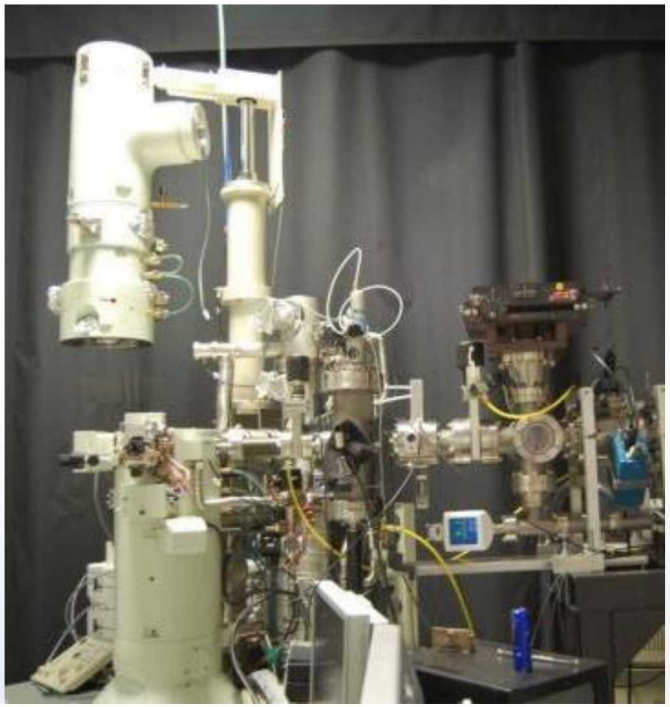
Exploring the Response of Materials to Radiation with *In situ* Microscopy

SAND2016-9693PE

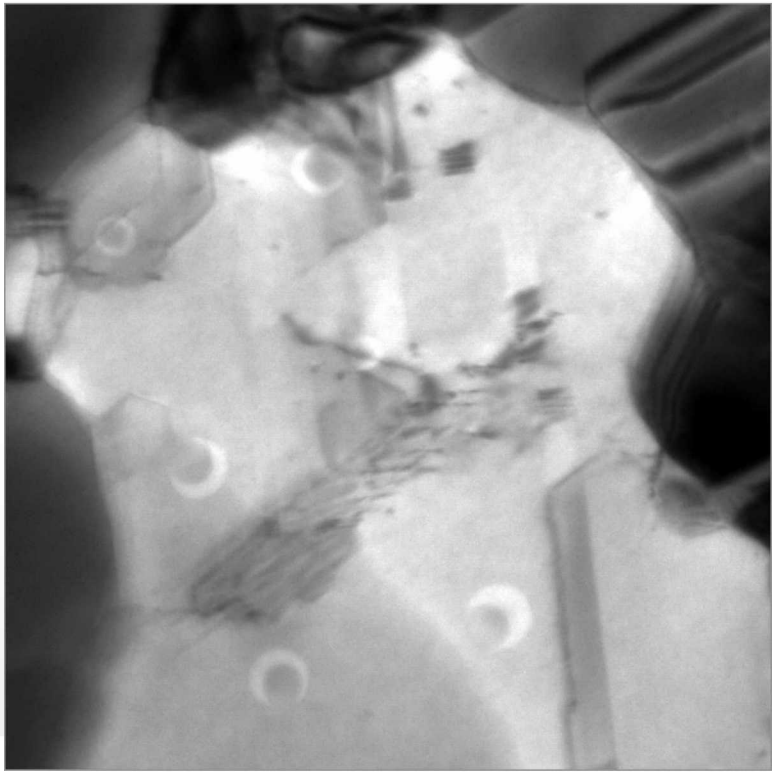
K. Hattar

Ion Beam Lab at Sandia National Laboratories

September 26, 2016



***In situ* TEM microscopy has recently undergone significant growth providing capabilities to investigate the structural evolution that occurs due to various extreme environments and combinations thereof**

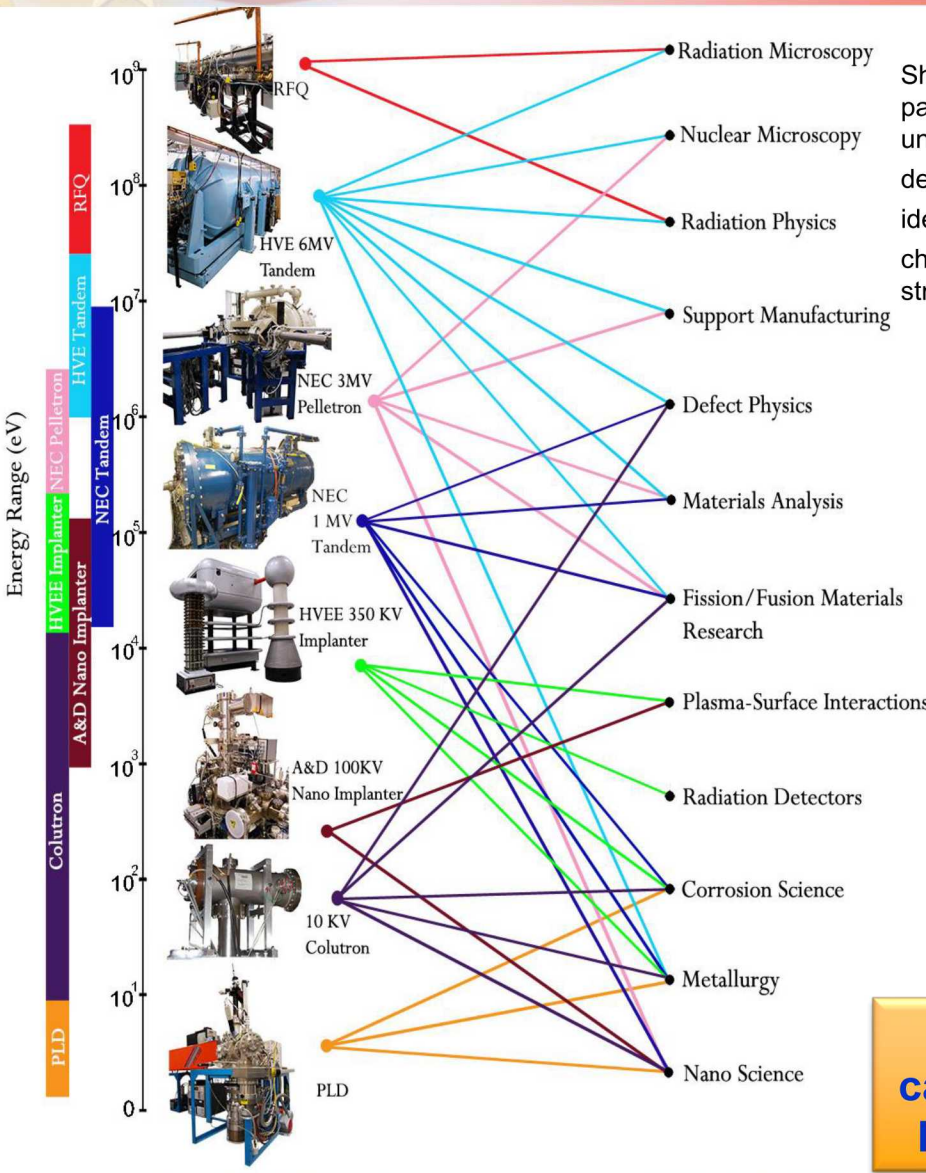


Collaborators:

- IBL: D.C. Bufford, D. Buller, C. Chisholm, B.G. Clark, J. Villone, G. Vizkelethy, B.L. Doyle, S. H. Pratt, & M.T. Marshall
- Sandia: B. Boyce, T.J. Boyle, P.J. Cappillino, J.A. Scott, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, E. Carnes, J. Brinker, D. Sasaki, J.A. Sharon, T. Nenoff, W.M. Mook, B.R. Muntifering, P. Feng, F.P. Doty, B.A. Hernandez-Sanchez, P. Yang, J-E Mogonye, S.V. Prasad , P. Kotula, & C. Snow
- External: A. Minor, L.R. Parent, I. Arslan, H. Bei, E.P. George, P. Hosemann, D. Gross, J. Kacher, & I.M. Robertson

This work was supported by the US Department of Energy, Office of Basic Energy Sciences.

Sandia's Ion Beam Laboratory

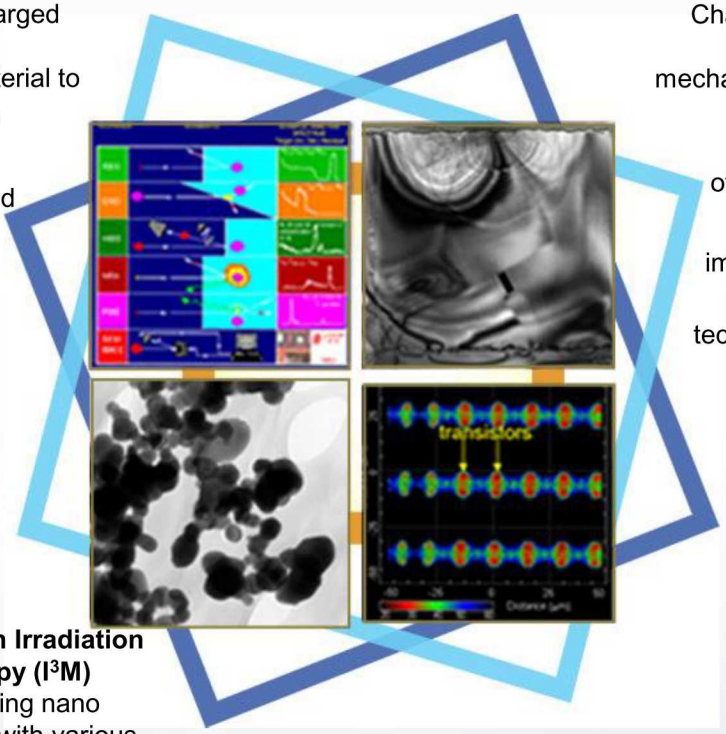


Ion Beam Analysis (IBA)

Shootinga charged particle at an unknown material to determine it's identity, local chemistry, and structure.

Ion Beam Modification (IBM)

Changing the optical, mechanical, and chemical properties of materials via ion implantation to meet technological needs



***In Situ* Ion Irradiation Microscopy (I^3M)**

Bombarding nano samples with various particles and observing the changes in real time to understand how materials will behave in extreme environments.

Radiation Effects Microscopy (REM)

Using ion emissions to determine the Radiation hardness of microelectronics, identifying potential weaknesses.

The IBL has a unique and comprehensive capability ion beam set including and *In situ* Ion Irradiation Transmission Electron Microscopy.

Benefits & Limitations of *in situ* TEM

Benefits

1. **Real-time nanoscale resolution observations of microstructural dynamics**

Limitations

1. **Predominantly limited to microstructural characterization**
   - Some work in thermal, optical, and mechanical properties
2. **Limited to electron transparent films**
   - Can often prefer surface mechanisms to bulk mechanisms
   - Local stresses state in the sample is difficult to predict
3. **Electron beam effects**
   - Radiolysis and Knock-on Damage
4. **Vacuum conditions**
   - 10^{-7} Torr limits gas and liquid experiments feasibility
5. **Local probing**
   - Portions of the world study is small

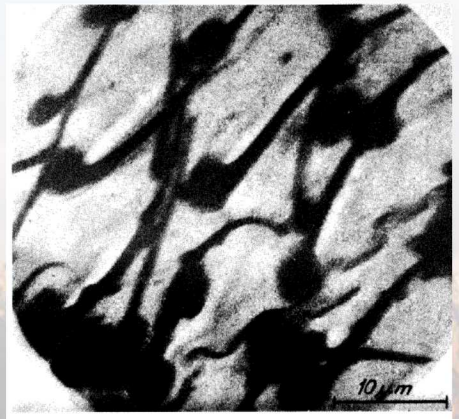


Fig. 6: Wing surface of the house fly.
(First internal photography, U = 60 kV, M_o = 2200)
(Driest, E., and Müller, H.O.: Z. Wiss. Mikroskopie 52, 53-57 (1935)

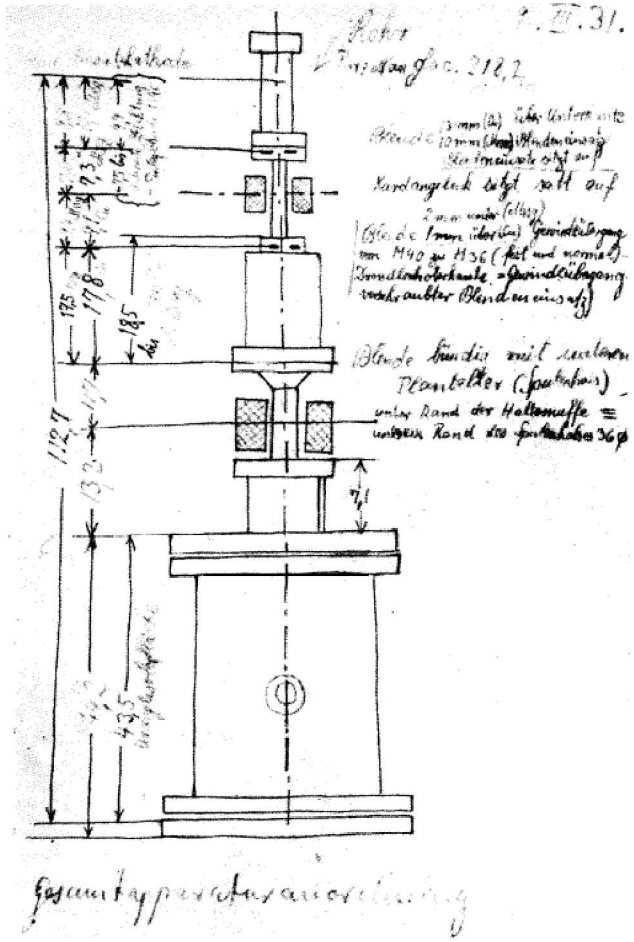
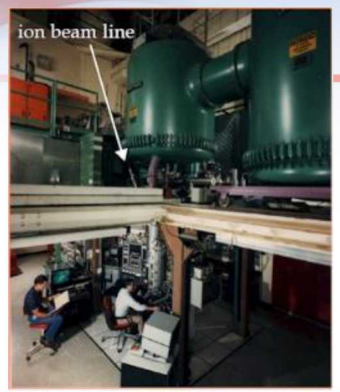
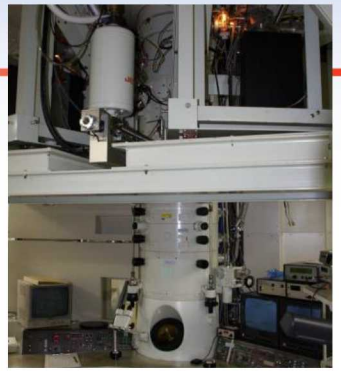


Fig. 2: Sketch by the author (9 March 1931) of the cathode ray tube for testing one-stage and two-stage electron-optical imaging by means of two magnetic electron lenses (electron microscope) [8].

History of *In situ* Ion Irradiation TEM



Courtesy of: J. Hinks



1930s… | 1960s | 1970s | 1980s | 1990s | 2000s | 2010s

1931
The invention of the TEM

1961
O^- emission reported from a TEM filament by Pashley, Presland, and Meneter at TI Labs, Cambridge, UK

1968
First TEM beamline combination by Thackery, Nelson, and Sansom at AERE Harwell, UK

1976
First HVEM with ion irradiation at UVA, USA

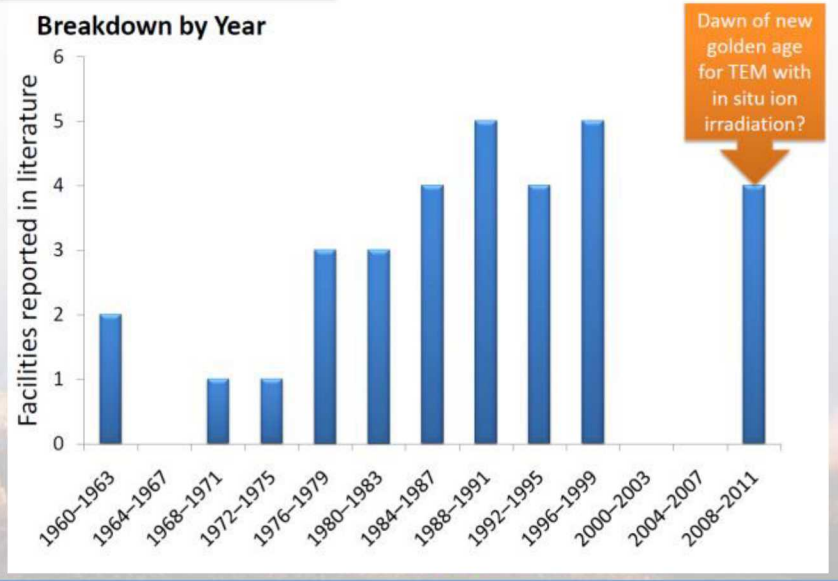
1978
First in-situ ion irradiation experiments at ANL

1990s
First dual beam system developed at JAERI and NIMS, Japan

Workshop on Ion Irradiation TEM
Huddersfield, UK (2008)
Albuquerque, USA (2011)
Saporro, Japan (2013)

"The direct observation of ion damage in the electron microscope thus represents a powerful means of studying radiation damage"

D.W. Pashley and A.E.B. Presland Phil Mag. 6(68) 1961 p. 1003



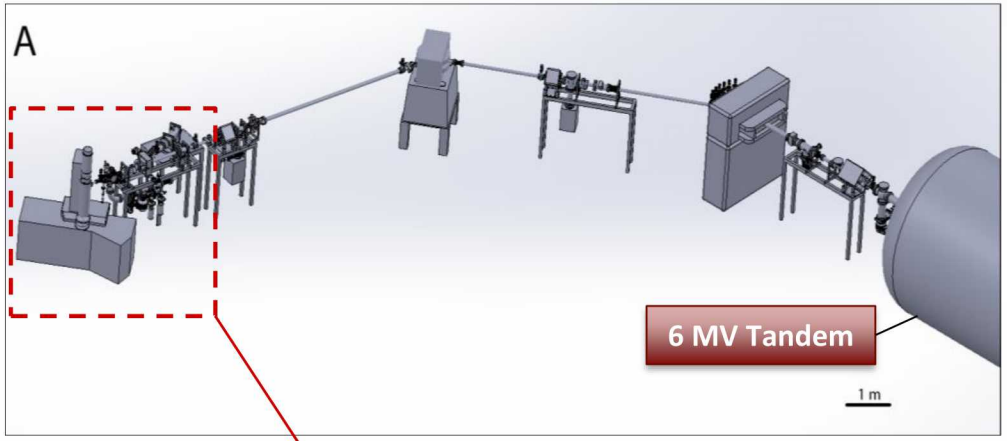
In situ Ion Irradiation TEM Facility

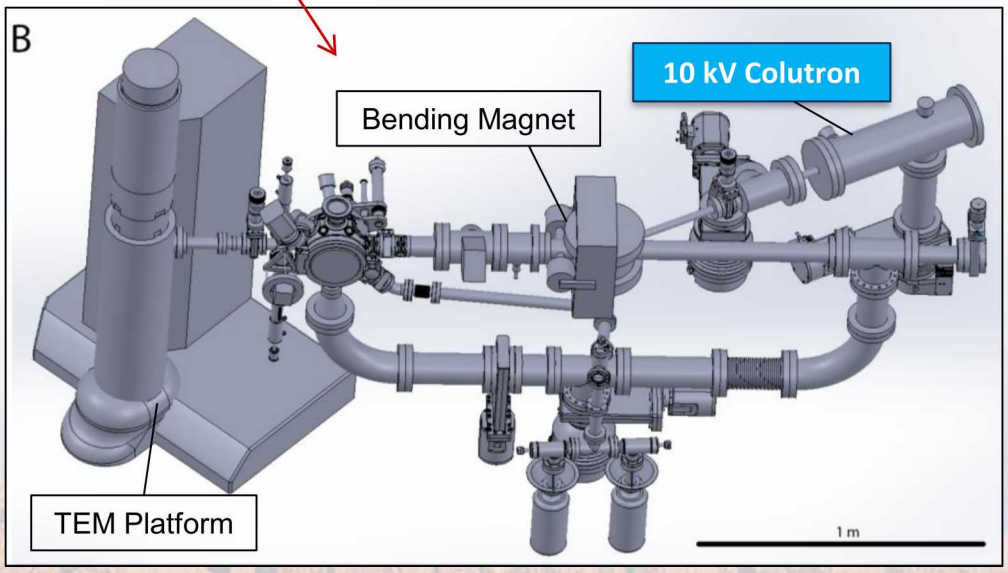
Proposed Capabilities

- 200 kV LaB_6 TEM
- Ion beams considered:
  - Range of Sputtered Ions
  - 10 keV D^{2+}
  - 10 keV He^{+}
- All beams hit same location

- Nanosecond time resolution (DTEM)
- Procession scanning (EBSD in TEM)
- *In situ* PL, CL, and IBIL

- *In situ* vapor phase stage
- *In situ* liquid mixing stage
- *In situ* heating
- Tomography stage (2x)
- *In situ* cooling stage
- *In situ* electrical bias stage
- *In situ* straining stage





Solid Works by: J. Davis

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Planned Development of the I^{3}TEM

Collaborators: D.L. Buller, B.G. Clark, D. Masiel, & B.L. Doyle

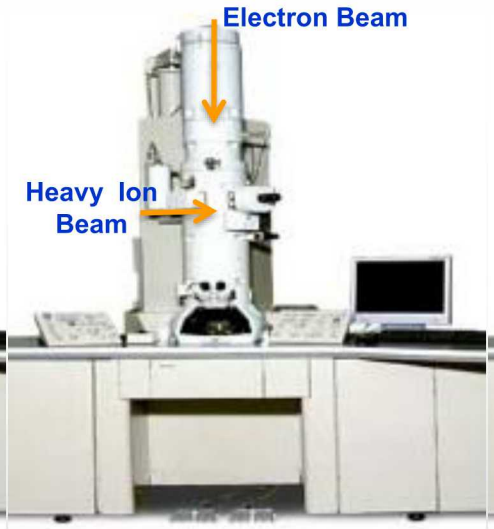
0th Generation

- 200 kV LaB_6 TEM
- *In situ* vapor phase stage
- *In situ* liquid mixing stage
- Simple tomography holder

Delivery October 29, 2010 &
Accepted January 31, 2011

1st Generation

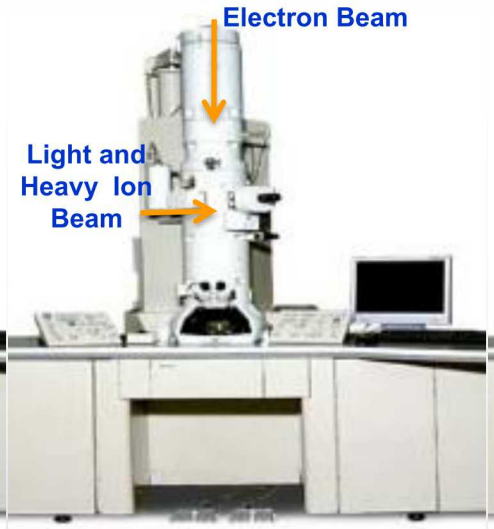
- Insertion of collimated Tandem ion beam at 0-9°
- Vacuum, electrical, and mechanical Isolation
- Adequate dosimetry



First Tandem beam into TEM
April 19, 2011

2nd Generation

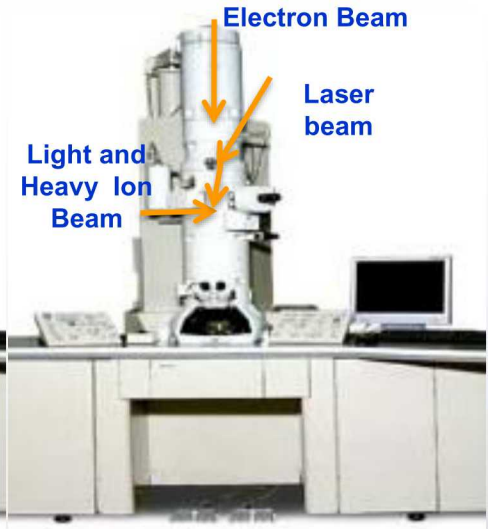
- Concurrent heavy and light ion implantation
- A Colutron ion beam will be bent in line with the Tandem beam.



First Colutron beam into TEM
October 3, 2012

3rd Generation

- Plan-view laser exposure and DTEM capabilities
- Provide ion and laser excitation sources with nanosecond timing



Optical port completed
Laser optics are being designed

Each facility addition will expand potential capabilities

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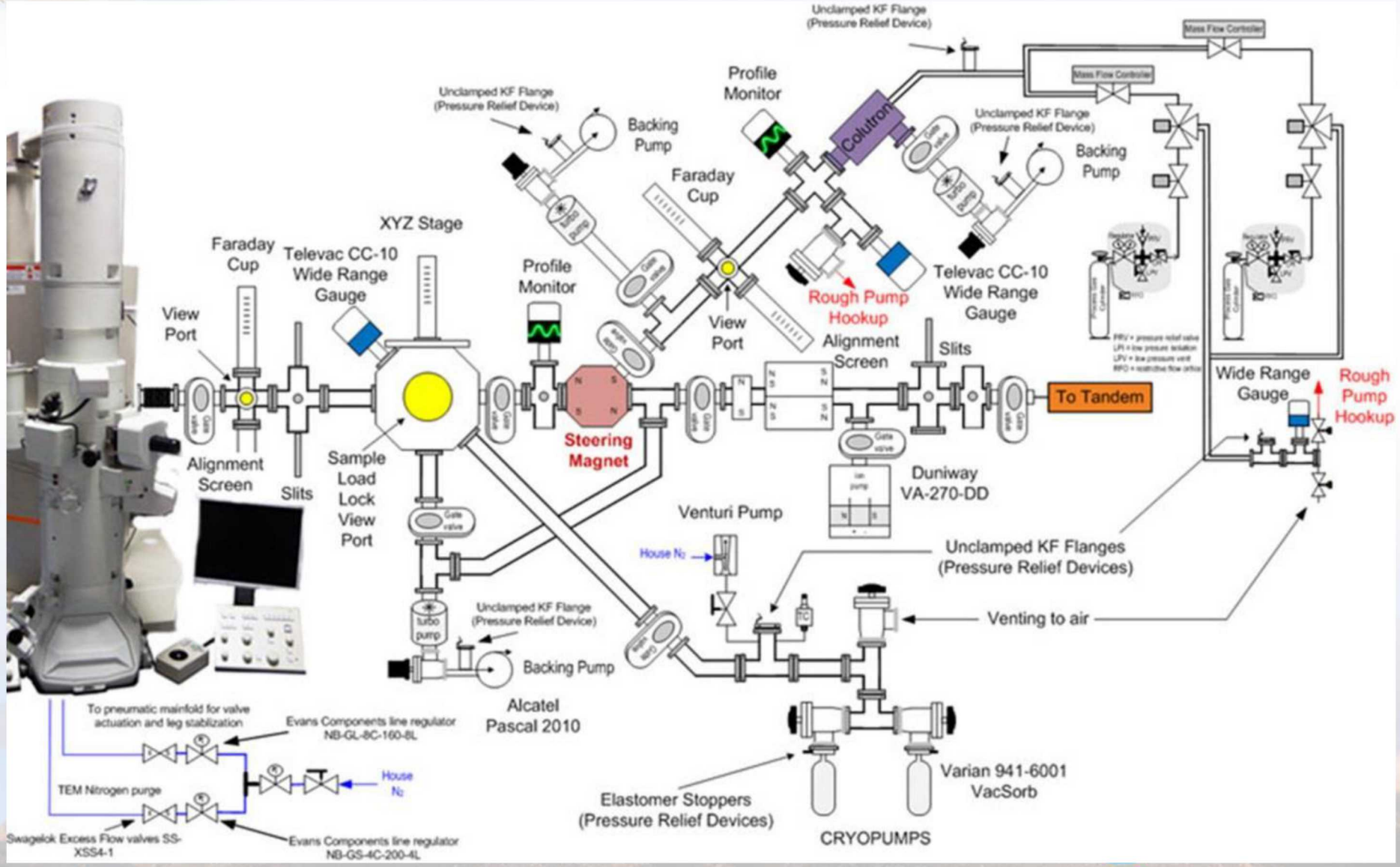
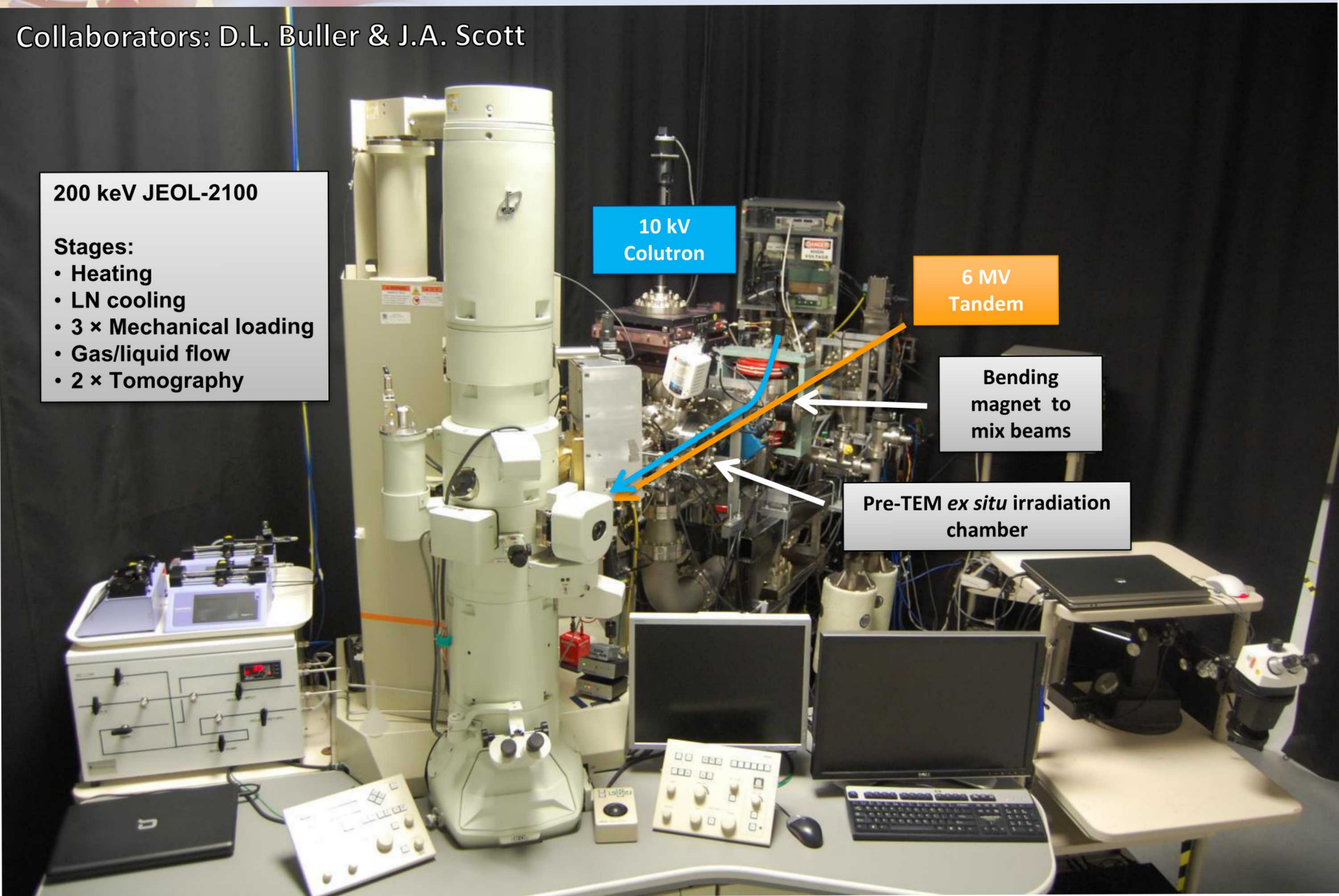
Schematic of the *In situ* TEM Beamline

Collaborators: M.T. Marshall J.A. Scott, & D.L. Buller



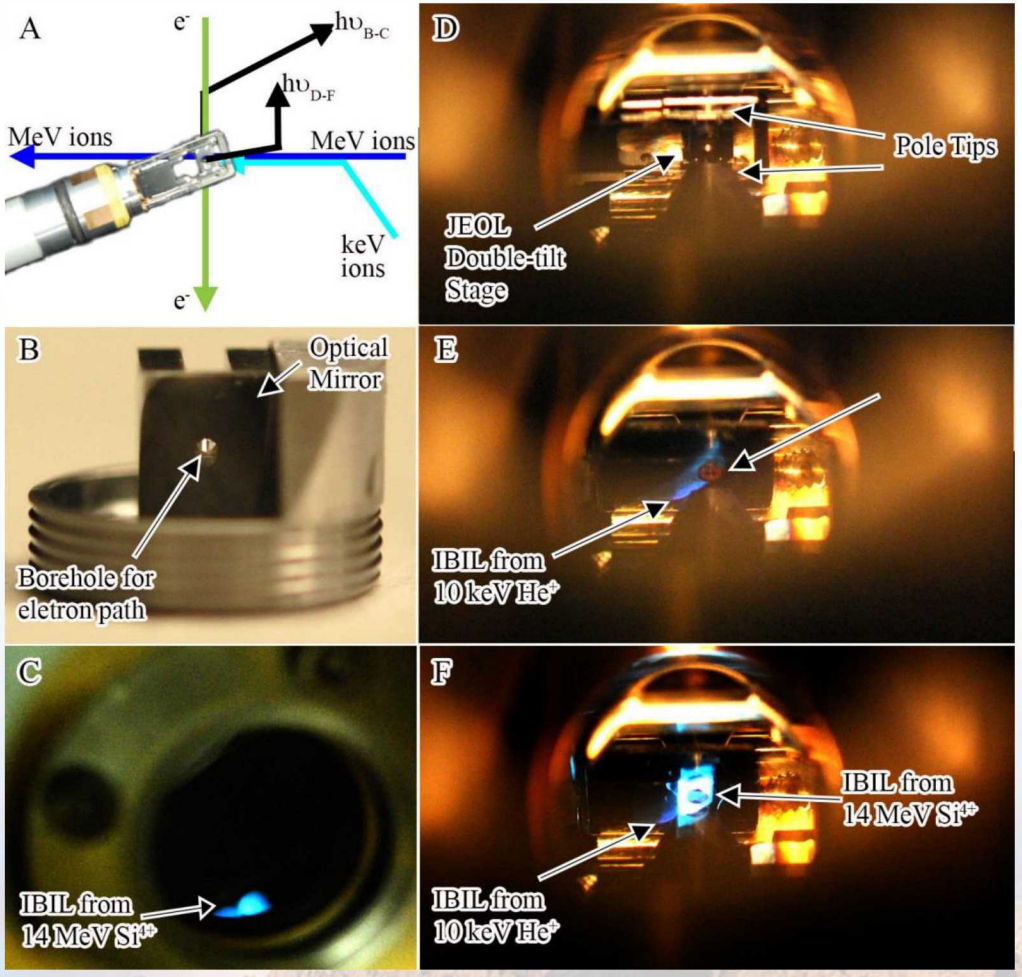
TEM Facility

Collaborators: D.L. Buller & J.A. Scott



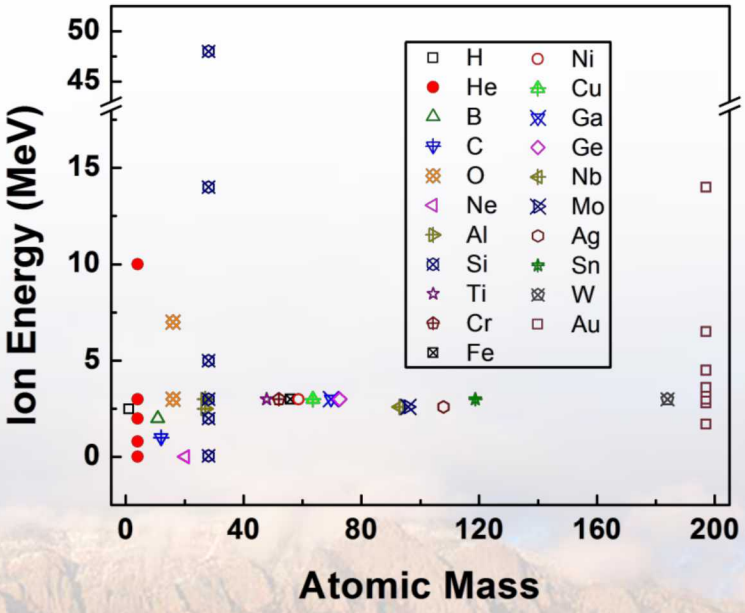
Visualizing Ion Beams in the TEM

IBIL from a quartz stage inside the TEM



Ion beam alignment at the sample confirmed using ion beam-induced luminescence

Ion species & energy introduced into the TEM



Hattar, et al, under review 2014.

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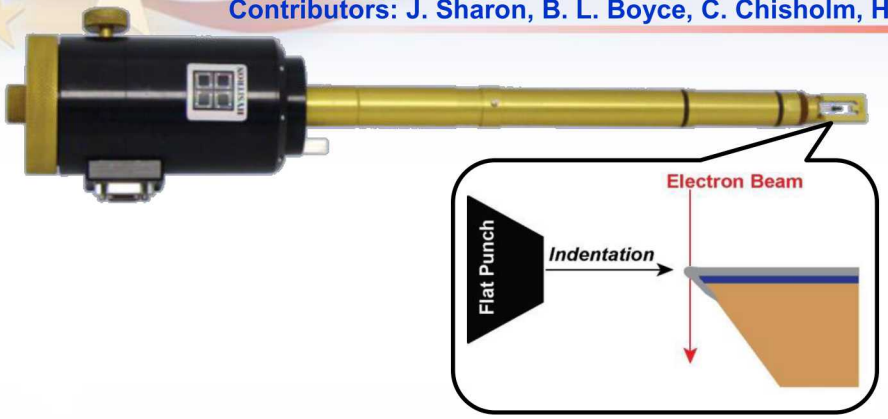
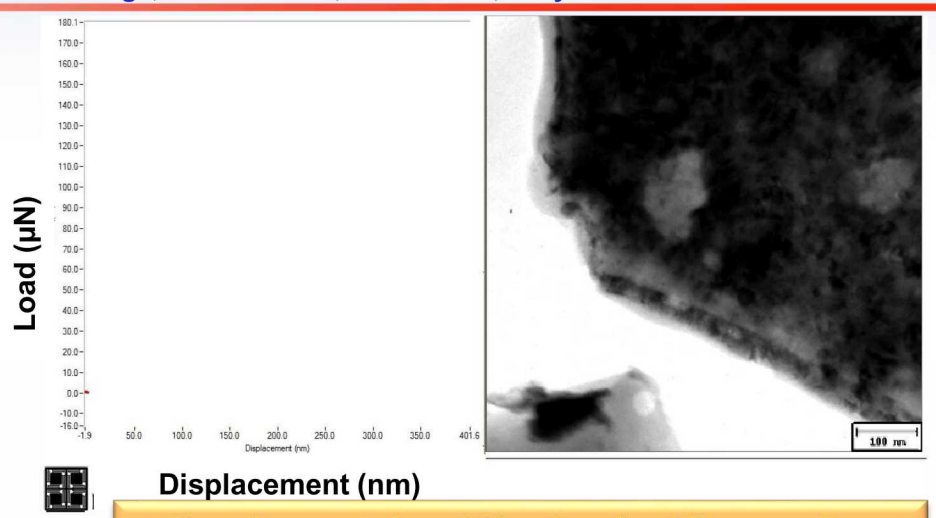
Next Steps: *In situ* TEM Quantitative Mechanical Testing

Contributors: J. Sharon, B. L. Boyce, C. Chisholm, H. Bei, E.P. George, P. Hosemann, A.M. Minor, & Hysitron Inc.

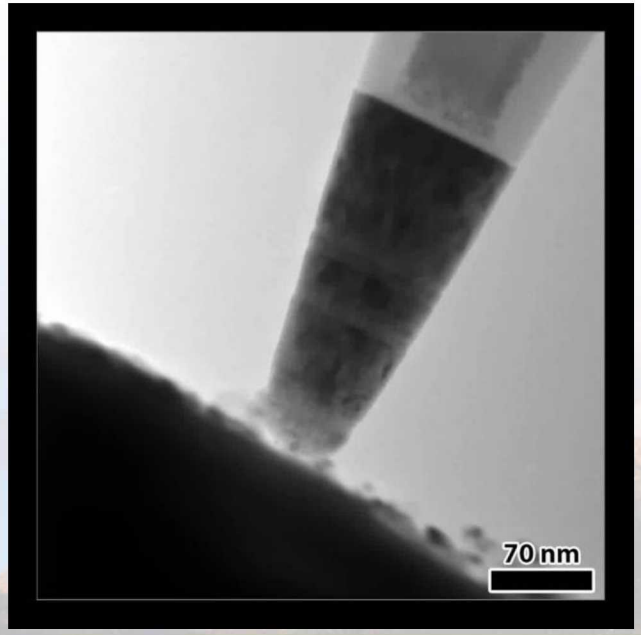


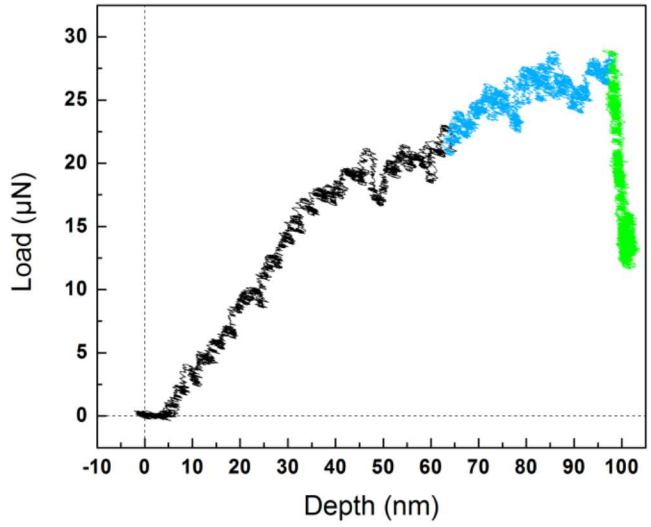


Range of Mechanical Testing Techniques

- Indentation
- Compression
- Tension
- Wear
- Fatigue
- Creep

Fundamentals of Mechanical Properties

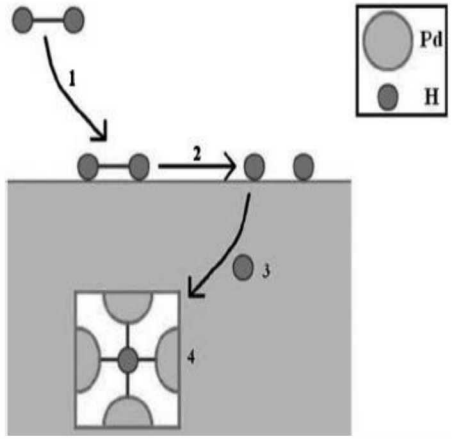




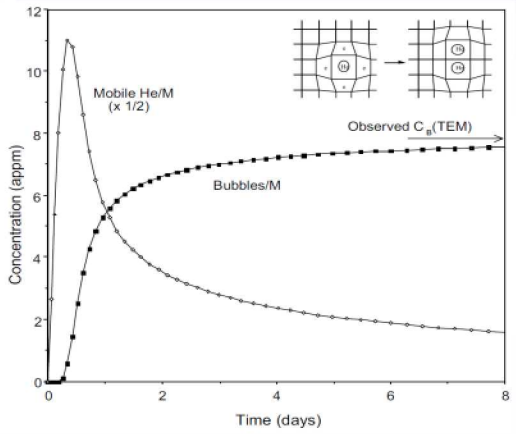
- 0.5 nm/s loading rate
- Trapezoid load function
- 60s load/60s hold/60s unload

Can *In situ* TEM Address Hydrogen Storage Concerns in Extreme Environments?

Contributors: B.G. Clark, P.J. Cappillino, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, L.R. Parent, I. Arslan. & Protochips, Inc.



R. Delmelle, J., Phys. Chem. Chem. Phys. (2011) p.11412



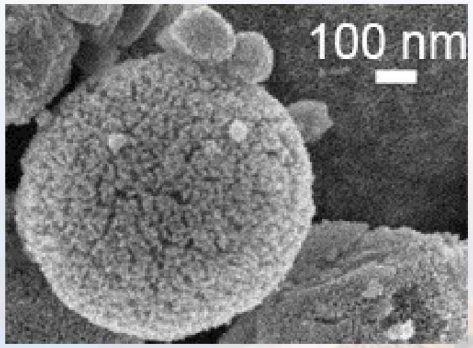
Cowgill, D., Fusion Sci. & Tech., 28 (2005) p. 539
Trinkaus, H. *et al.*, JNM (2003) p. 229
Thiebaut, S. *et al.* JNM (2000) p. 217

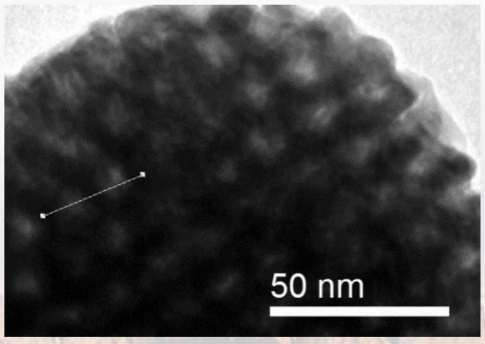
Vapor-Phase Heating TEM Stage

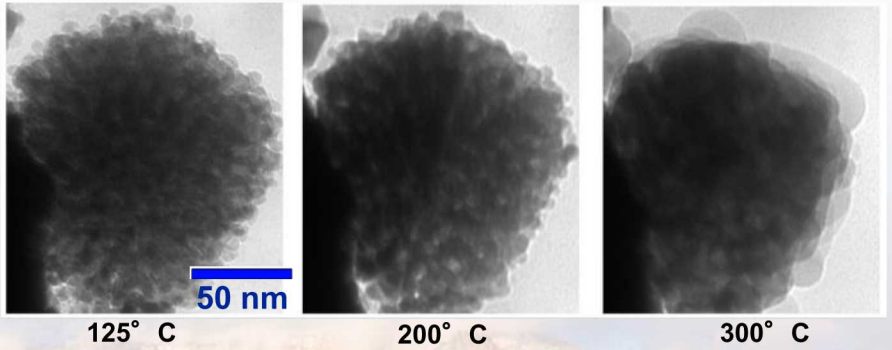
- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Compatible with MS and other analytical tools

- 1 atm H_2 after several pulses to specified temp.

Harmful effects may be mitigated in nanoporous Pd



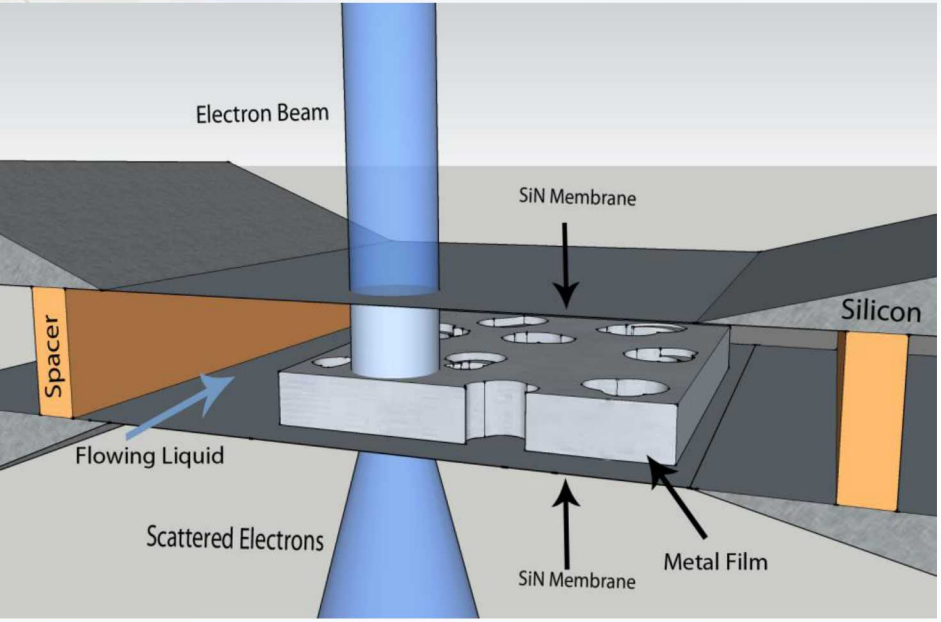




New *in situ* atmospheric heating experiments provide great insight into nanoporous Pd stability

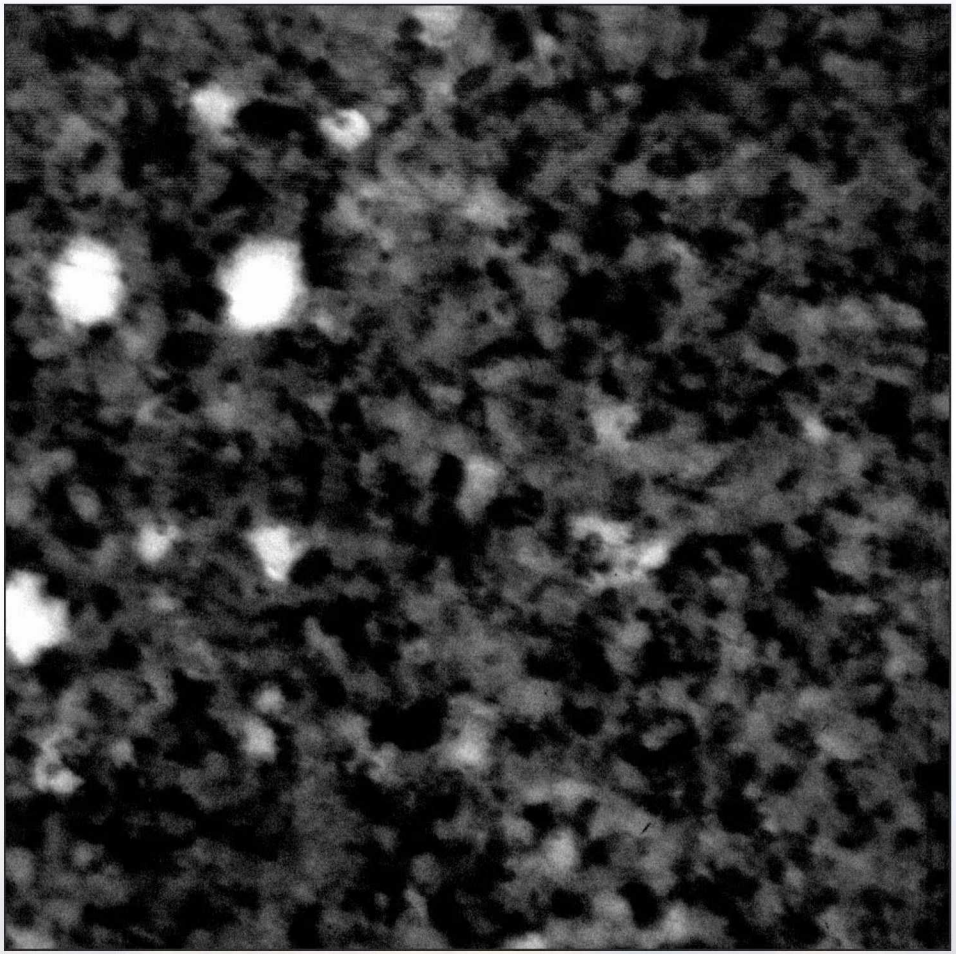
In situ TEM Corrosion

Contributors: D. Gross, J. Kacher, & I.M. Robertson



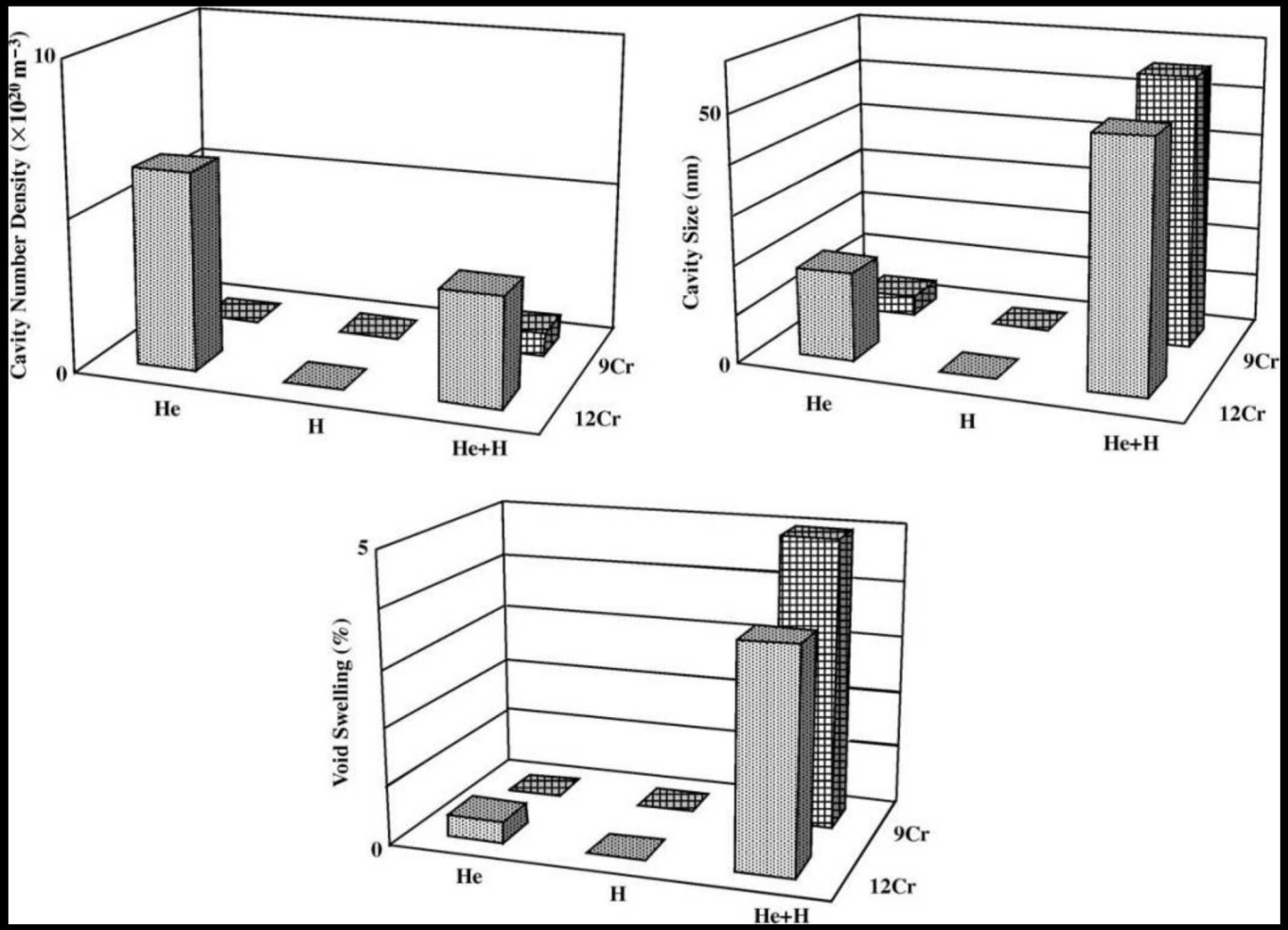
Microfluidic Stage

- Mixing of two or more channels
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Films can be directly deposited on the electron transparent SiN membrane



Pitting mechanisms during dilute flow of acetic acid over 99.95% nc-PLD Fe involves many grains.

H, He, and Displacement Damage Synergy



T. Tanaka et al. "Synergistic effect of helium and hydrogen for defect evolution under milt-ion irradiation of Fe-Cr ferritic alloys"
J. of Nuclear Materials 329-333 (2004) 294-298

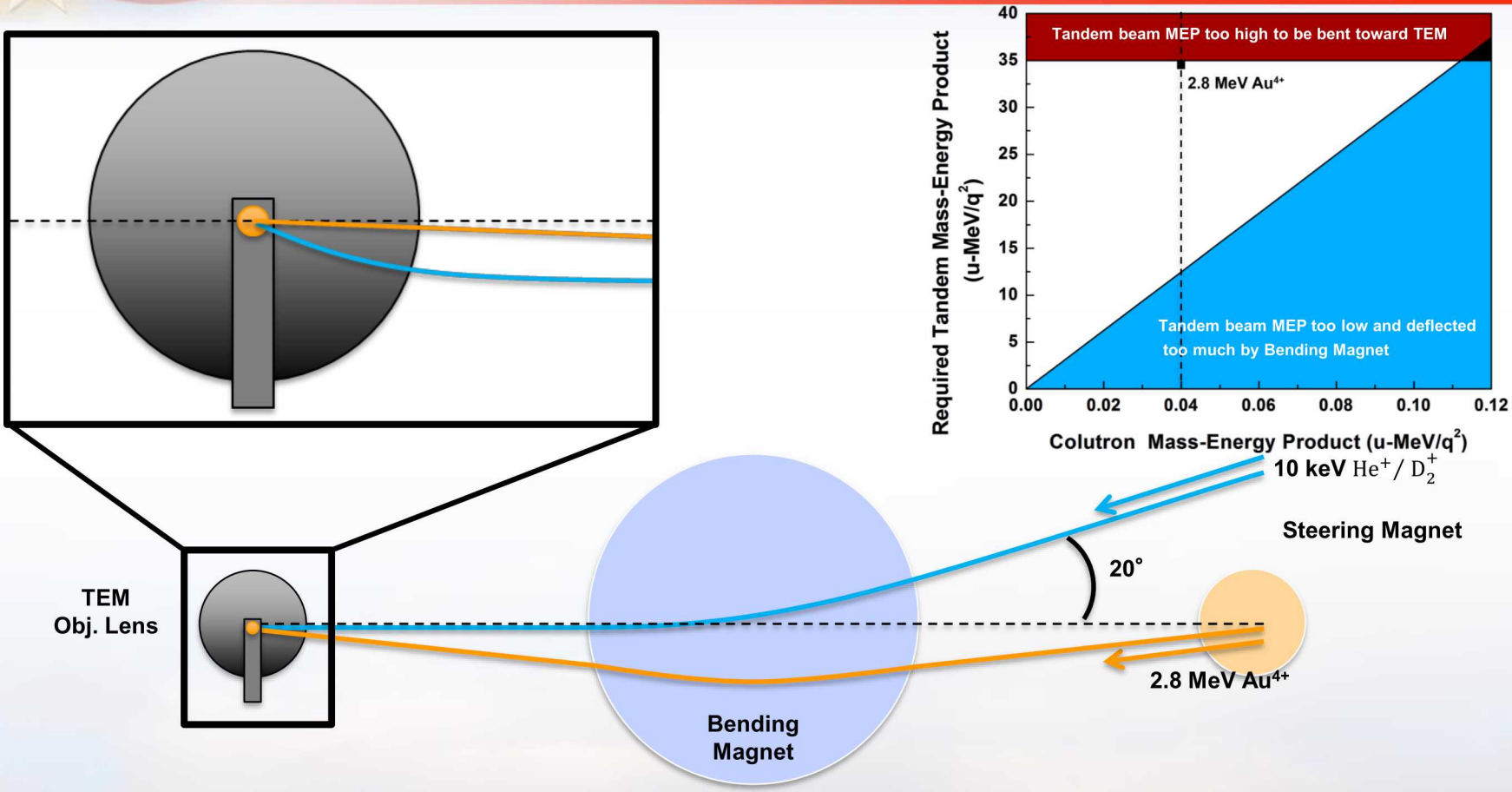
Coupling Effect

- H and He are produced as decay products
- The relationship between the point defects present, the interstitial hydrogen, and the He bubbles in the system that results in the increased void swelling has only been theorized.
- The mechanisms which governs the increased void swelling under the presence of He and H have never been experimental determined

Difficulty of performing triple-beam irradiation has resulted in a limited number of facilities world wide

Modeling Beam Mixing and Deflection

Collaborators: M. Steckbeck, D.C. Bufford, & B.L. Doyle

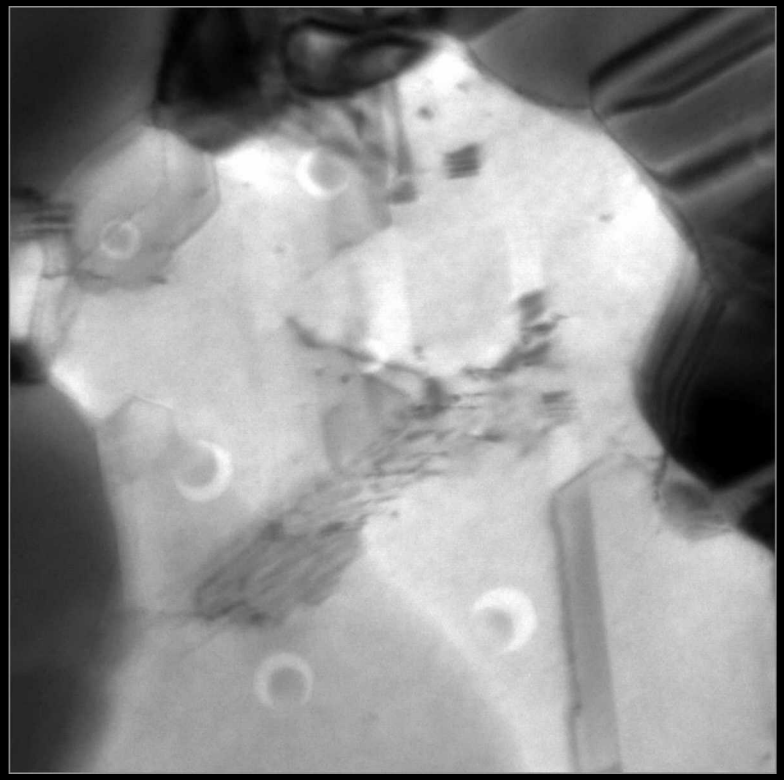


- Must compensate for deflection of Tandem beam by bending magnet Colutron beams deflected by the TEM objective lens
- Insignificant deflection of Tandem beams
- With 10 keV He/D_2 we can use Tandem beams ≳13 MeV/q^2
- Au, He, and D_2 ions all reach the sample concurrently

Single Ion Strikes

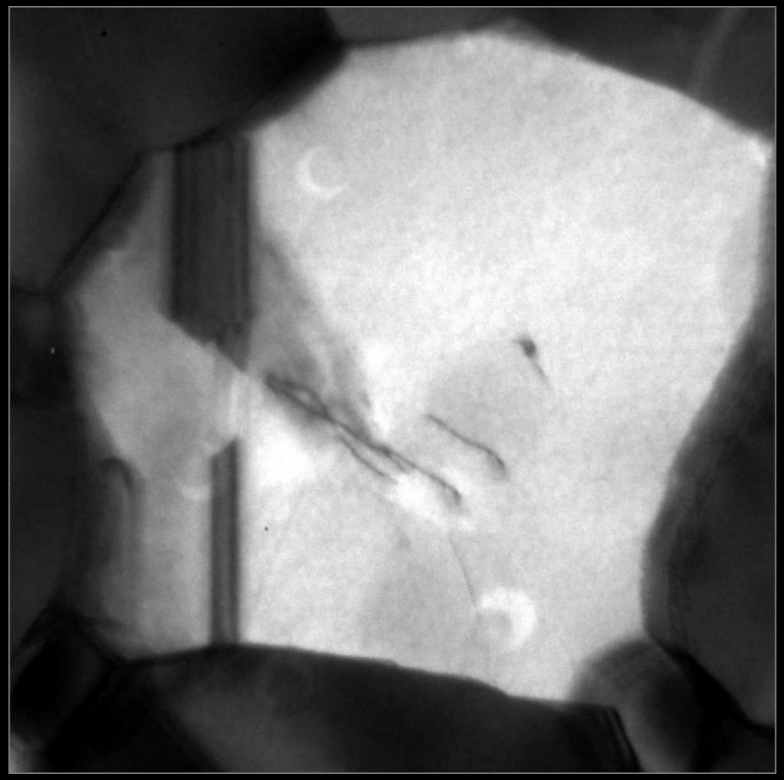
Collaborators: C. Chisholm , P. Hosemann, & A. Minor

7.9×10^9 ions/cm^2/s



vs

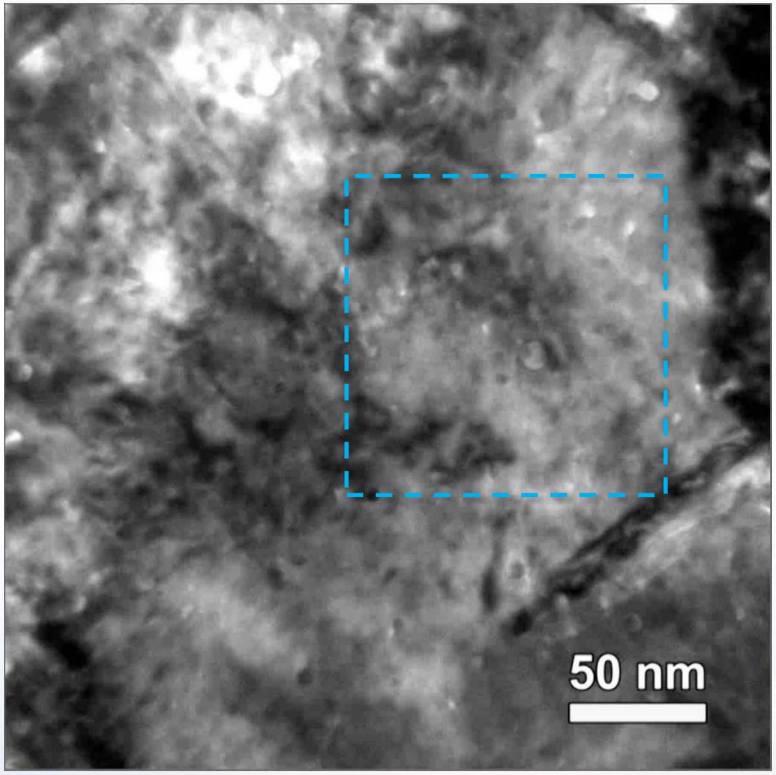
6.7×10^7 ions/cm^2/s

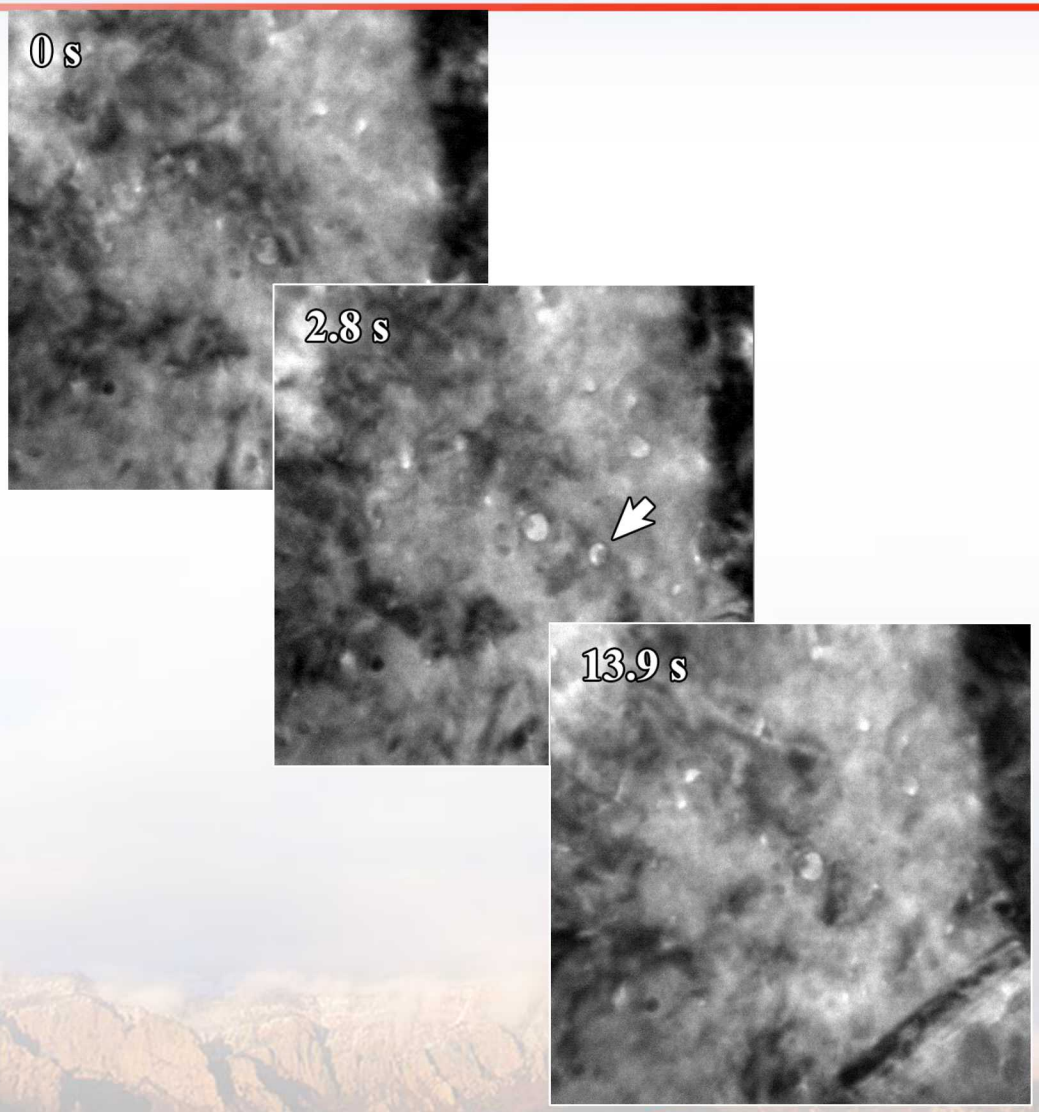


Improved vibrational and ion beam stability permits us to work at 120kx or higher permitting imaging of single cascade events

2.8 MeV Au^{4+} + 10 keV He^{+}/D_2^{+}

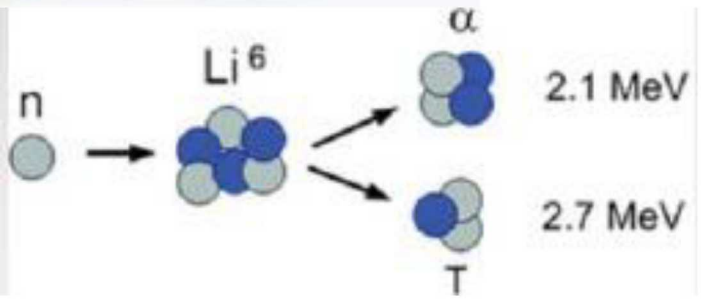
Video playback speed x1.5.



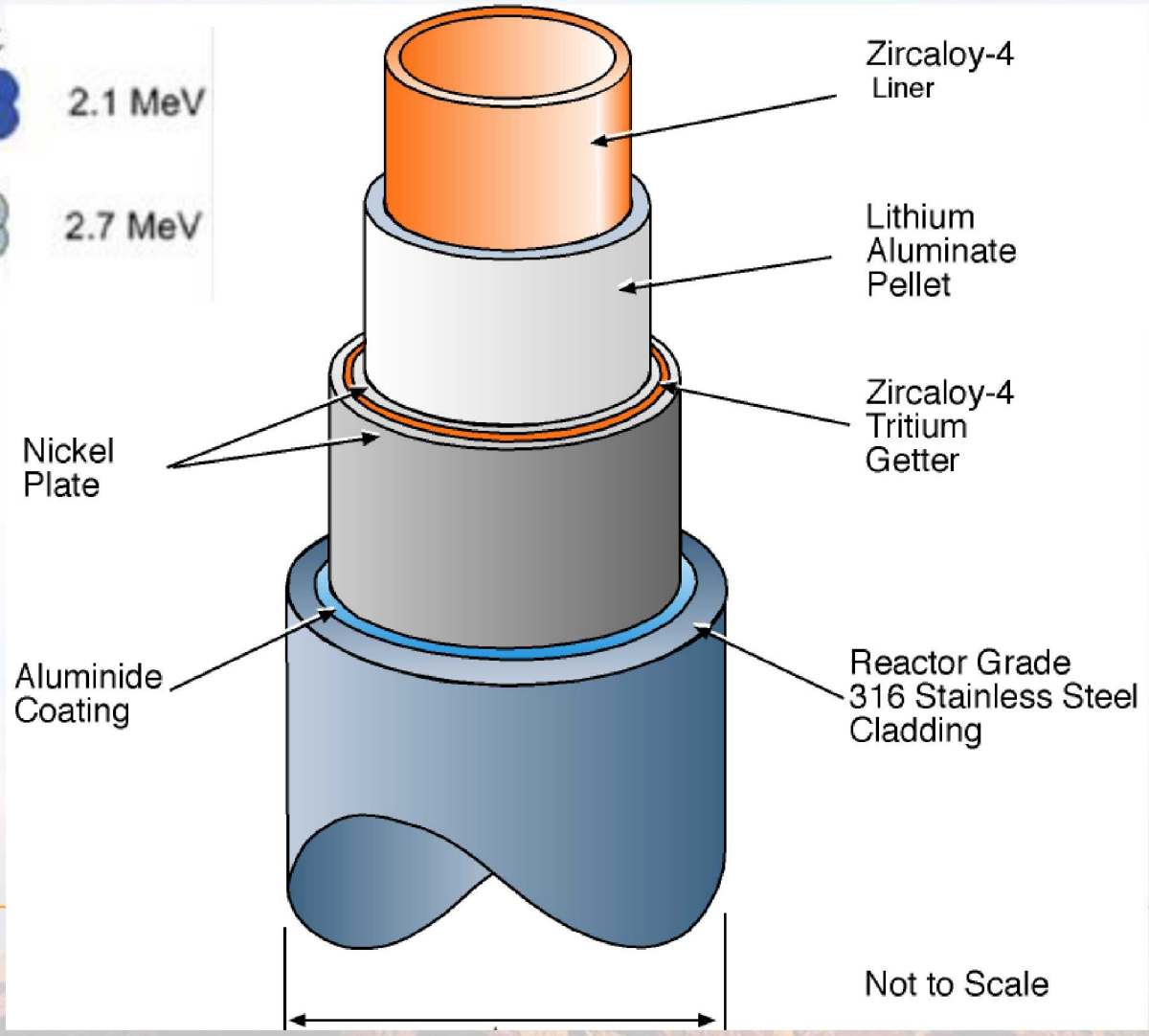


- **Approximate fluence:**
  - Au 1.2 $\times 10^{13}$ ions/cm^2
  - He 1.3 $\times 10^{15}$ ions/cm^2
  - D 2.2 $\times 10^{15}$ ions/cm^2
- **Cavity nucleation and disappearance**

Tritium Producing Burnable Absorber Rod (TPBAR)



- Displacement Damage
- Helium Implantation
- Tritium Implantation
- Elevated Temperatures



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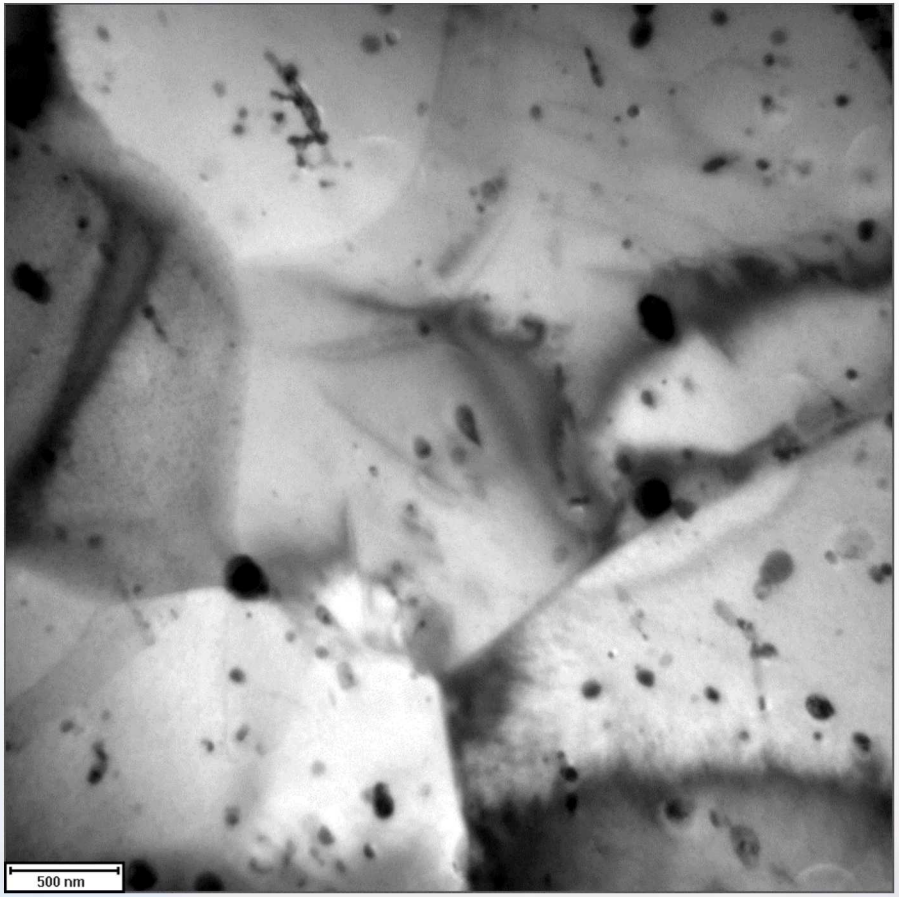
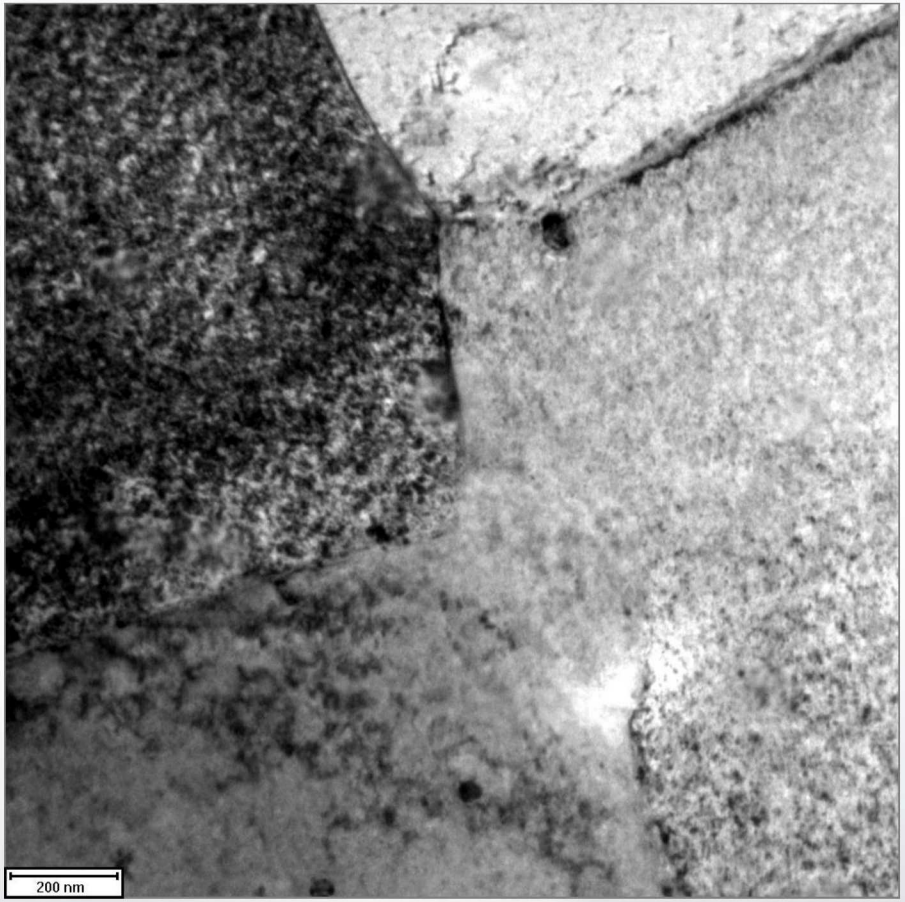
3 MeV Self Ion Irradiation at 310 C



Before Irradiation



After Irradiation
≈ 7 DPA

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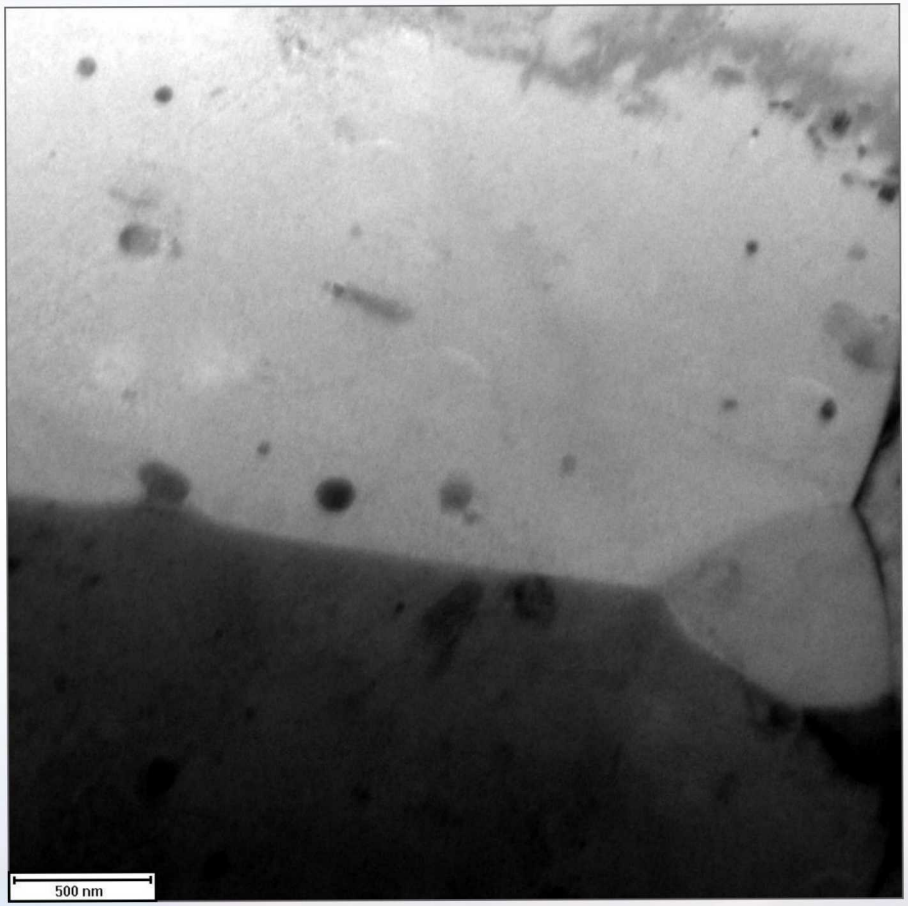
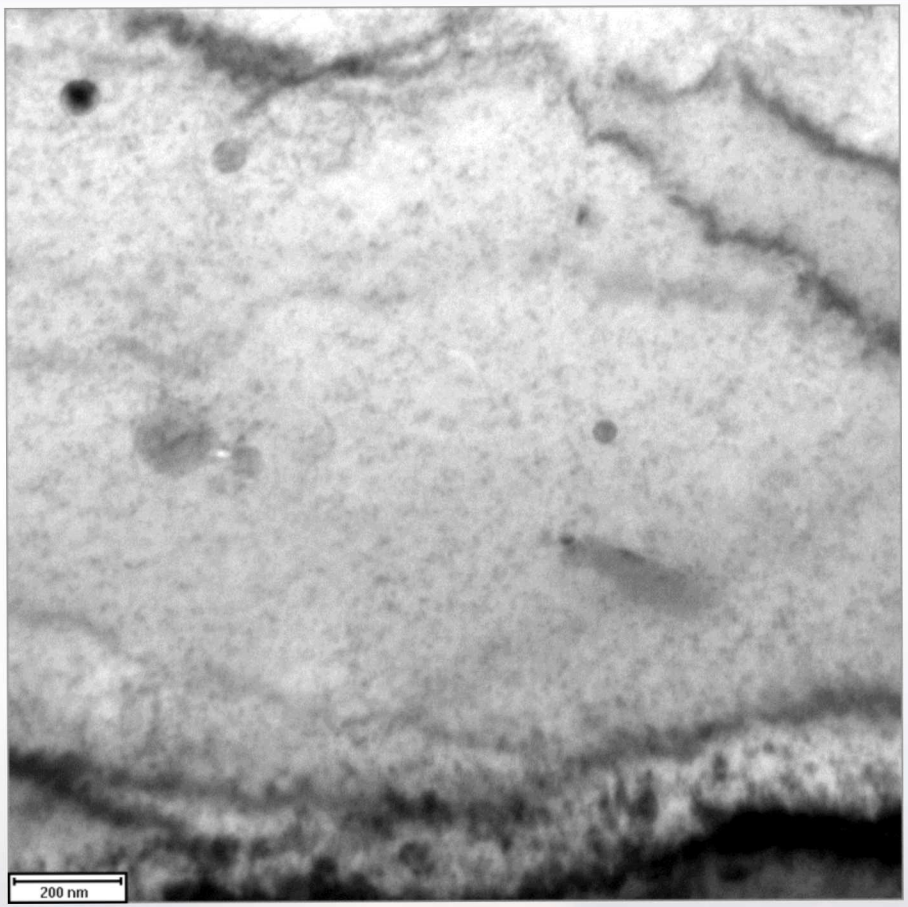
10 keV He^+ Implantation at 310 C

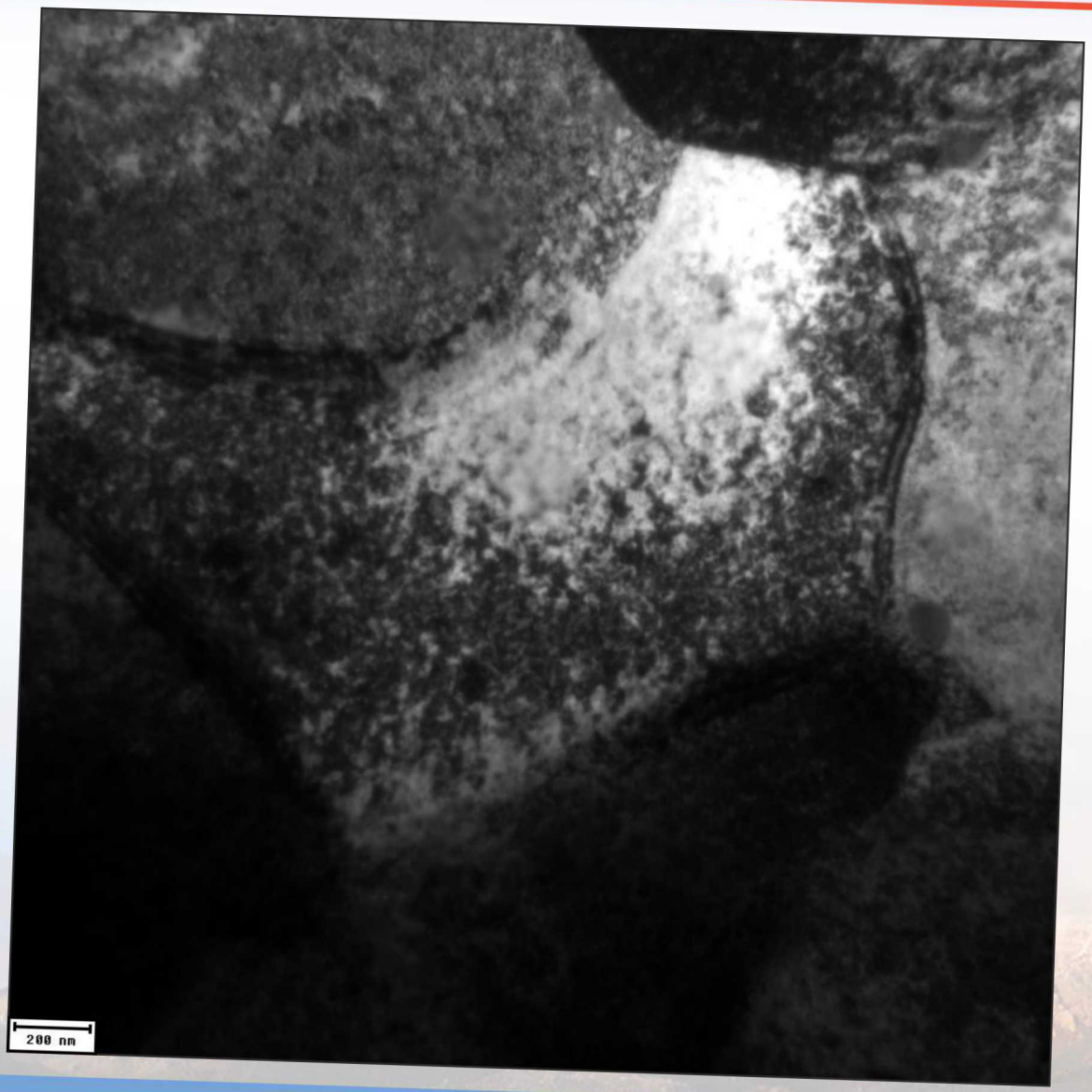


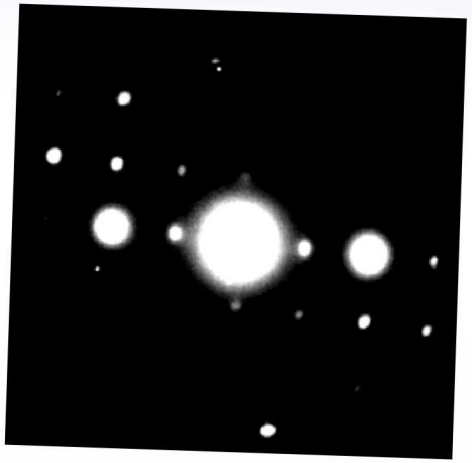
Before Implantation



After Implantation
Damage, No Cavities

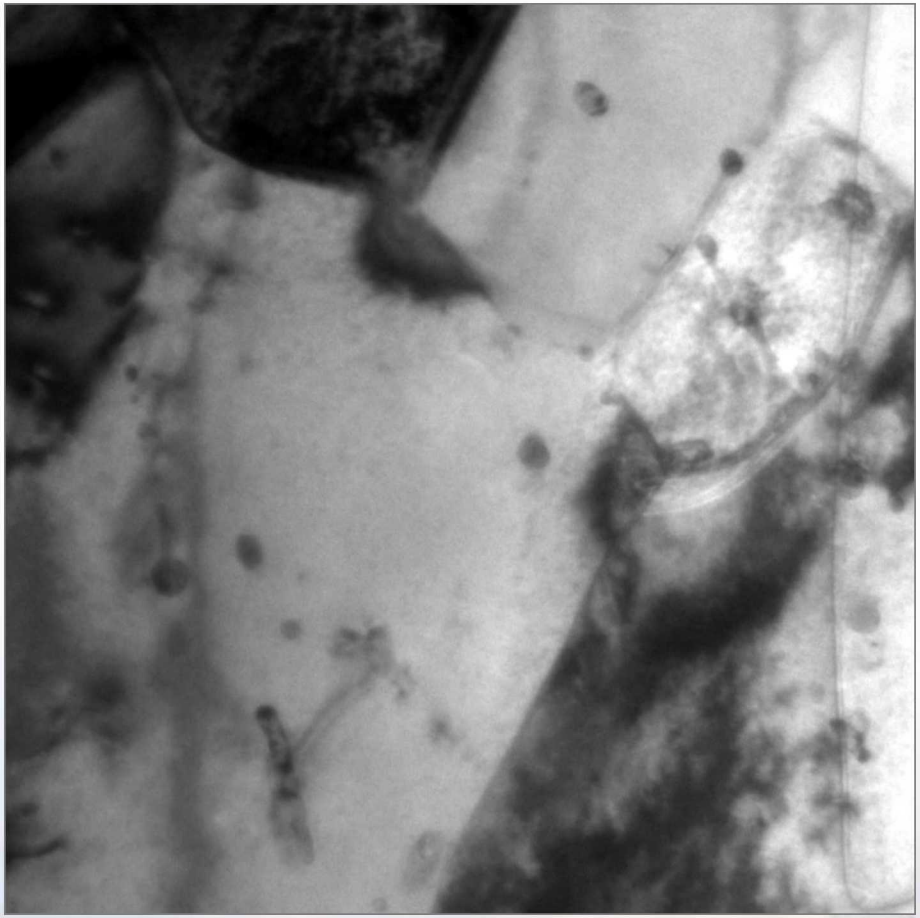
He^{+} Followed by Zr



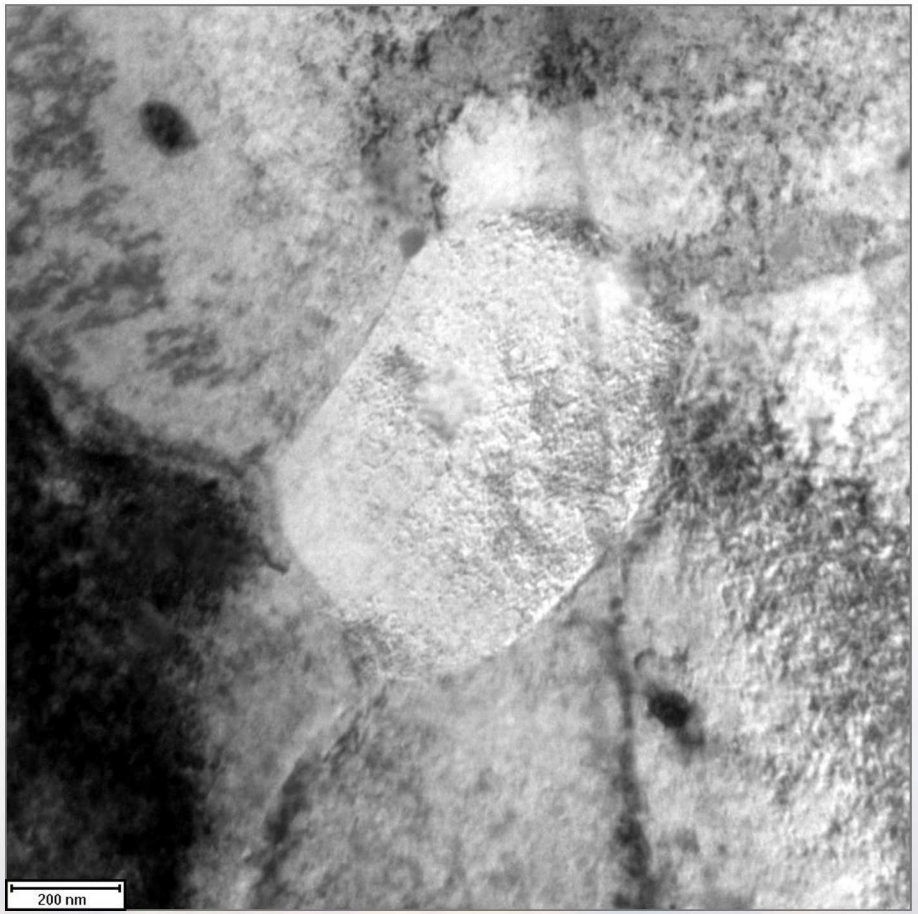


Two Beam
g = 0002

Concurrent He^+ Implantation and Self Ion Irradiation



Before Implantation/Irradiation



After Implantation/Irradiation
Damage, No Cavities

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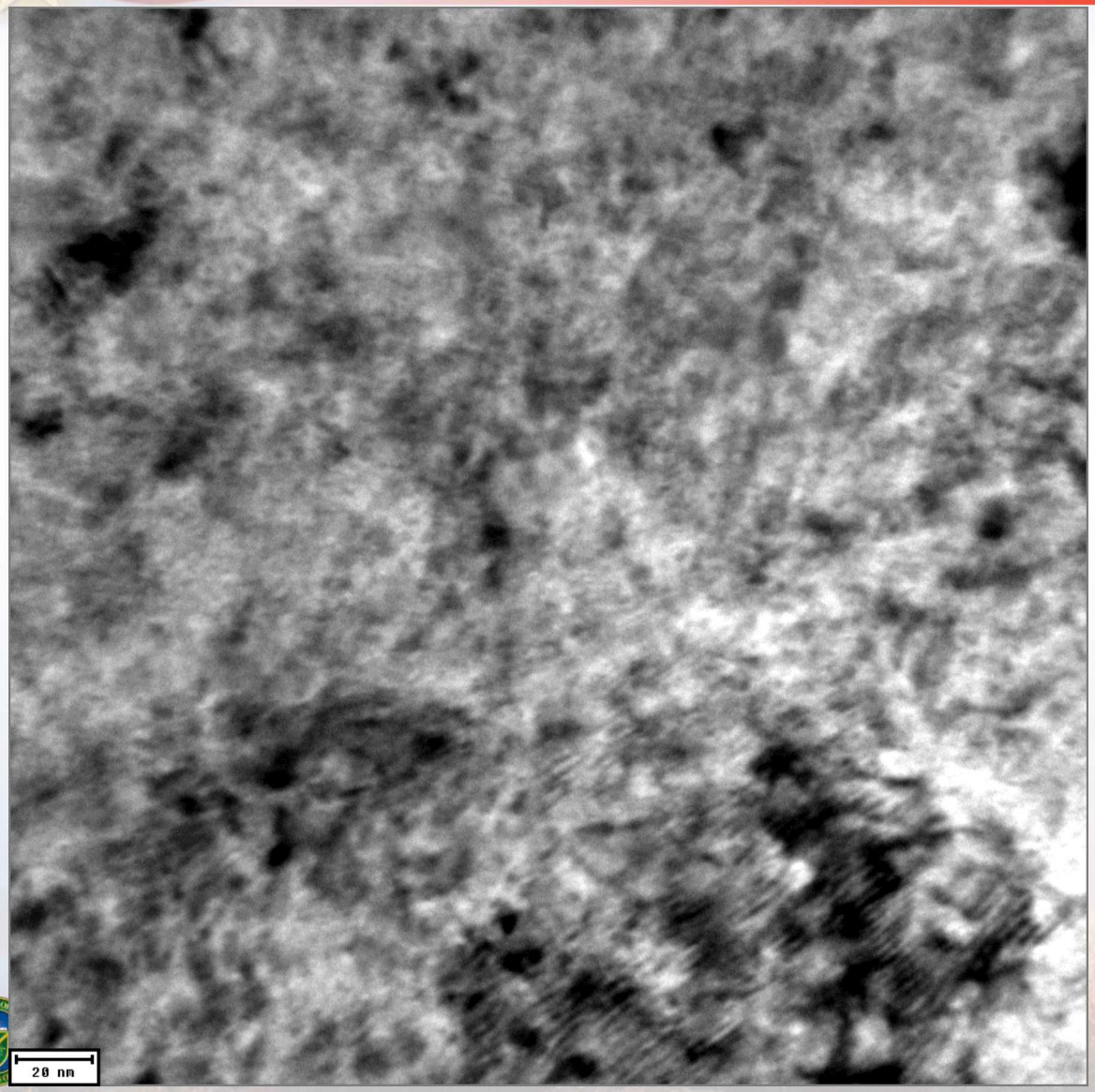
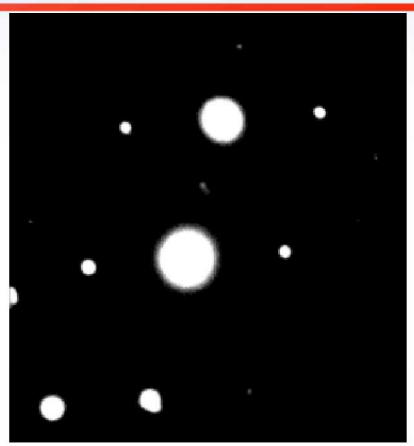
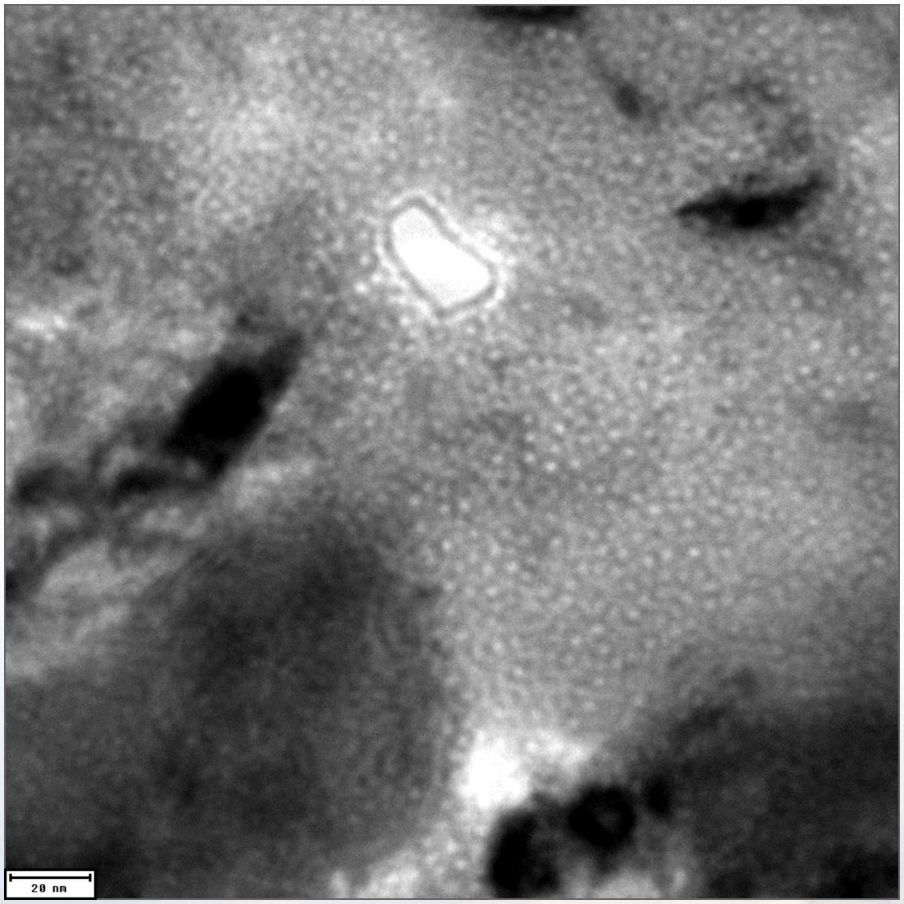
Concurrent D & He Implantation & Zr Irradiation



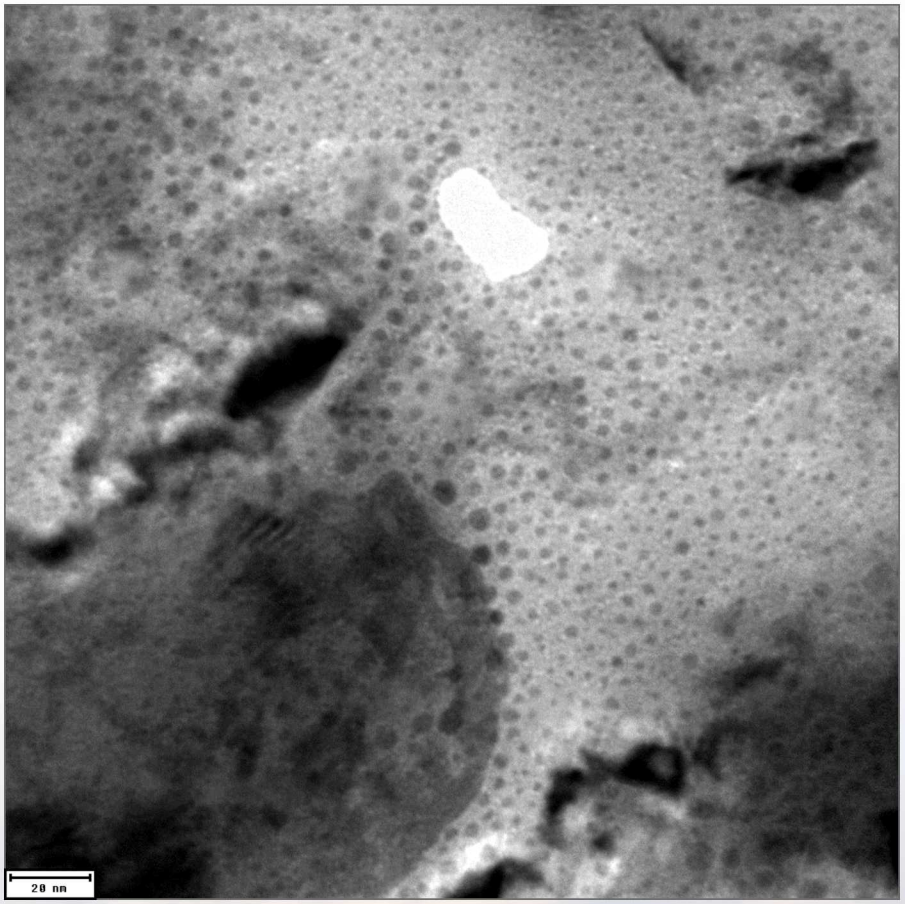


Two Beam
g = $1\bar{1}01$

Through Focus Images: 30 Days Later



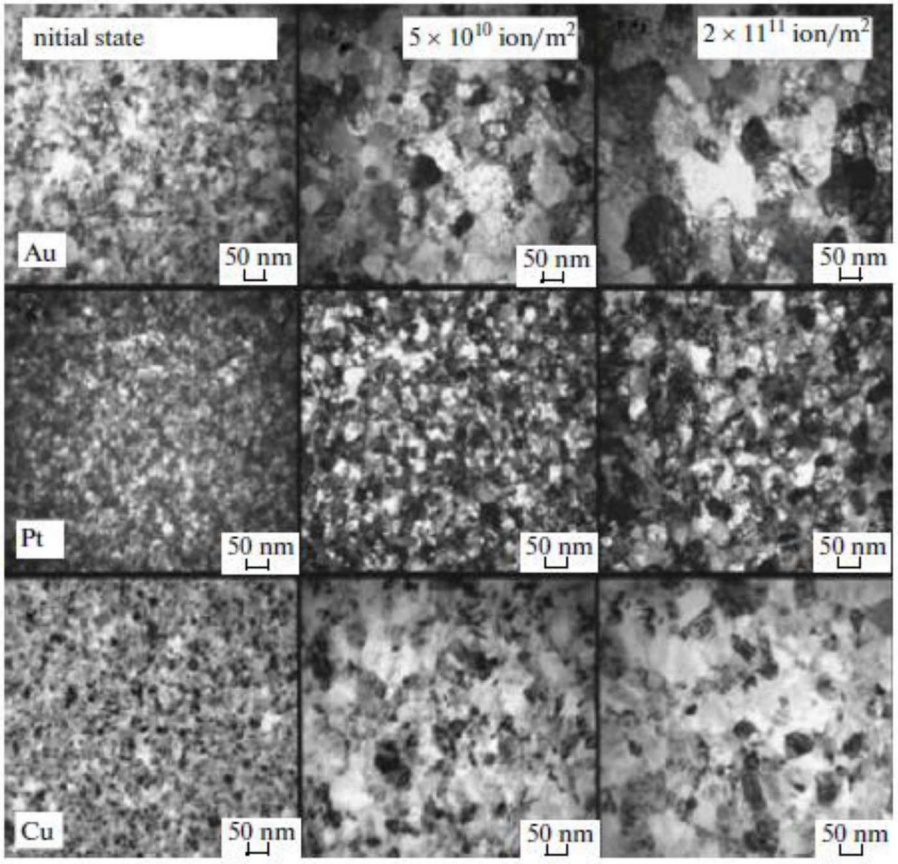
Under Focus



Over Focus

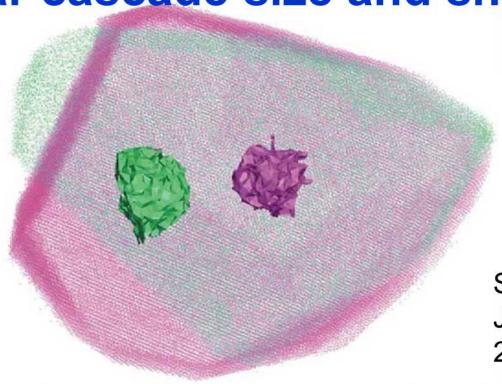
Radiation Tolerance from Nanostructured Metals

Variation in radiation tolerances



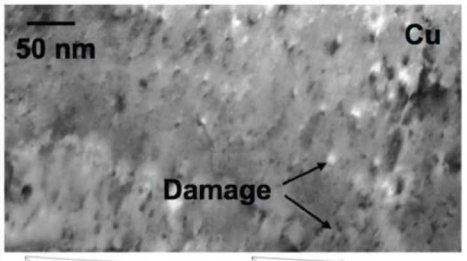
Kaomi et al., JAP: 2008. 104 073525

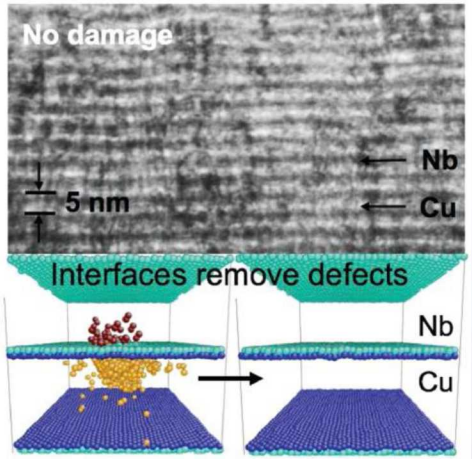
Similar cascade size and shape predicted



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.

Not necessarily the case if initial microstructural details and associated properties are considered

In Situ Irradiation

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

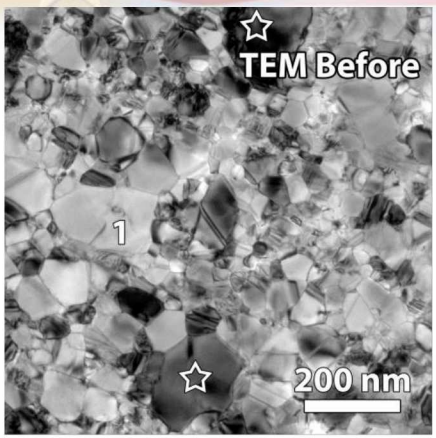
Locations of single ion strikes and resulting microstructural change captured.

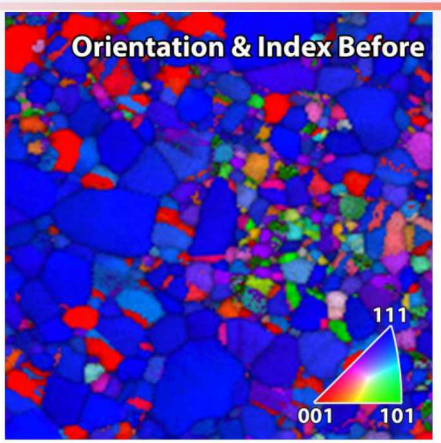
In situ ion irradiation TEM: 10 MeV Si into nanocrystalline Au.

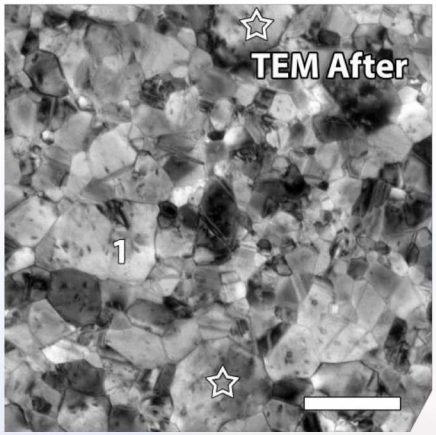
Playback at 2 × real time.

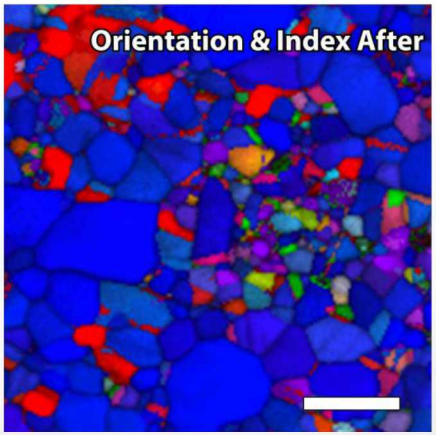
2× real time

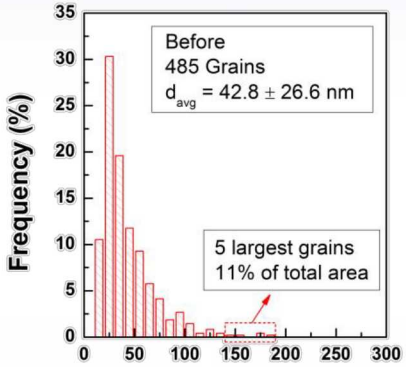
Quantification: Overall

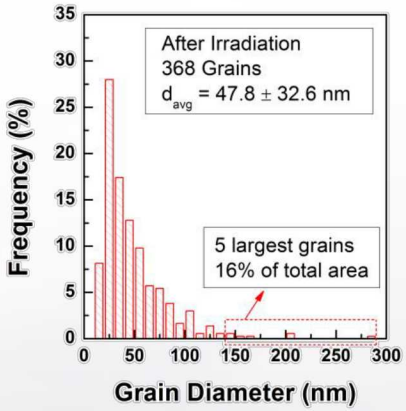


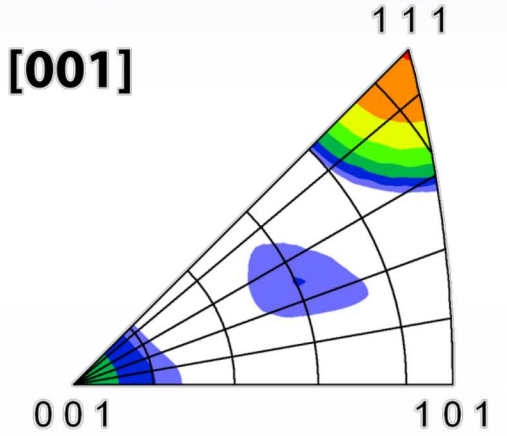


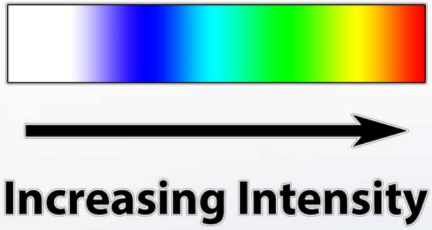












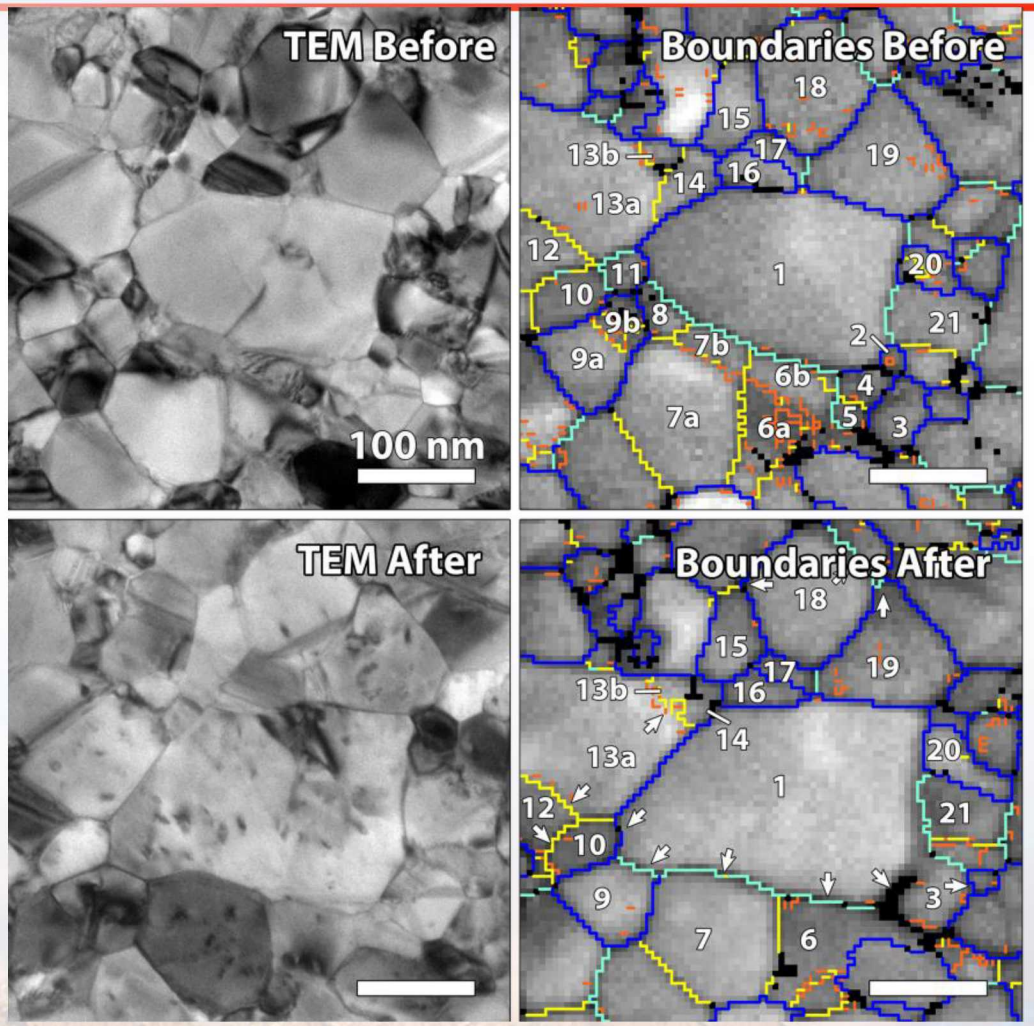
Bufford, *et al*, J Appl Phys, 2015.

- **Same area characterized before and after irradiation.**
  - Local grain size, orientation, boundary character
  - Hundreds of grains counted in minutes

Rapid quantification of statistically relevant numbers of grains and boundaries.

Quantification: Local

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

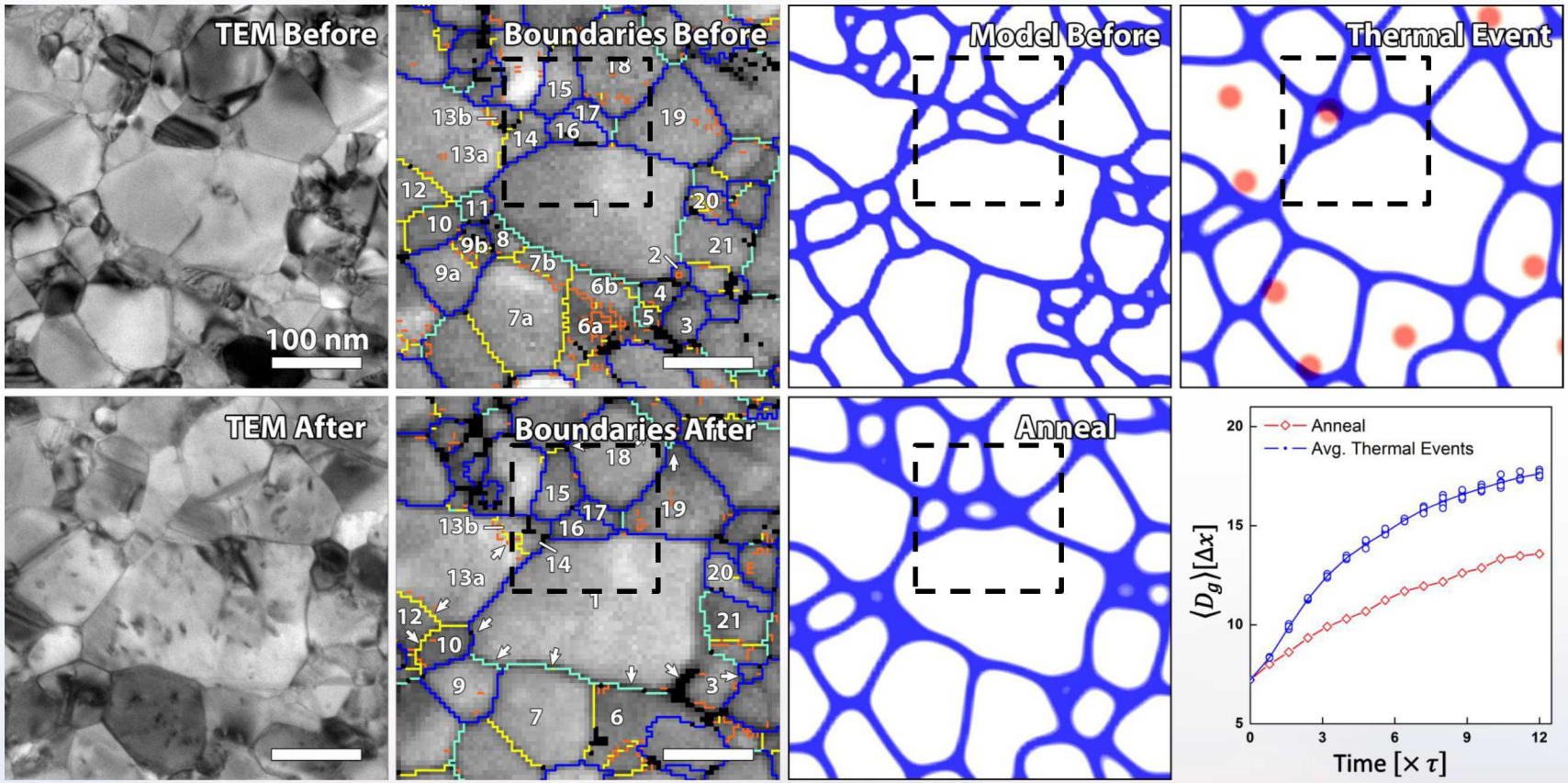


Bufford, *et al*, J Appl Phys, 2015.

3° ≤ φ < 15°
15° ≤ φ < 30°
30° ≤ φ

Individual grain boundary misorientation angle and axes quantified

Exp. & Model Comparison



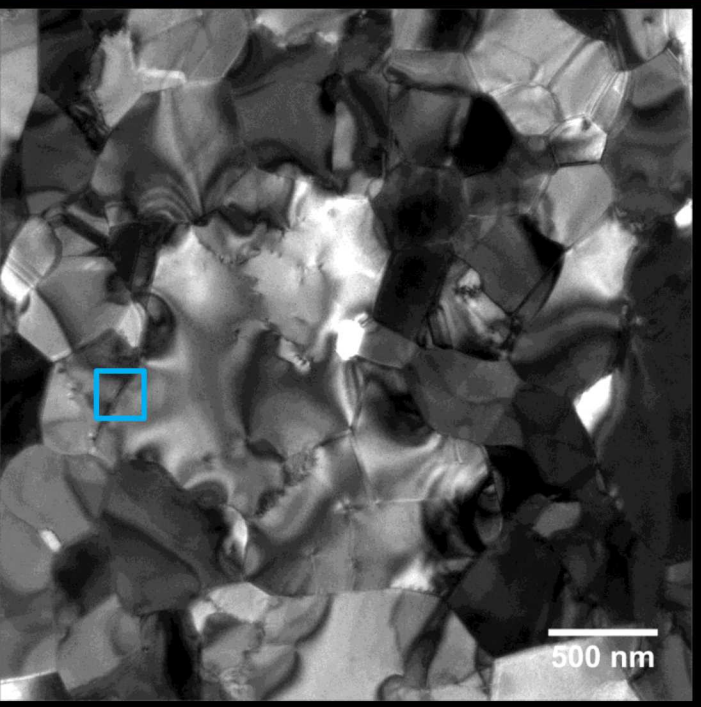
Bufford, *et al*, J Appl Phys, 2015.

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

Immobile boundaries suggest importance of non-thermally activated mobility

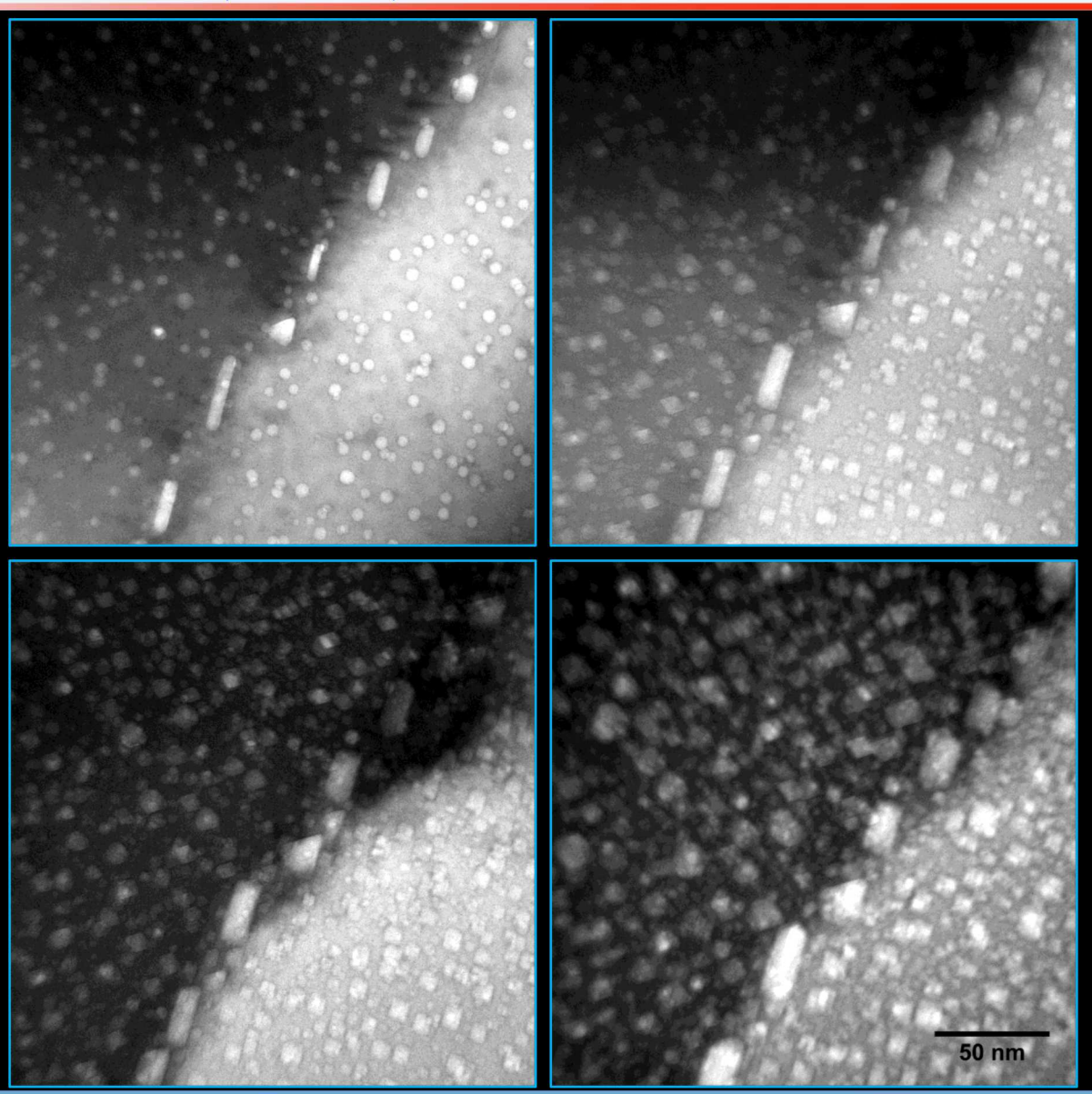
In situ Implantation

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



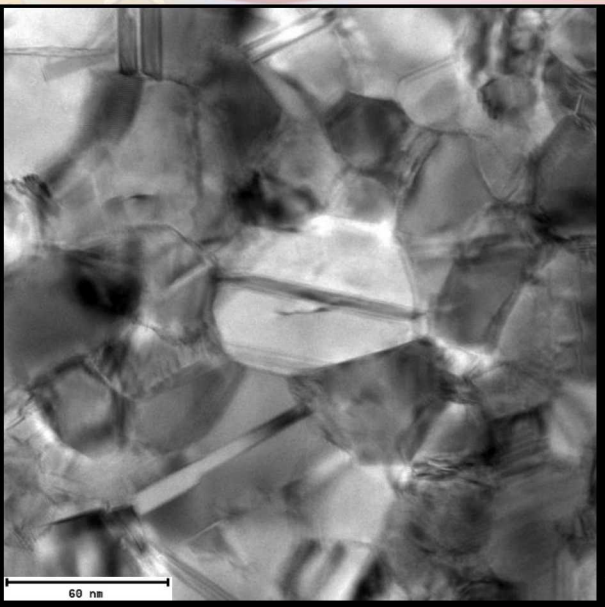
Gold thin-film implanted with 10keV He^{2+}

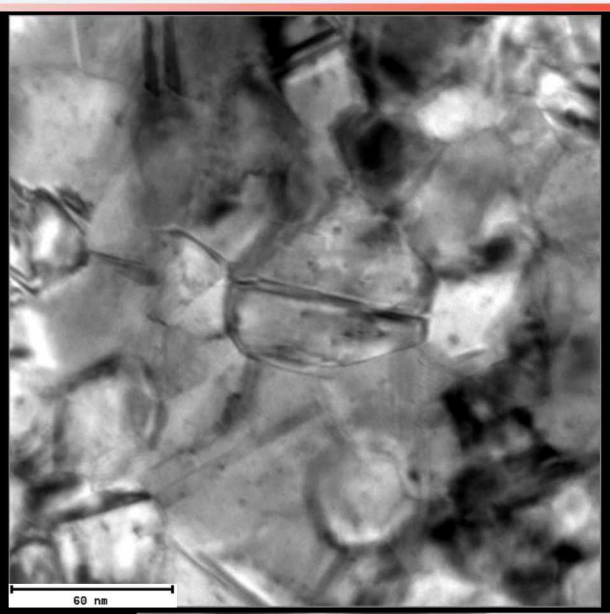
Result: porous microstructure



Nanocrystalline vs. Nanoporous Au results

Collaborators: N. Briot and T.J. Balk



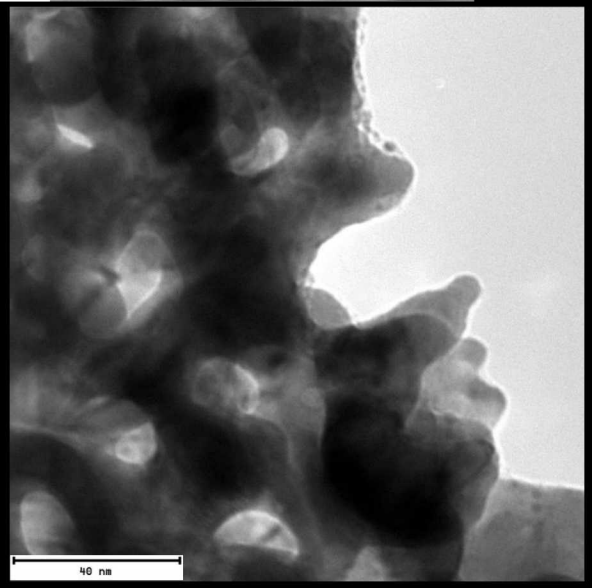


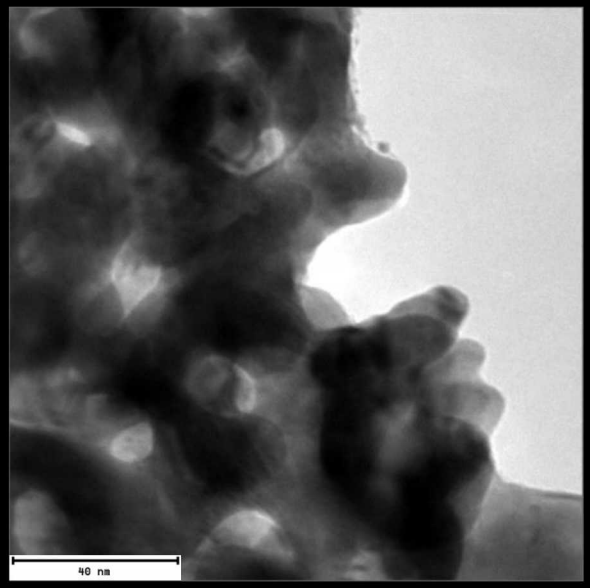
Nanoporous Au after ~ 6.6 dpa at 46 keV:

Melting of the ligament

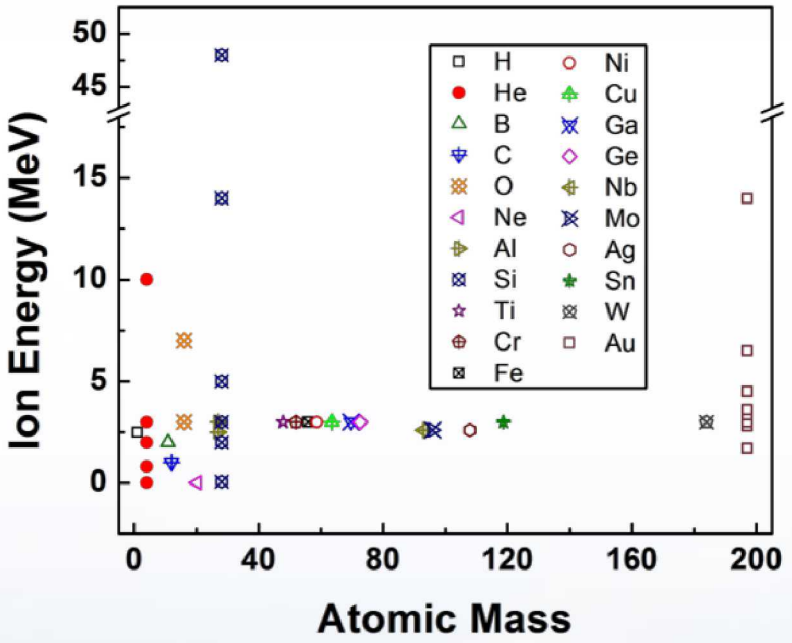
Nanocrystalline Au after ~ 0.5 dpa at 46 keV:

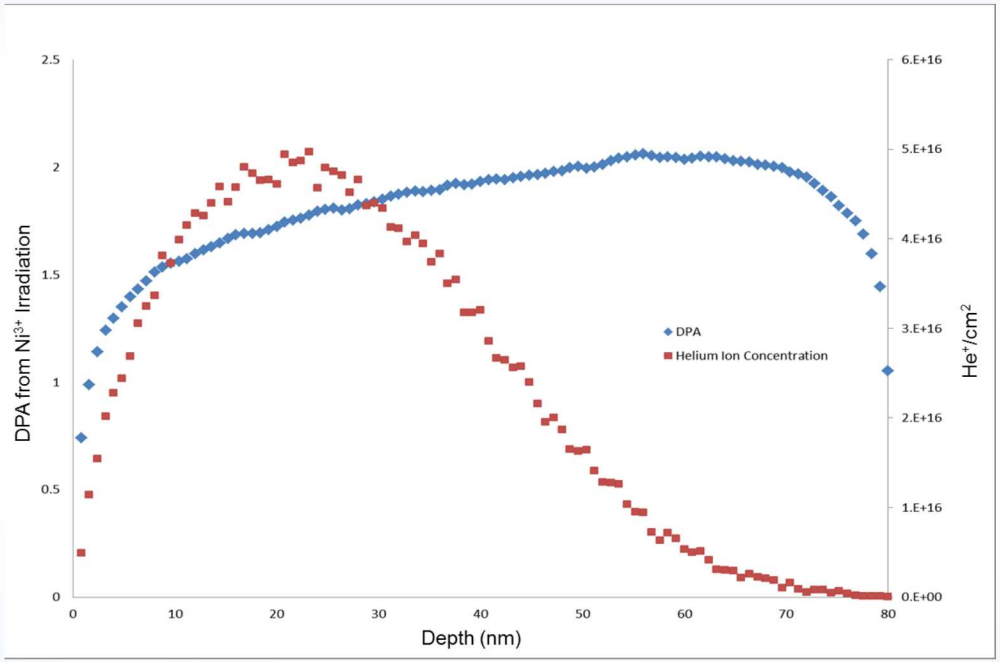
Lots of defects constantly created





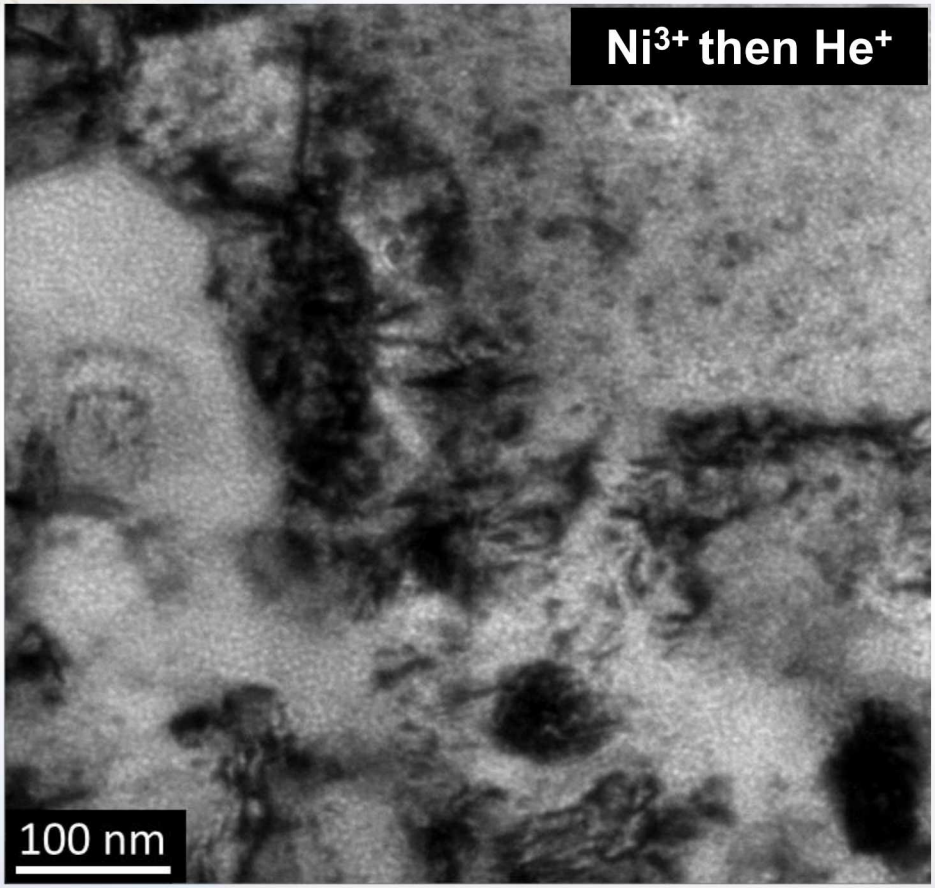
Ion Beam Conditions

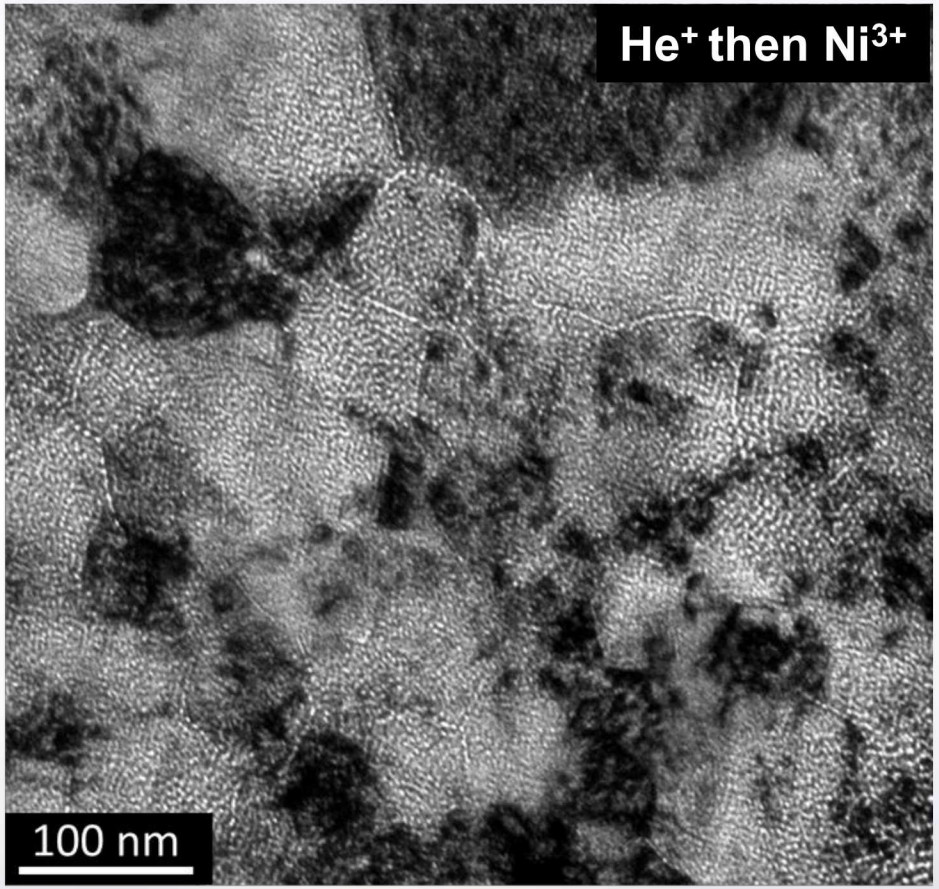




Order	Ni^{3+} Rate	Ni^{3+} damage	He Rate	He concentration
	ions/cm^{2}s	DPA	ions/cm^{2}s	ions/cm^2
Ni^{3+}, He^{+}	1.5 E11	1.8	2.6 E 13	3 E 16
He^{+}, Ni^{3+}	1.5 E11	0.7	5.5 E13	1 E 17

Irradiation / Implantation Sequence Effect on Cavity Structure

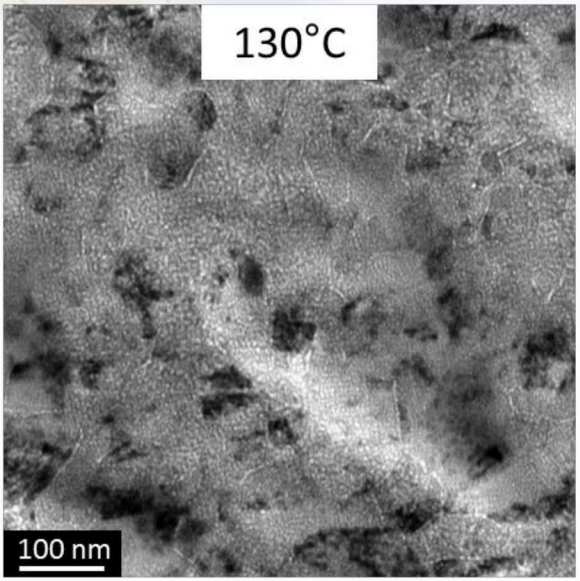


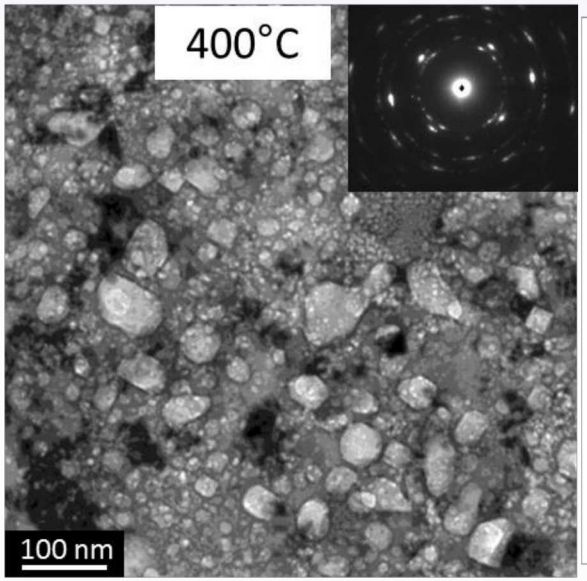


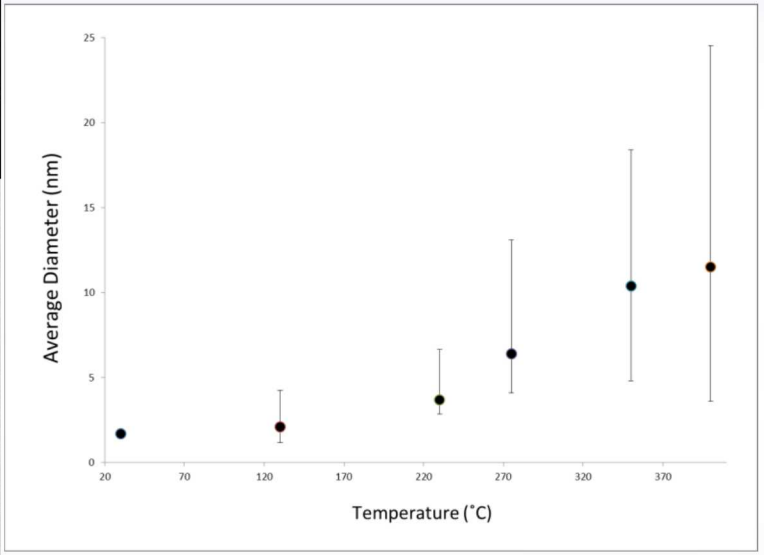
Evenly distributed cavaties over the entire grain structure

Apparent higher concentration of cavities along grain boundaries

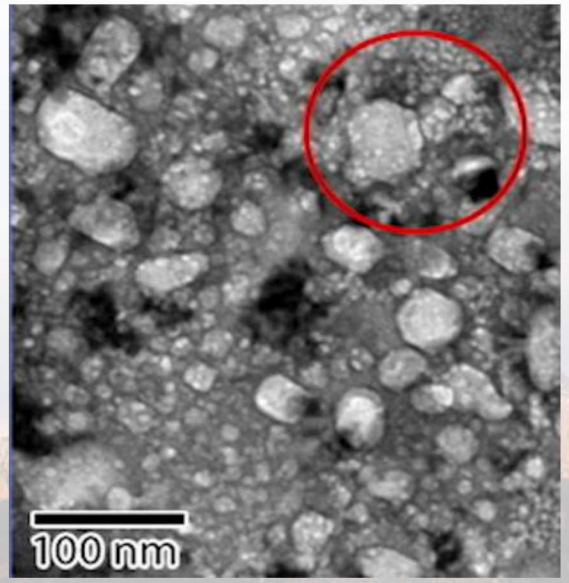
Cavity Growth during In-situ Annealing of 10 keV He^{+} Implanted and then 3 MeV Irradiated Ni^{3+}

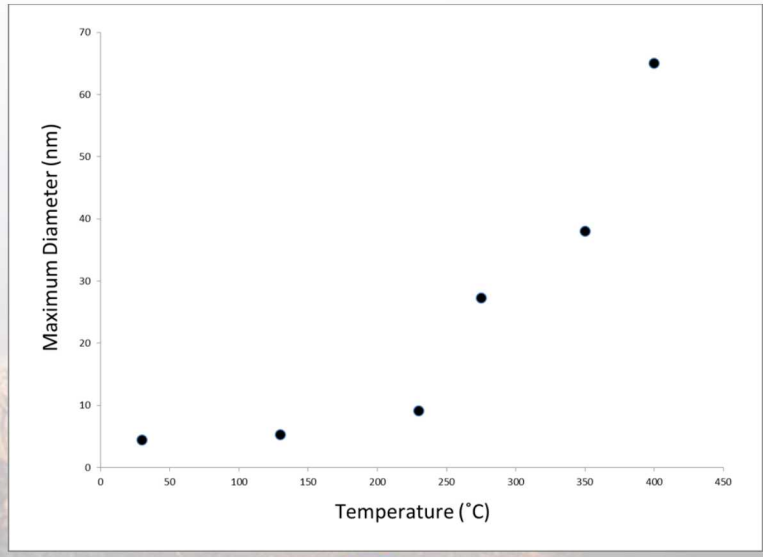






Bubble to cavity transition and cavity evolution can be directly studied

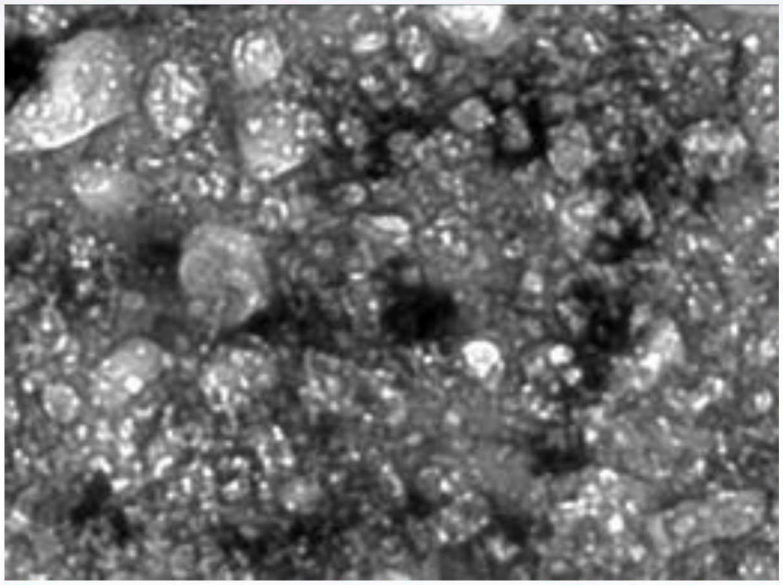


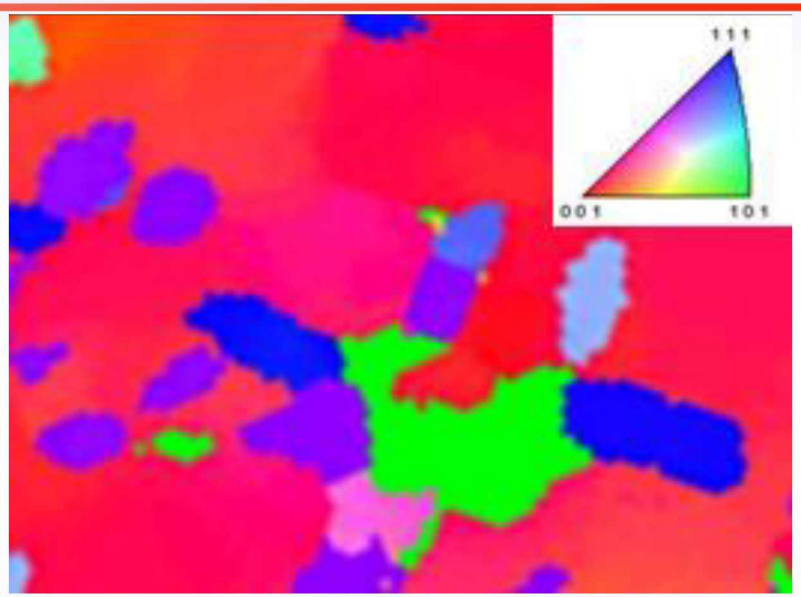


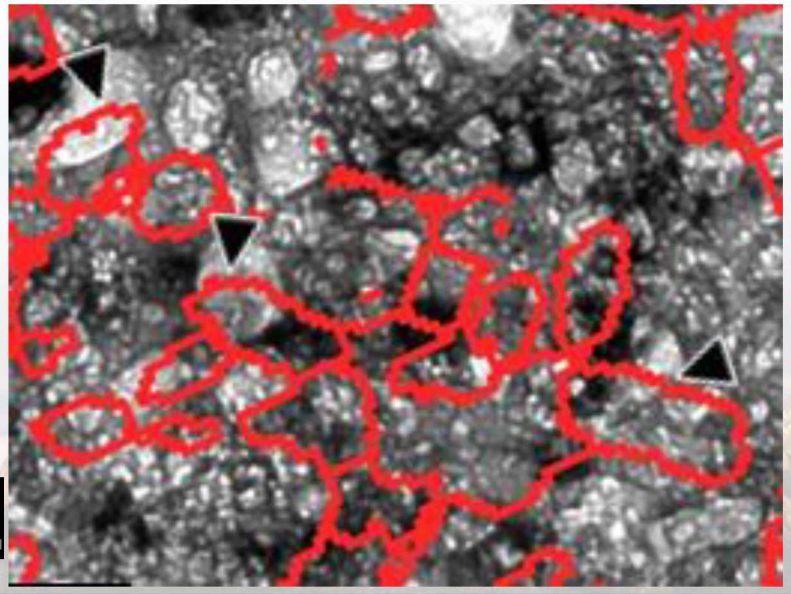
Precession Electron Diffraction Reveals Hidden Grain Structure

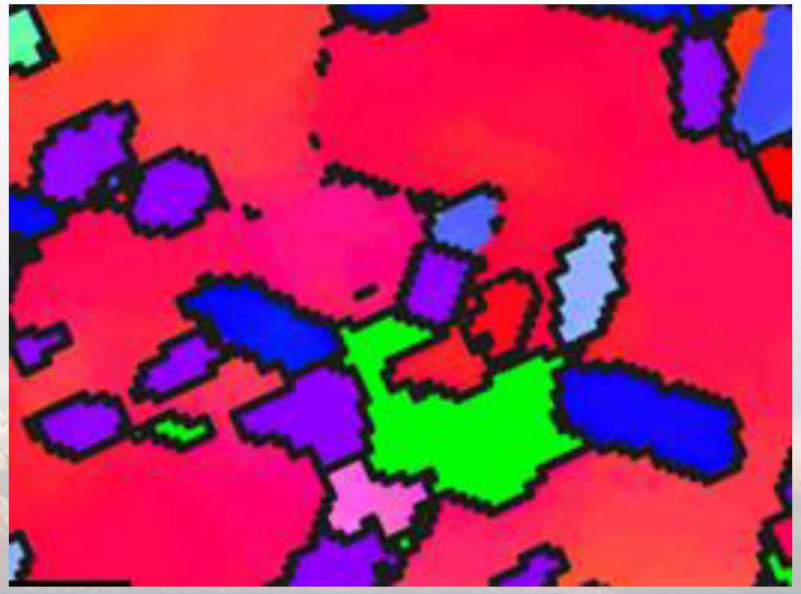
Cavities in helium implanted, self-ion irradiated,

nc nickel film annealed to 400 ˚C







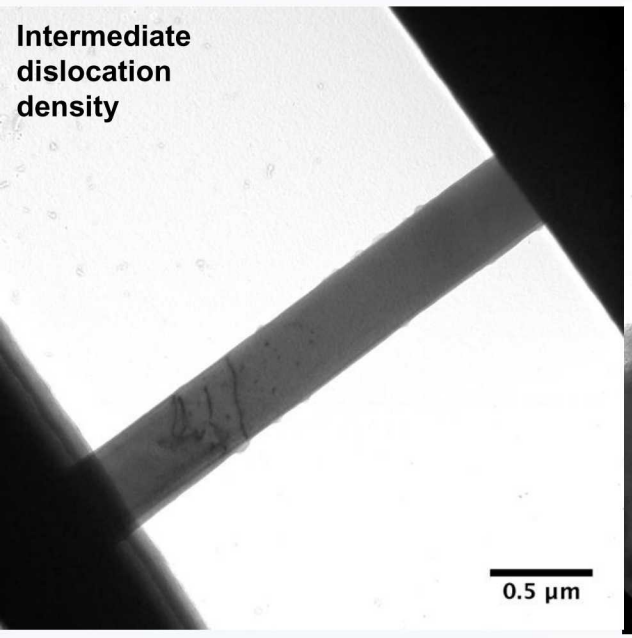


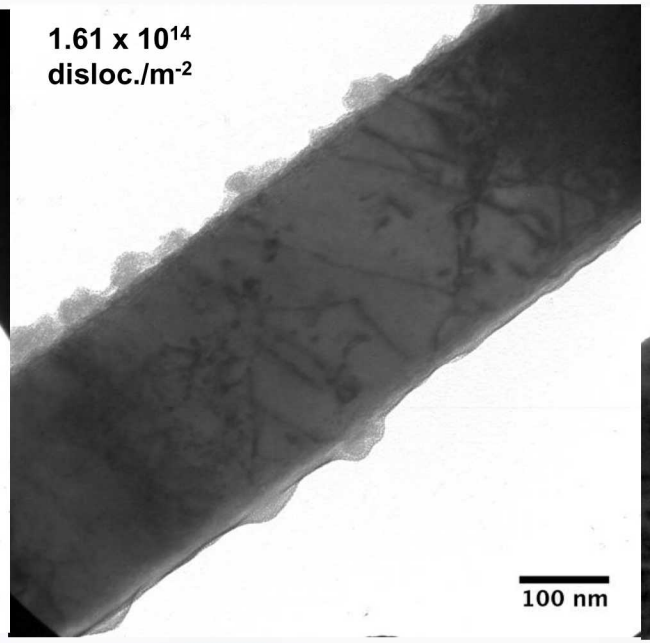
Cavities span multiple grains at identified grain boundaries

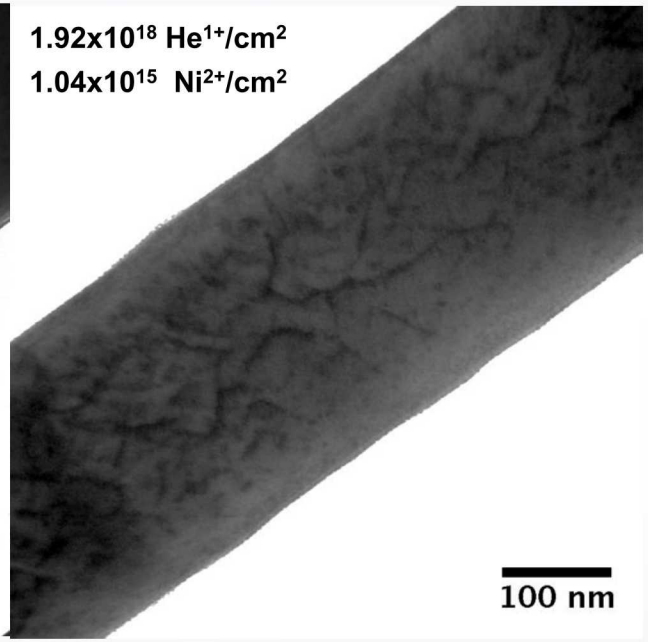
100 nm

Next Steps: *In situ* TEM Quantitative Mechanical Testing

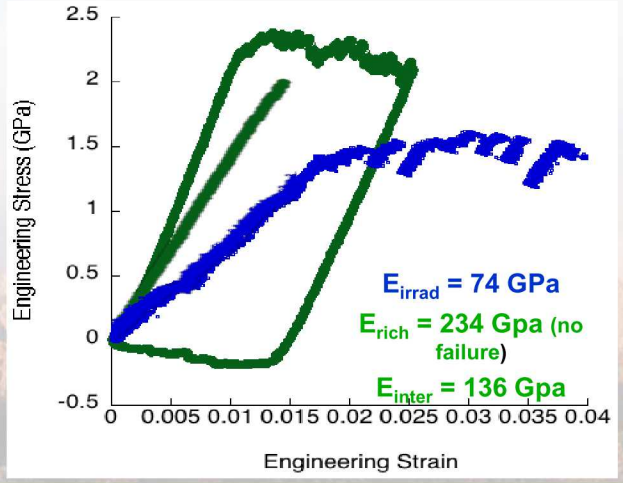
Contributors: C. Chisholm, H. Bei, E.P. George, P. Hosemann, & A.M. Minor







Work has started by looking sequentially at the quantitative effects of ion irradiation on mechanical properties utilizing in-situ ion irradiation TEM and in-situ TEM straining.



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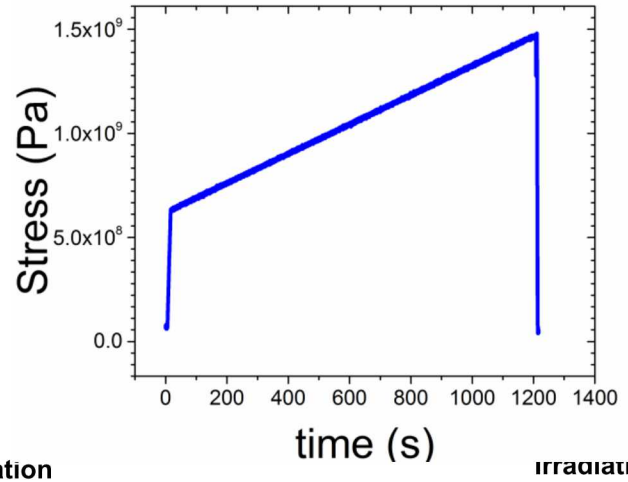
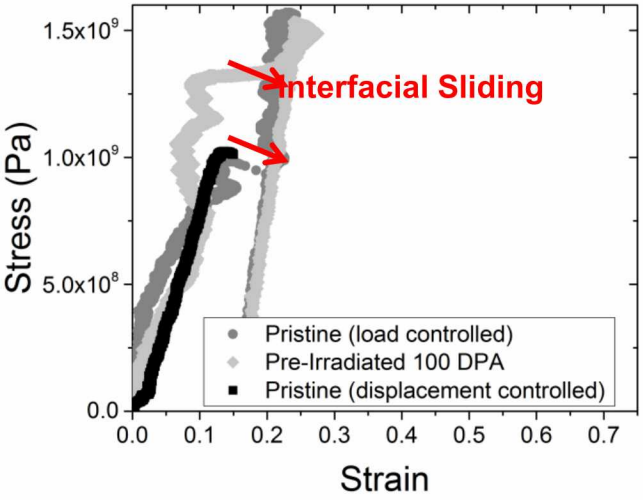
Irradiation Creep (4 MeV Cu^{3+} 10^{-2} DPA/s)

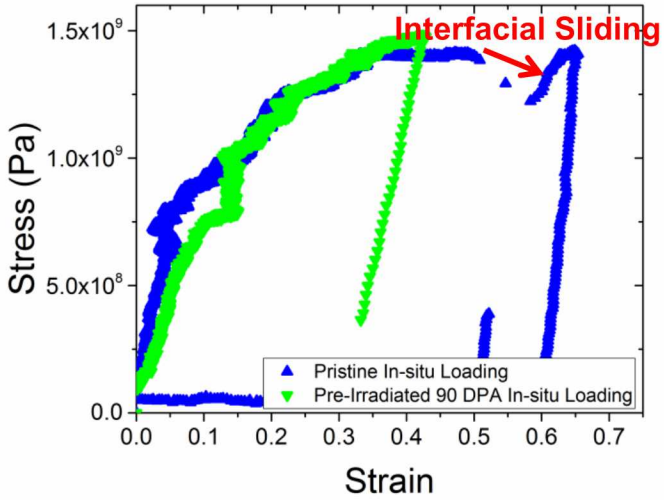
Controlled Loading Rate Experiments



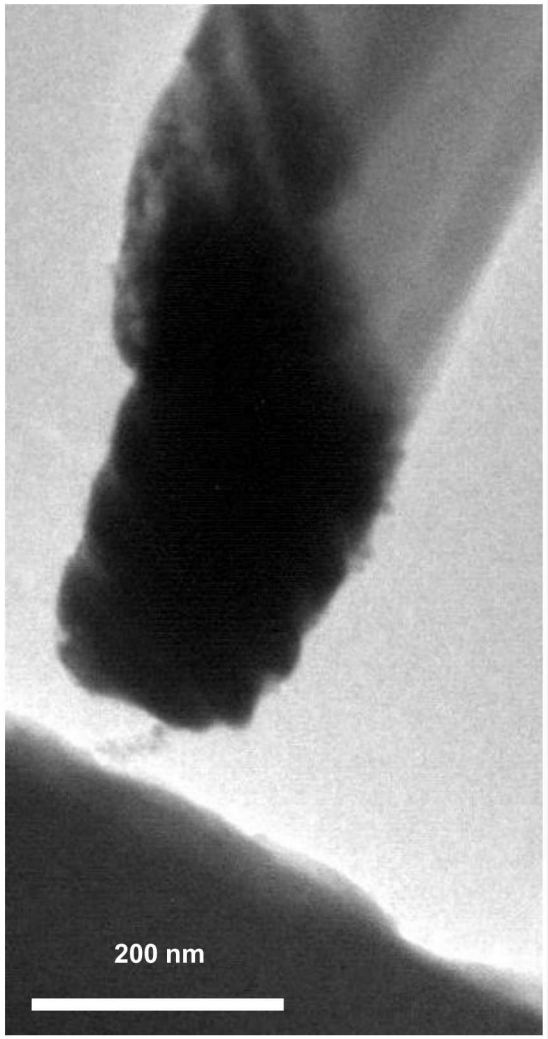
No Irradiation
(Loading rate 0.6 Mpa s^{-1})



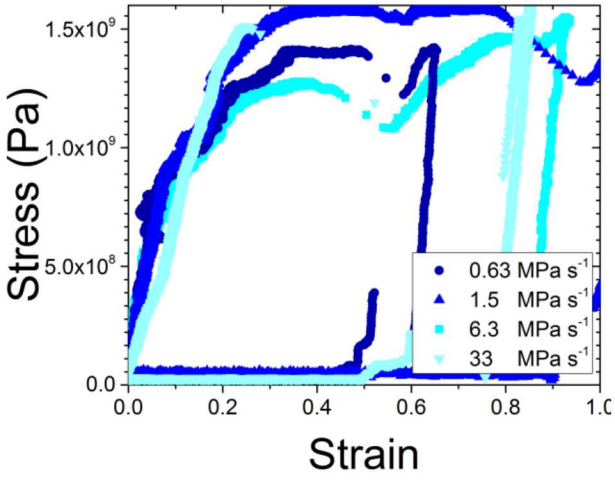
Irradiation Creep
(Loading rate 0.6 Mpa s^{-1})

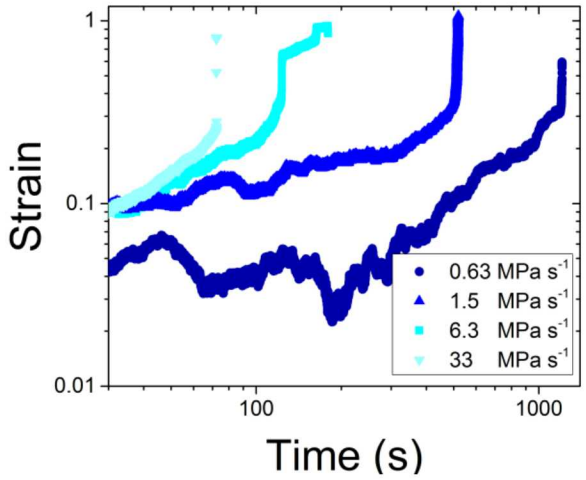


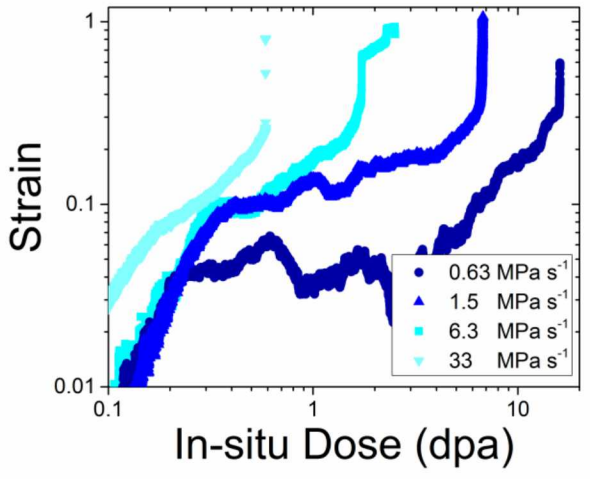
50 nm Cu-W multilayer
20 Min



Creep Response at Different Loading Rates



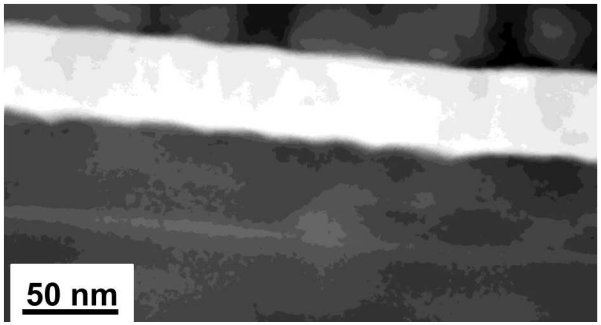




Significant creep observed at a fraction of the bulk yield strength

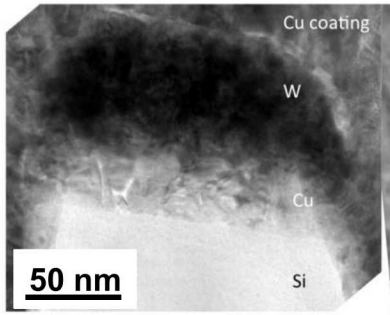
As-deposited Sample

ADF-STEM

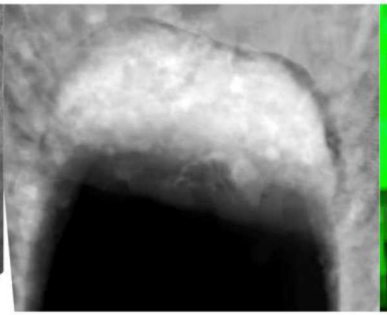


Post Creep Characterization

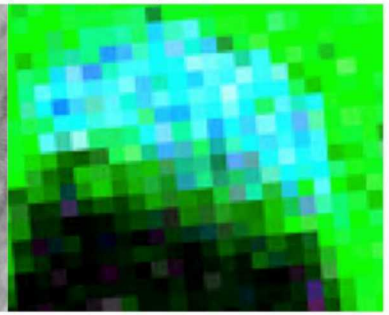
BF-TEM



ADF-STEM



EDS Map (W, Cu, Si)

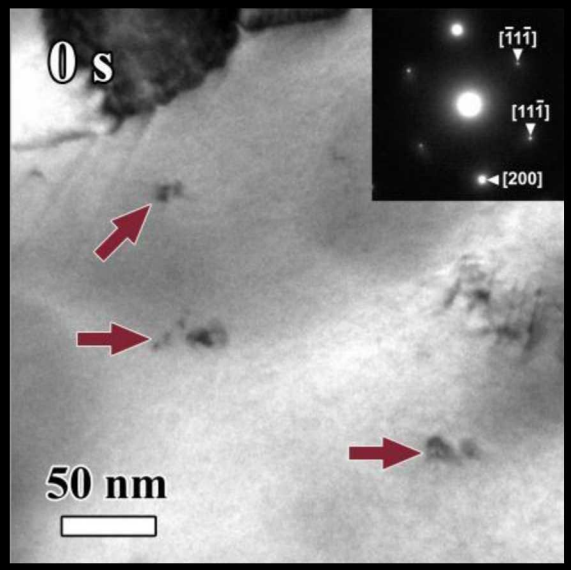


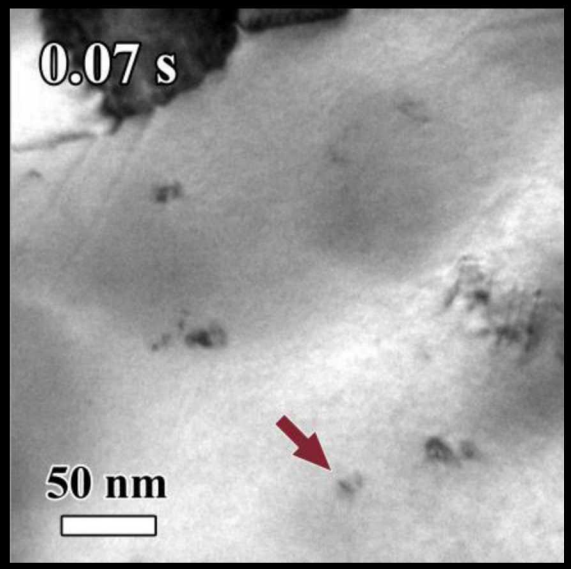
Compression (creep) only observed in Cu layer

48 MeV Si into Au Thin Foil

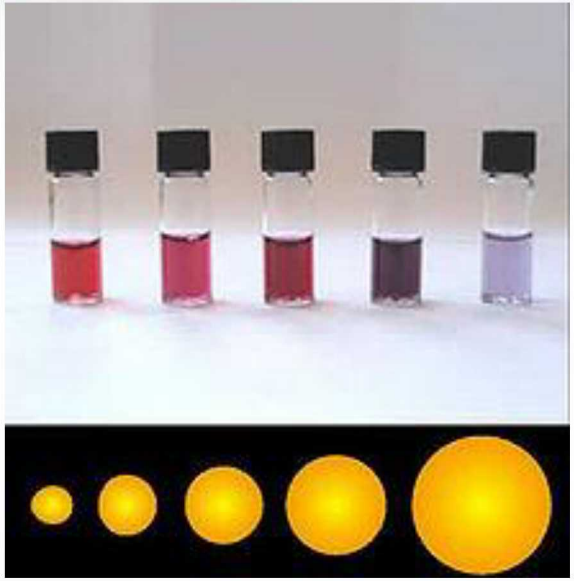
Sped up 8x

The majority of expected Si ion strikes are not observed. Those observed result in significantly smaller defect structures.



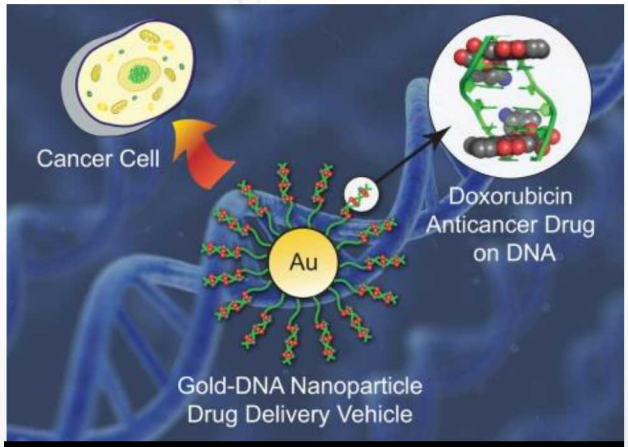


Nanoparticles in Extreme Environments

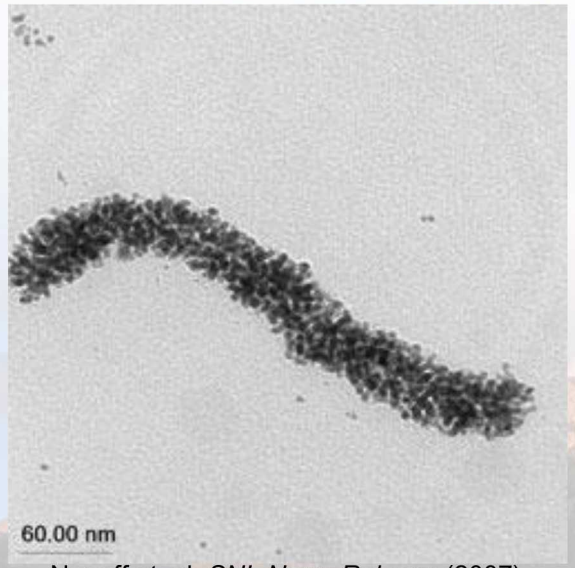


Au nanoparticles (Au NP) are of interest due to their unique optical, electronic, molecular-recognition, and catalytic properties

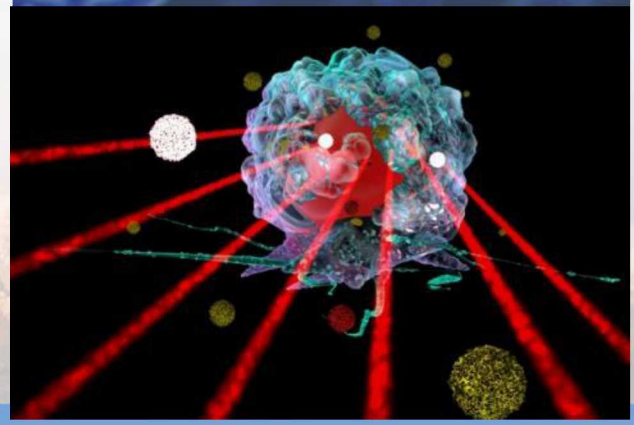
- Photothermal and laser ablation cancer therapies – concentrate IR into heat



- Tumor targeting and drug carriers – enhance dose delivery



- Catalysts for air-pollution control

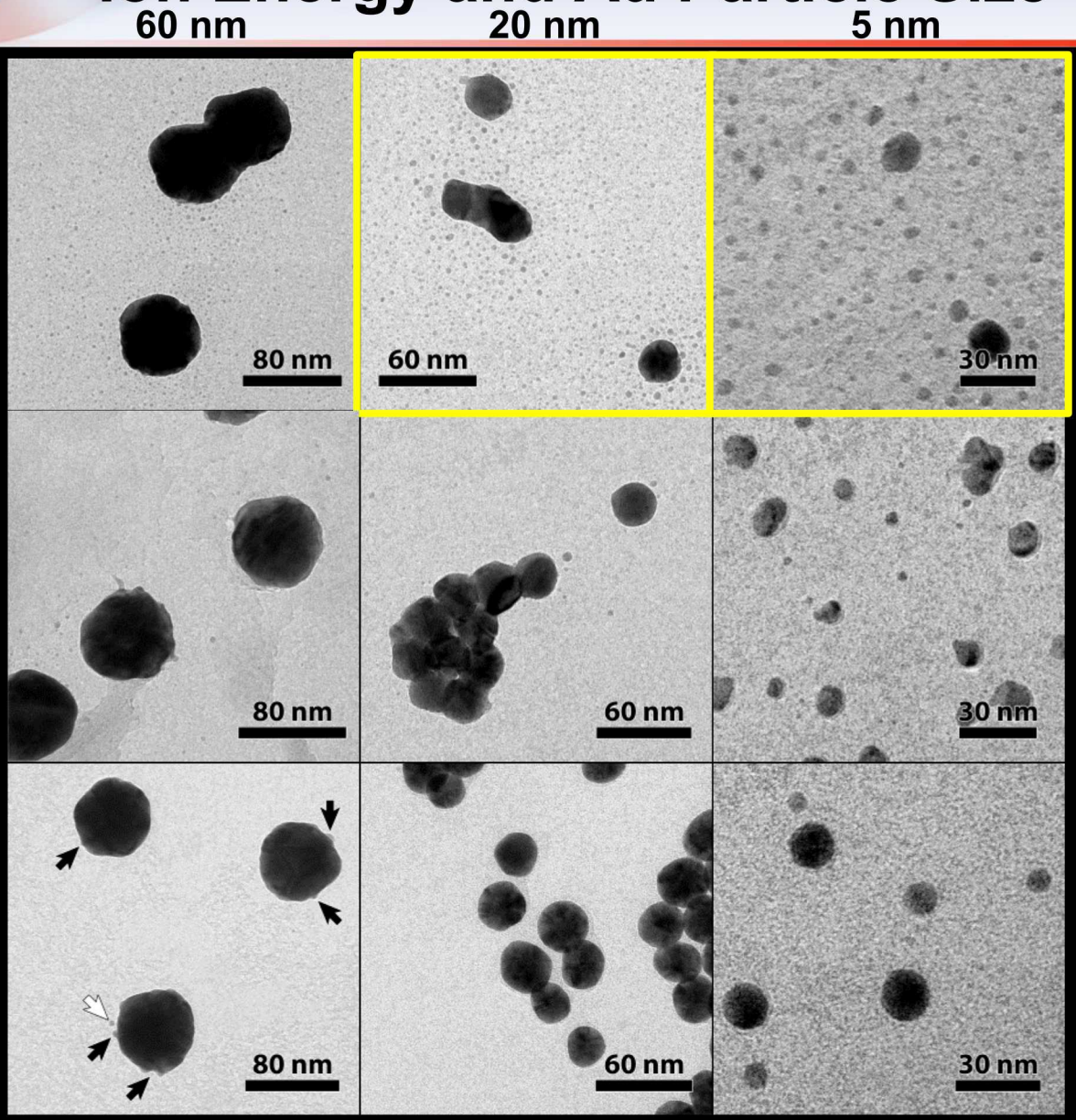


Do the unique properties of the NP withstand extreme radiation?

Nenoff et. al. *SNL News Release* (2007).

Cumulative Effects of Ion Irradiation as a Function of Ion Energy and Au Particle Size

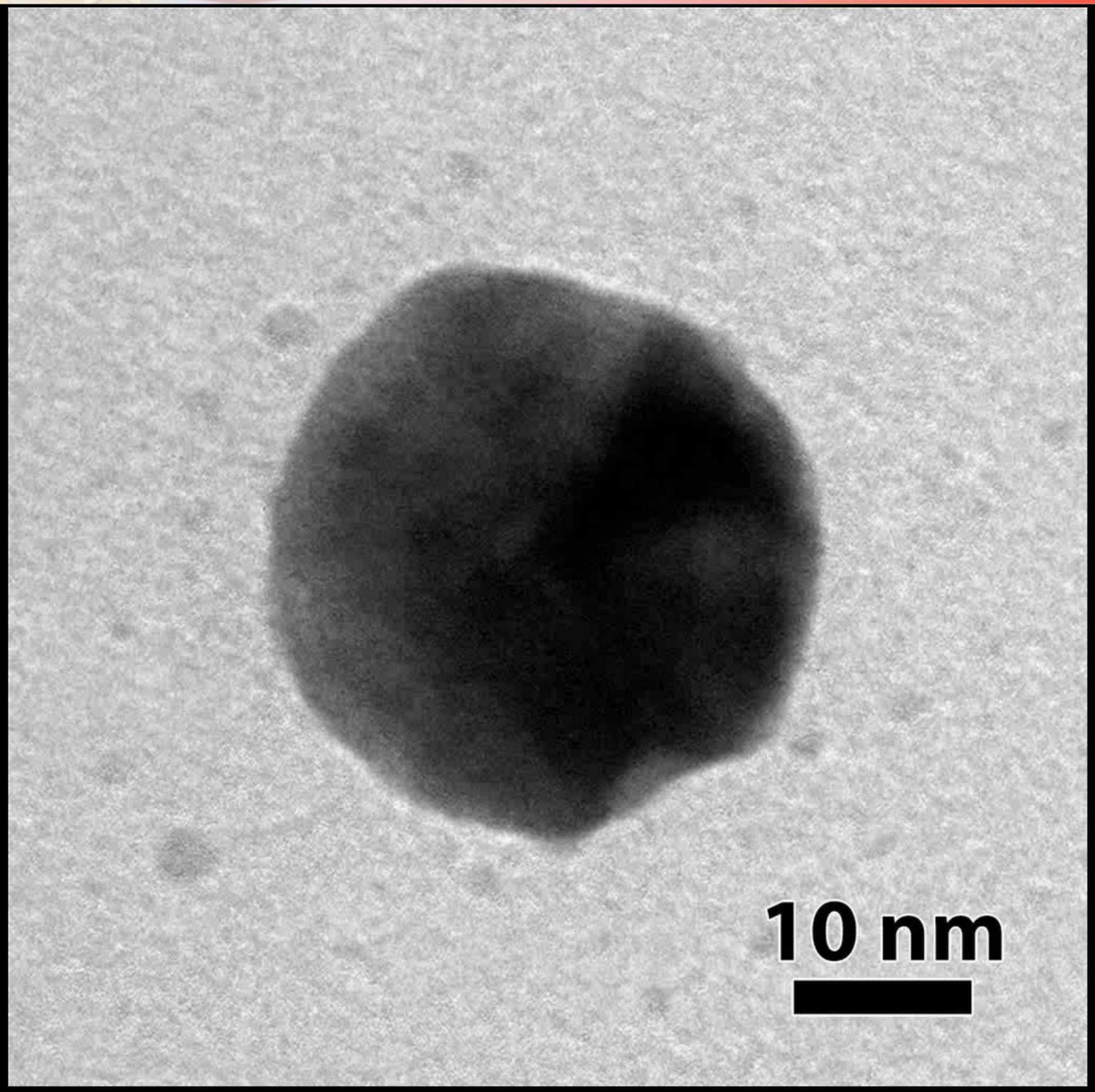
Collaborator: D.C. Bufford

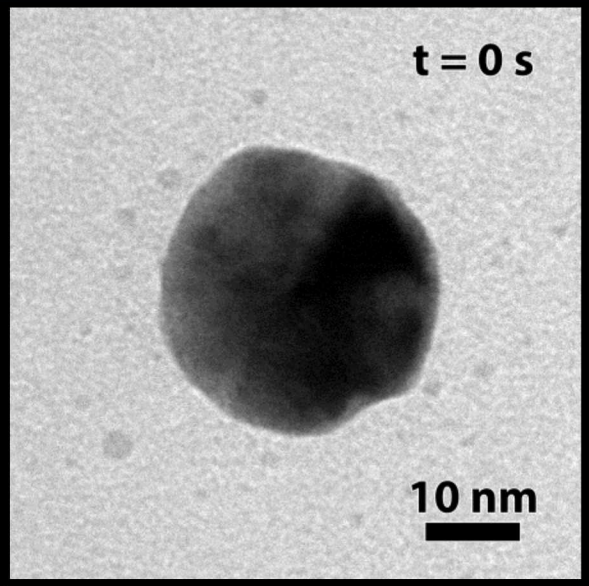


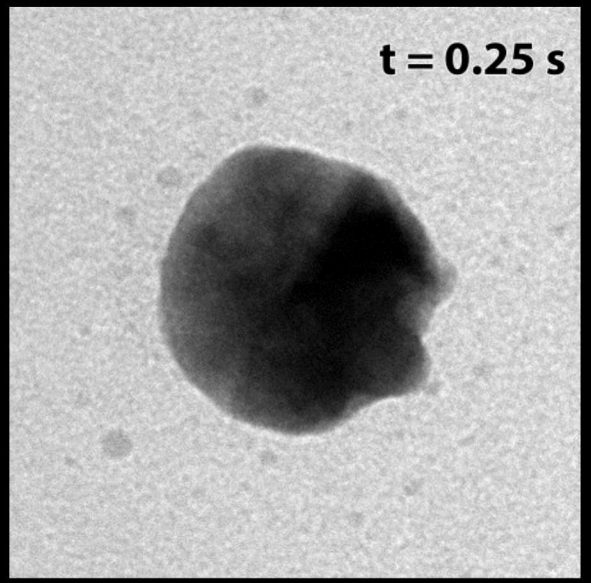
Particle and ion energy dictate the ratio of sputtering, particle motion, particle agglomeration, and other active mechanisms

Single Ion Effects with 46 keV Au^{1-} ions: 20 nm

Collaborator: D.C. Bufford

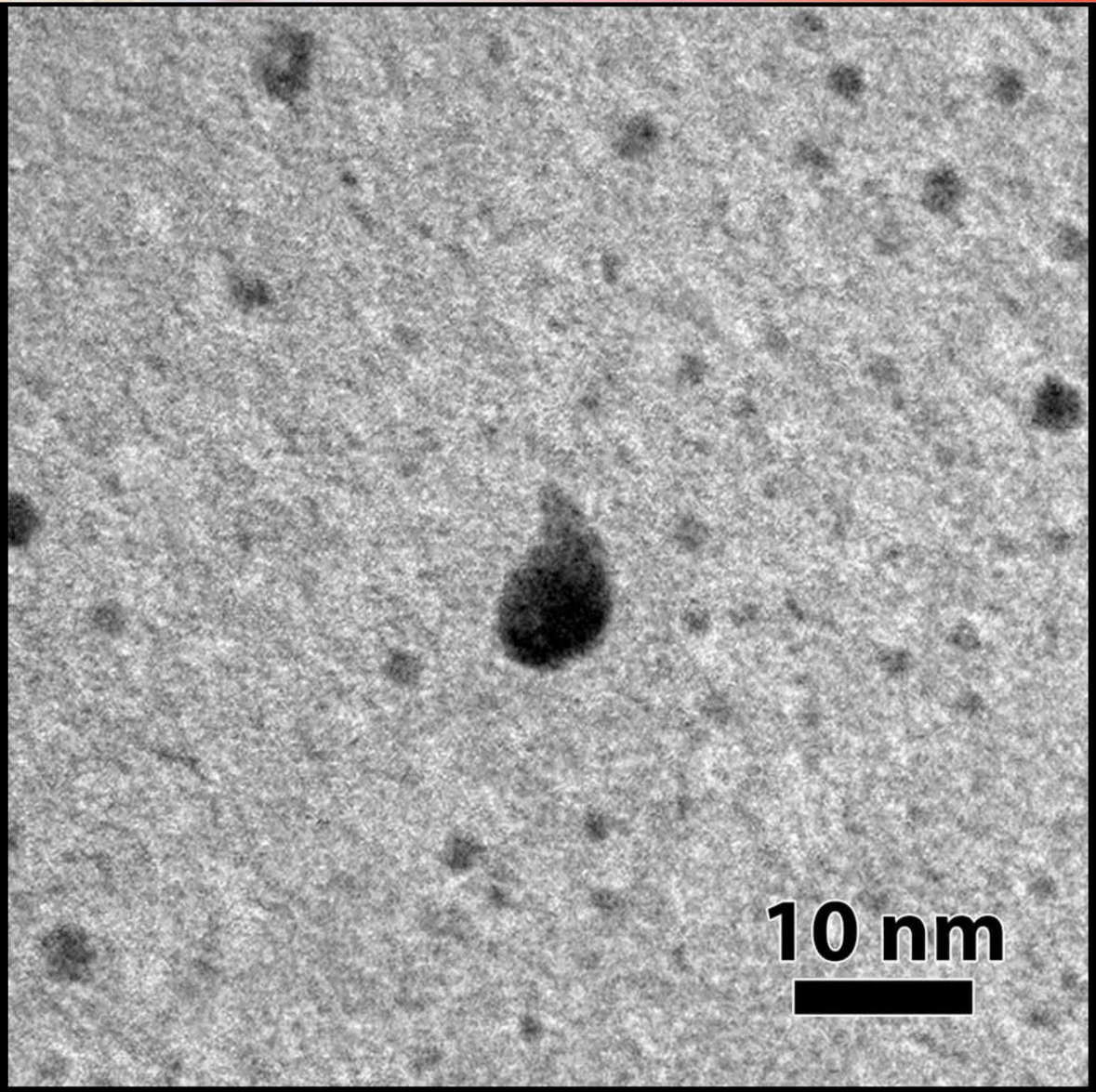


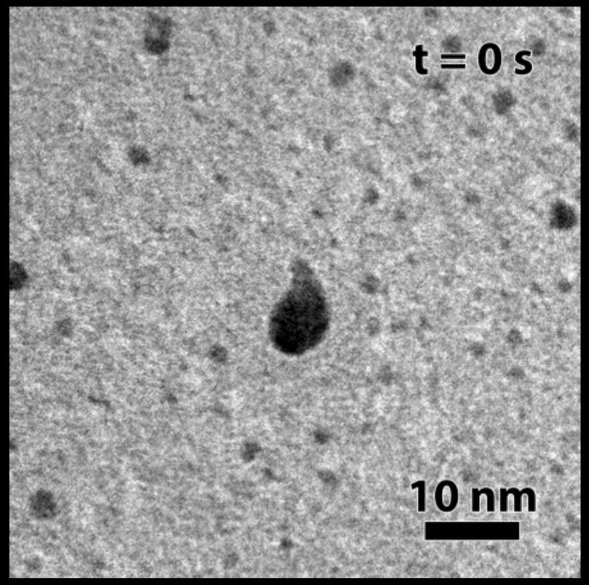


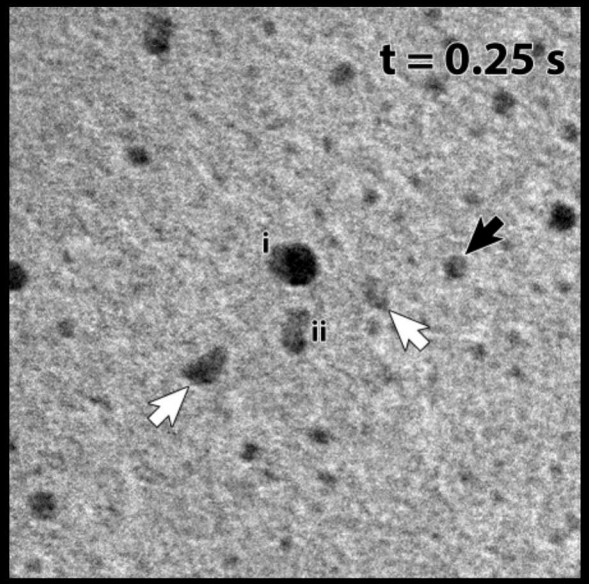


Single Ion Effects with 46 keV Au^{1-} ions: 5 nm

Collaborator: D.C. Bufford

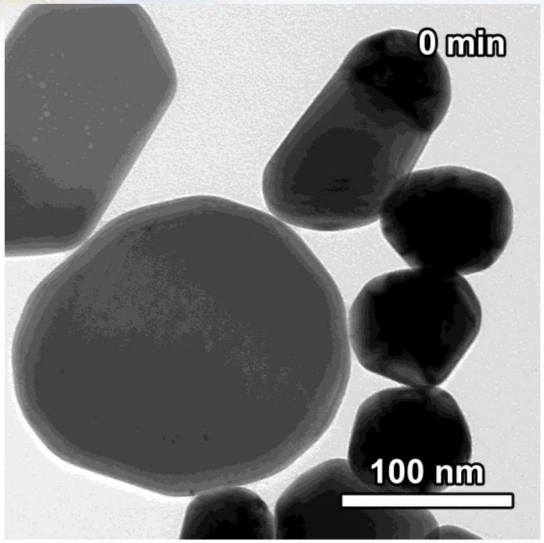


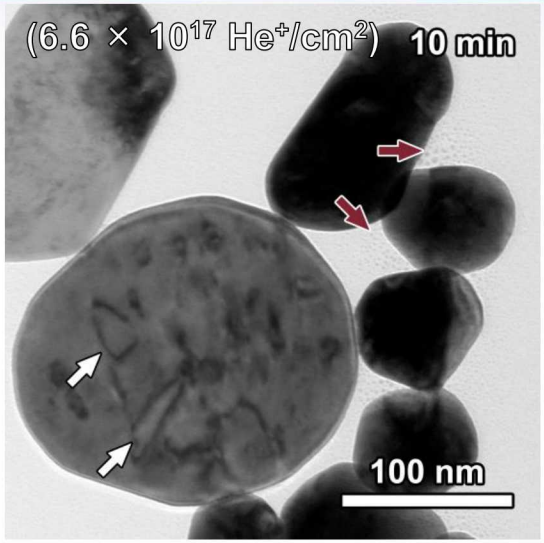


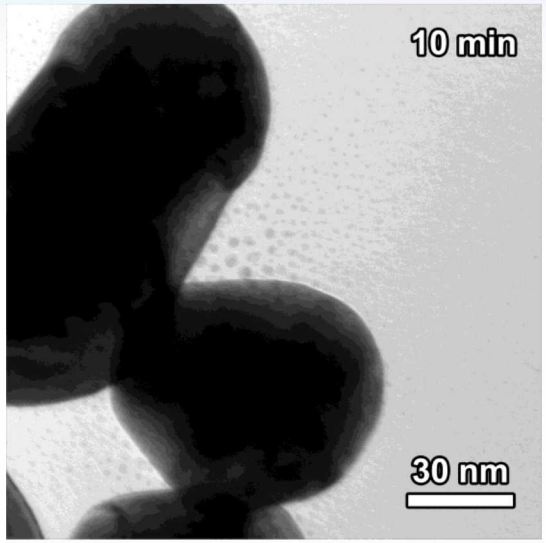


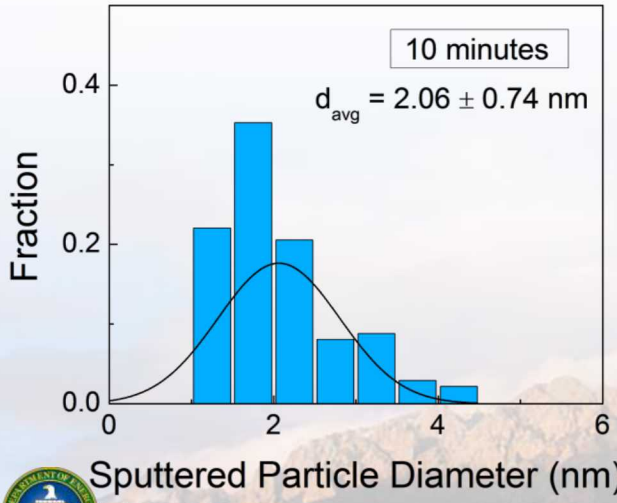
Formation of Dislocation Loops & Sputtered Particles due to He implantation

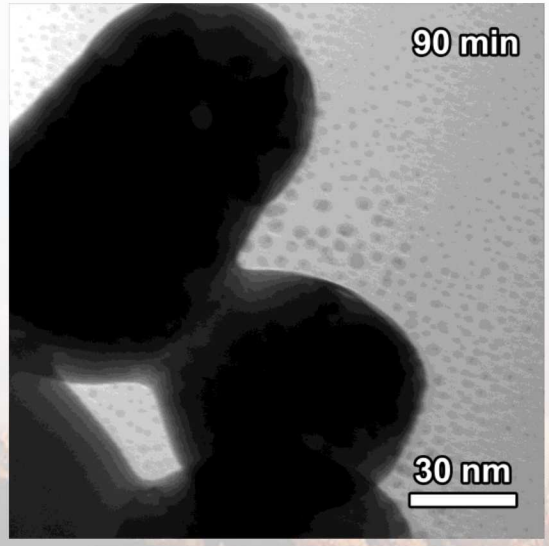
Collaborators: D.C. Bufford, S.H. Pratt & T.J. Boyle

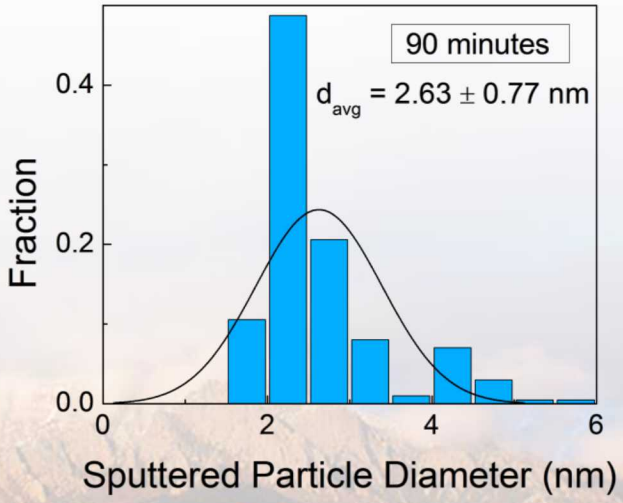










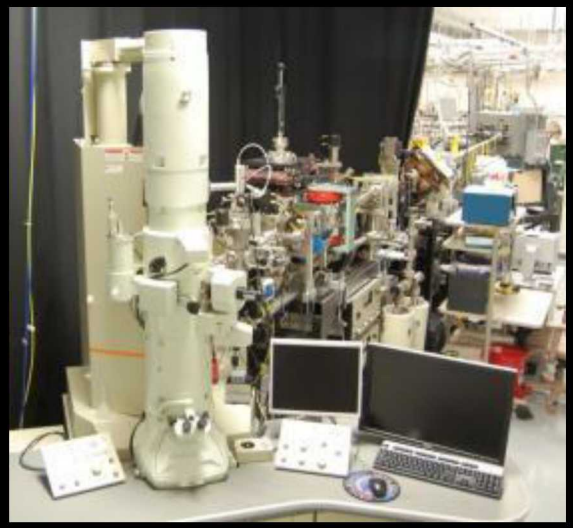


Sandia National Laboratories

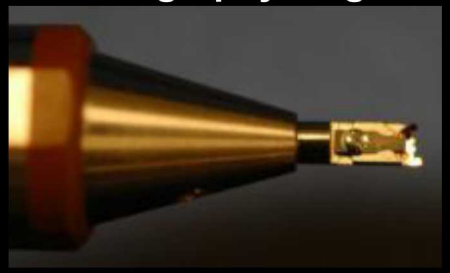
Advanced Microscopy Techniques Applied to Nanoparticles in Radiation Environments

Collaborators: S.H. Pratt & T.J. Boyle

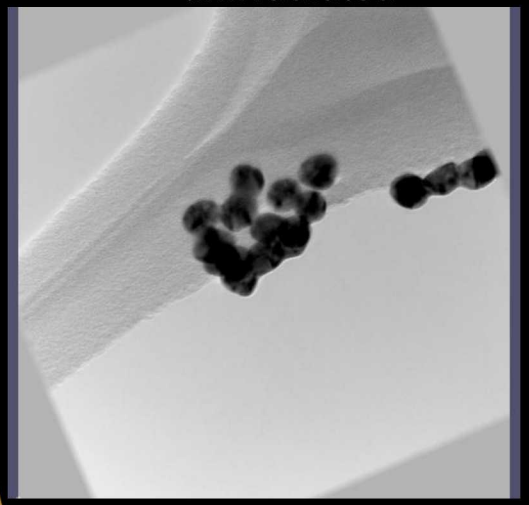
In situ Ion Irradiation TEM (I^3TEM)



Hummingbird tomography stage



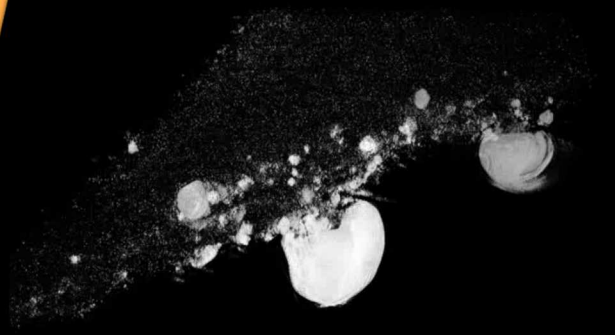
Aligned Au NP tilt series - unirradiated



Aligned Au NP tilt series - irradiated

Unirradiated Au NP model

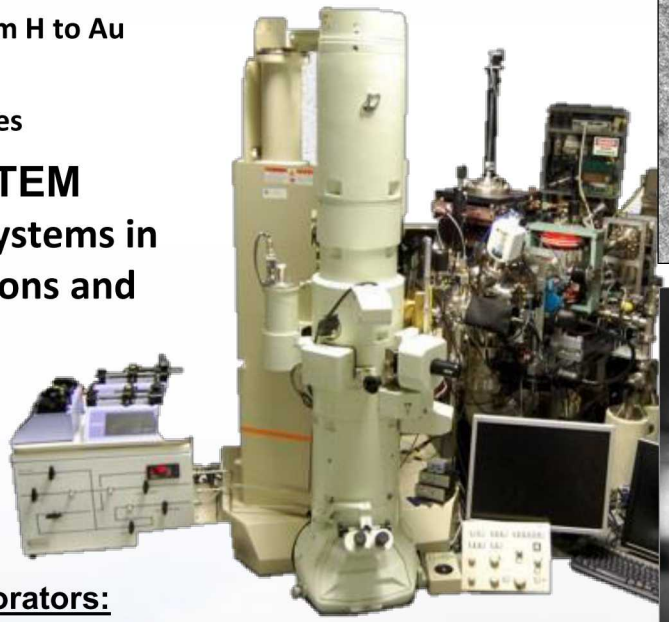
Irradiated Au NP model

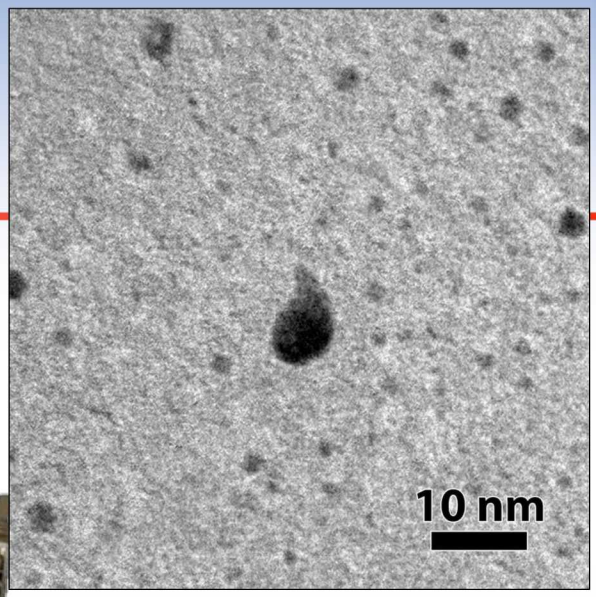


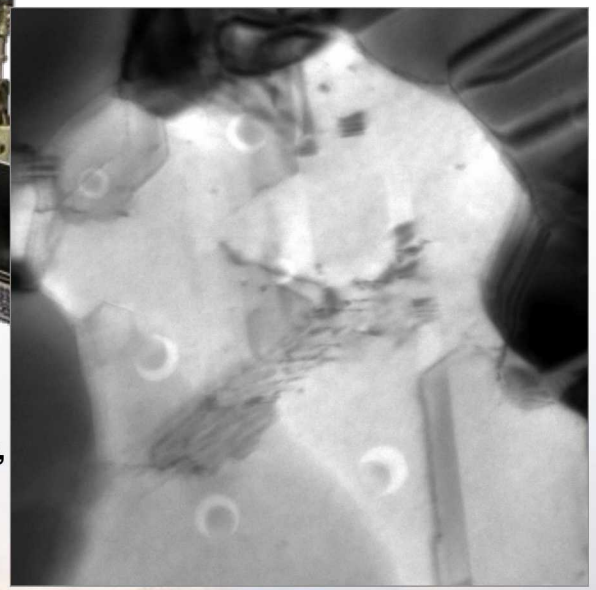
The application of advanced microscopy techniques to extreme environments provides exciting new research directions

Summary

- Sandia's I^{3}TEM is one of only two facilities in the US
  - Only facility in the world with a wealth of dual *in situ ion irradiation* capabilities
  - *In situ* high energy ion irradiation from H to Au
  - *In situ* gas implantation
  - 14 TEM stages with various capabilities
- Currently applying the current I^{3}TEM capabilities to various material systems in combined environmental conditions and expand the capabilities







Sandia's I^{3}TEM although still under development is providing a wealth of interesting initial observations

Collaborators:

- IBL: D.C. Bufford, D. Buller, C. Chisholm, B.G. Clark, J. Villone, G. Vizkelethy, B.L. Doyle, S. H. Pratt, & M.T. Marshall
- Sandia: B. Boyce, T.J. Boyle, P.J. Cappillino, J.A. Scott, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, E. Carnes, J. Brinker, D. Sasaki, J.A. Sharon, T. Nenoff, W.M. Mook, P. Feng, F.P. Doty, B.A. Hernandez-Sanchez, P. Yang, J-E Mogonye, S.V. Prasad , P. Kotula, S. Howell, T. Ohta, & T. Beechem
- External: A. Minor, L.R. Parent, I. Arslan, H. Bei, E.P. George, P. Hosemann, D. Gross, J. Kacher, & I.M. Robertson

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