SAND2018-2749C

Exceptional service in the national interest

Sandia National Laboratories

Deconvolution of Complex Environmental Effects Active in Nuclear Reactor Materials Through In-situ Ion Irradiation

Caitlin Taylor, Christopher Barr, Samuel Briggs, Brittany Muntifering, and Khalid Hattar

Sandia National Laboratories

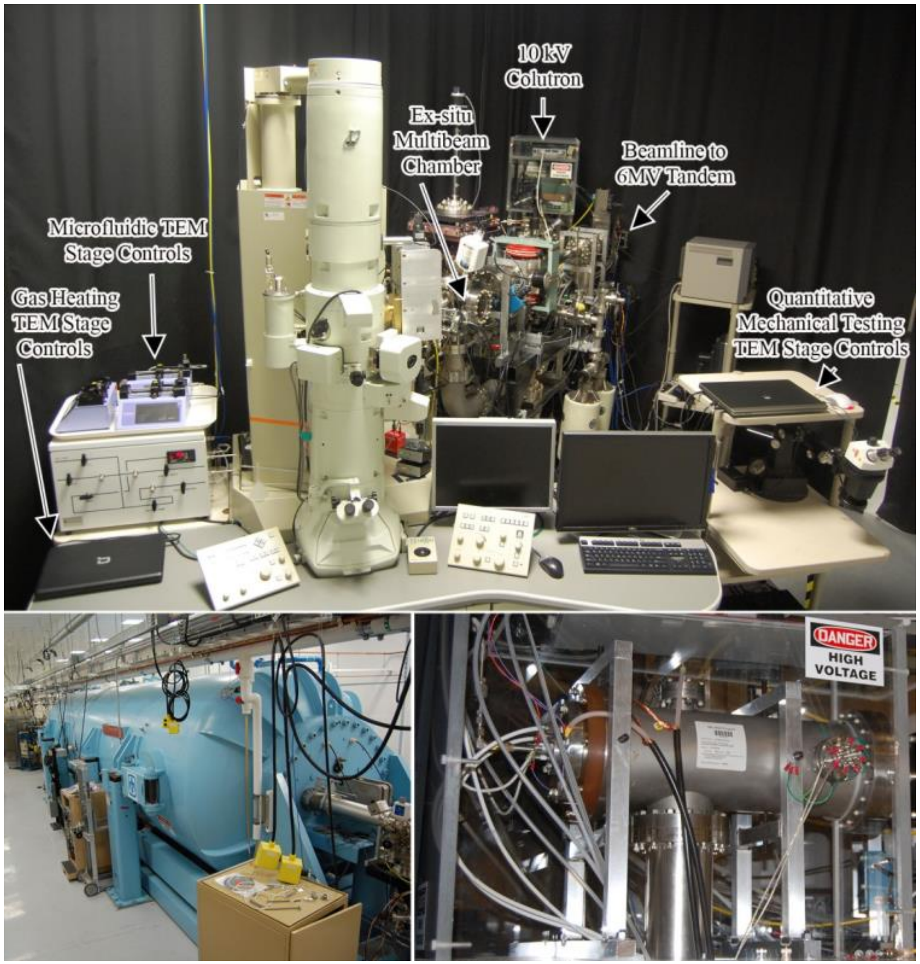
U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

Sandia National Laboratories is a multimission 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. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

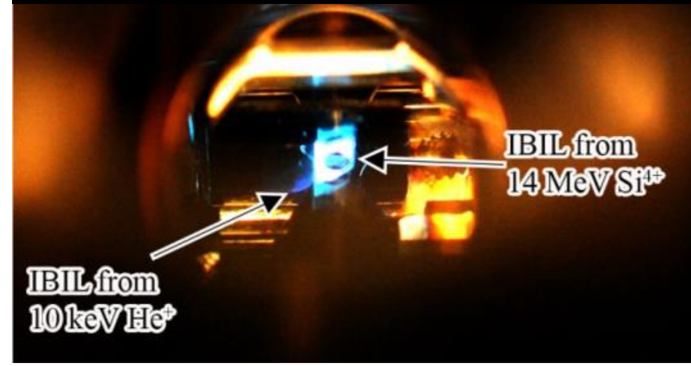
Sandia's Concurrent In-situ Ion Irradiation TEM Facility (I3TEM)

JEOL 2100 LaB6 microscope



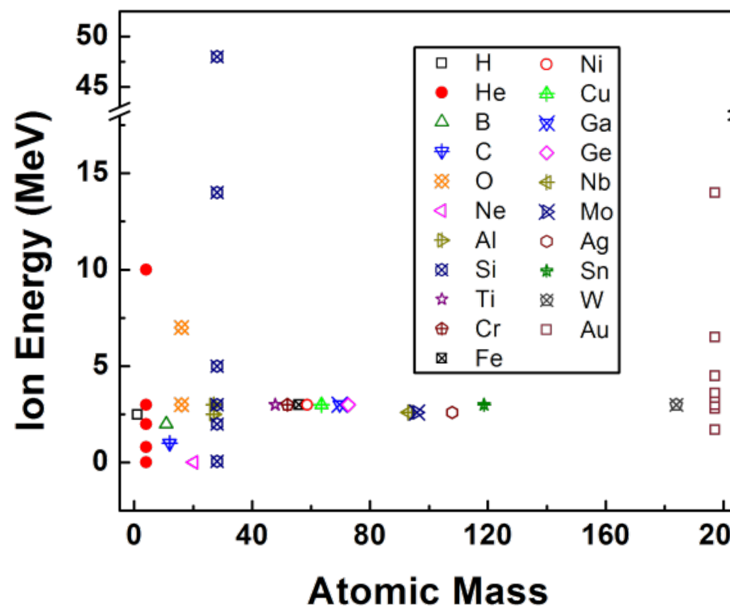
10 kV Colutron – 200 kV TEM - 6 MV Tandem

IBIL from a quartz stage inside the TEM

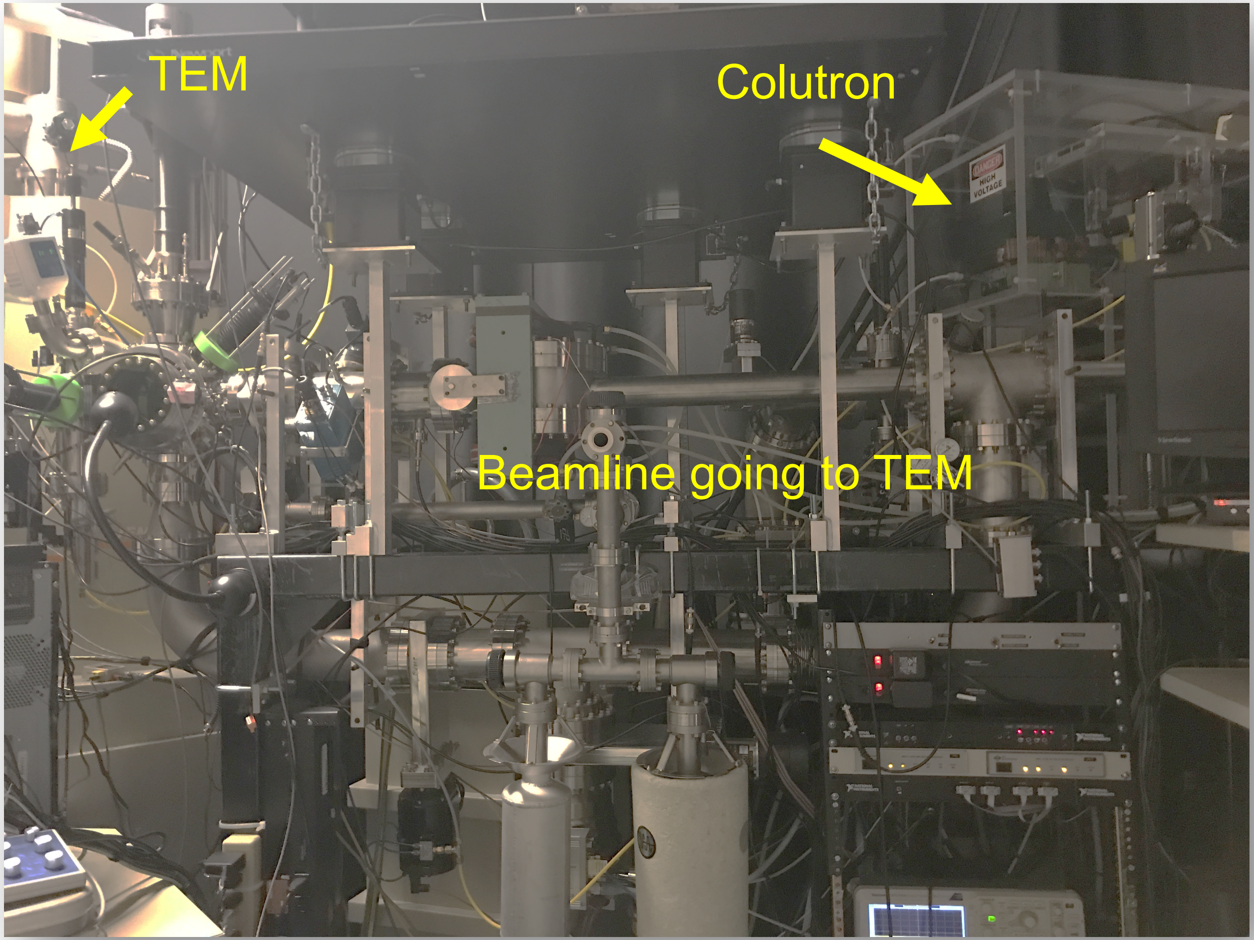


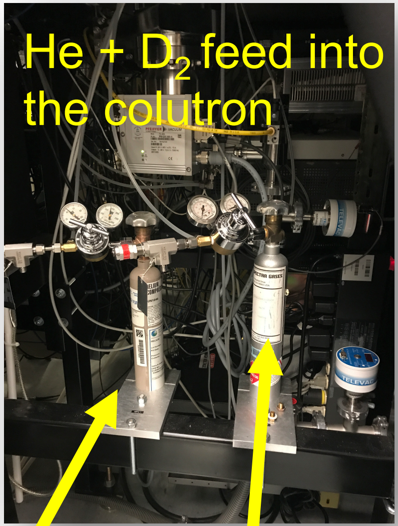
Direct real time observation of ion irradiation, ion implantation, or both with nanometer resolution

Ion species & energy introduced into the TEM



Sandia's I3TEM (side view of incoming beams)

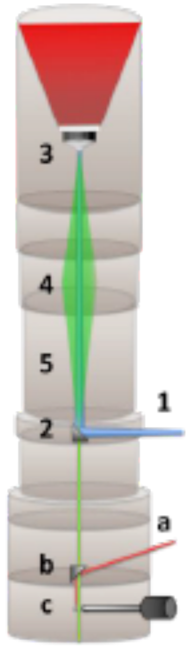




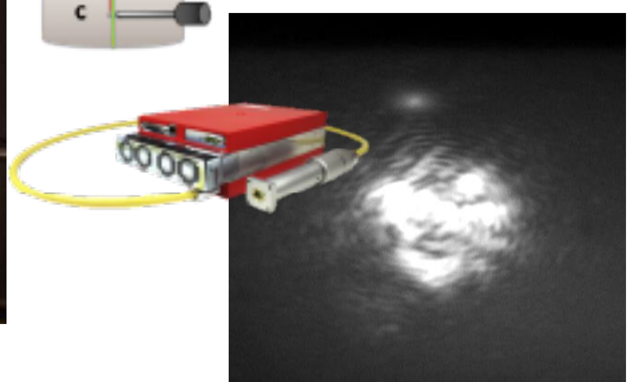
- JEOL 2100 TEM. Samples are tilted to +30º in x to face the ion beam
- Both He and D ions are part of the same ion beam (He + D_2)

Synergistic In-Situ Capabilities at I3TEM: Laser heating

Unique setups feasible when coupled with IR sample heating:



a. Adjustable power 1064 nm infrared specimen (IR) drive laser
b. IR laser is reflected directly onto the specimen with metal mirror
c. Heat specimens in in situ holders, which otherwise would not be possible

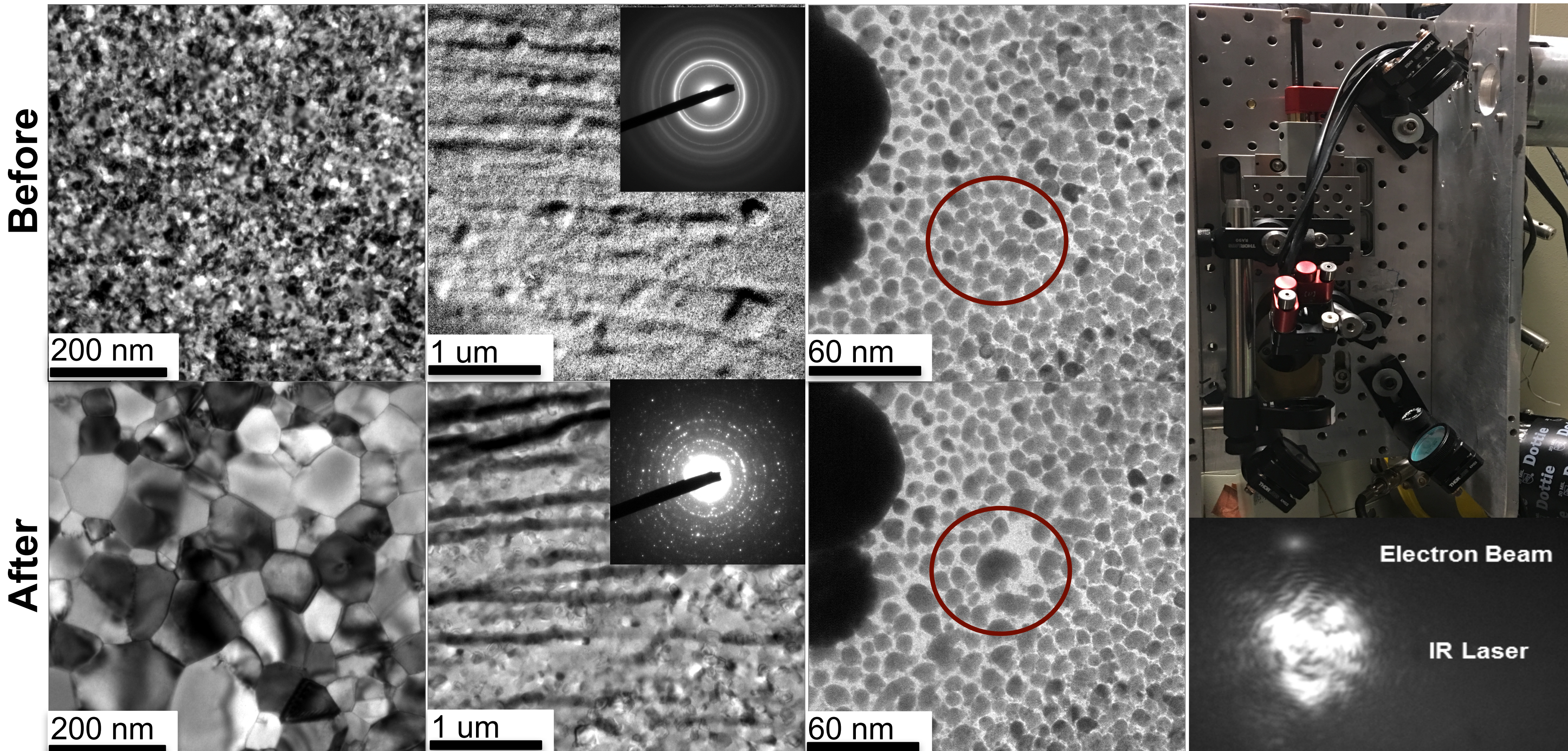


- Laser capabilities:
  - 2-20 Watts
  - Pulsed or continuous operation
  - Tunable spot size (typically 50 µm diameter spot size) – smaller/large possible
  - Positioning mirror, which can be used during laser operation

Synergistic In-Situ Capabilities at I3TEM: Heating via Laser Operation

Sandia National Laboratories

Pt Grain Growth | **Reactive Multilayer Films** | **Nanoparticle Sintering (20 nm Au**

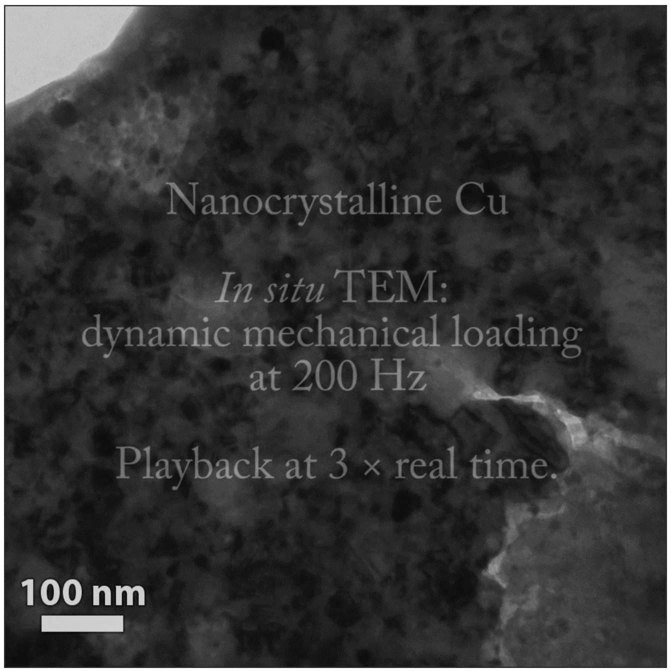


- **We can now introduce rapid thermal heating with any TEM stage or ion beam conditions**
- **Methods underway for accurate temperature determination**

Collaborator: P.M. Price, D. Adams, M. Abere, IDES (B.W. Reed)

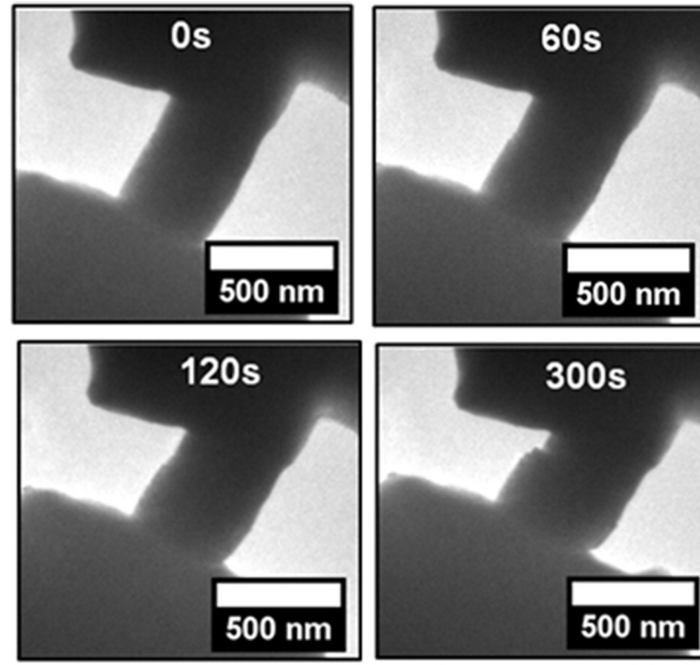
Synergistic In-Situ Capabilities at I3TEM: Mechanical Testing

- Quantified loading/fatigue/creep



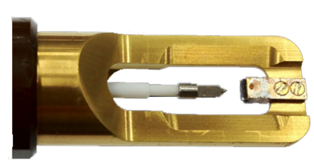
D. Bufford et al., Nanoletters (2016)

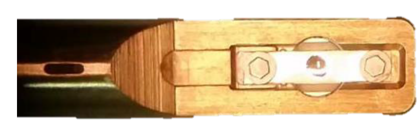
- Irradiation induced creep



G.S Jawaharram et al., Scripta Mat **148** (2018) 1-4

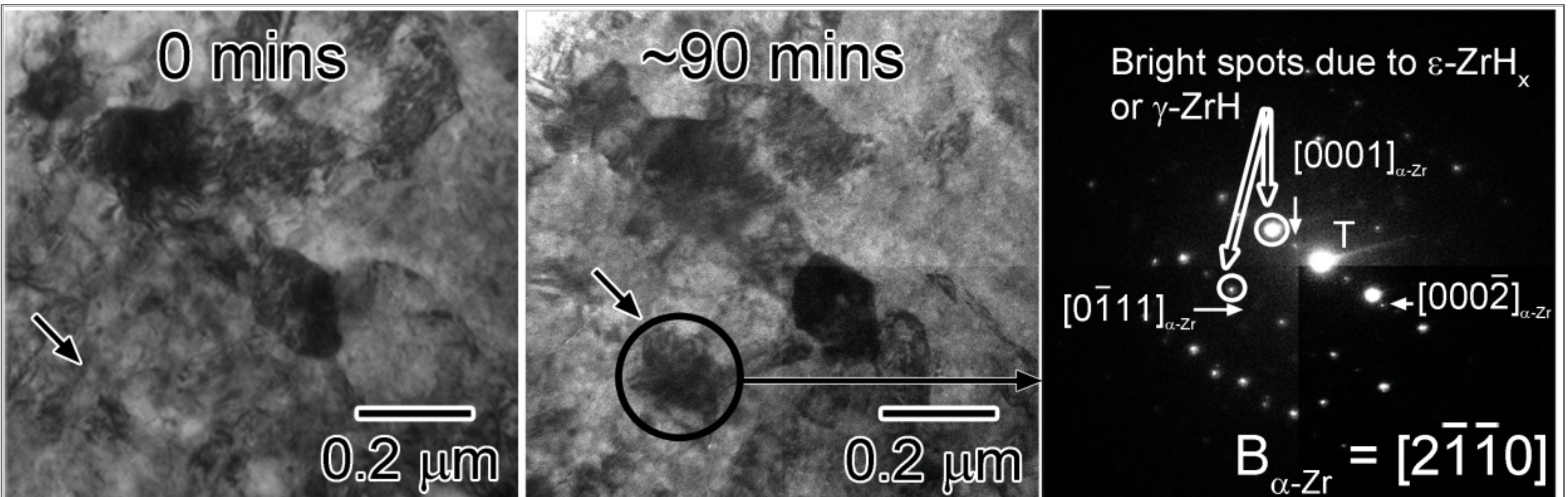
Stage options:
Hysitron PI-95 or Gatan heating/strain holder coupled with IR laser heating

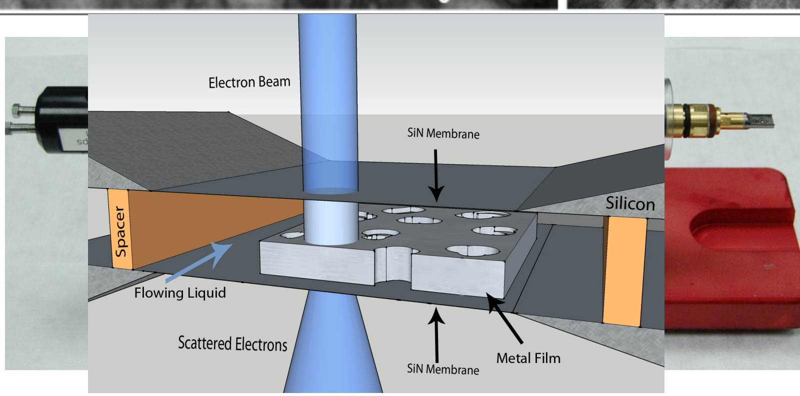




Synergistic *In-Situ* Capabilities at I3TEM: Corrosion in Zircaloy-2

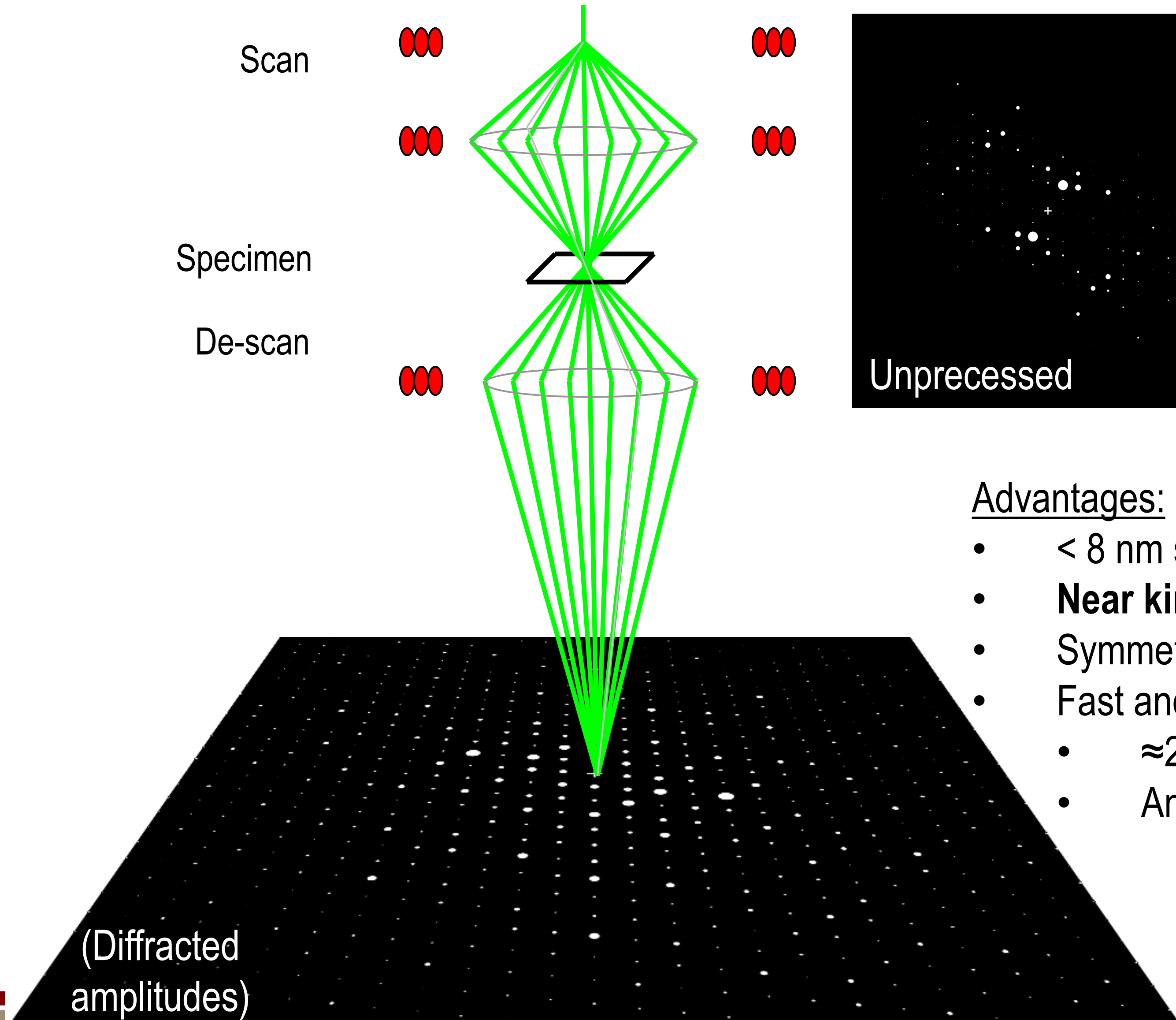
Absolute hydrogen pressure: 327 torr (~ 0.5 atm),
Ramp rate: 1 ^{o}C/s, **Final temperature:** ~ 400 C, **Dwell time:** ~ 90 mins

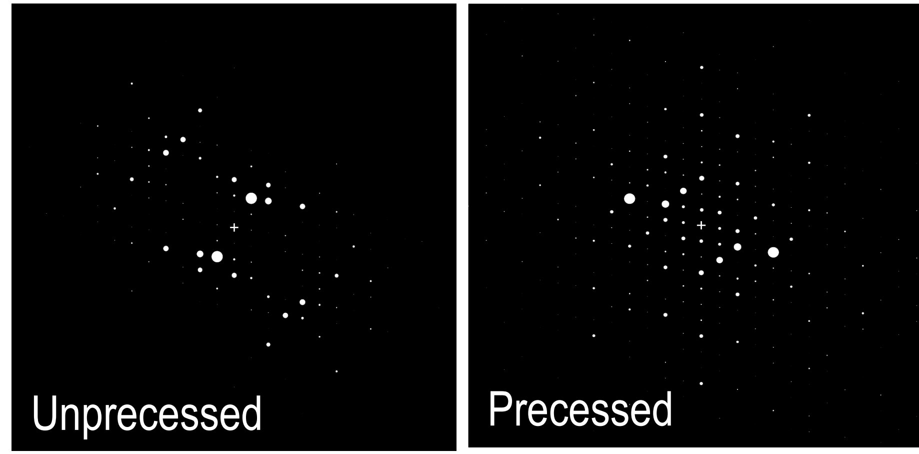




Hydride formation shown in TEM gas-cell stage at elevated temperature and hydrogen pressure

Synergistic Capabilities at I3TEM: Precession Electron Diffraction Microscopy (PED)





Advantages:

- < 8 nm spatial resolution (LaB6)
- **Near kinematical electron diffraction**
- Symmetry ambiguities are resolved
- Fast and automated acquisition
  - ≈200 grains in 15 min.
  - Angular resolution ≈1°

Chris Own, PhD Dissertation, Northwestern University, 2004

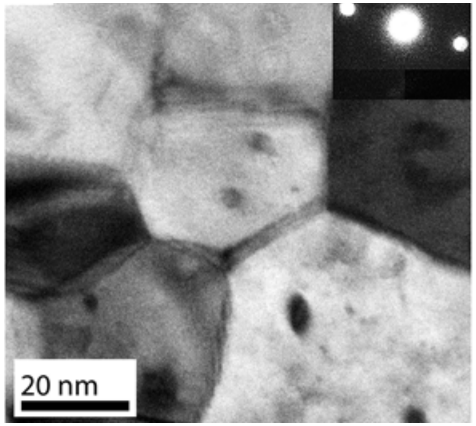
Slide courtesy: K. J. Ganesh and K. Hattar

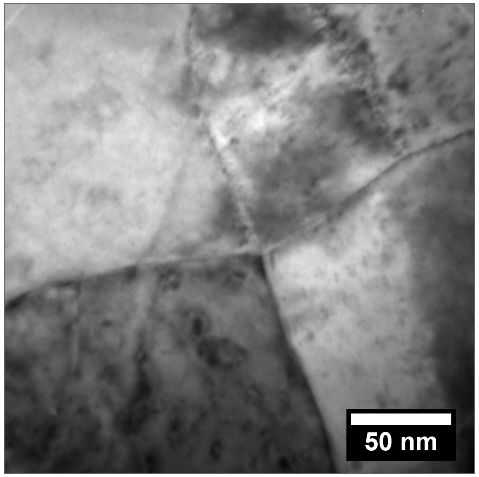
Two Case Studies of Irradiation Type Experiments Feasible at I3TEM

Most Common In-situ TEM Irradiation Setups

(1) Heavy ion irradiation into NC Pt

(2) Helium implantation and annealing in NC Pd – "Briefly"





All experiments (ion irradiation, helium implantation, and triple combined beam) can be coupled with other in-situ techniques previously detailed for additional complexity and simulated environments

Case 1: Grain Size Effects Under Irradiation in NC Pt

Project collaborators: Nan Li and Brad Boyce

NC Grain Size Effects: Motivation

- **Increase GB density →** numerous nanocrystalline studies indicating reduced defect size and/or density of defect (dislocations and voids) with NC grains
- Defect size and density reduction indicate improvements (reduction) in defect size and density from coarse grain counterparts
- **Very 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

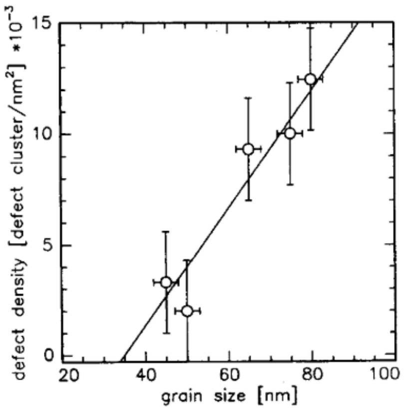
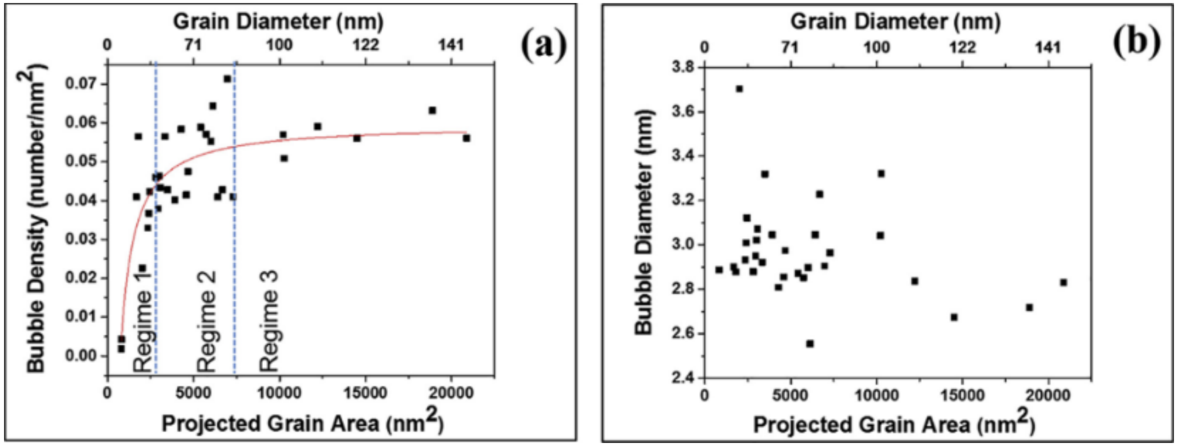


Fig. 8. Defect densities vs. grain size on the irradiated Pd sample (240 keV, 2×10^{16} Kr/cm^2).

Rose et al. (1997)

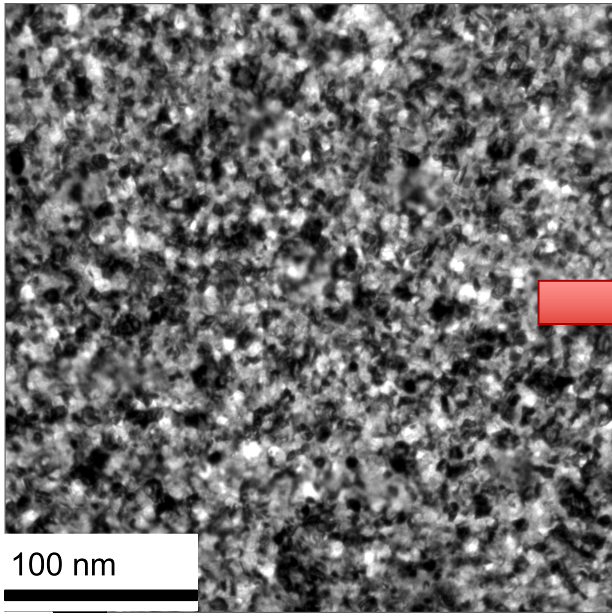
NC Fe Helium bubble trends at 427°C



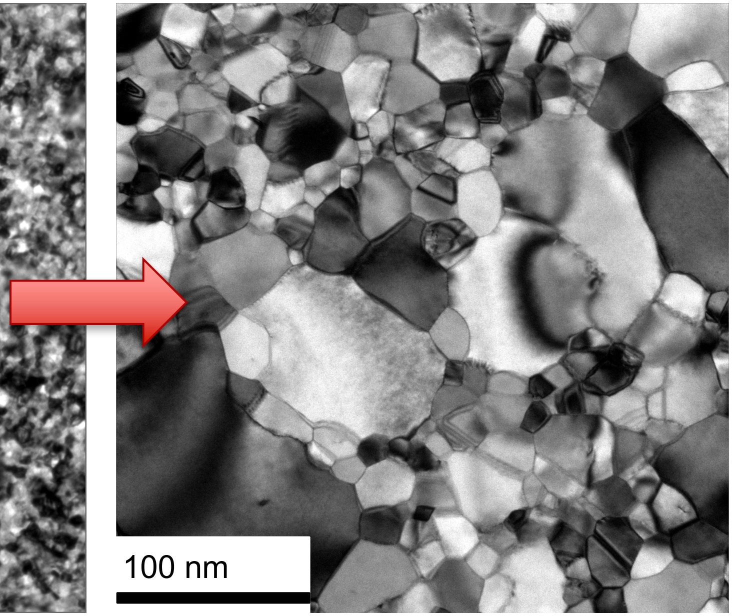
El-Atawani et al. **484** (2017) 236-244.

NC Grain Size Effects : Methodology

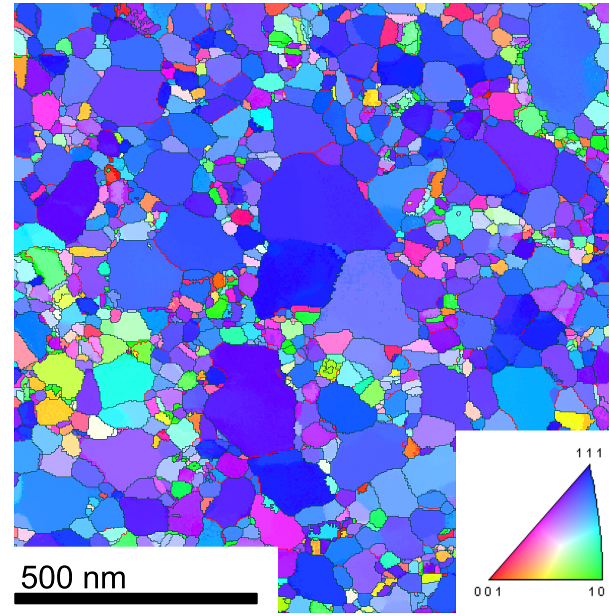
As-deposited NC Pt

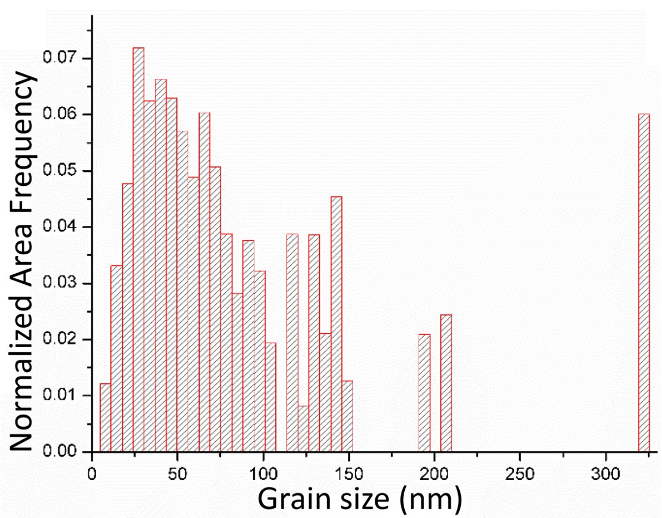


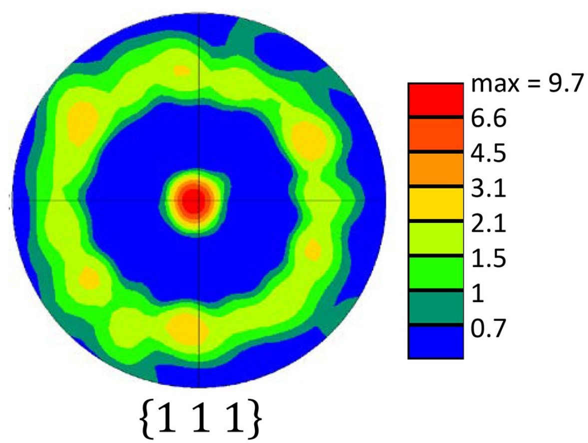
Annealed NC Pt (500 º C/2hrs)

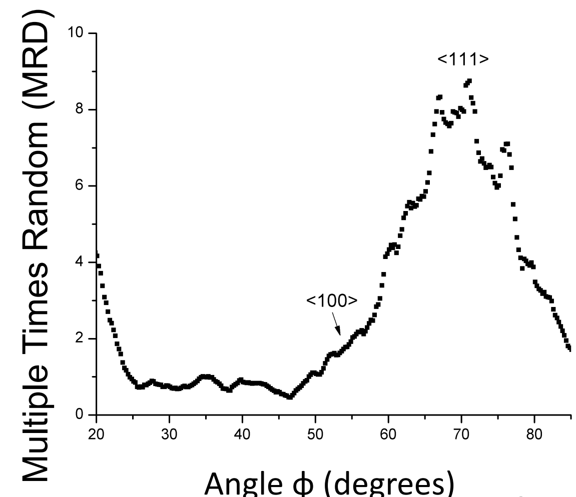


Annealed PED Map



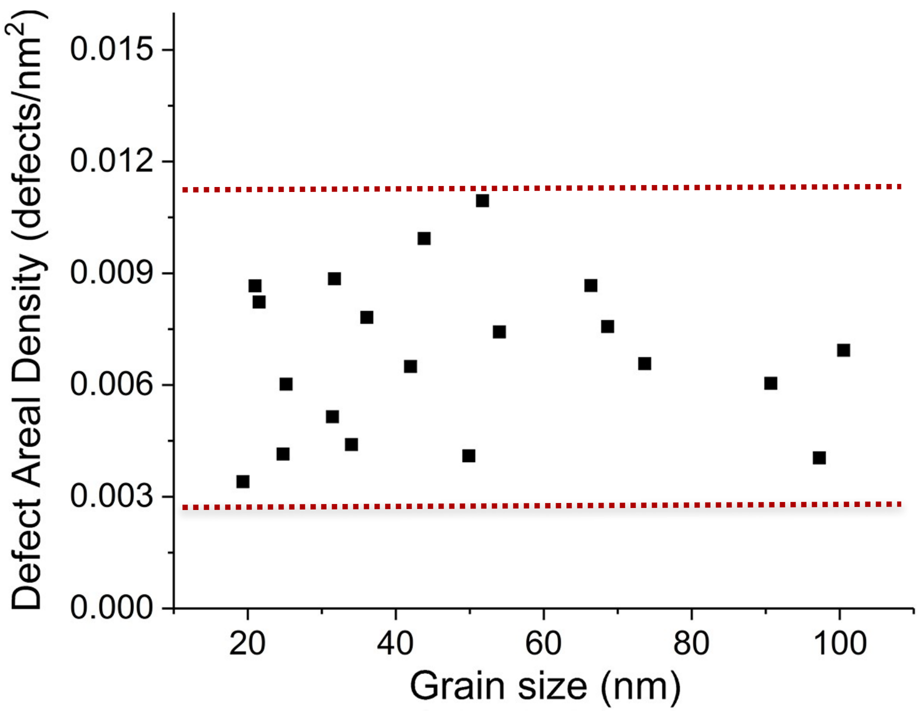


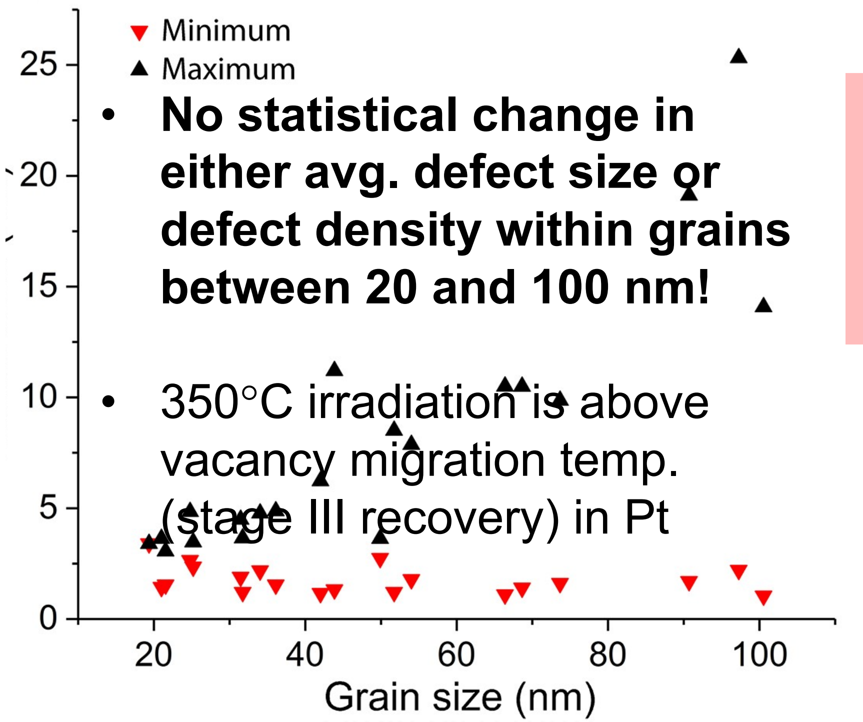




NC Grain Size Effects : Irradiation and Defect Trends

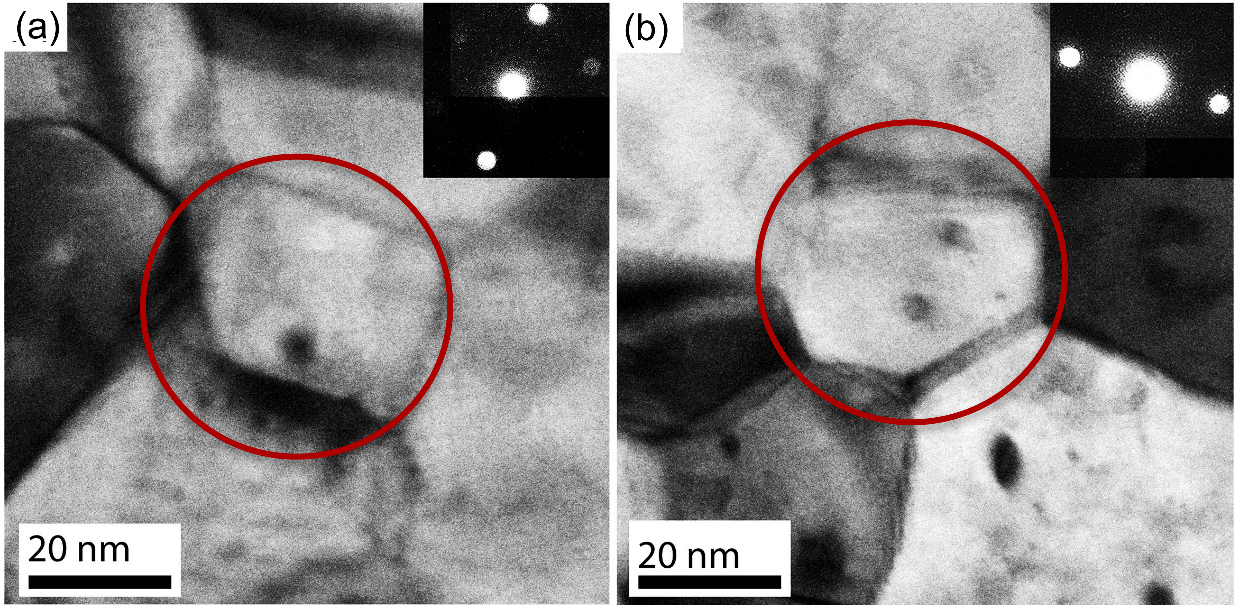
- In-situ TEM irradiations: 2.8 MeV Au^{4+} at a flux of 4.7×10^{10} ions/cm^{2}s to a fluence of $1.7 \times 10^{14} \frac{ions}{cm^2}$ at 350^{o}C using a Gatan double tilt heating stage
- **Average final dpa ≈ 3 dpa**
- NC grains with a slightly deviated g = <111> diffraction condition using two-beam 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



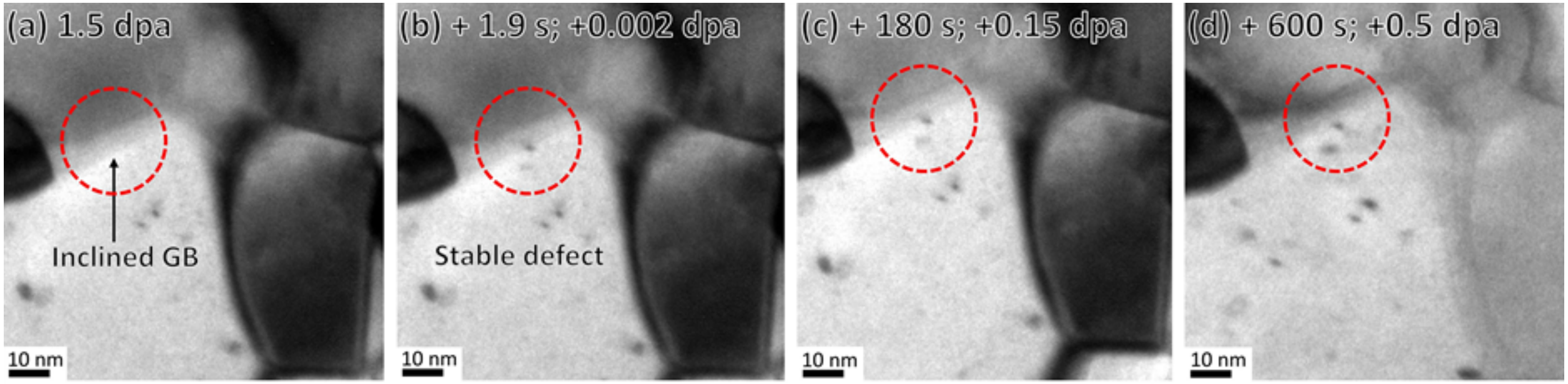
- Two grains → 20 nm in size (measured via mean intercept length)

- **How do you count the defects in these grains? 1 to 3 defects nearly full range of density!**
  - Grain A = 1 defect → 3.3 x 10^{-3} defects/nm^2
  - Grain B = 3 defects → 8.6 x 10^{-3} defects/nm^2

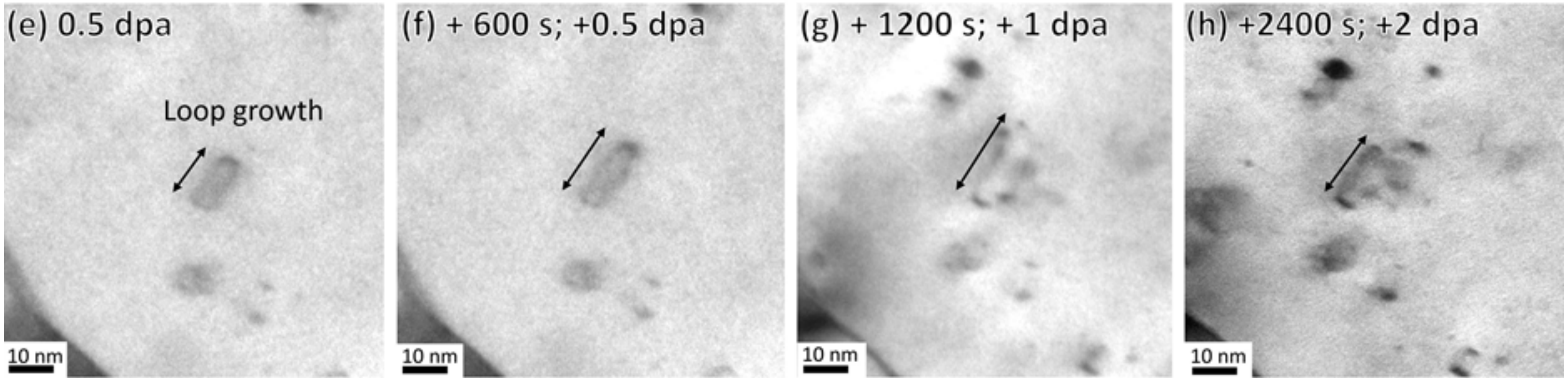
- **<u>With decreasing grain size, # of defects become both more challenging and critical for any type of accurate defect density</u>**

NC Grain Size Effects: Lack of Defect Density Reduction In-situ Observations

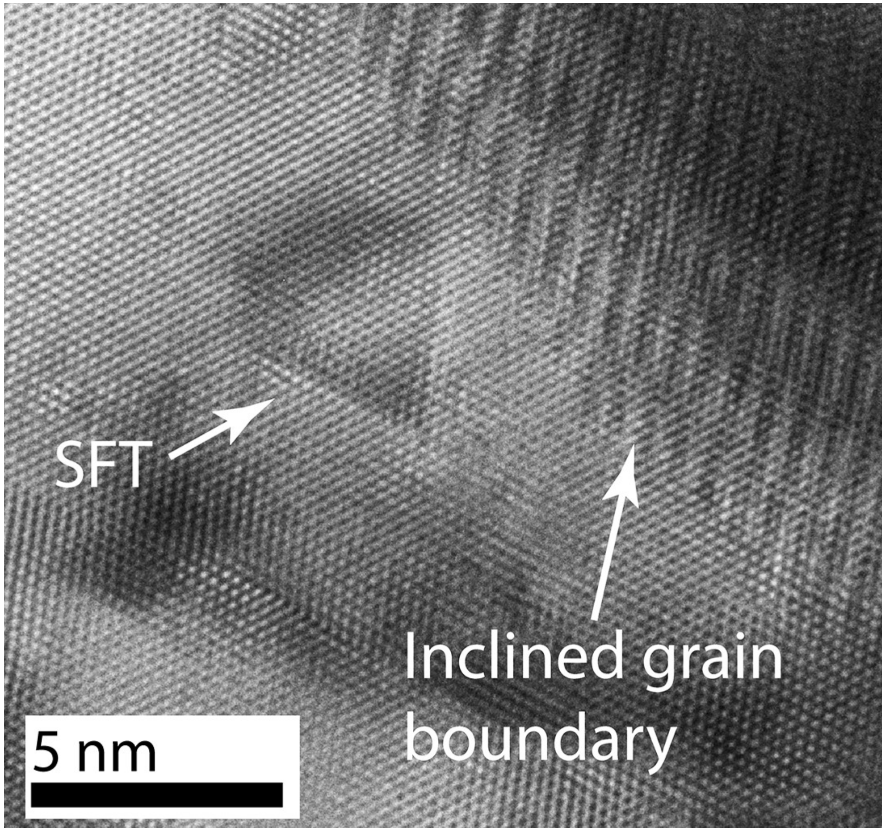
Defect evolution near GBs:



Defect evolution in grain interior

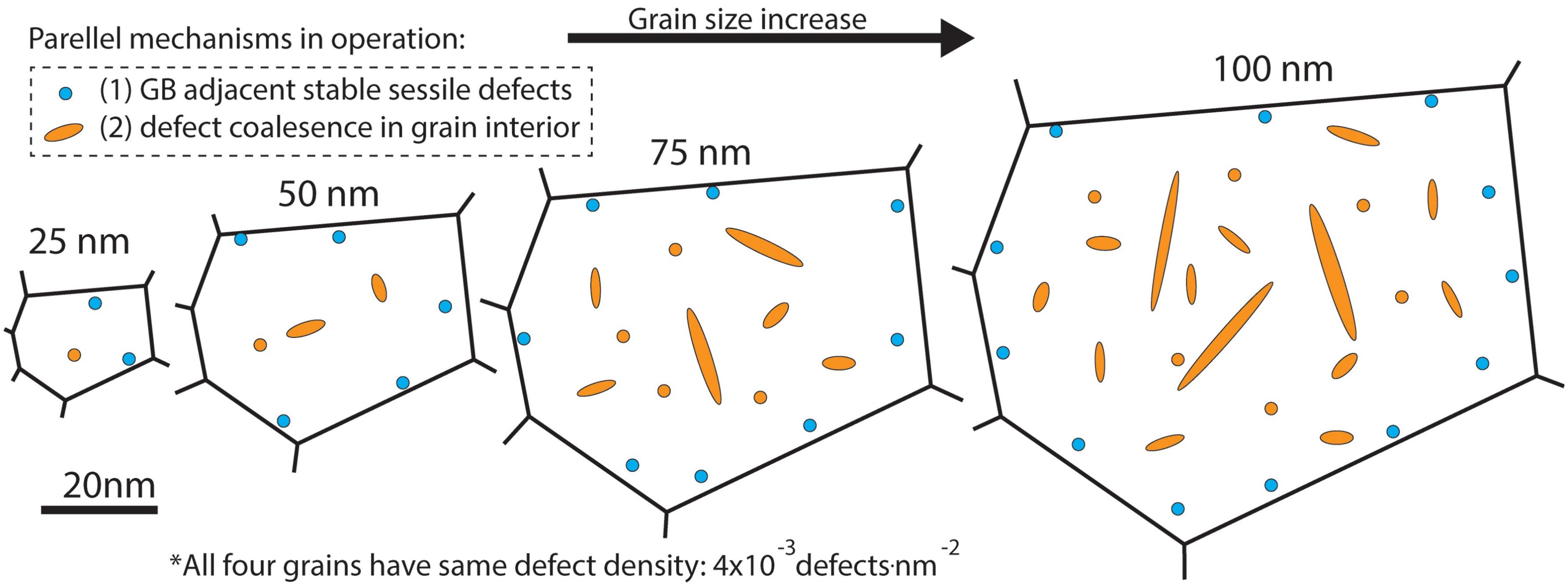


NC Grain Size Effects: Small loop stability near GBs



- 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!**

Case 2: Helium Bubble Nucleation and Growth in Pd

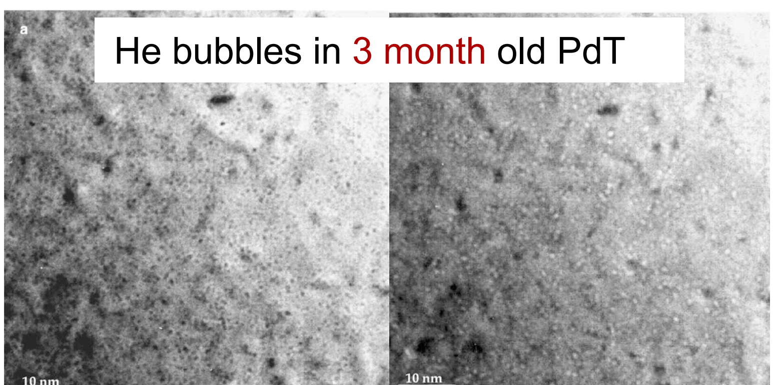
Palladium work completed by Caitlin Taylor (SNL), Collaboration with Joshua Sugar (SNL) and Dave Robinson (SNL)

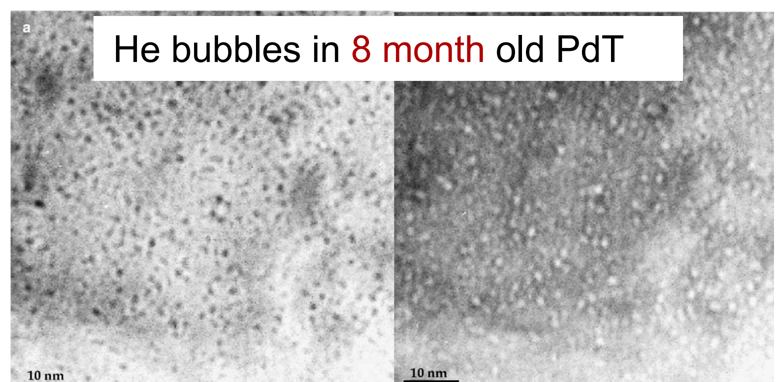
Palladium: Considered for solid-state ^{3}H storage →Result in ^{3}He bubbles and blistering

Sandia National Laboratories

- **Palladium is being considered for solid-state tritium storage**
- Helium accumulation
    - Due to rapid ^{3}He production in tritiated materials, **He bubble formation will eventually lead to blister formation**
    - Blisters bursting the surface release ^{3}He and ^{3}H from the material.

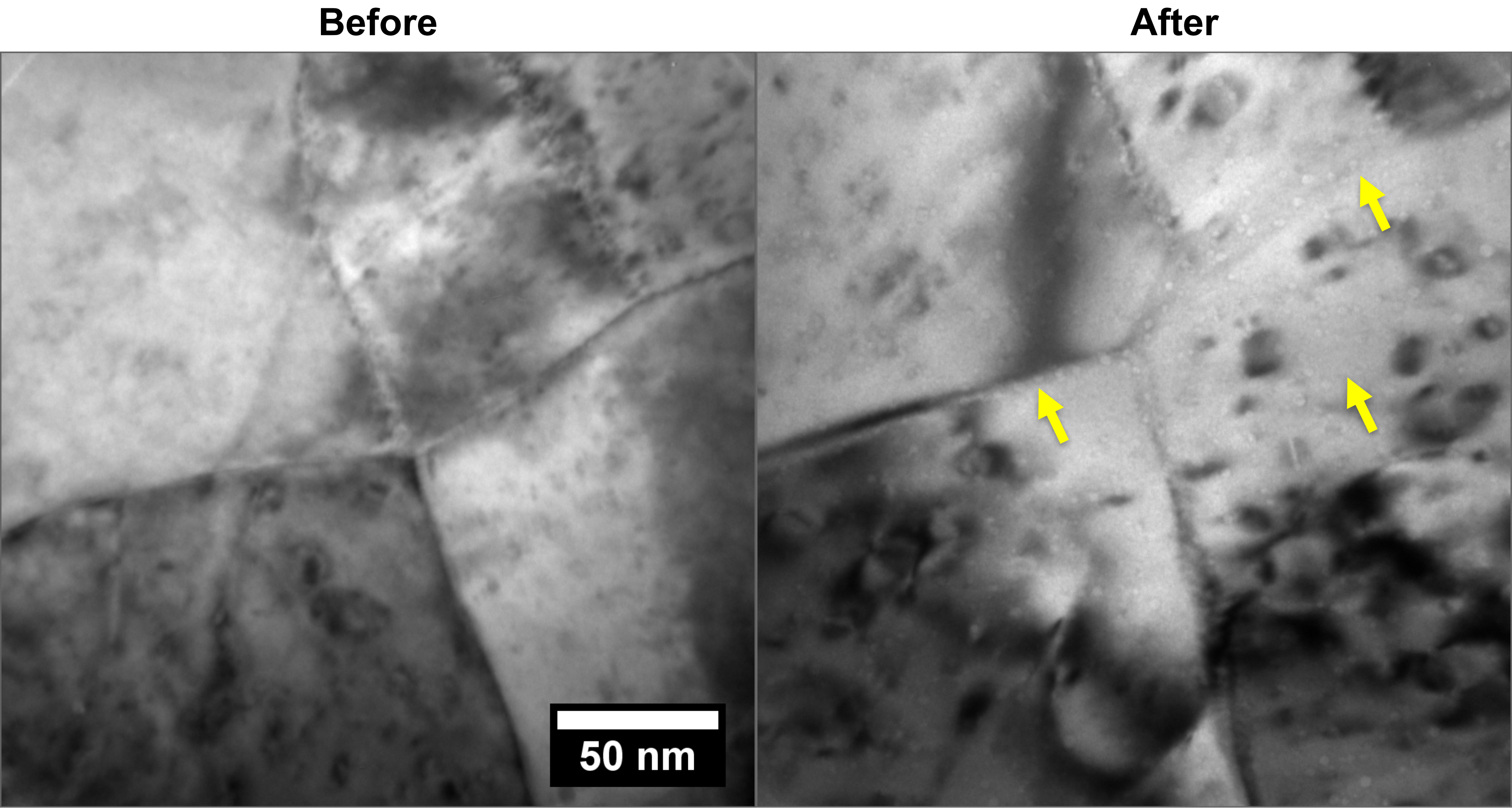
- **Bubble nucleation mechanisms in PdT are currently unknown. Necessary for predictive aging models**





Fabre et al, JNM 342 (2005) 101-107

Nanometer sized bubbles nucleated inside grains and at boundaries during in-situ 400^{o}C implantation



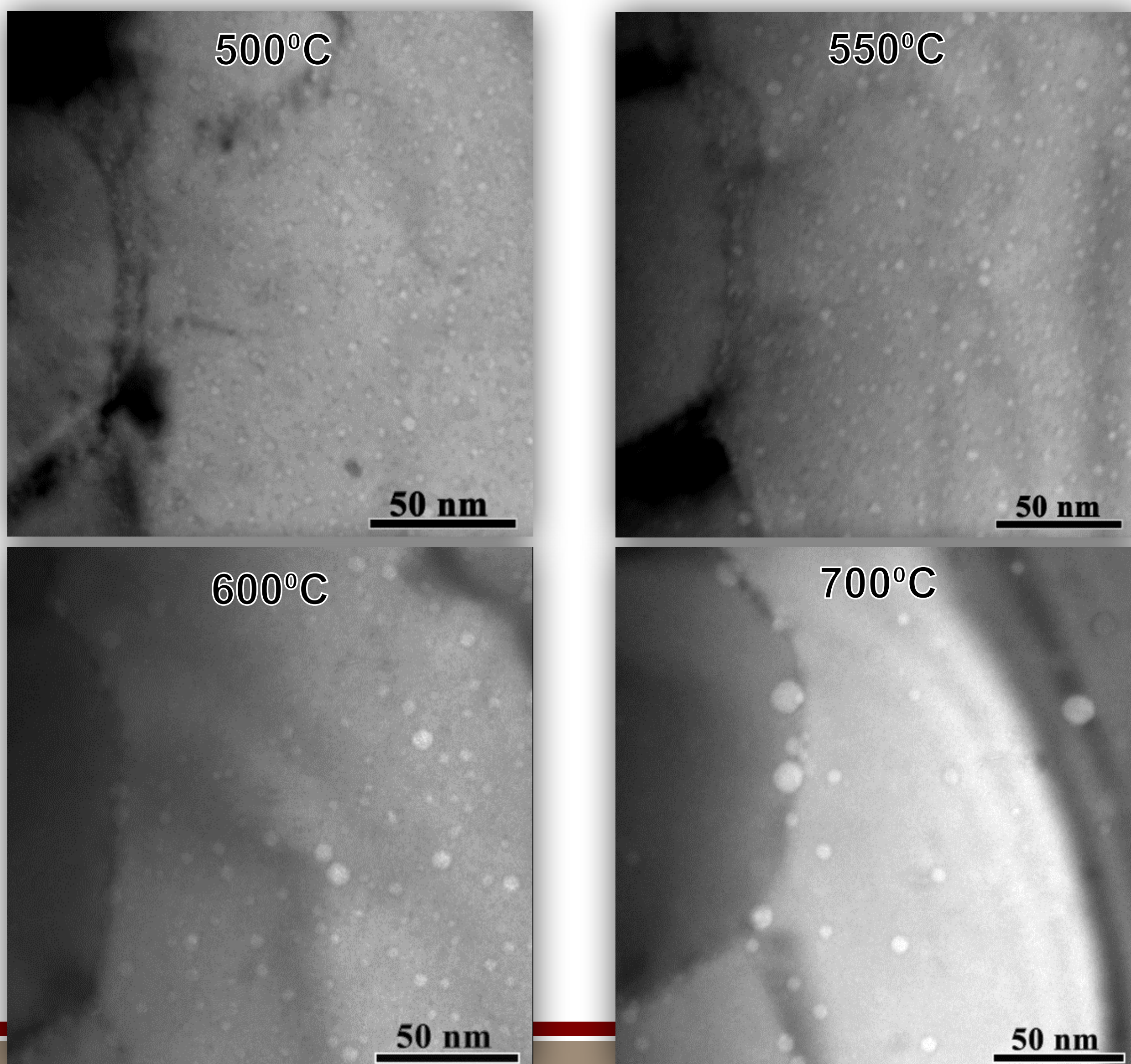
- During in-situ helium implantation, bubbles were observed forming simultaneously at the boundaries and inside the grains. 5 uA beam current, spot 10mm^2, 2 hrs total implantation
- Bubbles were an average of 1.9 nm in diameter after the implantation.

Bubble migration first observed at 450^{o}C, with bubble growth first being observed at 500^{o}C

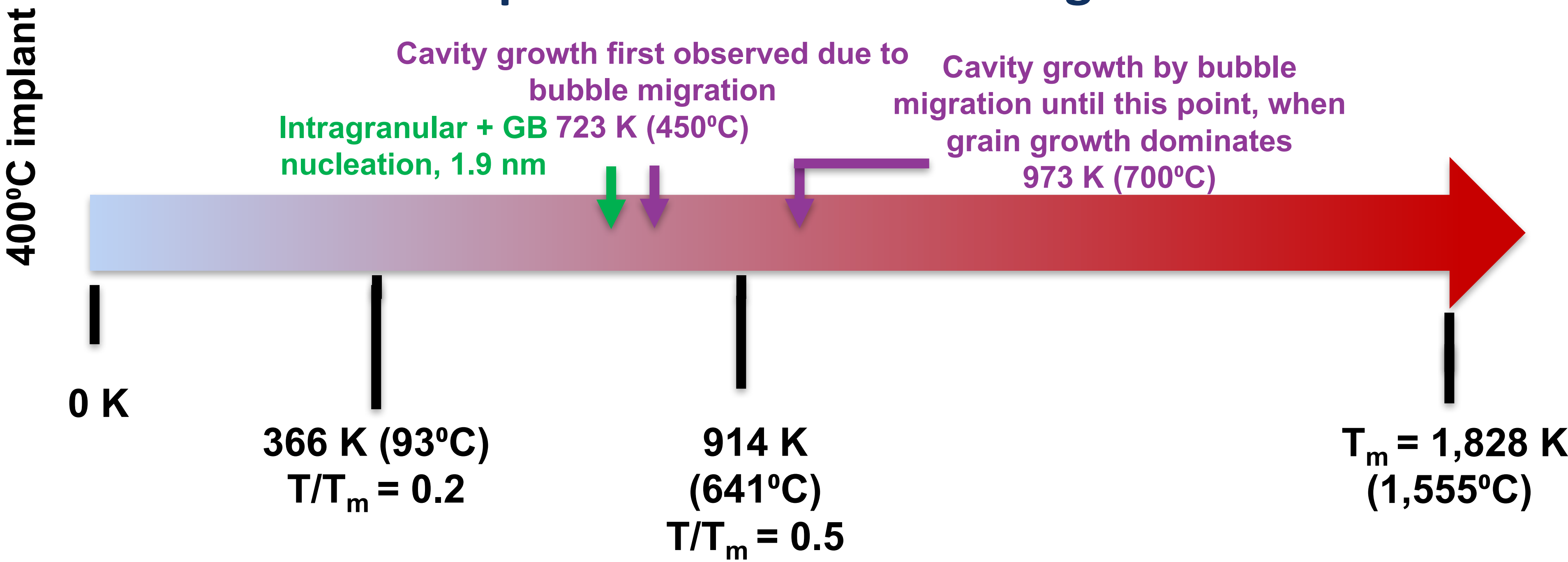
Sandia National Laboratories

Sample implanted @ 400^{o}C

Video 12x speed



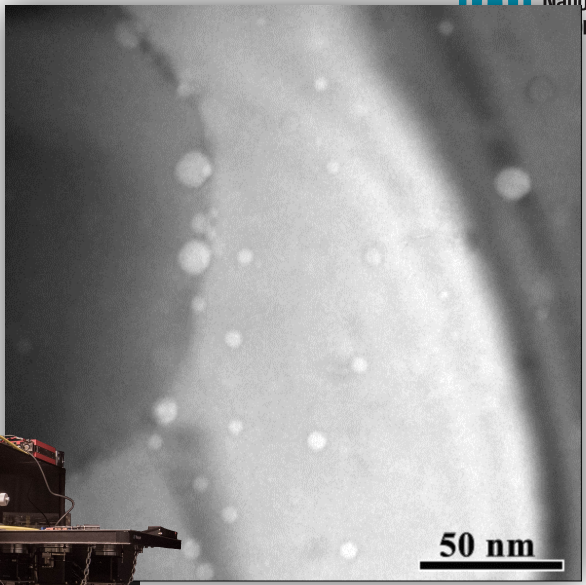
Summary of helium bubble kinetics observed during in-situ implantation and annealing

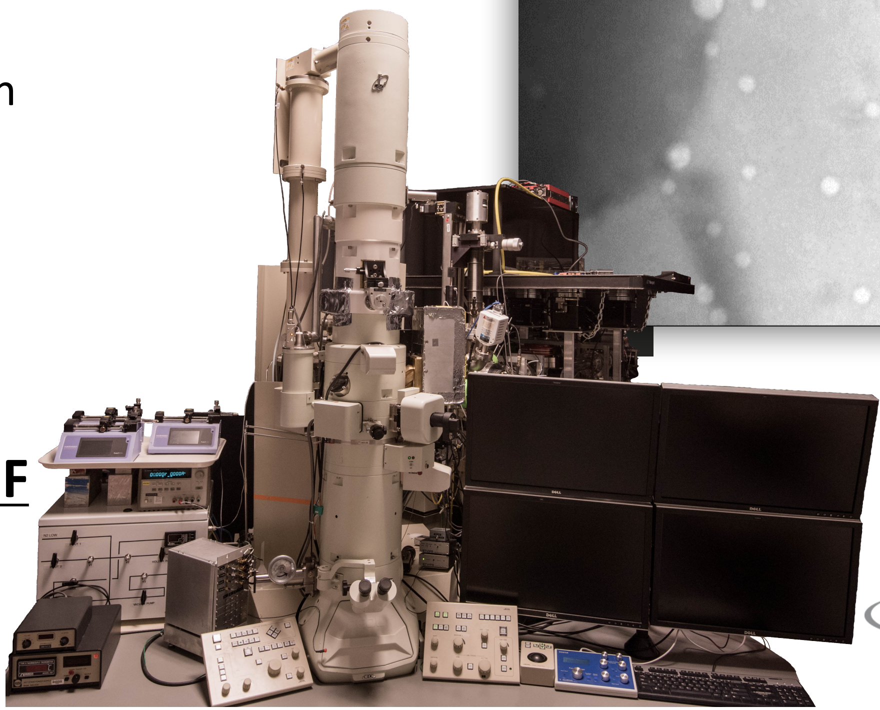


- At 400C, entire bubbles are observed migrating to the GB, promoting cavity growth at GBs.
- Intragranular growth is first observed at this temperature due to He or small He-V cluster diffusion from the lattice.
- At 450C, cavity migration occurs everywhere, eventually cavities become faceted and larger at GBs.

Summary

- Sandia's I3TEM unique CINT and NSUF facility
- Potential for wealth of new *in situ ion irradiation* capabilities
- *In situ* high energy ion irradiation to Au
- > 10 TEM stages with various capabilities
  - Mechanical to Liquid Cell microscopy feasible
- **<u>Facility accessible via NSUF and CINT via user proposals</u>**





Sandia's I^{3}TEM provides a wealth of interesting observations across various complex environments