Progress Towards *In situ* TEM Experiments in Combinations of Extreme Environments

SAND2014-20274C

K. Hattar, D.C. Bufford, M. Marshall, D.L. Buller, and B.L. Doyle

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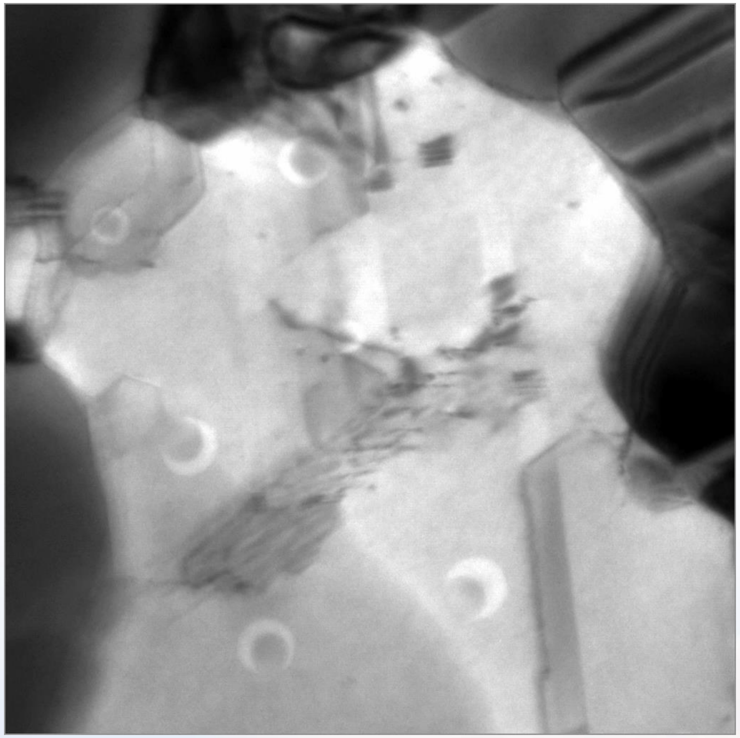
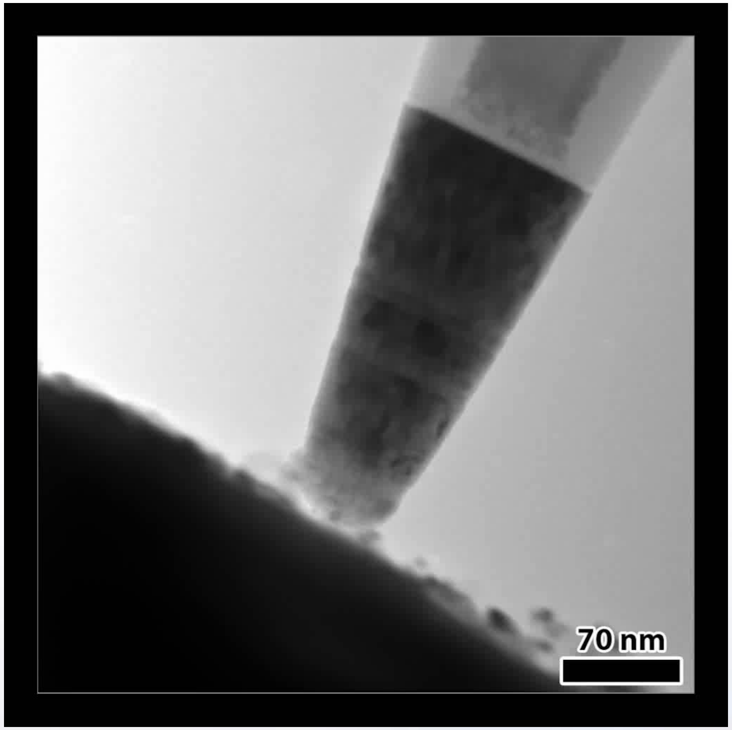


December 1, 2014



Collaborators:

- **Sandia: C. Chisholm, B.G. Clark, S. H. Pratt, 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**
- **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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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**
   - Portion of the world or even a 3mm disk studied is small

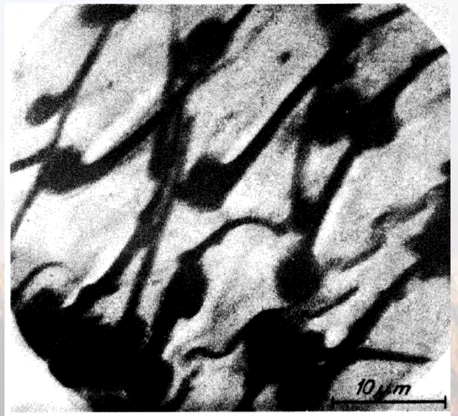


Fig. 6: Wing surface of the house fly.
(First internal photography, U = 60 kV, M_e = 2200)
(Driest, E., and Müller, H.O.: Z. Wiss. Mikroskopie 52, 53-57 (1935)

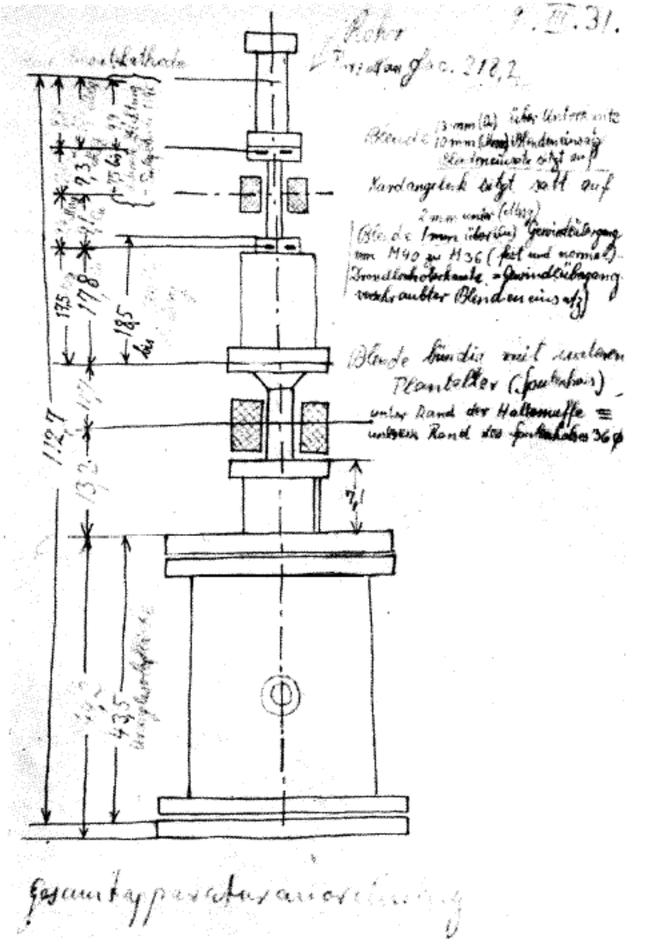
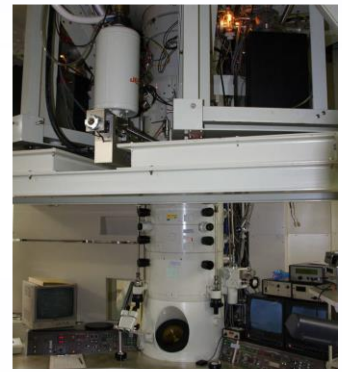


Fig. 2: Sketch by the author (9 March 1931) of the cathode ray tube for testing one-stage and two-stage electron-optical imaging by means of two magnetic electron lenses (electron microscope) [8].

E. Ruska Nobel lecture (1986)

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

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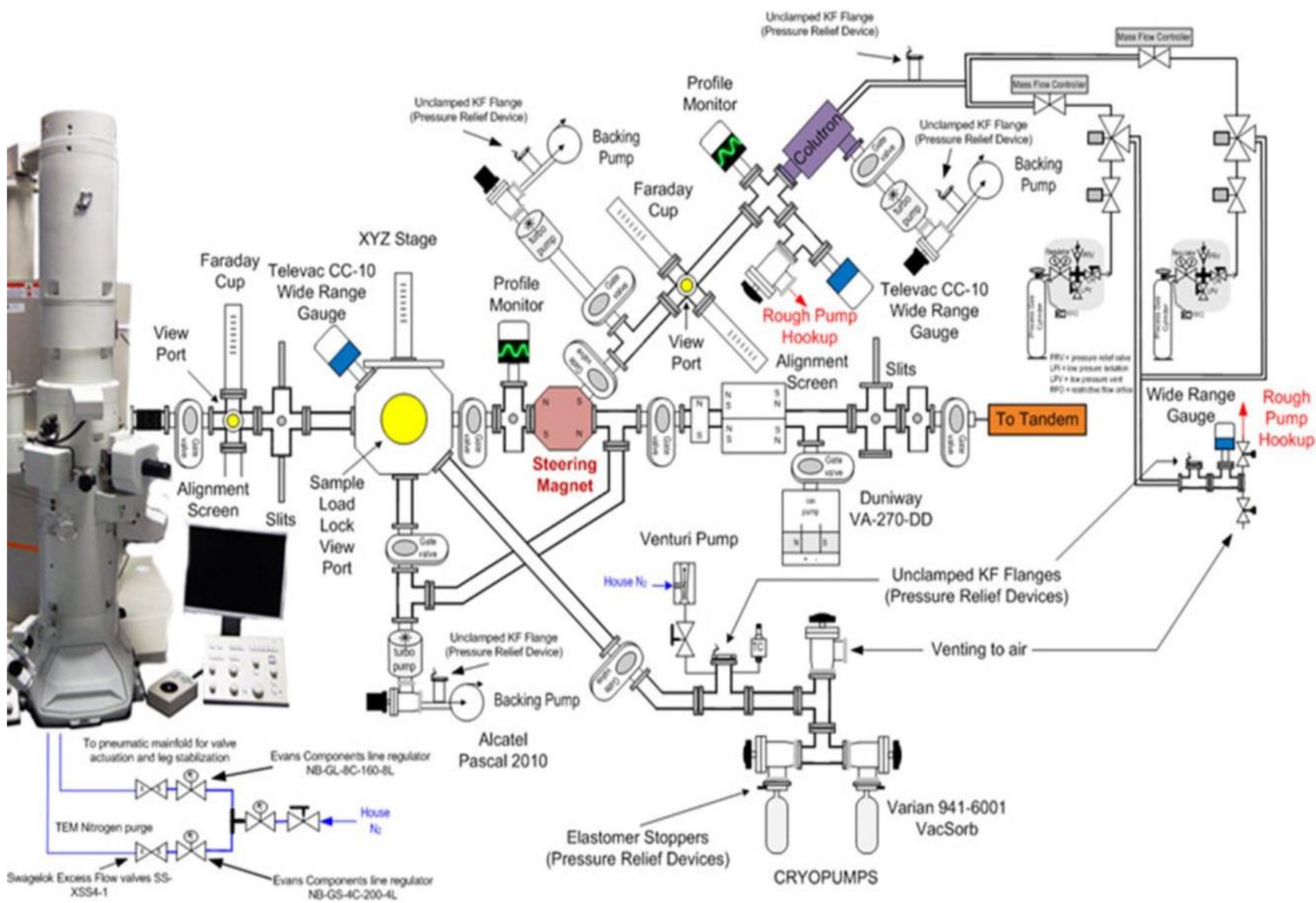
In situ Ion Irradiation TEM (I^3TEM) Facility

Collaborators: J.A. Scott

Proposed Capabilities

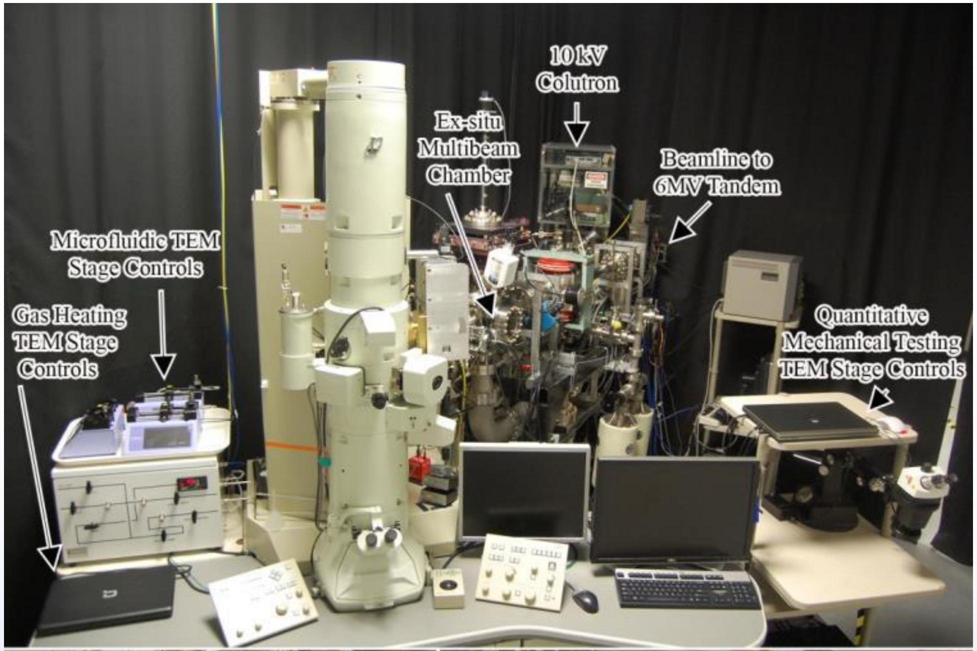
- 200 kV LaB_6 TEM
- Ion beams considered:
  - Range of Sputtered Ions
  - 10 keV D_2^+
  - 10 keV He^+
- All beams hit same location
- Nanosecond time resolution (DTEM)
- Procession scanning (EBSD in TEM)
- *In situ* PL, CL, and IBIL
- *In situ* vapor phase stage
- *In situ* liquid mixing stage
- *In situ* heating
- Tomography stage (2x)
- *In situ* cooling stage
- *In situ* electrical bias stage
- *In situ* straining stage

Schematic of the *In situ* TEM Beamline

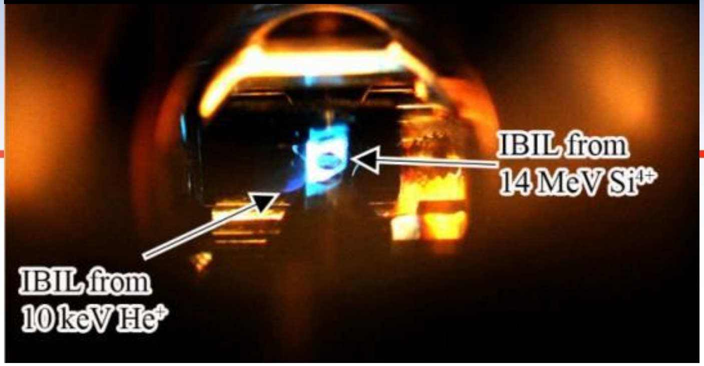


Sandia's Concurrent I³TEM Facility

10 kV Colutron - 200 kV TEM - 6 MV Tandem

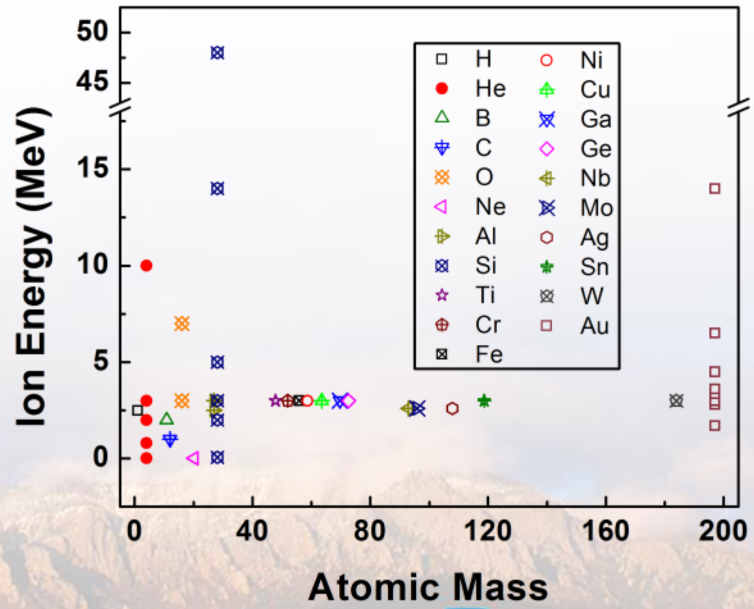


IBIL from a quartz stage inside the TEM



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

Ion species & energy introduced into the TEM



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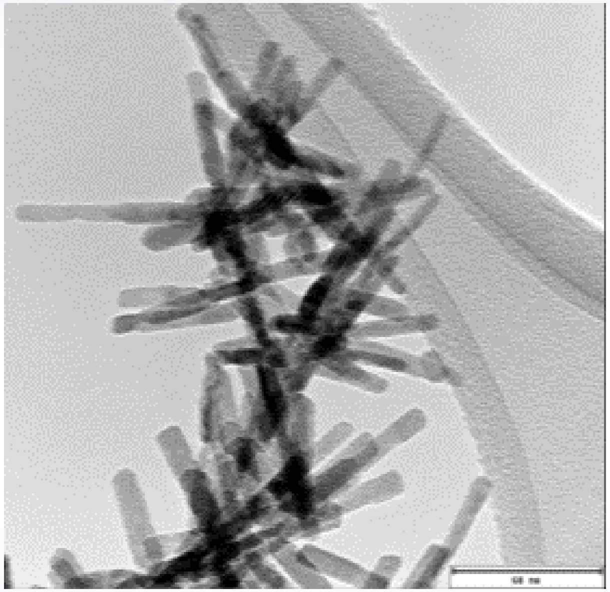
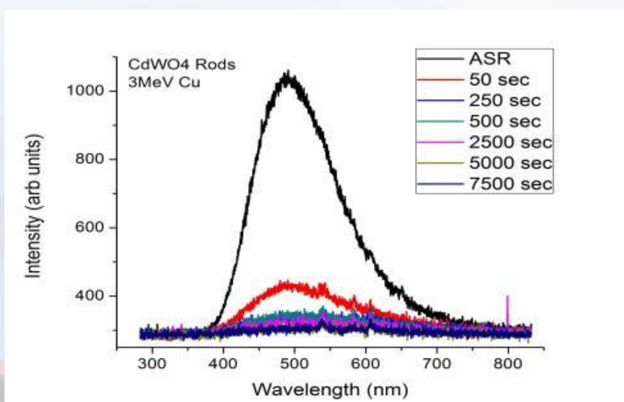
Radiation Tolerance is Needed in Advanced Scintillators for Non-proliferation Applications

Contributors: S.M. Hoppe, B.A. Hernandez-Sanchez, T. Boyle

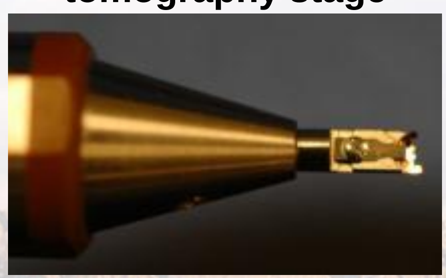


High-Z nanoparticles are promising, but are radiation sensitive

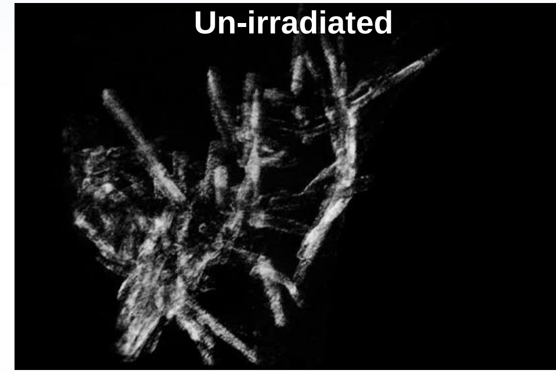


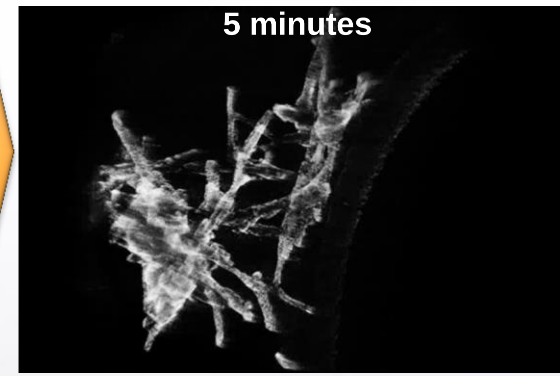
***In situ* Ion Irradiation TEM (I^3TEM)**

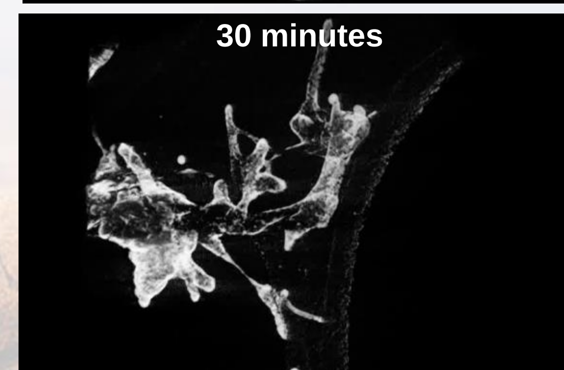
Hummingbird tomography stage



Tomography of Irradiated $CdWO_4$: 3 MeV Cu^{3+} at ~30 nA







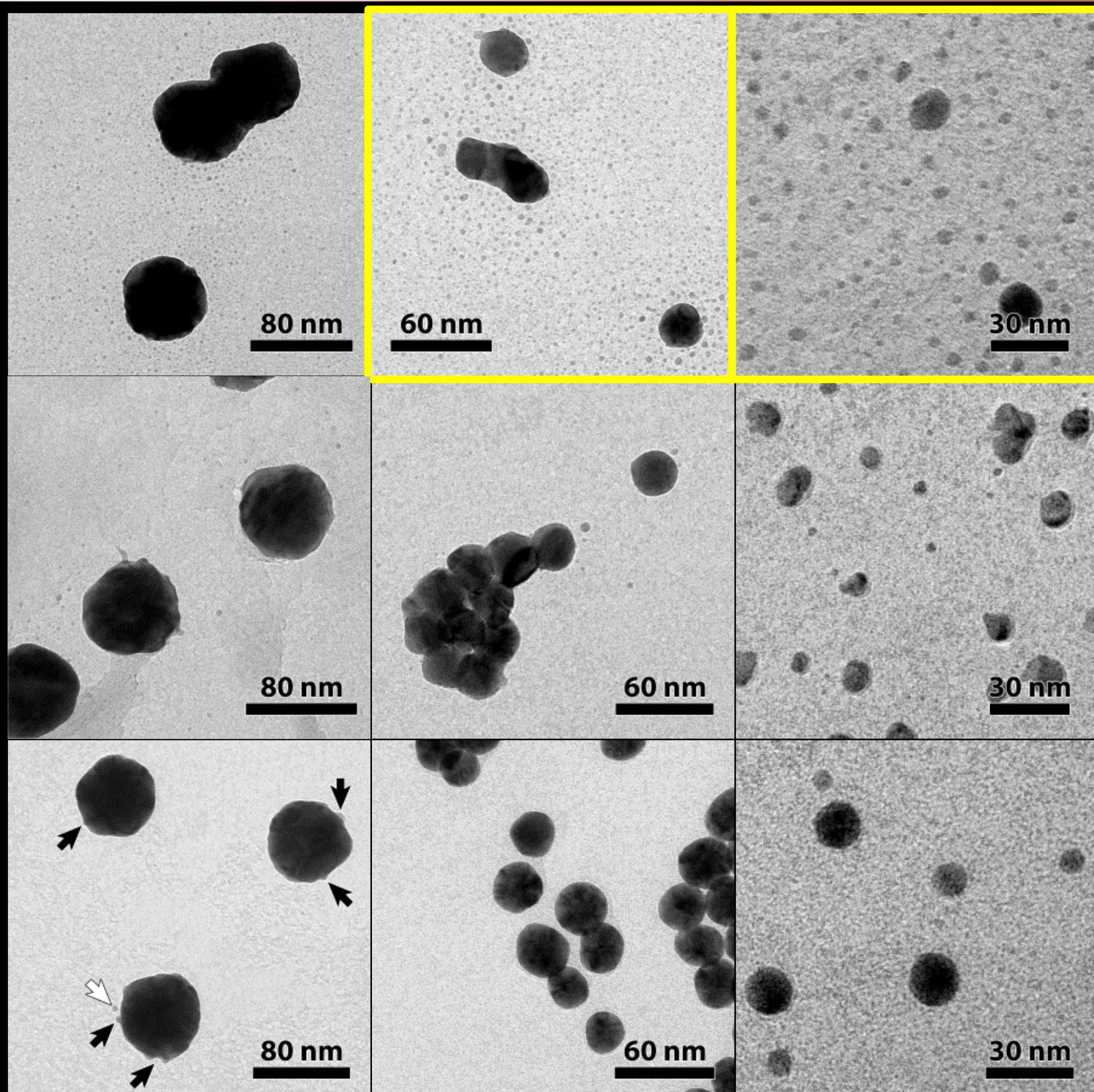
Cumulative Effects of Ion Irradiation as a Function of Ion Energy and Au Particle Size

60 nm | 20 nm | 5 nm

46 keV Au^{1-}
3.4×10^{14} /cm^2

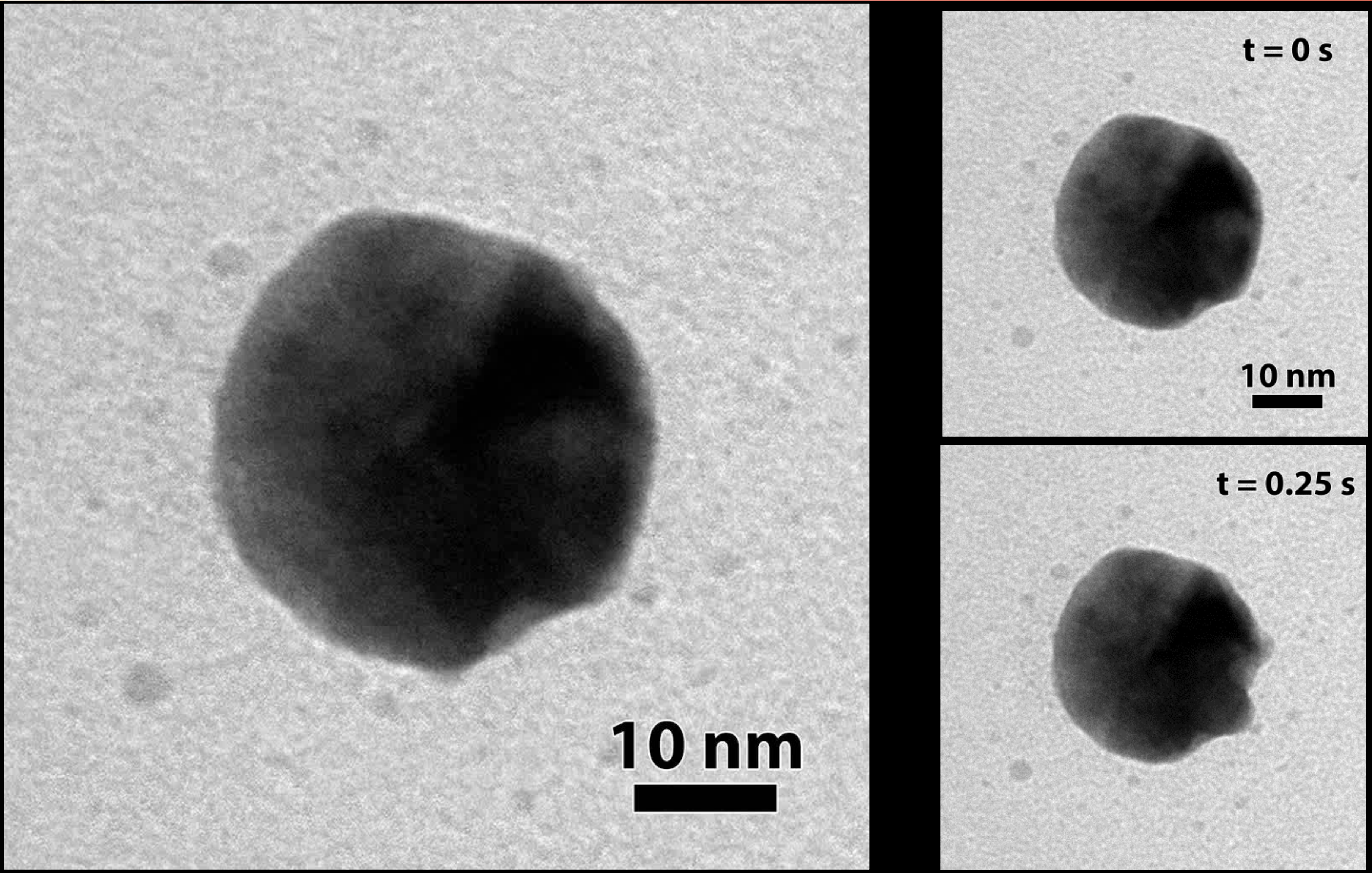
2.8 MeV Au^{4+}
4×10^{13} /cm^2

10 MeV Au^{8+}
1.3×10^{12} /cm^2

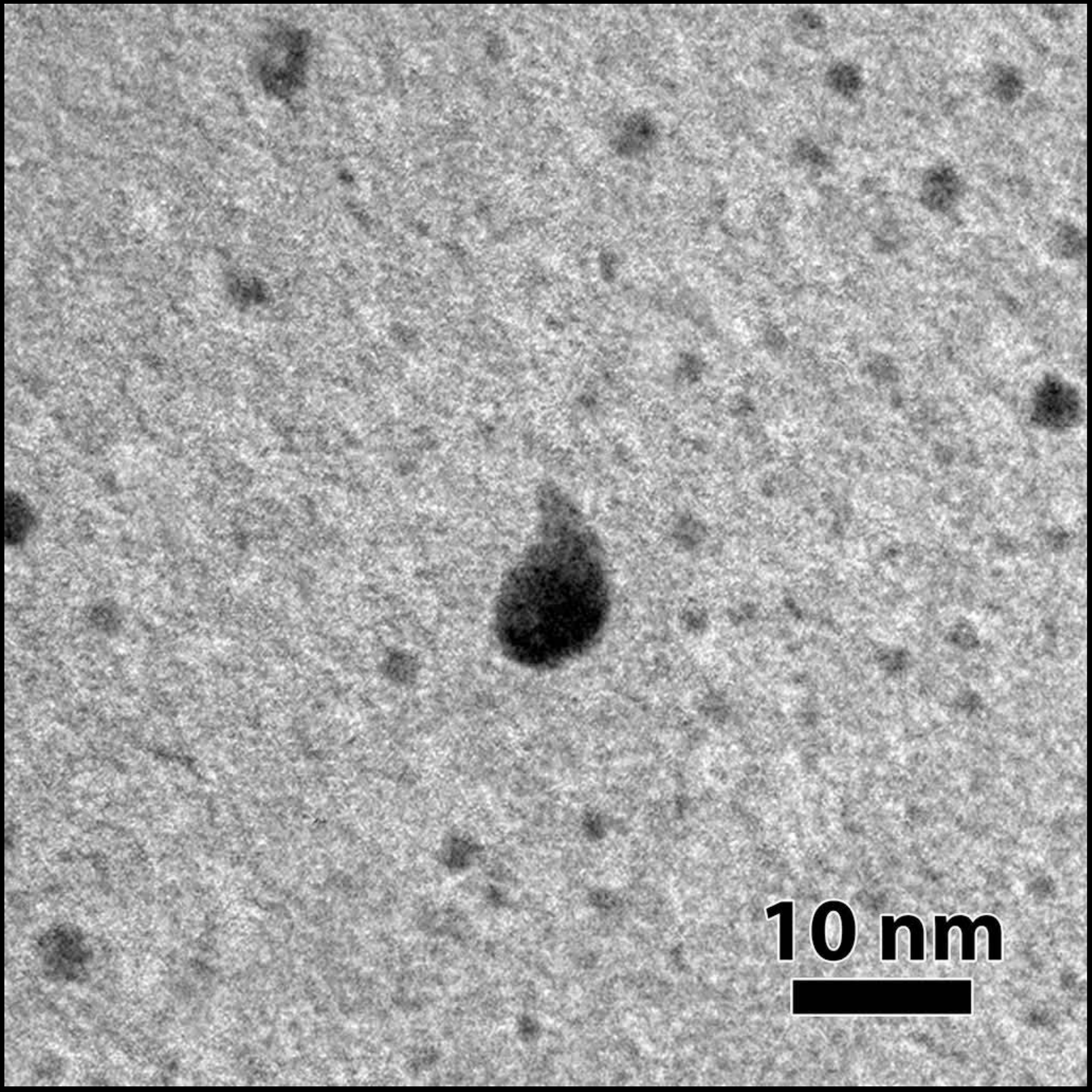


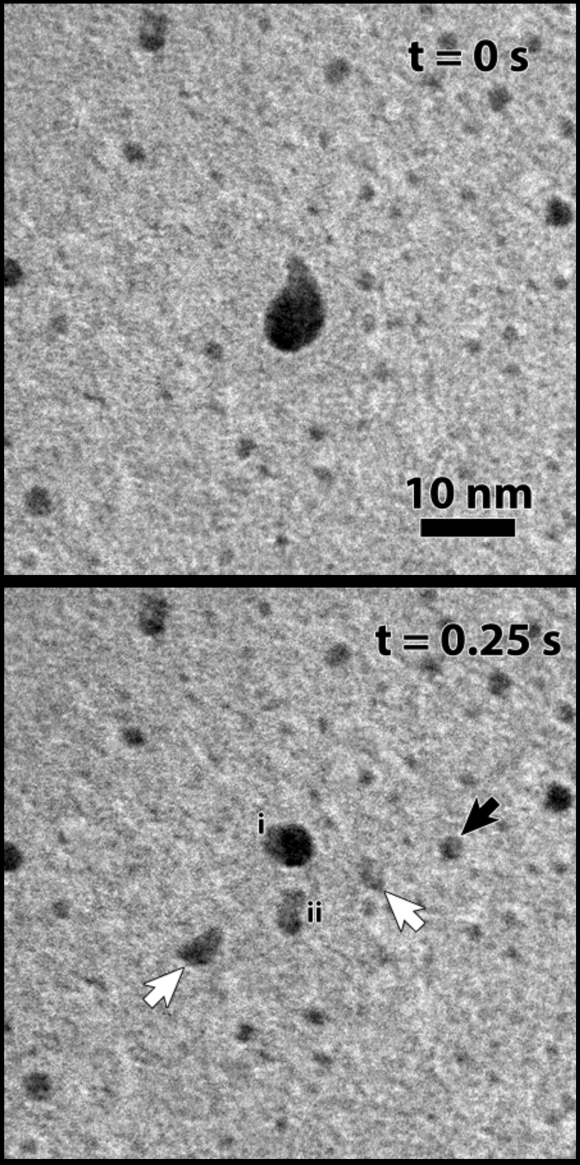
Particle and ion energy dictate the ratio of sputtering, particle motion, particle agglomeration, and other active mechanisms

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



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





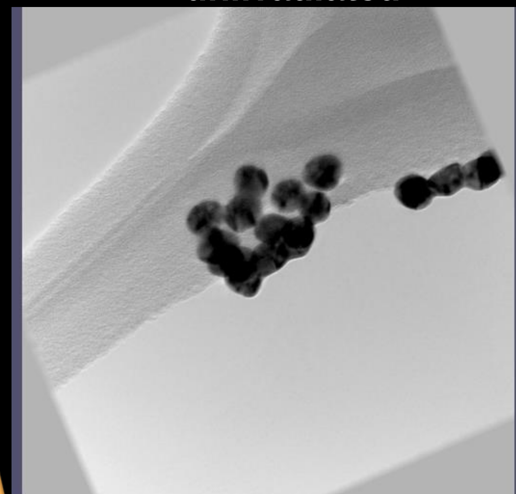
Advanced Microscopy Techniques Applied to Nanoparticles in Radiation Environments

Collaborators: S.M. Hoppe & T.J. Boyle

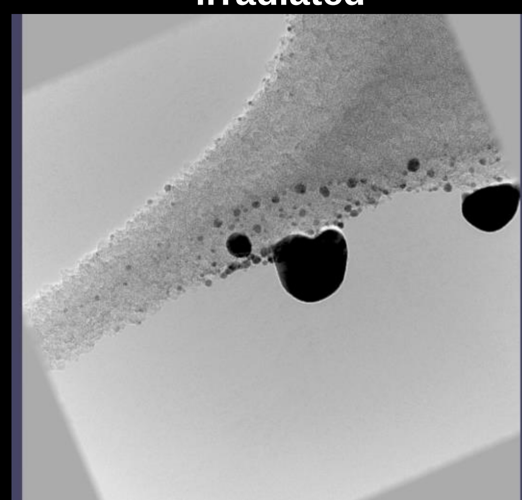
In situ Ion Irradiation TEM (I^{3}TEM)

Hummingbird tomography stage

Aligned Au NP tilt series - unirradiated

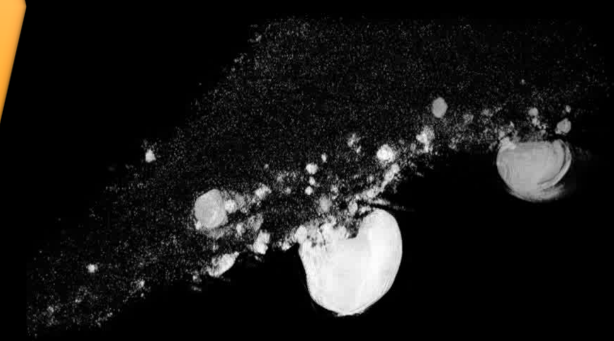


Aligned Au NP tilt series - irradiated



Unirradiated Au NP model

Irradiated Au NP model

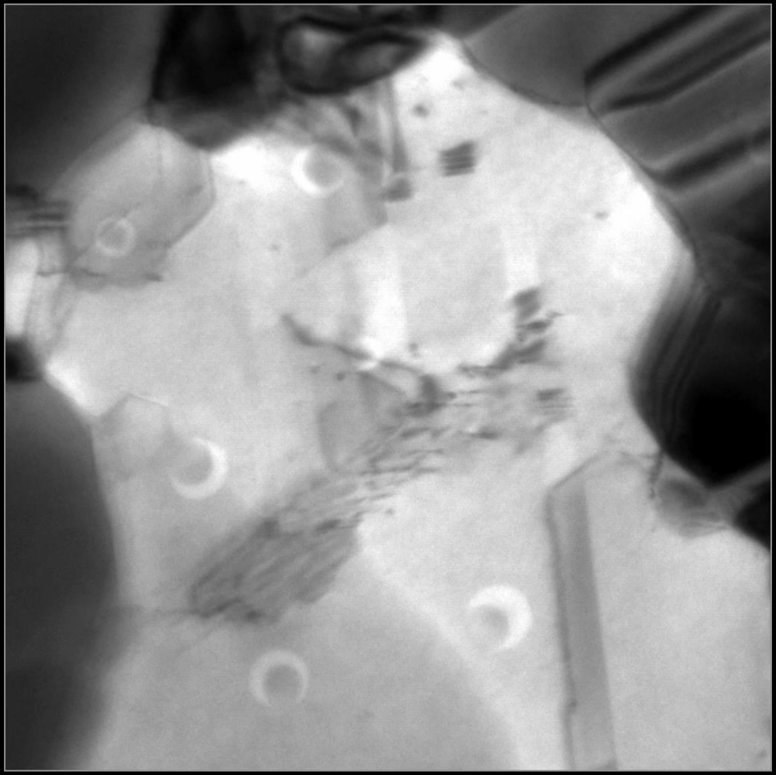


The application of advanced microscopy techniques to extreme environments provides exciting new research directions

Single Ion Strikes

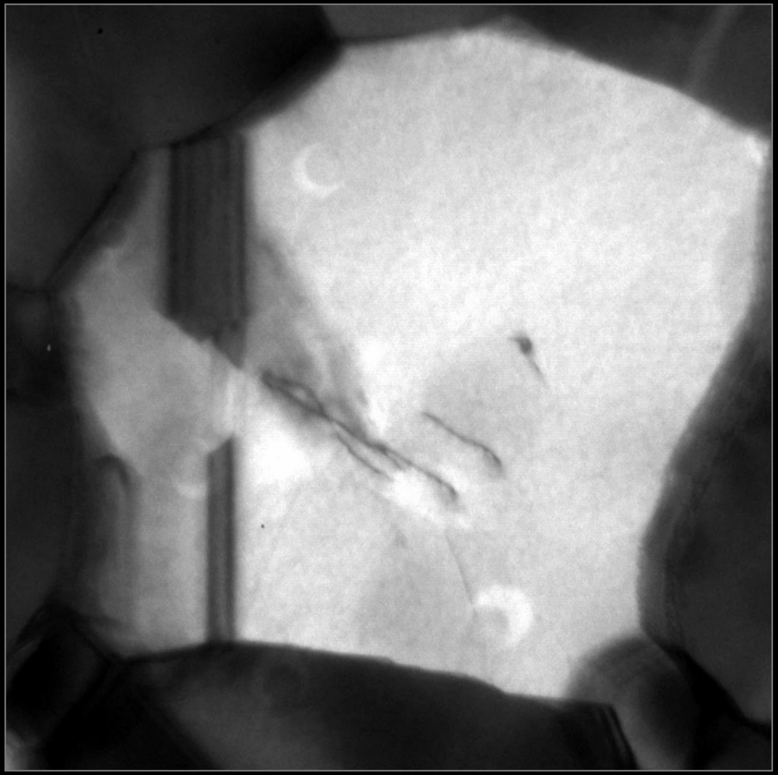
Collaborators: C. Chisholm & A. Minor

7.9 x 10^9 ions/cm^2/s



VS

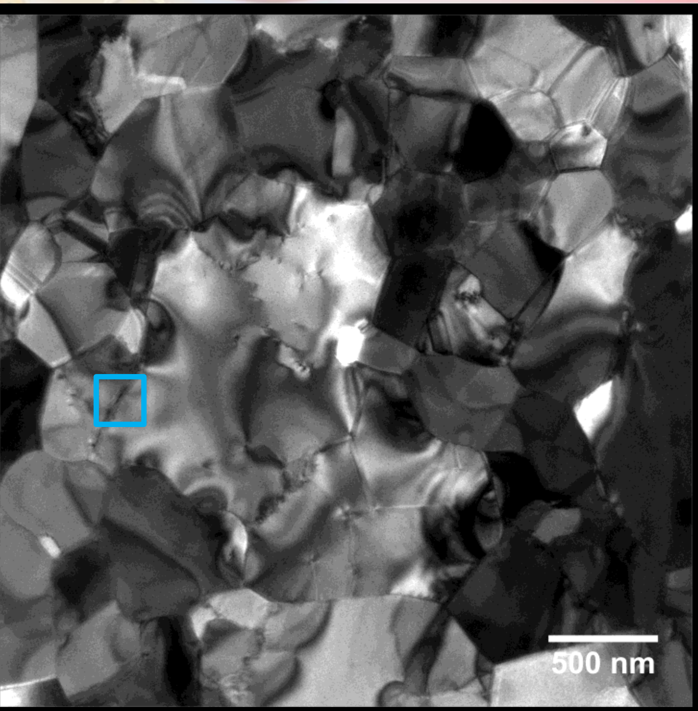
6.7 x 10^7 ions/cm^2/s



Improved vibrational and ion beam stability permits us to work at 400 kx or higher permitting imaging of single cascade events

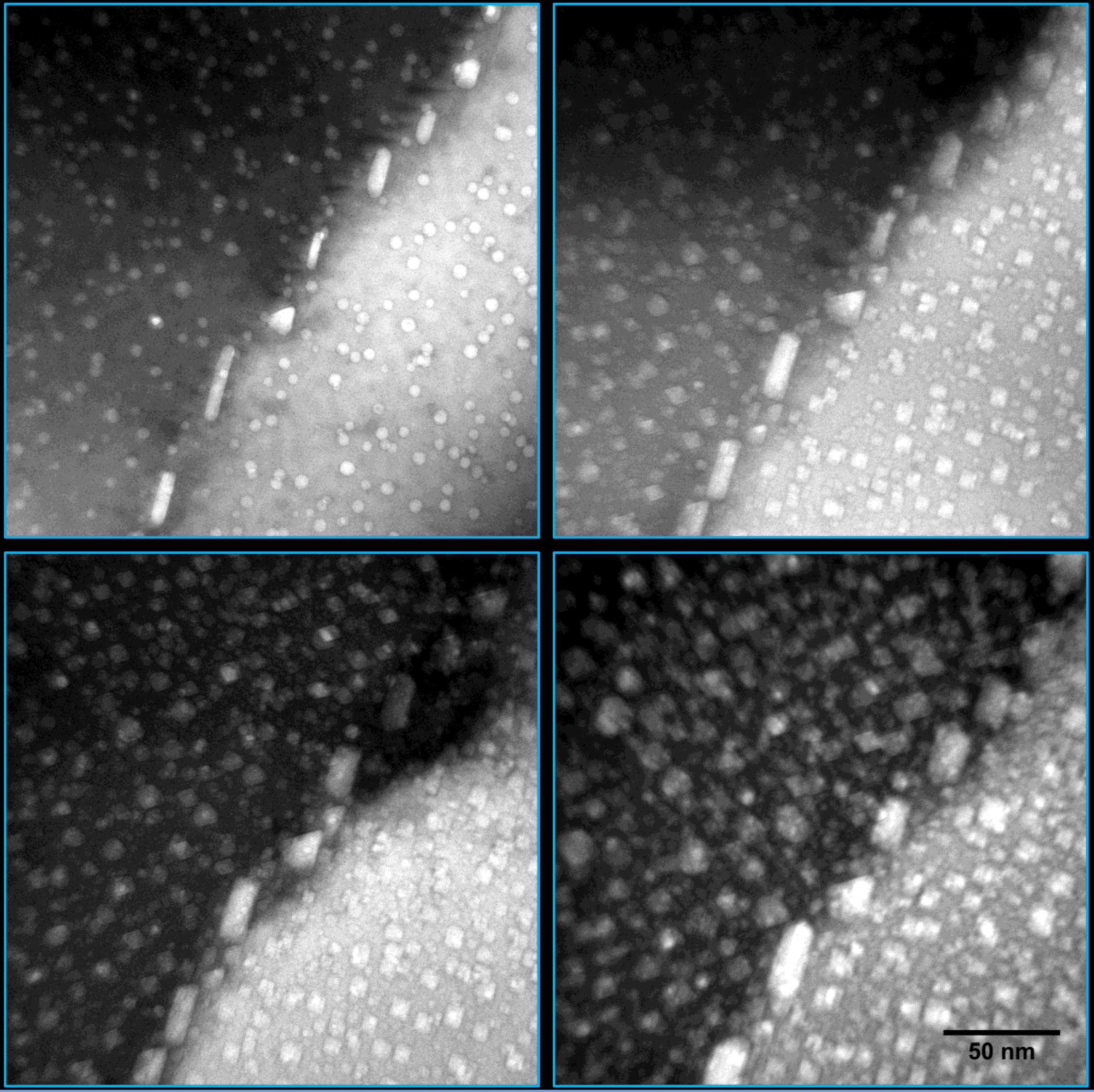
In situ Implantation

Collaborators: C. Chisholm & A. Minor

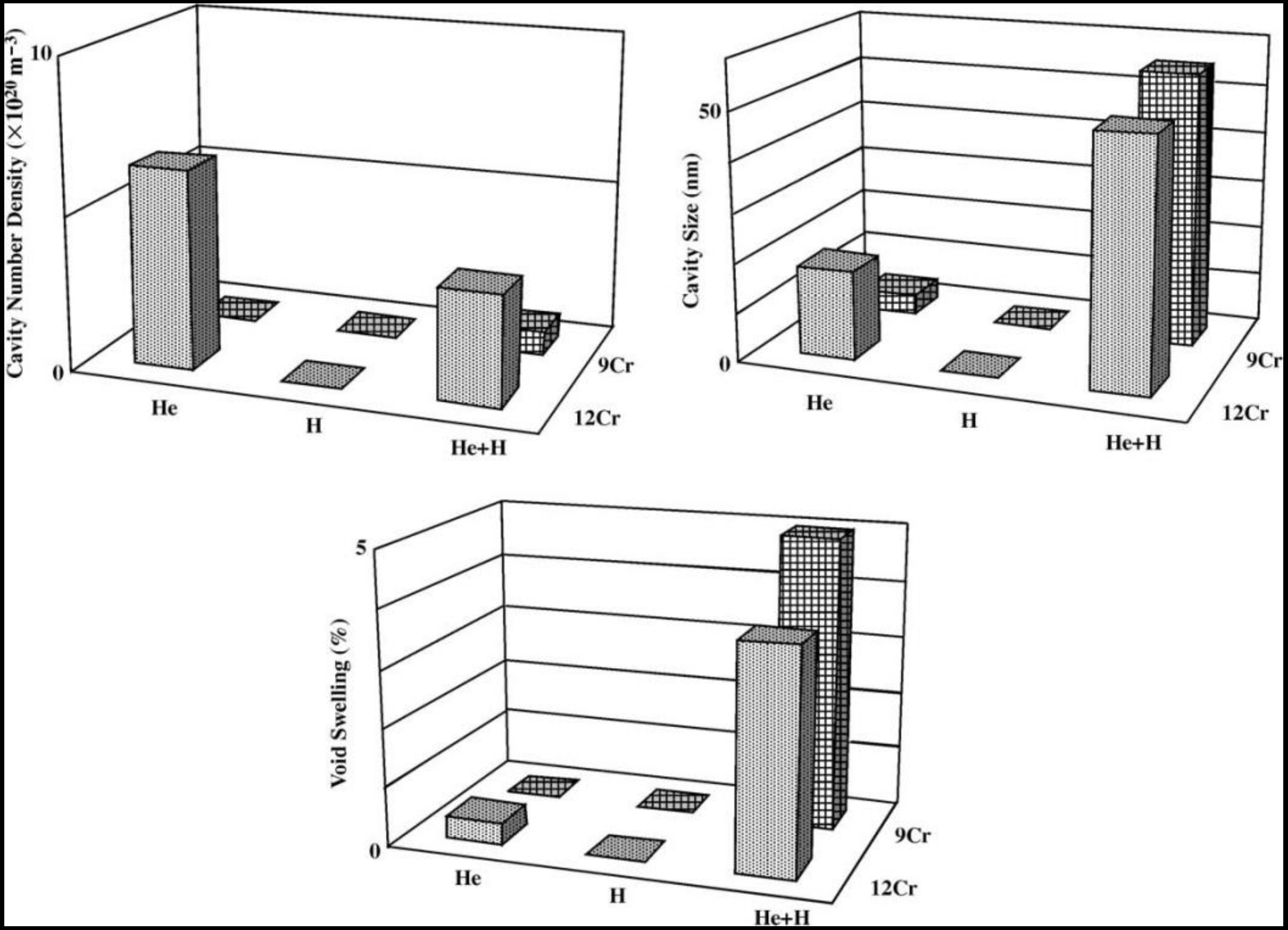


Gold thin-film implanted with 10keV He^{2+}

Result: porous microstructure



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"

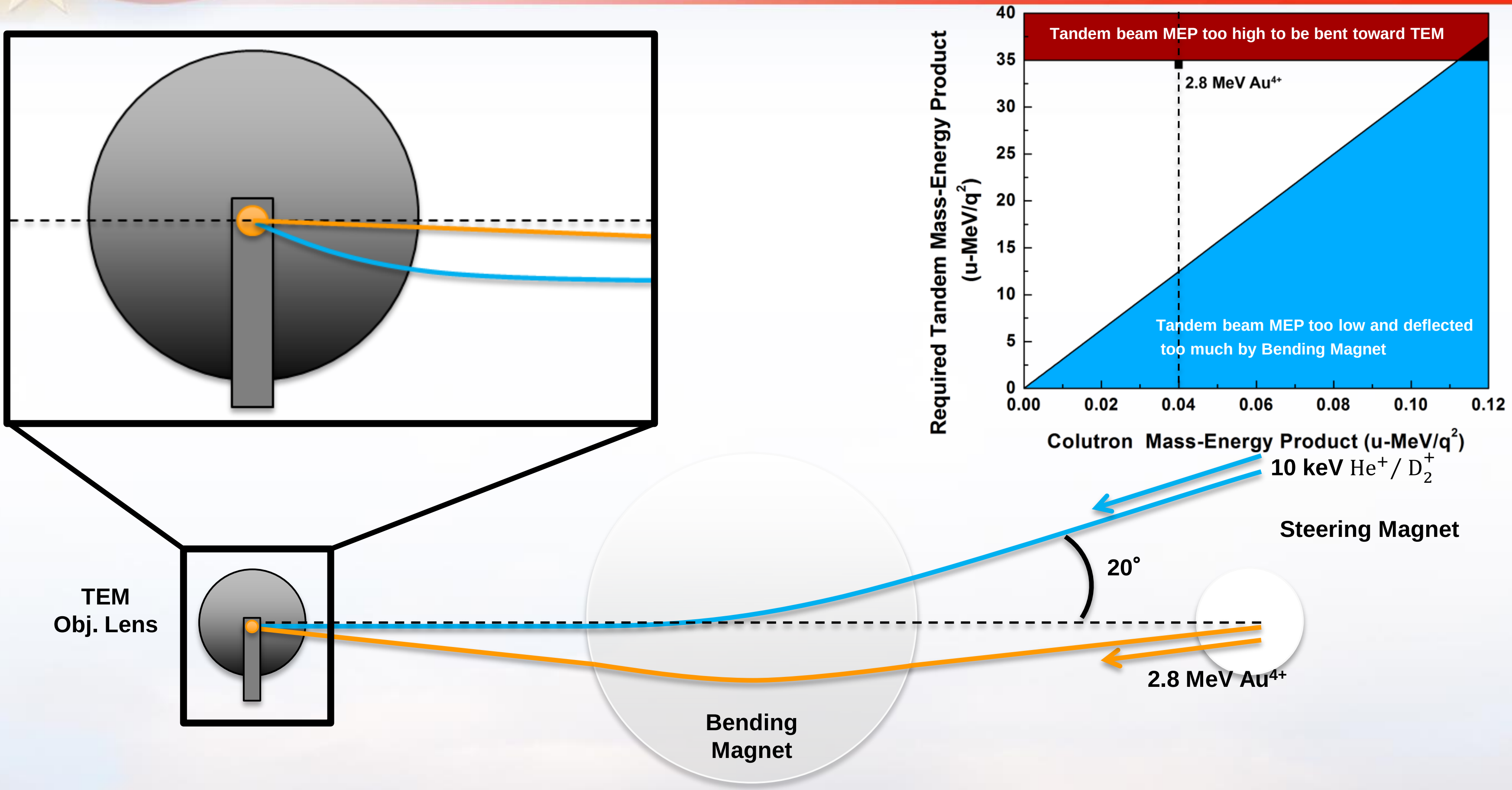
J. of Nuclear Materials 329-333 (2004) 294-298

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**

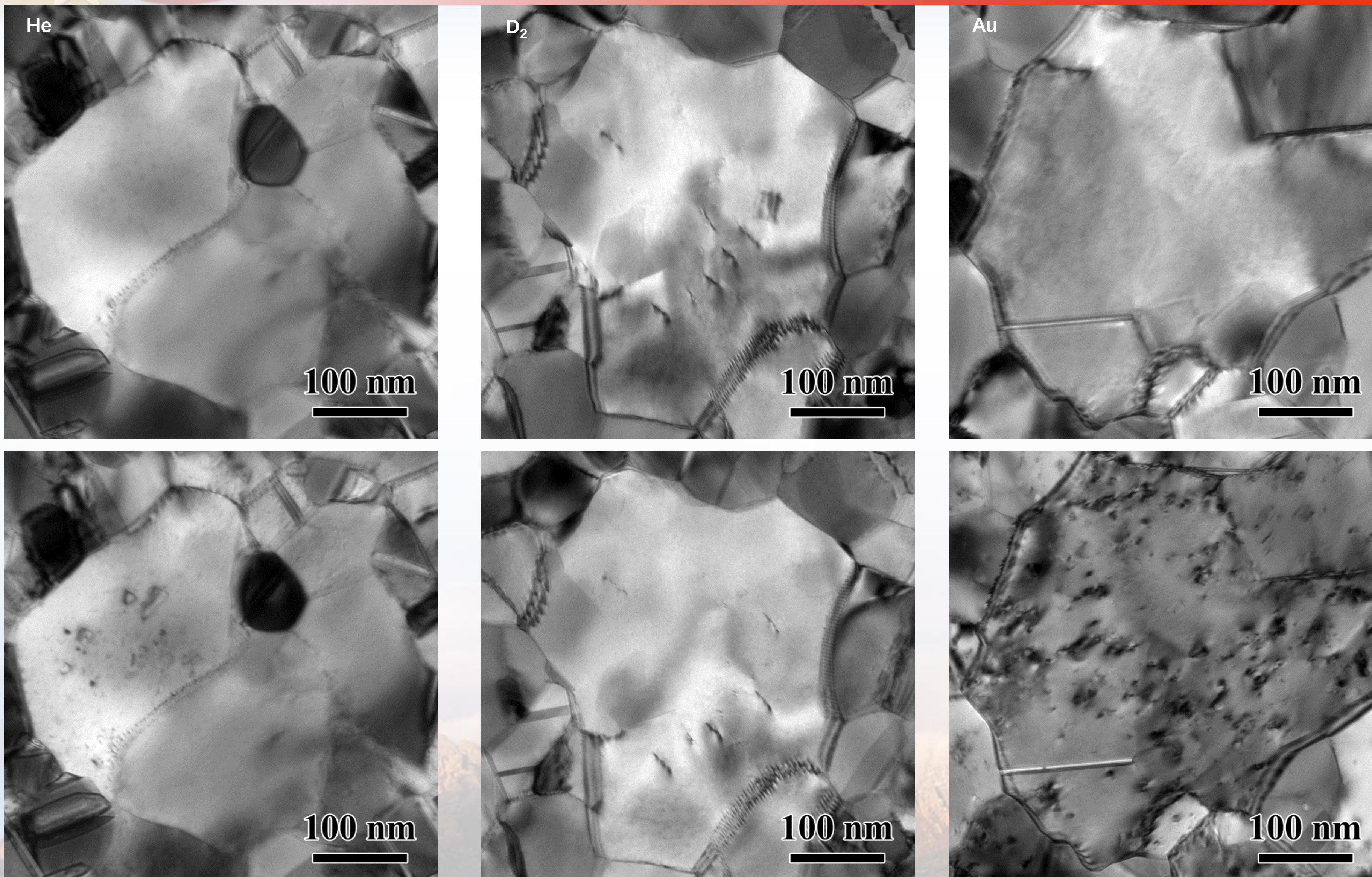
Difficulty of performing triple-beam irradiation has resulted in a limited number of facilities world wide

Modeling Beam Mixing and Deflection

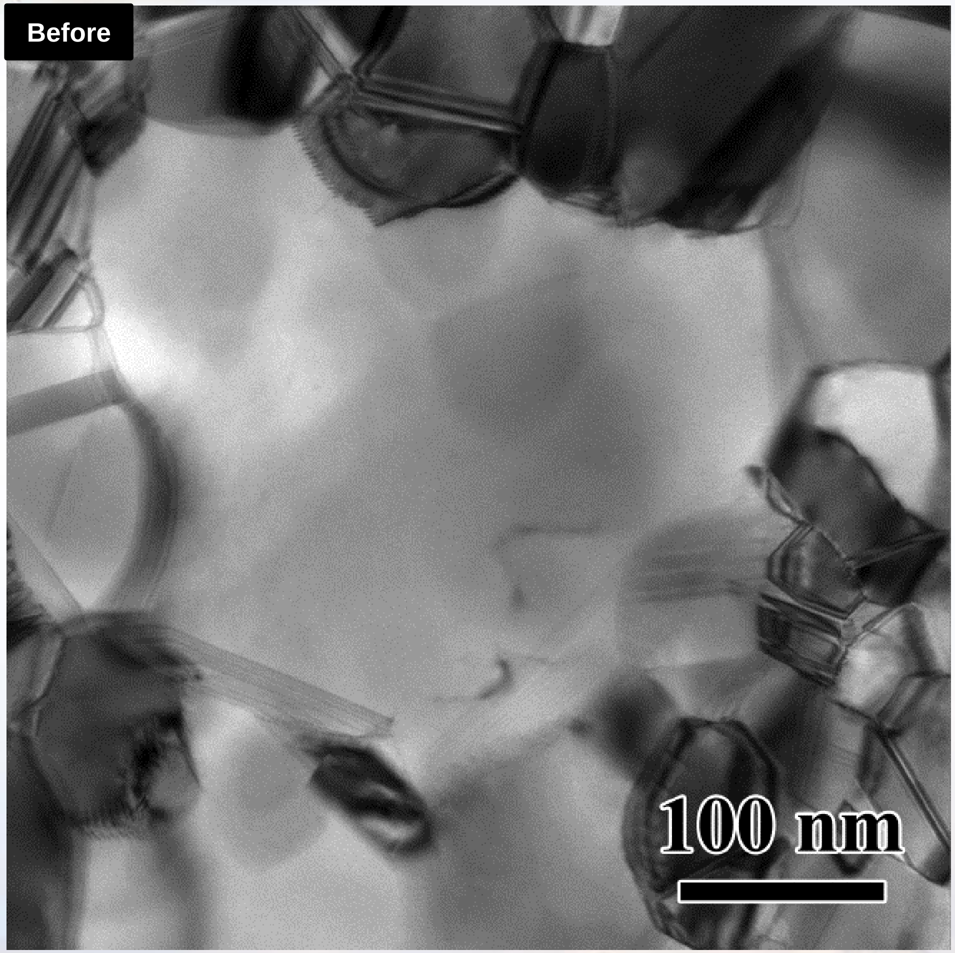


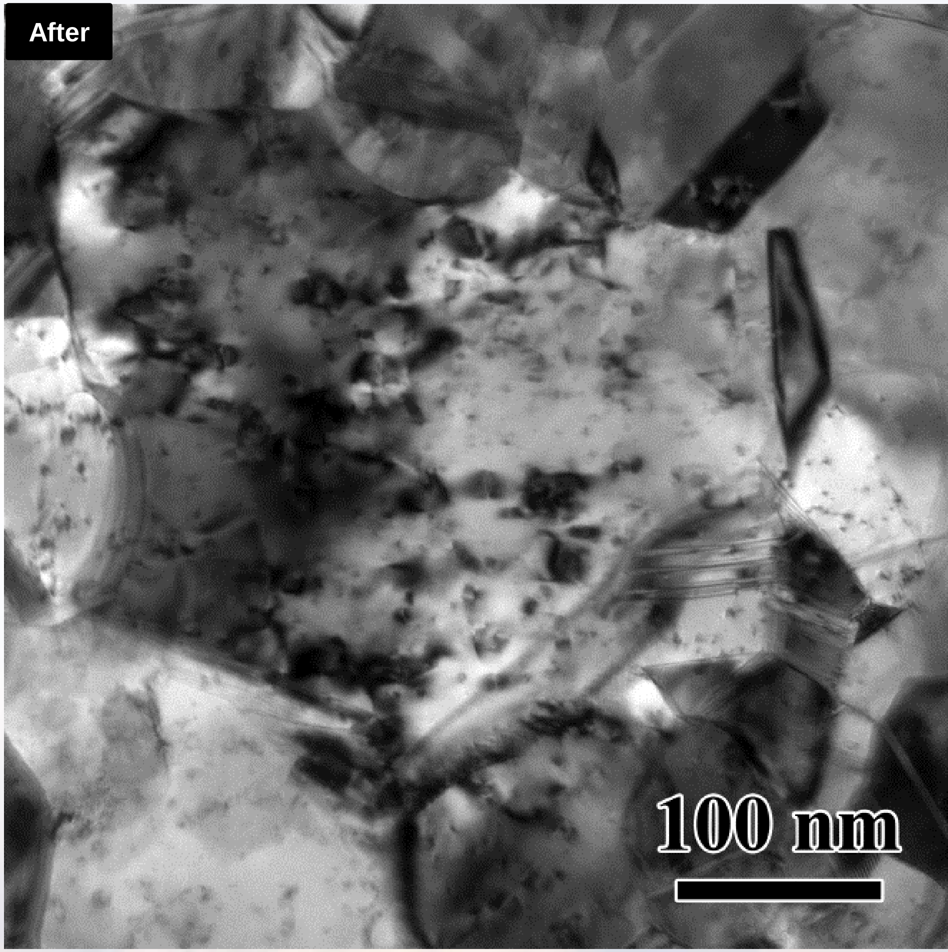
- Must compensate for deflection of Tandem beam by bending magnet
  Colutron beams deflected by the TEM objective lens
- Insignificant deflection of Tandem beams
- With 10 keV He/D_2 we can use Tandem beams ≳13 MeV/q^2
- Au, He, and D_2 ions all reach the sample concurrently

Aligned Individual Beams



Concurrent 10 keV He, 10 keV D_2, and 3 MeV Au



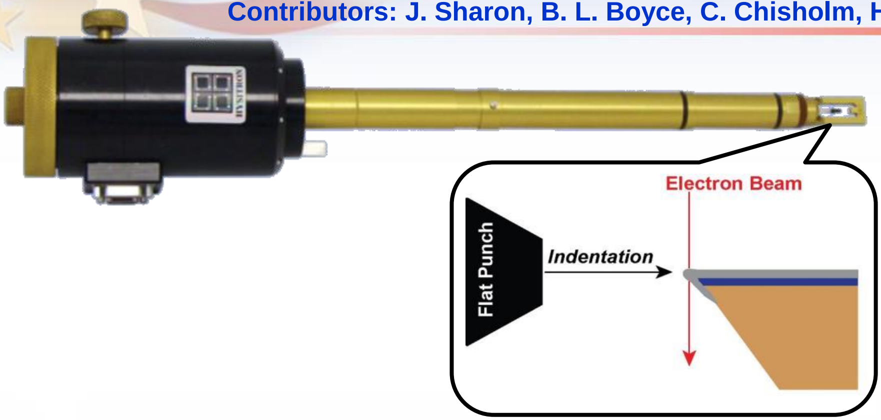


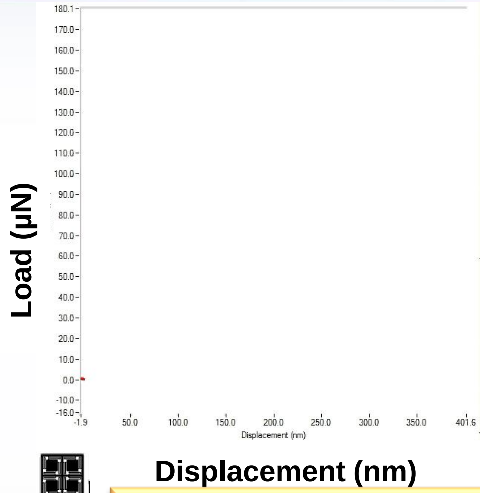
In-situ triple beam He, D_2, and Au beam irradiation has been demonstrated on Sandia's I^{3}TEM!

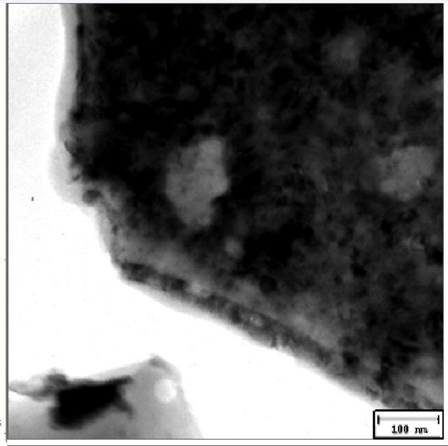
Extensive work is still needed to understand the defect strucutre evolution that has been observed.

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.



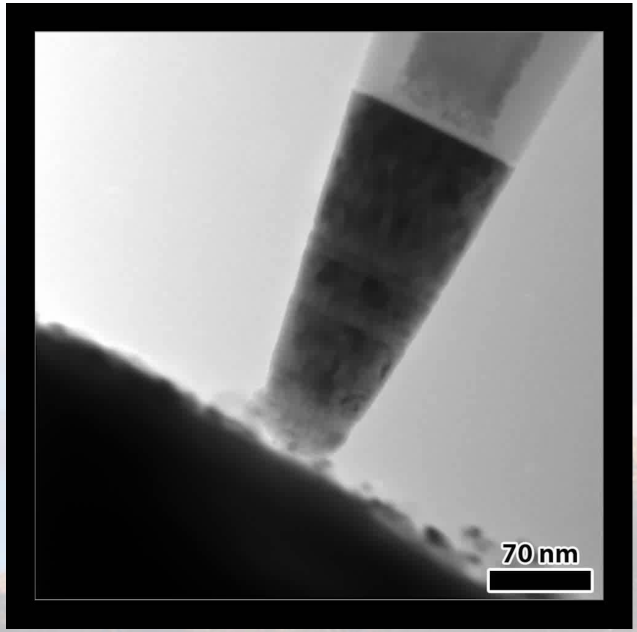


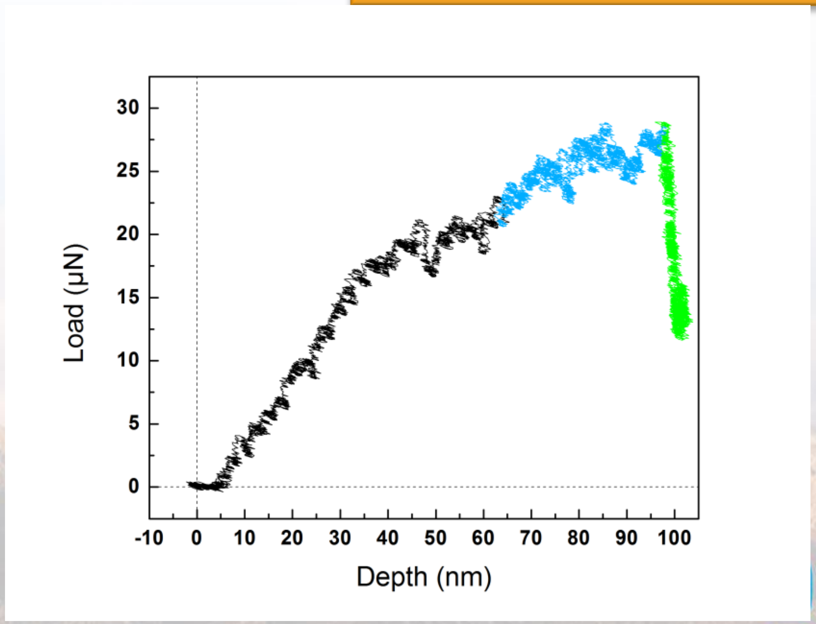


Range of Mechanical Testing Techniques

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

Fundamentals of Mechanical Properties

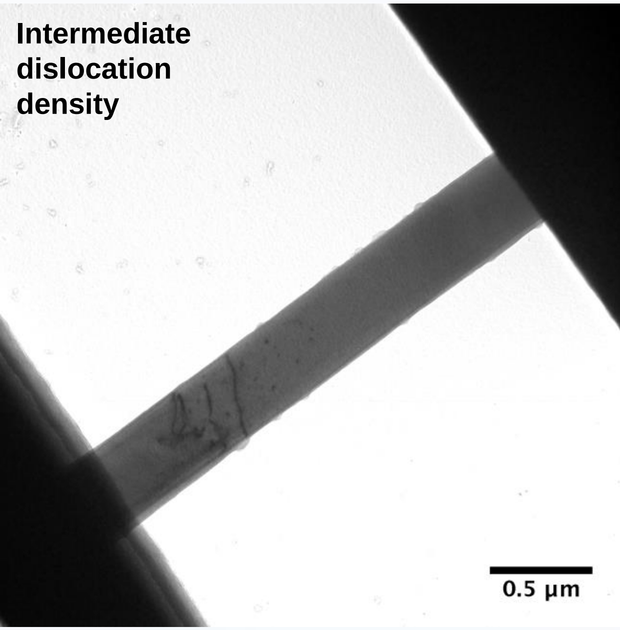


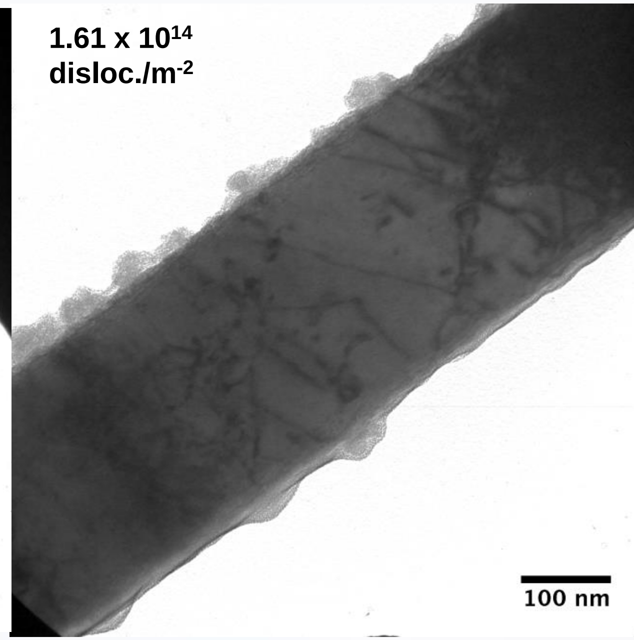


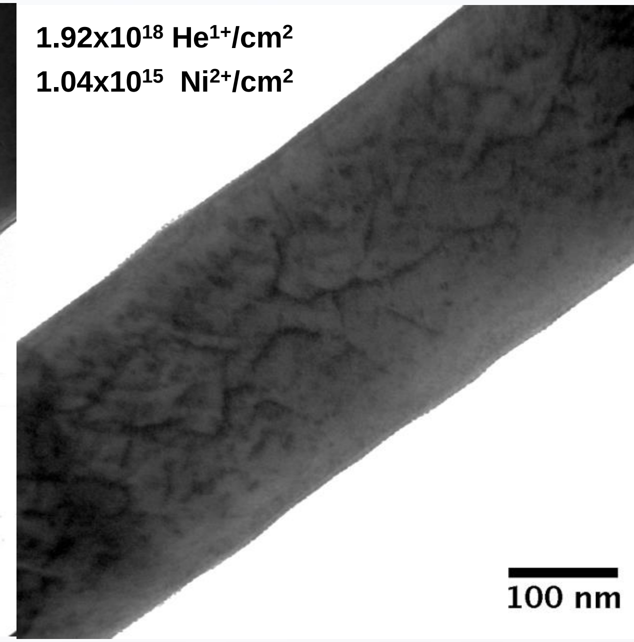
- 0.5 nm/s loading rate
- Trapezoid load function
- 60s load/60s hold/60s unload

Next Steps: *In situ* TEM Quantitative Mechanical Testing

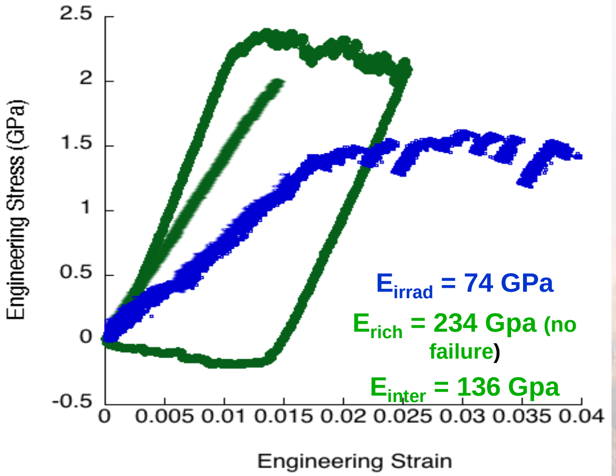
Contributors: C. Chisholm, H. Bei, E.P. George, P. Hosemann, & A.M. Minor





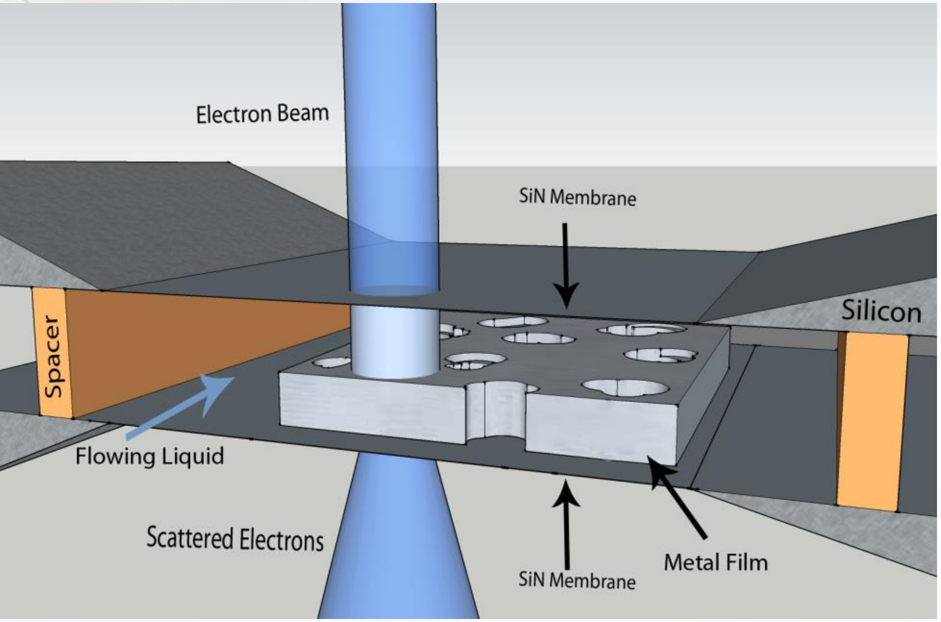


Work has started by looking sequentially at the quantitative effects of ion irradiation on mechanical properties utilizing in-situ ion irradiation TEM and in-situ TEM straining.



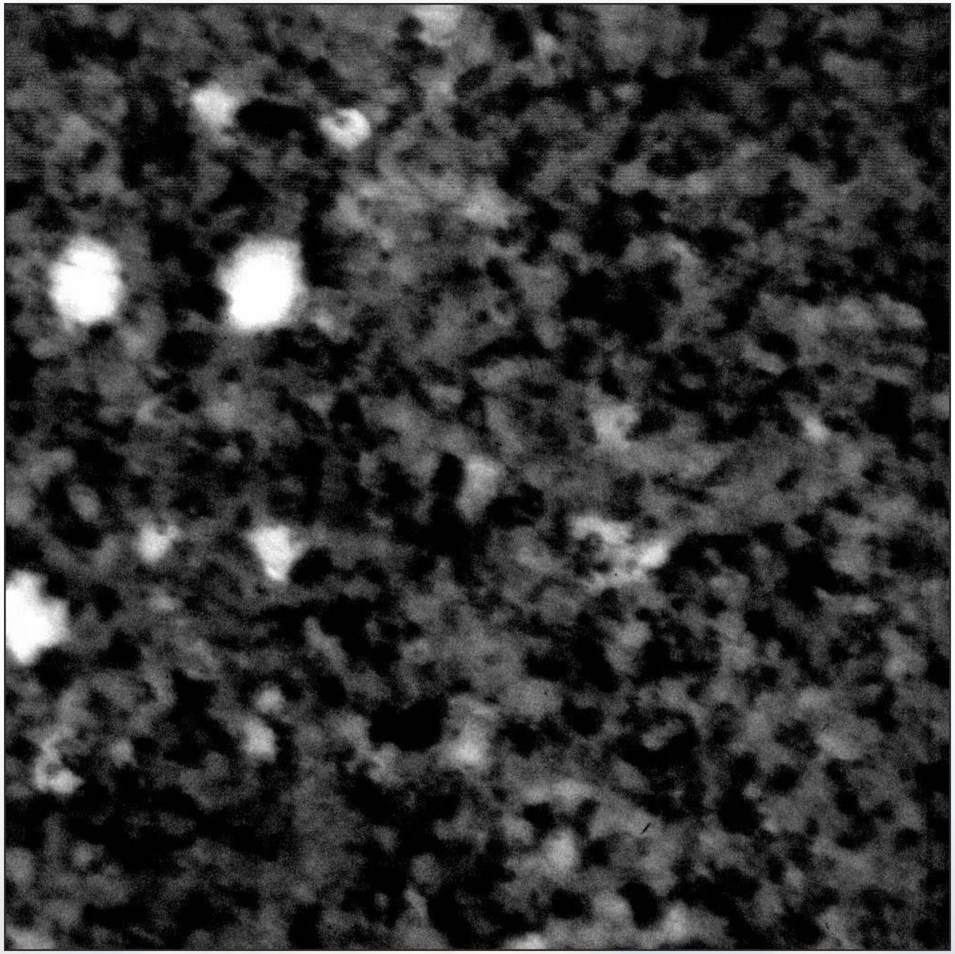
Next steps: *In situ* TEM Corrosion

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



Microfluidic Stage

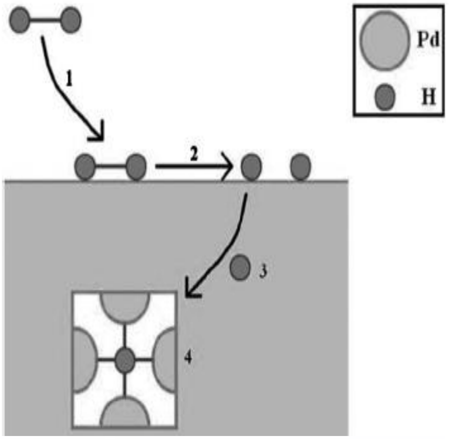
- **Mixing of two or more channels**
- **Continuous observation of the reaction channel**
- **Chamber dimensions are controllable**
- **Films can be directly deposited on the electron transparent SiN membrane**



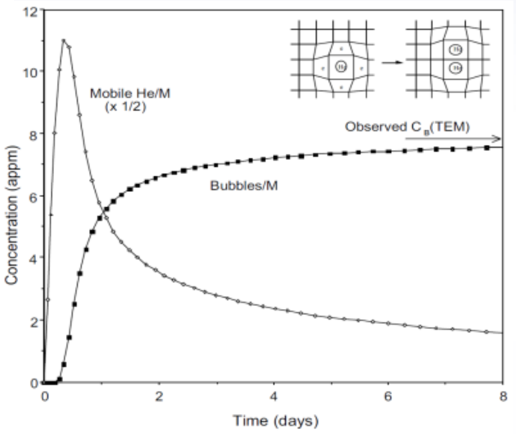
Pitting mechanisms during dilute flow of acetic acid over 99.95% nc-PLD Fe involves many grains.

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

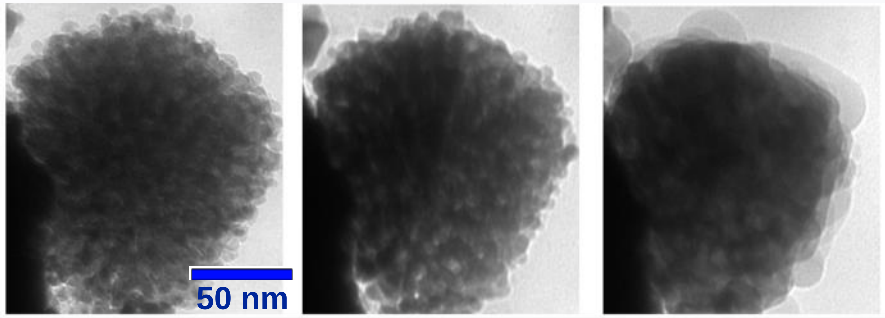


Cowgill, D., Fusion Sci. & Tech., 28 (2005) p. 539
Trinkaus, H. *et al*., JNM (2003) p. 229
Thiebaut, S. *et al*. JNM (2000) p. 217

Vapor-Phase Heating TEM Stage

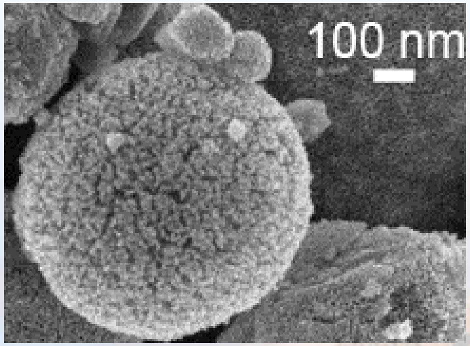
- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Compatible with MS and other analytical tools

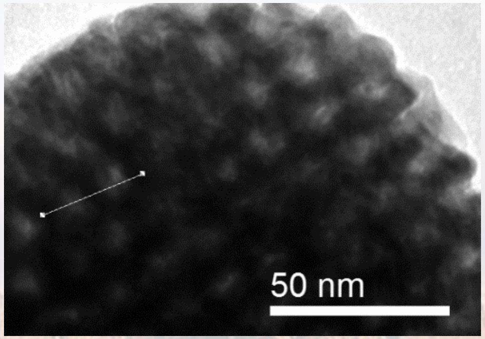
- 1 atm H_2 after several pulses to specified temp.



125° C 200° C 300° C

Harmful effects may be mitigated in nanoporous Pd

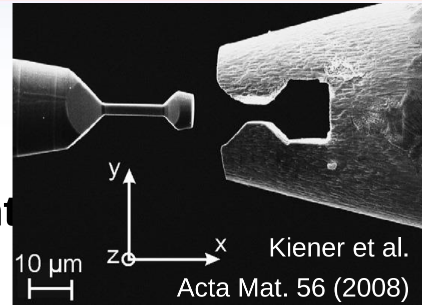


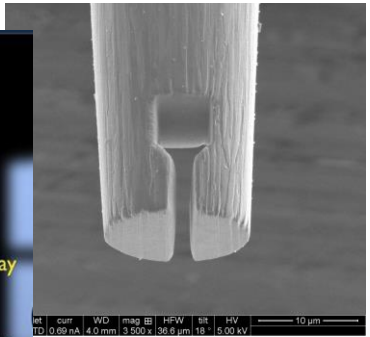


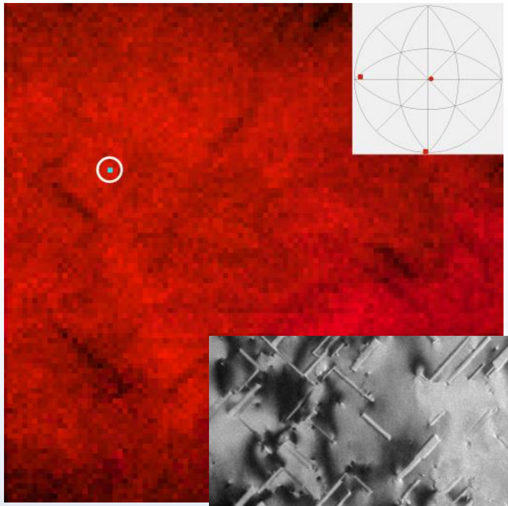
New *in situ* atmospheric heating experiments provide great insight into nanoporous Pd stability

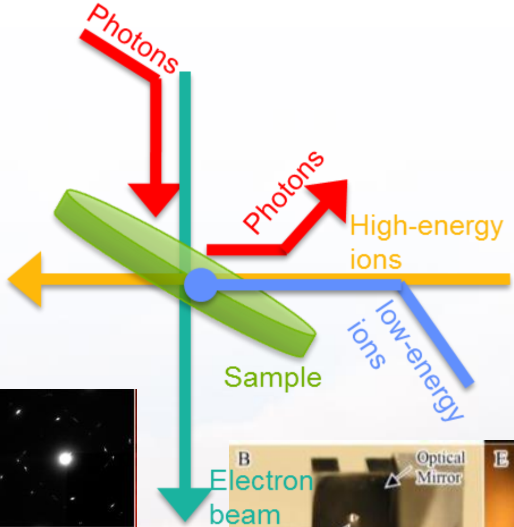
Future Directions Under Pursuit: Increased Data and Combinations of Enviroments

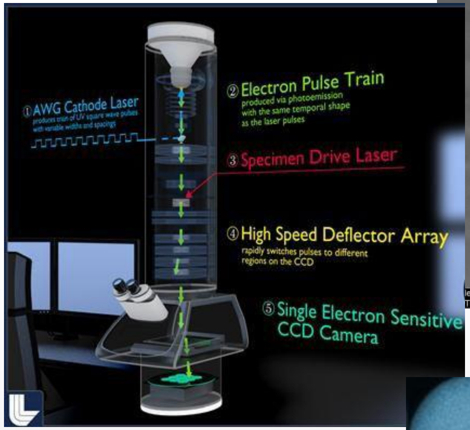
1. **In-situ TEM CL, IBIL (currently capable)**
2. ***In situ* ion irradiation TEM in liquid or gas (currently capable)**
3. **PED: Local texture characterization (recently installed)**
4. **Quantitative in-situ tensile/creep experiments (Sample in development**
5. **DTEM: Nanosecond resolution (laser optics needed)**

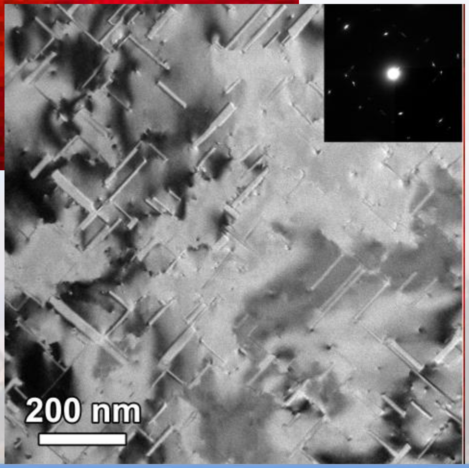


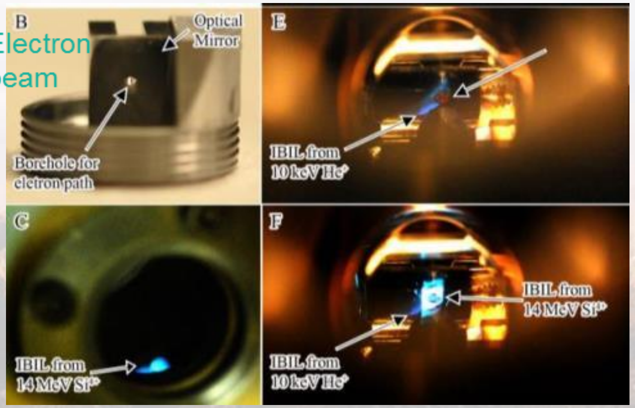


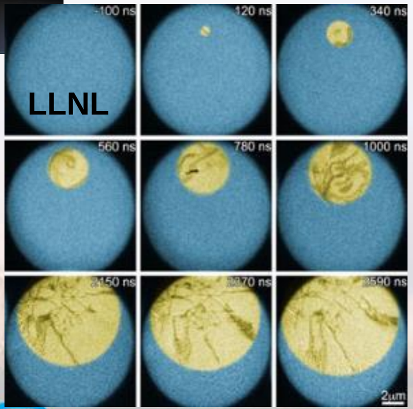












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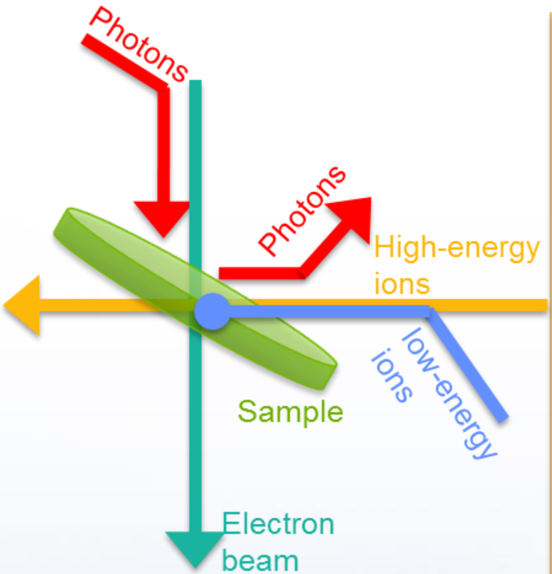
Summary



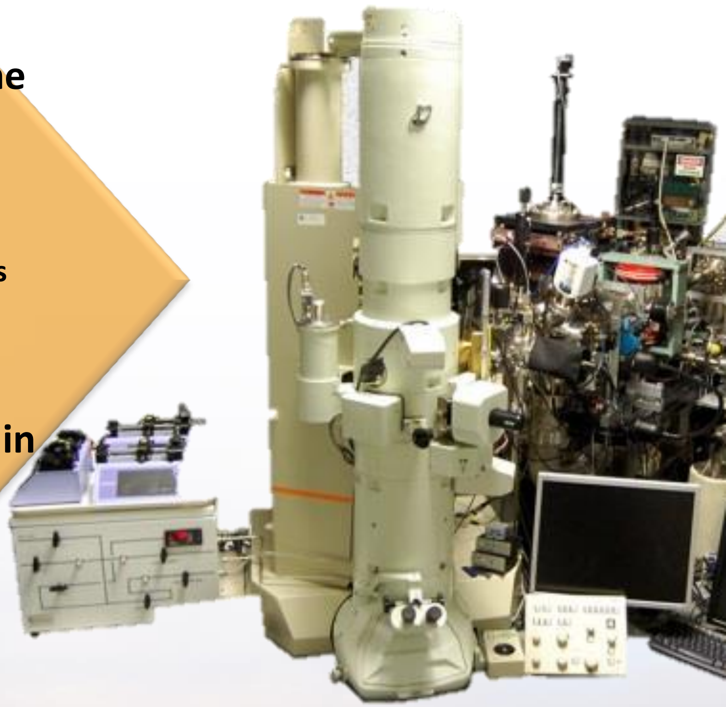
The Ion Beam Lab at Sandia National Laboratories applies a variety of nanoscale tools to a wealth of problems

Sandia's I^{3}TEM is one of a few in the world

- ***In situ* irradiation from H to Au**
- ***In situ* gas implantation**
- **Combinations of in-situ techniques**

I^{3}TEM can provide fundamental understanding to key mechanisms in a variety of extreme conditions

The I^{3}TEM capability are still being expanded…



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