



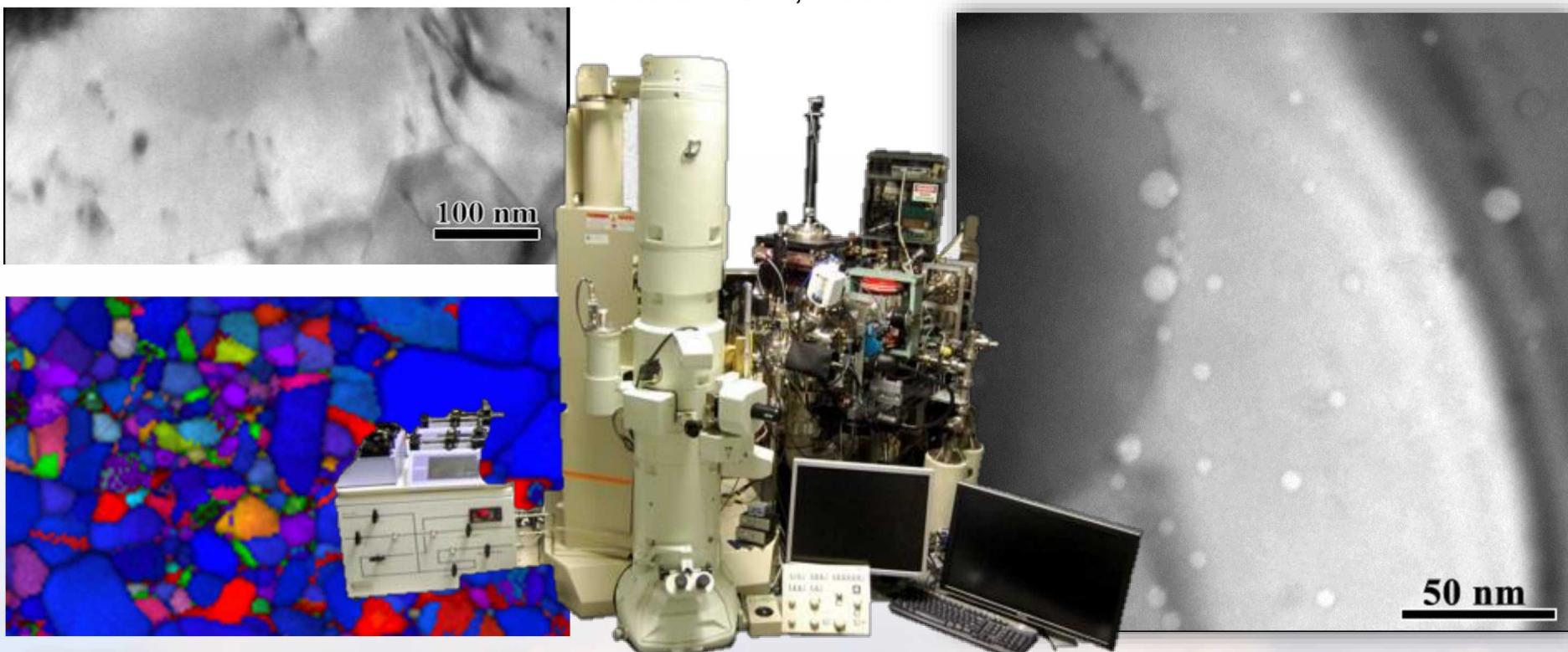
This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

Towards Controlling Grain Boundary Stability through Ion Beam Modification

SAND2018-12048C
NSLF
Nuclear Science User Facilities

K. Hattar, C. Barr, S. Briggs, B. Muntifering, D.C. Bufford, C. Taylor, N. Li, and A. Haque
Sandia National Laboratories, Los Alamos National Lab, Penn State University

October 17th, 2018



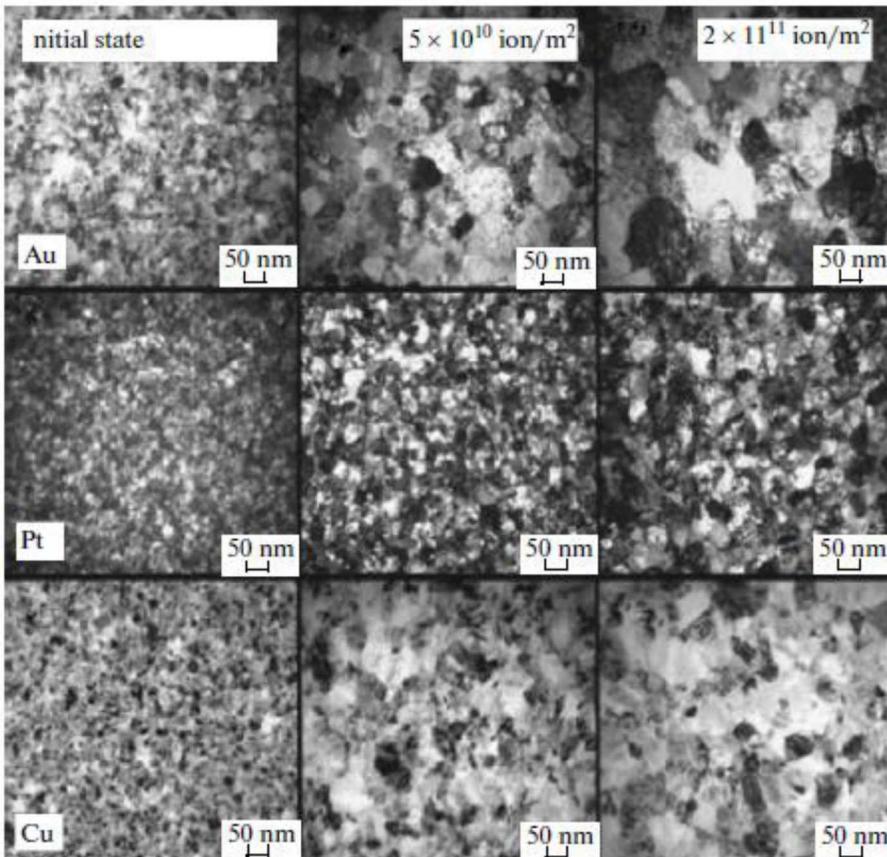
Collaborators:

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

This work was partially funded by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. 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. 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.

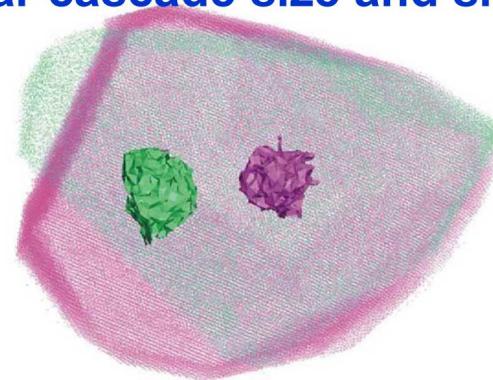
Section Summary: Radiation Tolerance from Nanostructured Metals

Variation in radiation tolerances



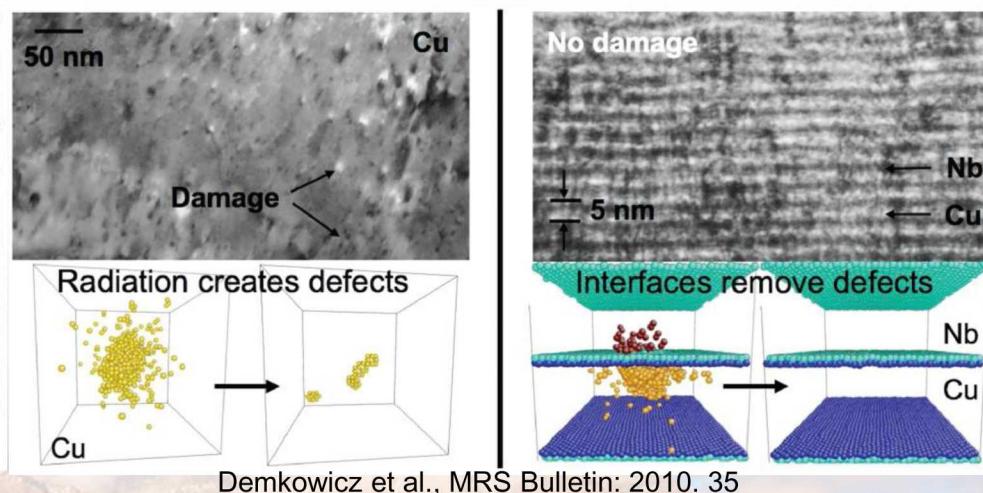
Kaomi et al., JAP: 2008. 104 073525

Similar cascade size and shape predicted



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

Nanolamellars are radiation tolerant



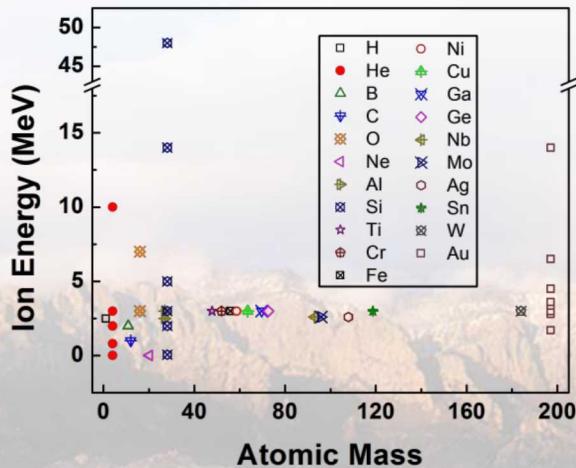
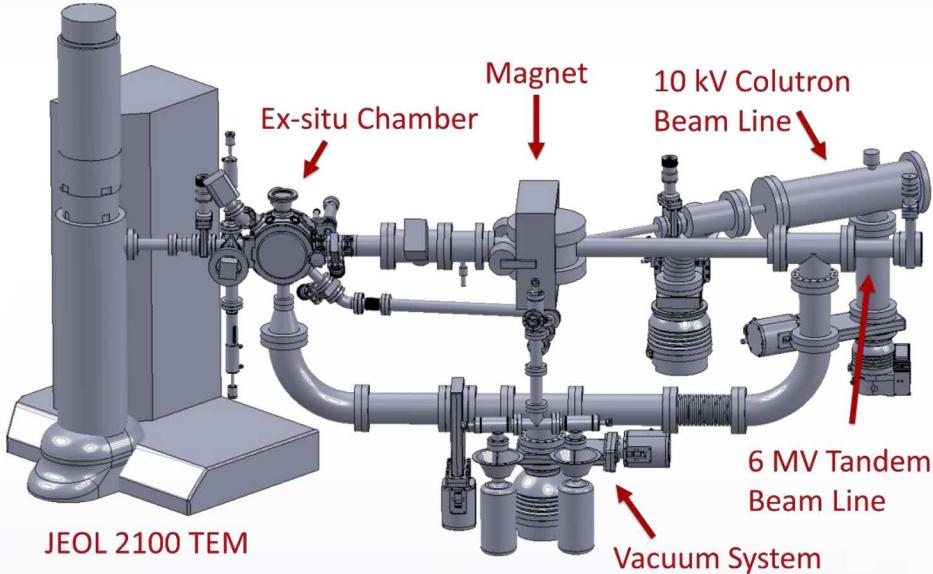
Demkowicz et al., MRS Bulletin: 2010. 35

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

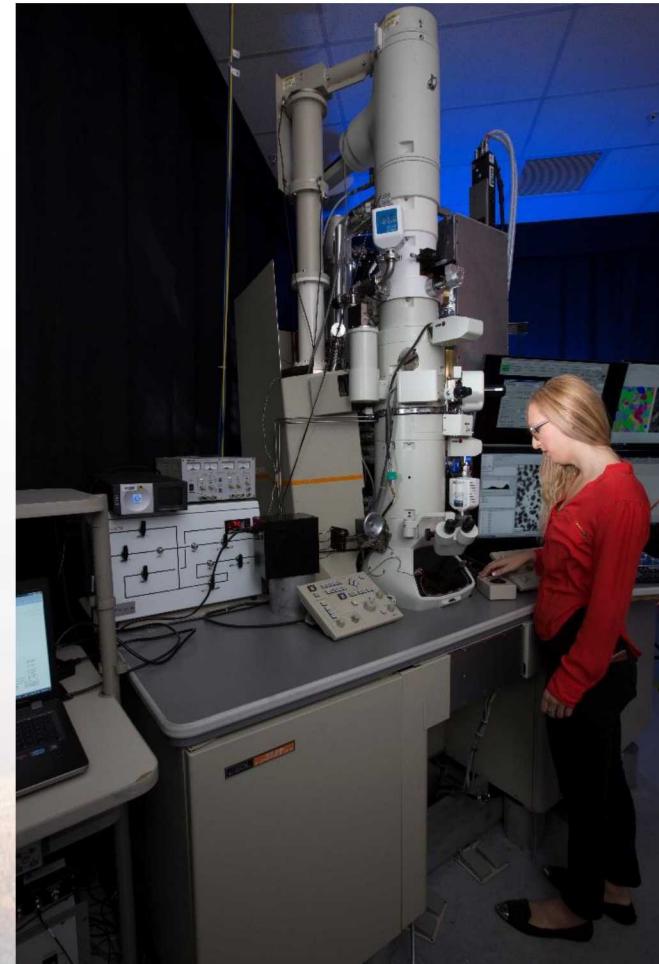
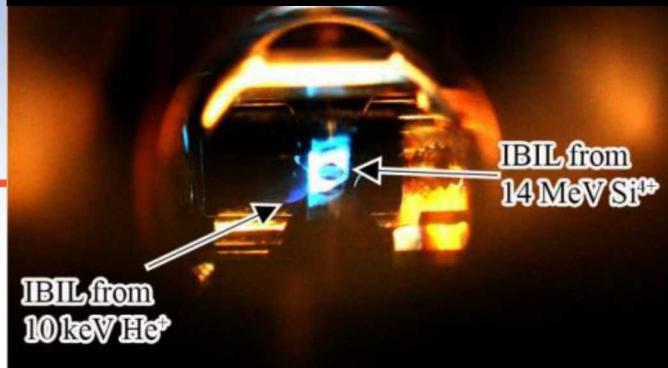
Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

Collaborator: D.L. Buller

10 kV Colutron - 200 kV TEM - 6 MV Tandem

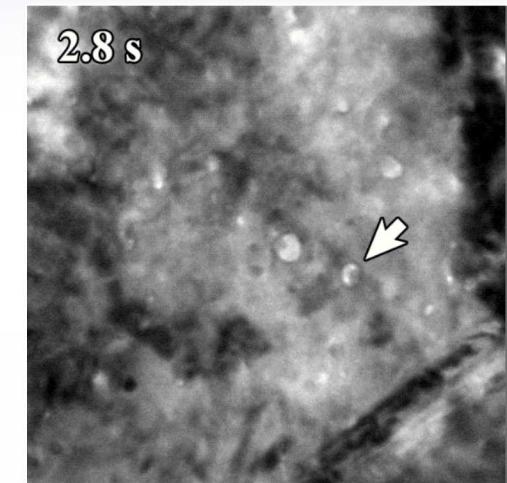
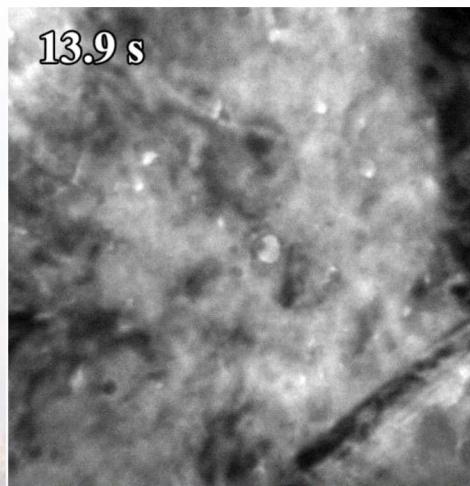
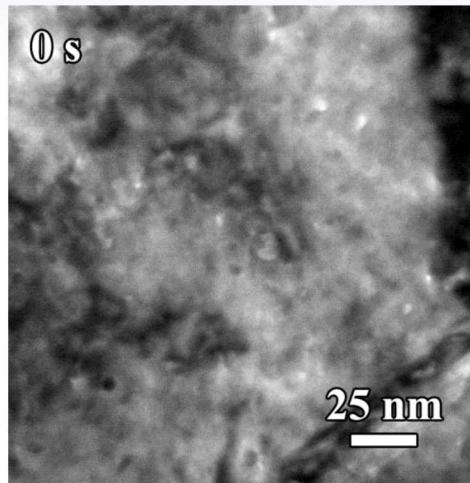
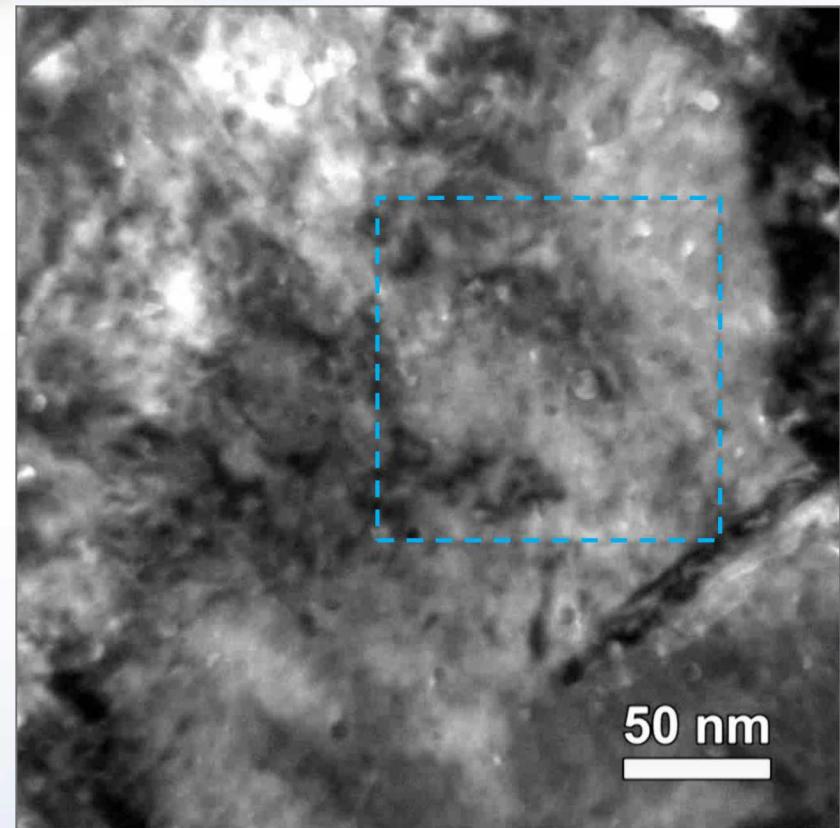


IBIL from a quartz stage inside the TEM



Simultaneous *In situ* TEM Triple Beam: 2.8 MeV Au⁴⁺ + 10 keV He⁺/D₂⁺

Collaborator: D.C. Bufford



In situ triple beam He,
D₂, and Au beam
irradiation is possible in
the Sandia's I³TEM!

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

Speed
x1.5

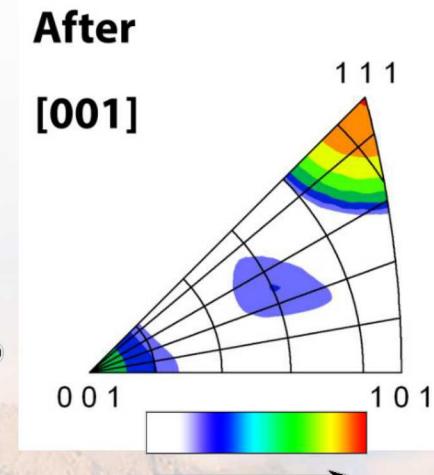
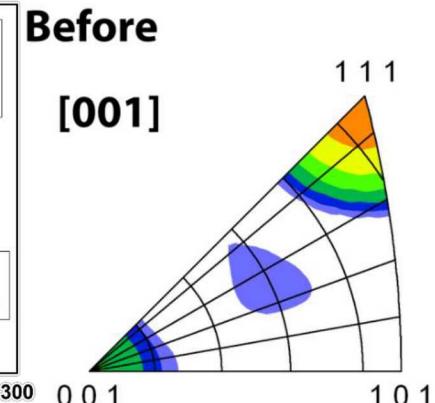
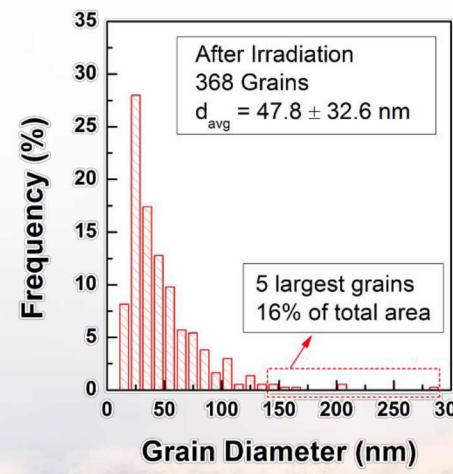
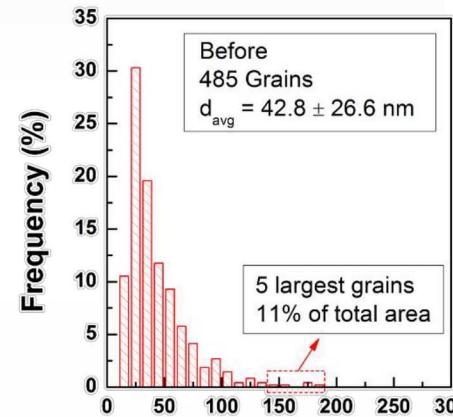
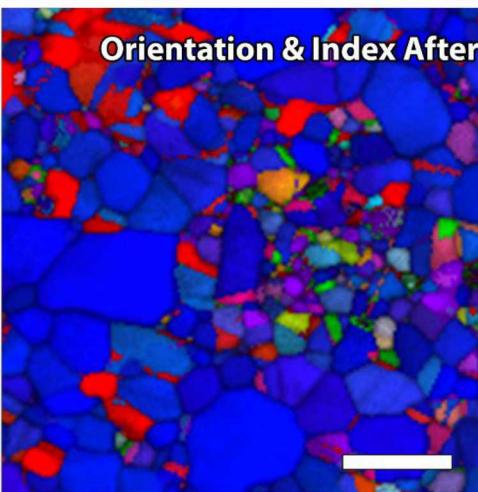
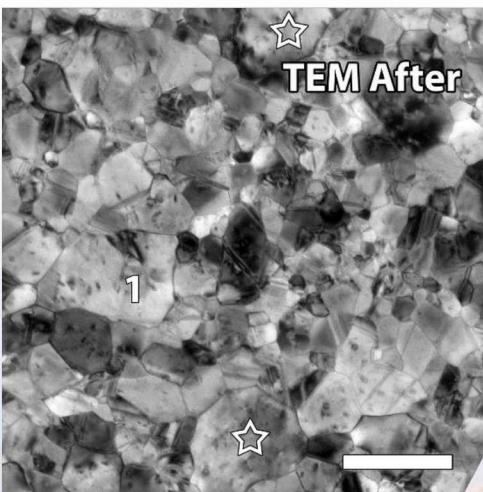
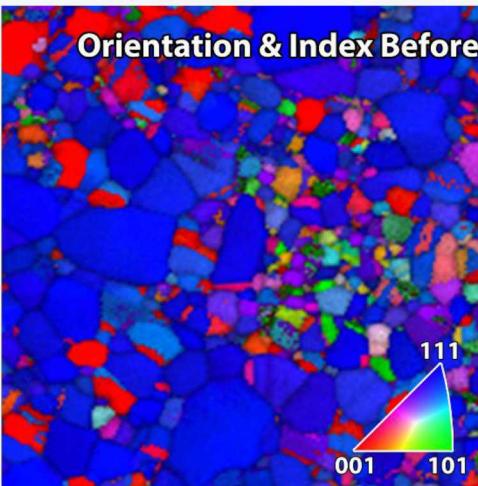
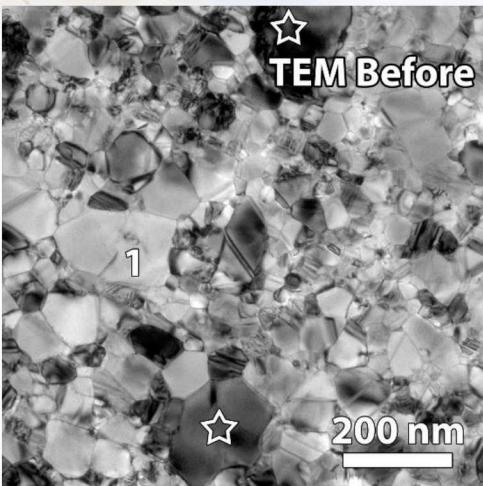
- Approximate fluence:
 - Au 1.2×10^{13} ions/cm²
 - He 1.3×10^{15} ions/cm²
 - D 2.2×10^{15} ions/cm²
- Cavity nucleation and disappearance



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Quantifying Grain Boundary Radiation Stability of Nanocrystalline Au

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



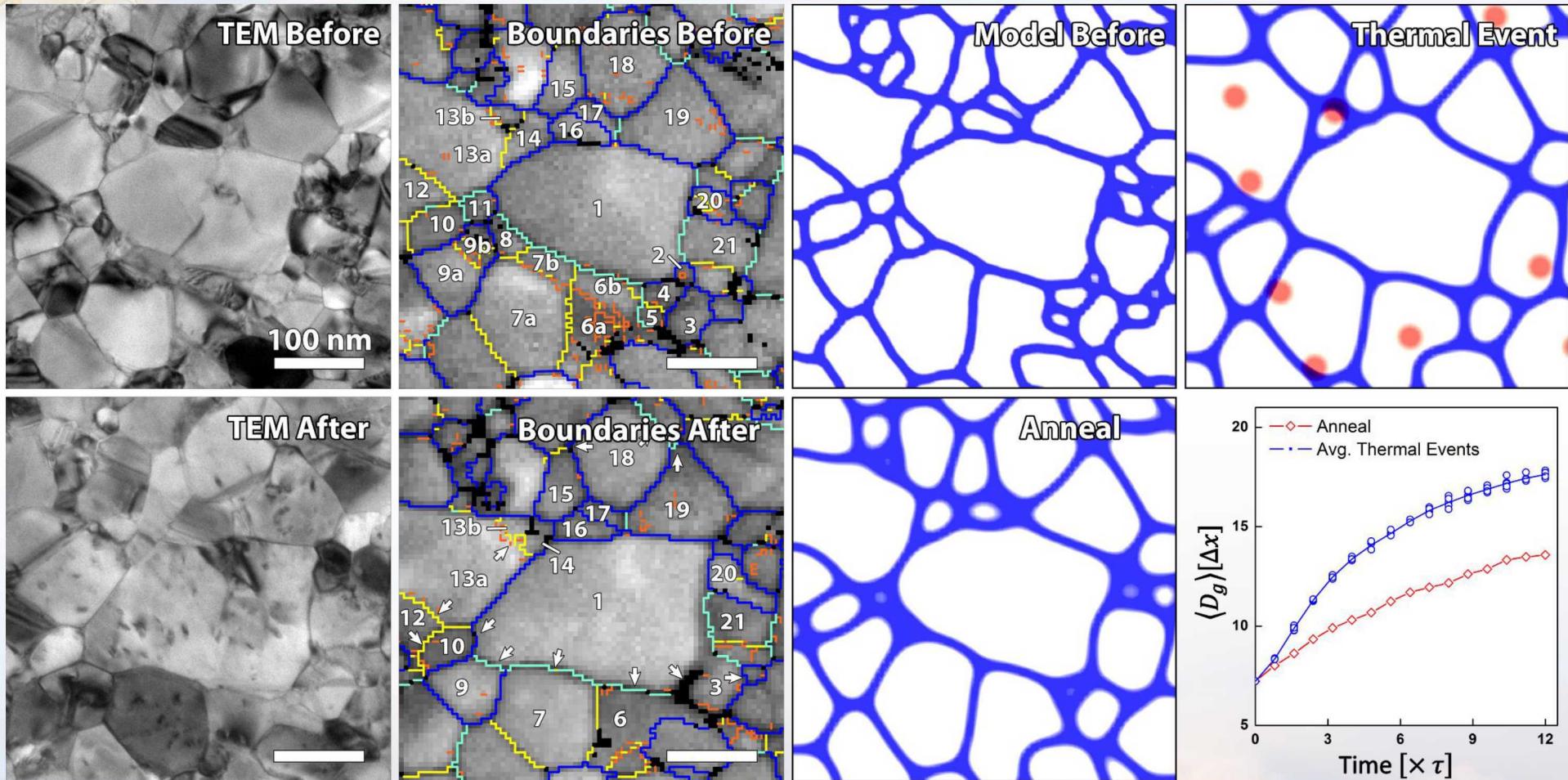
Any texture or grain boundary evolution can be directly observed and quantified



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Direct Comparison to Mesoscale Modeling

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



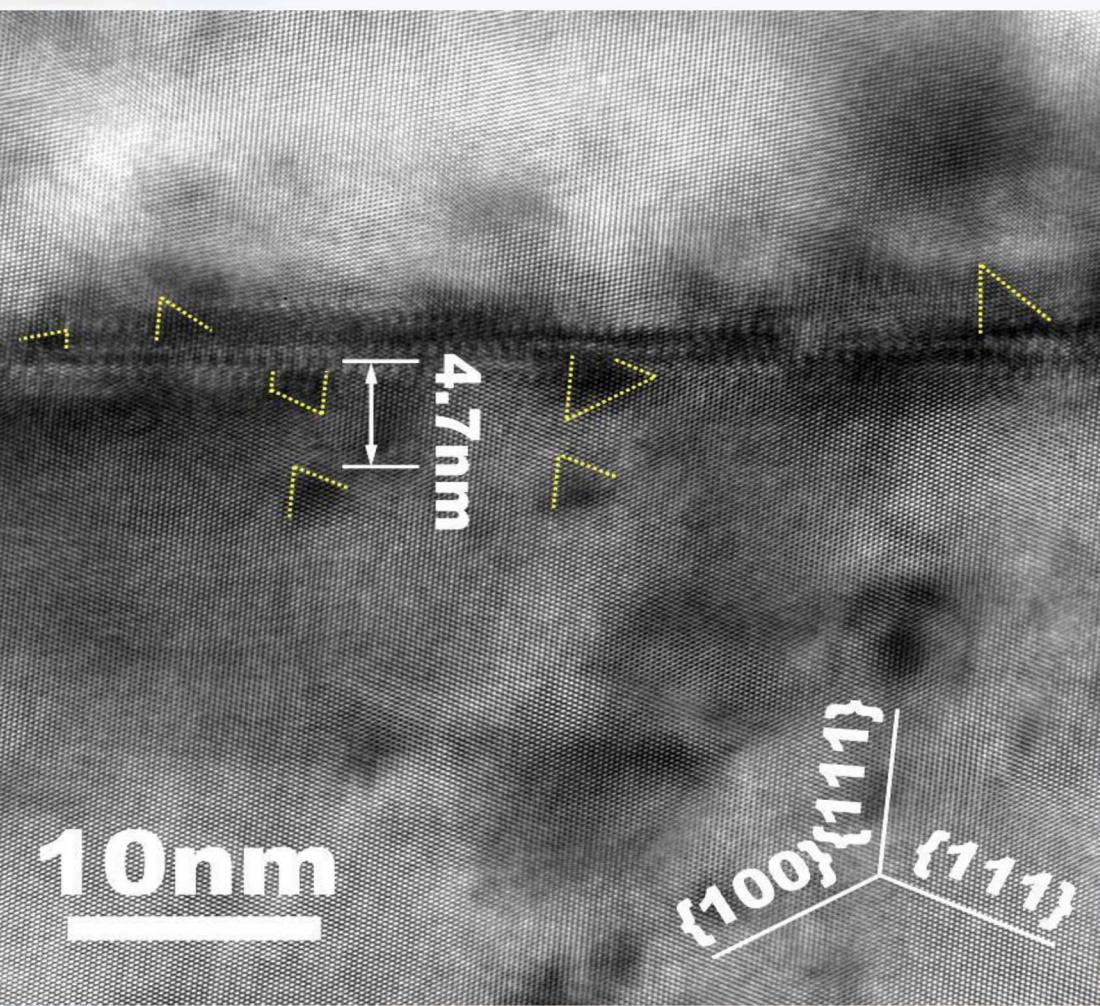
Because of the matching length scale, the initial microstructure can serve as direct input to either MD or mesoscale models & subsequent structural evolution can be directly compared.



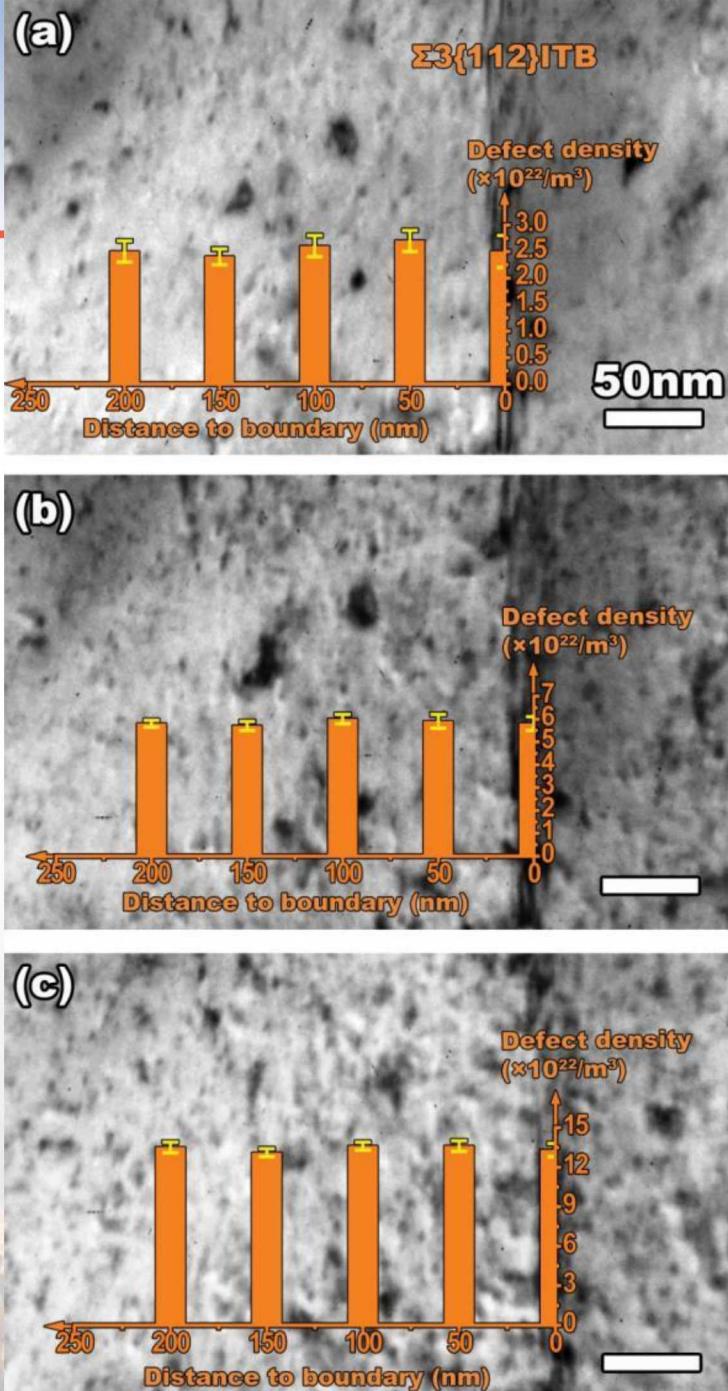
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Defects are Altered Little by the Presence of Grain Boundaries

Collaborators: N. Li & A. Misra



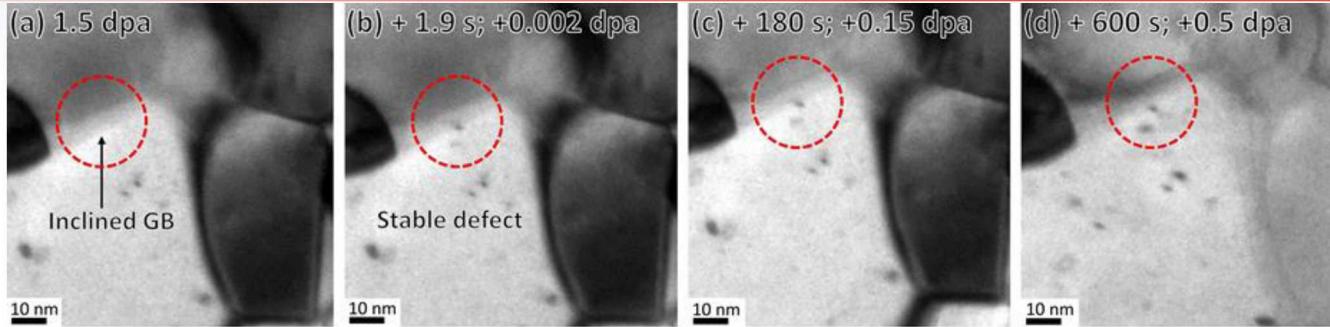
SFT appear to be directly at GB
No change in defect density is observed near GB



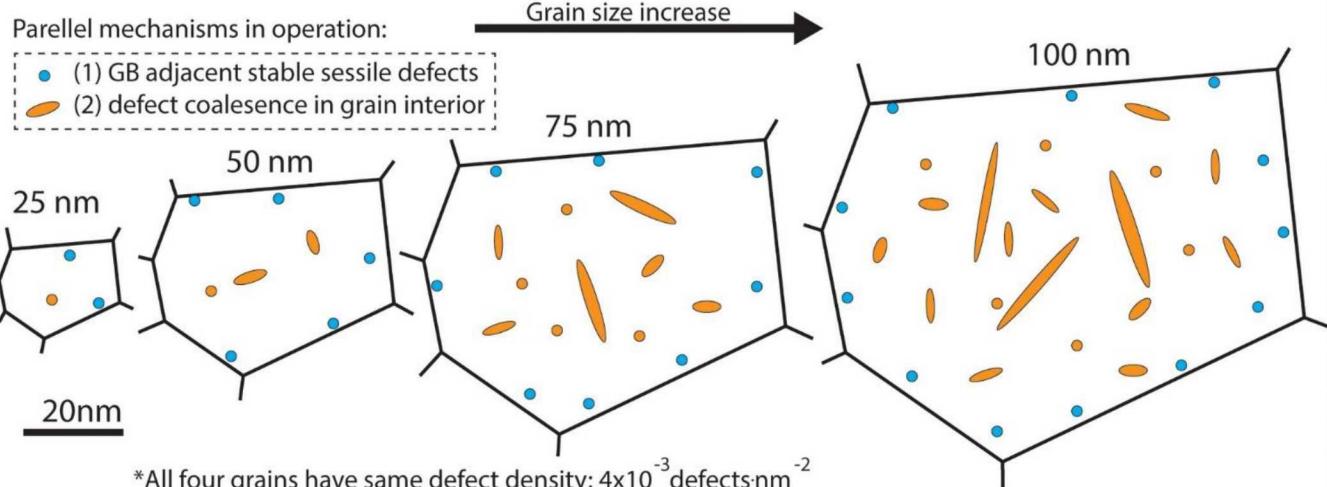
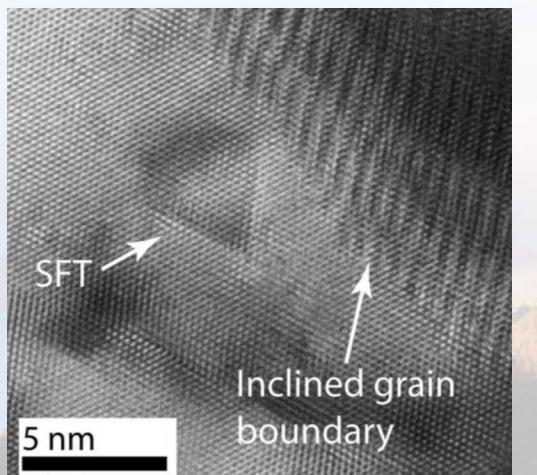
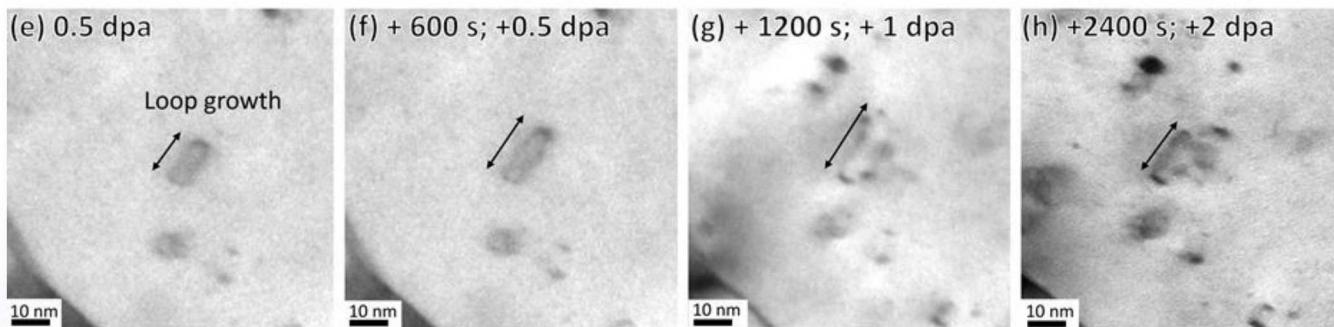
Breakdown of Increased Radiation Stability with Decreasing Grain Size

Collaborators: C.M. Barr

Defect evolution near GBs:



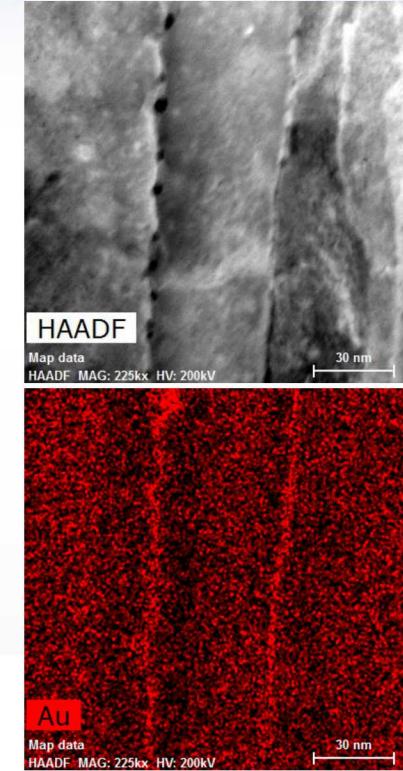
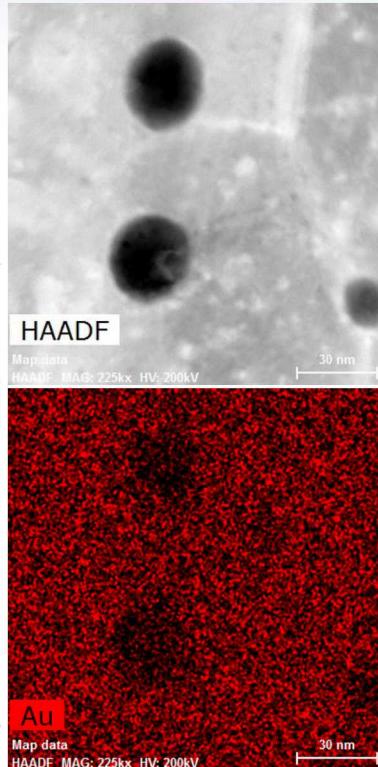
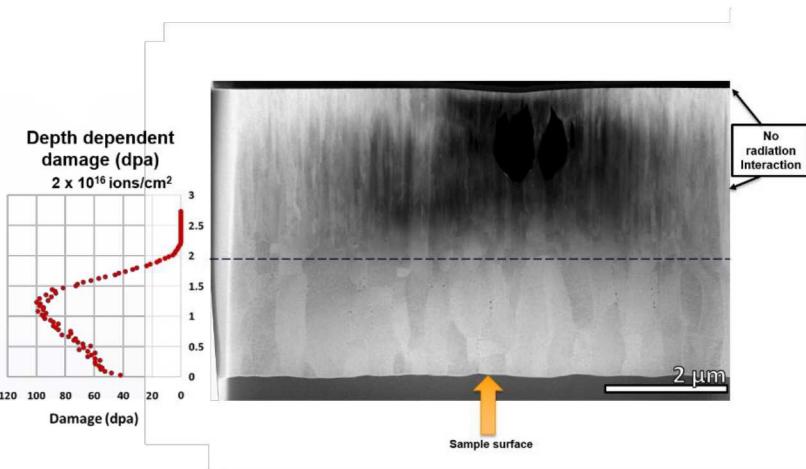
Defect evolution in grain interior:



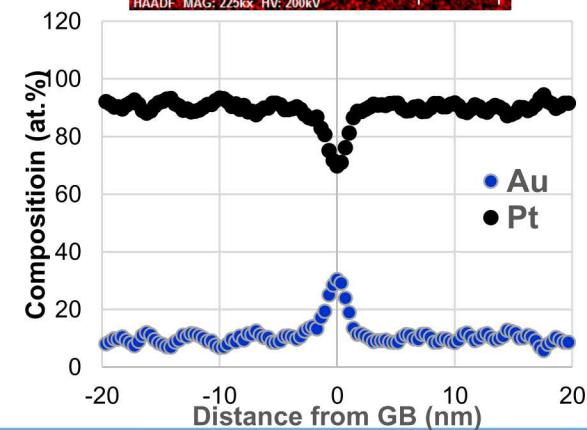
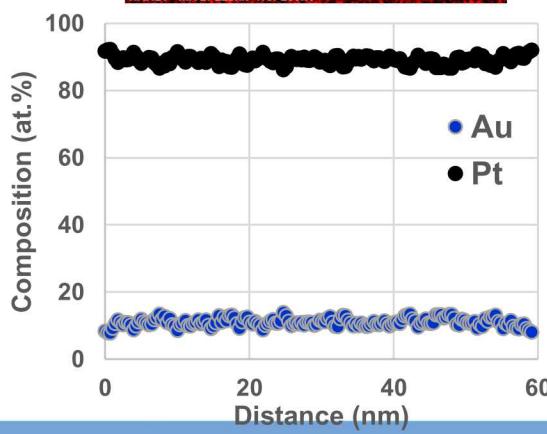
Exploring the Radiation Stability of Thermally Stable Nanocrystalline Metals

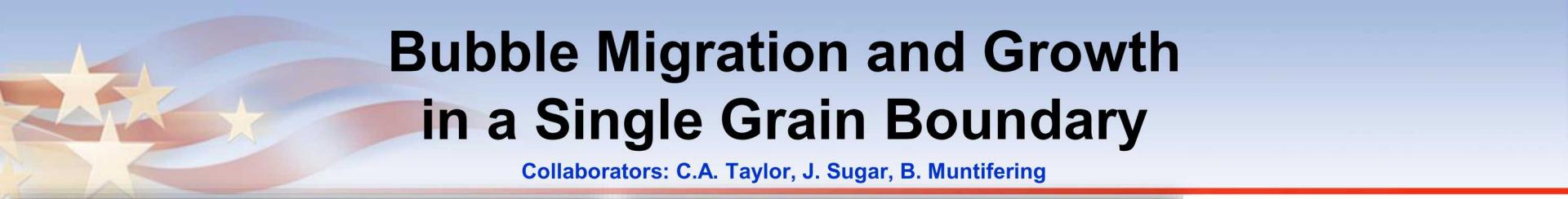
Collaborators: C.M. Barr, D. Adams

- Pre-Irradiation Heat Treatment:**
500° C/2hrs in vacuum furnace
- Irradiation conditions:** 20 MeV Au⁴⁺;
No external heating (RT plus beam heating)



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

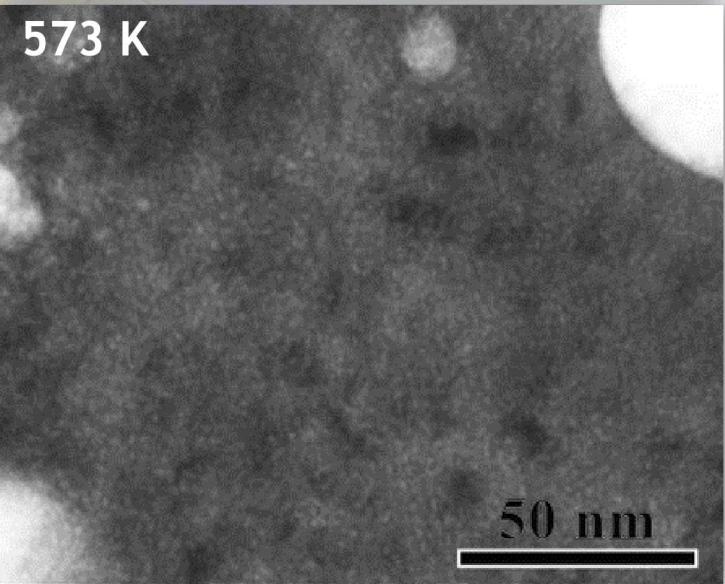




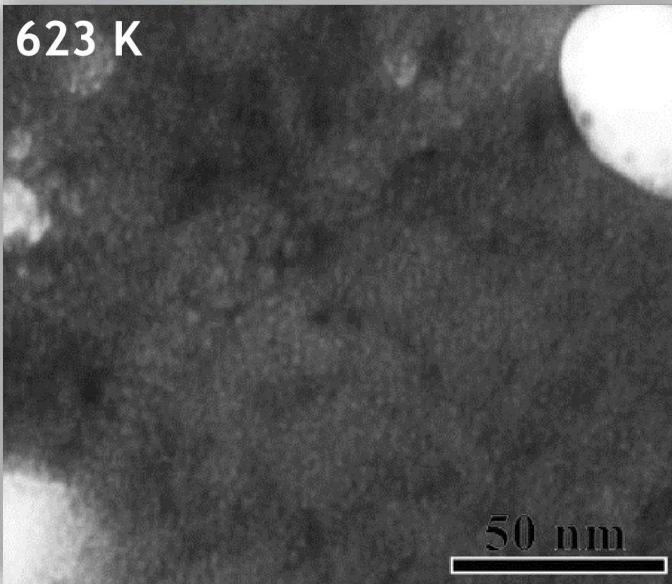
Bubble Migration and Growth in a Single Grain Boundary

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

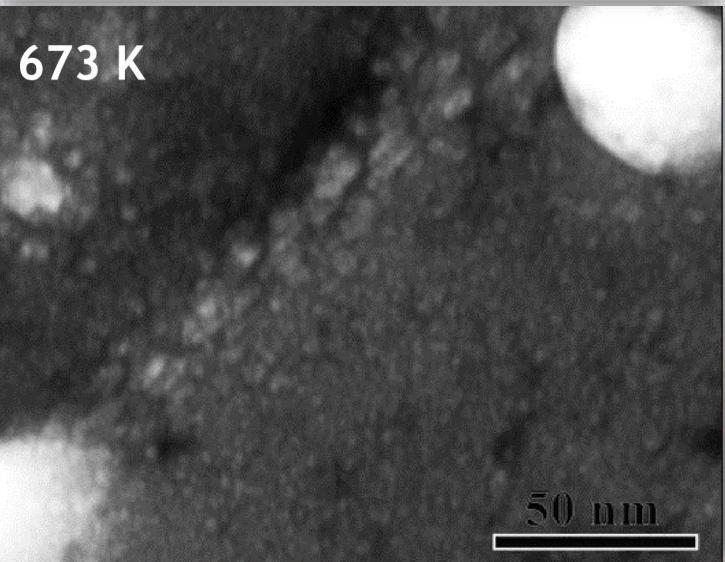
573 K



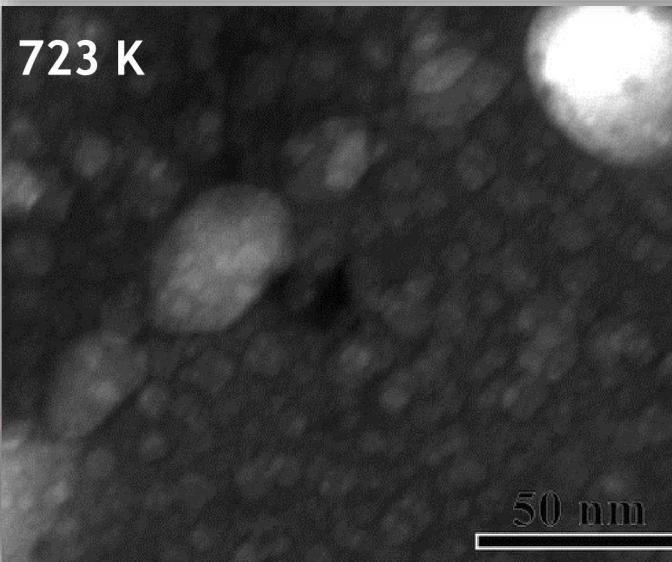
623 K



673 K



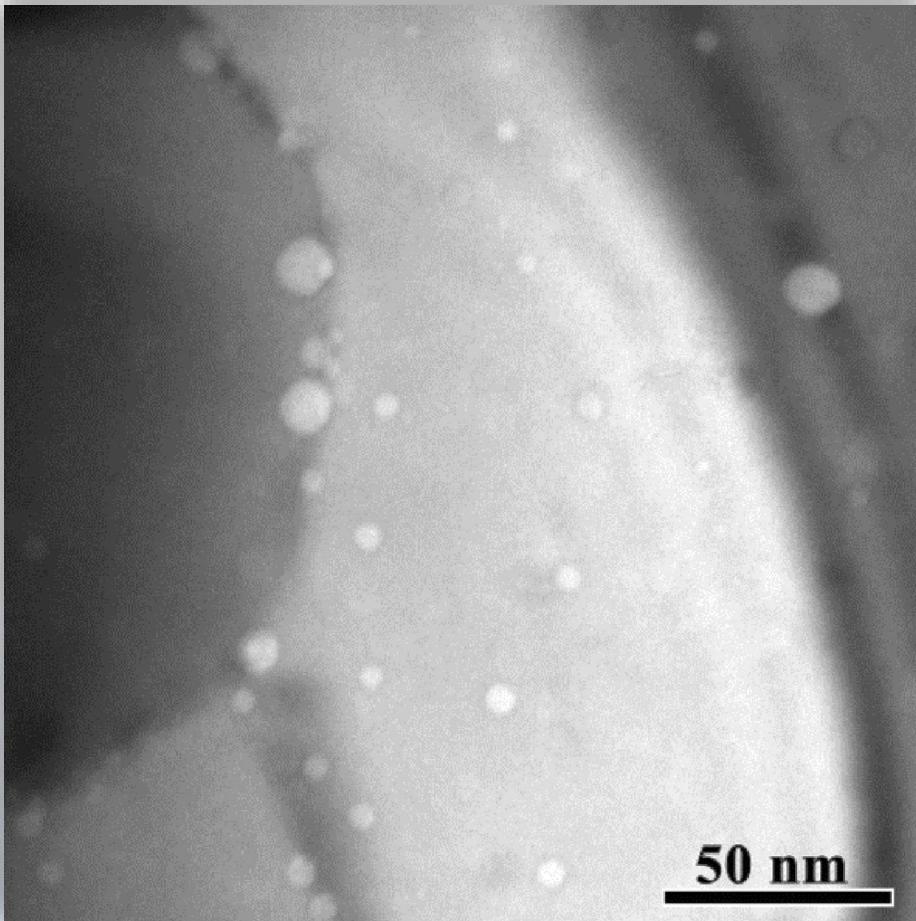
723 K



- 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

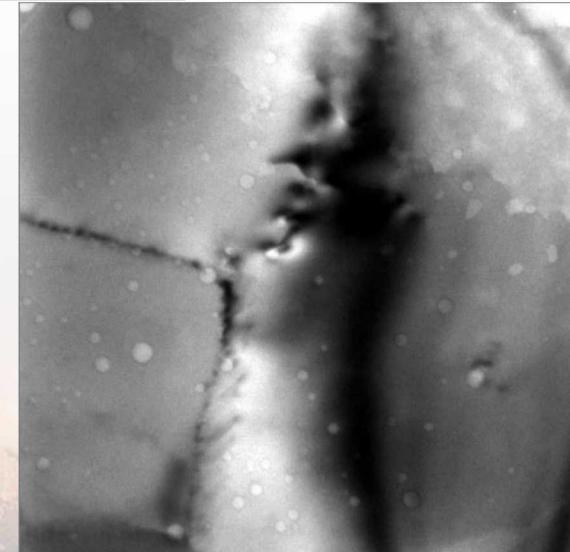
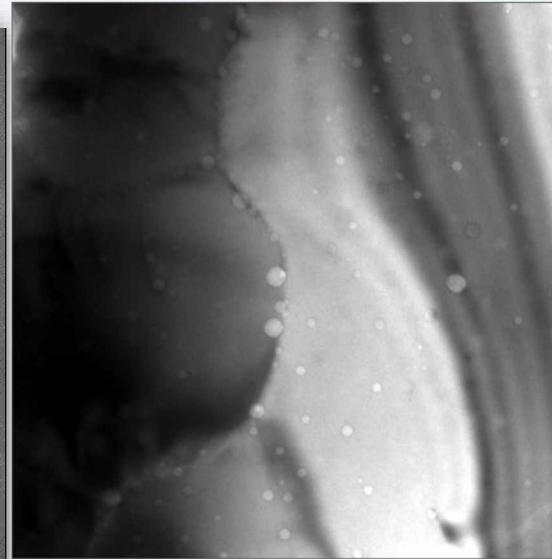
Collaborators: C.A. Taylor, J. Sugar, B. Muntifering



Cavities effect grain boundary mobility, triple junction angle



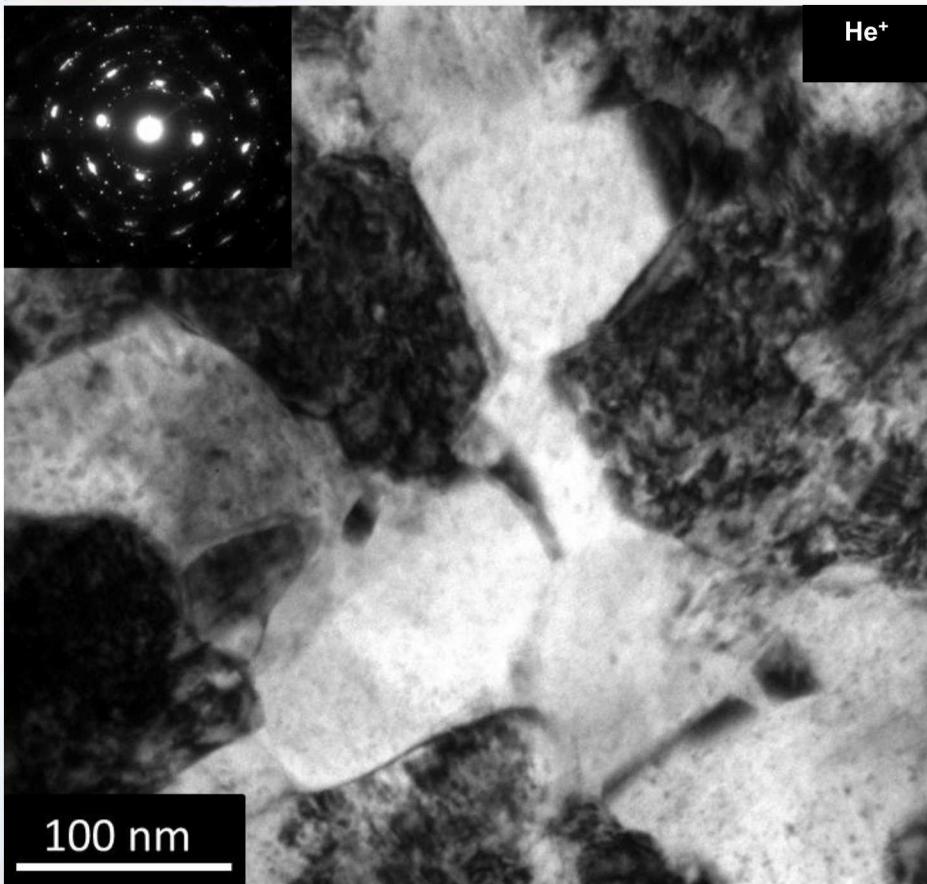
grain boundary motion alters cavity coalescence



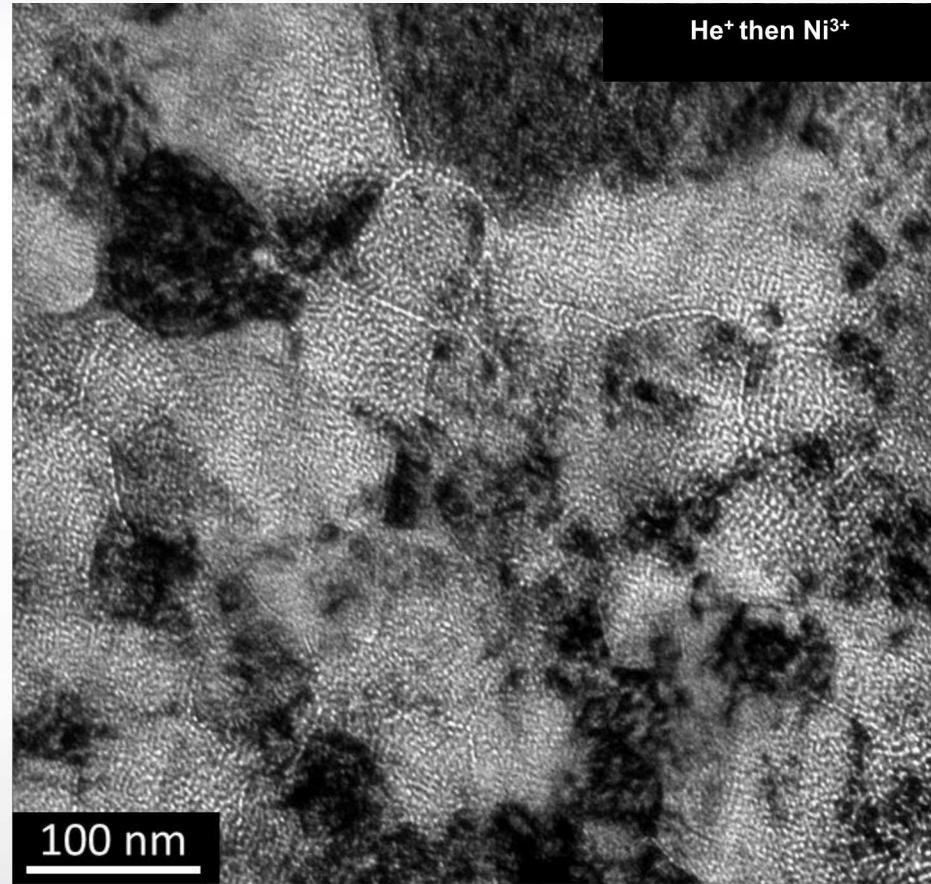
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Heterogeneous Bubble Formation under Some Radiation Sequences

Collaborator: B. Muntifering & J. Qu



$10^{17} \text{ He}^+/\text{cm}^2$
Visible damage to the sample



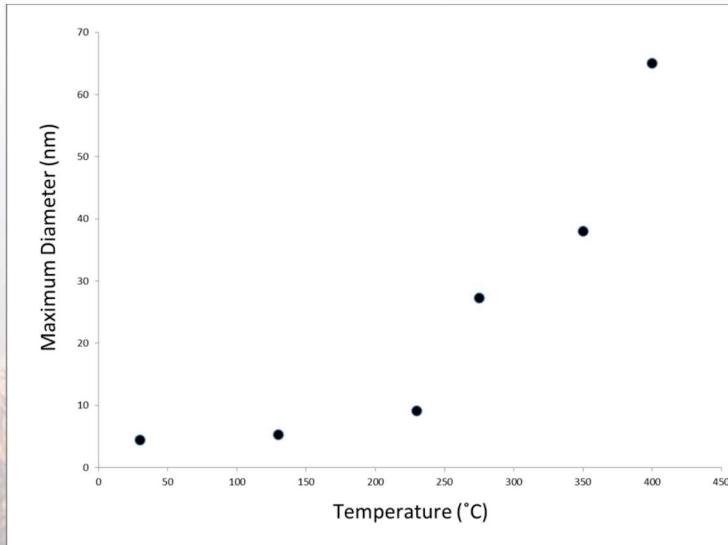
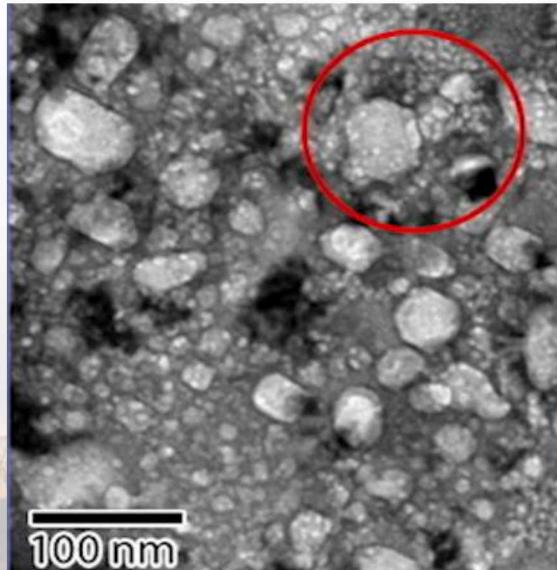
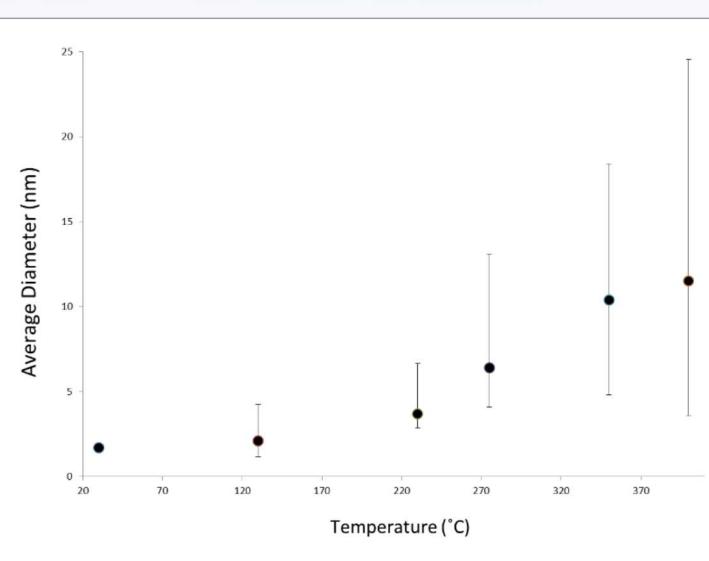
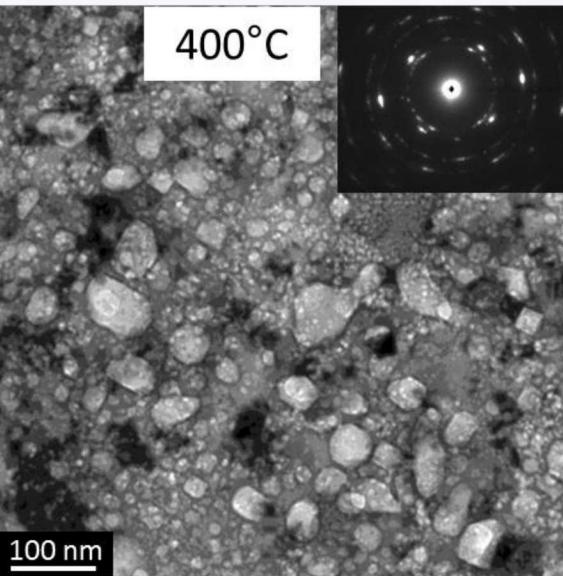
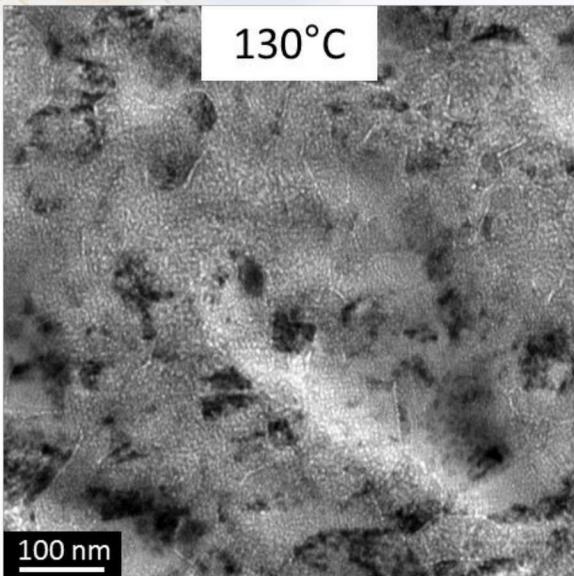
0.7 dpa Ni³⁺ irradiation
High concentration of cavities along grain boundaries



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Cavity Growth during *In situ* Annealing of 10 keV He⁺ Implanted and then 3 MeV Irradiated Ni³⁺

Collaborator: B. Muntifering & J. Qu



Bubble to cavity transition and cavity evolution can be directly studied

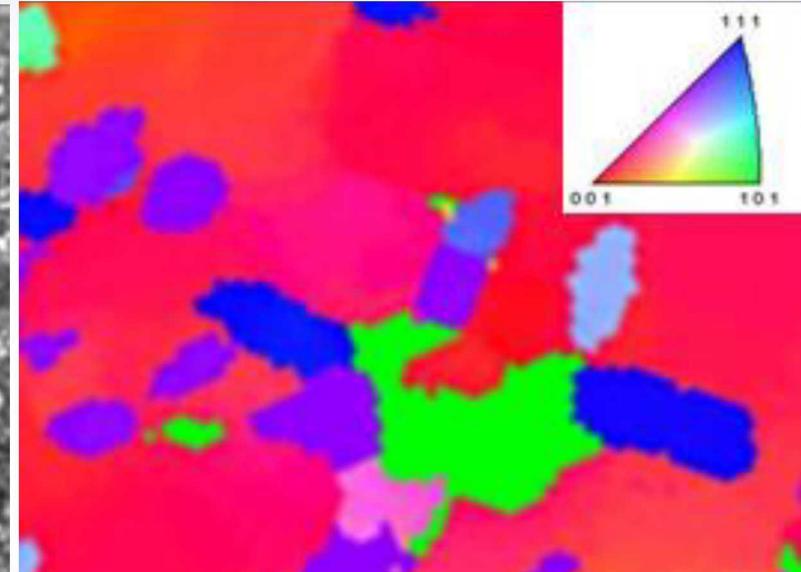
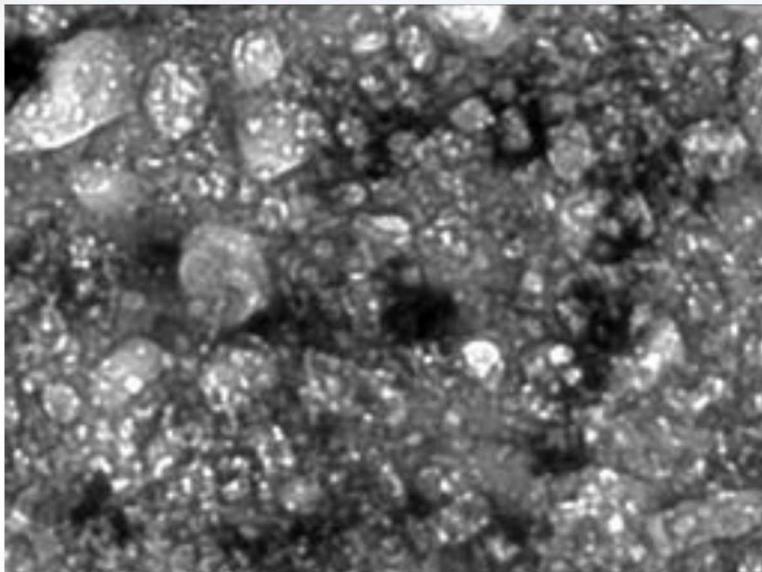




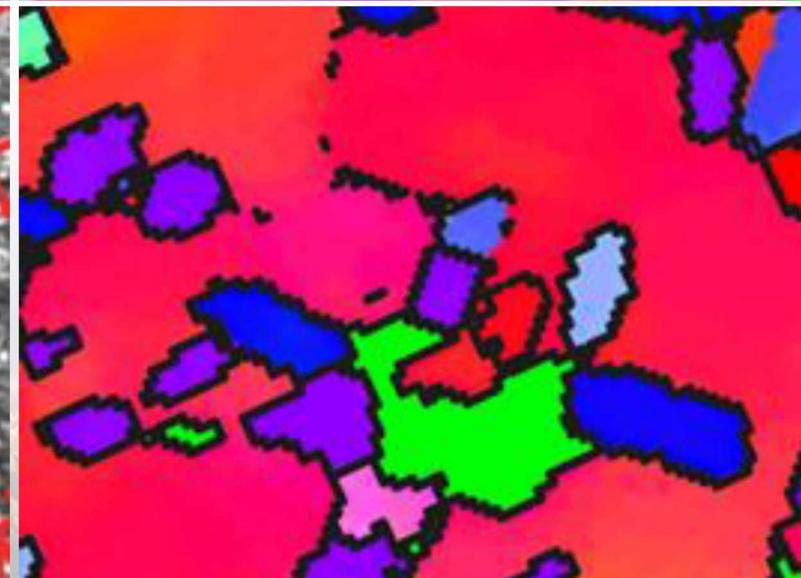
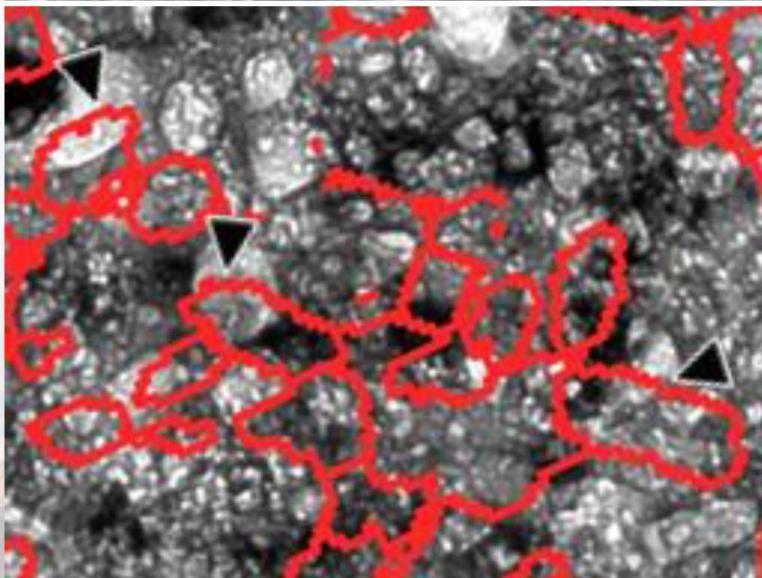
Precession Electron Diffraction Reveals Hidden Grain Structure

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



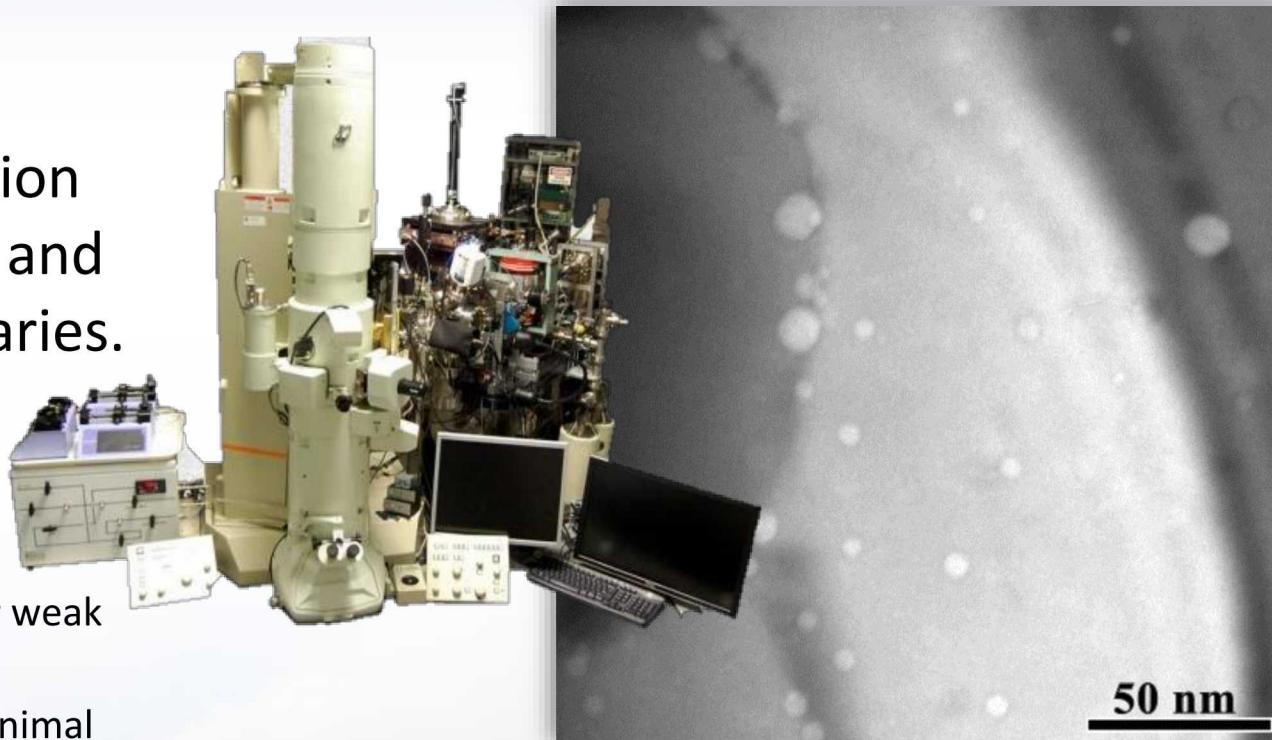
100 nm

Summary

- Sandia's I³TEM 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
- Might be tailorable



The response of grain boundaries to radiation damage is complex and depends on many variables.
There is a possibility to CONTROL them!

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



This work was partially funded by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. 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. 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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BACK UP SLIDES

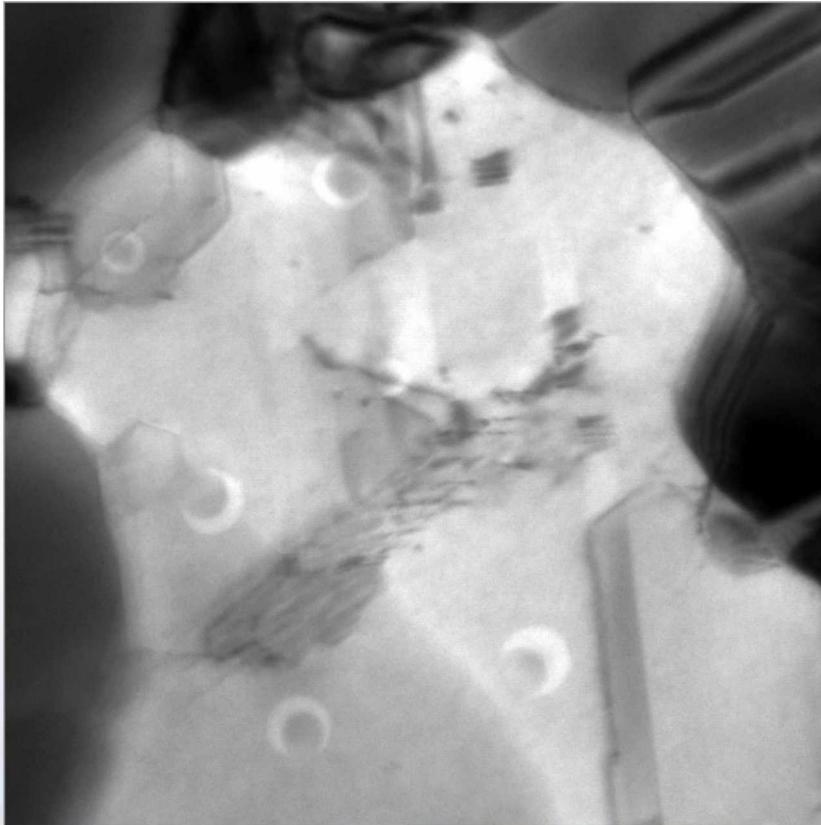


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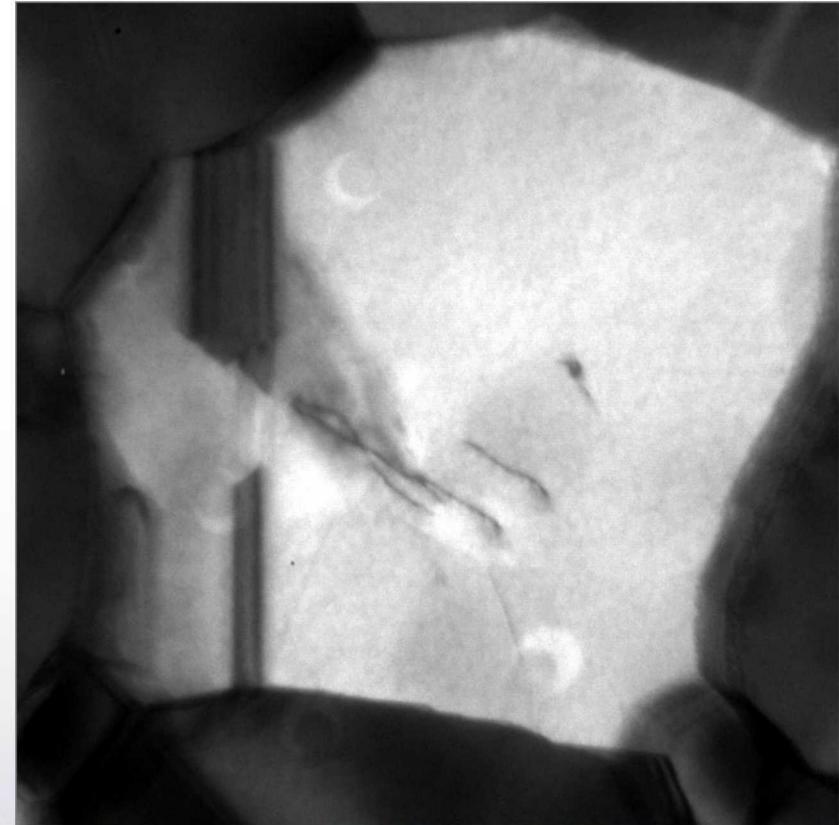
Dose Rate Effects in Nanocrystalline Metals

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

7.9×10^9 ions/cm²/s



6.7×10^7 ions/cm²/s



VS

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

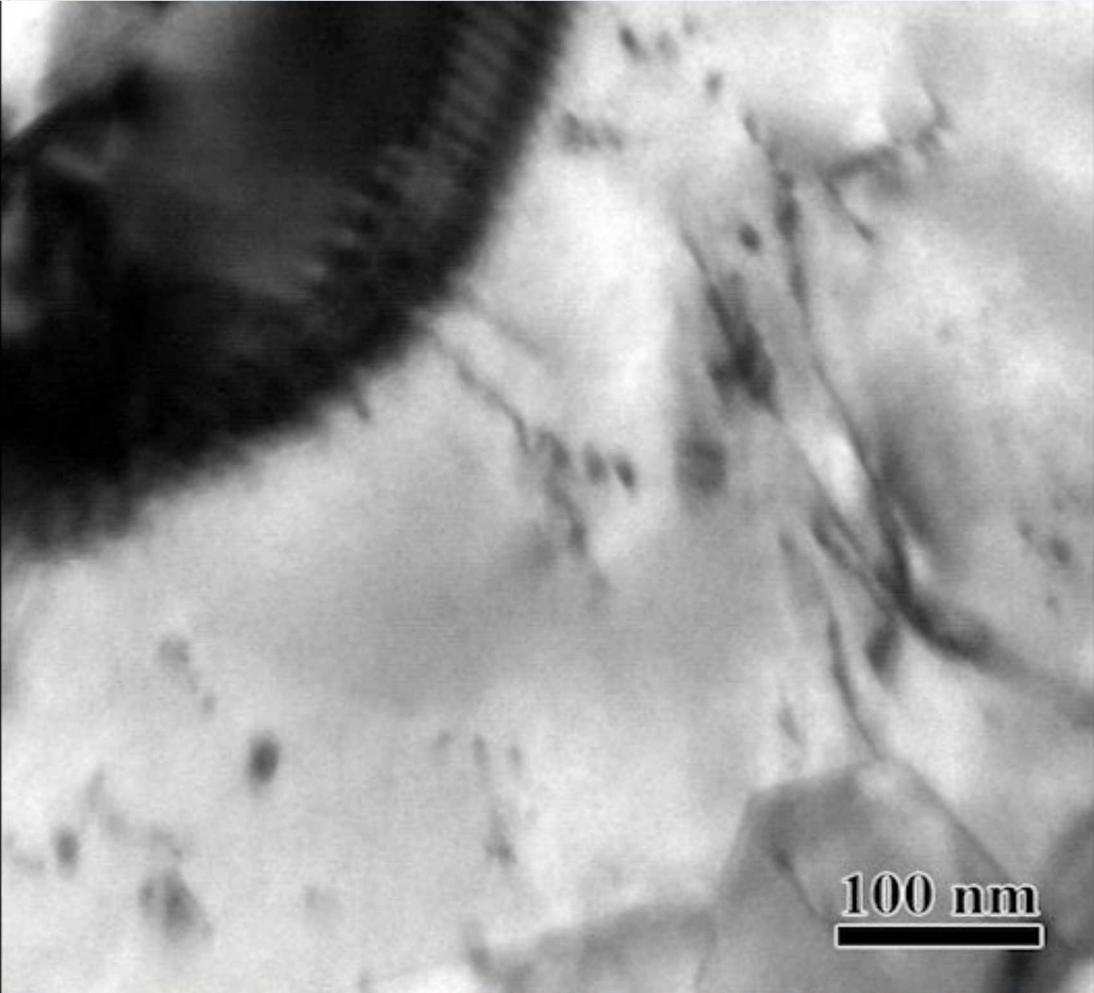


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1D Brownian Motion in Real Time

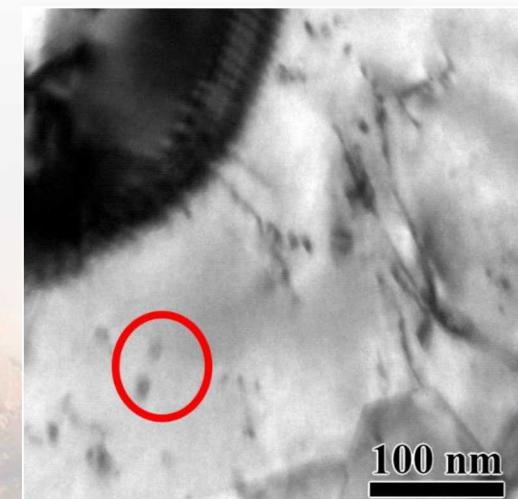
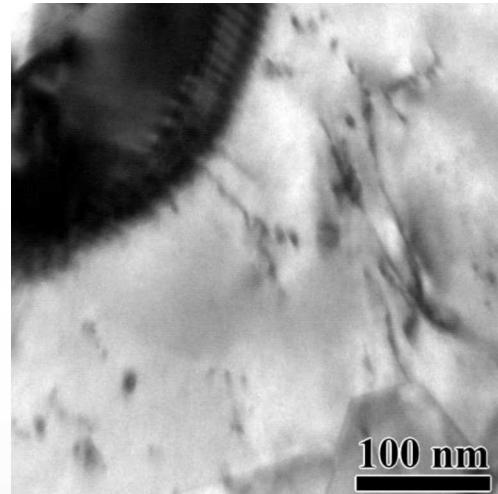
Collaborator: D.C. Bufford



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



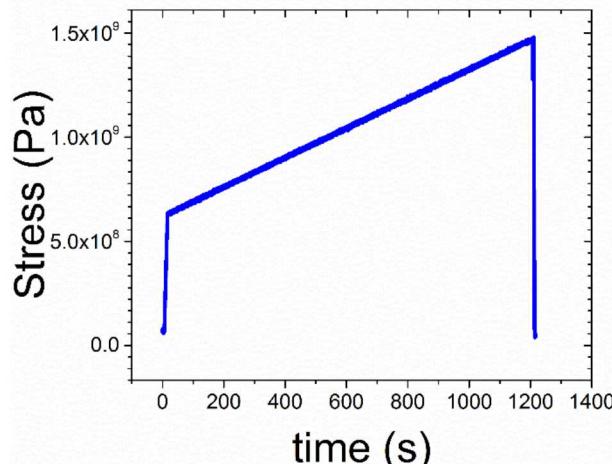
Triple beam condition:
2.8 MeV Au^{4+} + 10 keV He^+ / D_2^+



Irradiation Creep (4 MeV Cu³⁺ 10⁻² DPA/s)

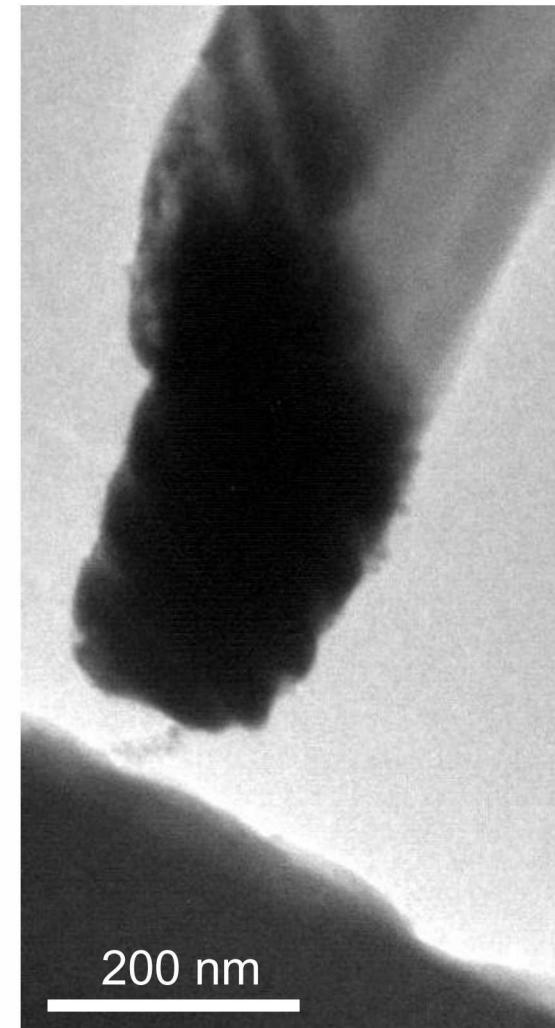
Contributors: G.S. Jawaharam, S. Dillon & R.S. Averback

Controlled Loading Rate Experiments

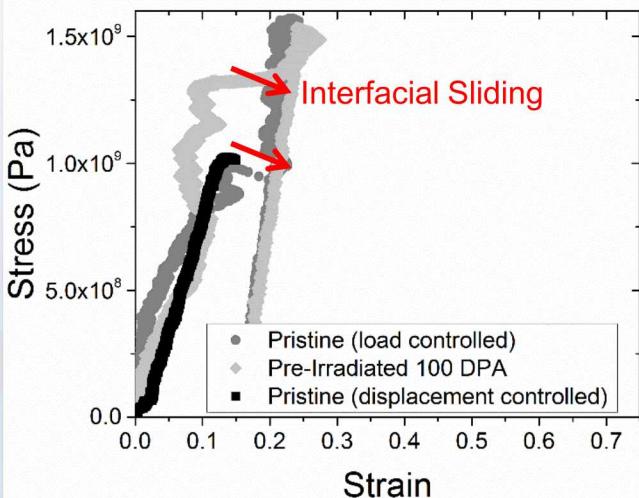


In-situ TEM
radiation
creep is
feasible!

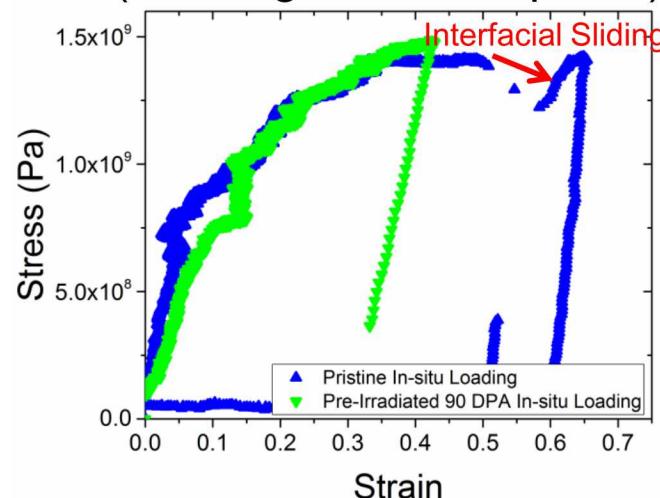
50 nm Cu-W multilayer
20 Min



No Irradiation
(Loading rate 0.6 Mpa s⁻¹)



Irradiation Creep
(Loading rate 0.6 Mpa s⁻¹)



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

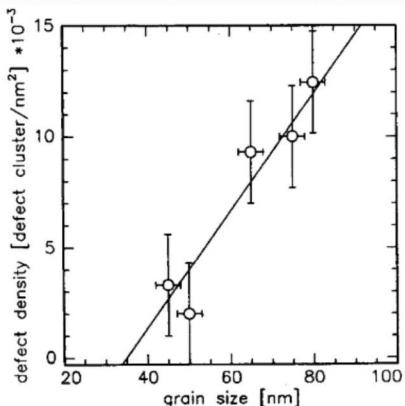
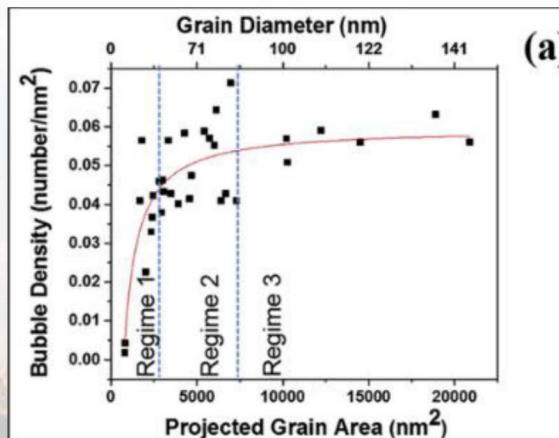


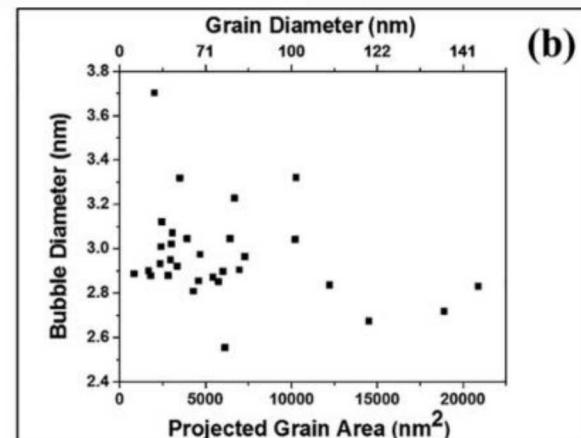
Fig. 8. Defect densities vs. grain size on the irradiated Pd sample (240 keV, 2×10^{16} Kr/cm²).

Rose et al. (1997)

NC Fe Helium bubble trends at 427° C



(a)



(b)

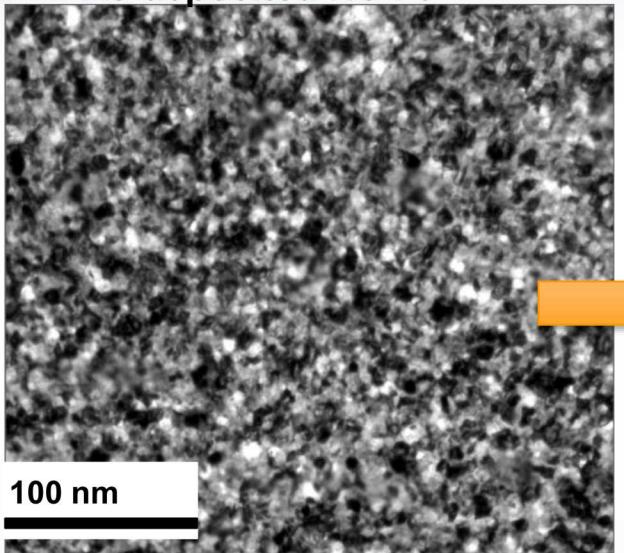
El-Atawani et al. 484 (2017) 236-244.



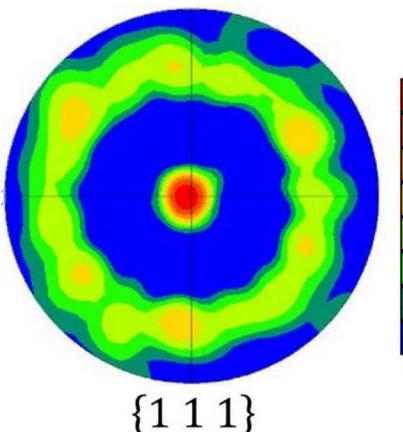
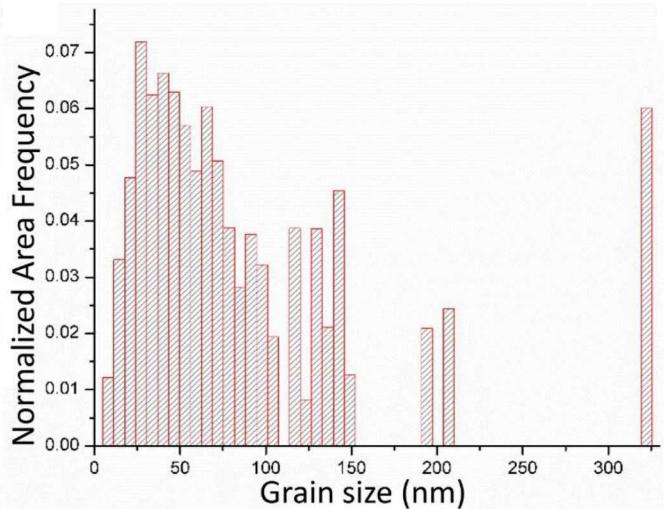
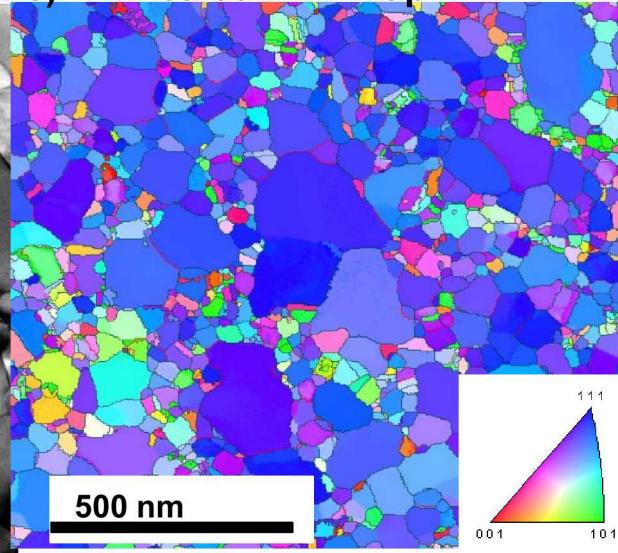
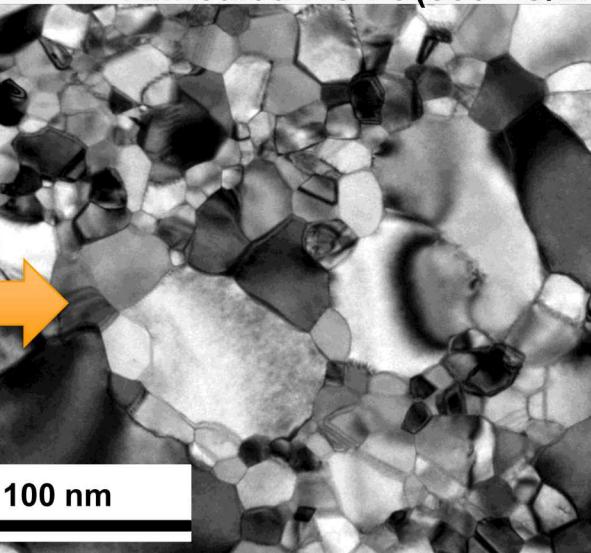
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NC Grain Size Effects: Methodology

As-deposited NC Pt



Annealed NC Pt (500 °C/2hrs) Annealed PED Map



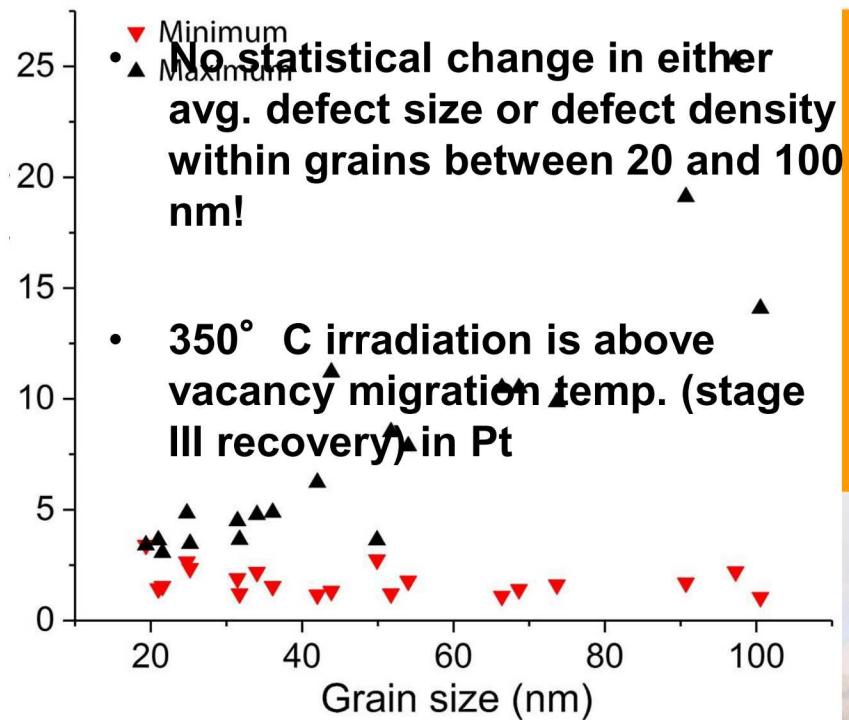
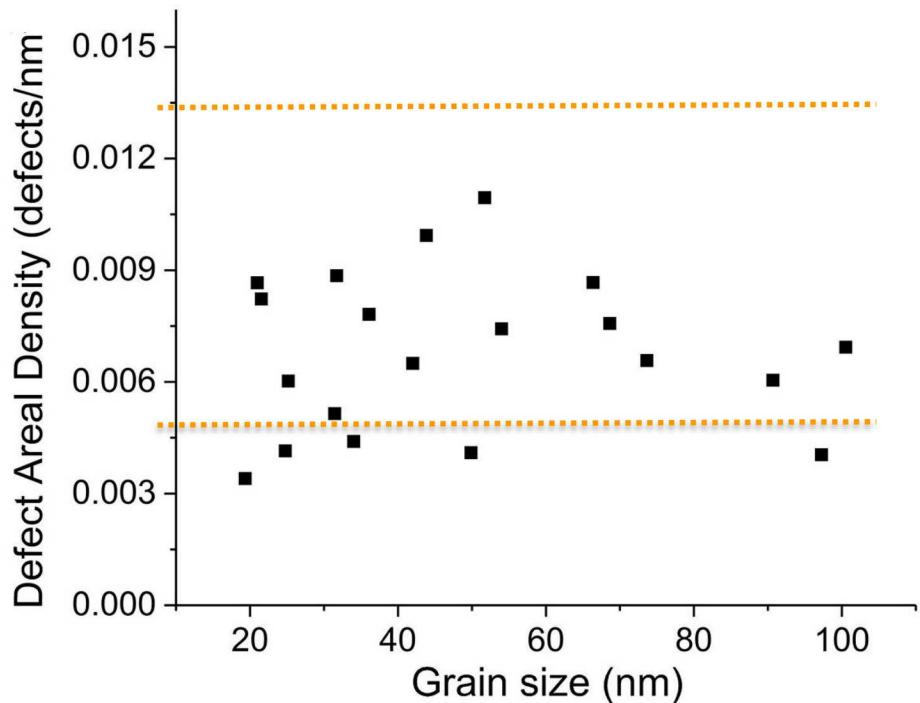
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NC Grain Size Effects : Irradiation and Defect Trends

- In-situ TEM irradiations: 2.8 MeV Au^{4+} at a flux of 4.7×10^{10} ions/cm²s to a fluence of 1.7×10^{14} ions/cm² at 350°C using a Gatan double tilt heating stage

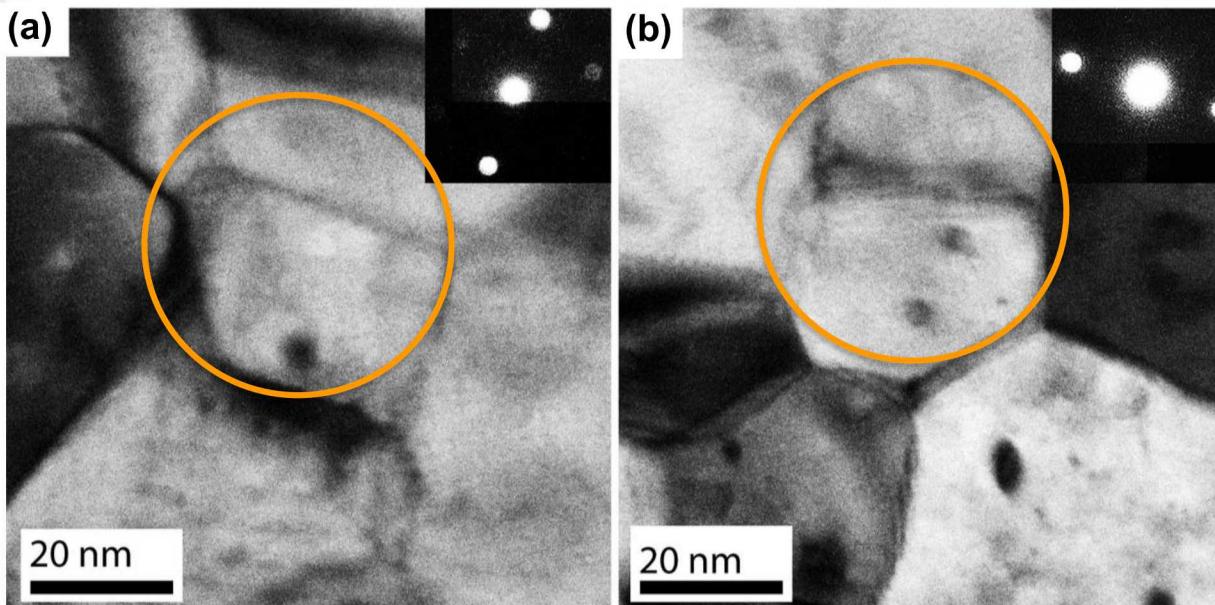
- Average final dpa ≈ 3 dpa

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/nm²
 - Grain B = 3 defects → 8.6×10^{-3} defects/nm²
- With decreasing grain size, # of defects become both more challenging and critical for any type of accurate defect density



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

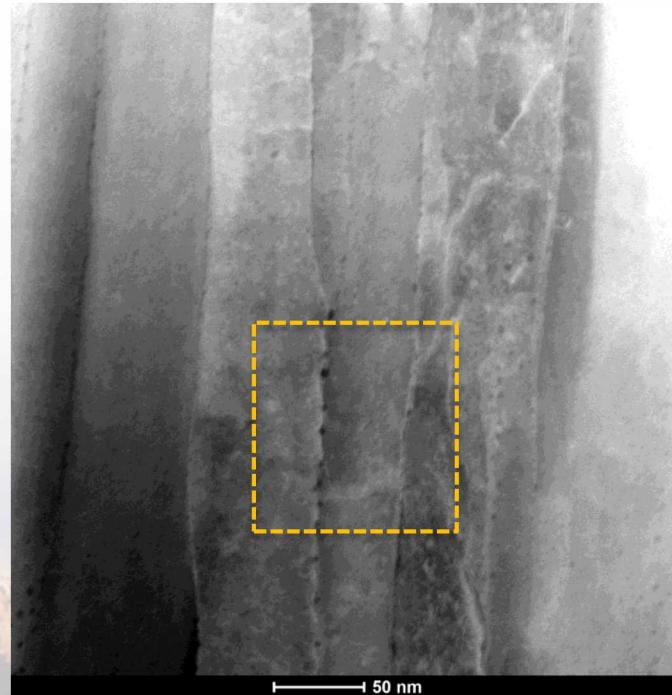
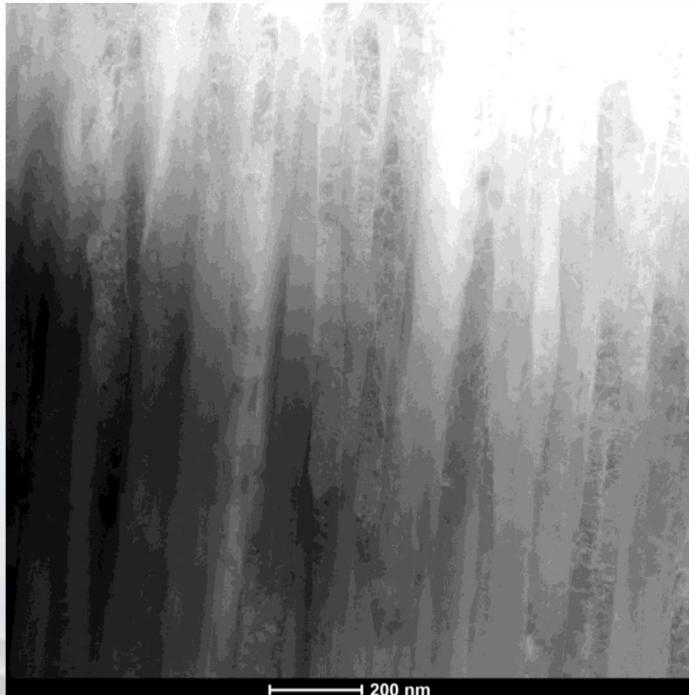
Christopher Barr and Khalid Hattar



Sandia National Laboratories

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 \approx 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

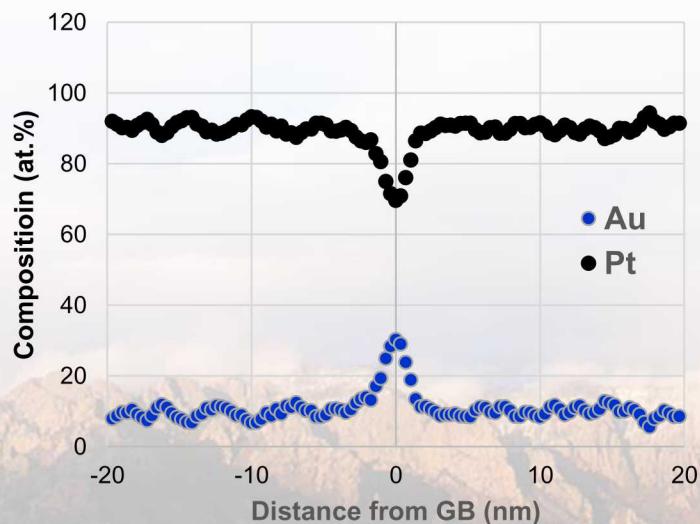
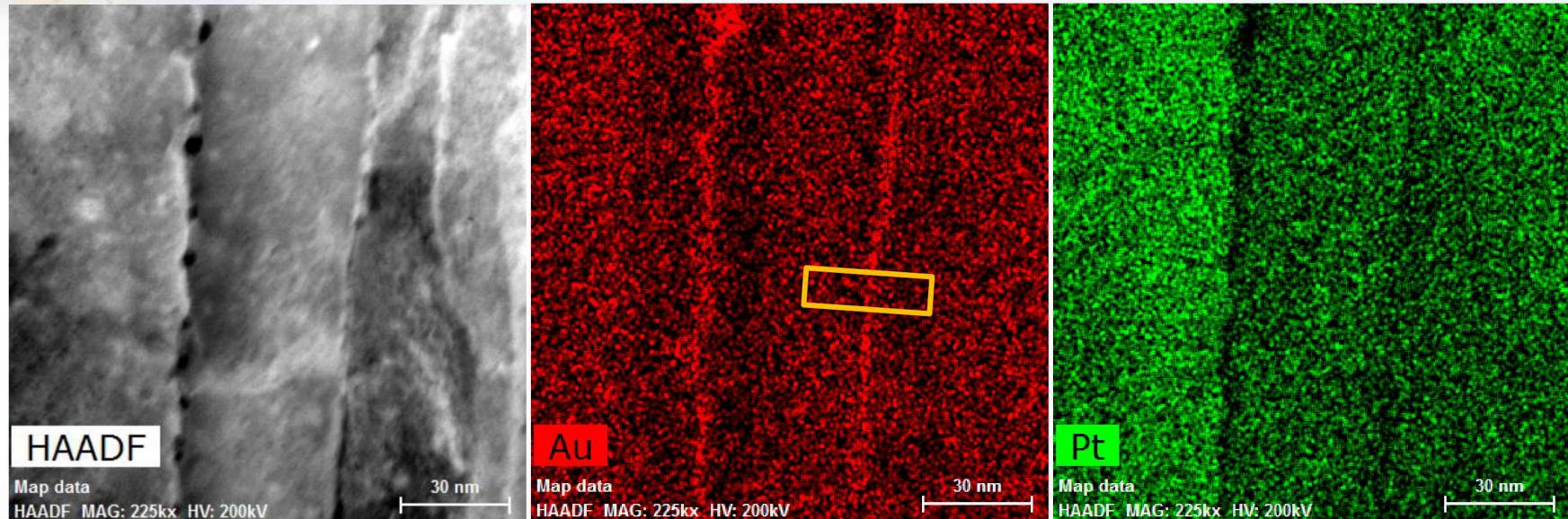


*Box is STEM-
EDS area



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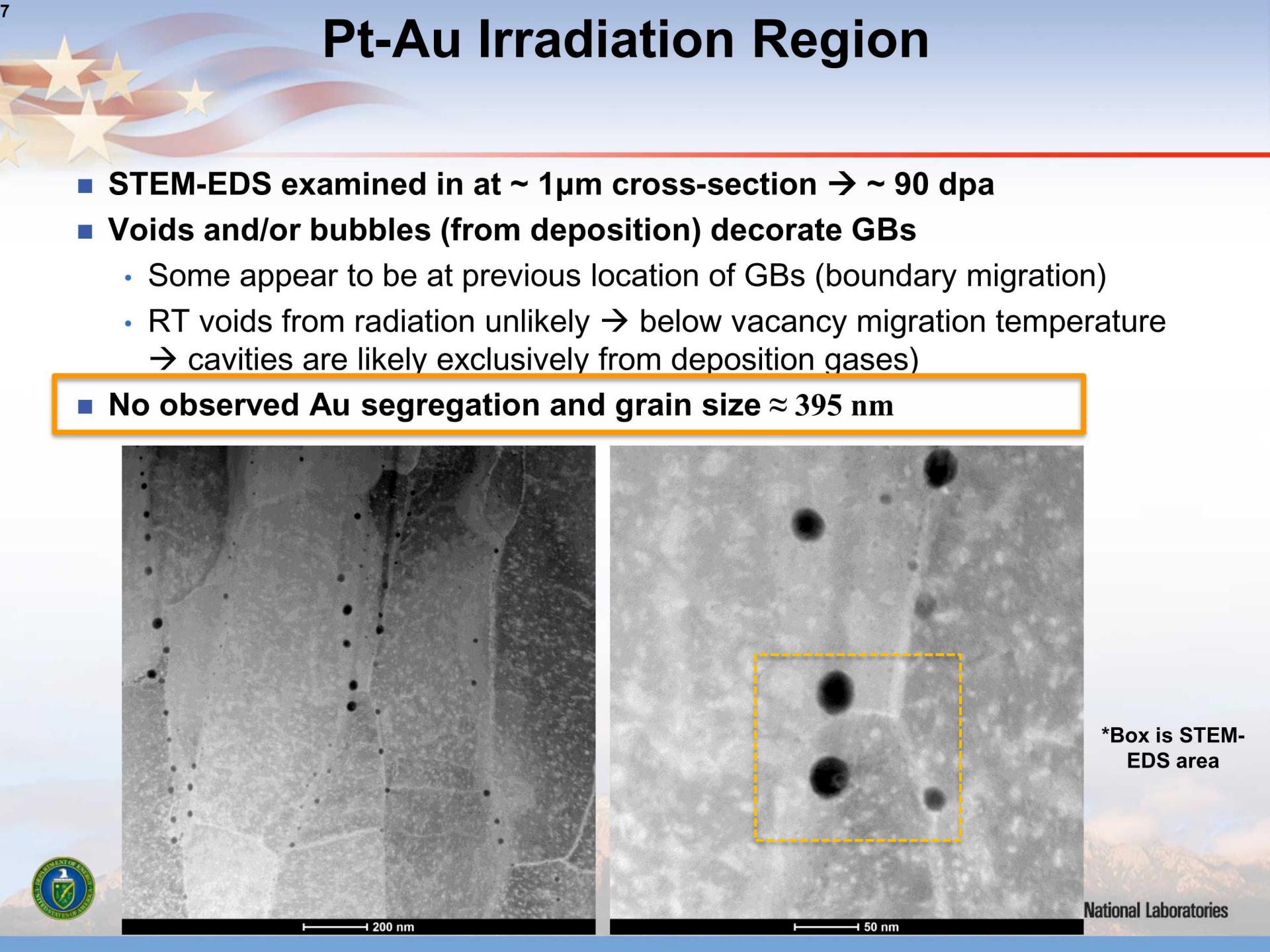
Pt-Au Thermal Region: ~ 30-35 at.% Au at GBs



*Box is representative STEM-EDS line scan from map

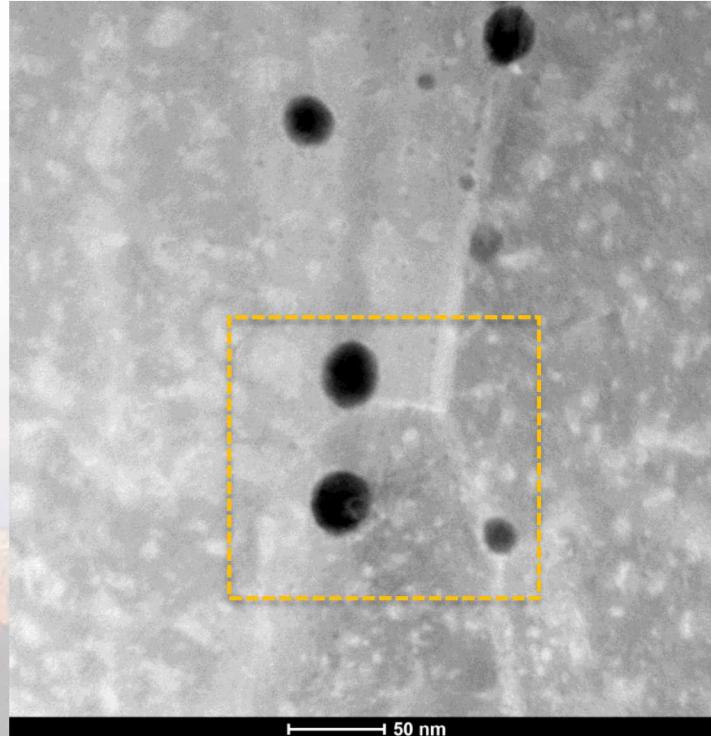
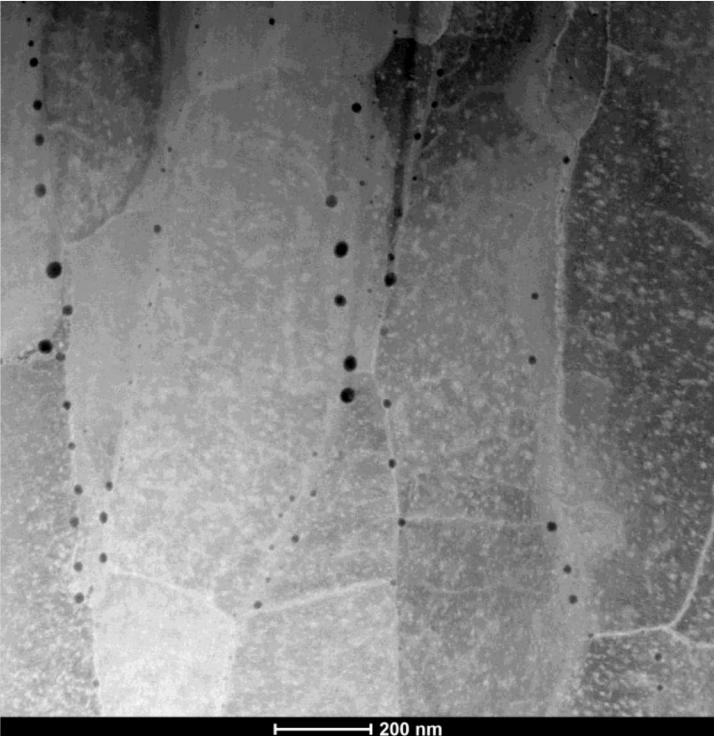


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Pt-Au Irradiation Region

- STEM-EDS examined in at $\sim 1\mu\text{m}$ cross-section $\rightarrow \sim 90$ 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 \rightarrow below vacancy migration temperature
 \rightarrow cavities are likely exclusively from deposition gases)
- **No observed Au segregation and grain size ≈ 395 nm**

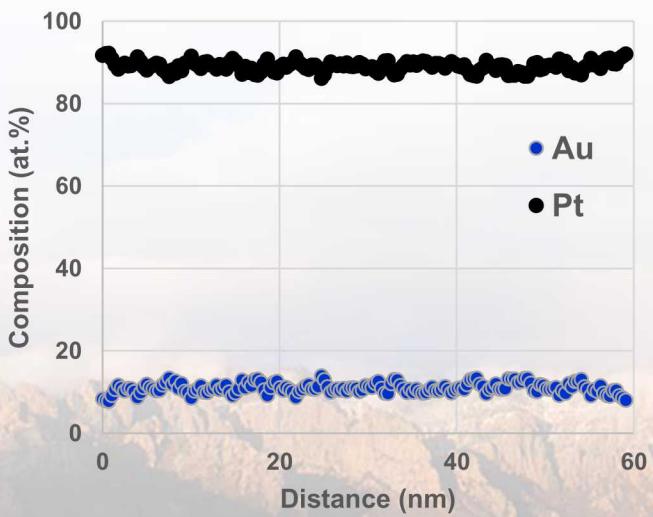
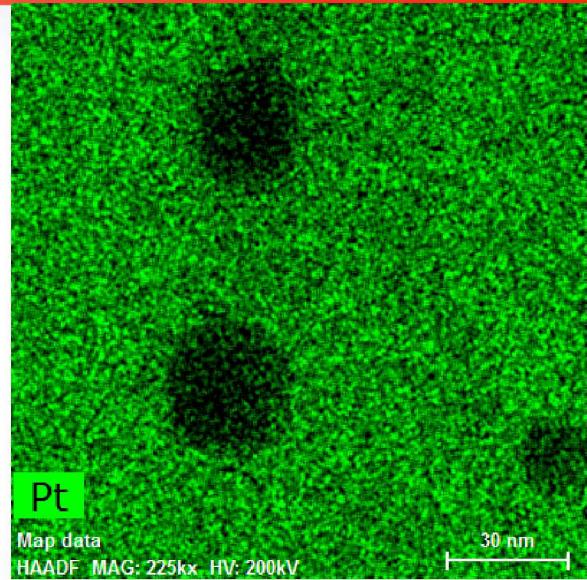
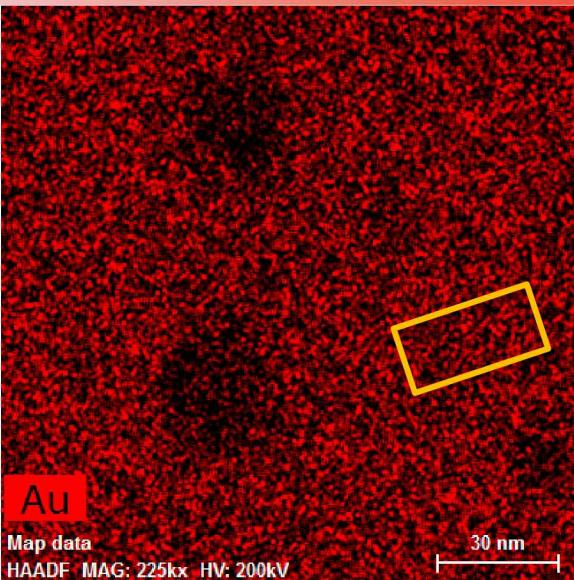
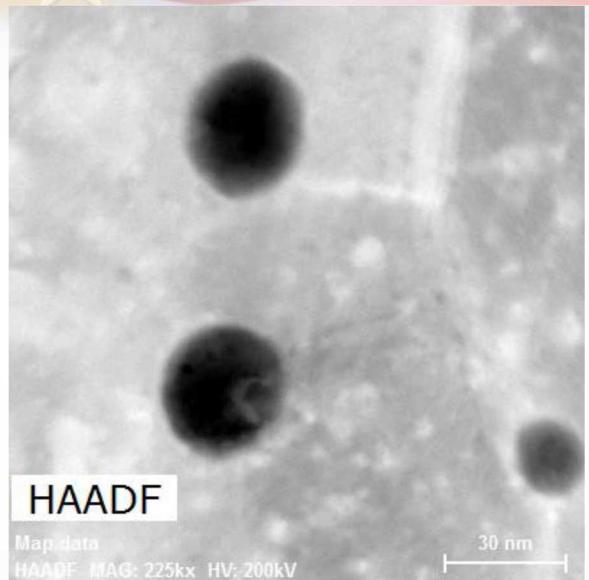


*Box is STEM-
EDS area



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Pt-Au Irradiation Region



*Box is representative STEM-EDS line scan from map



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Lack of radiation tolerance: ion beam mixing?

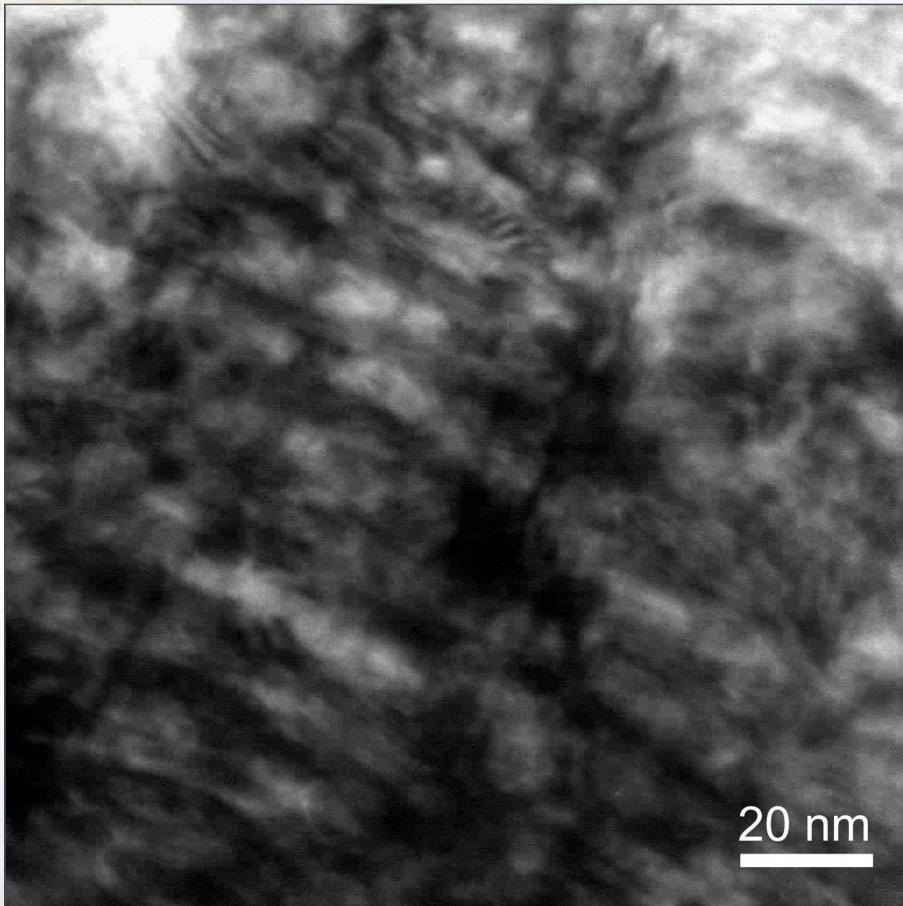
- **Grain growth (50 nm to 395 nm) in the irradiation zone**
- **Why hasn't the Au at the GB (confirmed prior to irradiation) stopped GB migration during irradiation?**
- **Effect of ion beam mixing during RT irradiation**
 - **Immiscible alloy system → possible that this induced full mixing (Au at GB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



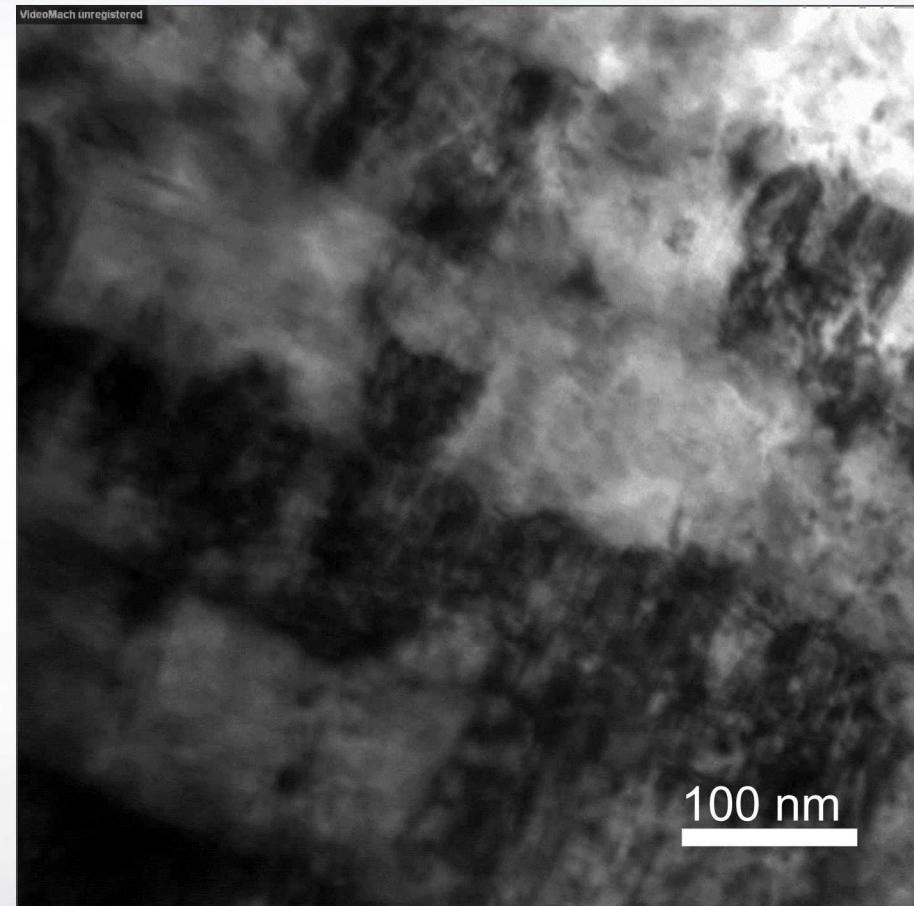
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Layer Deposition to Provide Defect Sinks

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



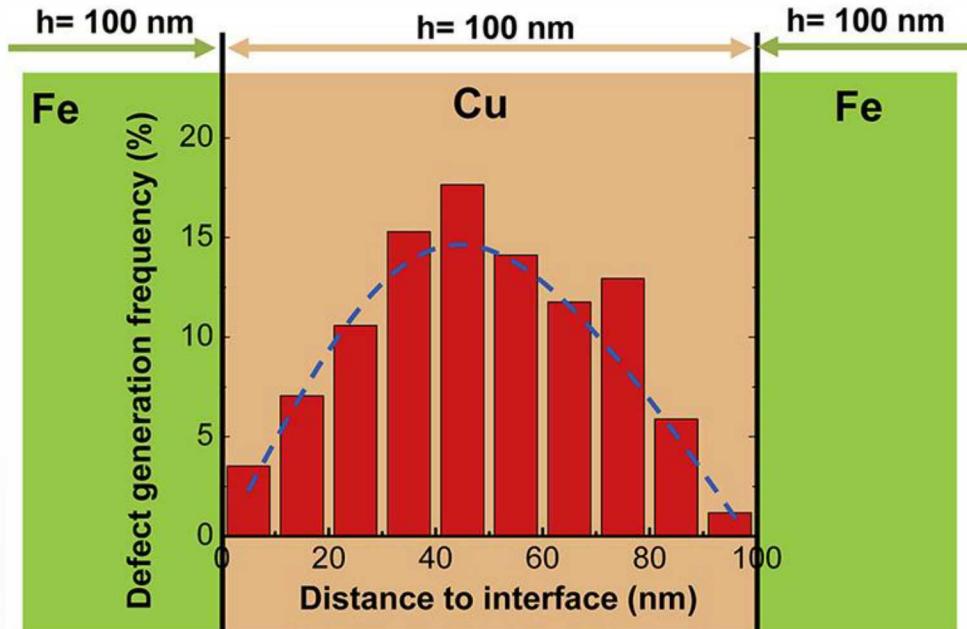
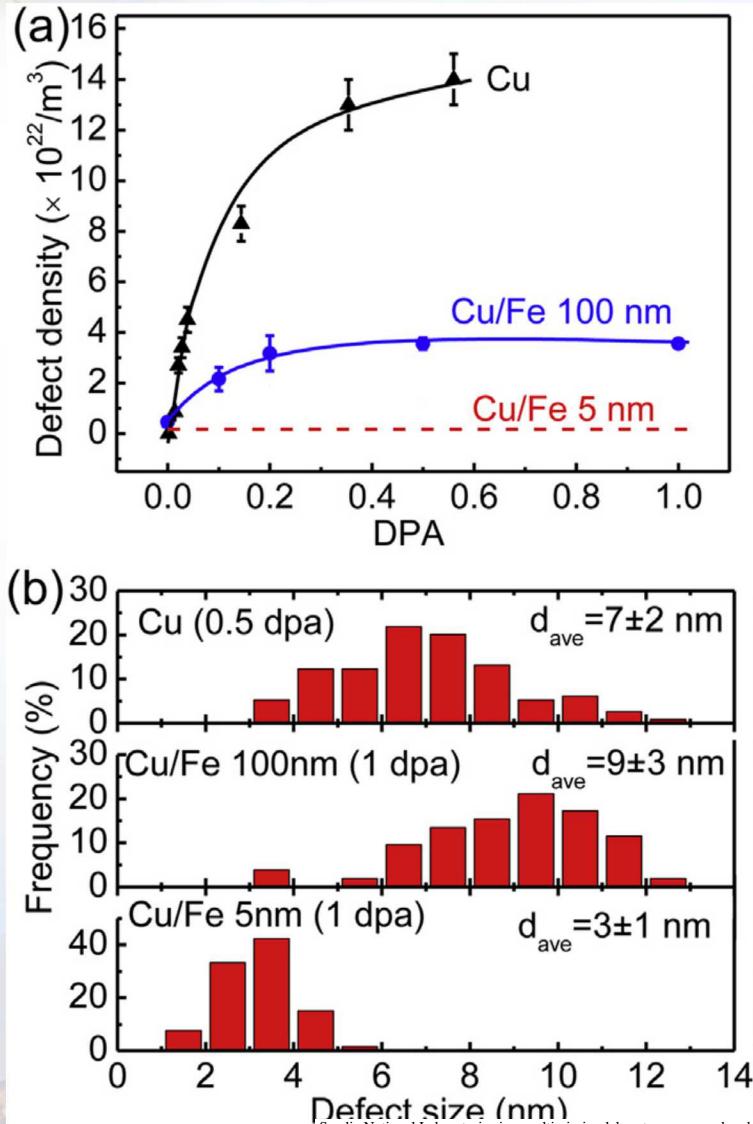
Film deposition of layered structures can also provide an opportunity for radiation defect sinks



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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.

