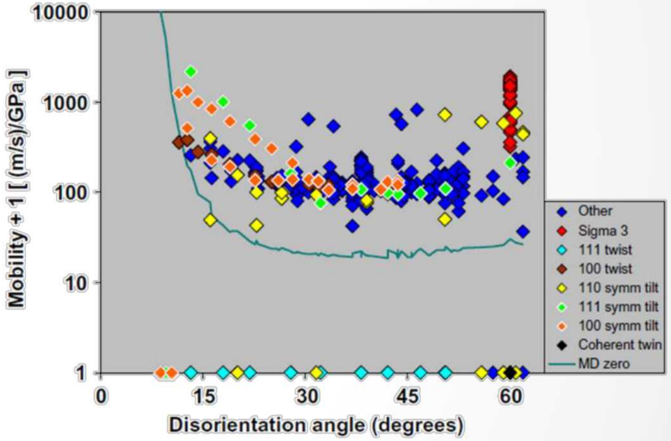
How does structure affect mechanical properties, and how do both change over time?

At the grain boundary level

- Slip transfer and modification of boundaries
- Fatigue-induced grain boundary migration and coarsening
- Radiation-induced grain boundary migration and coarsening



Gokhale, *et al*, Soft Mater, 2013.

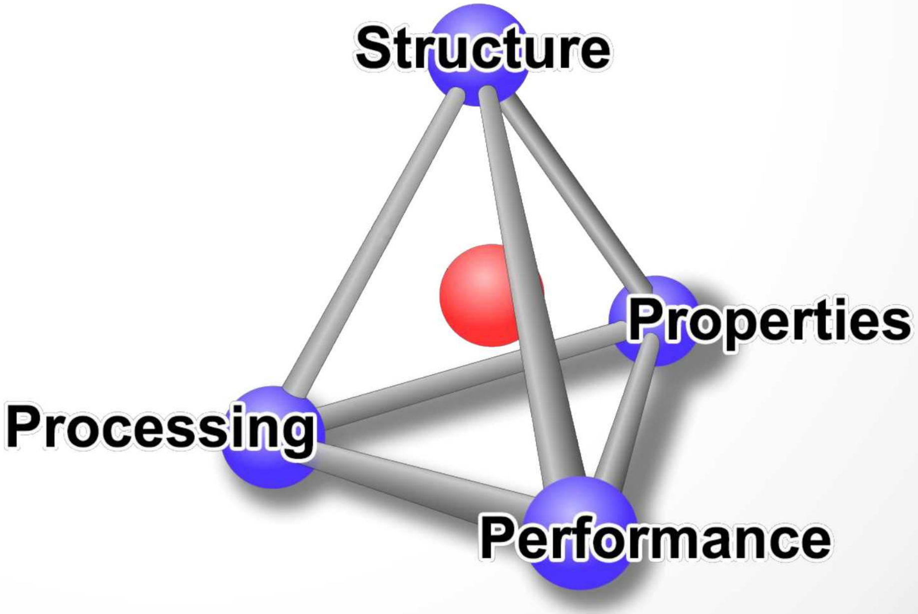


Olmstead, *et al*, Acta Mater, 2009.

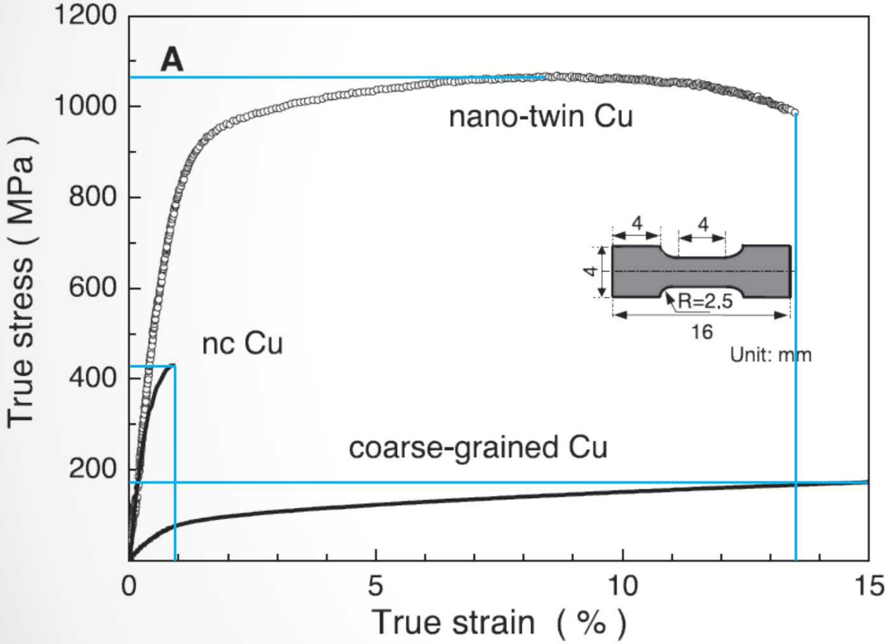
How do we quantify these processes in nanocrystalline metals, and what do we learn in doing so?

Roadmap

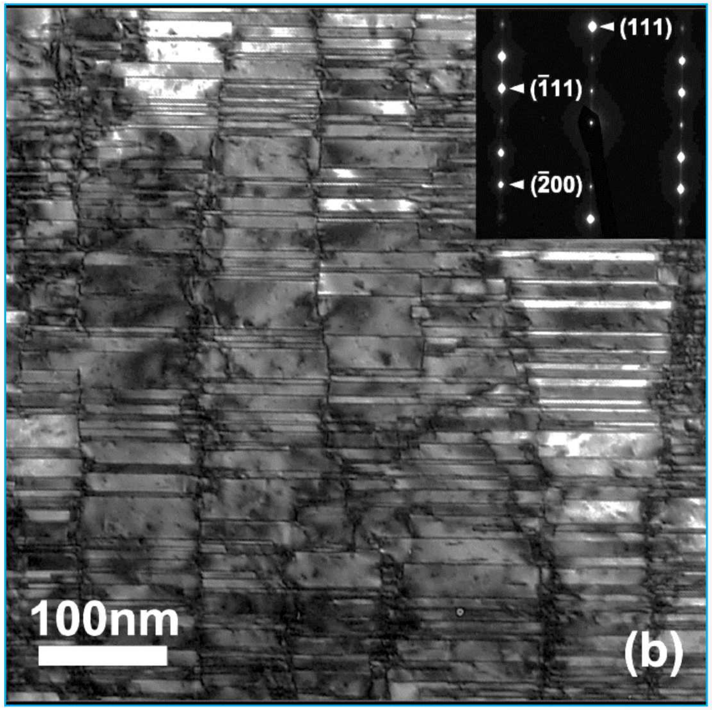
- Mechanical Loading
  - Grain boundary – dislocation interactions
  - Nanoscale Fatigue
- Irradiation
  - Grain growth



Slip transfer in nanotwinned metals



Lu, et al, *Science* 2004.



Bufford, et al. *Acta Mater*, 2011.

- Microstructures dominated by twins spaced <100 nm
- High mechanical strength
- Considerable plasticity

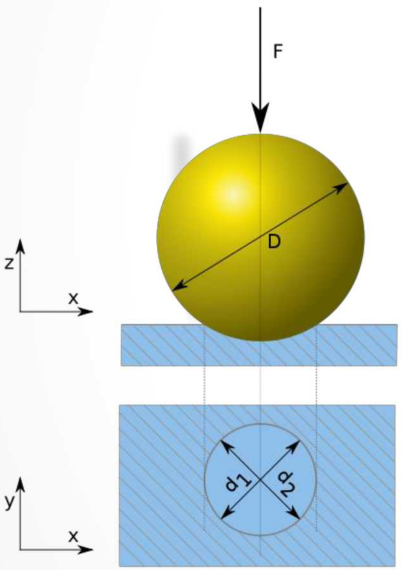
What is happening at the grain boundary level?

Macro/Microindentation

Sandia National Laboratories

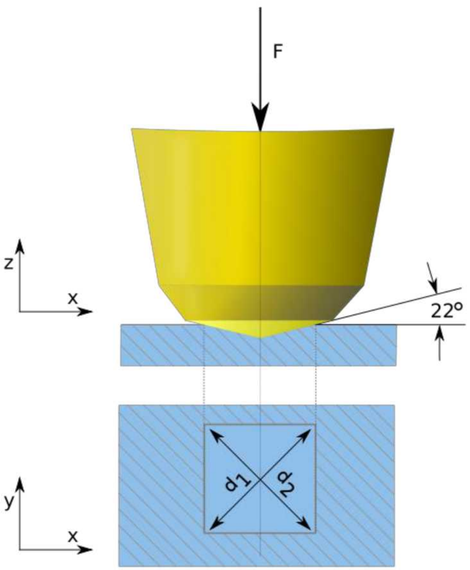
- Apply a static load
- Measure residual indentation area
- Depths from tens of μm to mm

Lokilech, *via* Wikimedia Commons.



Brinell

A1, *via* Wikimedia Commons.



Vickers

Diamond Vickers Tip

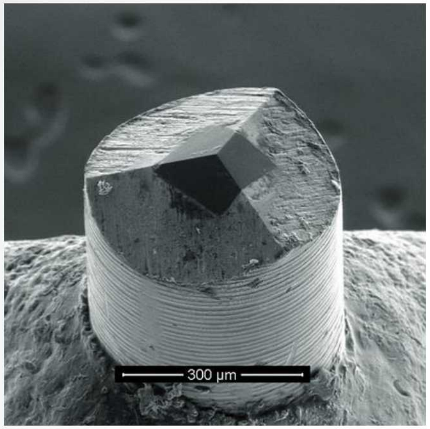
R. Tanaka, *via* Wikimedia Commons.

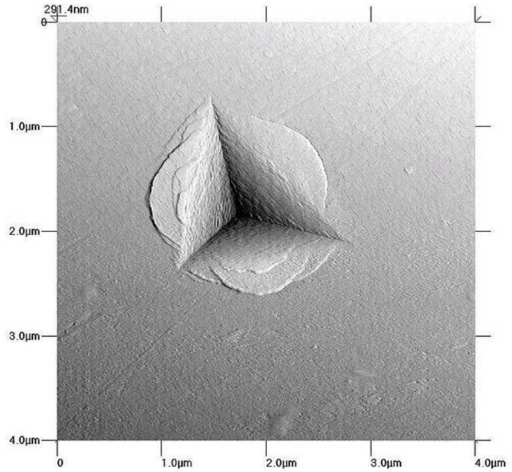
- Hardness, $H = \frac{P_{max}}{A_r}$

Infer bulk properties from local resistance to plastic deformation in “small” volumes.

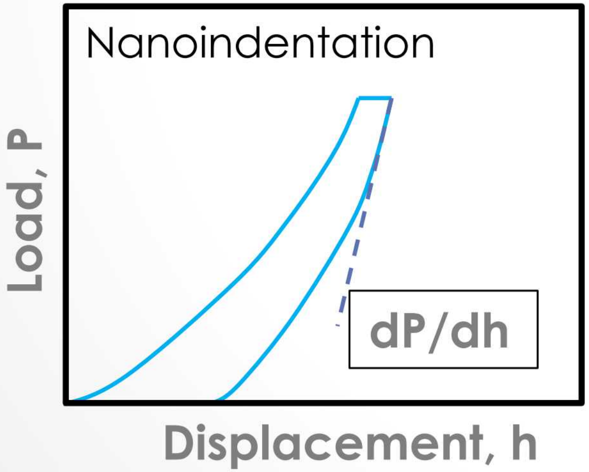
Nanoindentation

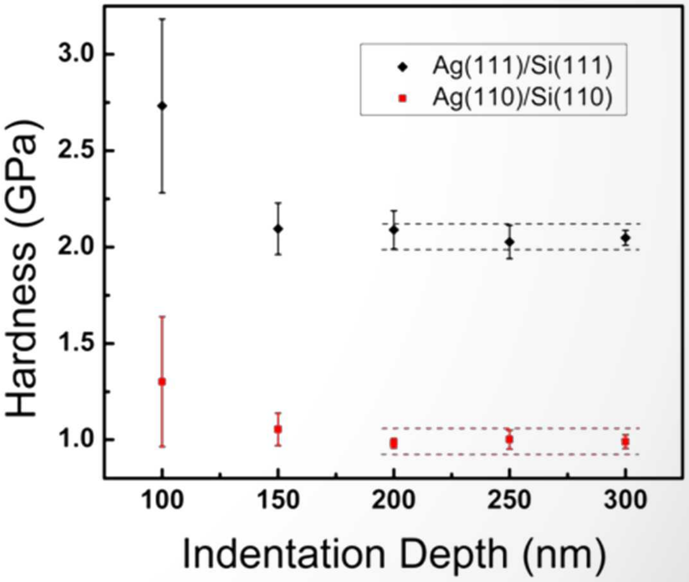
Sandia National Laboratories





J. Puthoff, *via* Wikimedia Commons.

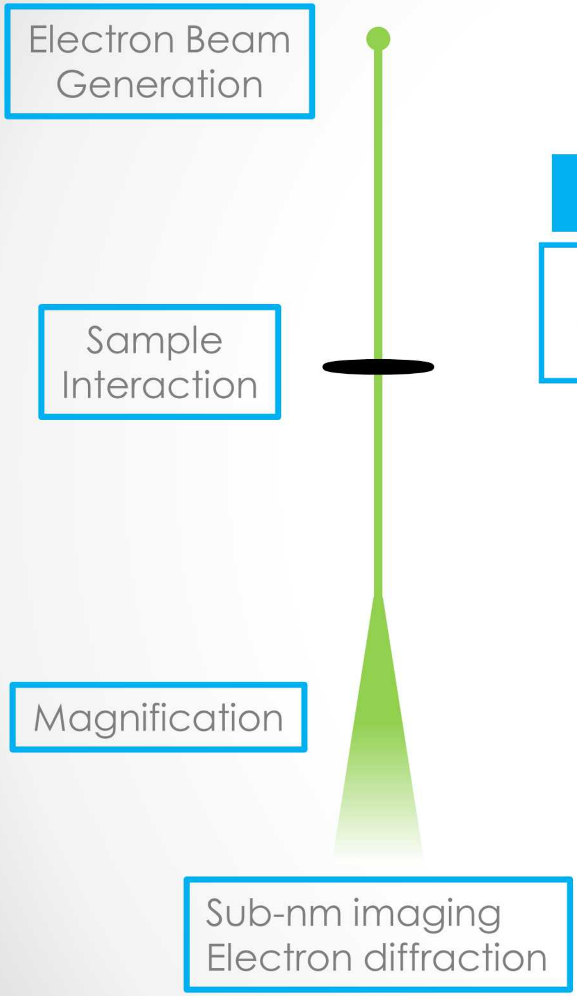




Quantified mechanical response at the nanoscale.

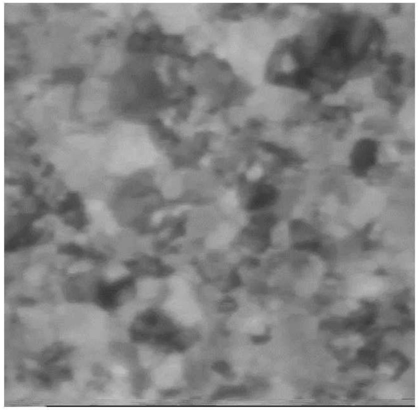
TEM and *In Situ* Experiments

Sandia National Laboratories



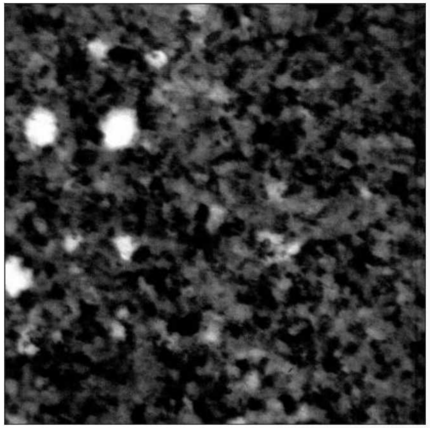
Thermal

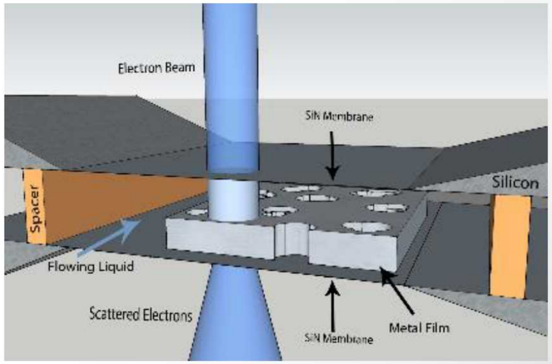
Effects of temperature on microstructural evolution up to 800 °C



Environmental

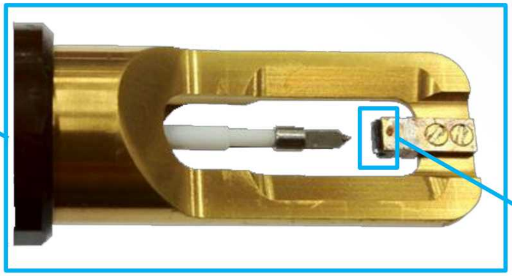
Effects of corrosion and gas loading at the grain level

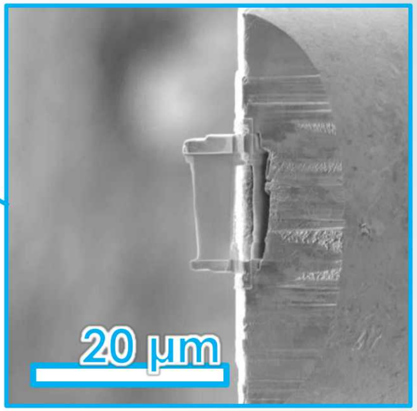




Enables real-time visualization of samples under various stimuli

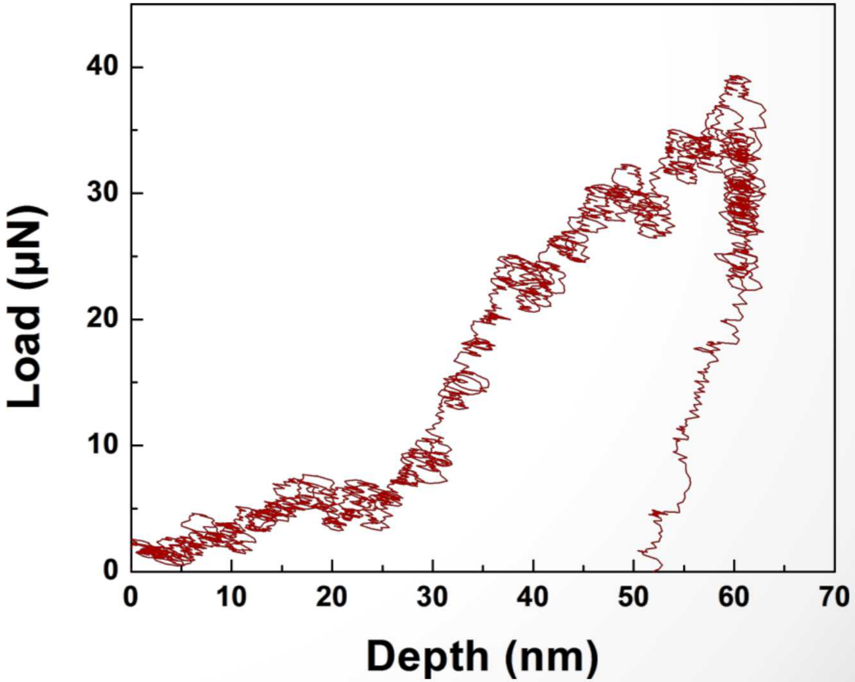
In Situ Nanoindentation





Hysitron PI95 *In Situ* Nanoindentation TEM Holder

- Sub nanometer displacement resolution
- Quantitative force information with µN resolution



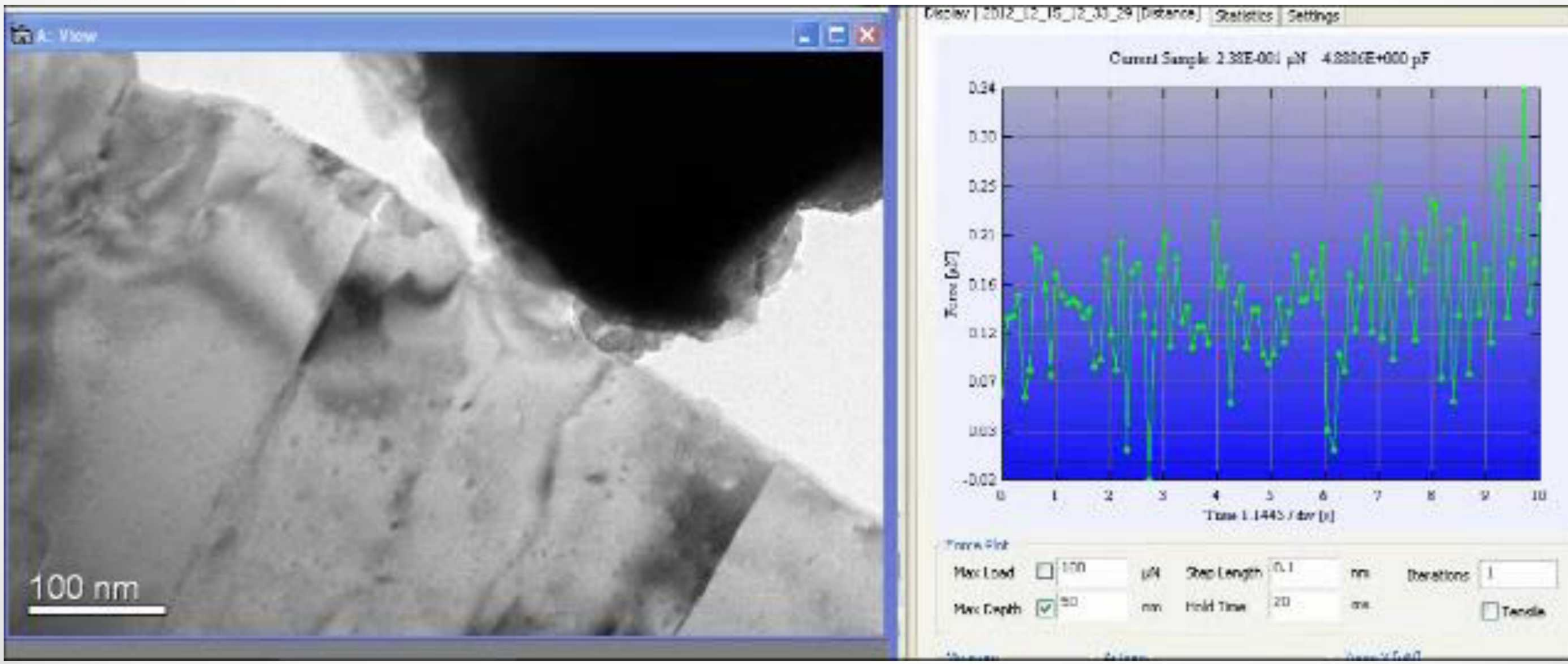
Direct correlation of mechanical loading and defect behaviors.

Plasticity in Al

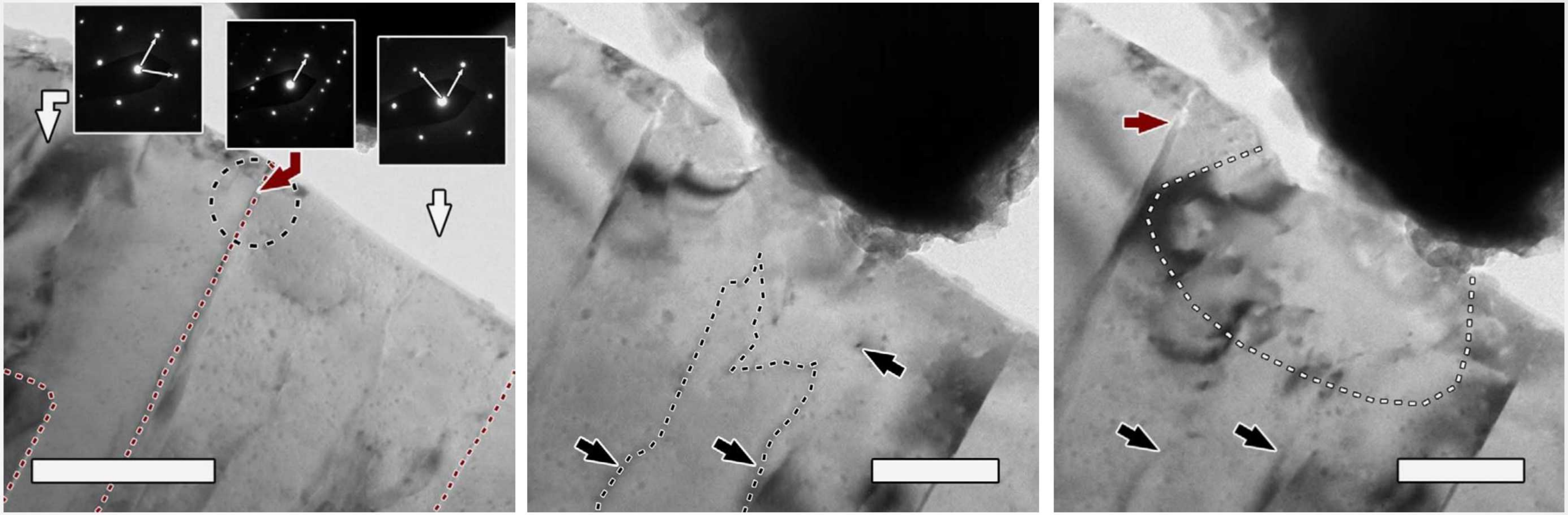
Sandia National Laboratories

- *In situ* nanoindentation of Al near a Σ3{112} twin boundary
- Initial cycle

Video playback ×3



Plasticity in Al



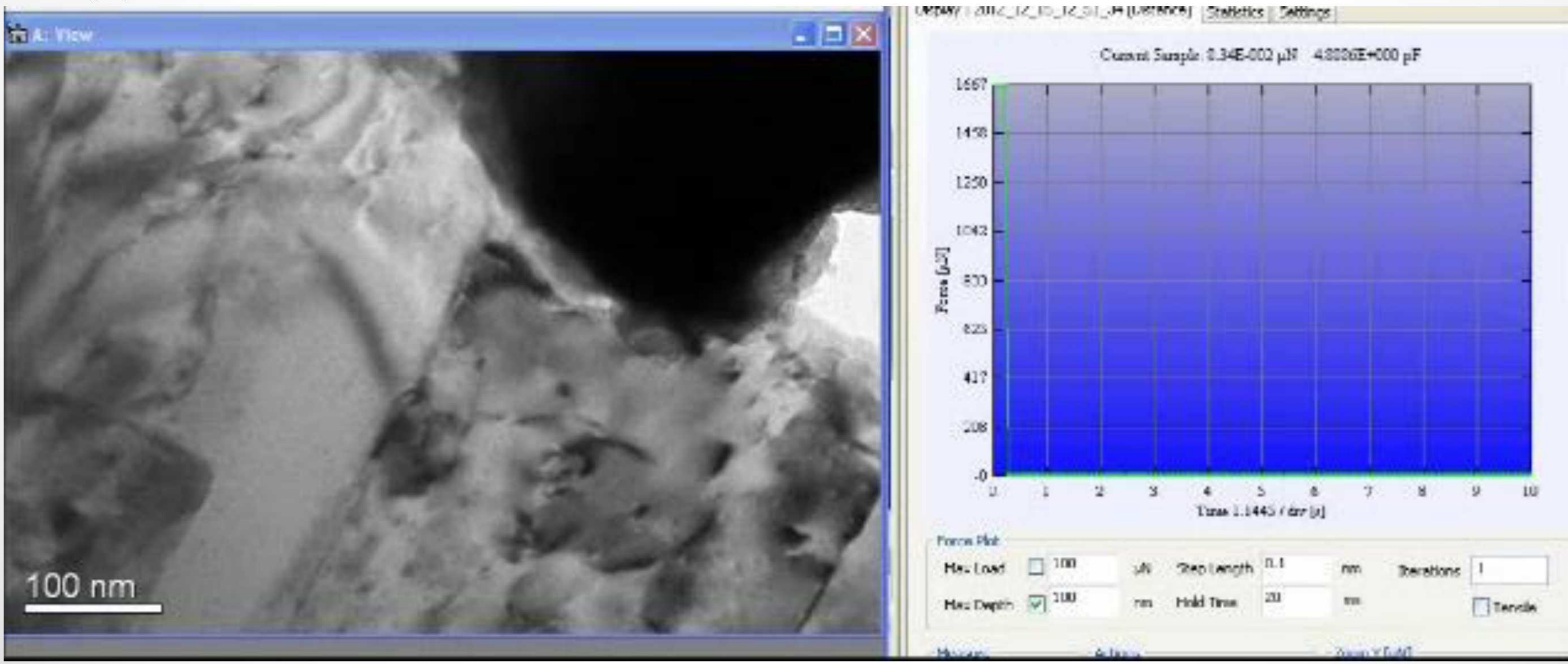
- Grains initially contain threading dislocations and small dislocation loops
- Movement of existing dislocations observed
- Deformation confined by twin boundaries to single grain

Confinement of dislocation activity suggests barrier (Hall-Petch) strengthening.

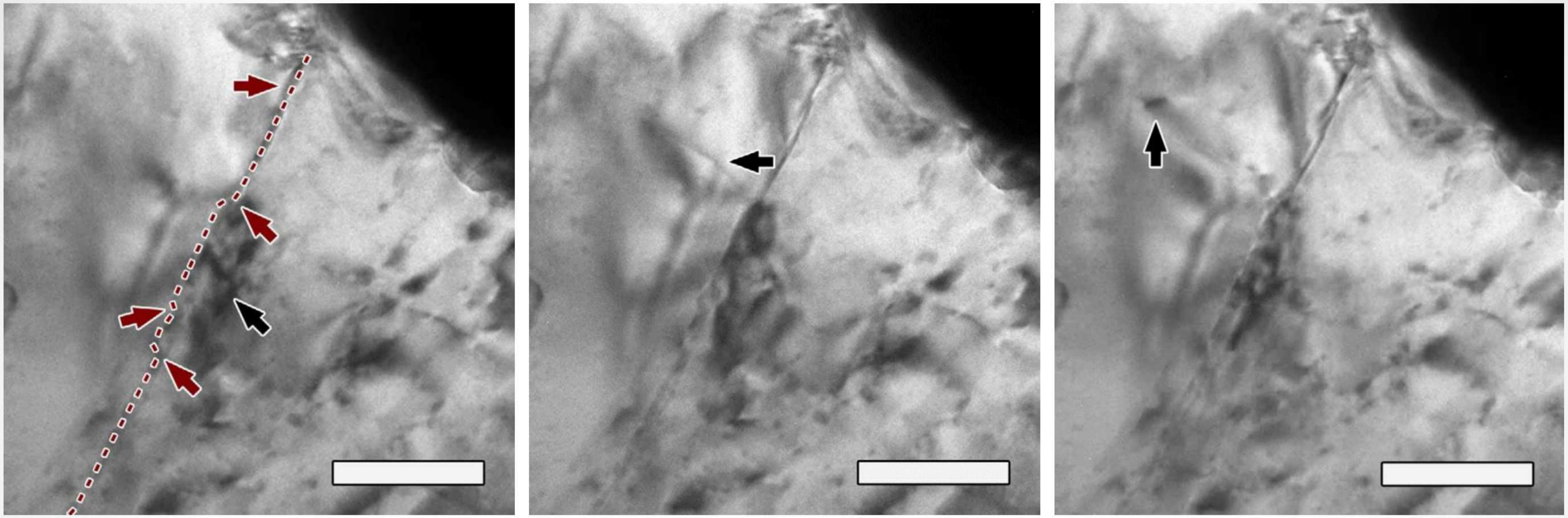
Dislocation Transmission

- In situ nanoindentation of Al near a Σ3{112} twin boundary
- Cycle #4, after 3 previous cycles to progressively higher loads

Video playback ×3



Dislocation Transmission



Bufford, *et al.*, Nat Commun 2014.

- Boundary deformed by dislocation interactions in previous cycles
  - But no obvious plastic deformation in adjacent grain
- First *observable* plasticity event in second grain captured
- Measured forces associated with the event

Known bicrystal geometry and quantitative force measurements provide bounds for determining likely dislocation reactions and estimating associated local stresses.

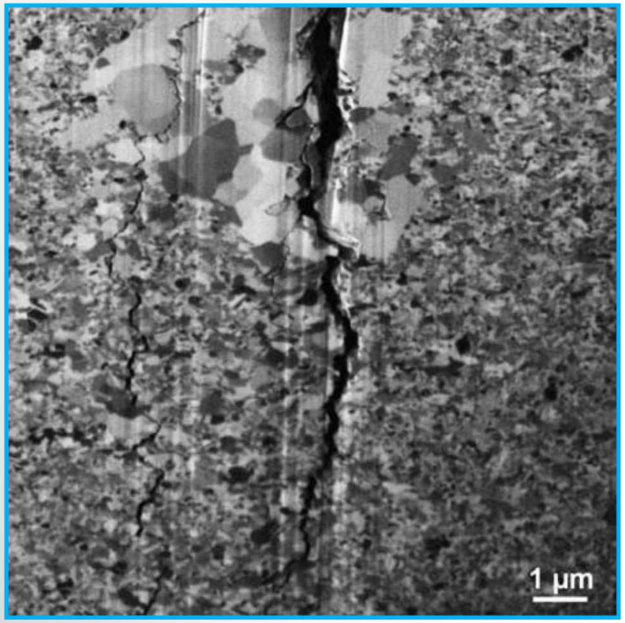
Slip transfer in nanotwinned metals

- Grain boundary structure change resulting from dislocation pile-up
- Dislocation transmission appears to be spatially correlated with defects formed in the boundary

Fatigue-induced grain growth

Fatigue: Progressive microstructural change with cyclic loading

Execcharter, 2011.



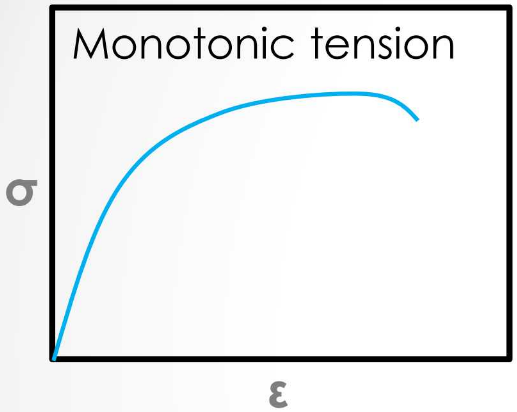
Padilla and Boyce, Exp Mech 2006.

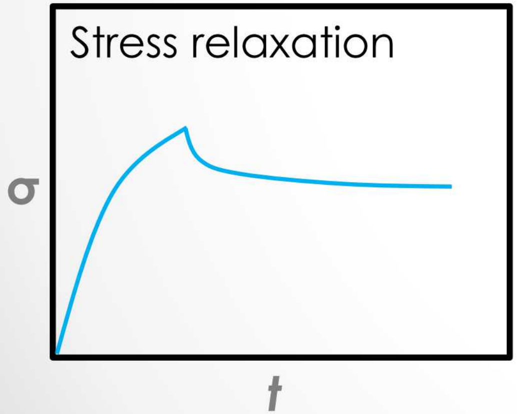
In nanocrystalline metals…

- Locations of grain growth and cracks are correlated
- Substantial hurdle to practical adoption of these materials

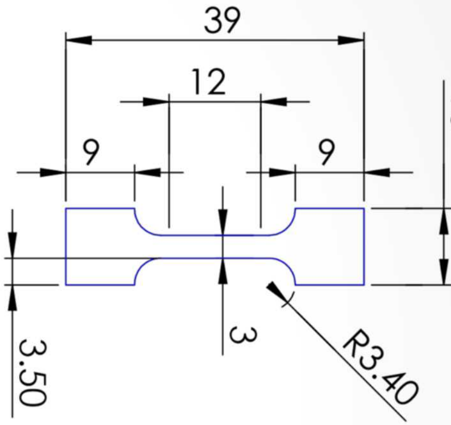
Which one comes first?

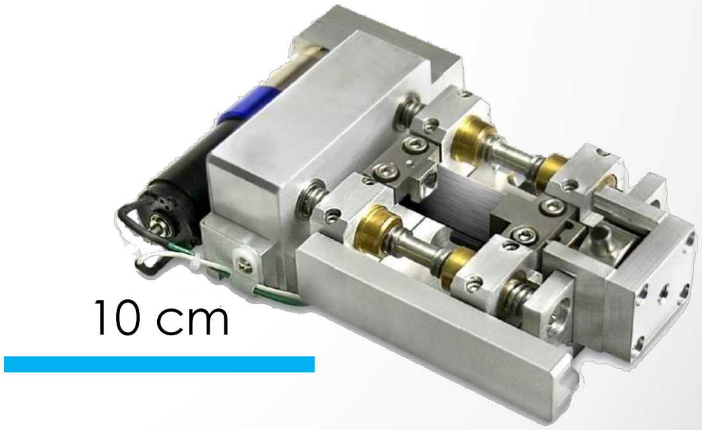
Tensile Testing





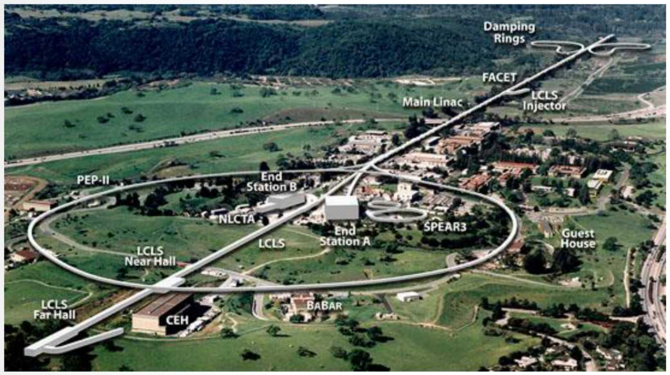
- Lots of information:
  - E, σ_y, σ_{UT}, elongation, toughness, n, m
- More information:
  - m, ΔV, creep

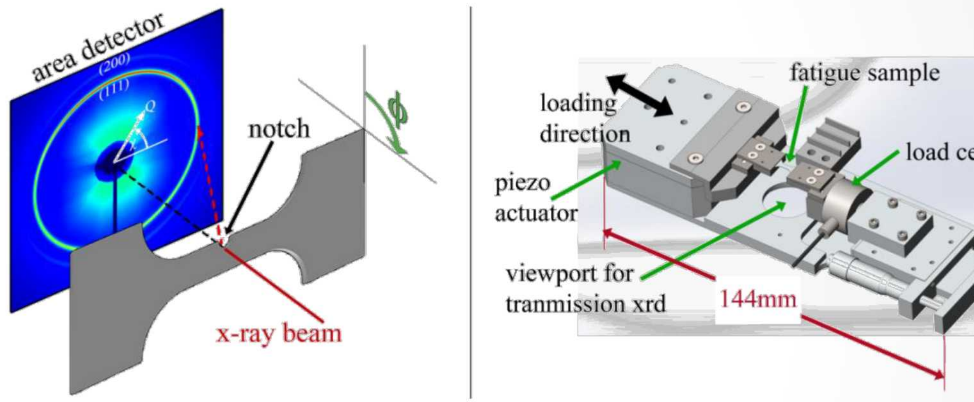




Bulk Approach

Collaborators: A. Mehta, D. Van Campen, T.A. Furnish, B.L. Boyce





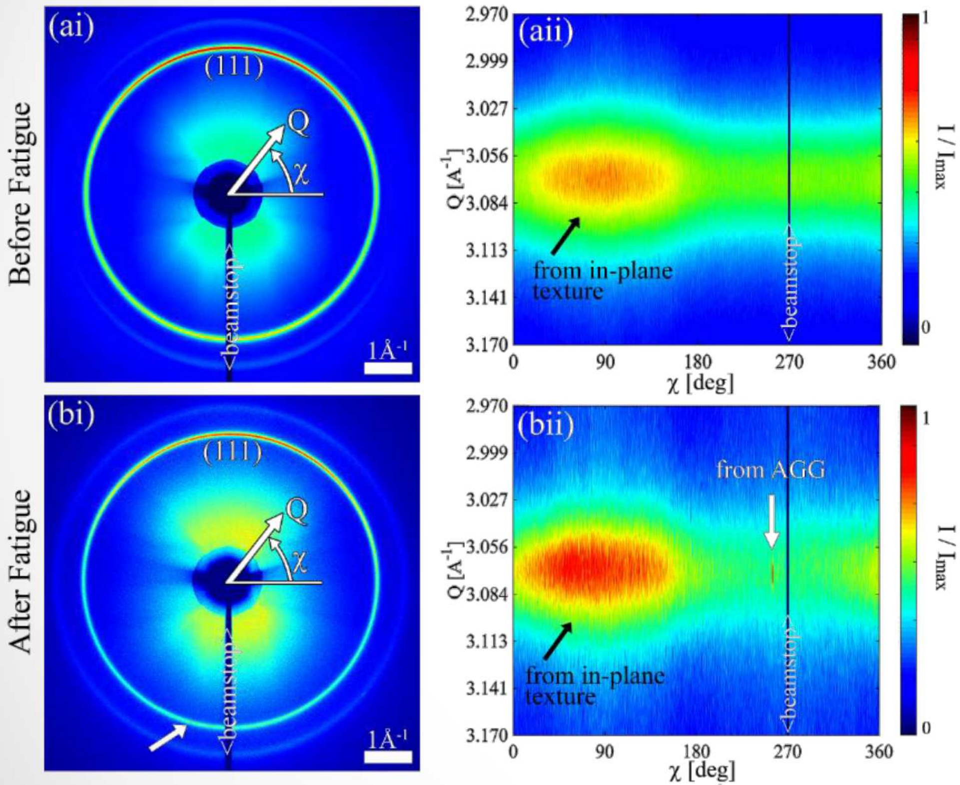
Furnish, et al, J Mater Sci 2017.

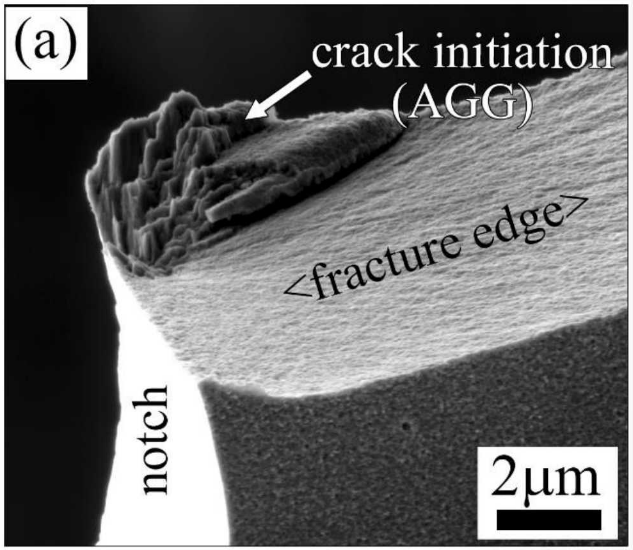
- Fatigue performed *in situ* during X-ray diffraction analysis
- Experiments performed at the Stanford Synchrotron Radiation Lightsource (SSRL)

Which comes first, grain growth or crack initiation?

Bulk Approach

Collaborators: A. Mehta, D. Van Campen, T.A. Furnish, B.L. Boyce





Furnish, et al, J Mater Sci 2017.

- Patterns processed and analyzed for outliers
- Fatigue-induced grain growth captured before failure

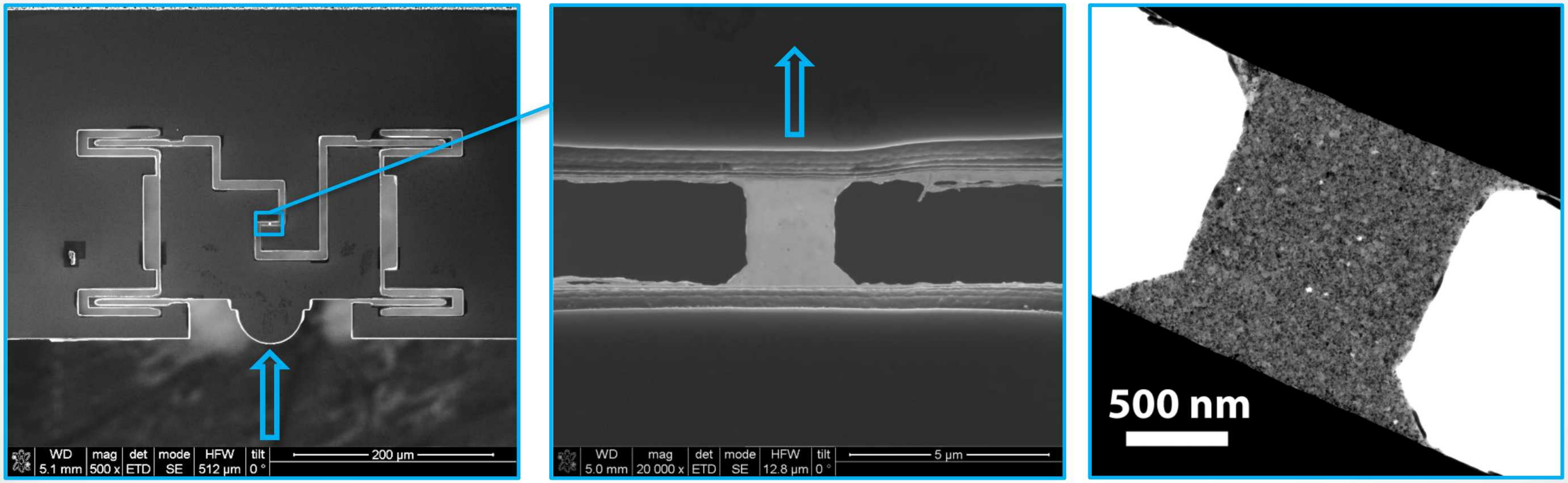
- **Detection of abnormalities in only ~0.00001% of the sampled volume!**
- **Analysis of fracture surface indicates that grain growth preceded crack initiation**

Small-Scale Approach

Sandia National Laboratories

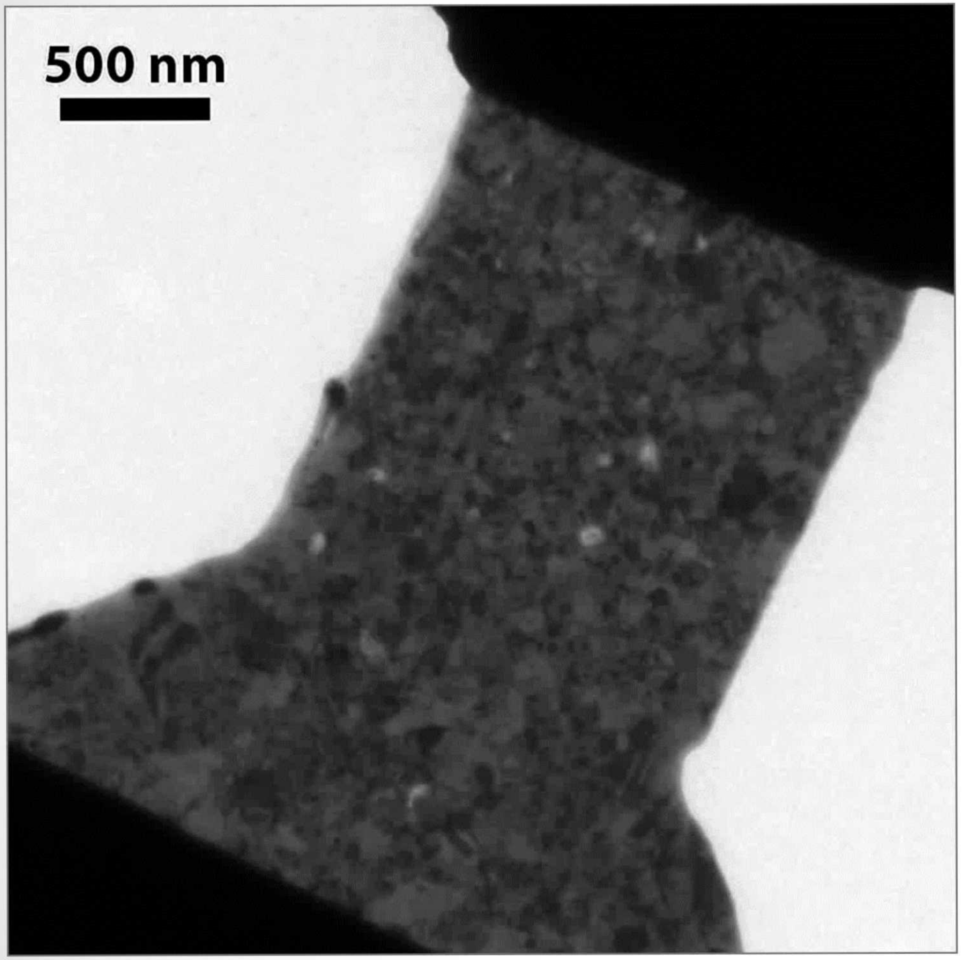
- "Push-to-Pull" devices
  - Commercial (Hysitron) microfabricated Si test frames
  - Cu film (75 nm) floated onto device, then FIB milled

Collaborators: D. Adams, K. Hattar, W. Mook, C. Sobczak

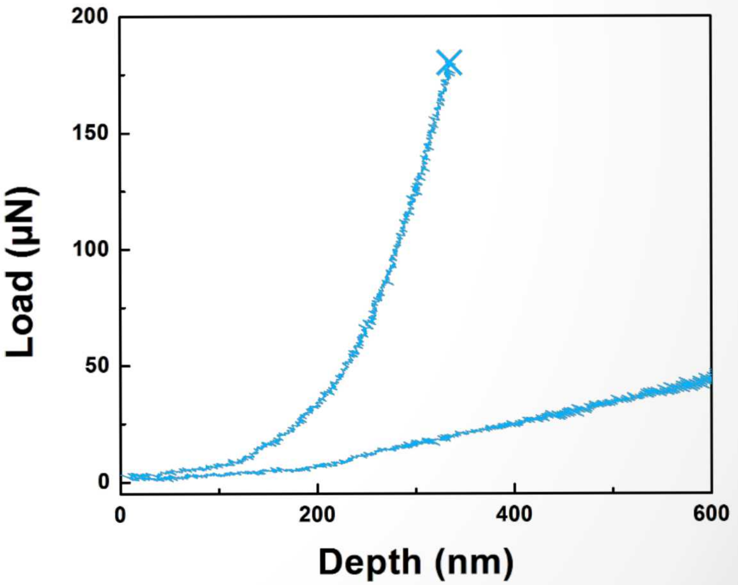


- Thin foil geometry not ideal for mechanics, but is electron-transparent
- Nearly pure tension, uniform cross sectional area, stable load frame

Monotonic

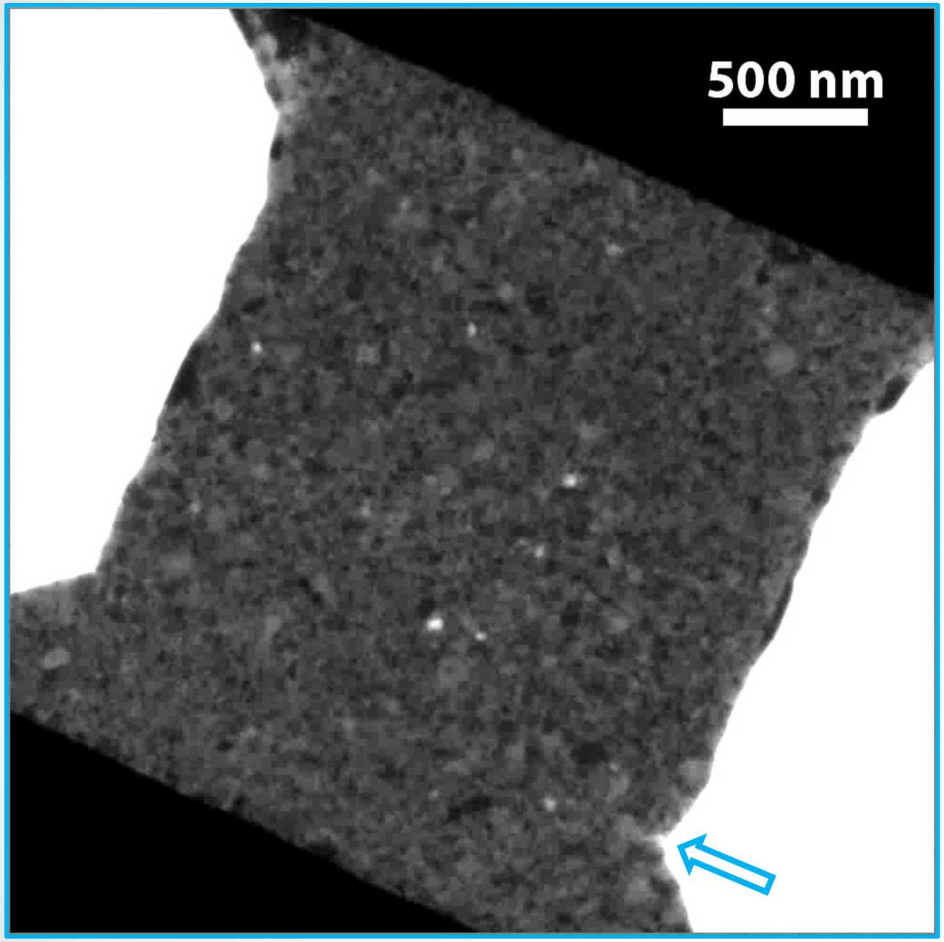


- Monotonic loading
  - Negligible plasticity before failure
  - Rapid crack propagation



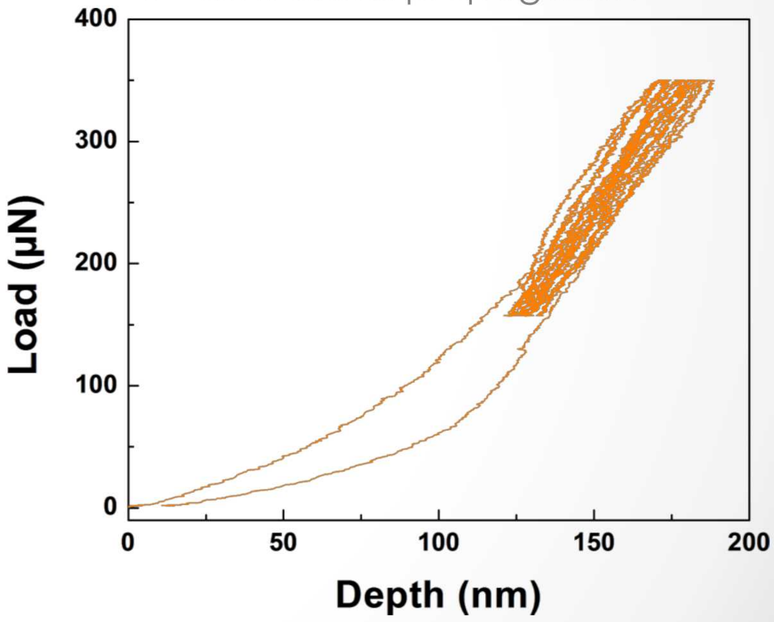
Successful quantitative tensile testing of a micrometer-scale sample!

Low Cycle Fatigue *In Situ*



Video playback ×10

- Cyclic loading:
  - Crack initiated in previous monotonic test
  - 9 cycles to ~87.5% of that load
  - 50 % unloading
  - Slow crack propagation



- **Direct measurements of crack growth**
- **Little structural evolution at the crack tip**

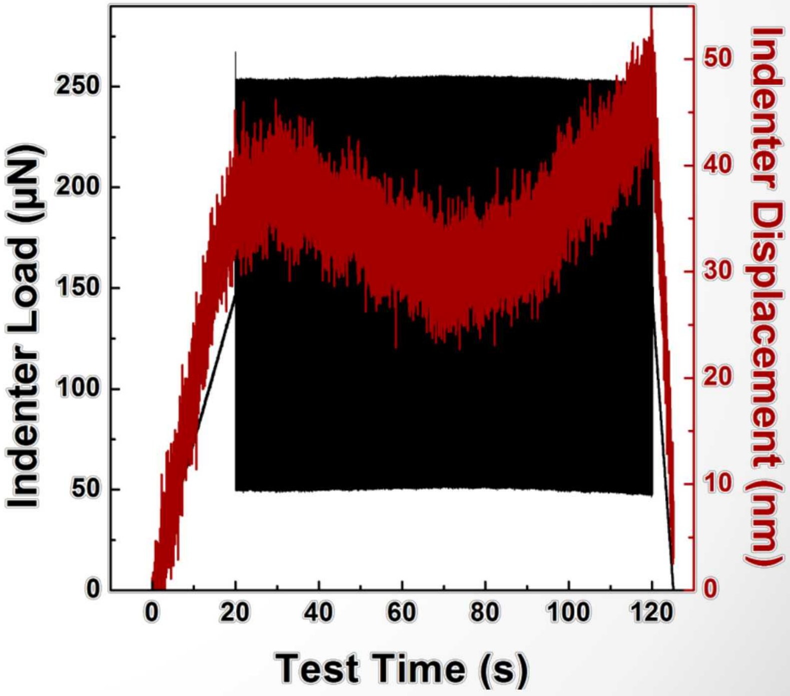
High Cycle Fatigue *In Situ*

Sandia National Laboratories

Collaborators: S.A. Asif, D. Stauffer, B. Boyce, K. Hattar, W. Mook

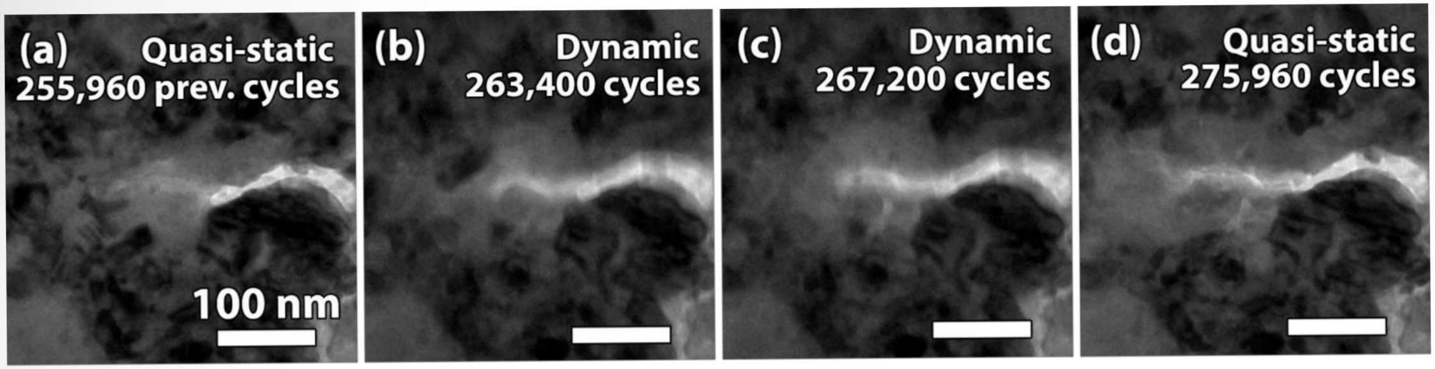
Video playback ×3

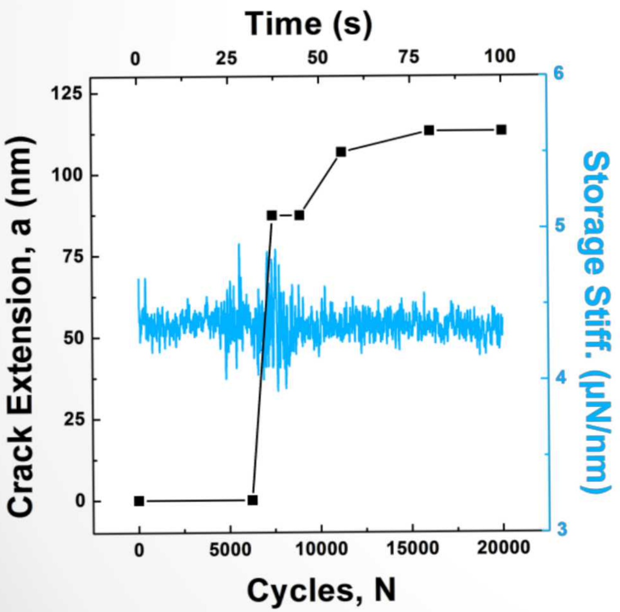
- Cyclic loading:
  - 200 hz
- Structural change at crack tip captured

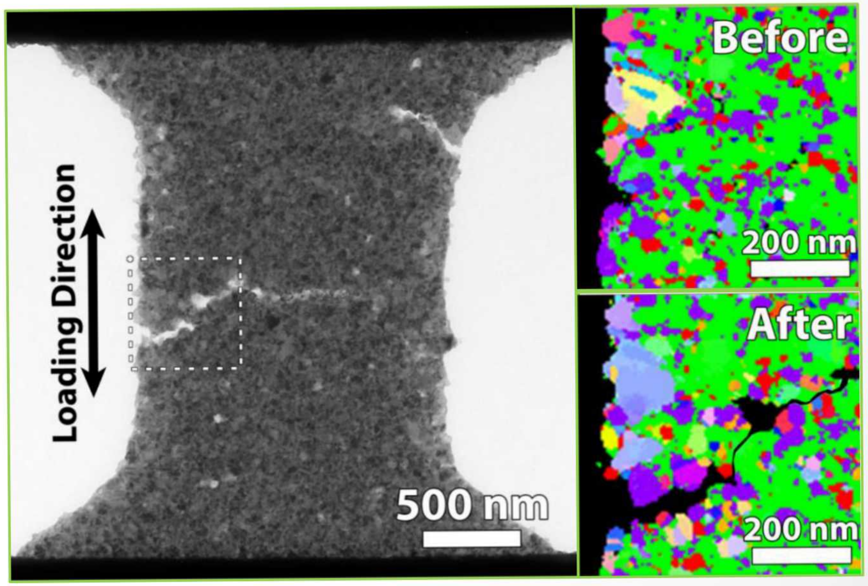


Crack propagation and grain boundary activity captured

Crack Growth Quantified







Bufford, et al, Nano Lett 2016.

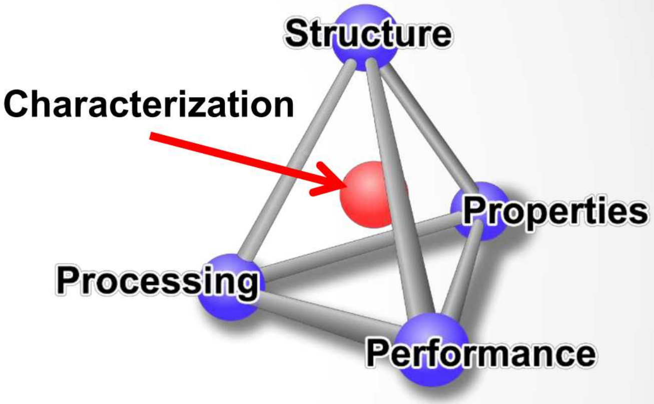
- Average crack growth rate measured at 6×10^{-12} m/cycle!
- Evidence of fatigue-induced grain growth.

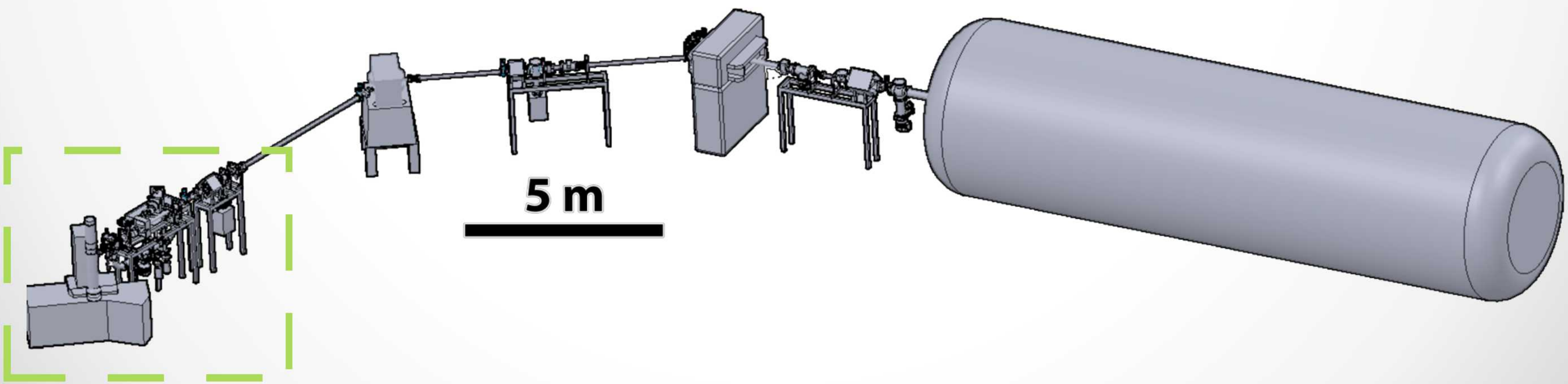
Fatigue-induced grain growth

- Captured local fatigue-induced structural changes as they occur
- Ongoing work to reveal orientation dependencies at the grain boundary level

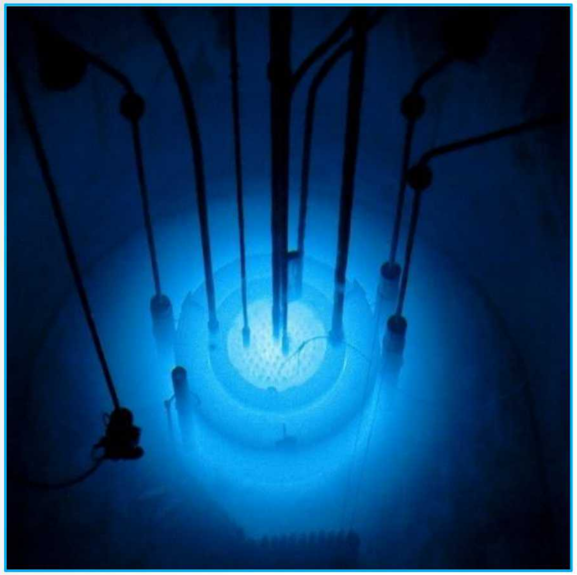
Roadmap

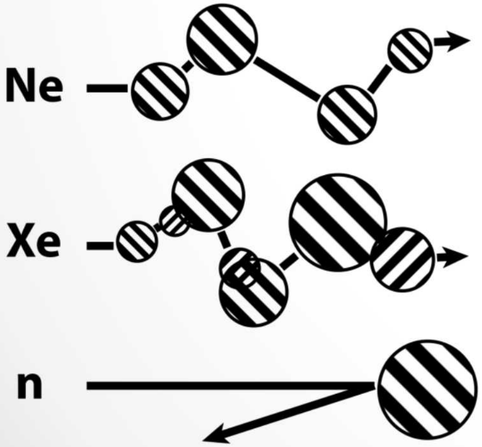
- Mechanical Loading ✓
  - Grain boundary – dislocation interactions
  - Nanoscale Fatigue
- Irradiation
  - Grain boundary migration





Radiation-Induced Grain Growth

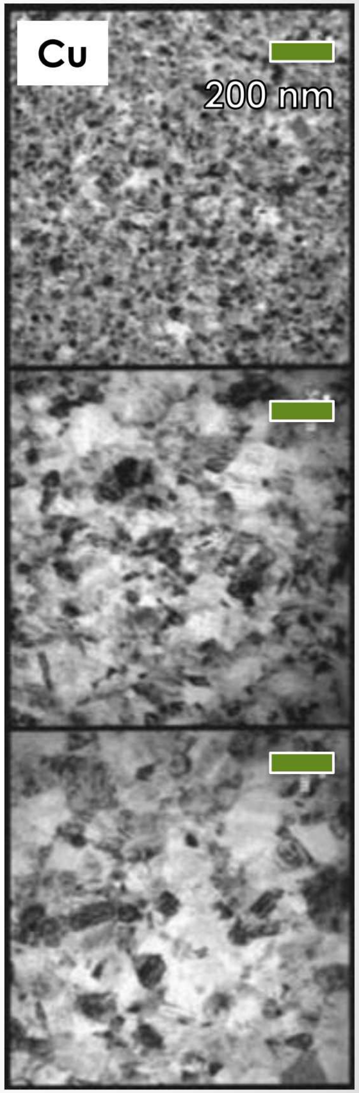




Averback, J Nucl Mater, 1994.

Nanocrystalline metals:

- Exemplary mechanical properties
- Abundant sinks for structural and chemical defects
- Ideal candidates for radiation-tolerant materials?



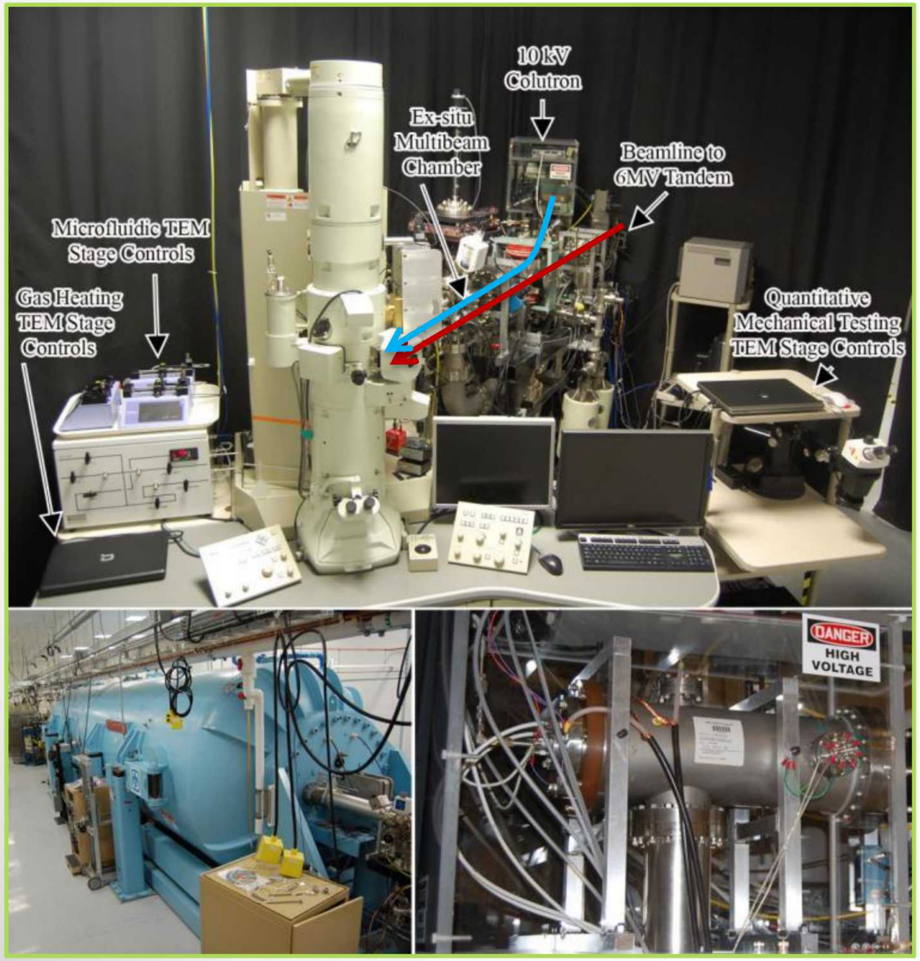
Kaoumi, *et al*, J ASTM Intl, 2006.

What are the relationships among ion damage, grain boundary character, and grain growth?

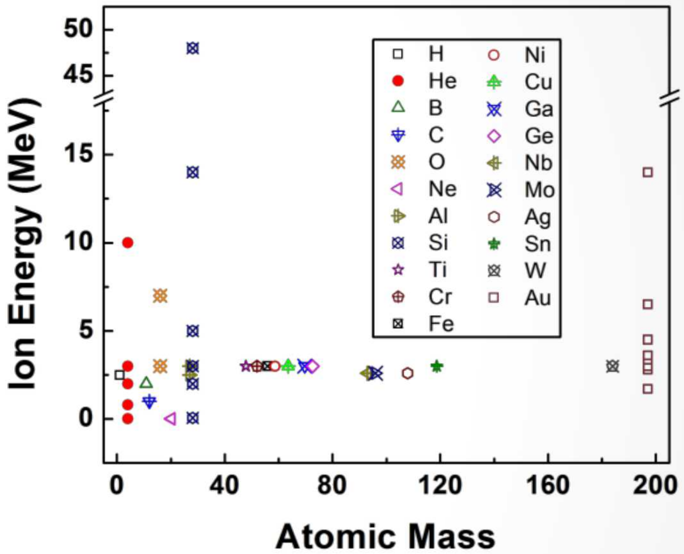
Sandia's *In situ* Ion Irradiation TEM (I^3TEM)

Collaborators: D. Buller, K. Hattar, J. Scott

10 kV Colutron - 200 kV TEM - 6 MV Tandem



Ion species & energy introduced into the TEM



Direct real time observation of ion irradiation, ion implantation, or both with nanometer resolution.

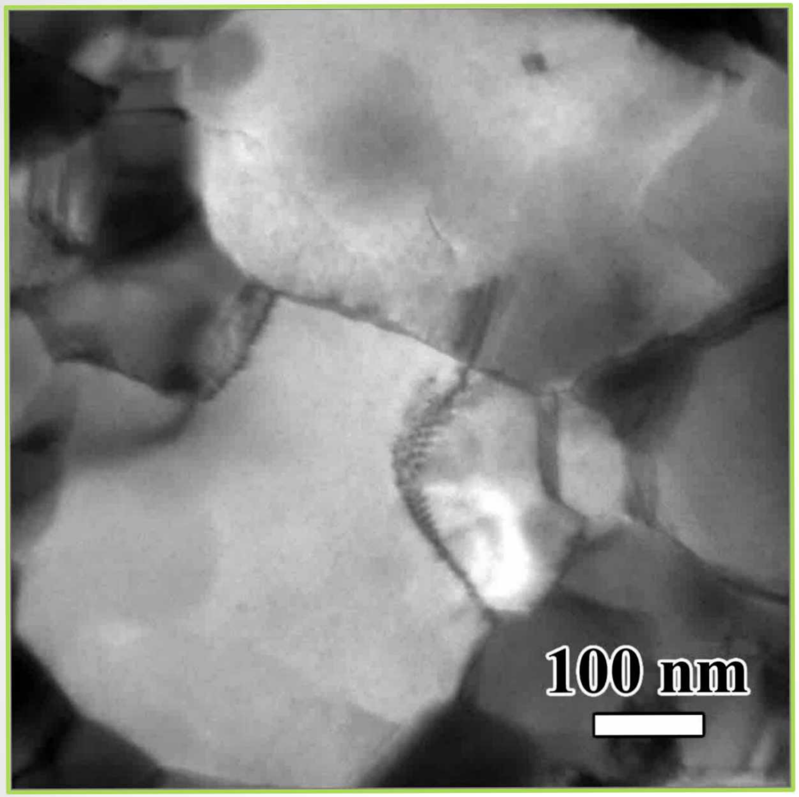
Similar beams can be directed to the TEM and end stations.

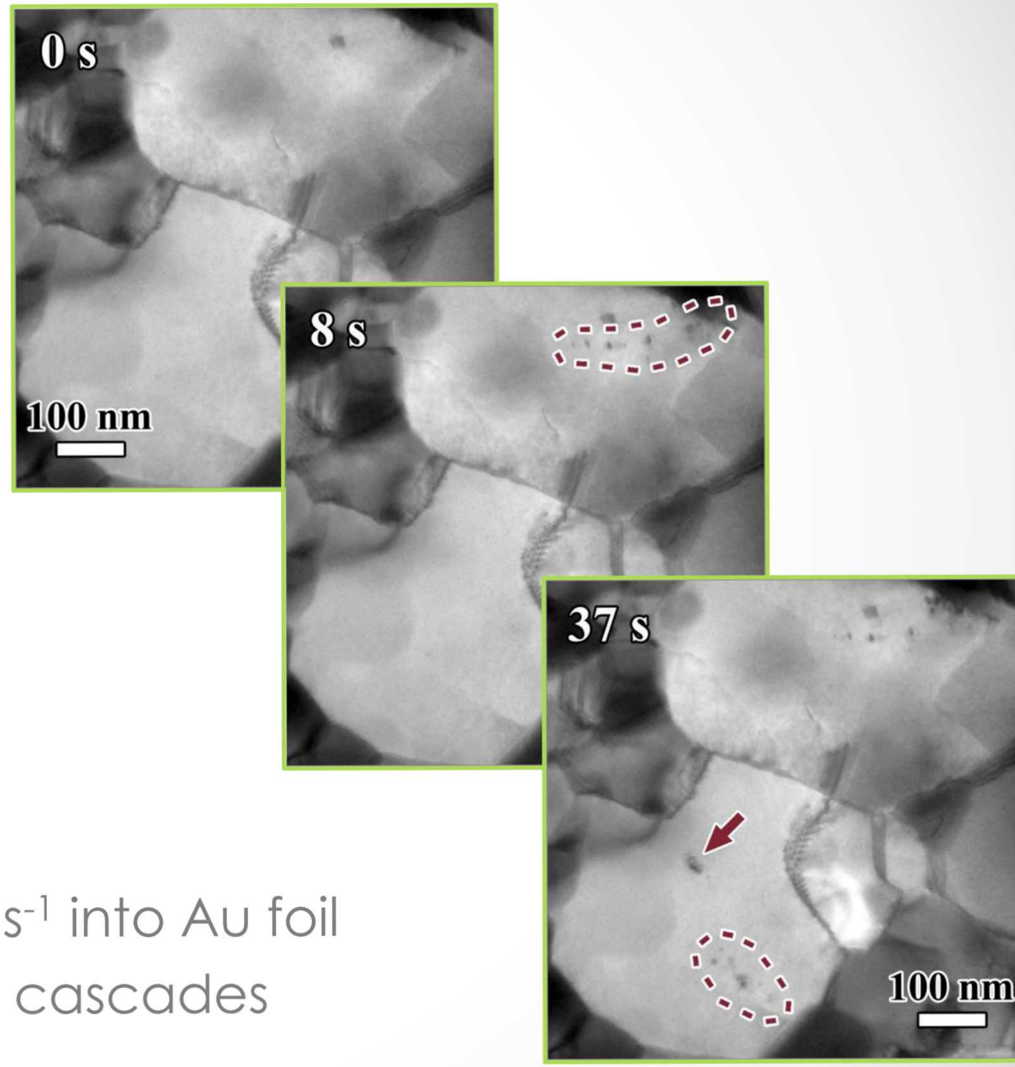
Hattar, *et al*, Nucl Instr Meth Phys Res B, 2014.

In Situ Irradiation: 3.6 MeV Au^{6+}

Sandia National Laboratories

Video speed ×5.





- Au^{6+} at 2.1×10^{8} ions cm^{-2} s^{-1} into Au foil
- Large defect clusters from cascades

What happens near grain boundaries?

Hattar, *et al*, Nucl Instr Meth Phys Res B, 2014.

Boundary Migration

Sandia National Laboratories

- Au foil during bombardment with 10 MeV Si^{3+}
- ~22 s of 4000s total experiment time

In situ ion irradiation TEM: 10 MeV Si into nanocrystalline Au.

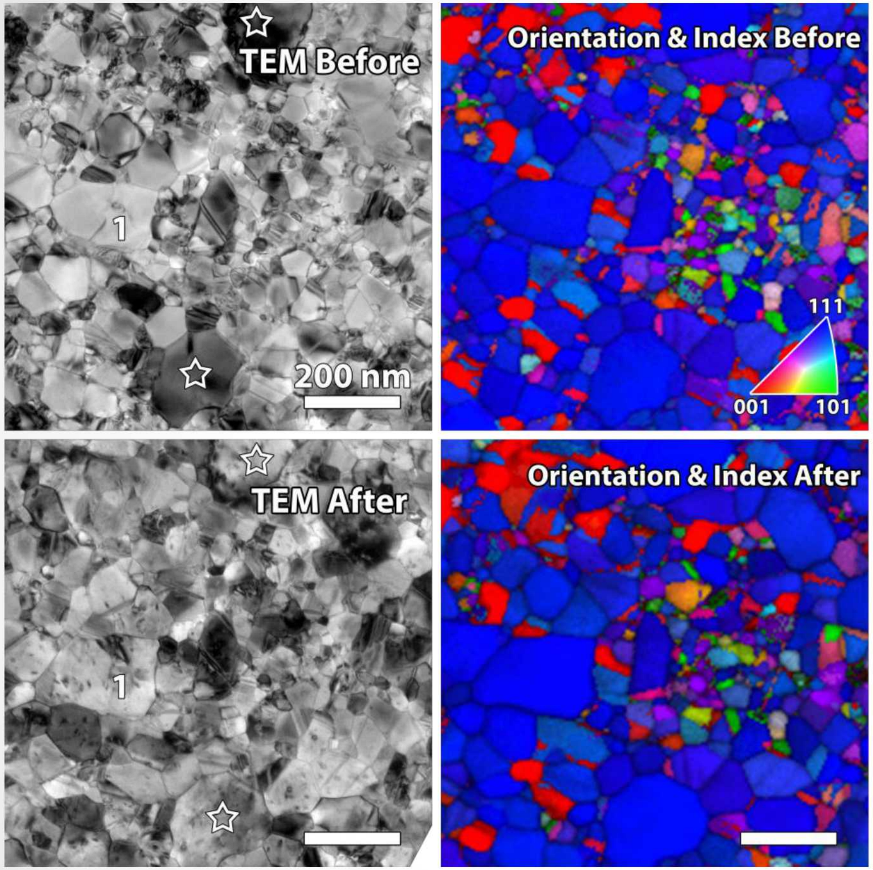
Playback at 2 × real time.

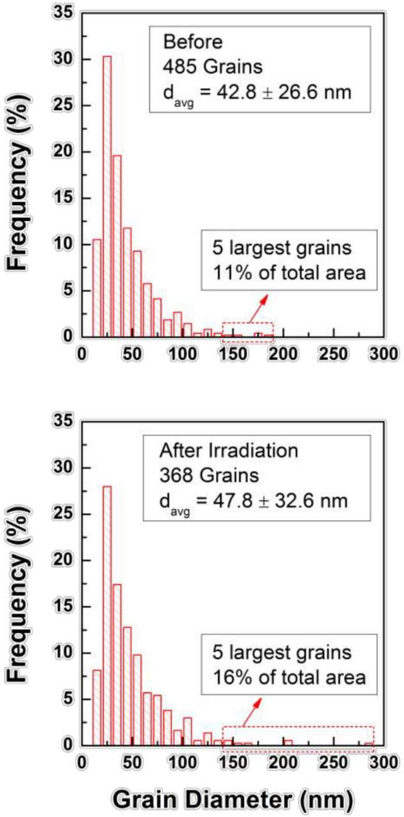
2× real time

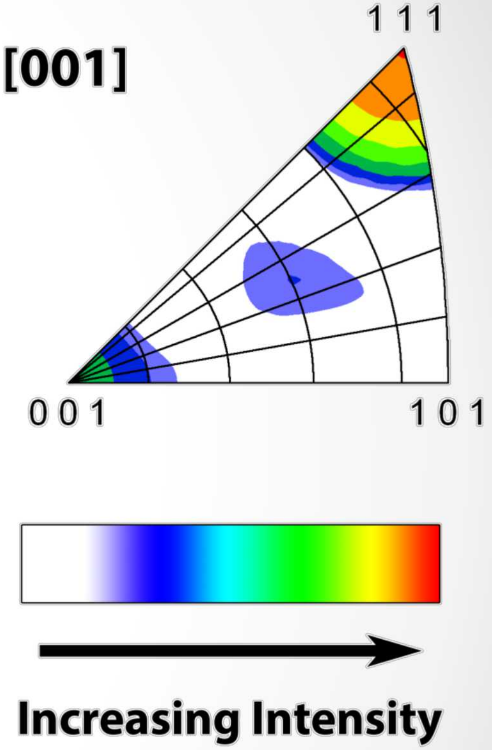
Structural evolution on the timescale of seconds after ion strikes.

Quantification: Overall

Bufford, *et al*, J Appl Phys, 2015.







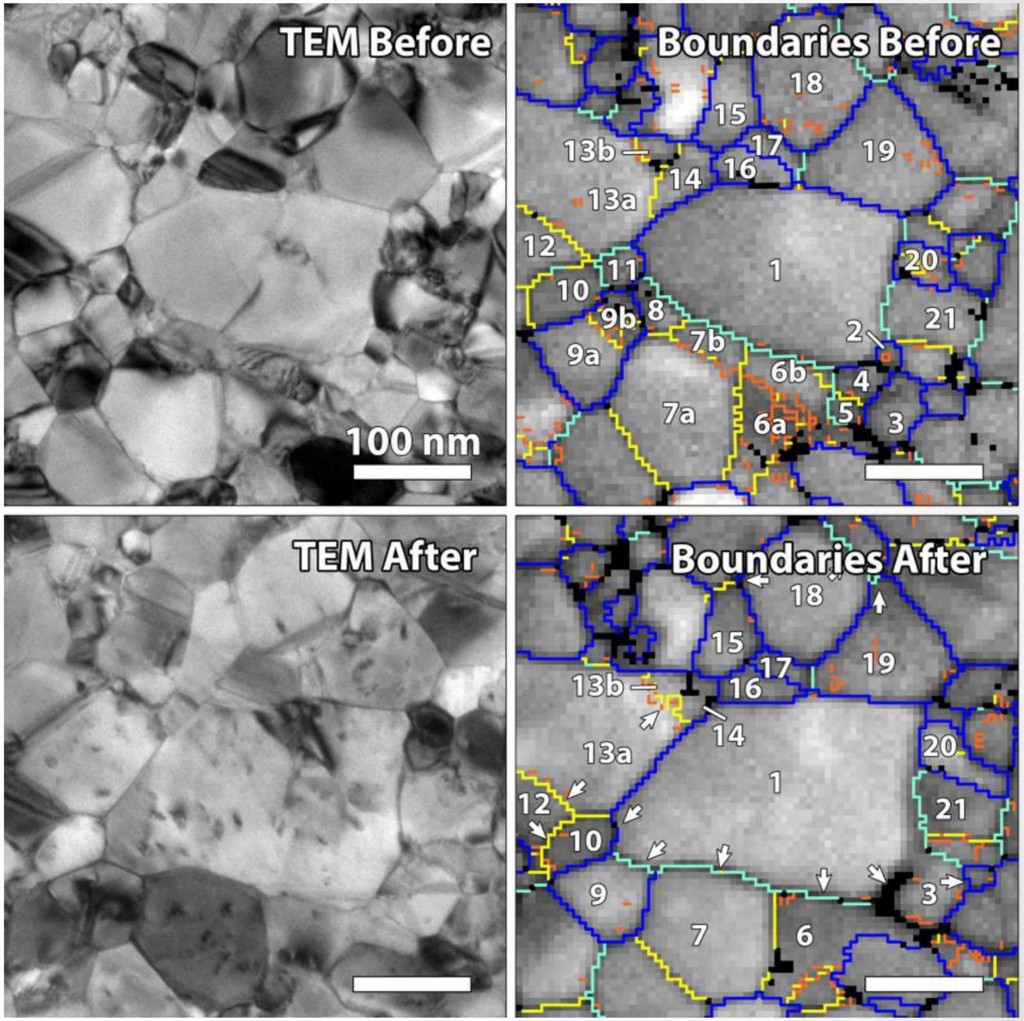
- Same area characterized before and after irradiation.
  - Hundreds of grains counted in minute
  - Grain size, orientation, boundary characters

Rapid quantification of statistically relevant numbers of grains and boundaries.

Individual Boundaries

- The same grains identified before and after irradiation
- Individual grain boundary misorientation angles and axes quantified
- Correlation of GB properties and radiation-induced changes

$\phi < 3°$
$3° \leq \phi < 15°$
$15° \leq \phi < 30°$
$30° \leq \phi$

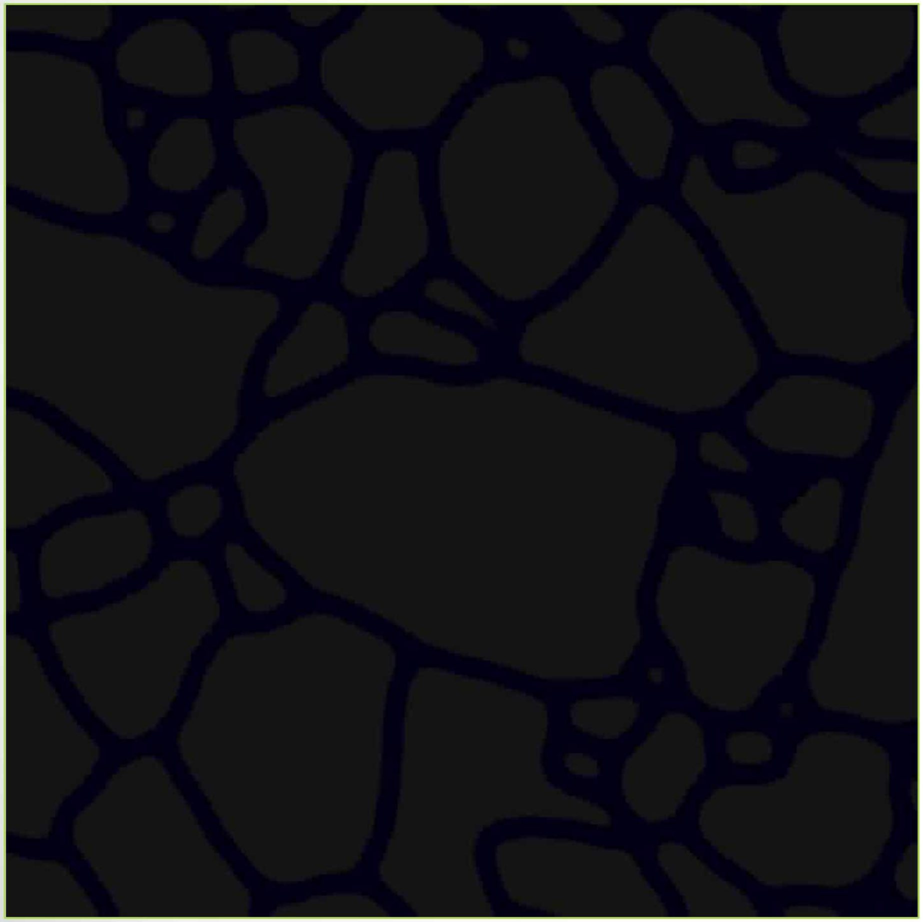


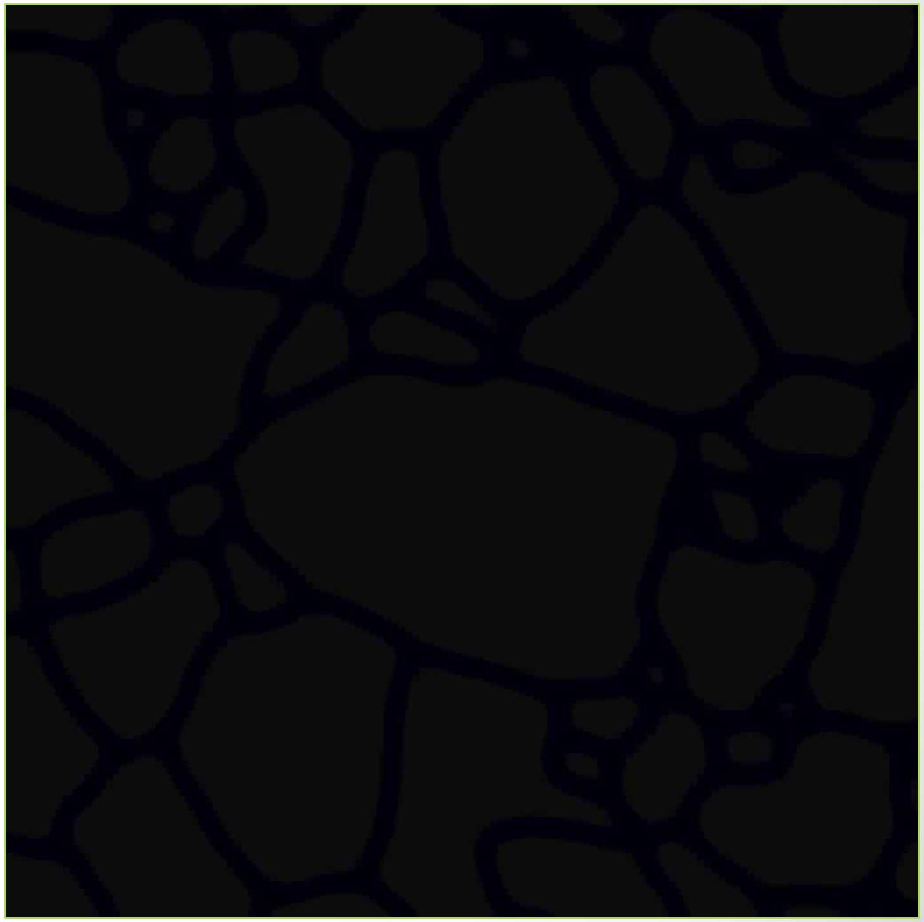
Bufford, *et al*, J Appl Phys, 2015.

Changes in individual boundaries quantified!

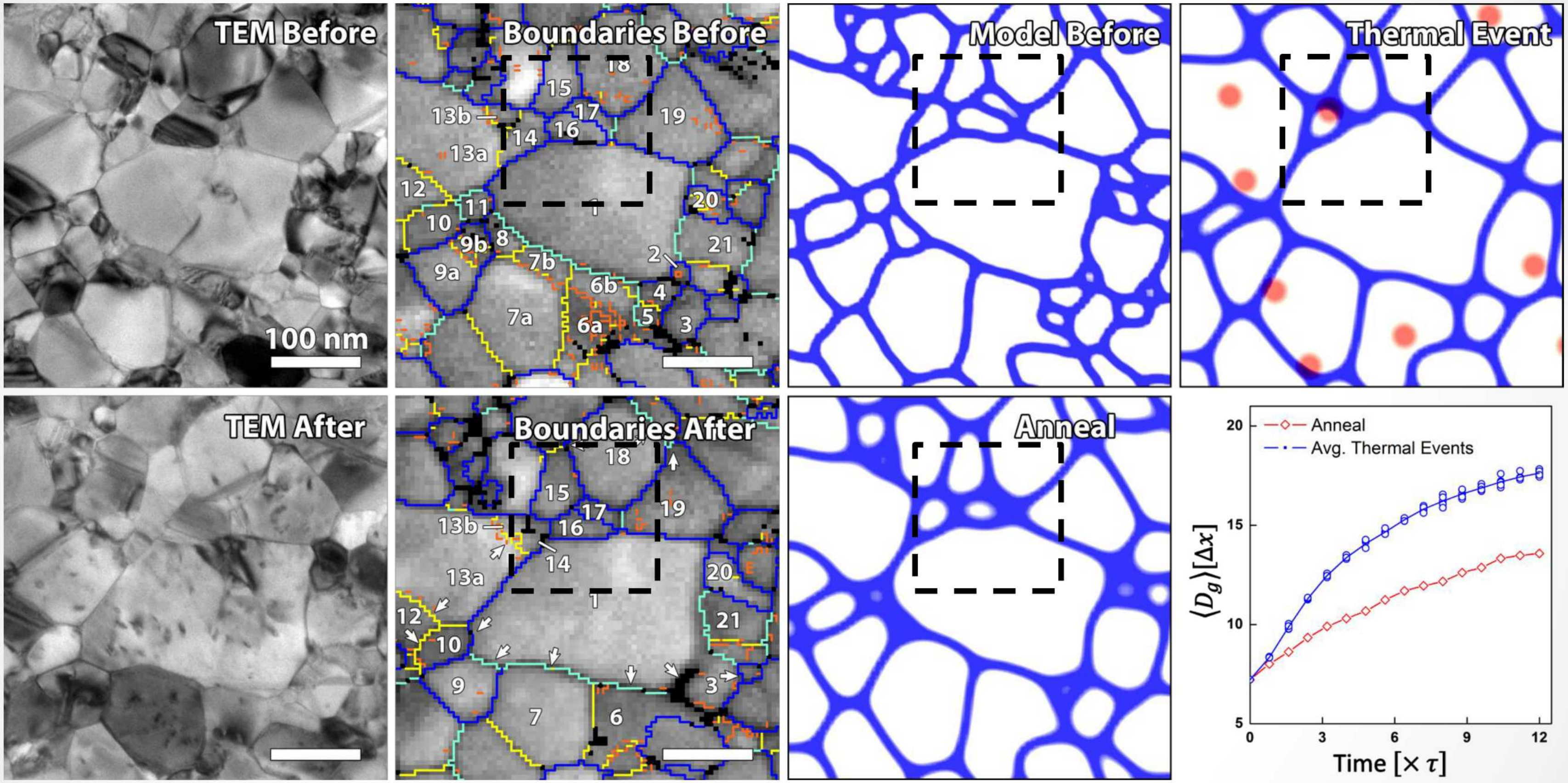
Simulated Irradiation and Annealing

Collaborators: F.F. Abdeljawad and S.M. Foiles





Exp. & Model Comparison



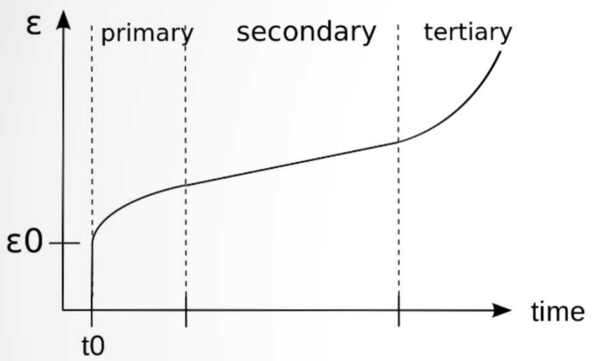
Bufford, *et al*, J Appl Phys, 2015.

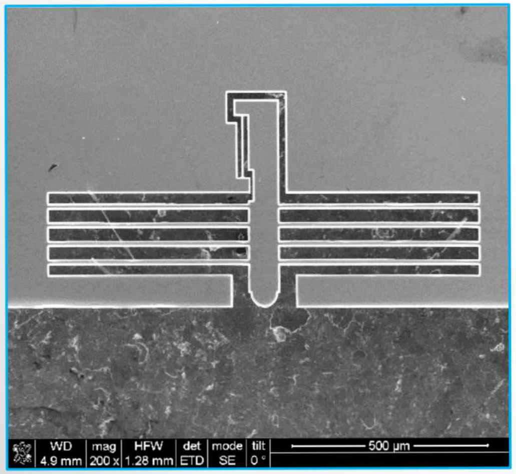
- Overall scaling laws appear consistent
- Subtle deviations from homogenous grain growth

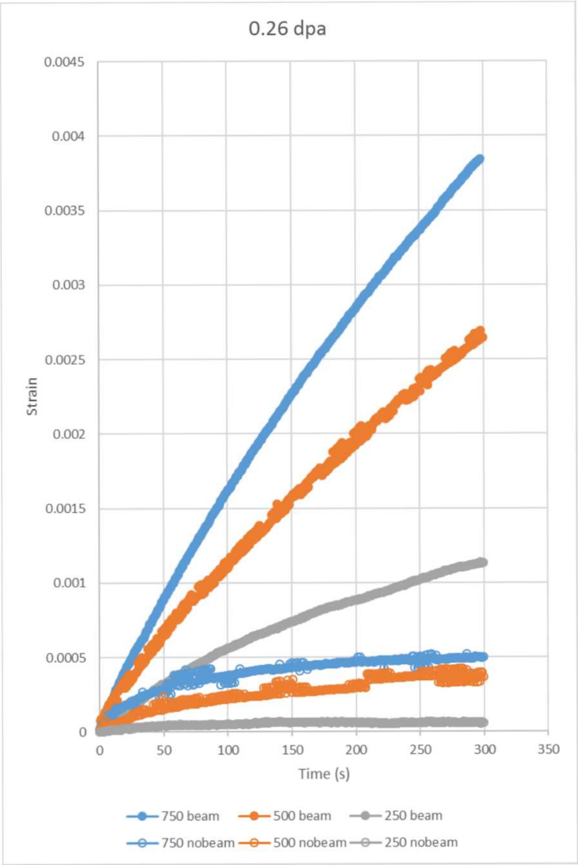
Stable boundaries suggest importance of non-thermally activated mobility.

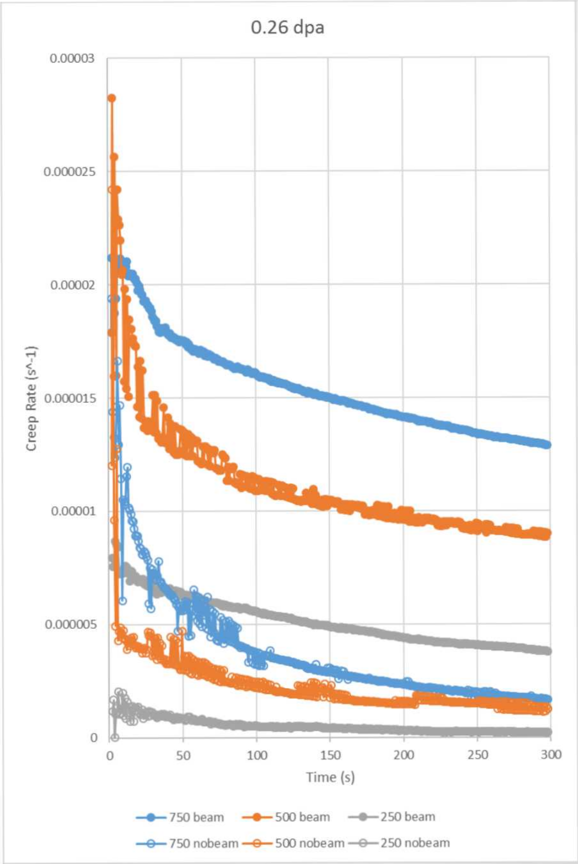
Irradiation Creep

Collaborators: B. Wang, A. Haque, K. Hattar

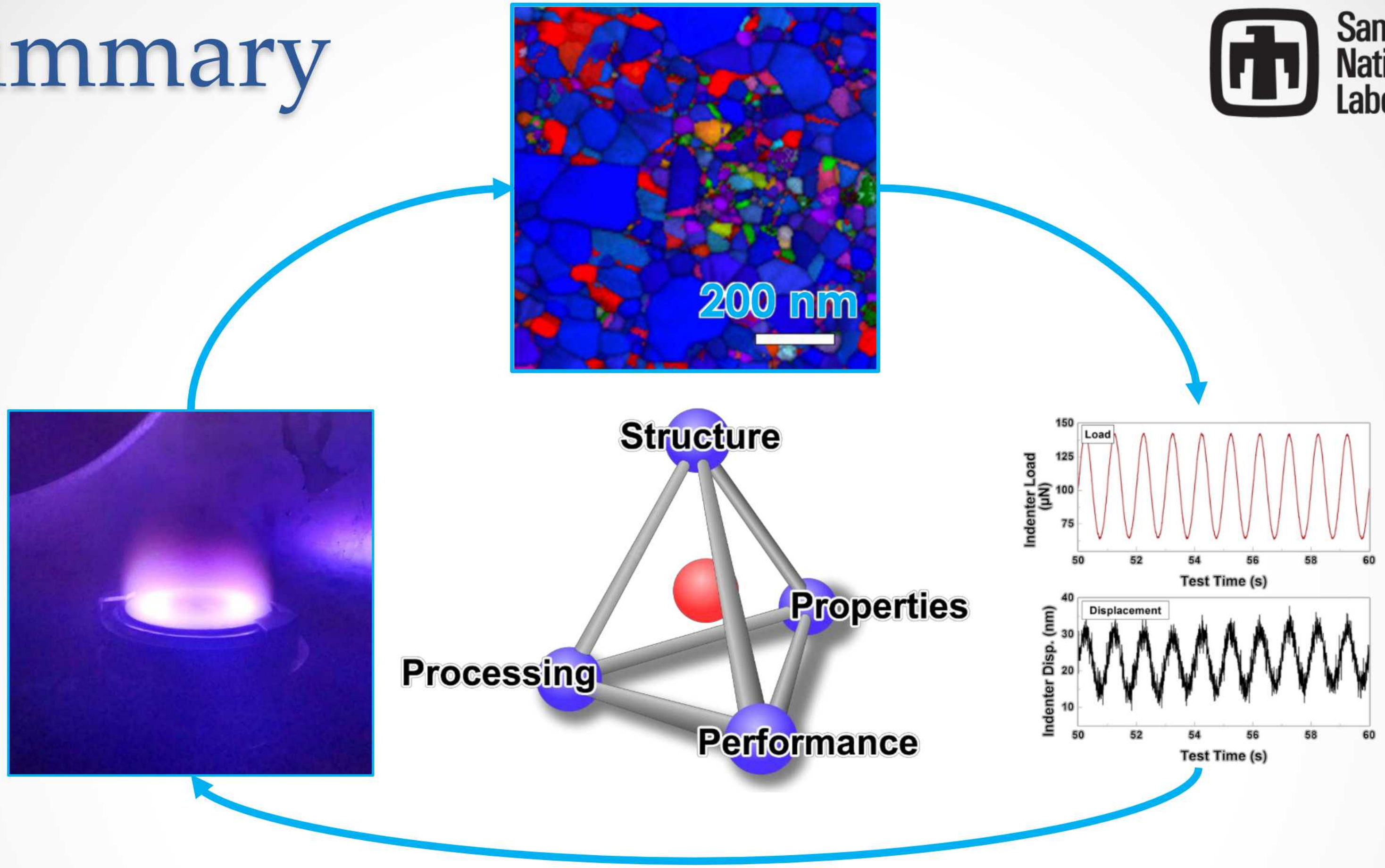








Summary

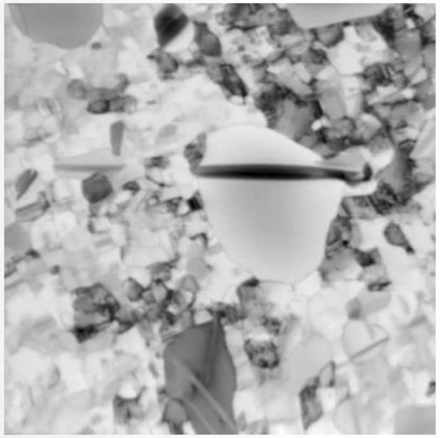


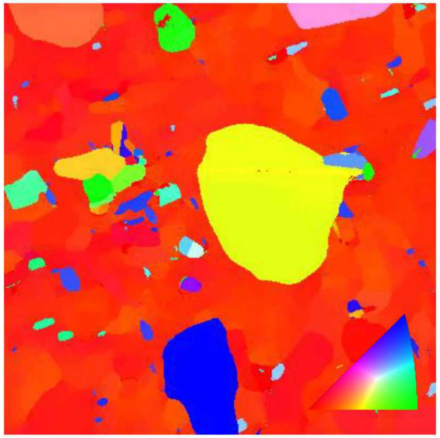
Revealed and quantified structural evolution at the grain boundary level under mechanical loading and irradiation.

Coming full-circle to produce better materials.

The Future

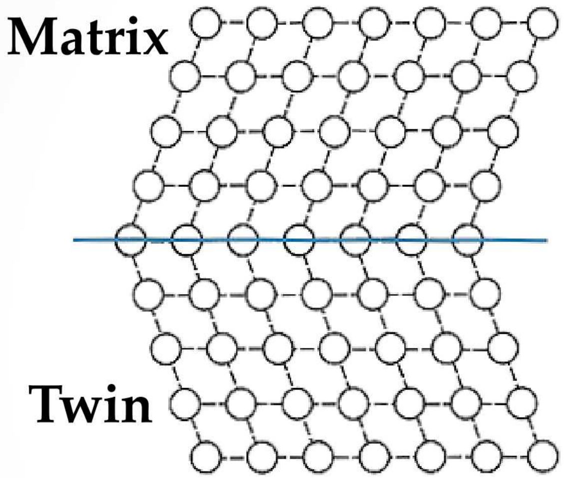
- Halting grain growth in nanocrystalline metals
- Connecting grain-level processes to bulk fracture
- Understanding and improving nanomaterials for energy
- 3D and multi-modal analysis



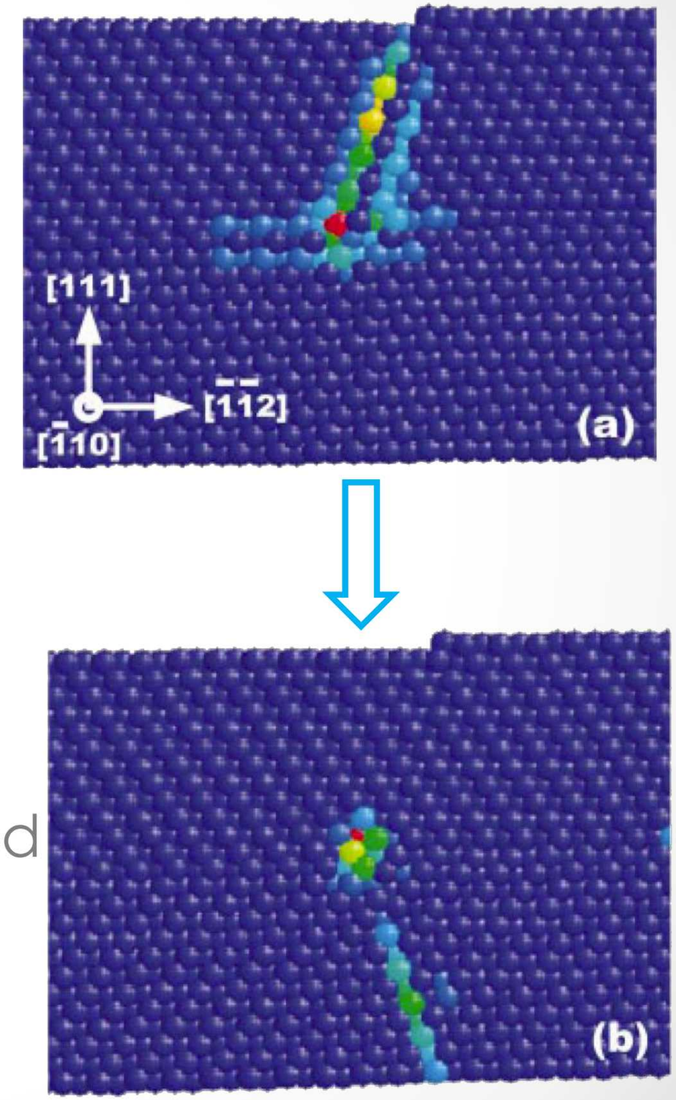


Acknowledgements: **IBL**: D.L. Buller, B.L. Doyle, C. Gong, K. Hattar, M.T. Marshall, B.R. Muntifering, M. Steckbeck. **Sandia**: F.F. Abdeljawad, D.P. Adams, B. Boyce, T.J. Boyle, S.M. Foiles, H. Lim, W.M. Mook, J.A. Scott, J.A. Sharon, S.H. Pratt, M. Rye, C. Sobczak. **External**: S. Bhowmick, L. Kuhn (Hysitron), A. Minor, C. Chisholm, P. Hosemann (UC Berkley), Z. Bi, Q.X. Jia, Y. Liu, (Los Alamos National Lab), A. Darbal (AppFive), D. Kaoumi (University of South Carolina), A. Leff (Drexel University), Y. Zhu, H. Wang, X. Zhang (Texas A&M University). Work performed by DCB at Sandia was fully supported by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science under proposal #U2014A0026.

Twins and other grain boundaries



Imperial College Rock Library.

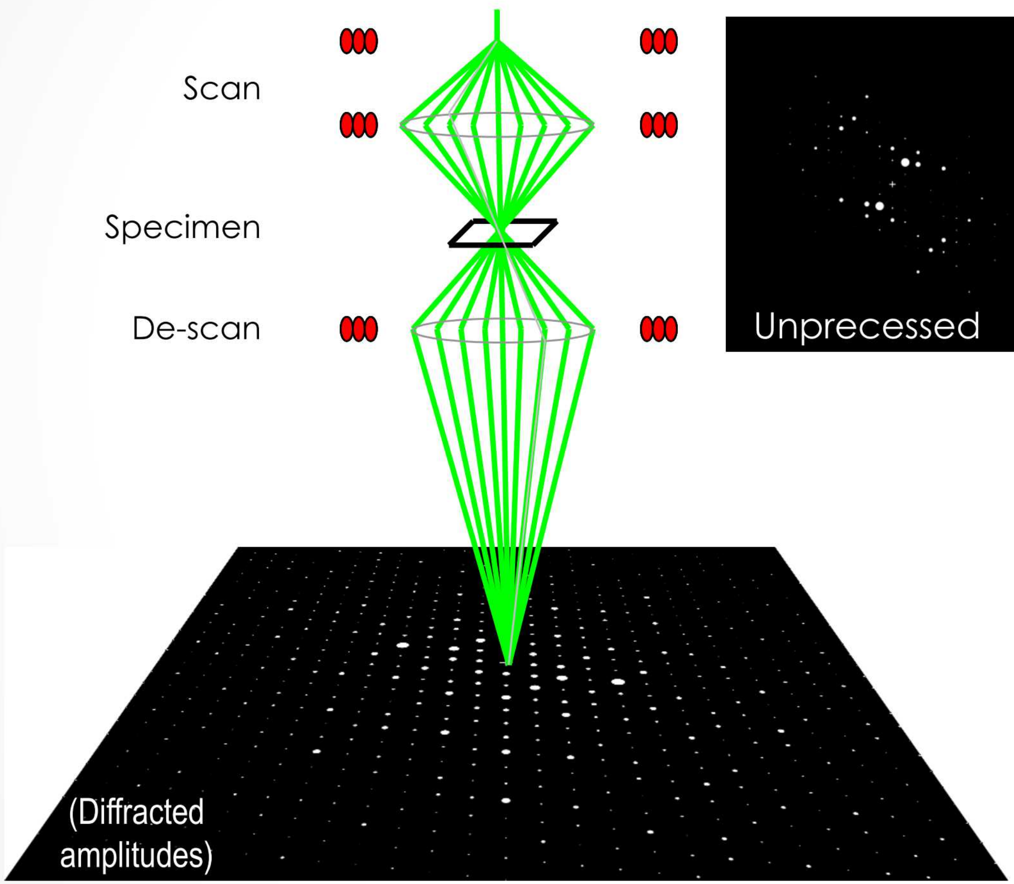


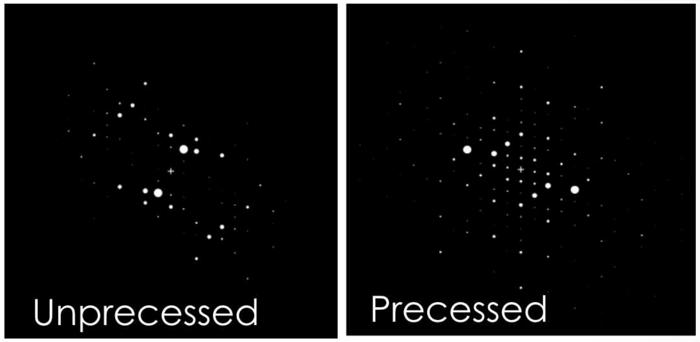
Zhang, et al, *Appl Phys Lett* 2004.

- Twin boundaries are less disordered than most other boundaries
- During simulated deformation:
  - Existing dislocation cross slips at high stress
  - Sessile dislocation left at the boundary

Precession Electron Diffraction Microscopy

Collaborators: K.J. Ganesh, S. Rajasekhara, P.J. Ferreira



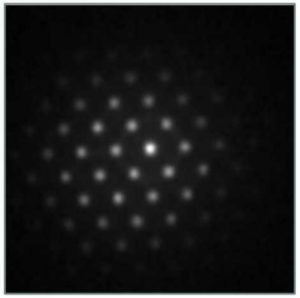


Advantages:

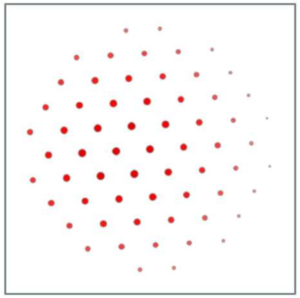
- < 10 nm spatial resolution
- Near kinematical electron diffraction
- Symmetry ambiguities are resolved
- Fast and automated acquisition
  - ~200 grains in 15 min.

Slide courtesy of K.J. Ganesh

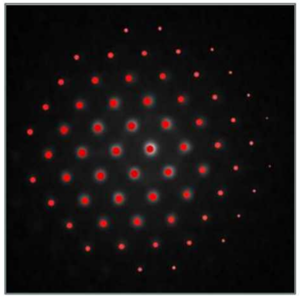
Approach: Experimental



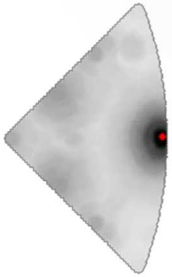
Experimental Pattern



Theoretical Template

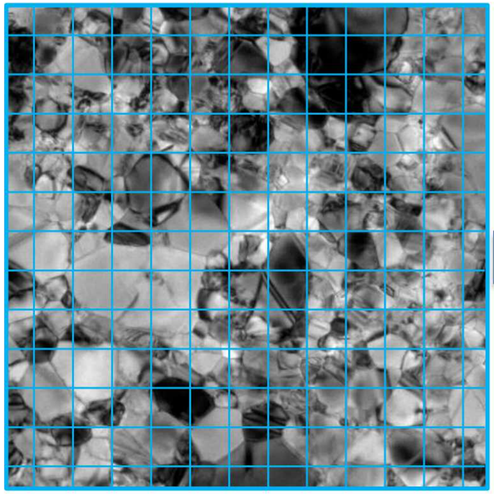


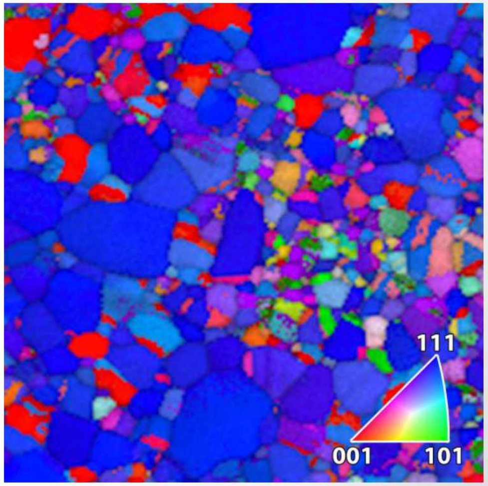
Template Matched



Point Mapped To IPF

- Automated diffraction orientation mapping
  - Point by point grid of orientations mapped
  - 5 nm resolution
- Analogous to EBSD



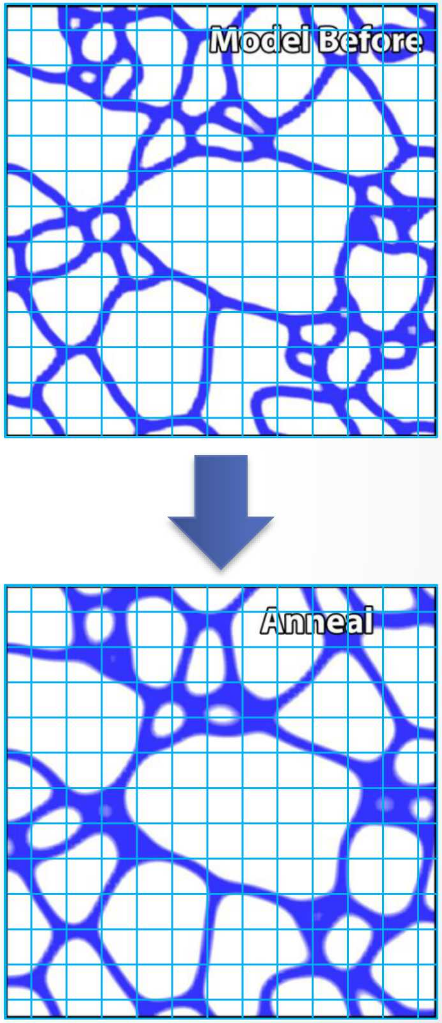


Point diffraction data

Approach: Modeling

Sandia National Laboratories

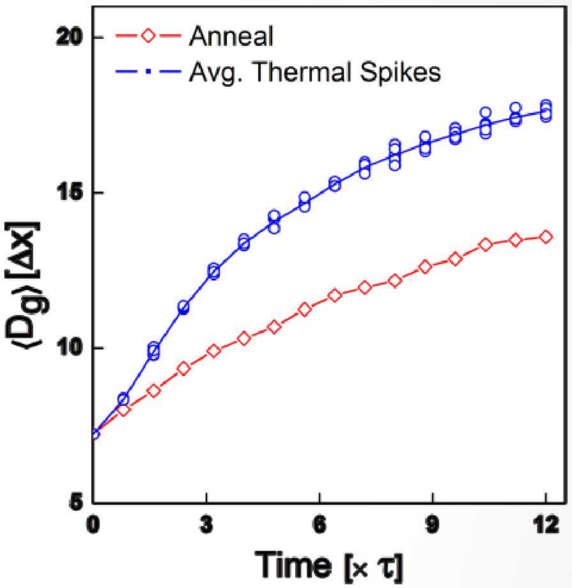
- What is phase field modeling?
  - Mathematical model for solving interfacial problems, like solidification, growth, etc.
- Example grain growth model
  - Thermodynamic free energy function
    - $dF = d(\gamma A) = \gamma dA$ (γ: GB energy, A: GB area)
  - Model for kinetics
    - $V = M\gamma h$ (M: GB mobility, h: GB curvature)
  - Solve at each pixel for a predetermined timestep
- See Abdeljawad and Foiles, Acta Mater, 2015 for more information

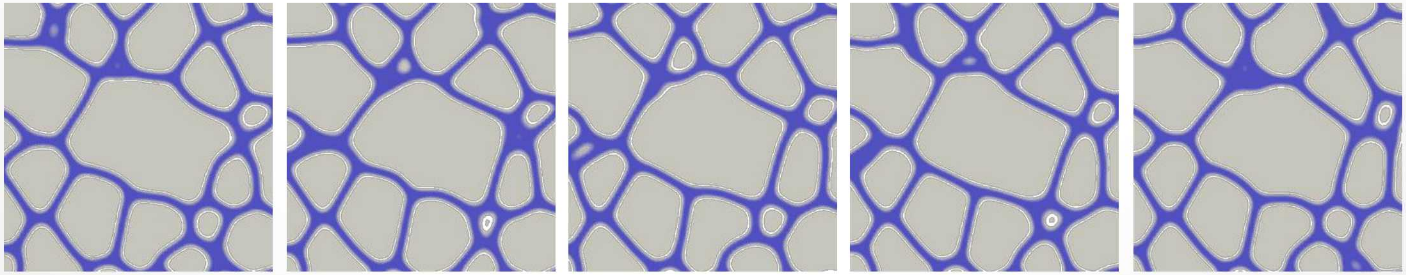


Can directly use experimental maps as input structures, and then compare evolutions!

Model Data Analysis

- During simulated annealing grain growth scales approximately with $T^{1/2}$
  - Expected for homogenous grain growth
- During simulated irradiation, grain growth scales with $T^{1/n}$, where $n \approx 3$
  - Initially faster, but stagnates sooner





Growth time scaling

Fraction of ion strikes that intersect grain boundaries (thus contributing to grain growth):

$$f_{\mathrm{GB}} = \frac{\pi D^2 \frac{d_{\mathrm{spike}}}{2}}{\frac{\pi D^3}{6}} = \frac{3 d_{\mathrm{spike}}}{\boxed{D}},$$

- Incorporation of this *D* term leads to scaling proportional to $t^{(1/3)}$.
- Consistent with experimental observations.

$$\frac{dD}{dt} = \frac{\Phi \chi \delta}{N_{\mathrm{at}}} \frac{3 d_{\mathrm{cas}}}{\boxed{D}} \left[\frac{4 \gamma V_{\mathrm{at}} N_{\mathrm{at}} \nu}{D k_B} \frac{\sqrt{\frac{3}{5}} \Gamma\left(\frac{8}{3}\right) k_B^{8/3}}{10 \pi C_0^{2/3} \kappa_0} \frac{Q^{5/3}}{E_a^{8/3}} \right].$$

$$D^2 dD = \left[12 \gamma d_{\mathrm{spike}} \Phi \chi \delta \frac{V_{\mathrm{at}} \nu \sqrt{\frac{3}{5}} \Gamma\left(\frac{8}{3}\right) k_B^{5/3}}{10 \pi C_0^{2/3} \kappa_0} \frac{Q^{5/3}}{E_a^{8/3}} \right] dt.$$

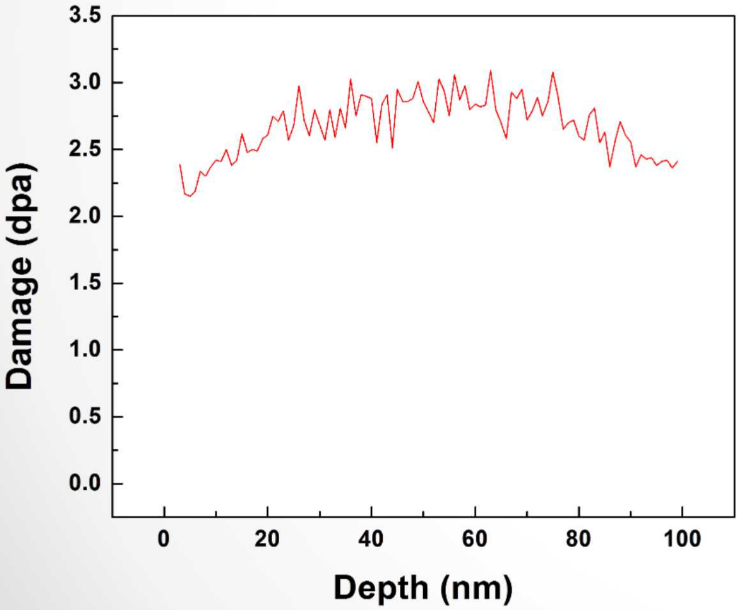
$$\boxed{D^3 - D_0^3} = \left[36 \gamma d_{\mathrm{spikes}} \chi \delta \frac{V_{\mathrm{at}} \nu \sqrt{\frac{3}{5}} \Gamma\left(\frac{8}{3}\right) k_B^{5/3}}{10 \pi C_0^{2/3} \kappa_0} \frac{Q^{5/3}}{E_a^{8/3}} \right] \Phi t$$

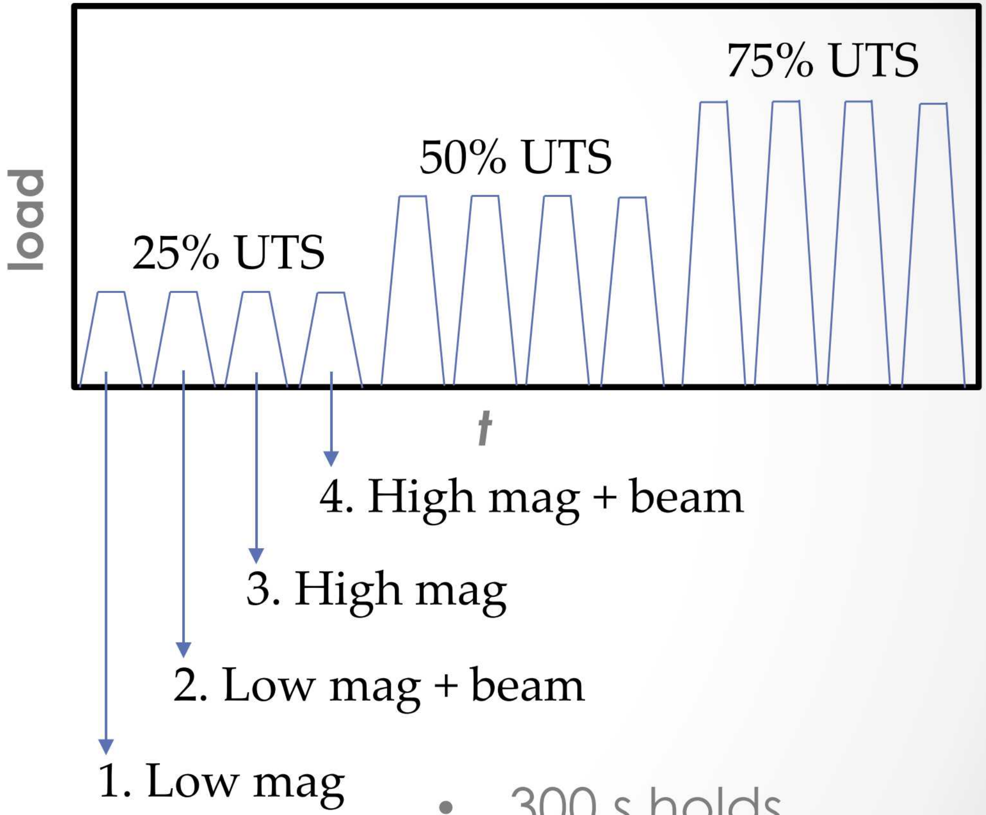
$$= K \Phi t.$$

Kaoumi, *et al*, J Appl Phys, 2008.

Irradiation Creep

- 1.4 MeV Zr
- 4.6 × 10^{13} and 10^{14} ions cm^{-2}
- Nominally 0.26 and 2.6 dpa
- 4.2 × 10^{10} ions cm^{-2} s^{-1}
- 2.5 × 10^{-4} dpa s^{-1}

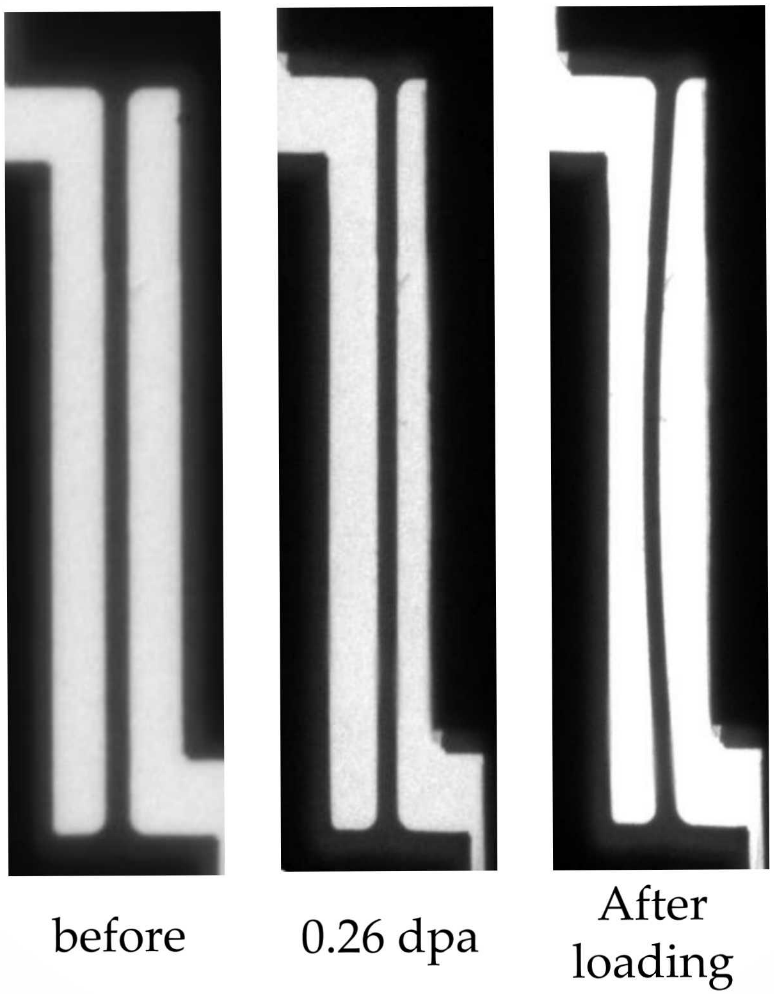




- 300 s holds
- 7.5 × 10^{-2} dpa $cycle^{-1}$

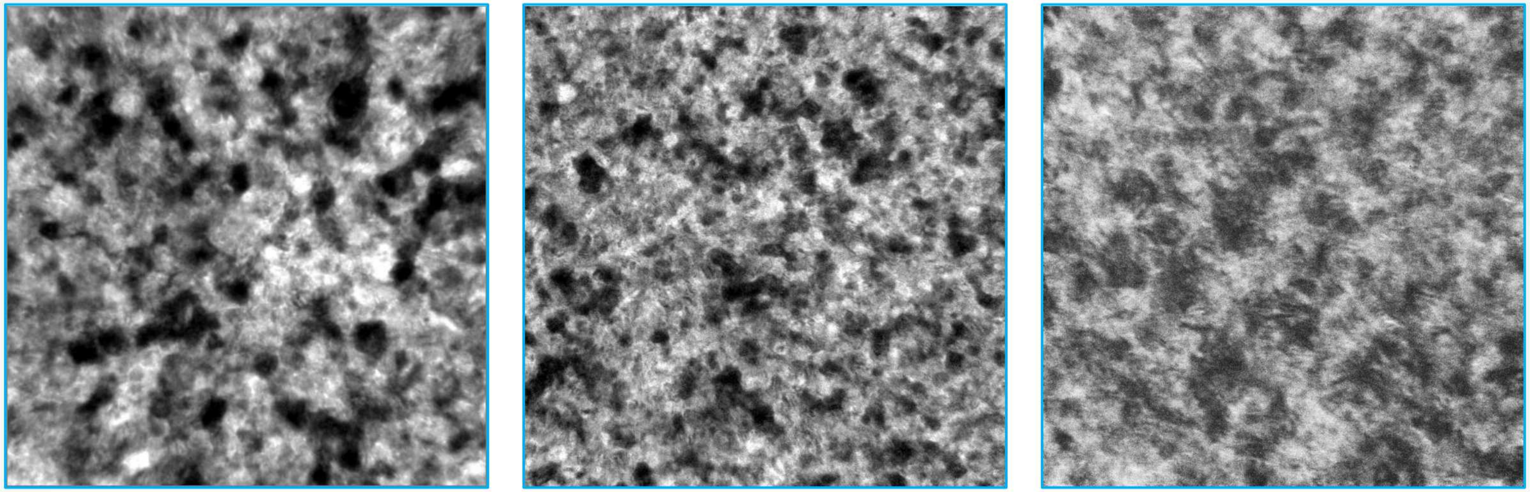
Measuring overall changes and microstructural changes with and without irradiation.

Elongation: 0.26 dpa

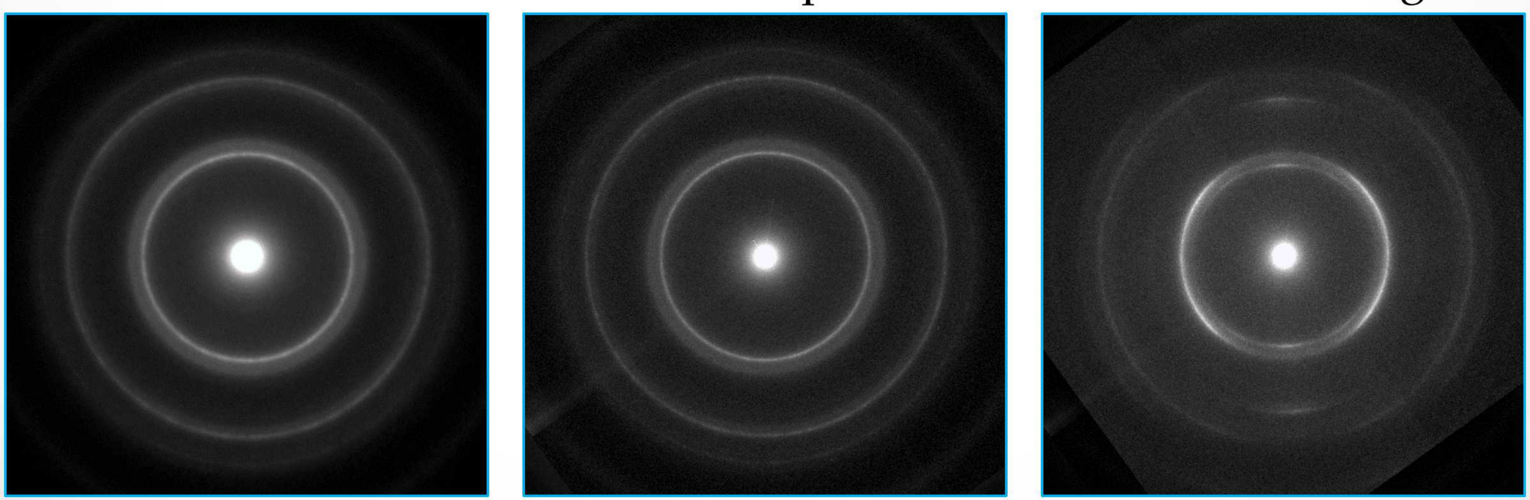


Minimal change with irradiation alone, but clear extension during loading.

Structure: 0.26 dpa



before | 0.26 dpa | After loading



Little change with irradiation, but deformation texture developed during loading.