This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

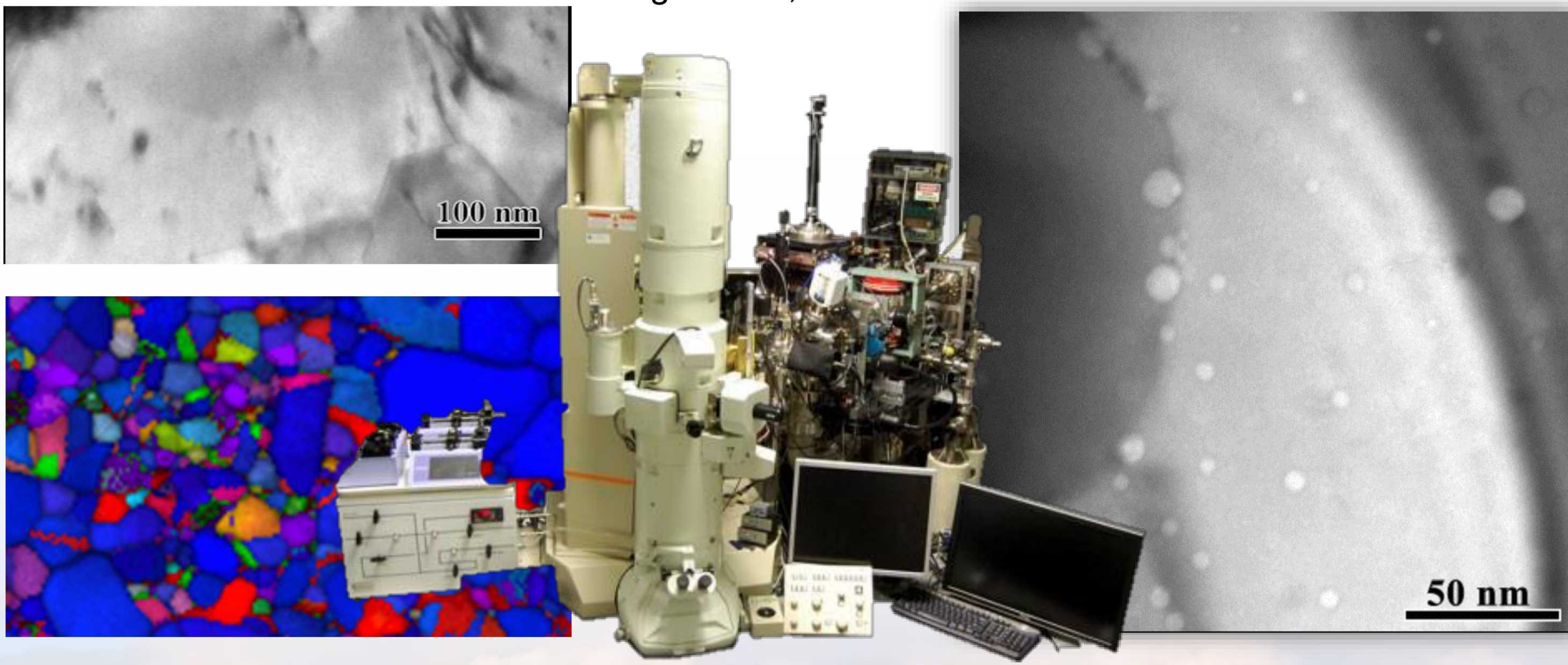
SAND2018-9393C

Exploring the Interplay Between Grain Boundaries and Radiation Damage

C. Barr, B. Muntifering, C. Taylor, D.C. Bufford, D. Adams, and K. Hattar

Sandia National Laboratories

August 13th, 2018



Collaborators:

- D.L. Buller, C. Chisholm , P. Hosemann, A. Minor, F. Abdeljawad, S.M. Foiles, J. Qu, Sugar, P. Price, M. Abere, S. Briggs, D.B. Robinson, N. Li, A. Misra, Y. Chen, N. Li, D. Bufford, X. Zhang

This work was partially funded by the Division of Mate… …eering, Office of Basic Energy Sciences, U.S. Department of Energy. This work was performed, in part, … …t of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE's National Nuclear Security Administration under contract DE-NA-0003525. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government.

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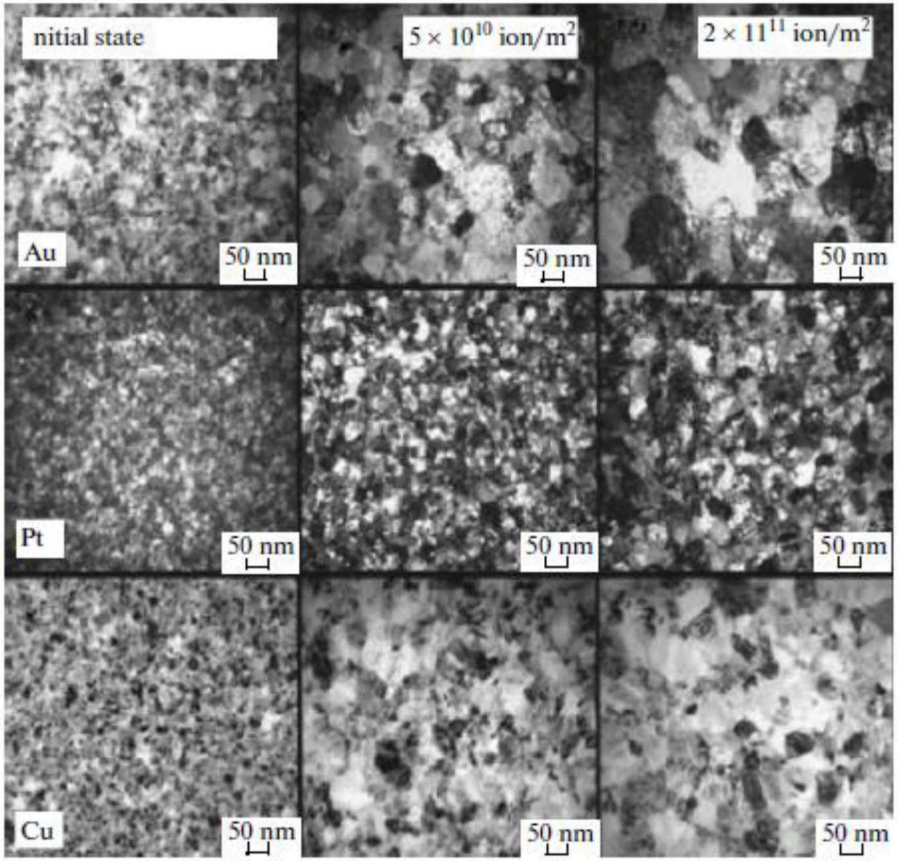
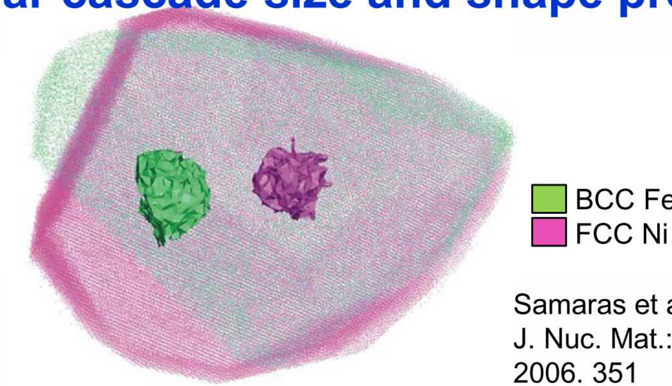
Section Summary: Radiation Tolerance from Nanostructured Metals

Variation in radiation tolerances



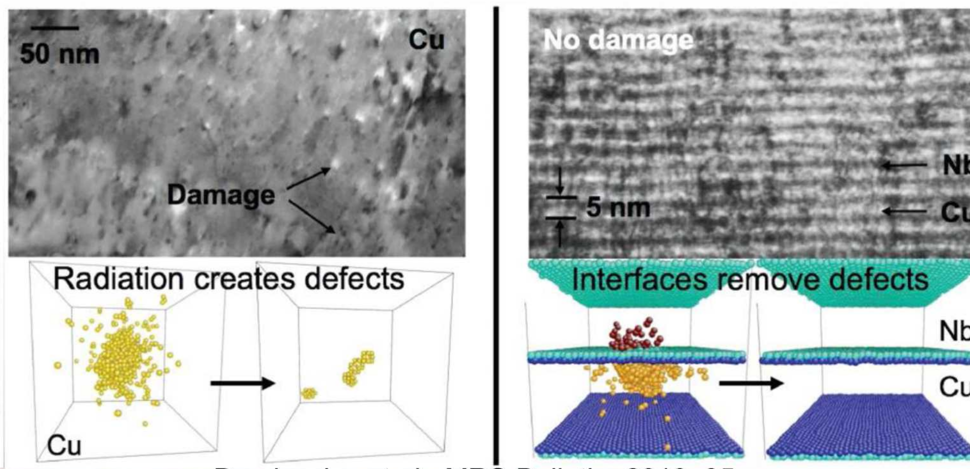
Kaomi et al., JAP: 2008. 104 073525

Similar cascade size and shape predicted



Samaras et al., J. Nuc. Mat.: 2006. 351

Nanolamellars are radiation tolerant



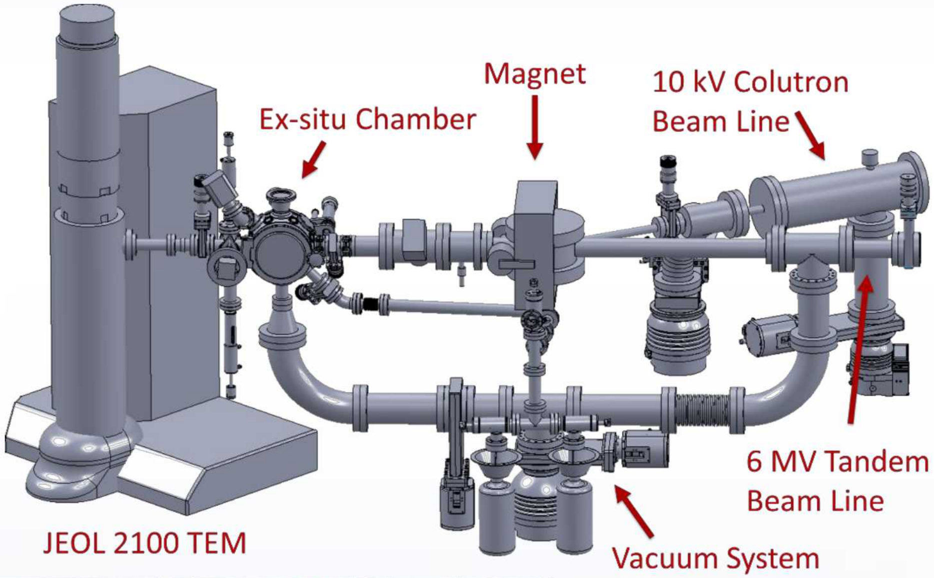
Demkowicz et al., MRS Bulletin: 2010. 35

To a first order mean grain size comparison, these reports appear conflicting.
Initial microstructural details and associated properties need to be considered!

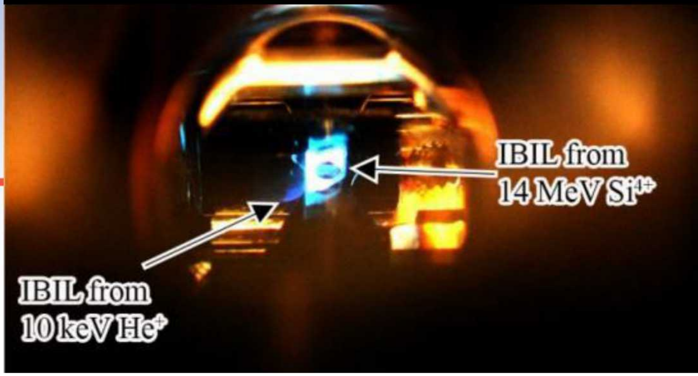
Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

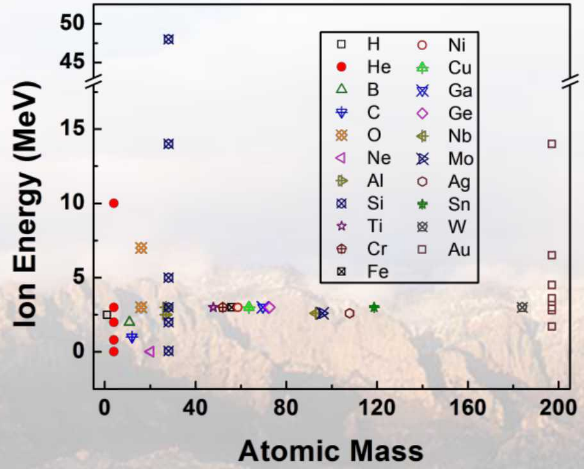
Collaborator: D.L. Buller

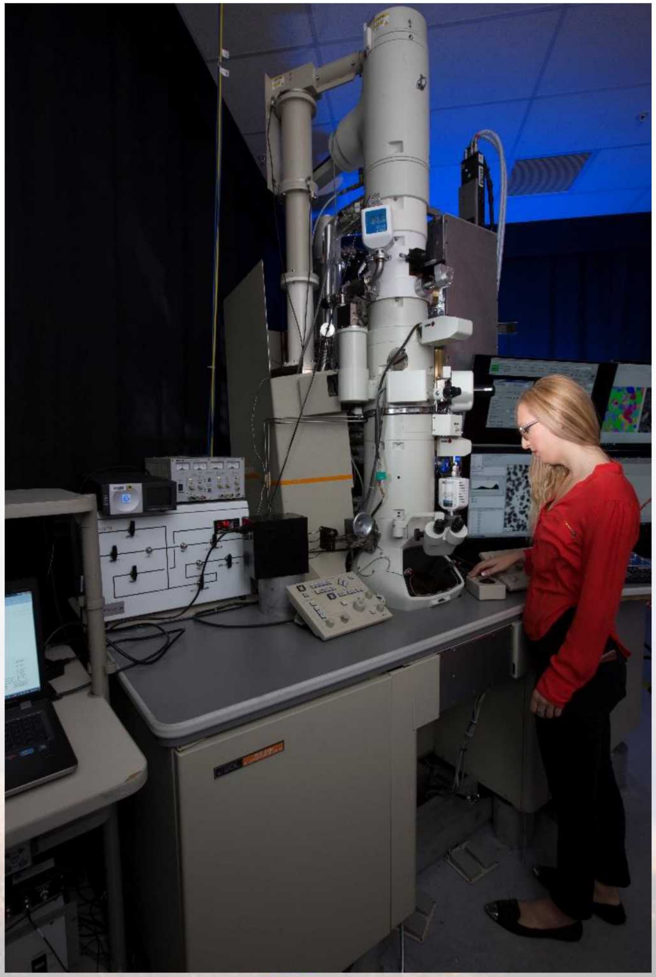
10 kV Colutron - 200 kV TEM - 6 MV Tandem



IBIL from a quartz stage inside the TEM



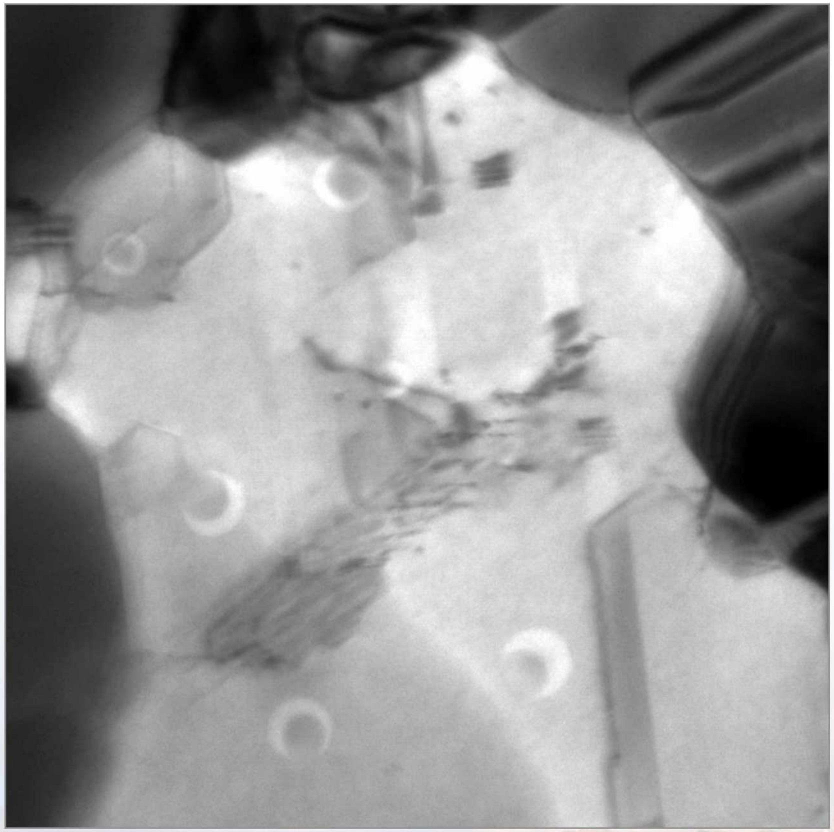




Dose Rate Effects in Nanocrystalline Metals

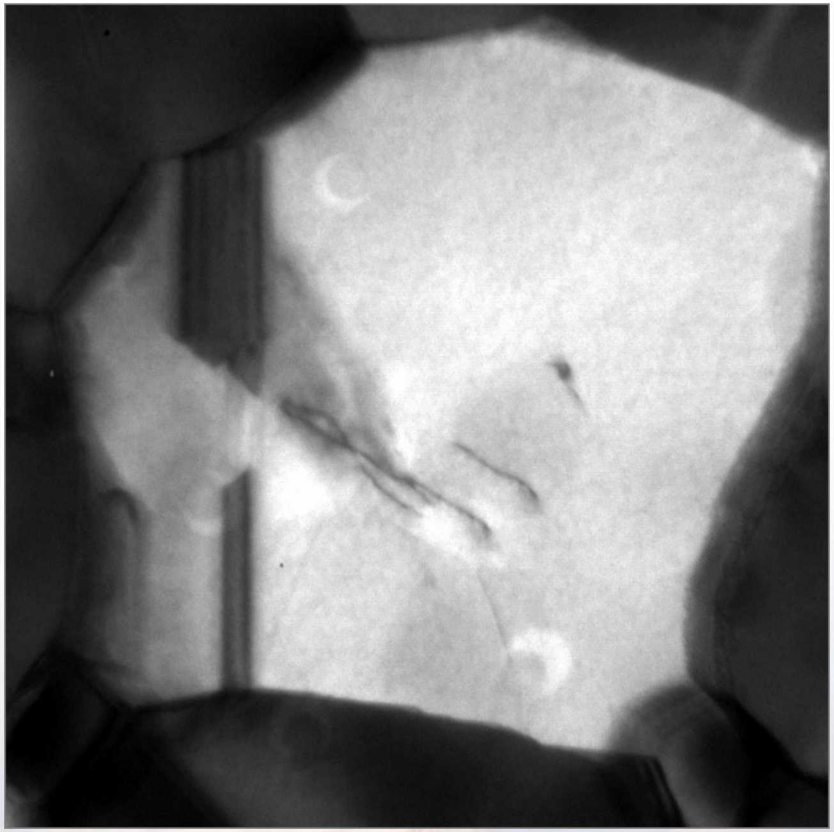
Collaborators: C. Chisholm , P. Hosemann, & A. Minor

7.9×10^9 ions/cm^2/s



VS

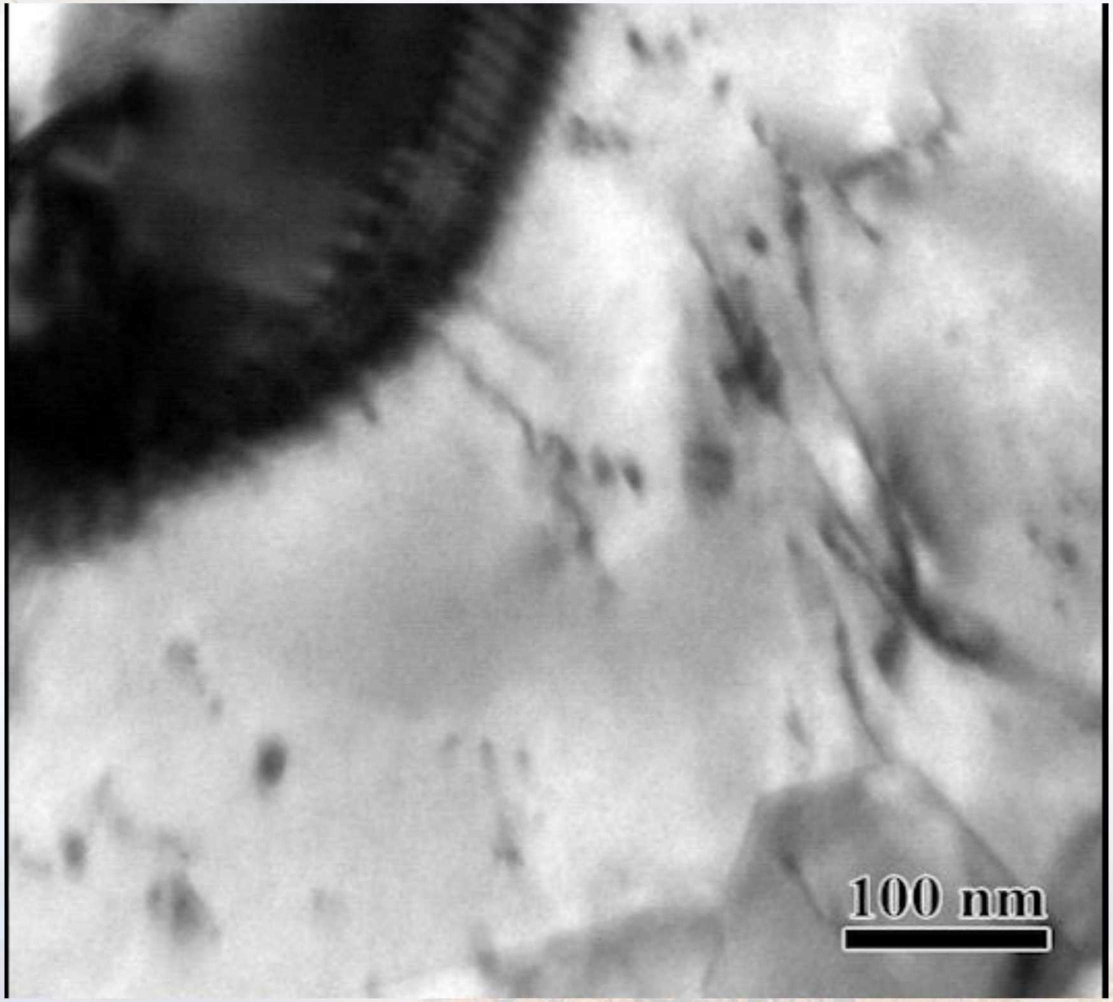
6.7×10^7 ions/cm^2/s



Improved vibrational and ion beam stability permits us to work at 120kx or higher permitting imaging of single cascade events

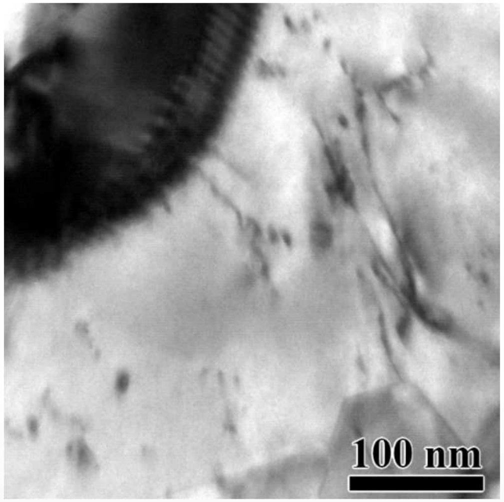
1D Brownian Motion in Real Time

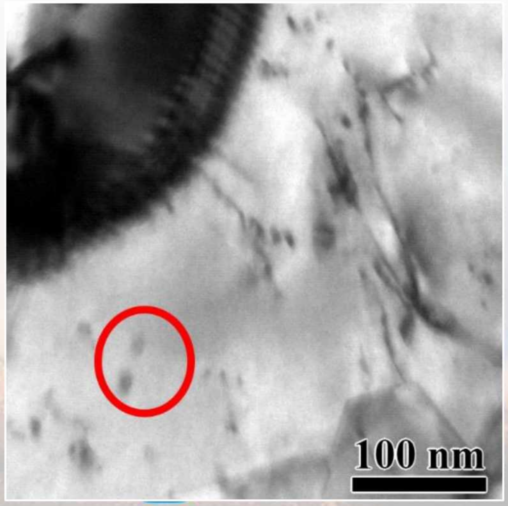
Collaborator: D.C. Bufford



Triple beam condition:

2.8 MeV Au^{4+} + 10 keV $\mathbf{He}^{+}/\mathbf{D}_2^{+}$

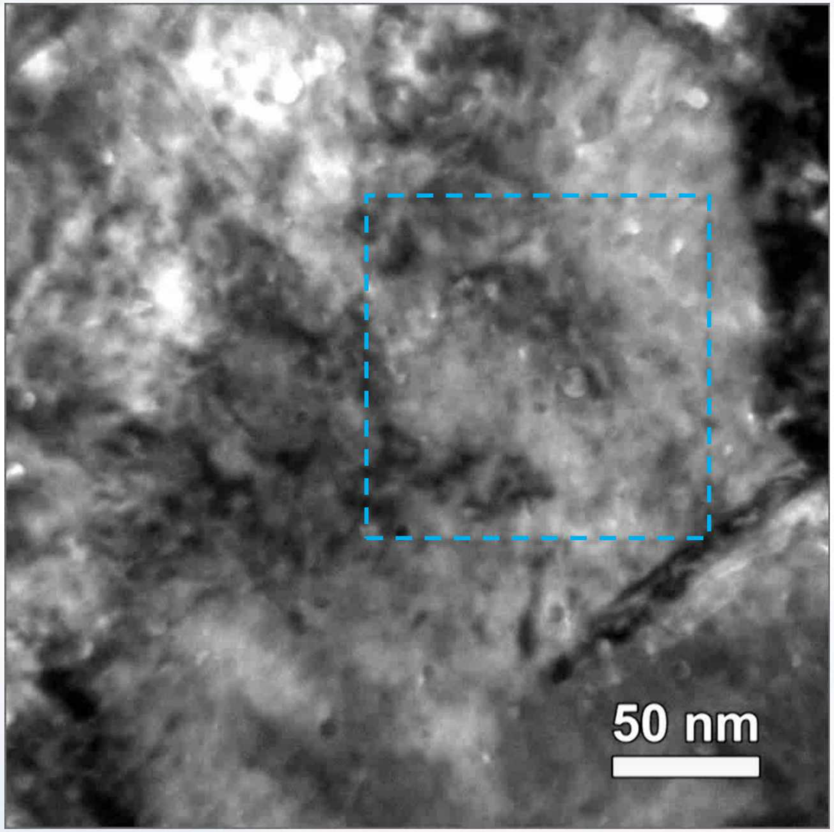


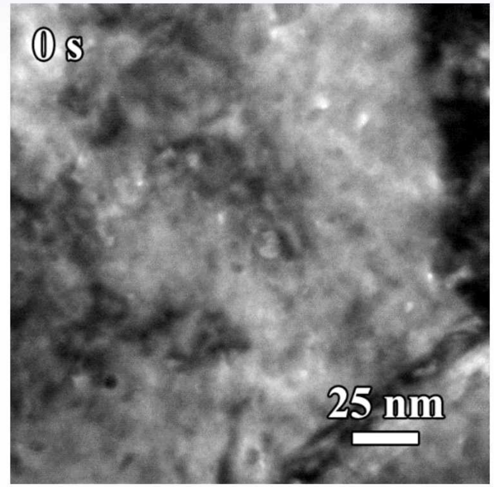


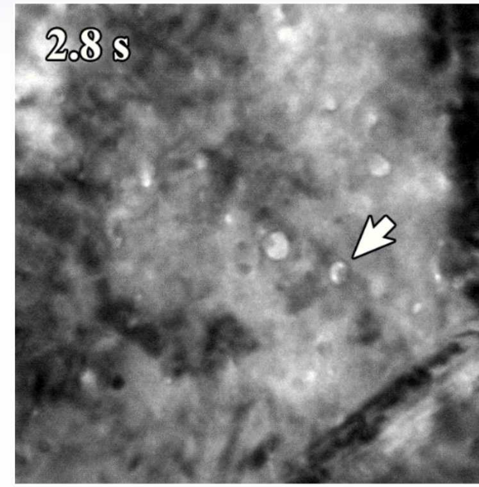
- **Dislocation loop moves between two pinning sites**
  - ~30 nm apart

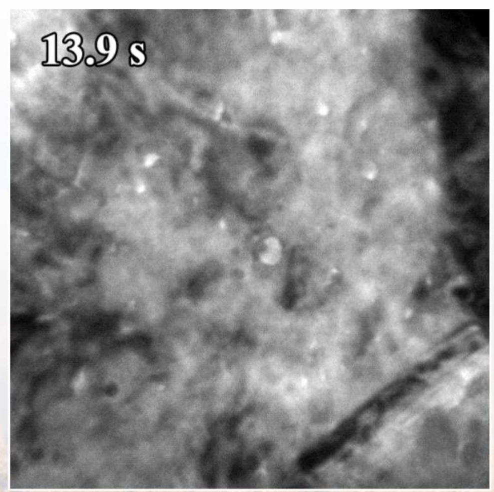
Simultaneous *In situ* TEM Triple Beam: 2.8 MeV Au^{4+} + 10 keV He^{+}/D_2^{+}

Collaborator: D.C. Bufford









In situ triple beam He, D_2, and Au beam irradiation is possible in the Sandia's I^3TEM!

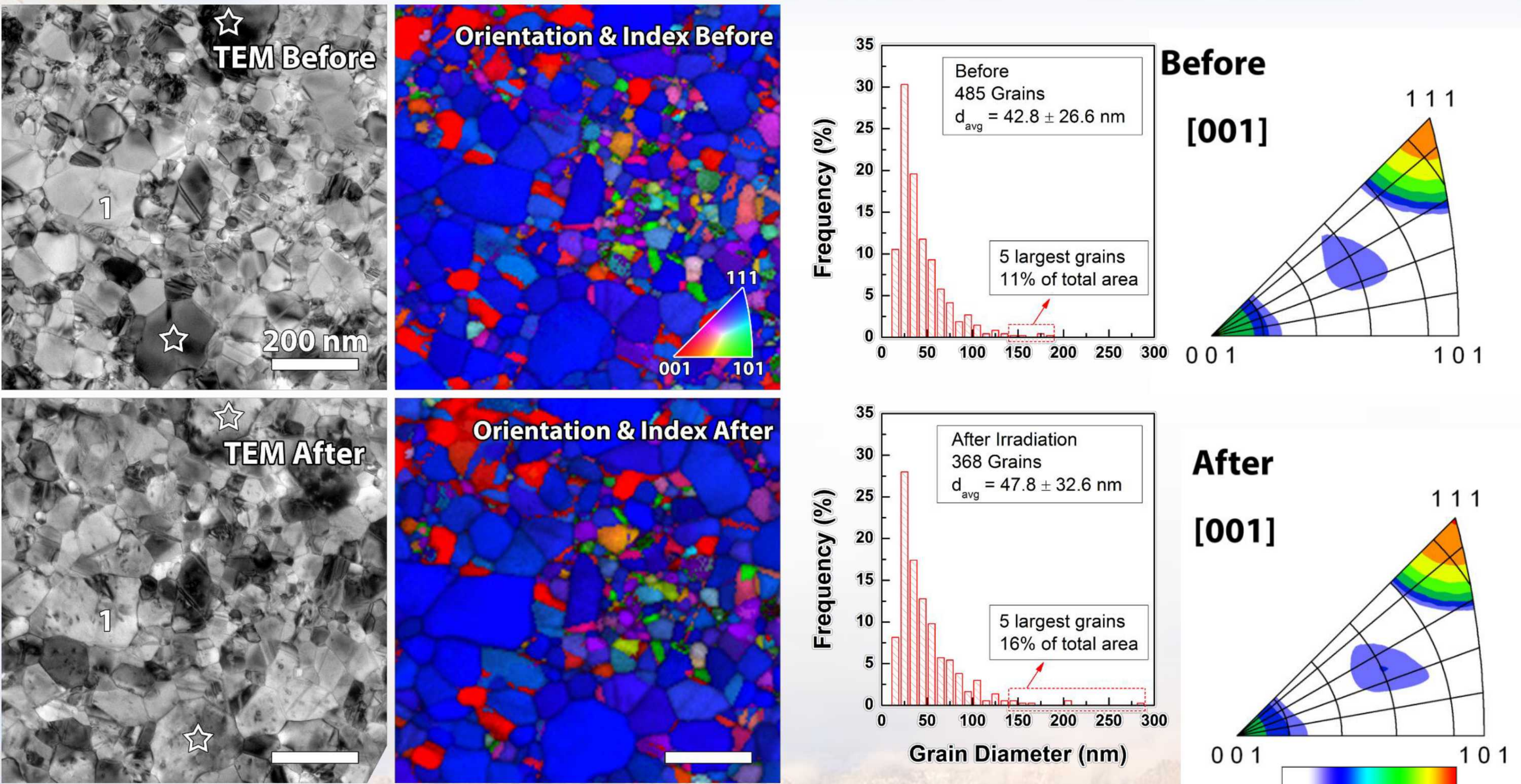
Intensive work is still needed to understand the defect structure evolution that has been observed.

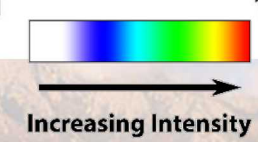
Speed x1.5

- **Approximate fluence:**
  - Au 1.2 $\times 10^{13}$ ions/cm^2
  - He 1.3 $\times 10^{15}$ ions/cm^2
  - D 2.2 $\times 10^{15}$ ions/cm^2
- **Cavity nucleation and disappearance**

Quantifying Grain Boundary Radiation Stability of Nanocrystalline Au

Collaborators: D.C. Bufford, F. Abdeljawad, & S.M. Foiles





10 MeV Si

Any texture or grain boundary evolution can be directly observed and quantified

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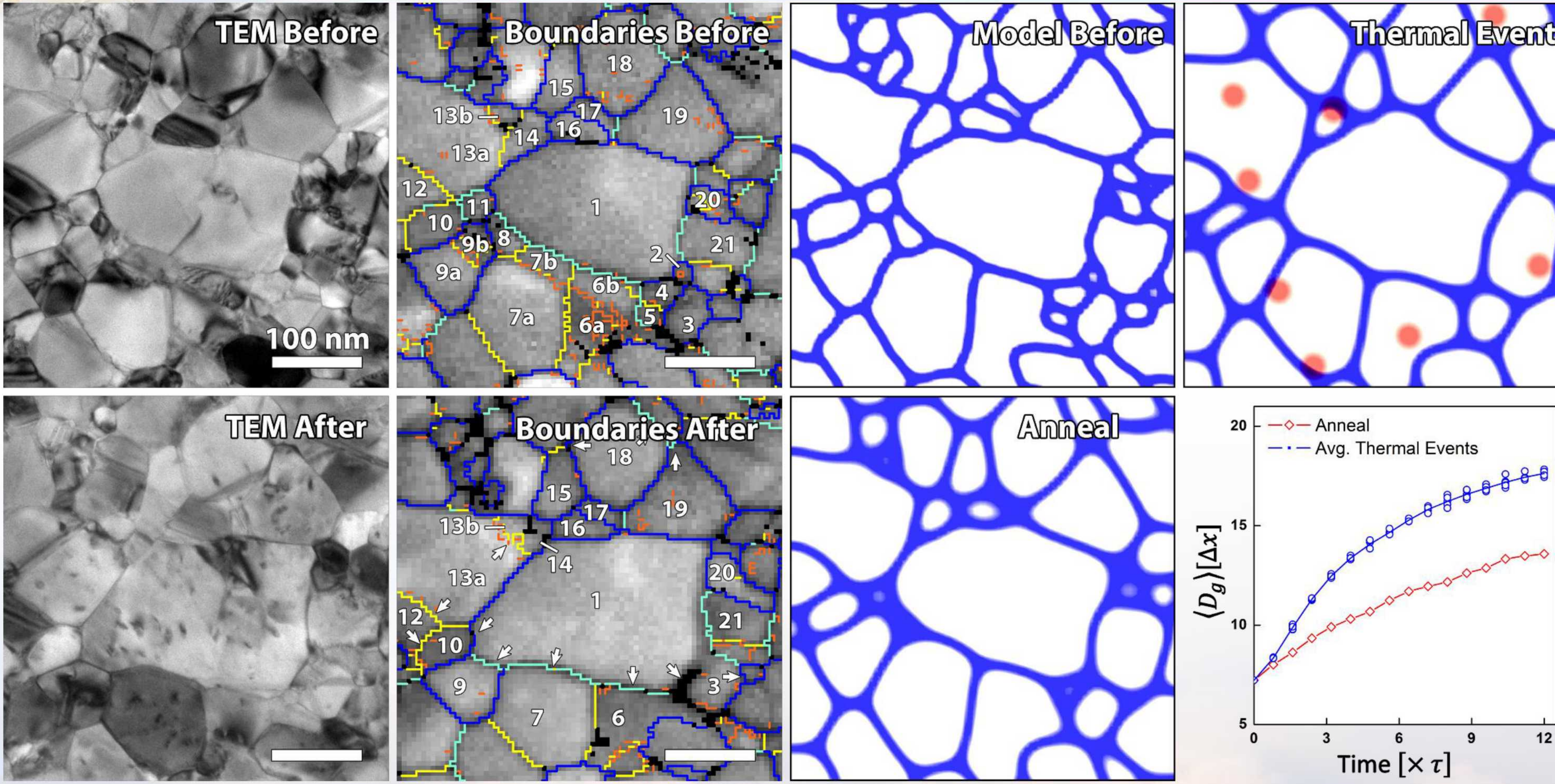
Direct Comparison to Mesoscale Modeling

Collaborators: D.C. Bufford, F. Abdeljawad, & S.M. Foiles

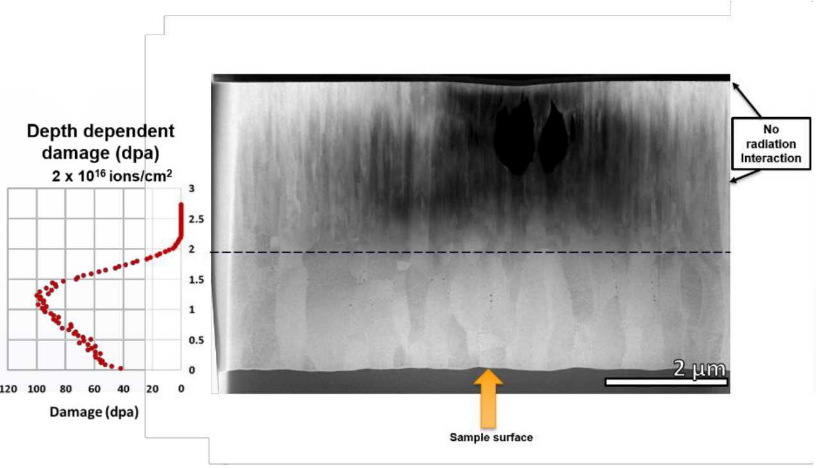


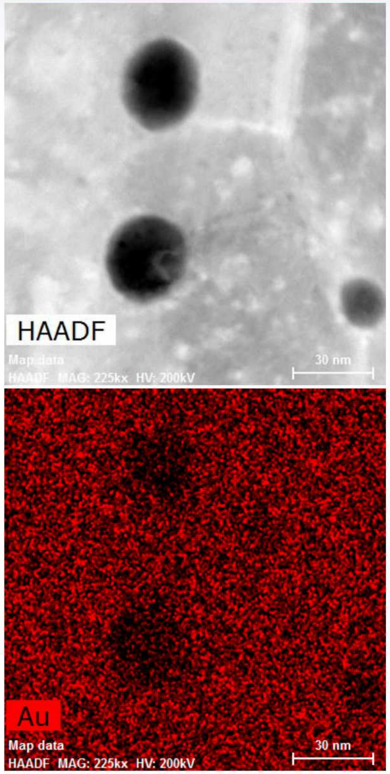
Because of the matching length scale, the initial microstructure can serve as direct input to either MD of mesoscale models & subsequent structural evolution can be directly compared.

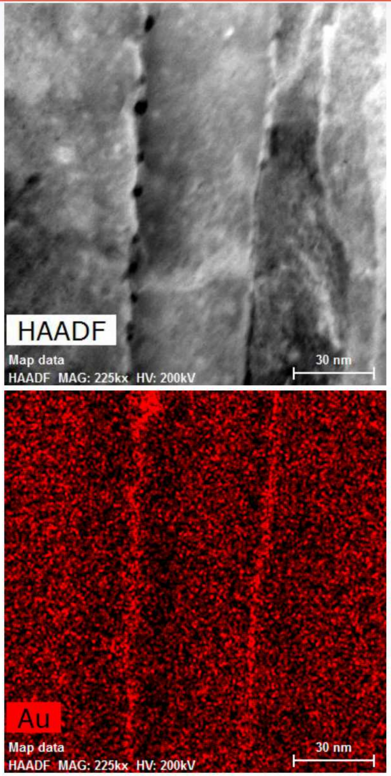
Exploring the Radiation Stability of Thermally Stable Nanocrystalline Metals

Collaborators: C.M. Barr, D. Adams

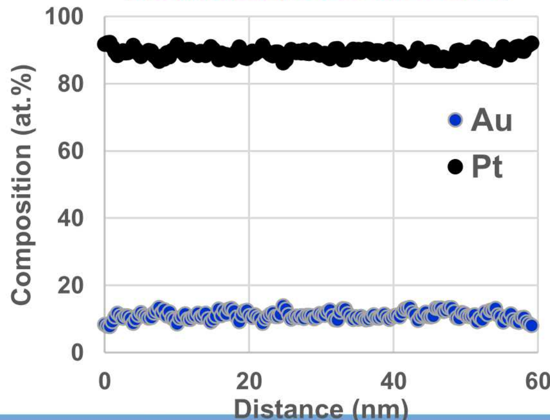
- **Pre-Irradiation Heat Treatment**: 500° C/2hrs in vacuum furnace
- **Irradiation conditions**: 20 MeV Au^{4+}; No external heating (RT plus beam heating)

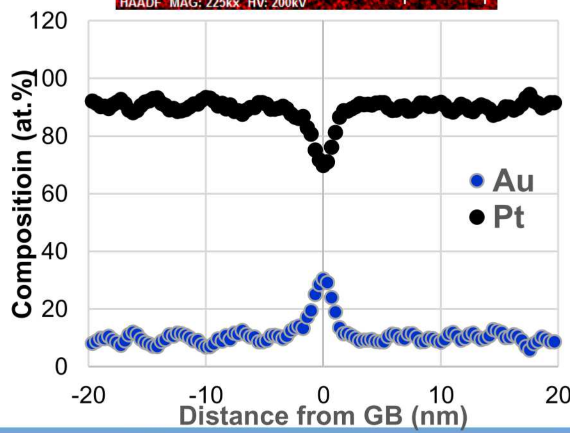






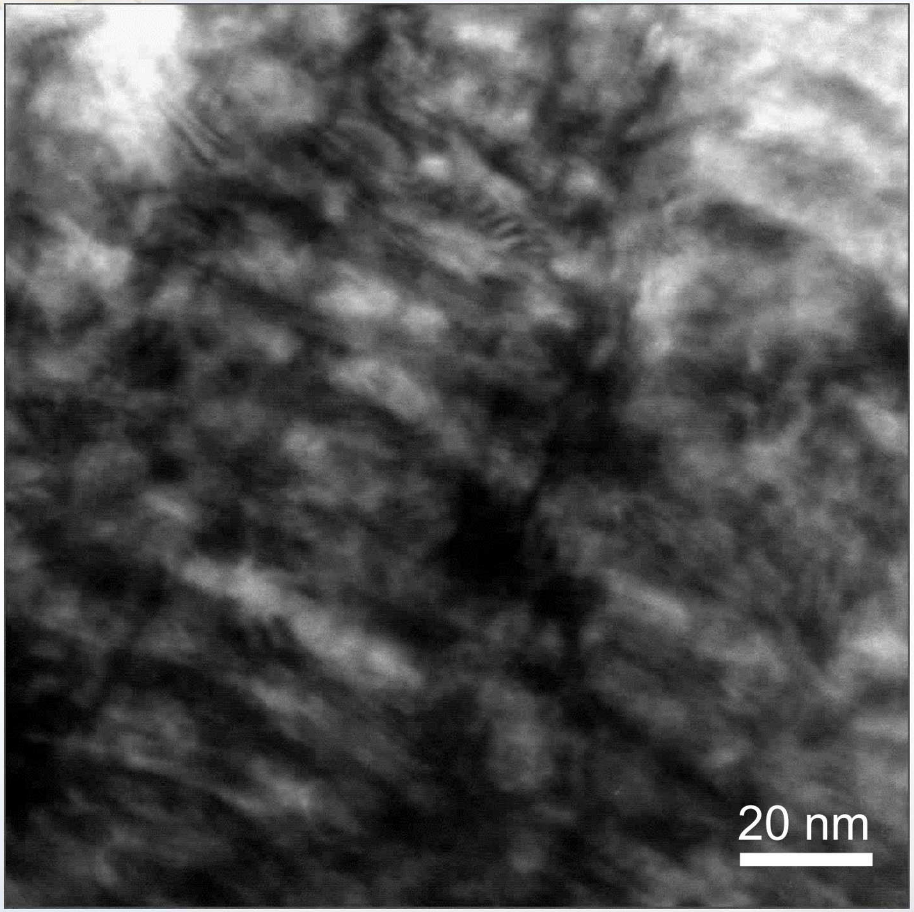
Thermal Stability ≠ Radiation Stability at least not in Pt-10Au

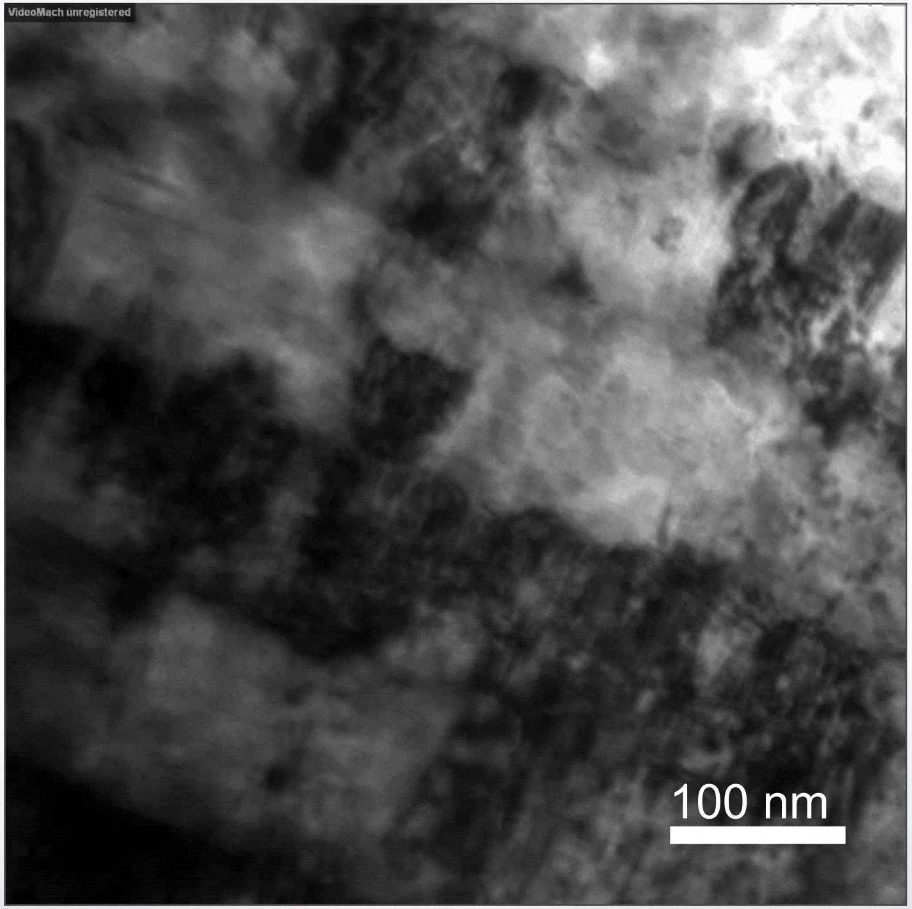




Layer Deposition to Provide Defect Sinks

Collaborators: Y. Chen, N. Li, D. Bufford, & X. Zhang



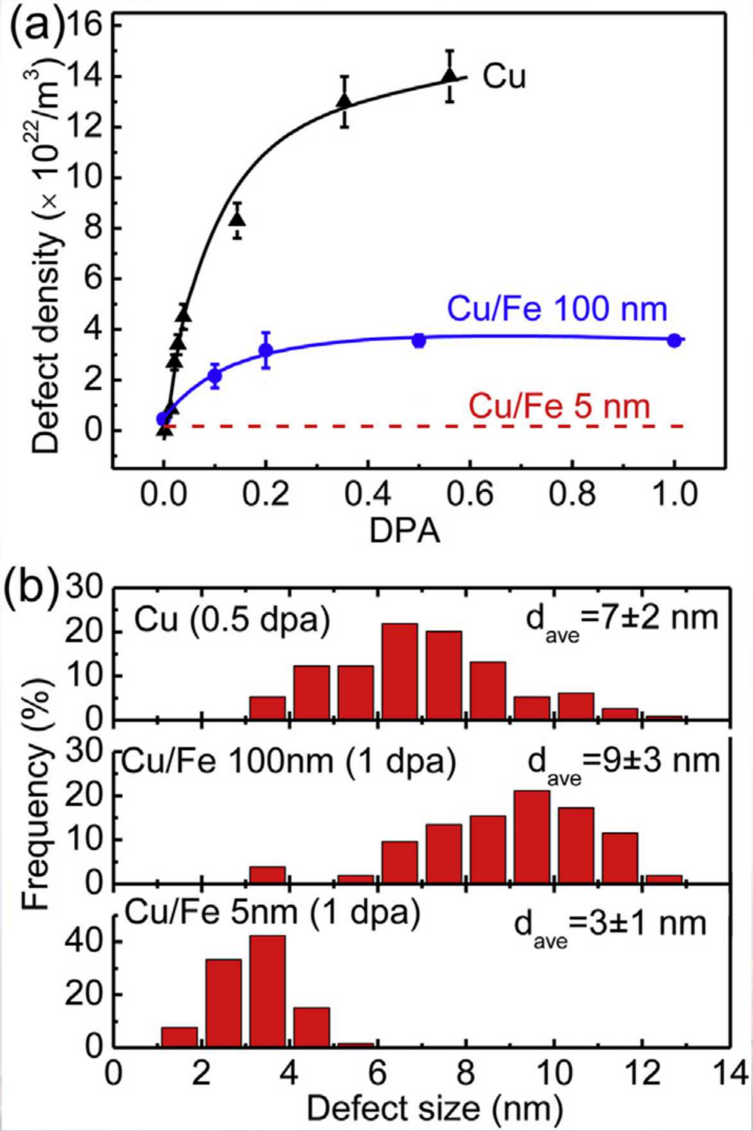


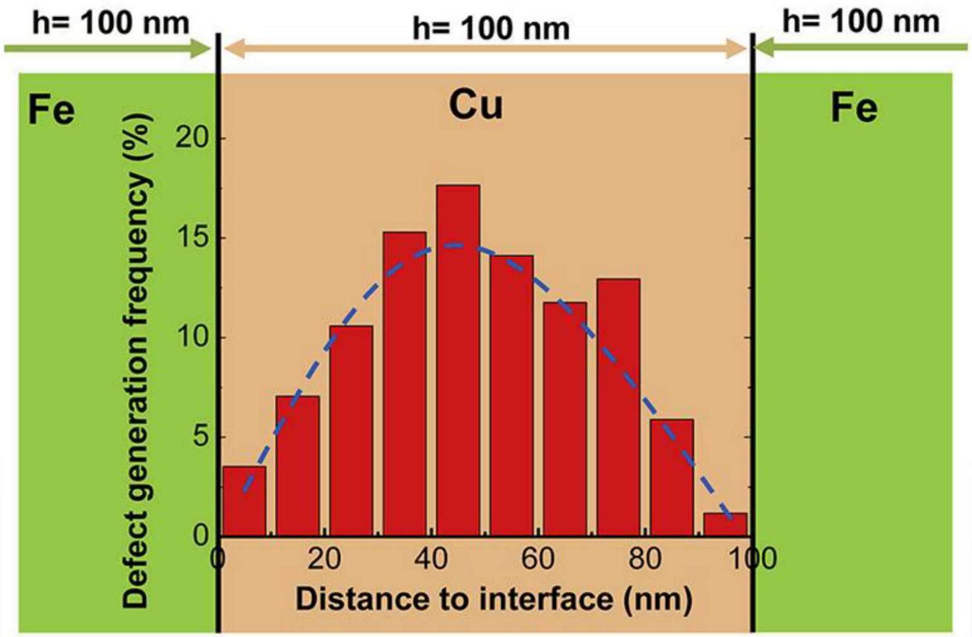
In situ **TEM self-ion irradiation of Cu/Fe 100 nm and 5 nm multilayers with 3 MeV Cu**

Film deposition of layered structures can also provide an opportunity for radiation defect sinks

Layer Deposition to Provide Defect Sinks

Collaborators: Y. Chen, N. Li, D. Bufford, & X. Zhang

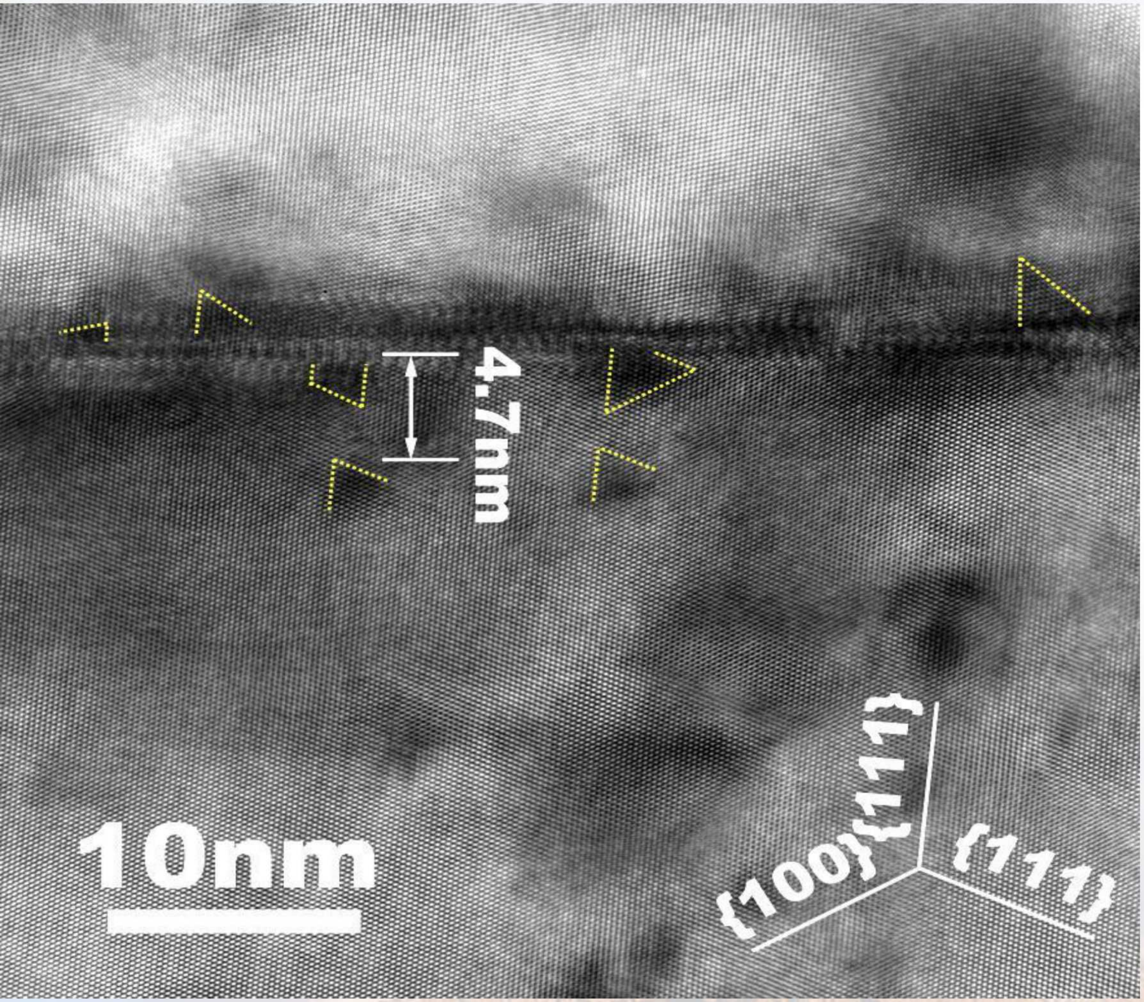


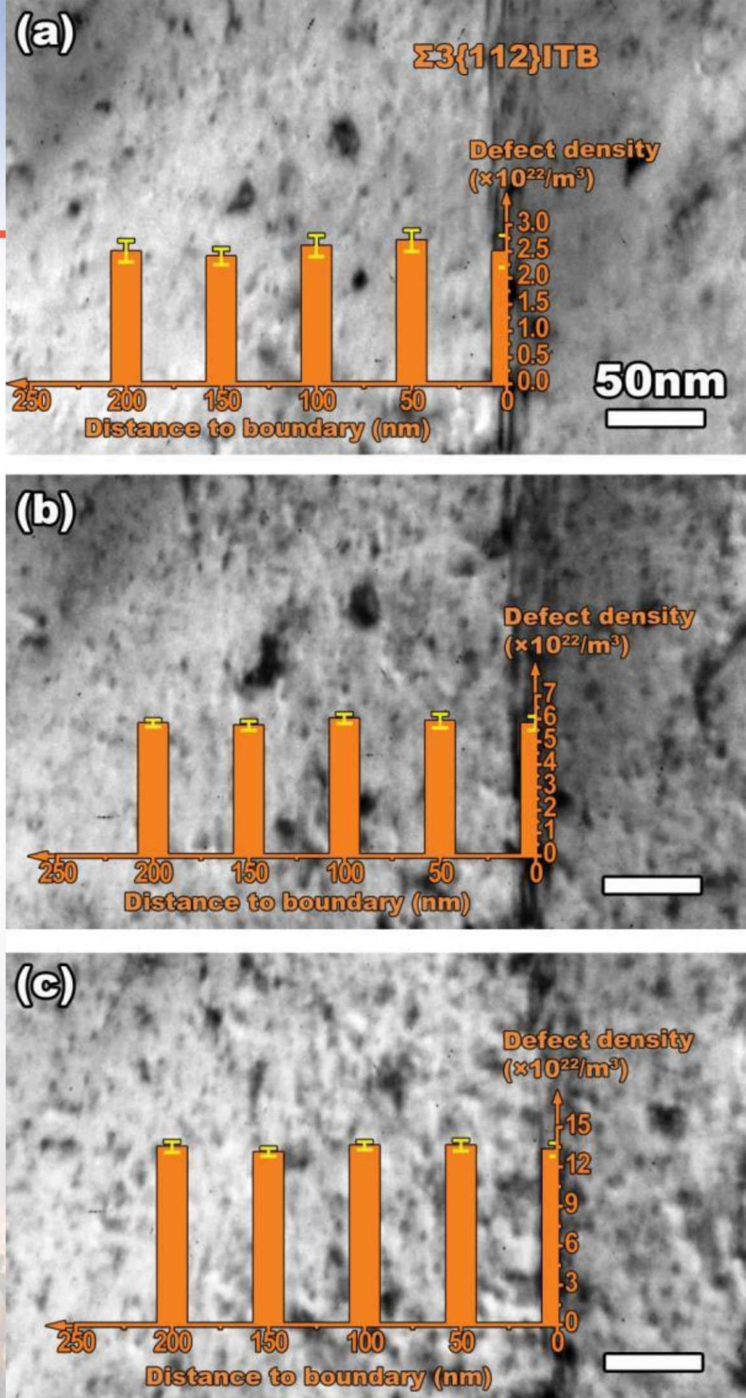


- Layered structure provides significant improvement over either pure system.
- The radiation tolerance improves with decreasing layer thickness.

Defects are Altered Little by the Presence of Grain Boundaries

Collaborators: N. Li & A. Misra





SFT appear to be directly at GB

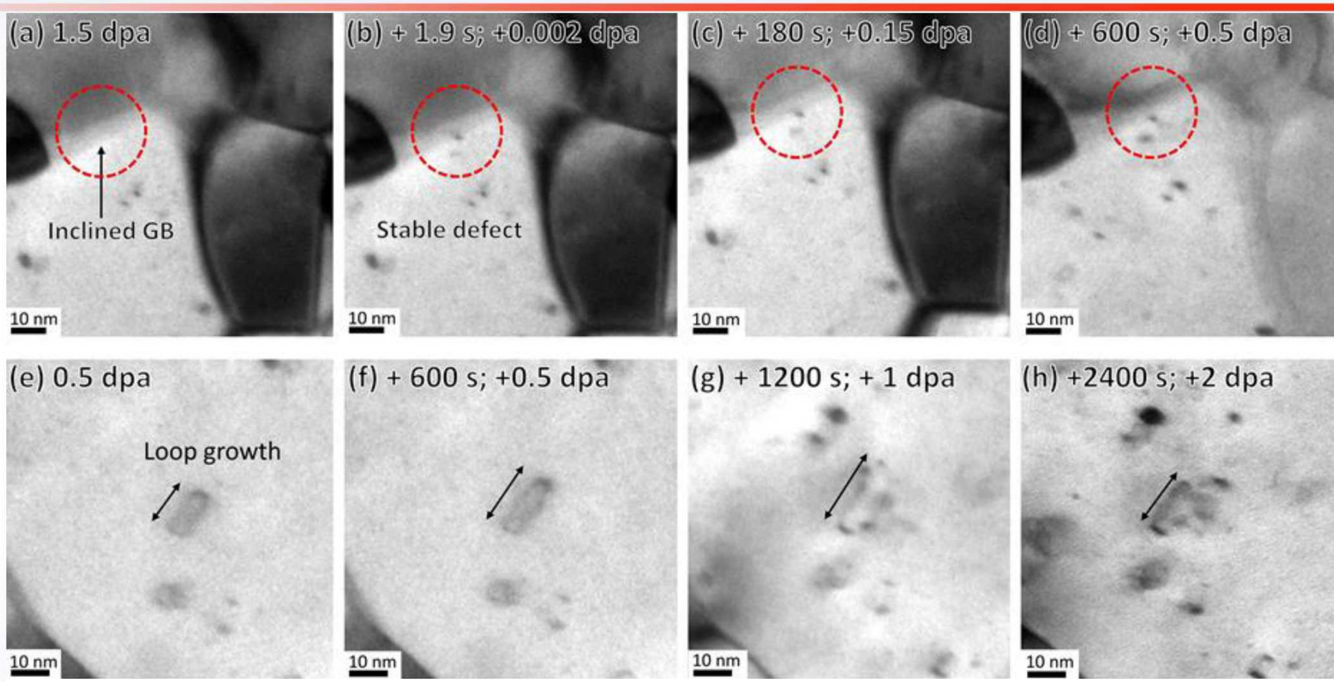
No change in defect density is observed near GB

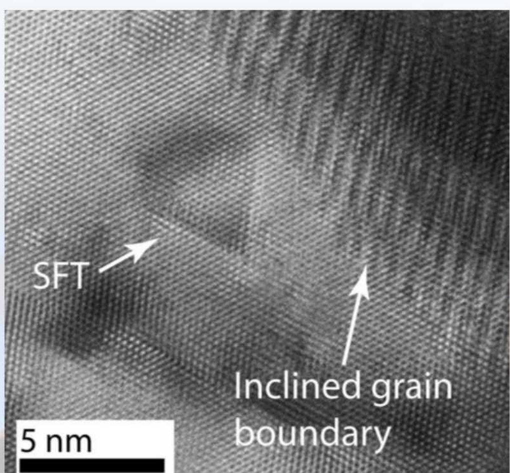
Breakdown of Increased Radiation Stability with Decreasing Grain Size

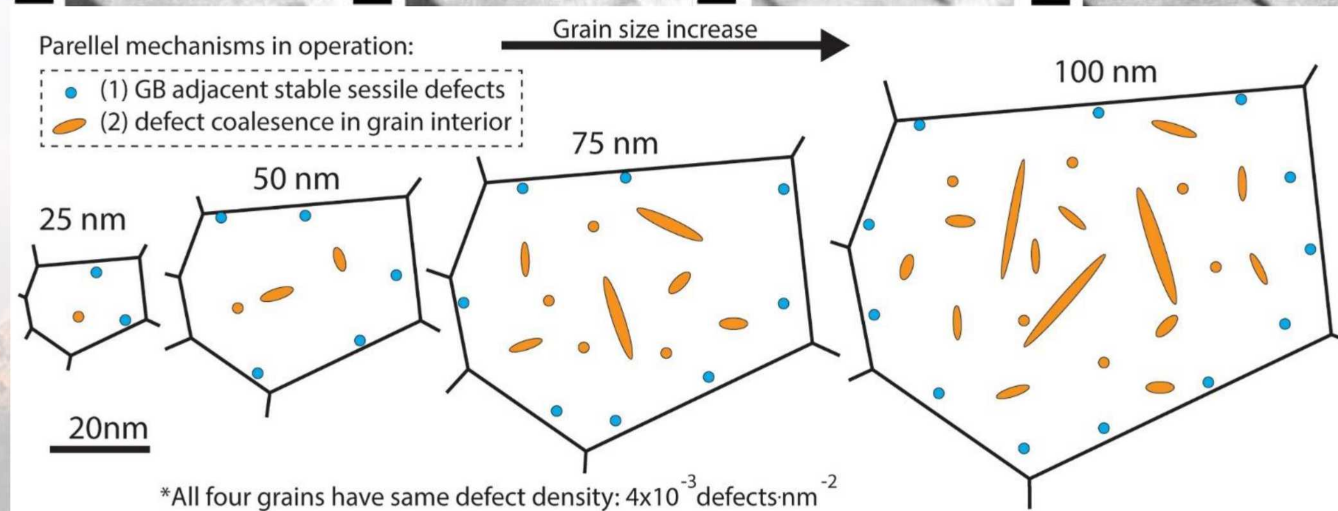
Collaborators: C.M. Barr

Defect evolution near GBs:

Defect evolution in grain interior:

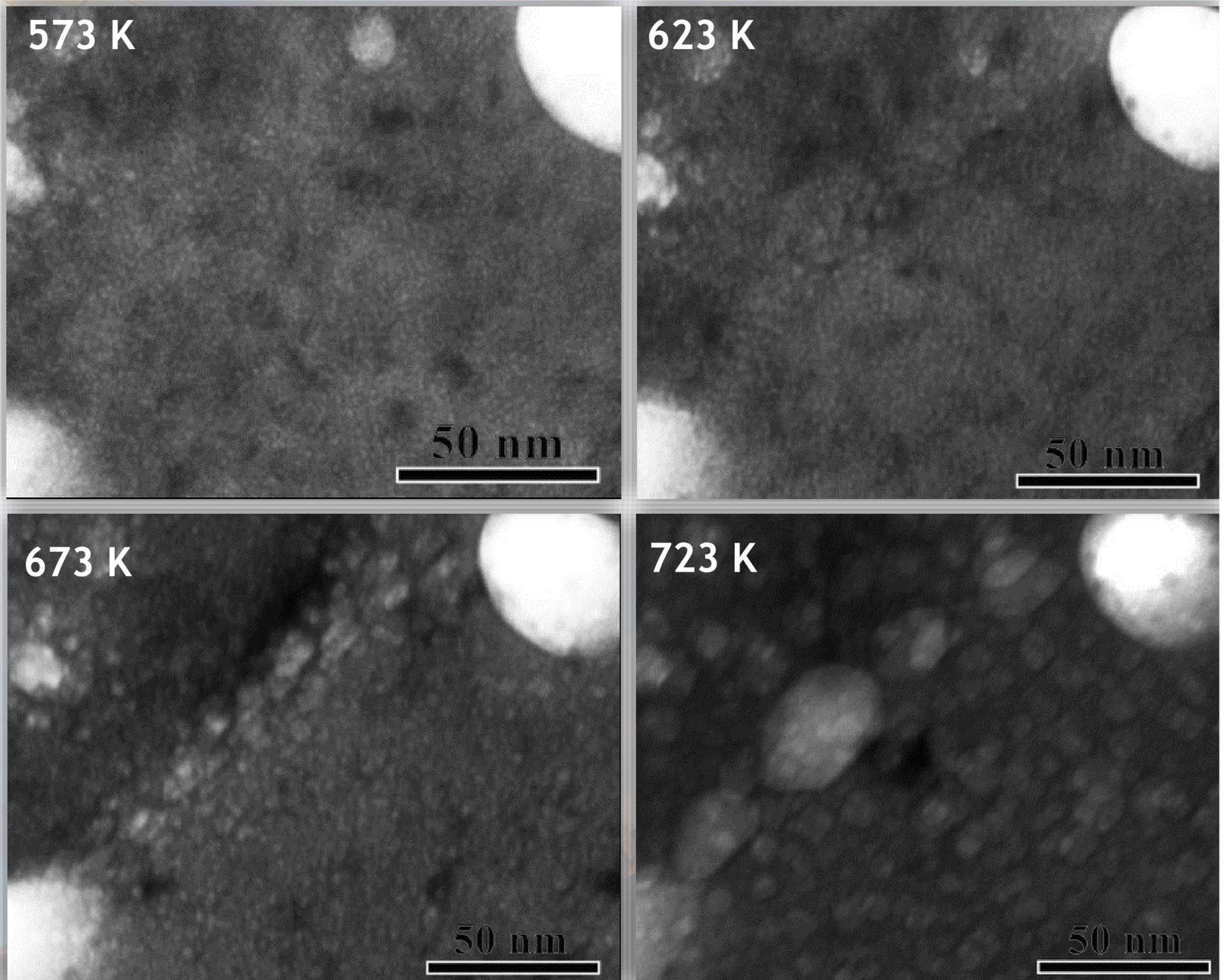






Bubble Migration and Growth in a Single Grain Boundary

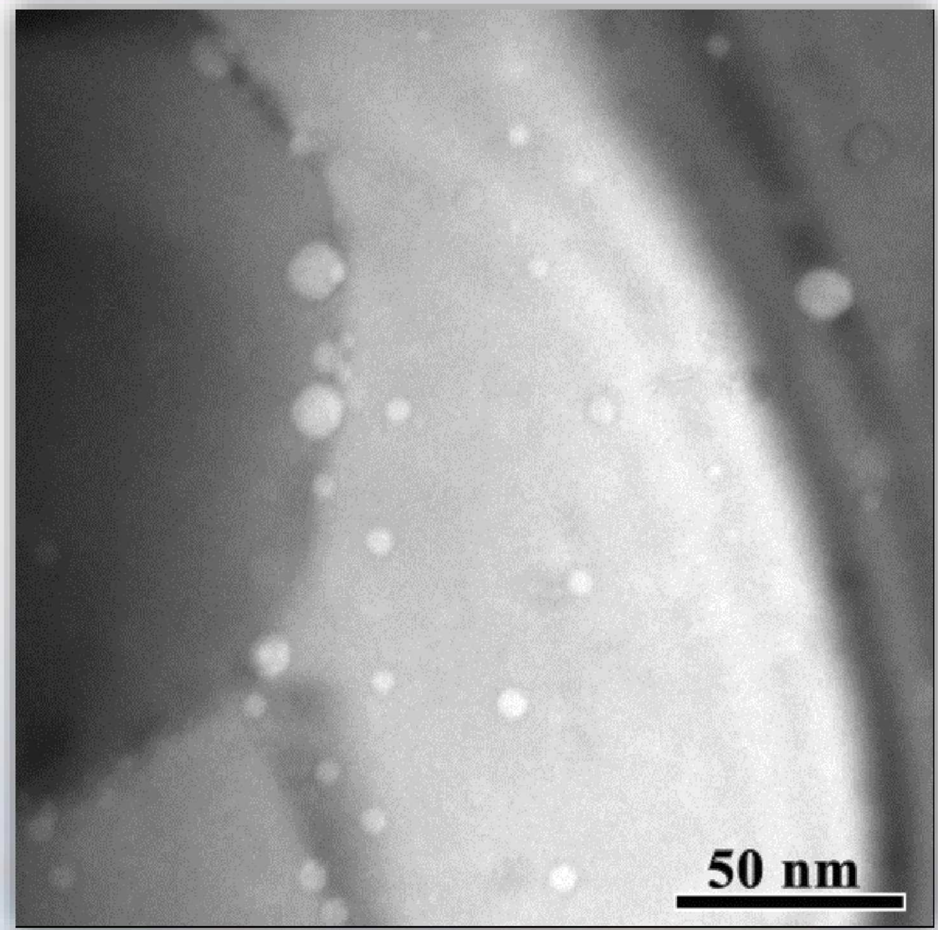
Collaborators: C.A. Taylor, J. Sugar, B. Muntifering

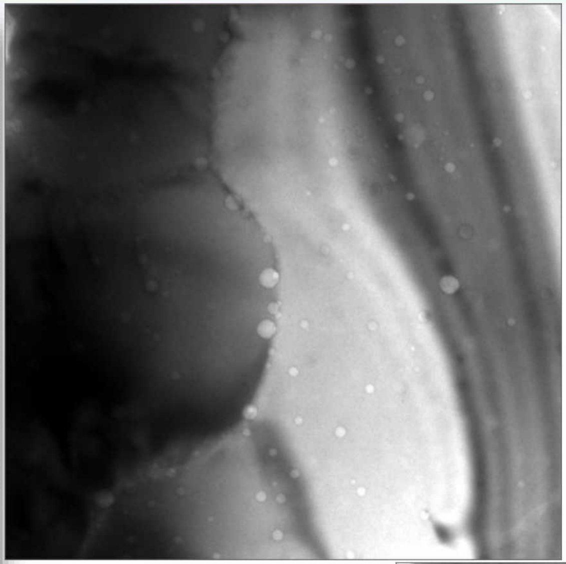


- 573K: Growth at GB from diffusion of interstitial He or small He-V clusters
- 623 K: Growth at GB from diffusion of interstitial He or small He-V clusters
- 673 K: Bubble migration to GB & cavity growth, intragranular growth from diffusion of interstitial He or small He-V clusters
- 723 K: Blister formation at boundaries, intragranular faceted cavity growth.

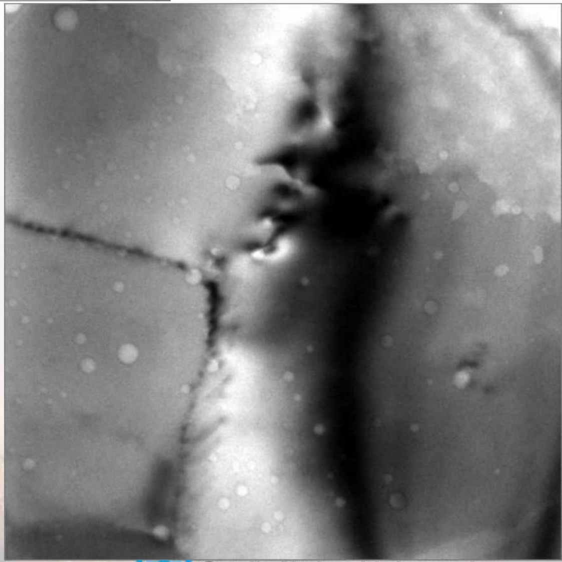
Cavities Role and Grain Boundary Motion are Corelated

Collaborators: C.A. Taylor, J. Sugar, B. Muntifering





Cavities in helium implanted, Pd foil during annealing at 700 ˚C

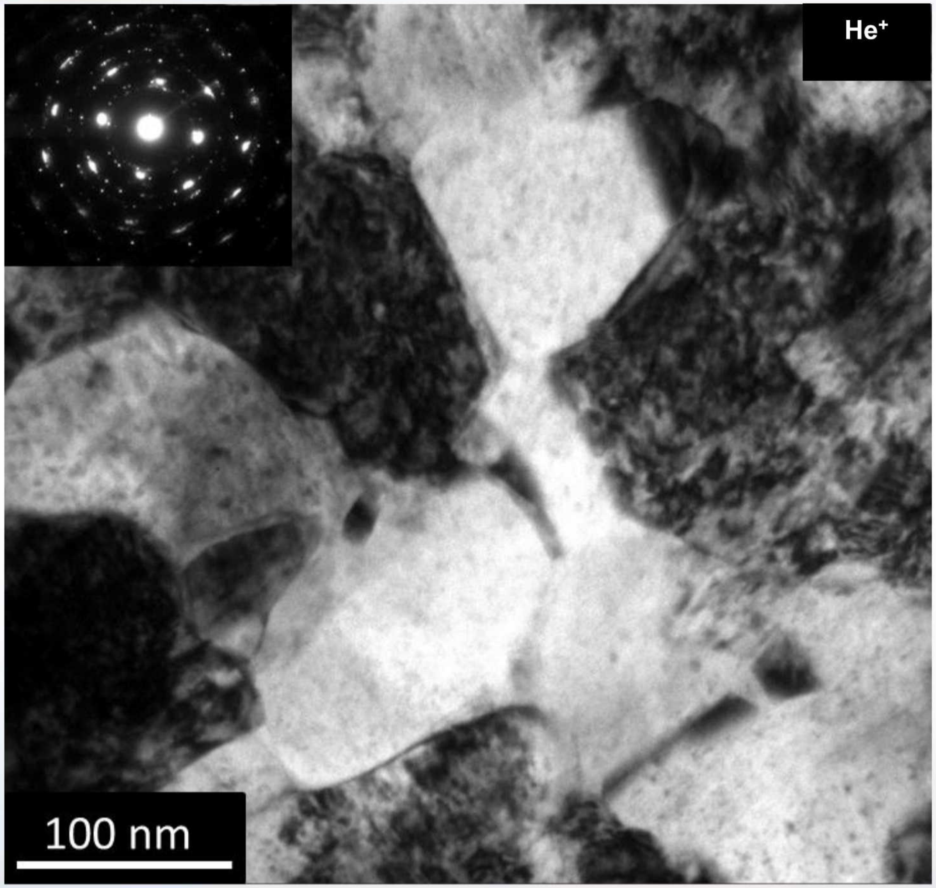


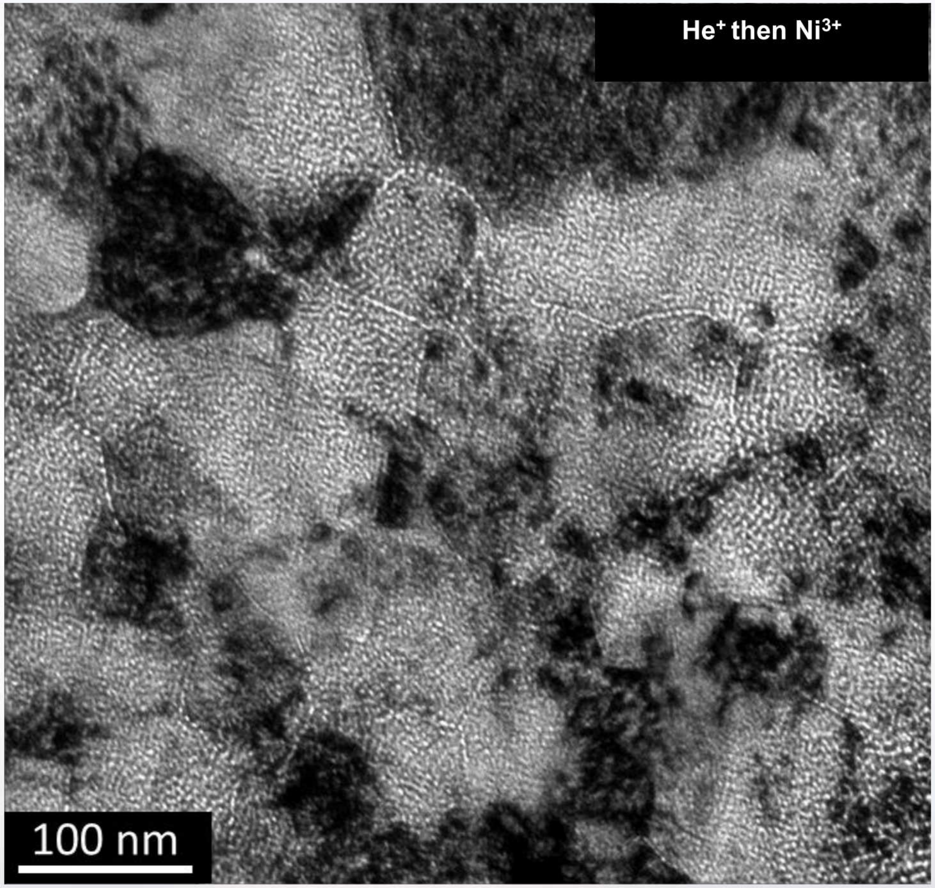
Cavities effect grain boundary mobility, triple junction angle

grain boundary motion alters cavity coalescence

Heterogeneous Bubble Formation under Some Radiation Sequences

Collaborator: B. Muntifering & J. Qu





10^{17} He^+/cm^2

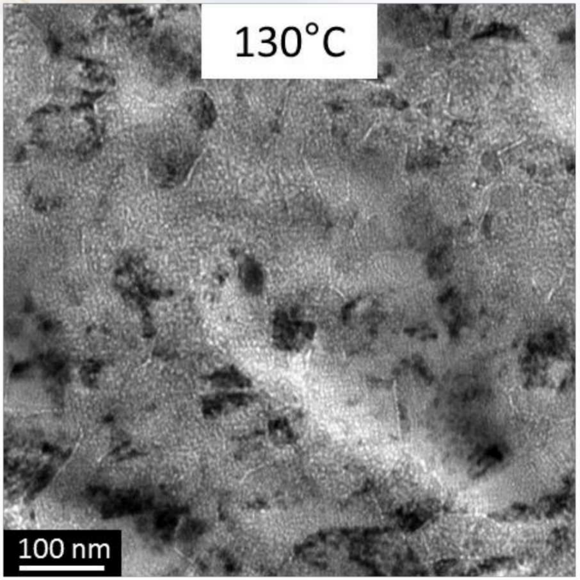
Visible damage to the sample

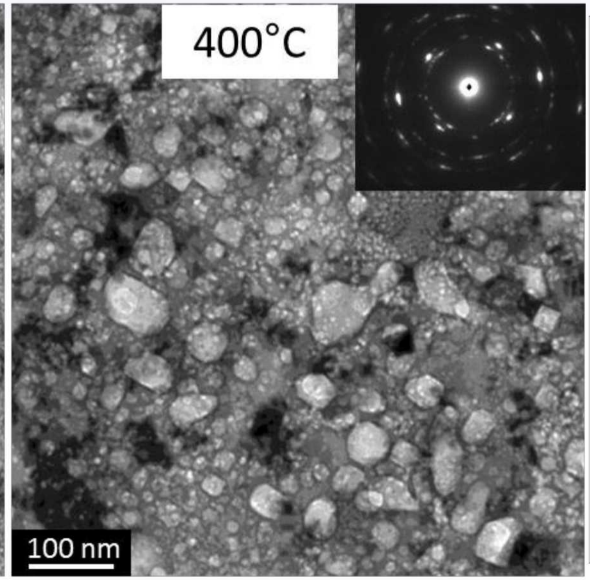
0.7 dpa Ni^{3+} irradiation

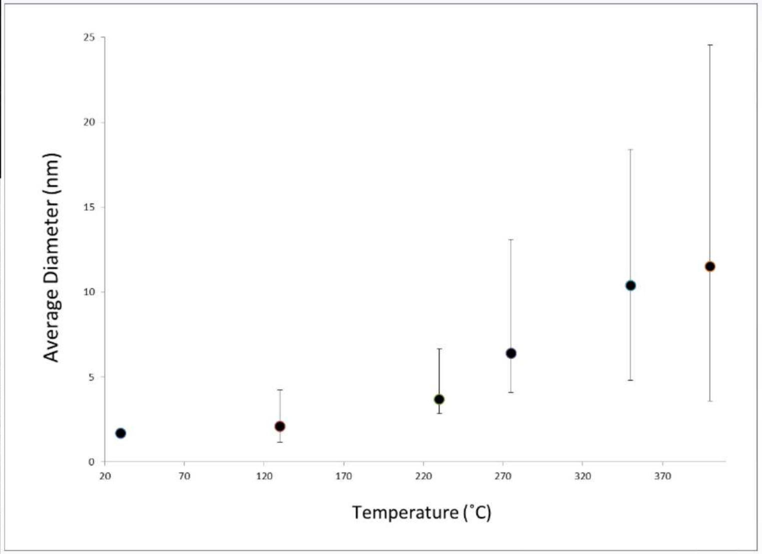
High concentration of cavities along grain boundaries

Cavity Growth during *In situ* Annealing of 10 keV He^{+} Implanted and then 3 MeV Irradiated Ni^{3+}

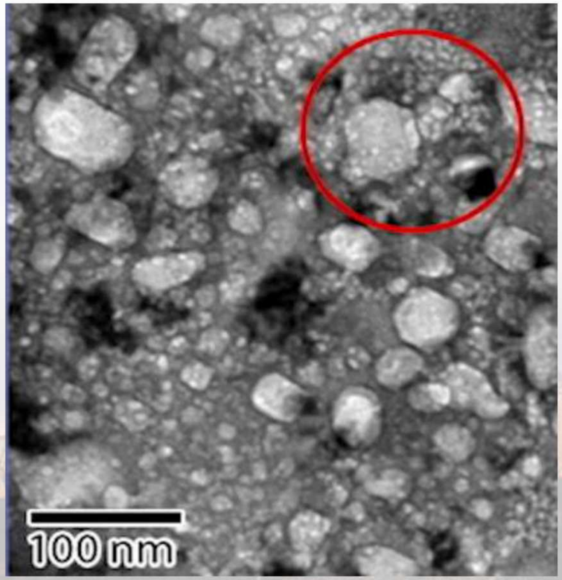
Collaborator: B. Muntifering & J. Qu

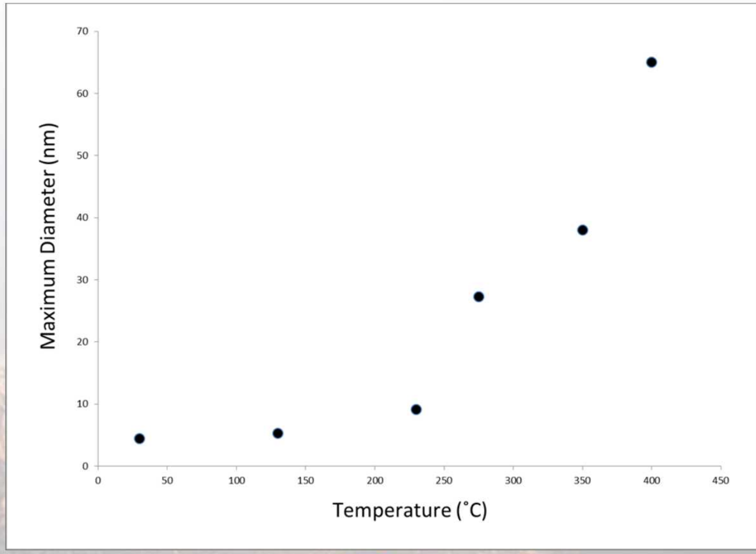






Bubble to cavity transition and cavity evolution can be directly studied





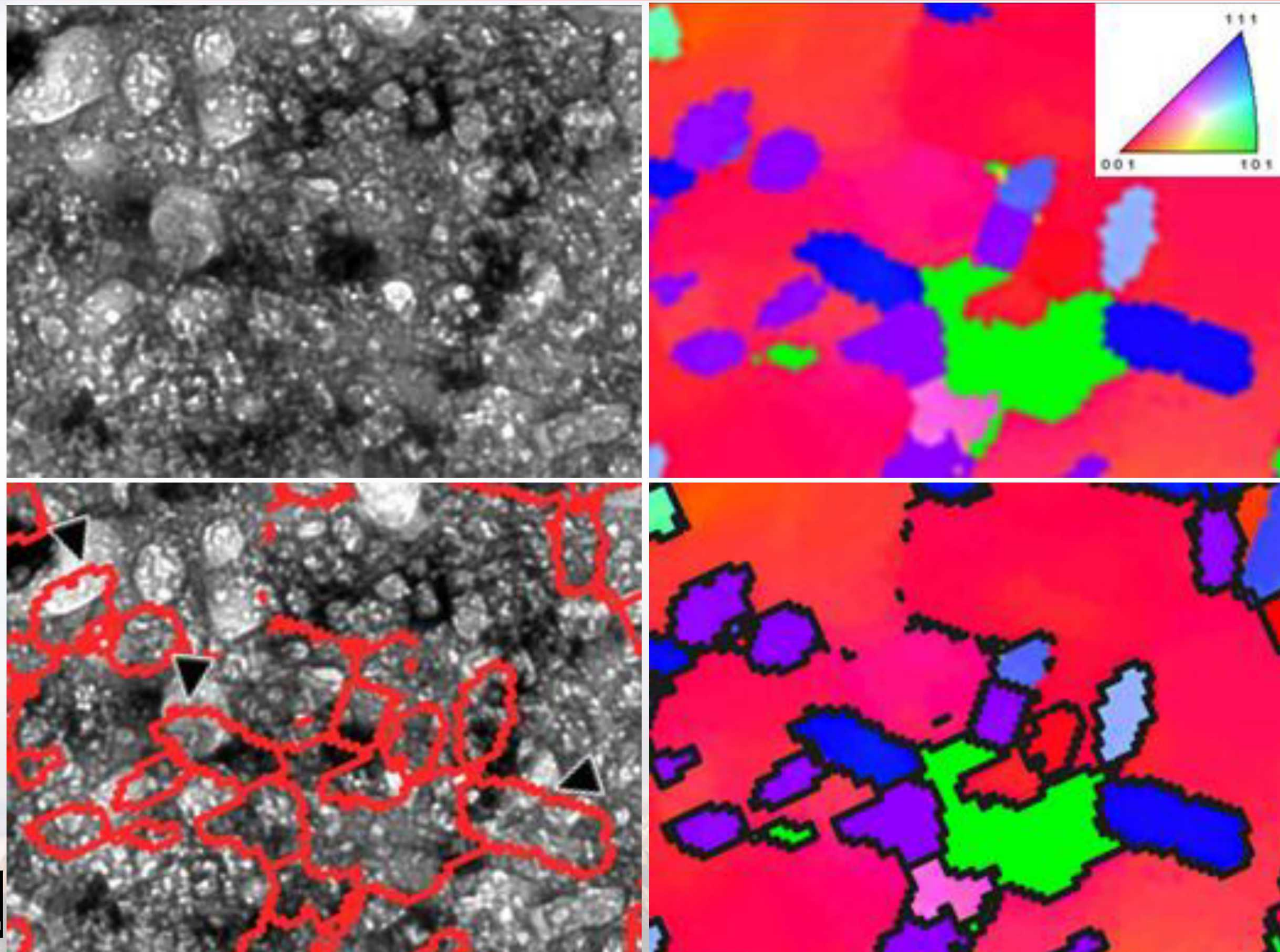
Precession Electron Diffraction Reveals Hidden Grain Structure

Collaborator: B. Muntifering & J. Qu

Cavities in helium implanted, self-ion irradiated,

nc nickel film annealed to 400 ˚C

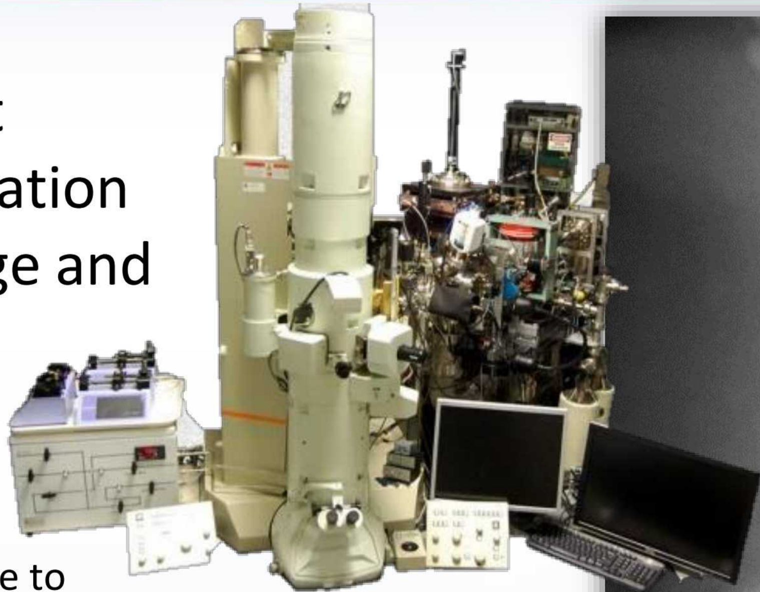
Cavities span multiple grains at identified grain boundaries

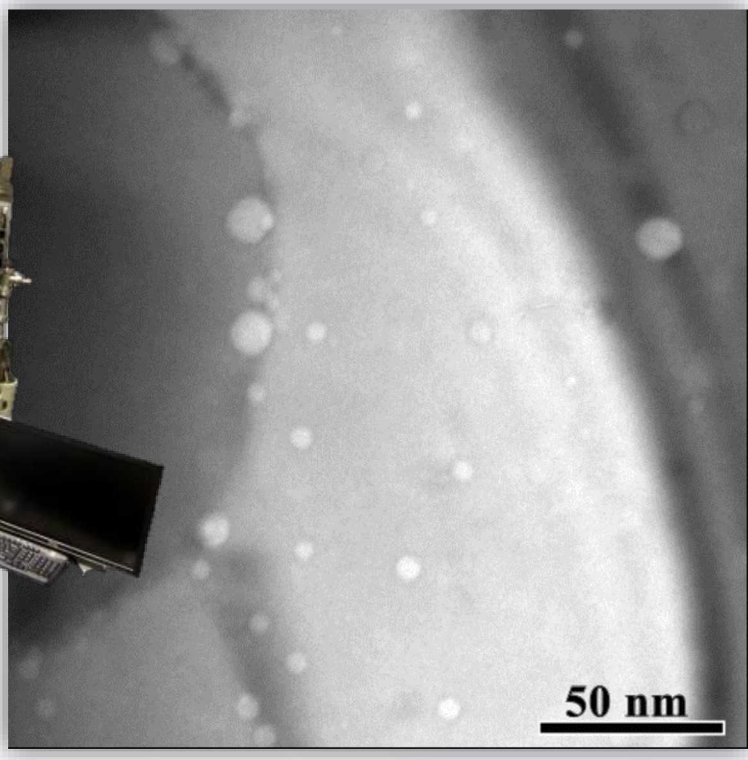


100 nm

Summary

- Sandia's I^3TEM is a unique facility that permits the exploration of radiation damage and various grain boundaries.
- Grain Boundaries:
  - Can become mobile due to radiation damage
  - Can act as a strong sink or weak structural sinks
  - Can provide intense or minimal radiation induced segregation





The response of grain boundaries to radiation damage is complex and depends on many variables!

Collaborators:

- D.L. Buller, C. Chisholm , P. Hosemann, A. Minor, F. Abdeljawad, S.M. Foiles, J. Qu, Sugar, P. Price, M. Abere, S. Briggs, D.B. Robinson, N. Li, A. Misra, Y. Chen, N. Li, & X. Zhang

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Sandia National Laboratories

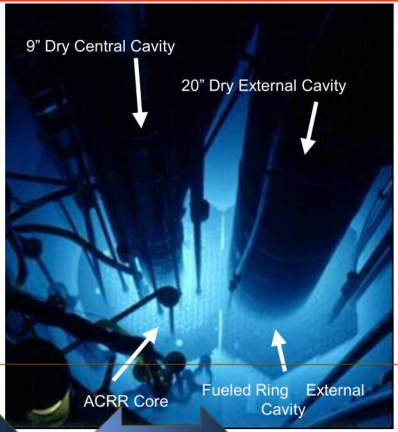
Sandia's USER Capabilities

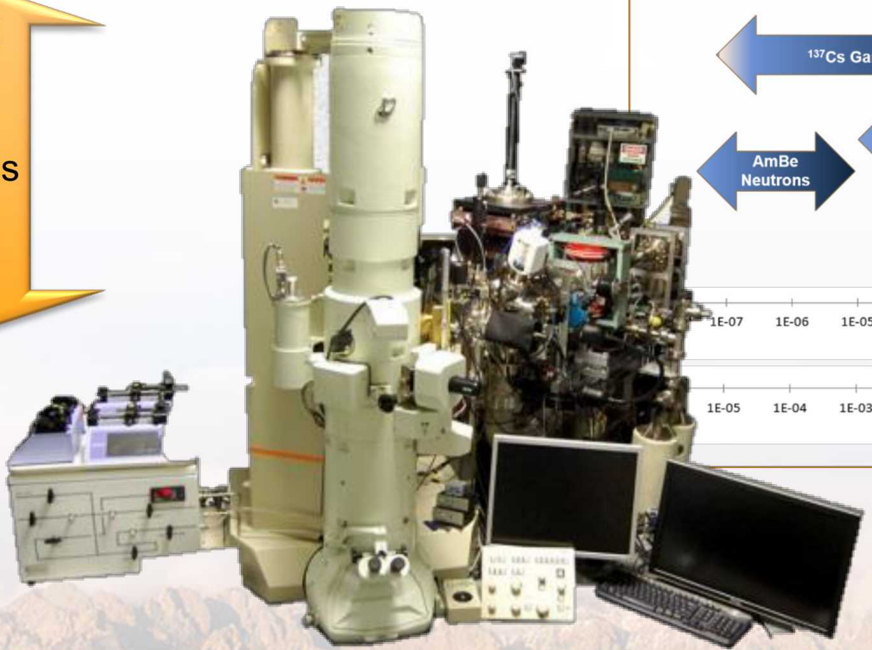
Core Facility - SNL

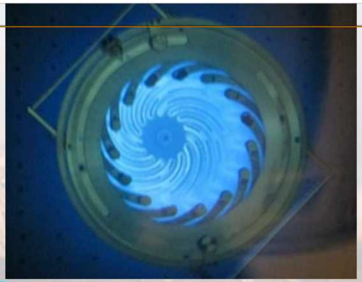
- Nanophotonics & Optical Nanomaterials
- Soft- Biological & Composite Nanomaterials
- Quantum Materials
- In-situ Characterization and Nanomechanics

Gateway Facility - LANL

D. Hanson, W. Martin, M. Wasiolek





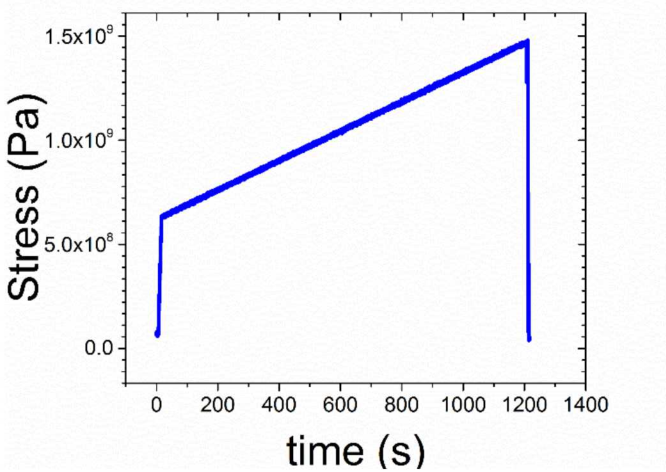


BACK UP SLIDES

Irradiation Creep (4 MeV Cu^{3+} 10^{-2} DPA/s)

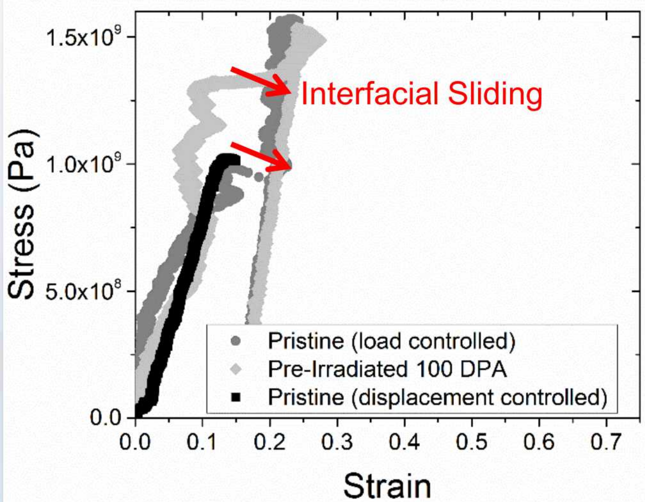
Contributors: G.S. Jawaharram, S. Dillon & R.S. Averback

Controlled Loading Rate Experiments

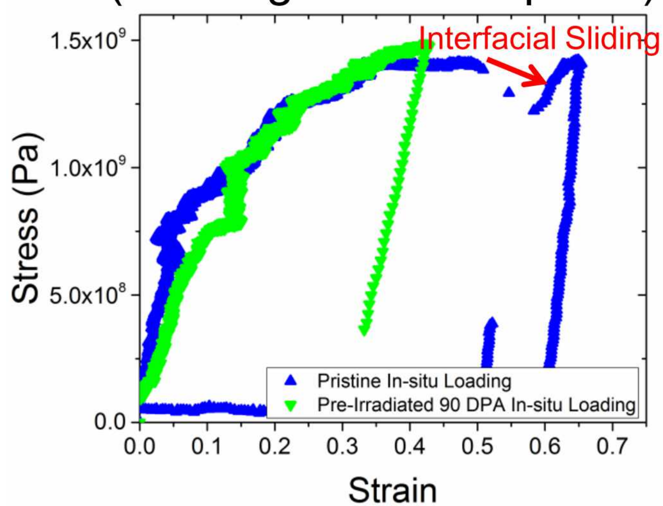


In-situ TEM radiation creep is feasible!

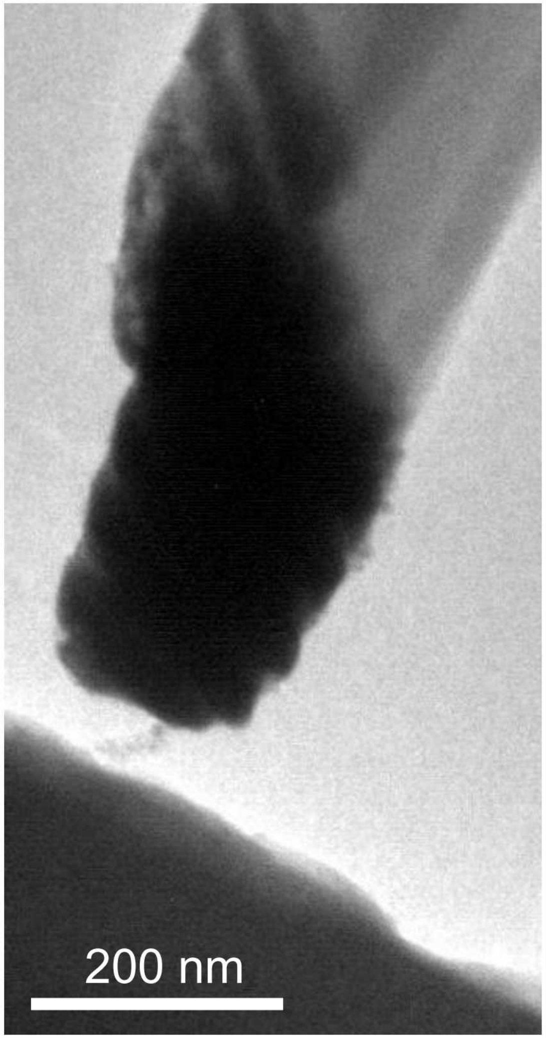
No Irradiation
(Loading rate 0.6 Mpa s^{-1})



Irradiation Creep
(Loading rate 0.6 Mpa s^{-1})



50 nm Cu-W multilayer
20 Min



NC Grain Size Effects: Motivation

- Increase GB density → numerous nanocrystalline studies indicating reduced defect size and/or density of defect
- Limited exploration within the nanocrystalline regime

Can we use in-situ TEM irradiation coupled with defect quantification to understand if any variation exist within nanocrystalline regime (grain size less 100 nm)?

NC Pd defect trends at RT

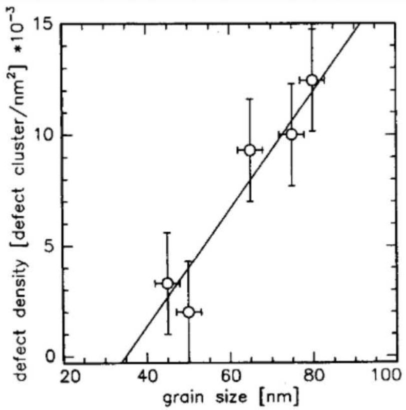
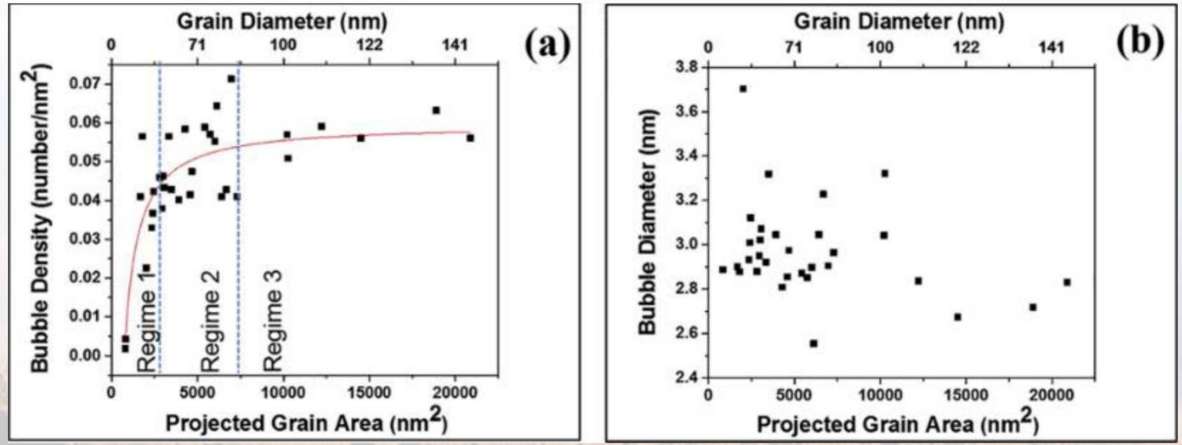


Fig. 8. Defect densities vs. grain size on the irradiated Pd sample (240 keV, 2×10^{16} Kr/cm^2).

Rose et al. (1997)

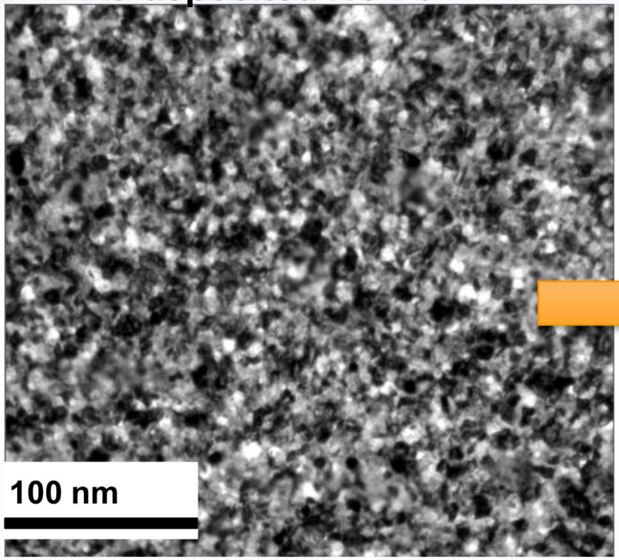
NC Fe Helium bubble trends at 427° C

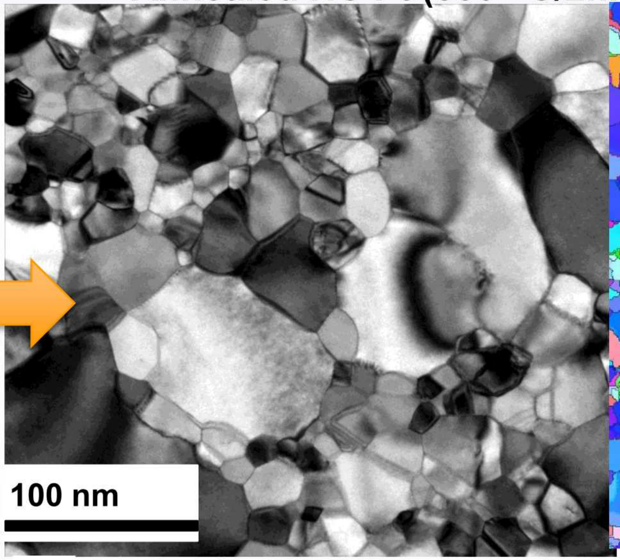


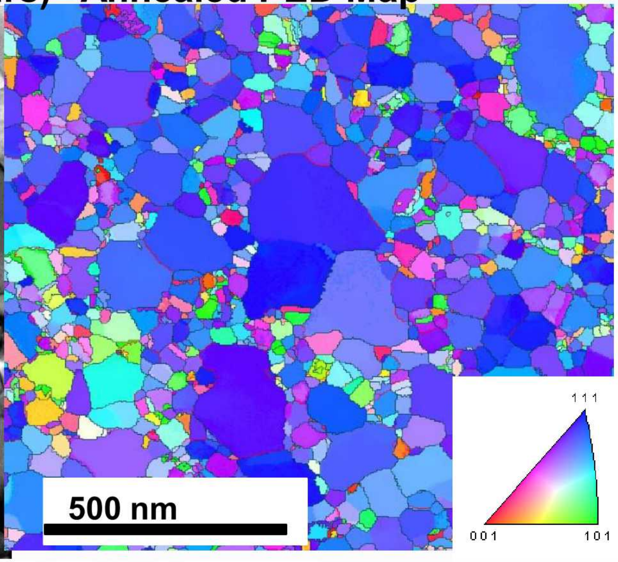
El-Atawani et al. 484 (2017) 236-244.

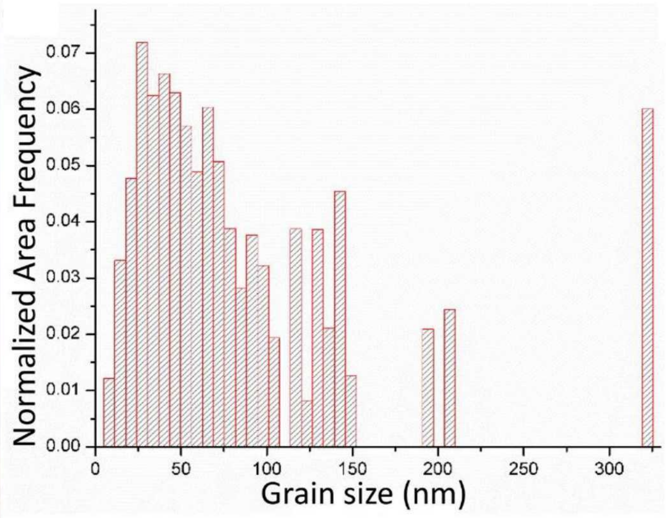
NC Grain Size Effects: Methodology

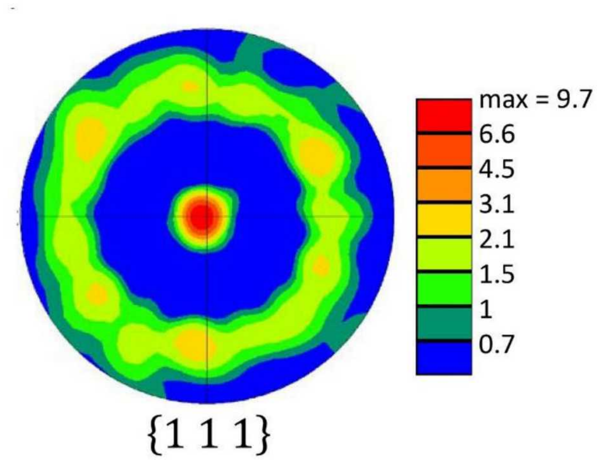
As-deposited NC Pt **Annealed NC Pt (500 º C/2hrs)** **Annealed PED Map**







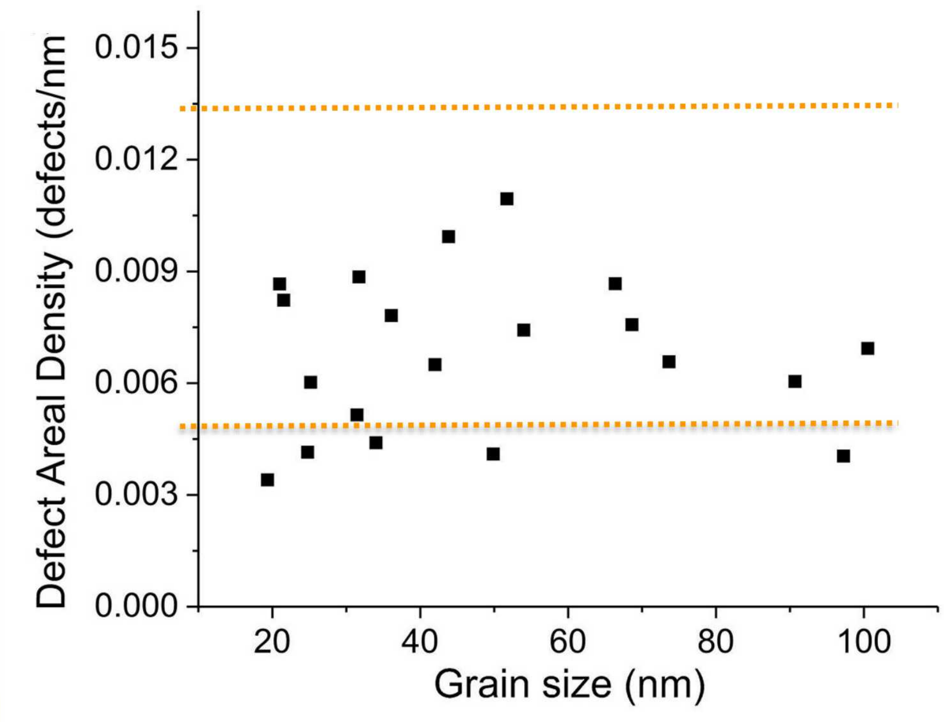


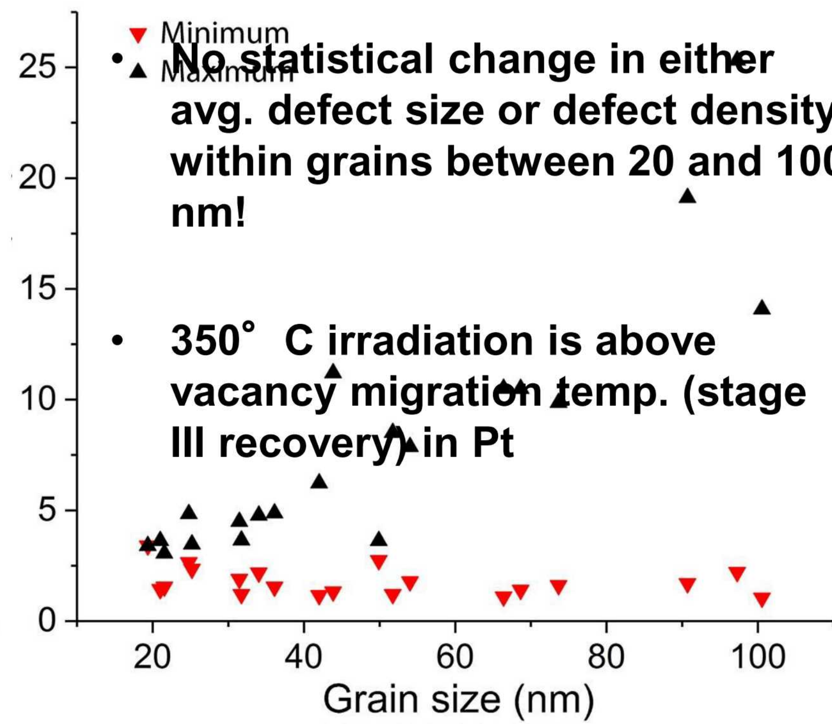


NC Grain Size Effects : Irradiation and Defect Trends

- **In-situ TEM irradiations: 2.8 MeV Au^{4+} at a flux of 4.7 x 10^{10} ions/cm^2s to a fluence of 1.7 x 10^{14} $\frac{ions}{cm^2}$ at 350^{0}C using a Gatan double tilt heating stage**
- **Average final dpa ≈ 3 dpa**

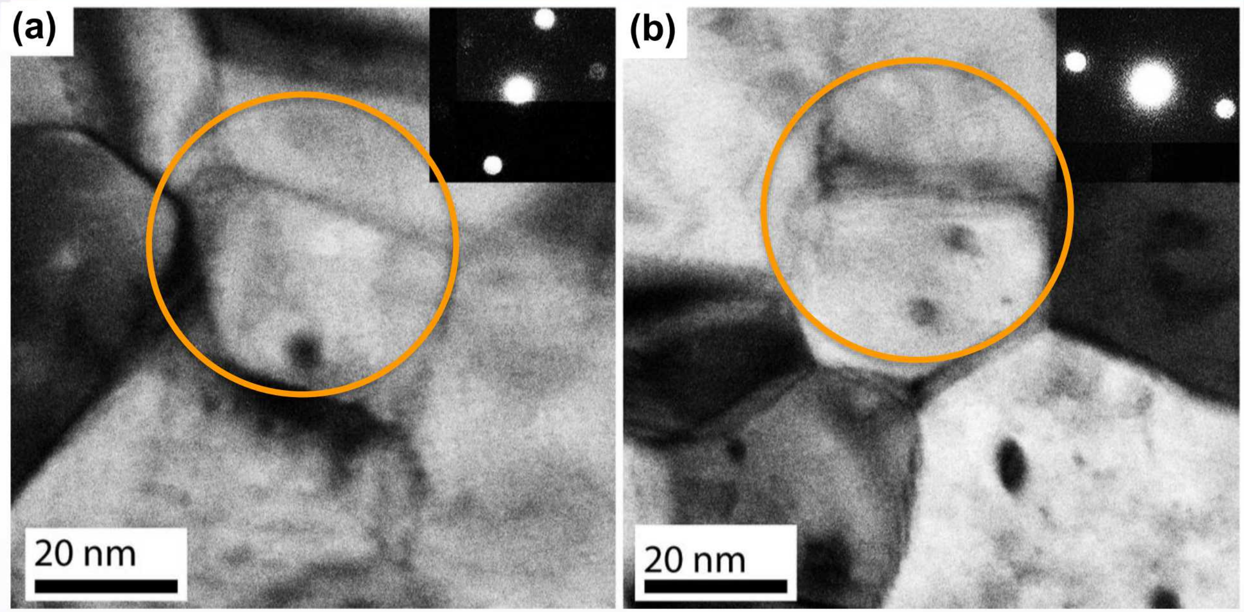
NC grains with a slightly deviated g = <111> diffraction condition using two-beam kinematic bright-field TEM utilizing processed nanobeam electron diffraction





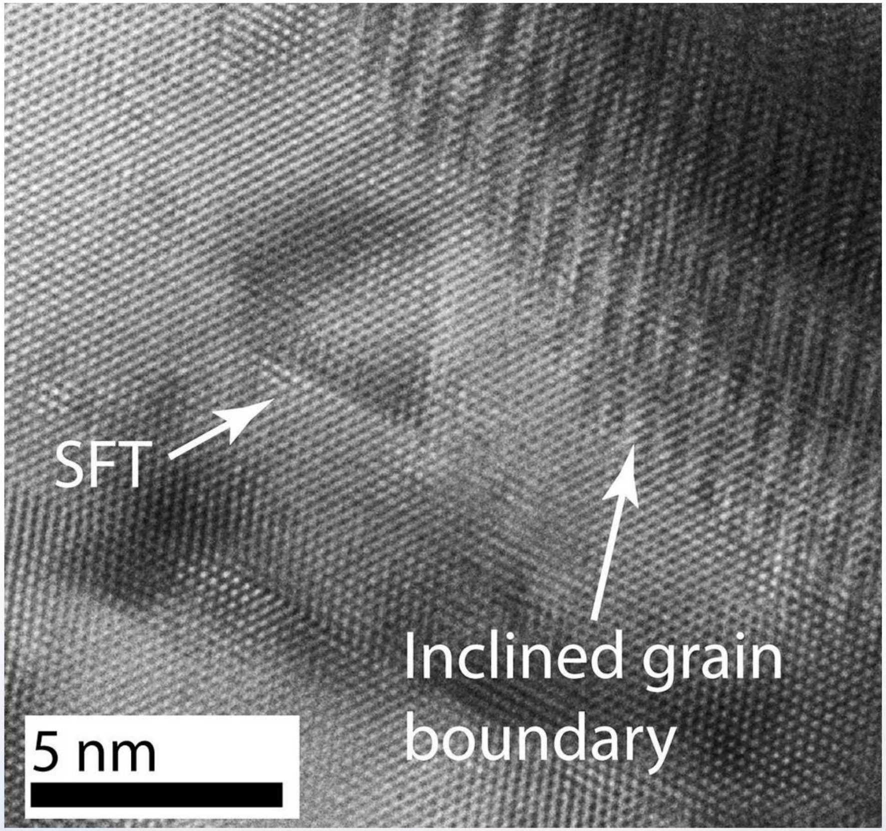
- **No statistical change in either avg. defect size or defect density within grains between 20 and 100 nm!**
- **350° C irradiation is above vacancy migration temp. (stage III recovery) in Pt**

NC Grain Size Effects: Cautionary Tale in Defect Counting



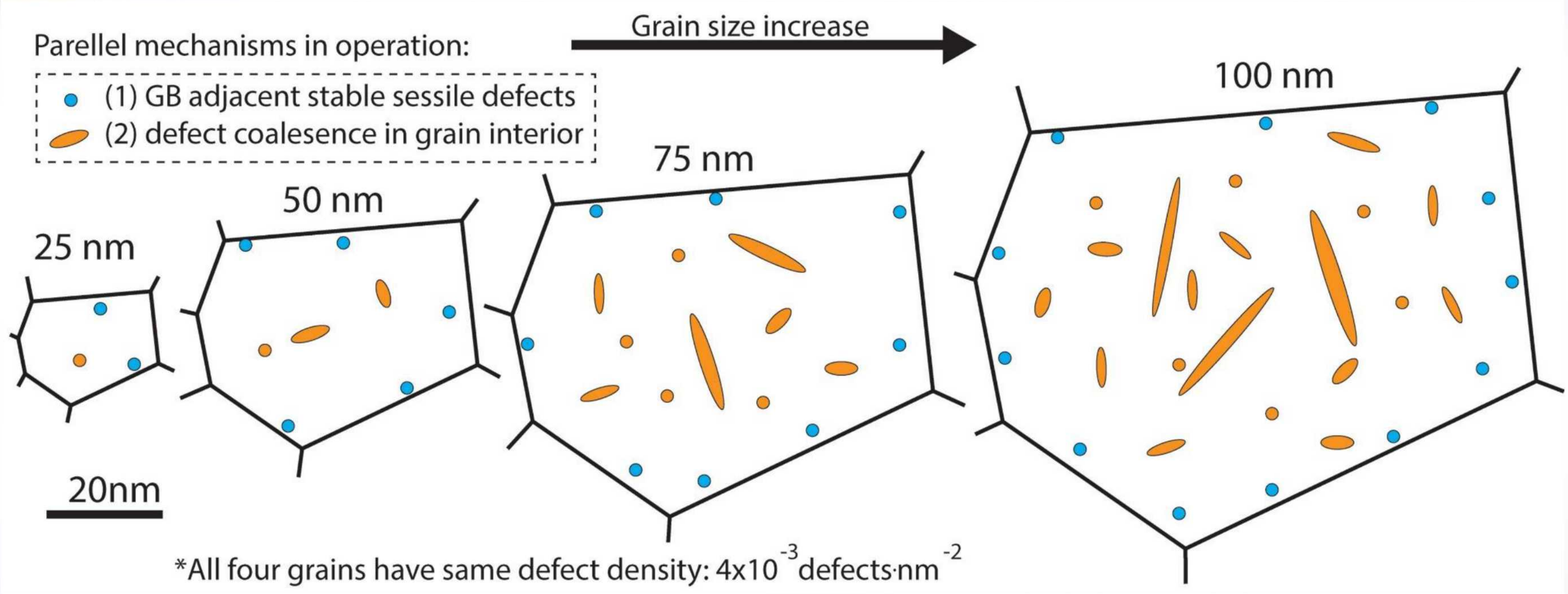
- Two grains → 20 nm in size (measured via mean intercept length)
- How do you count the defects in these grains? 1 to 3 defects nearly full range of density!
  - Grain A = 1 defect → 3.3×10^{-3} defects/nm^2
  - Grain B = 3 defects → 8.6×10^{-3} defects/nm^2
- **With decreasing grain size, # of defects become both more challenging and critical for any type of accurate defect density**

NC Grain Size Effects: Small loop stability near GBs



- Stable sessile defect clusters (1 to 3 nm) adjacent to most GB
- As previously shown, only a small number of these stable defects can impact (increase defect density in small > 40 nm grains)
- "Small stable" defects → majority assumed to be SFTs

NC Grain Size Effects: Proposed Mechanism



- We propose a threshold or breakdown of a reduction in irradiation induced defect density in NC Pt → Analogous to the classic Hall-Petch strengthening breakdown within the nanocrystalline regime

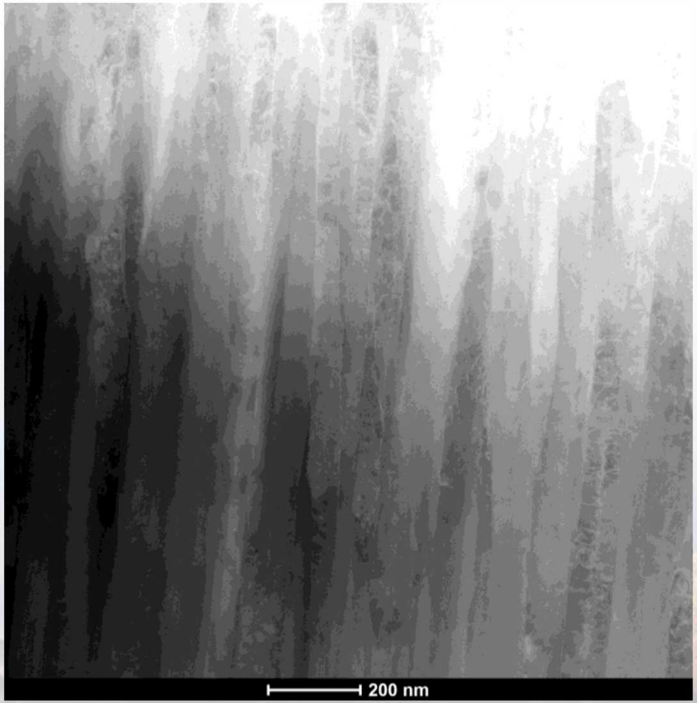
- Results observed in thin film Pt highlight a lack of radiation tolerance (as defined as a reduction in defect size and density) between 100 and 20 nm
- Only one temperature, flux, fluence, material system → more efforts to expand beyond this limited scope!

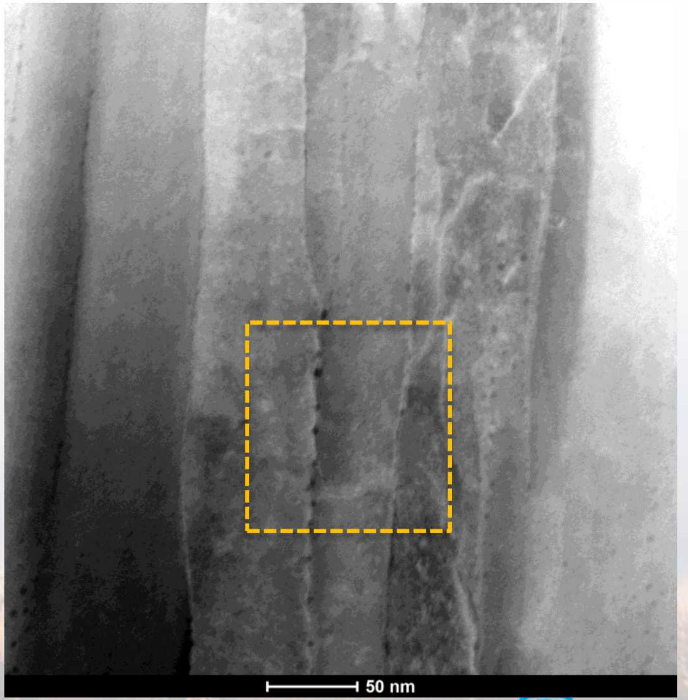
Exploring the effects of ion beam irradiation and mixing on the grain size stability in immiscible binary nanocrystalline alloys

Christopher Barr and Khalid Hattar

Pt-Au Below End of Range: Thermal Region

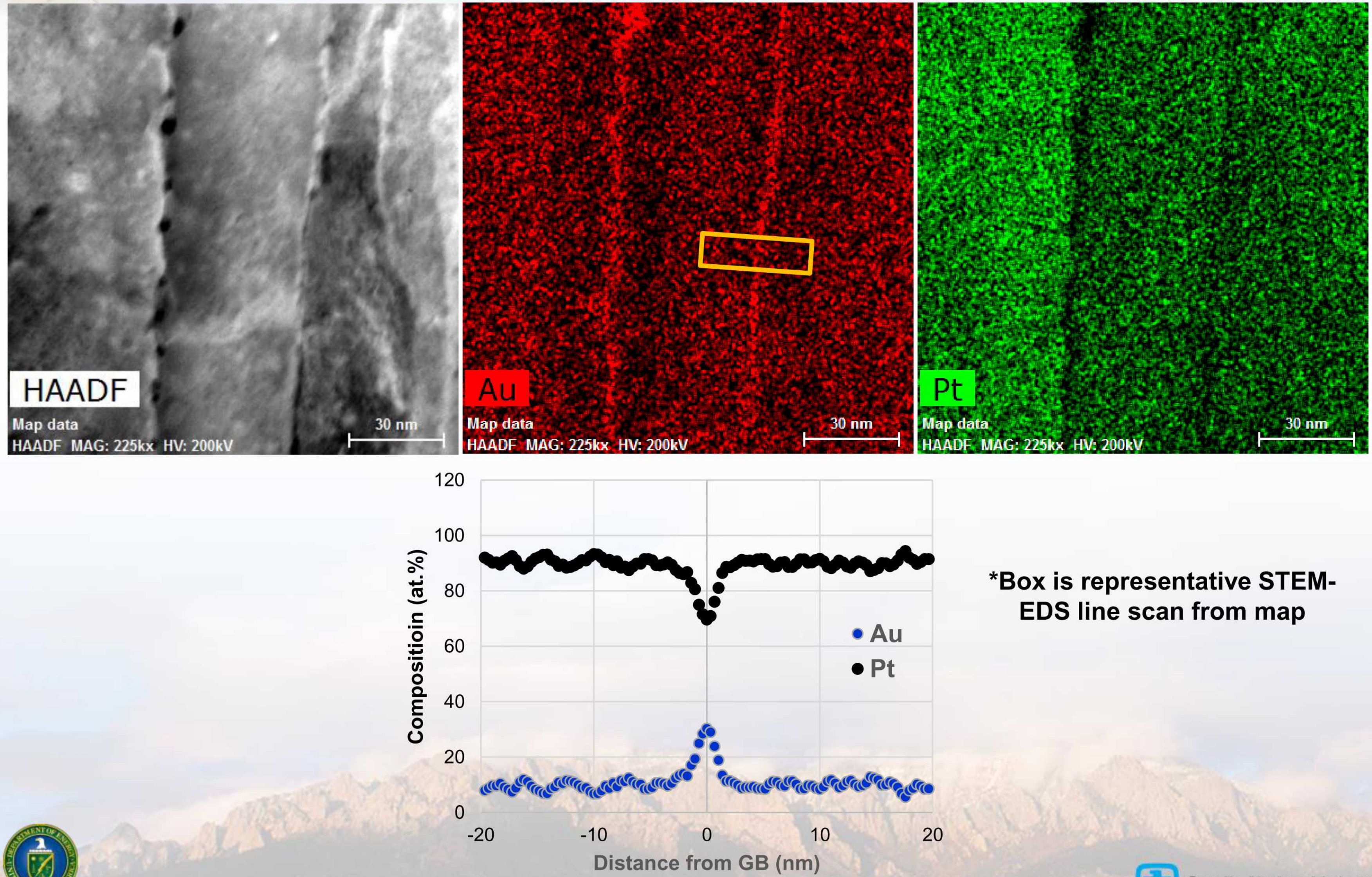
- **Prior to irradiation: 500C – 2hrs (known to induce Au segregation at GBs)**
- **Au at GBs still exist post irradiation (in regions not irradiated – beyond end of range) → Grain size ≈ 50 nm**
  - Au segregation thought to be major factor in thermally stabilizing GBs (i.e. prior SNL studies)
- **STEM-EDS (shown in next slide) examined in region outside radiation depth**





***Box is STEM-EDS area**

Pt-Au Thermal Region: ~ 30-35 at.% Au at GBs

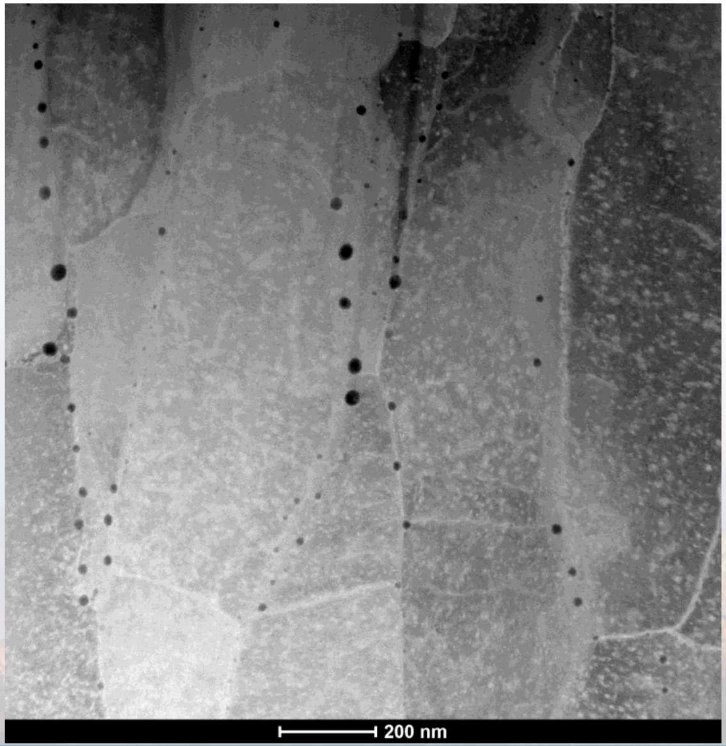


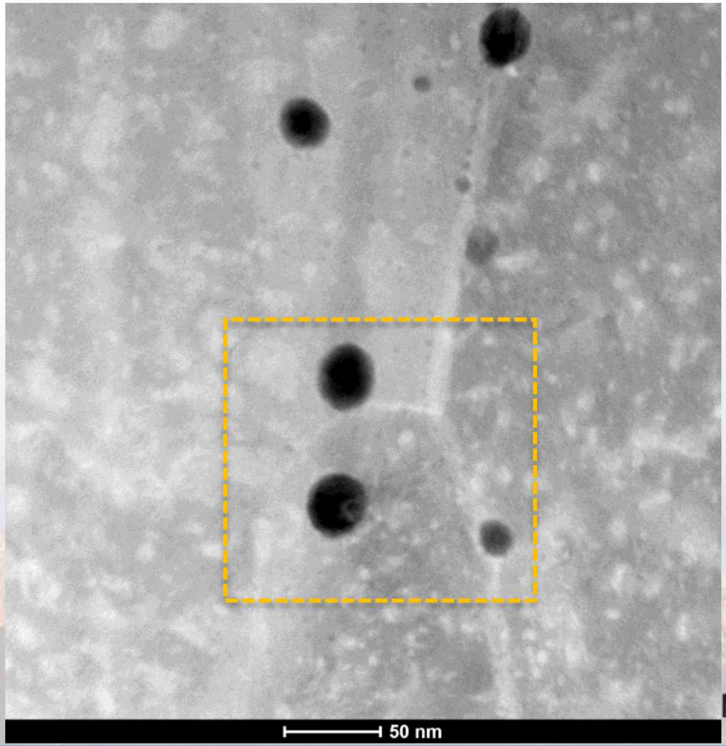
*Box is representative STEM-EDS line scan from map

Sandia National Laboratories

Pt-Au Irradiation Region

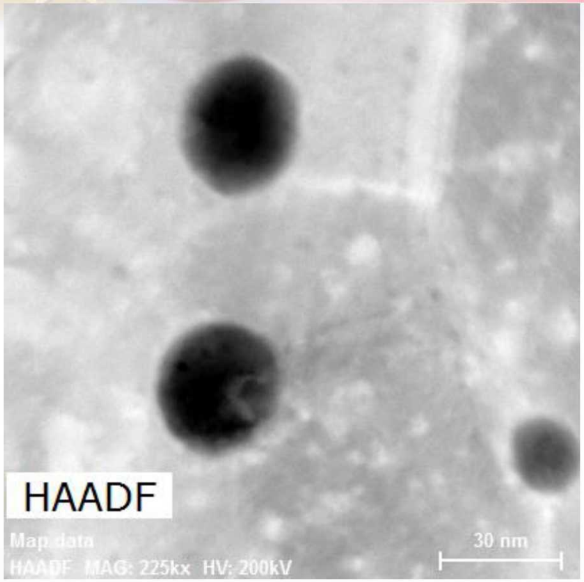
- **STEM-EDS examined in at ~ 1µm cross-section → ~ 90 dpa**
- **Voids and/or bubbles (from deposition) decorate GBs**
  - Some appear to be at previous location of GBs (boundary migration)
  - RT voids from radiation unlikely → below vacancy migration temperature → cavities are likely exclusively from deposition gases)
- **No observed Au segregation and grain size ≈ 395 nm**

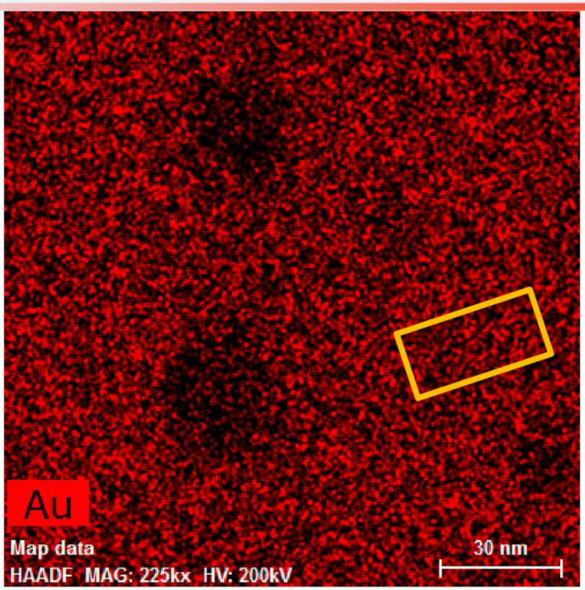


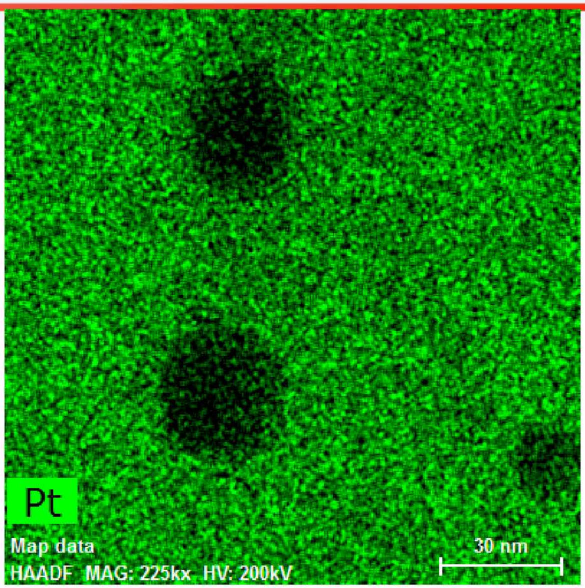


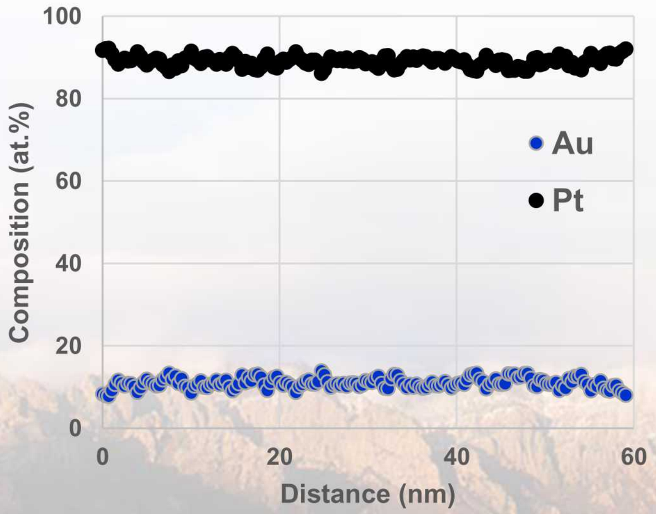
*Box is STEM-EDS area

Pt-Au Irradiation Region









*Box is representative STEM-EDS line scan from map

Lack of radiation tolerance: ion beam mixing?

- Grain growth (50 nm to 395 nm) in the irradiation zone
- Why hasn't the Au at the GB (confirmed prior to irradiation) stopped GB migration during irradiation?
- Effect of ion beam mixing during RT irradiation
  - Immiscible alloy system → possible that this induced full mixing (Au at GB prior to irradiation to uniform distribution after irradiation)
  - Grains do not grow during prolonged 500C thermal anneal