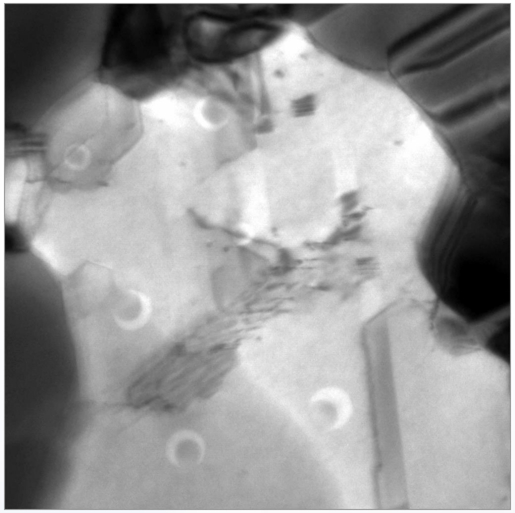
In situ Nanoscale Testing to Validate and Elucidate Mechanism for Predictive Modeling

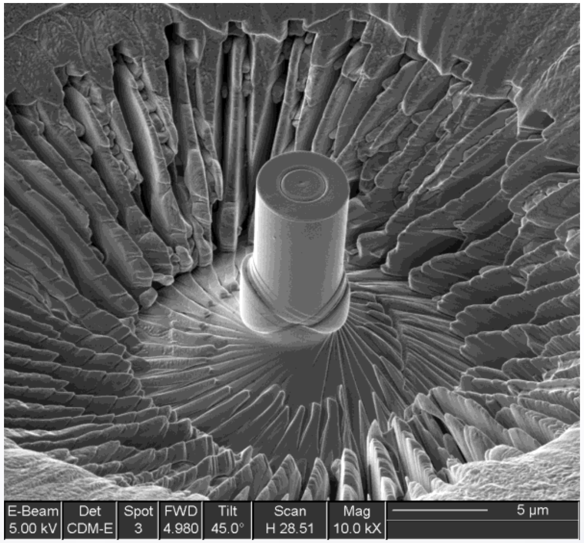
SAND2014-17696PE

K. Hattar

Sandia National Laboratories

September 11, 2014





Collaborators:

- IBL: D. Bufford, D. Buller, S. Pratt, S. Rajasekhara, J. Villone, and all the IBL staff
- Sandia: T.E. Buchheit, B. Boyce, T.J. Boyle, F.P. Doty, P. Feng, S. Goods, B.A. Hernandez-Sanchez, A.C. Kilgo, P.G. Kotula, J. Puskar, M.J. Rye, J.A. Scott, P. Yang
- External: N. Li, A. Misra, L.N. Brewer, S. Maloy, A. McGinnis, P. Rossi, Protochips Inc.

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

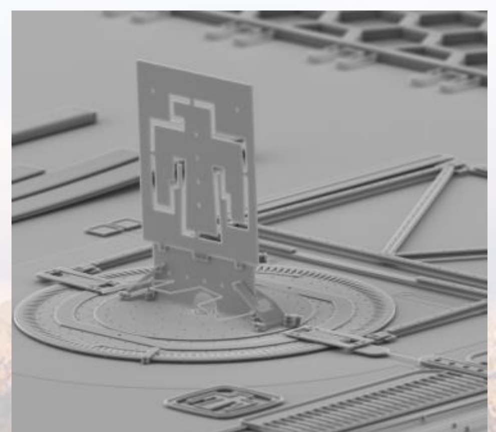
What is Sandia National Laboratories?


96% of total NW parts

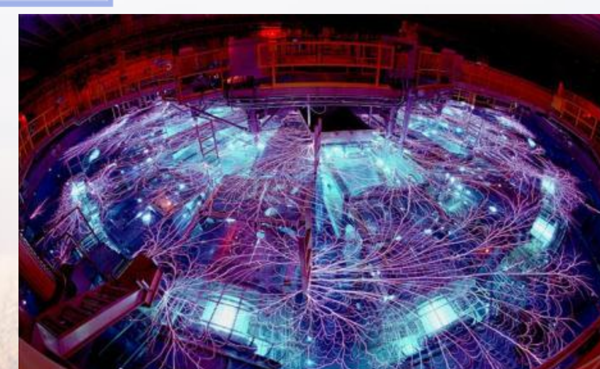
4 Mission Areas

- Nuclear Weapons
- Defense Systems and Assessments
- Energy, Resources, and Nonproliferation
- Homeland Security and Defense

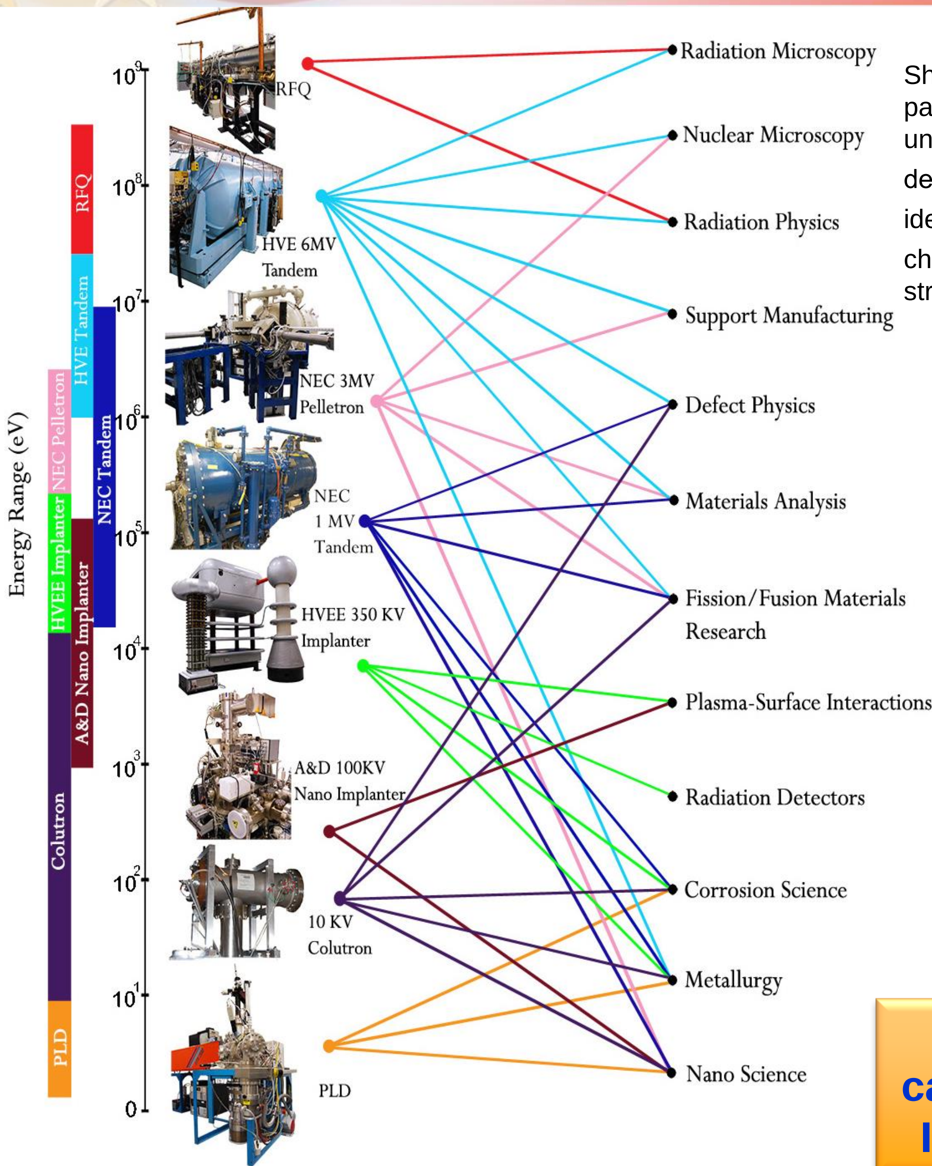

Sled track


Clean room invented at SNL in 1963


Renewable and alternative energy


Z machine:
the world’s most powerful X-ray source

Sandia's Ion Beam Laboratory

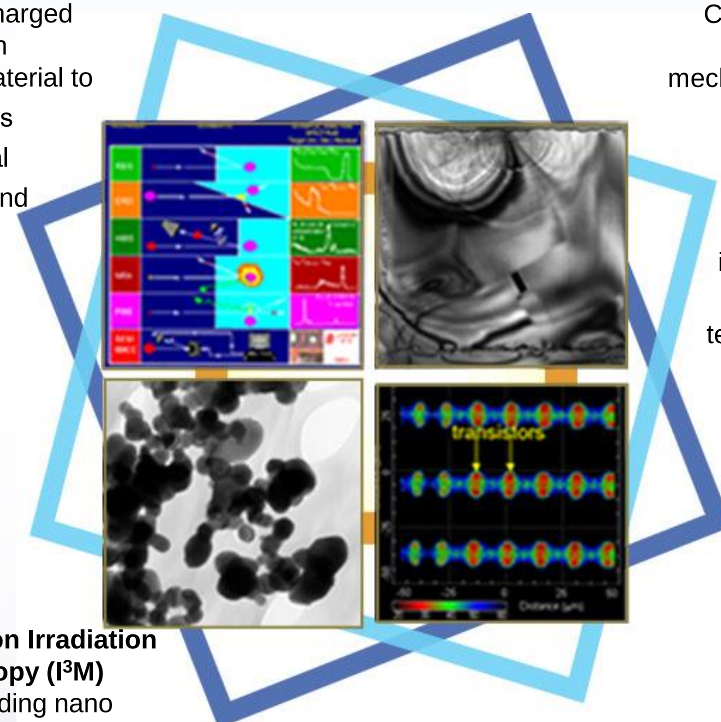


Ion Beam Analysis (IBA)

Shooting a 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 (I³M)**

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.

Potential Evolution of System Design

Use the Nearest Stone

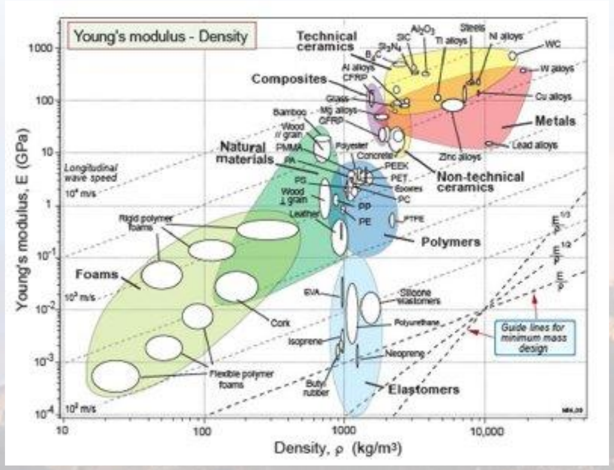
to

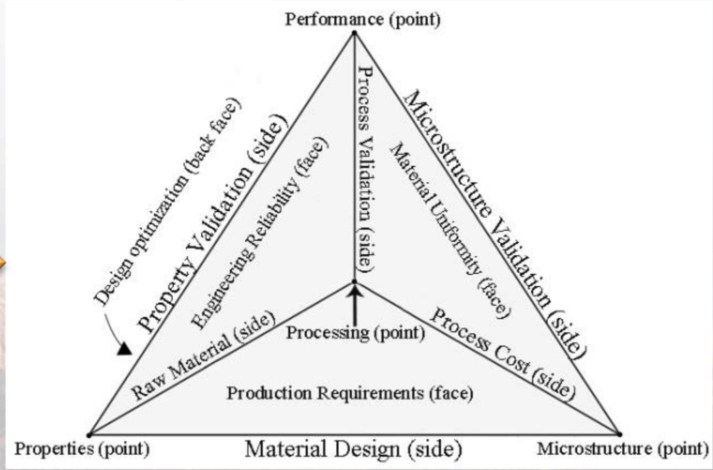
- Radar charts and Ashby plots of current material
- Accelerated and field testing
- **Scientist create a new materials. Engineers find an application**

Materials by Design

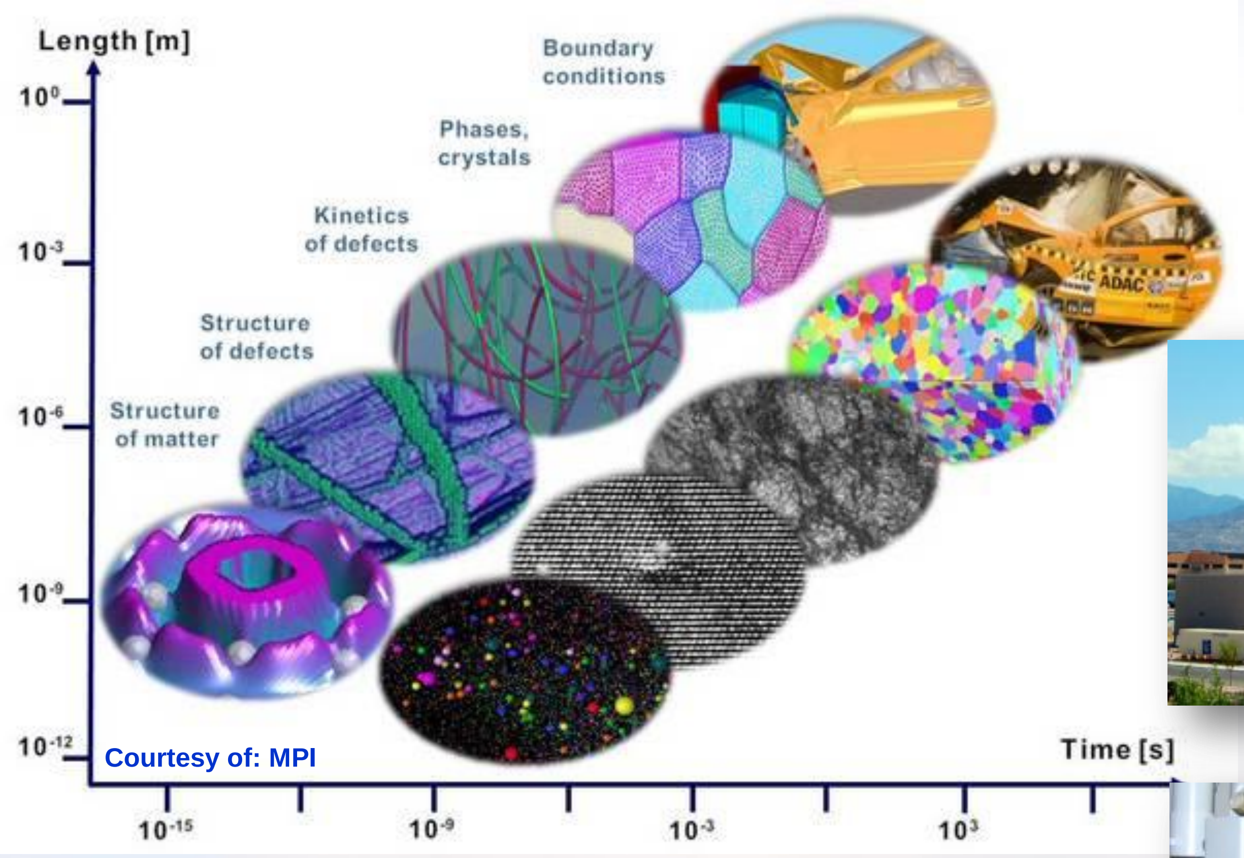
- **Physics-based approach**
- **Requires multiscale modeling**
- **Engineers require given properties, Scientists tailor the chemistry and microstructure to achieve it.**

Great vision! We are making strides, but we are not there yet



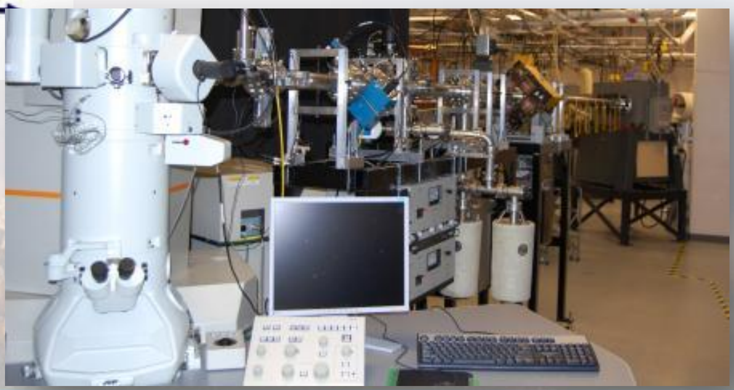


Investigating the nm Scale to Understand the km Scale to Understand Materials Response in the Extremes



Ion Beam Lab (IBL)

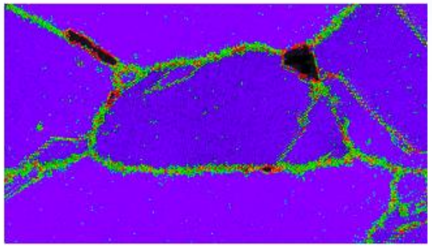
In situ Ion Irradiation TEM (I^3TEM)

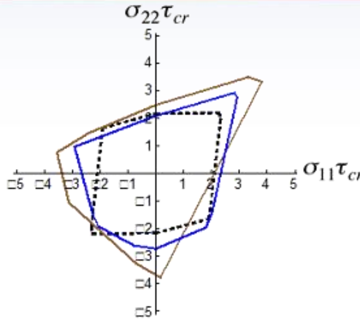


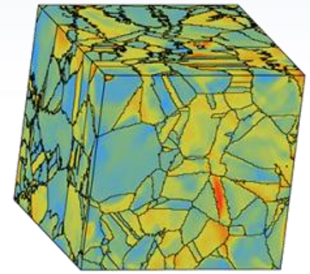
To develop predictive physics-based models, a fundamental understanding of the structure of mater, defects, an the kinetics of structural evolution in the environments of interest are needed

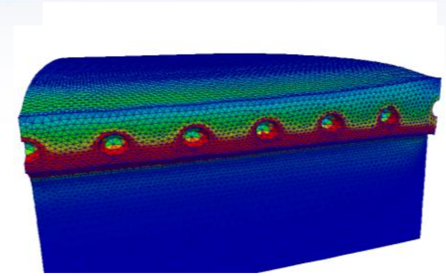
Approach: Multiscale simulation & experiments are needed to understand and predict the sources of material variability

simulations

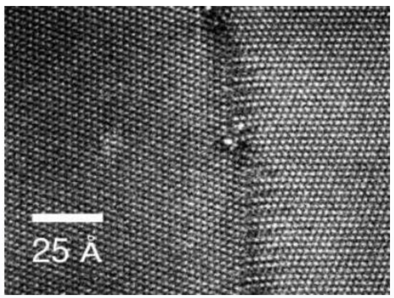


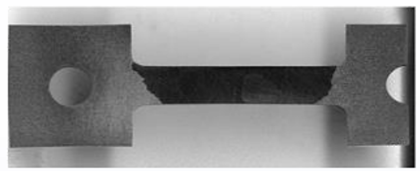


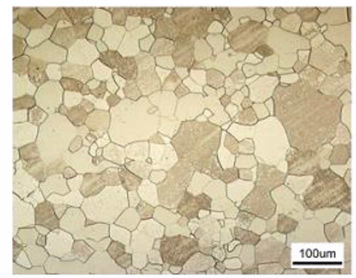


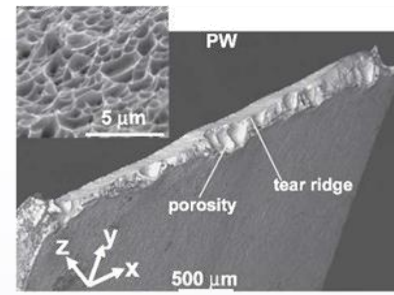


experiments









Atomic scale phenomena
10^{-9} m 10^{-9} s

Single crystal behavior
10^{-6} m 10^{0} s

Microstructural effects
10^{-3} m 10^{3} s

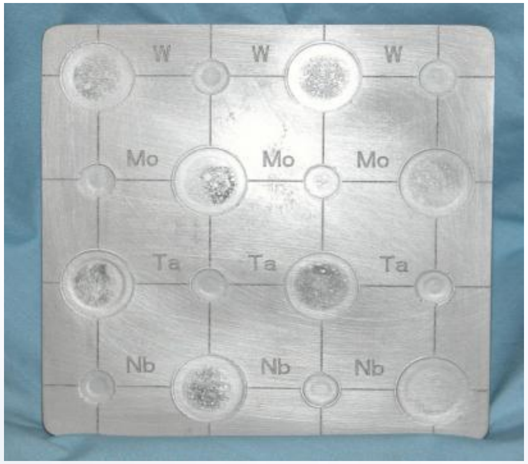
Material performance
10^{0} m 10^{6} s

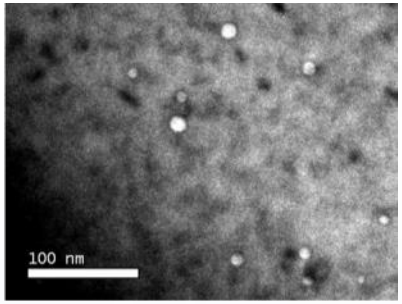
Atoms-up: Develop physics-based models to provide scientific insight

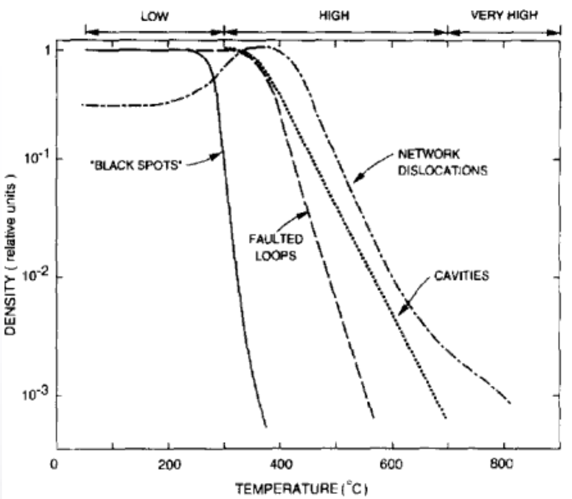
Continuum-down: Augment engineering-scale models to provide improved fidelity

Sandia's Approach to Rapid Material Validation for Advanced Materials Necessary for New Reactors

- Advanced materials are needed
- Several theories exist for the desired microstructure
- New materials have been made
- Current neutron fluxes require decades for testing





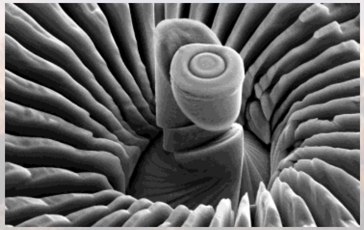


Microstructural Characterization (XTEM)

Mechanical Properties (small-scale testing)

Local Composition (Material Design) + Local Microstructural Control (Ion Irradiation)

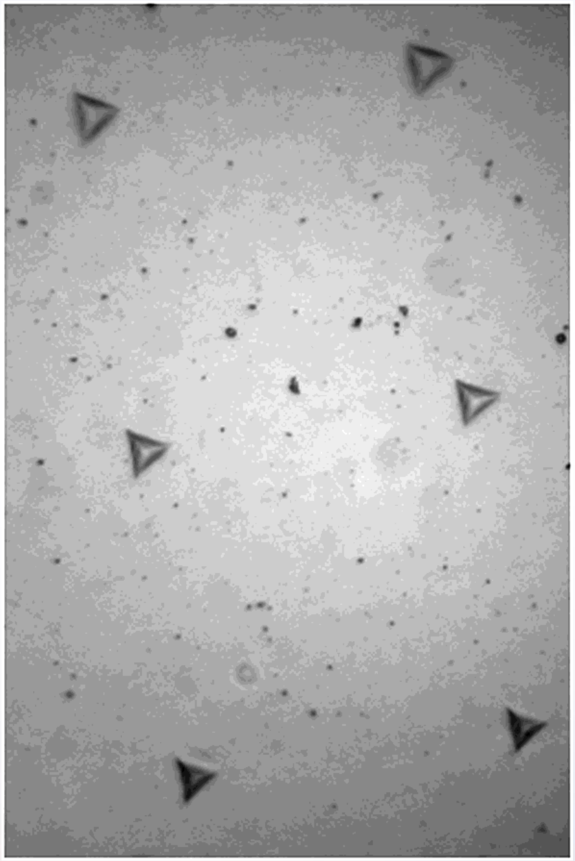
Validating Comparison to Neutron Irradiation Experiments + Investigation into new materials



Testing of Irradiated Stainless Steels

Collaborators: L.N. Brewer, T.E. Buchheit and A.J. Kilgo

- Micropillar is difficult for many polycrystalline materials
  - Due to the dependence of FIB milling rate on orientation



To validate the approach:

1. Metals previously tested by Neutron Irradiation must be tested
2. The effect of temperature and various ion characteristics must be considered

Thus, we irradiated

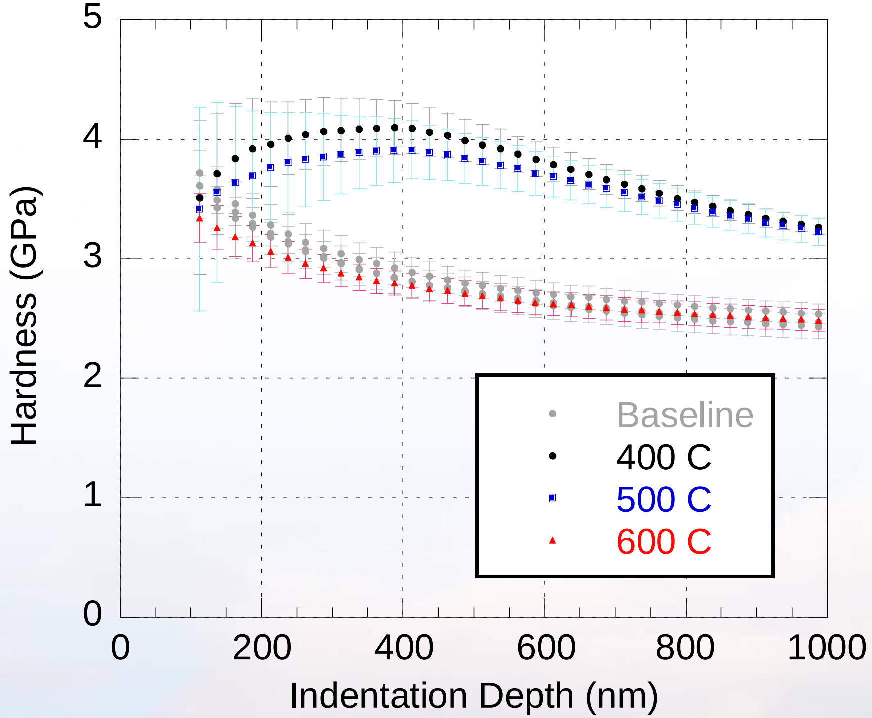
- 420, 409, and 316L SS
- Approximately 10 dpa, 40 dpa, and 100 dpa
- Temperatures of 400 ºC, 500 ºC, and 600 ºC

Three steel compositions were irradiated under various conditions. Nanoindentation was selected as the optimal small scale testing method.

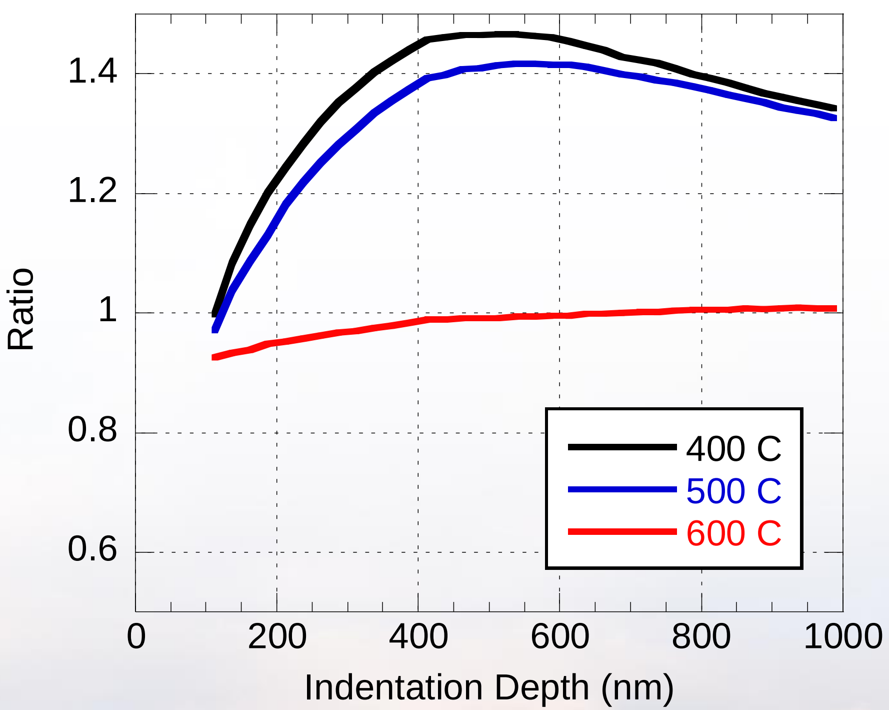
Berkovich Indentation of 100 dpa Irradiated Samples

Collaborators: L.N. Brewer, T.E. Buchheit and A.J. Kilgo

Hardness vs. Indentation Depth
Comparison of 100 dpa measurements



Baseline to Implanted Region Hardness Ratio vs. Indentation Depth - 100 dpa experiments



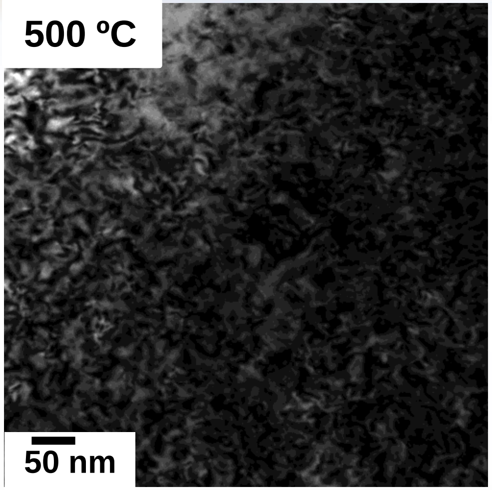
At 100 dpa, the hardness difference between 400 ºC and 500 ºC sample and the control microstructure has increased.

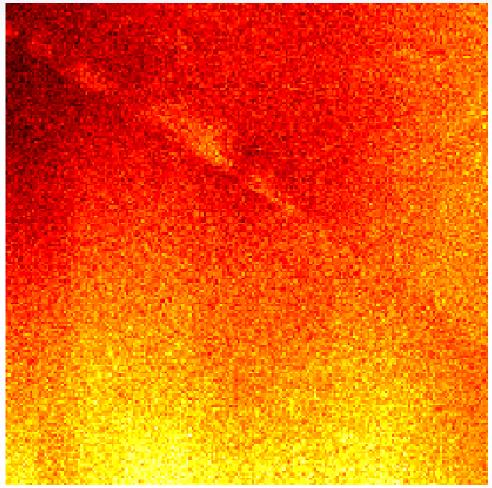
Sandia National Laboratories

Microstructural Evolution between 500 ºC and 600 ºC

316L Stainless Steel: 100dpa, 20 MeV Nickel Ions

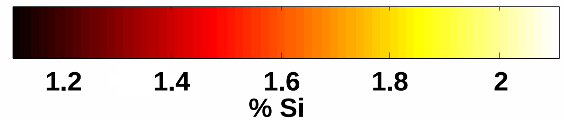
Collaborators: L.N. Brewer, A.J. Kilgo, P. Kotula

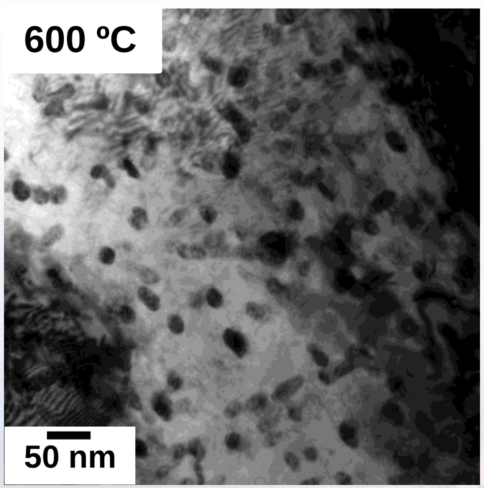


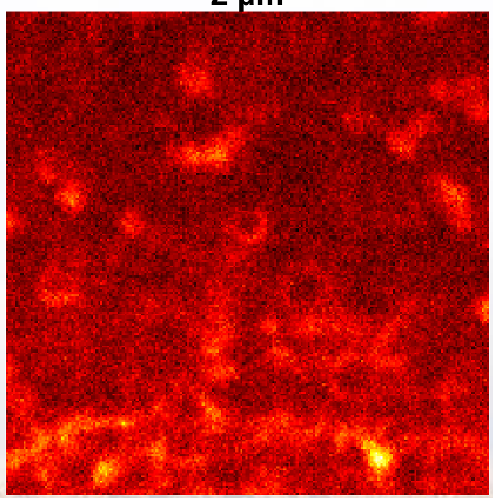


2 µm

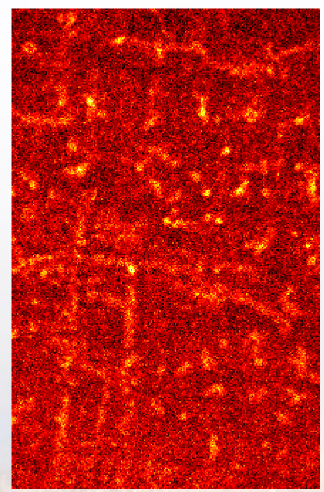
- Large number of small defects present in the irradiated region
- No significant segregation of either the Ni or Si constituents





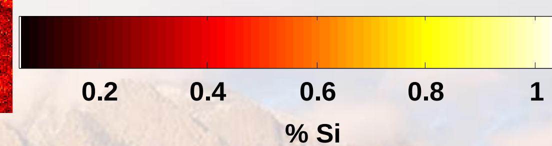


500 nm



1µm x 2µm

- Voids are formed and are self-ordered
- Significant segregation of either the Ni or Si constituents



Ni and Si rich regions appear to self-organize and sometimes surround voids at 600 °C, but not 500 °C

Sandia National Laboratories

Micropillar Compression Experiments

Collaborators: M.J. Rye, L.N. Brewer, B. Boyce

Sample Preparation:

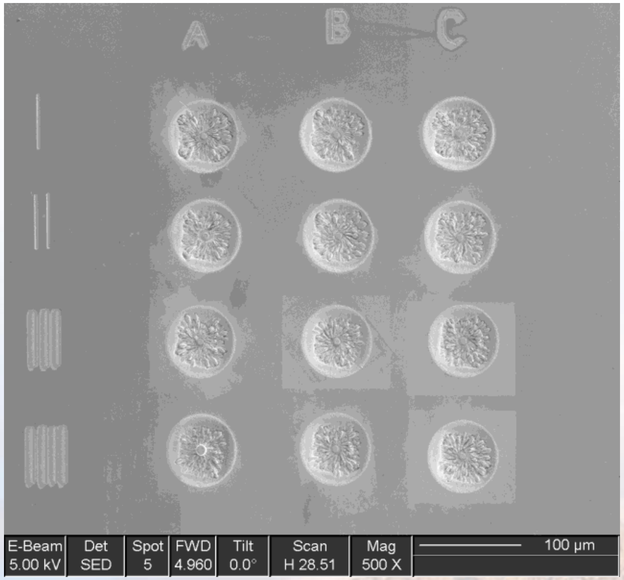
- Copper single crystals (FCC)
- Different crystallographic orientations: (100), (110), and (111)
- Self-ion Implants at 30 MeV to 0 (control), 50 dpa, and 100 dpa.

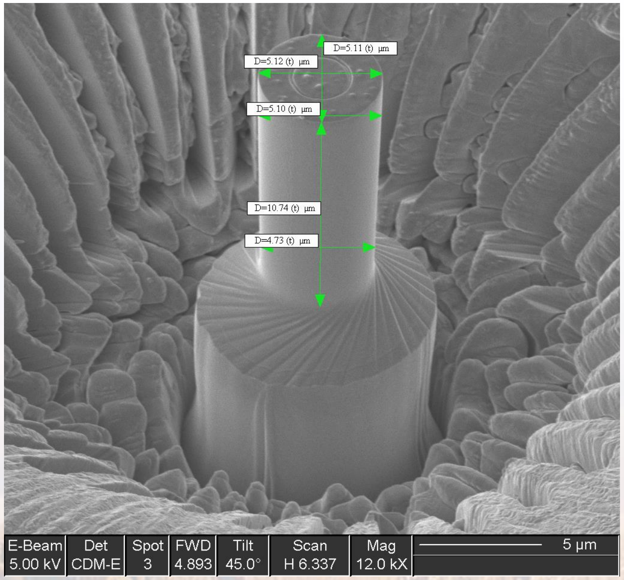
Pillar Manufacturing:

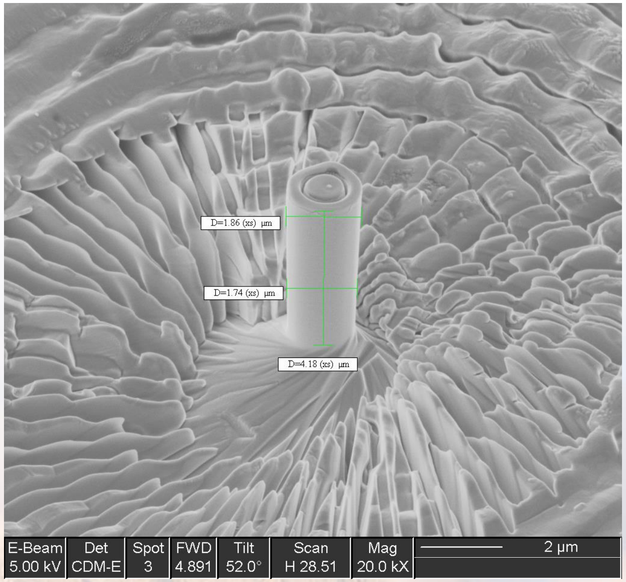
- We employed Uchic's FIB lathe machining process for straight-walled cylinders.
- Array of at least 9 nominally identical pillars tested per condition to assess statistical variability.
- Height varies from 4 µm to 10 µm

Compression Testing:

- Hysitron Performech Nanoindenter permits <1 nm and <1 µN resolution.
- 25 µm flat ended cone indenter in feedback displacement control, rather than typical force control.
- Pillars compressed 10% strain at a strain rate of $0.025\ s^{-1}$.

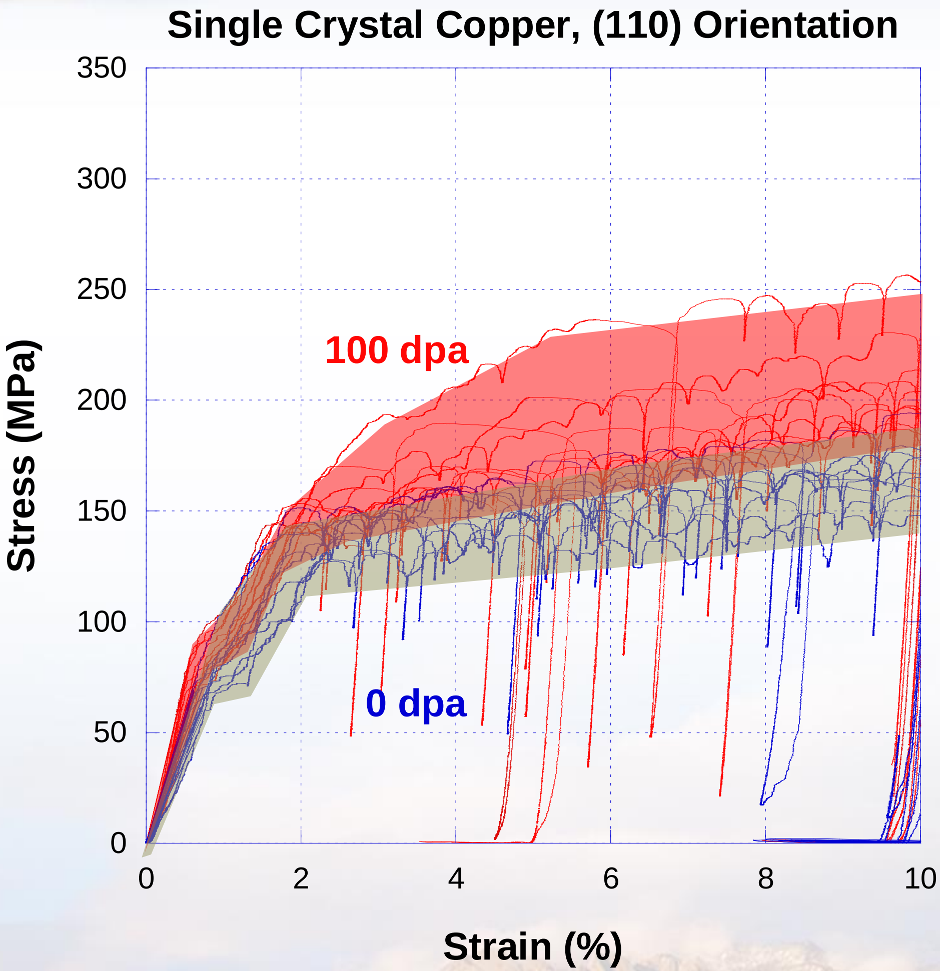


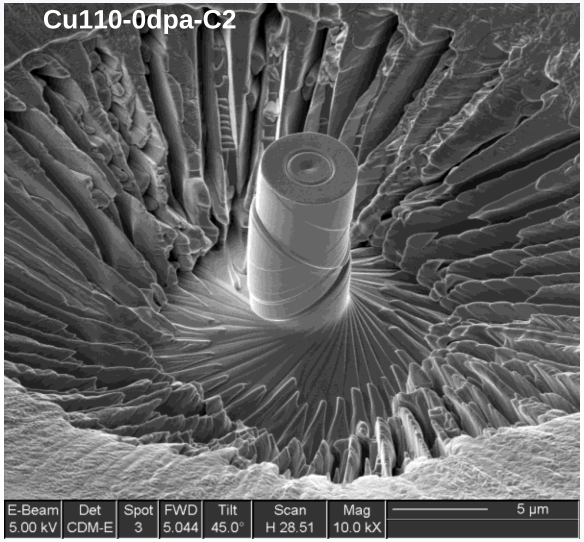


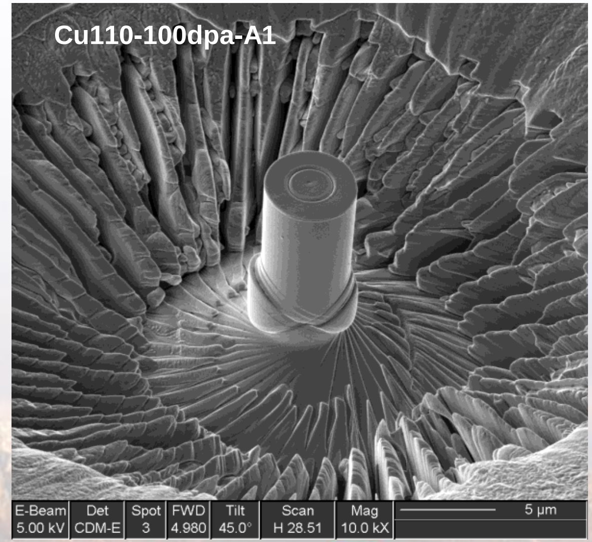


Large Micropillar Compression

Collaborators: M.J. Rye, L.N. Brewer, B. Boyce





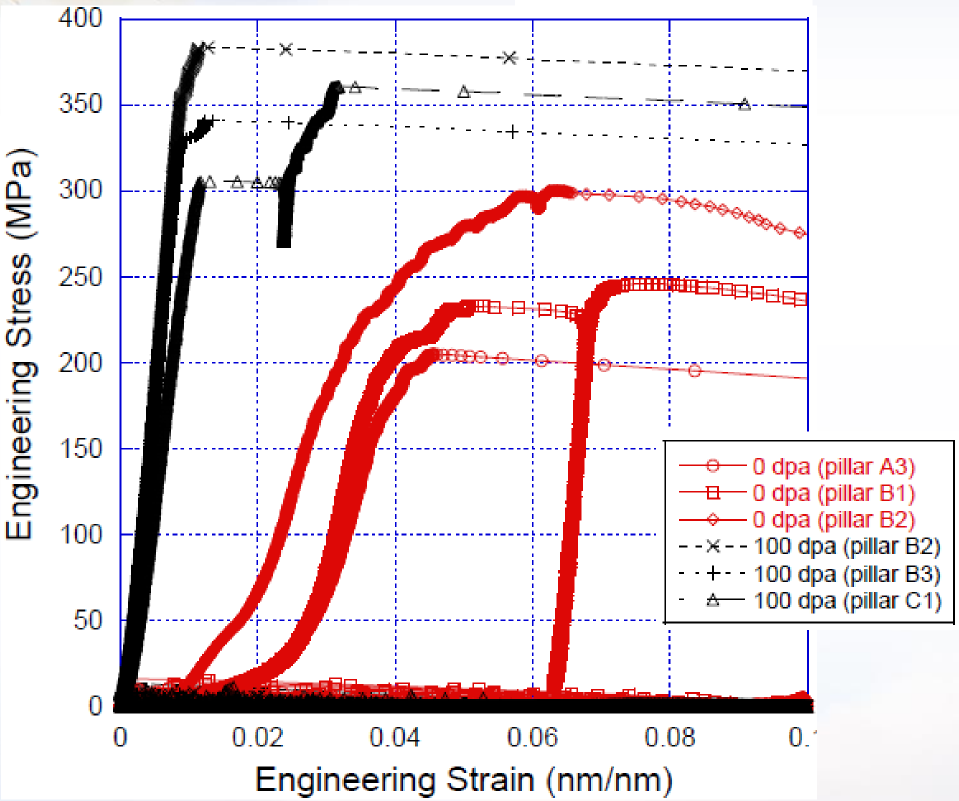


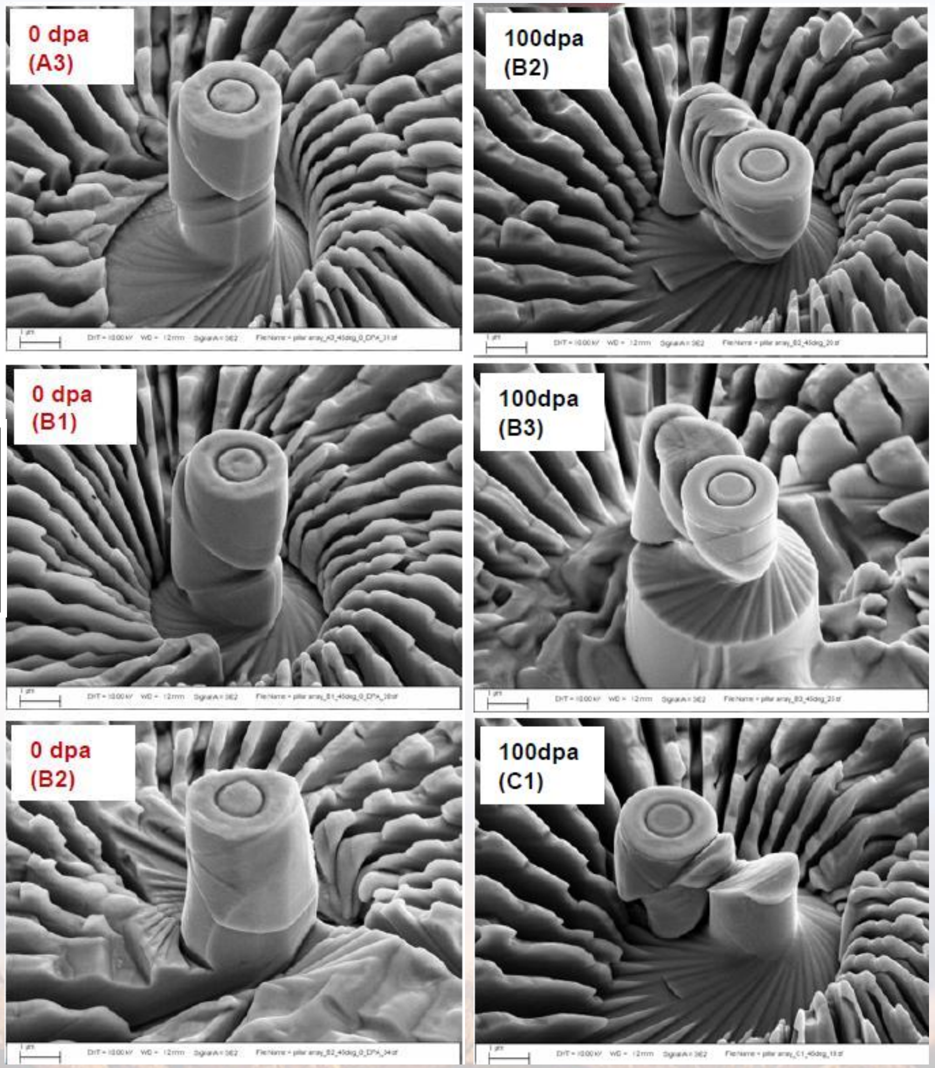
Minimal difference between the control and irradiated 10 µm-tall pillars. Slip occurred in the bottom fraction of the pillars.

Sandia National Laboratories

Intermediate Micropillar Compression

Collaborators: M.J. Rye, L.N. Brewer, B. Boyce

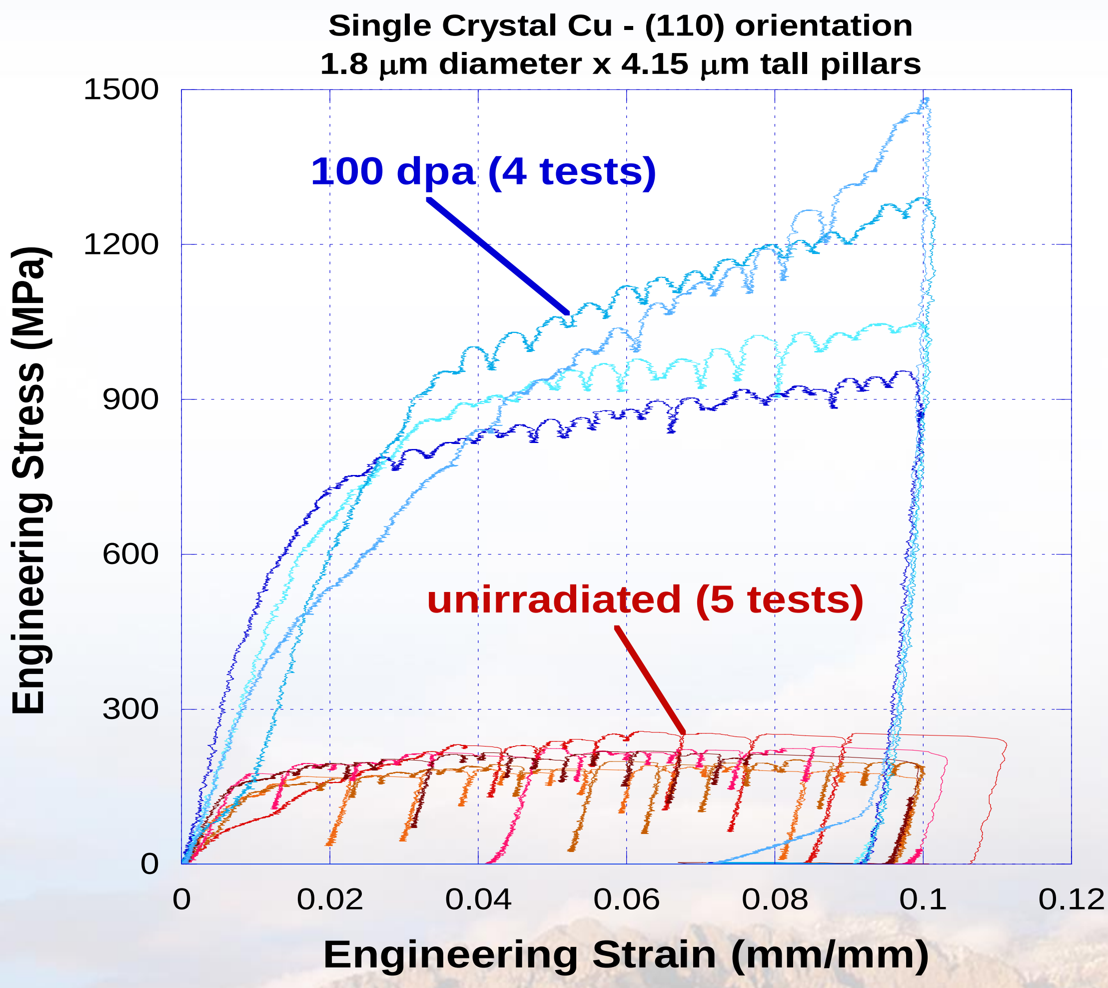


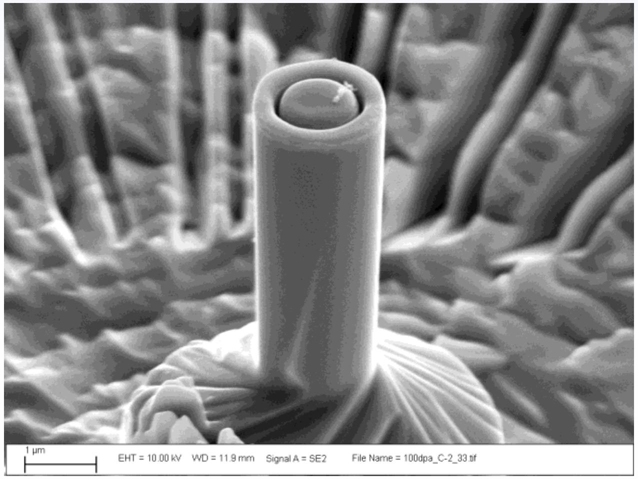


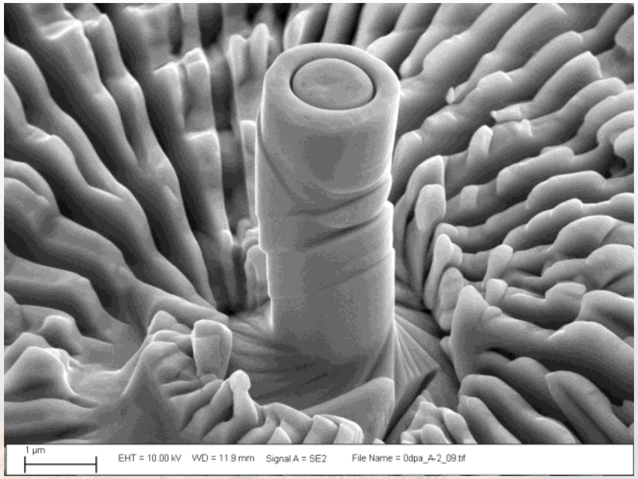
5 µm-tall pillars show greater distinction with catastrophic failure

Small Micropillar Compression

Collaborators: M.J. Rye, L.N. Brewer, B. Boyce







Initial tests indicate that the 4 μm-tall pillars are 5 times stronger and show no signs of slip band formation

Sandia National Laboratories

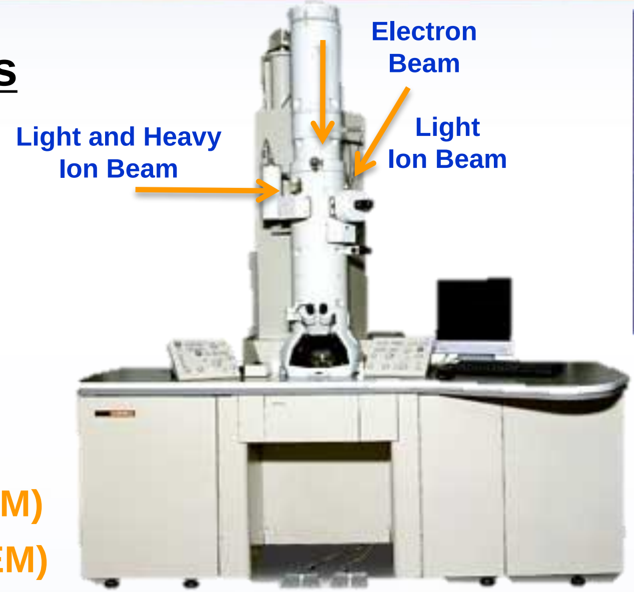
In situ Ion Irradiation TEM Facility

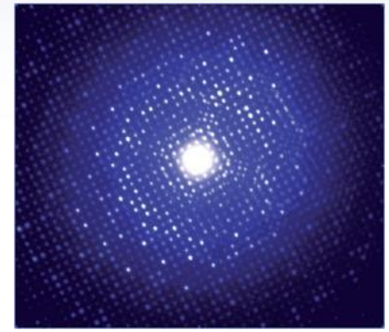
Proposed Capabilities

- 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

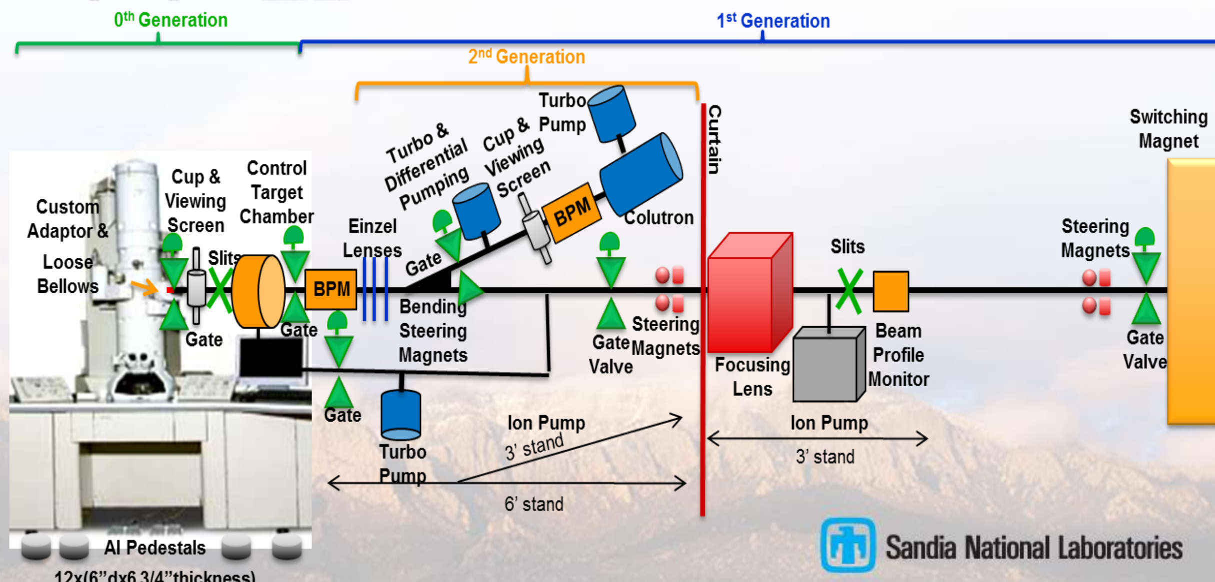
- *In situ* vapor phase stage
- *In situ* liquid mixing stage
- *In situ* heating
- Tomography stage (2x)
- *In situ* cooling stage
- *In situ* electrical bias stage
- *In situ* straining stage





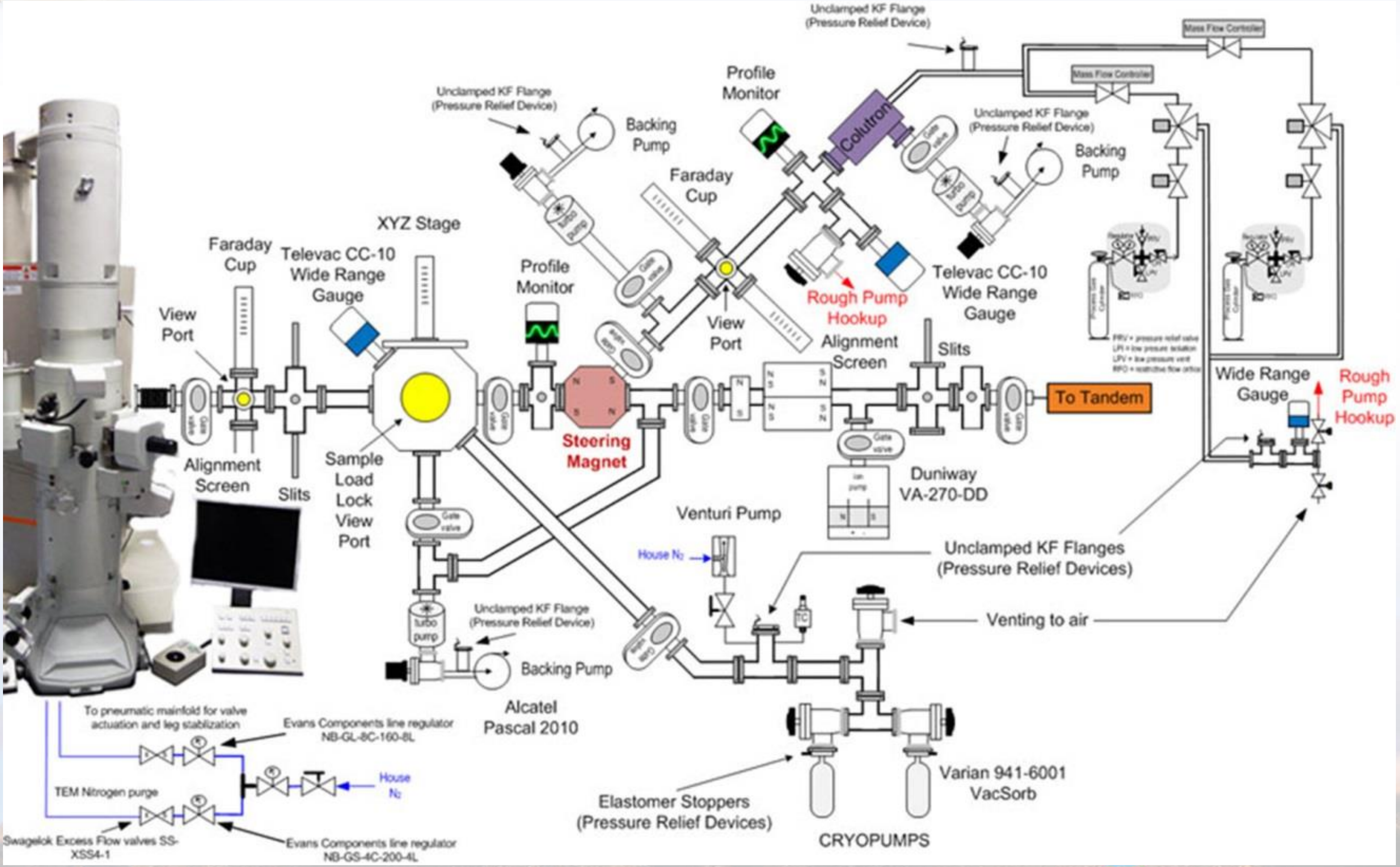
TVIPS

Hummingbird



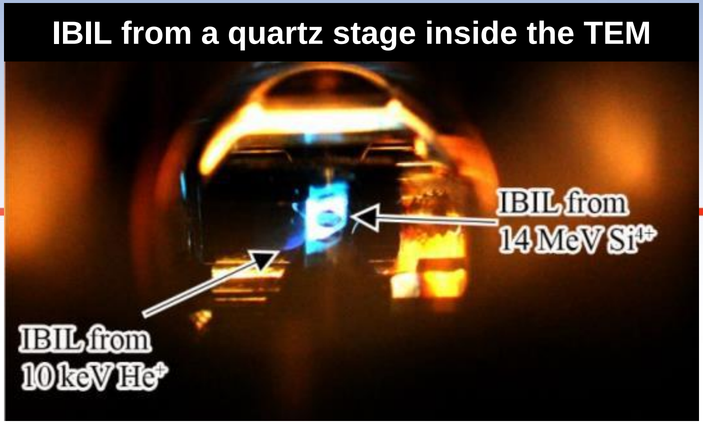
Sandia National Laboratories

Schematic of the *In situ* TEM Beamline



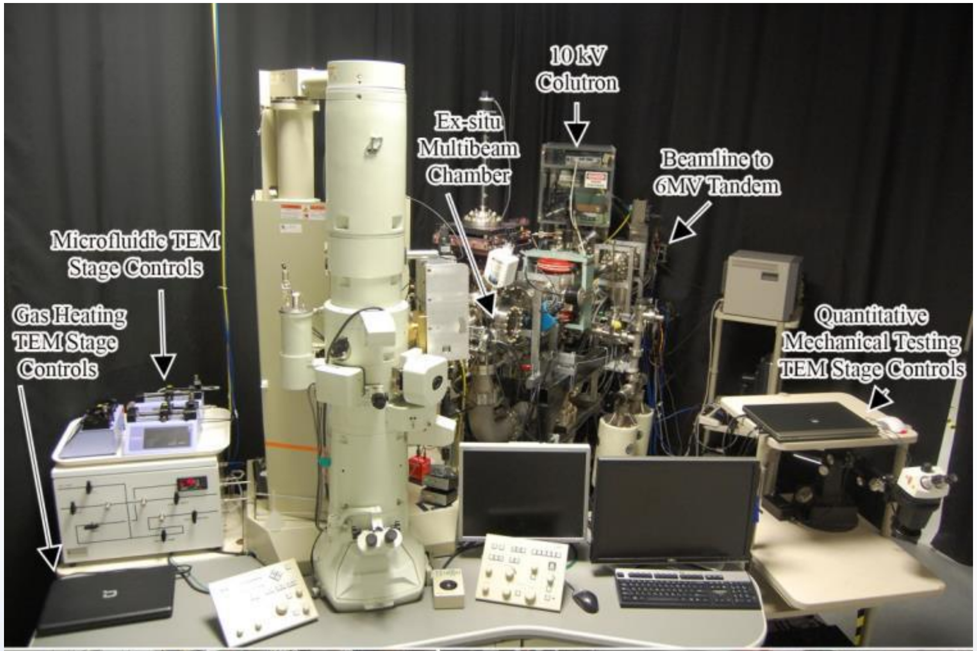
Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

Collaborator: D.L. Buller

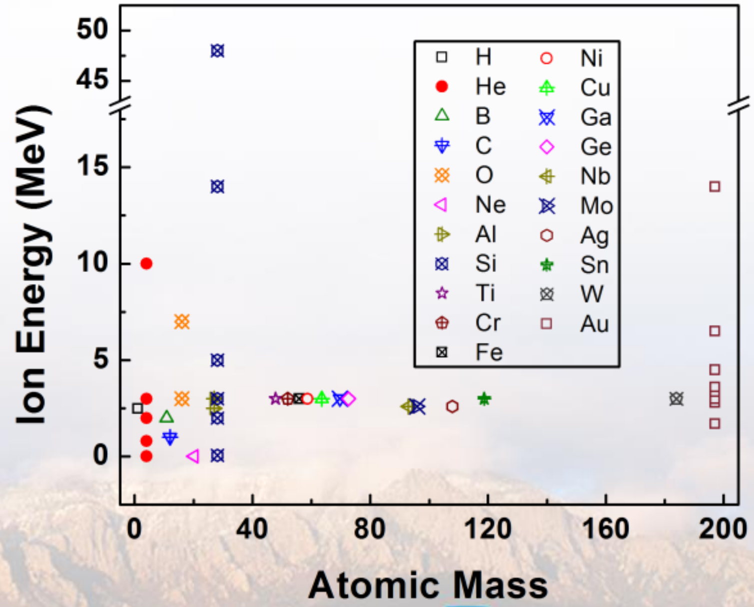


Direct real time observation of ion irradiation,
ion implantation, or both with nanometer resolution

10 kV Colutron - 200 kV TEM - 6 MV Tandem



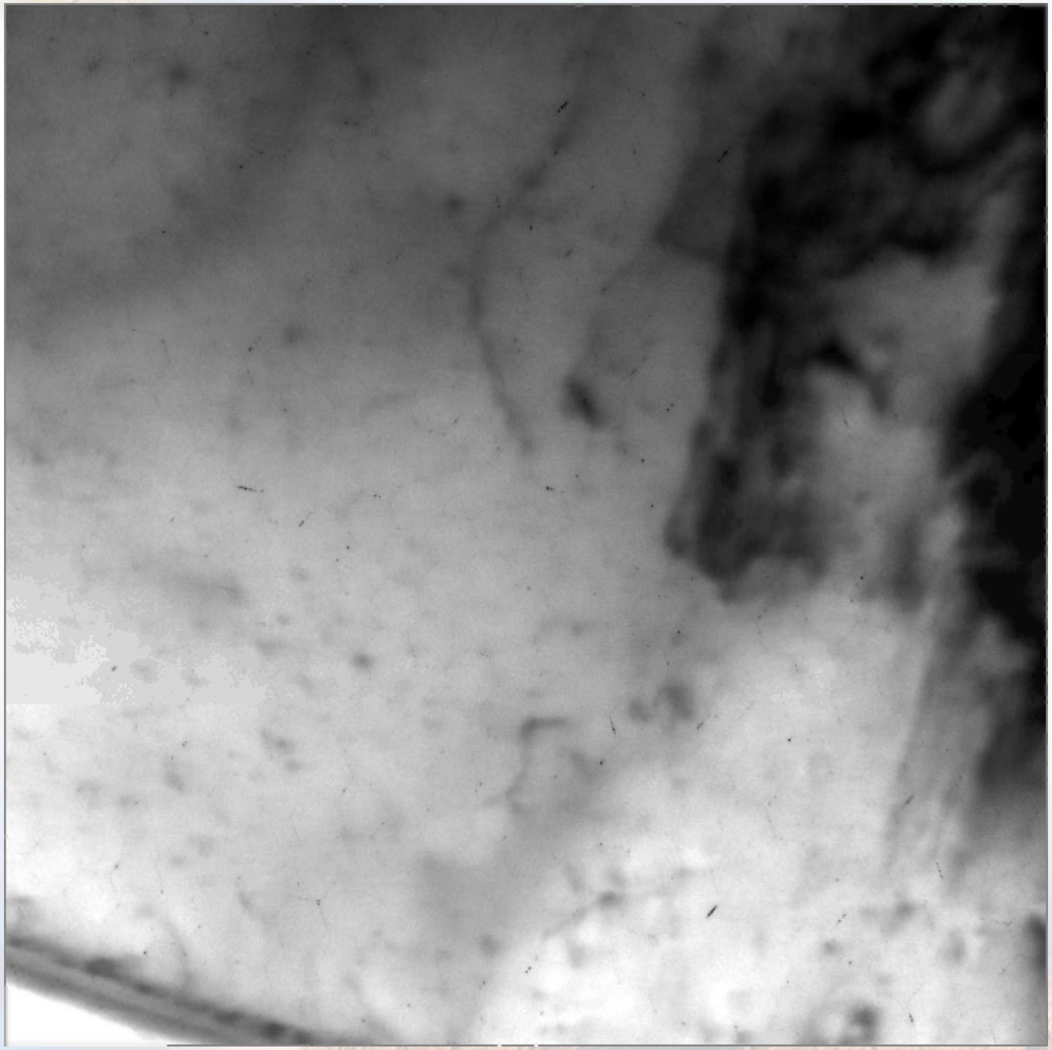
Ion species & energy introduced into the TEM

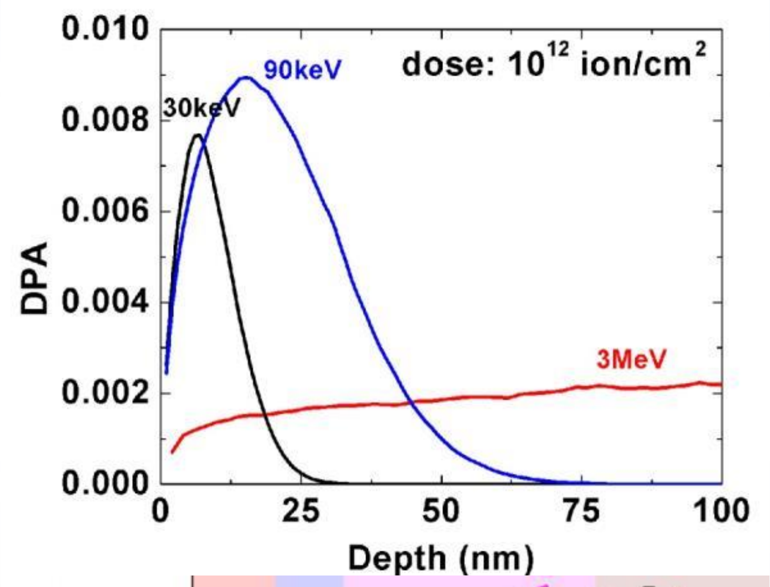


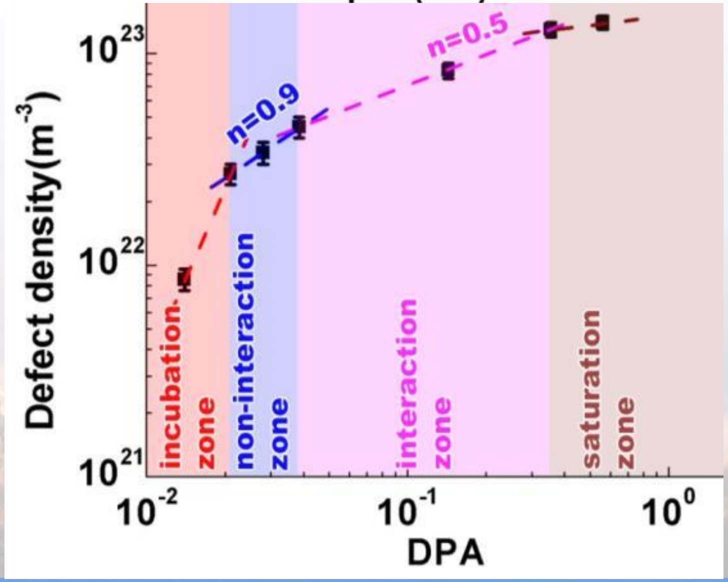
Sandia National Laboratories

Quantifying Defect Evolution in Irradiated Cu

Collaborators: N. Li & A. Misra

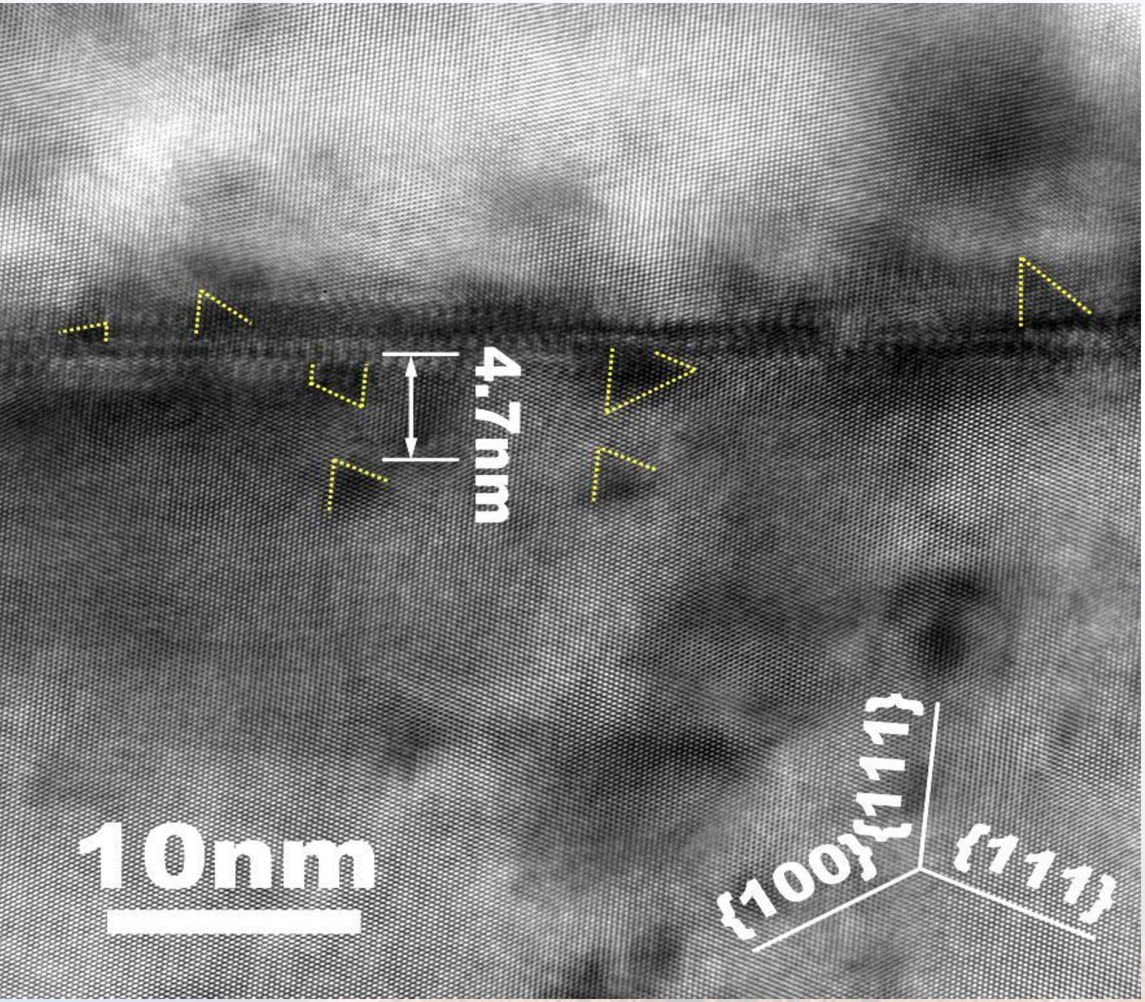


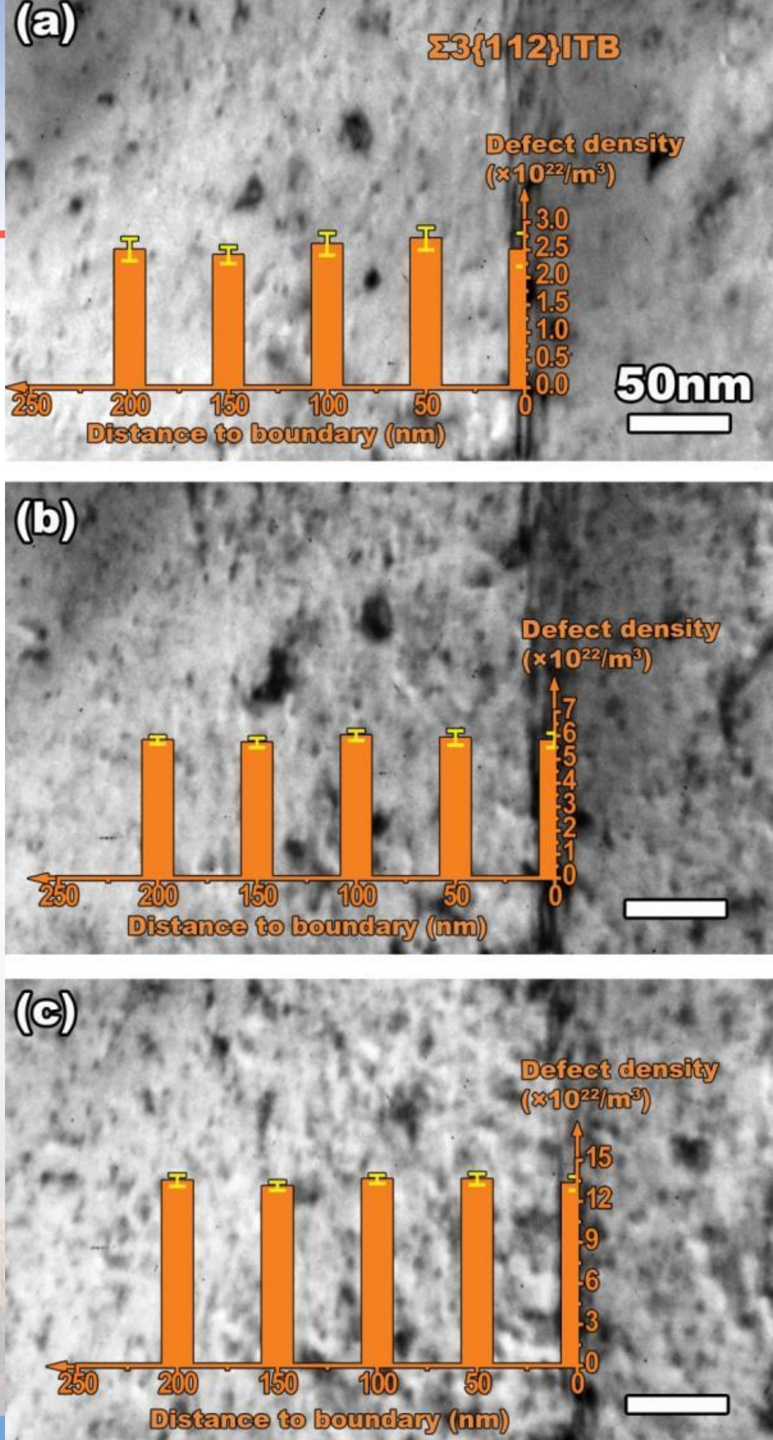




Defects are Altered Little by the Presence of Grain Boundaries

Collaborators: N. Li & A. Misra

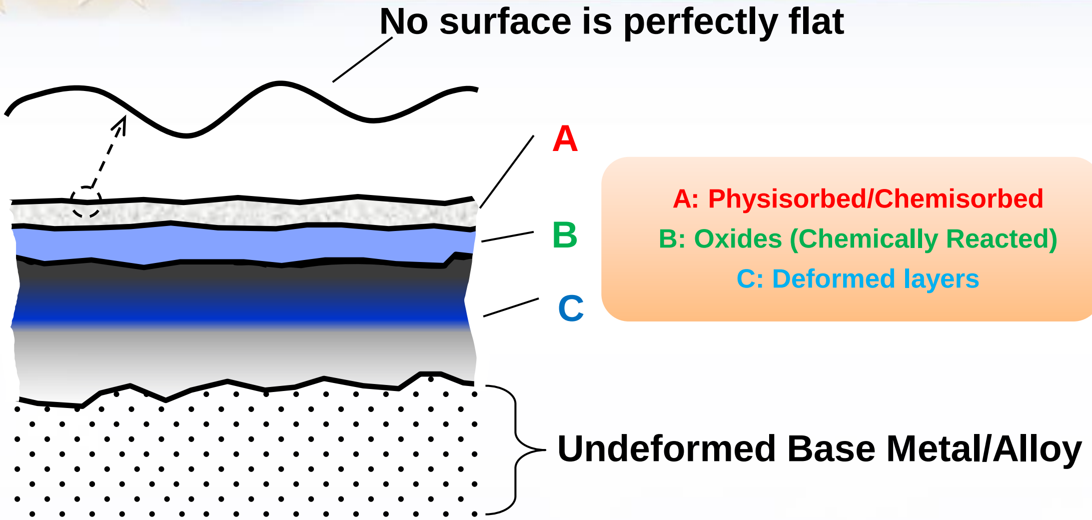




SFT appear to be directly at GB

No change in defect density is observed near GB

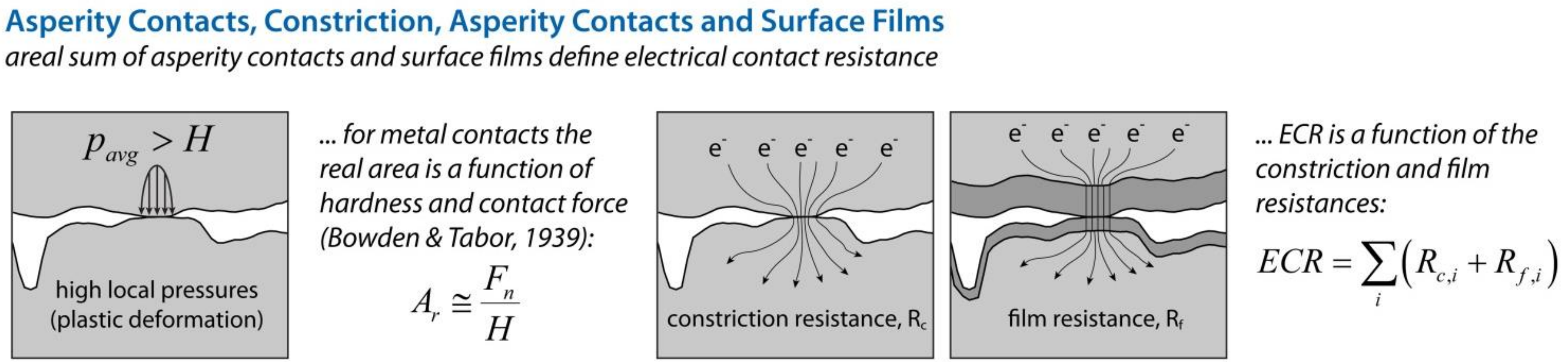
Tailoring Wear Properties in Au Sliding Contacts



Real area of contact (A_r) to be minimized for low adhesion

(Low Adhesive Wear)

Or maximized for reduced electrical contact resistance (ECR)



Archard, Journal of Applied Physics (1953) 24:981

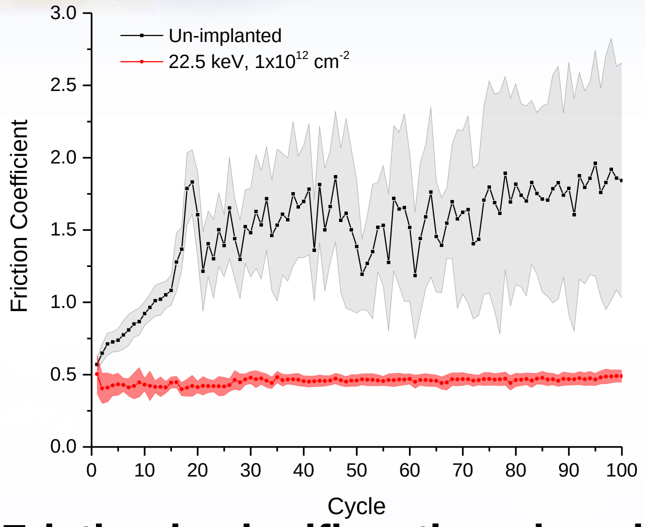
R. Holm, Electrical Contacts Handbook (1958) Berlin: Springer-Verlag

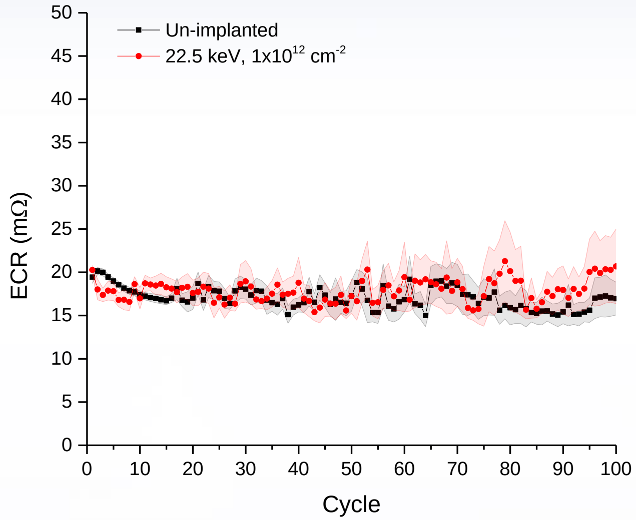
Greenwood & Williamson, Proc. Royal Society (1966) A295:300

T.W. Scharf & S.V. Prasad, Journal of Material Science (2013) 48:511-531

ECR and Wear Measurements

Collaborators: J-E Mogonye & S.V. Prasad

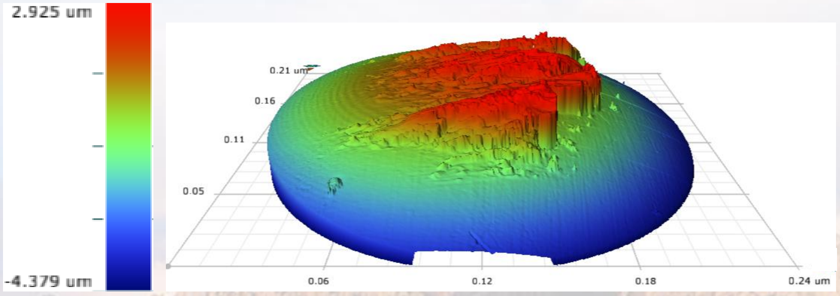




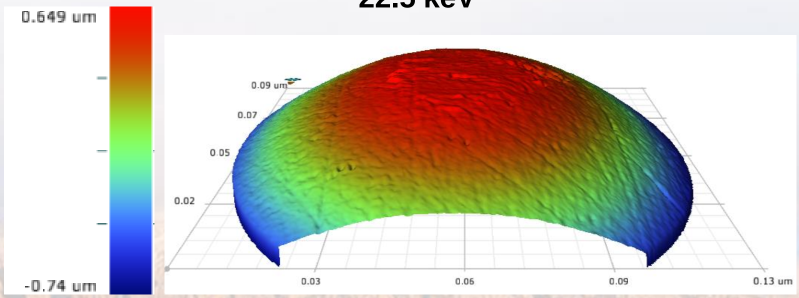
Friction is significantly reduced with ^{3}He implantation while maintaining ECR performance

Scanning white light interferometer topographical construction of riders after 100 cycles

Rider after 100 cycles against Un-implanted Au



Rider after 100 Cycles against Au implanted to 1E12 cm^{-2} @ 22.5 keV



Wear is significantly reduced with minimal effect in ECR

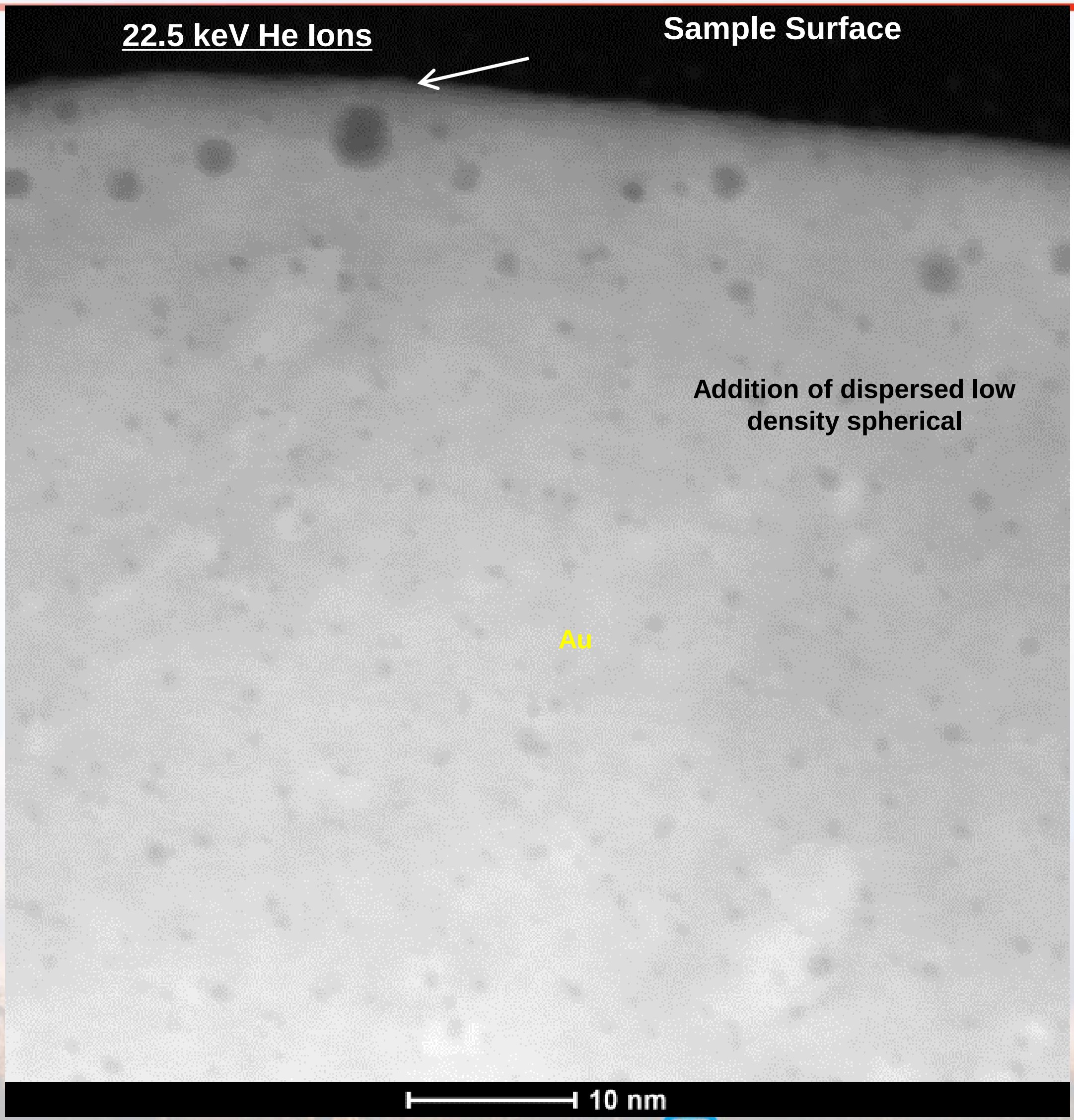
Sandia National Laboratories

Modeling and STEM of He Implantation

Collaborators: J-E Mogonye & S.V. Prasad

- Simulations: SRIM 2008
- Monte-Carlo simulation of kinematic interaction based on empirical data fitted functions
  - Input variables of target material include density, AMU, and thickness.
  - Input variables of ions include AMU, energy, and angle of incidence.
  - Assumes isotropic material, thus no consideration for channeling effects

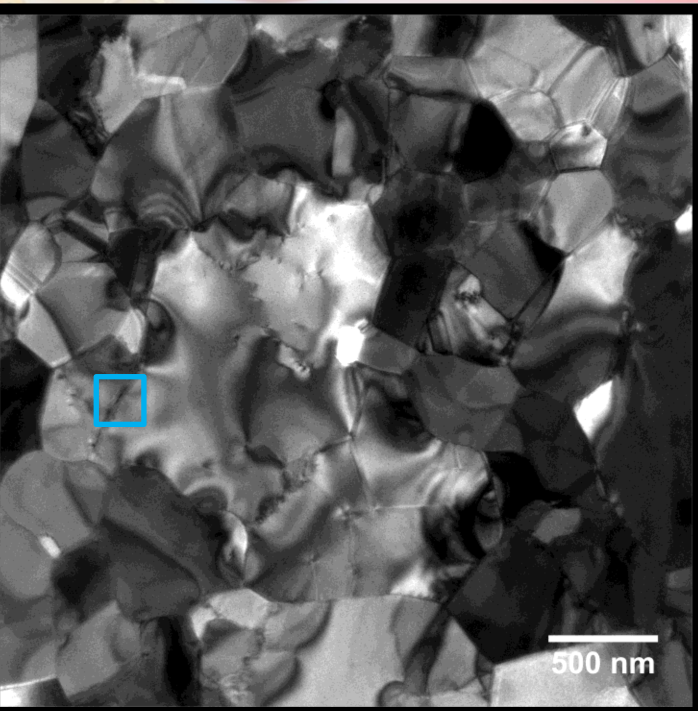
- AC-STEM used to observe the distribution of implanted bubbles
- Bubble locations are in good agreement with SRIM ion range predictions

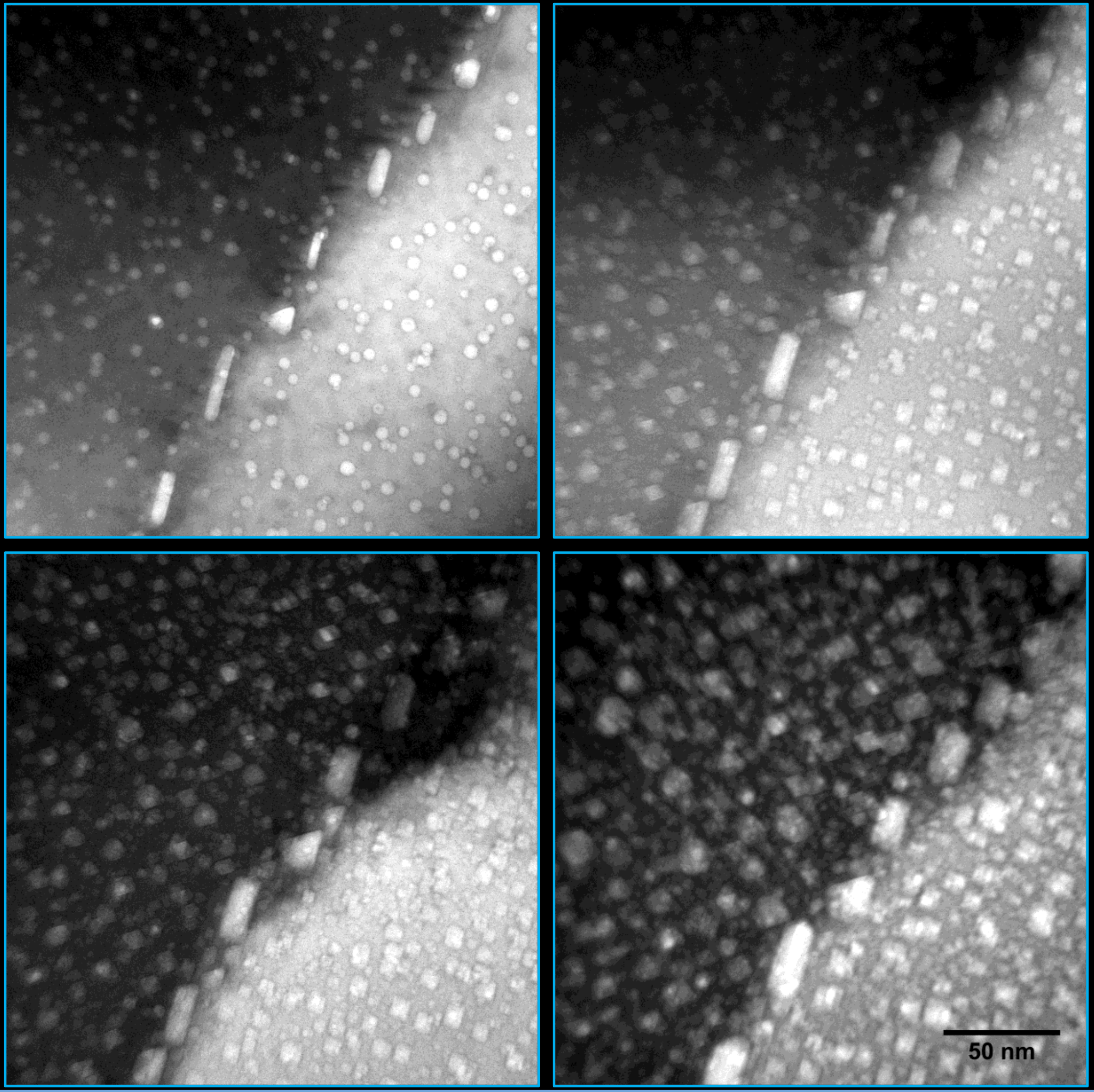


He implantation result in small dispersed spherical structures assumed to be He bubbles. Dispersion and depth can be tailored

In situ Implantation

Collaborators: C. Chisholm & A. Minor

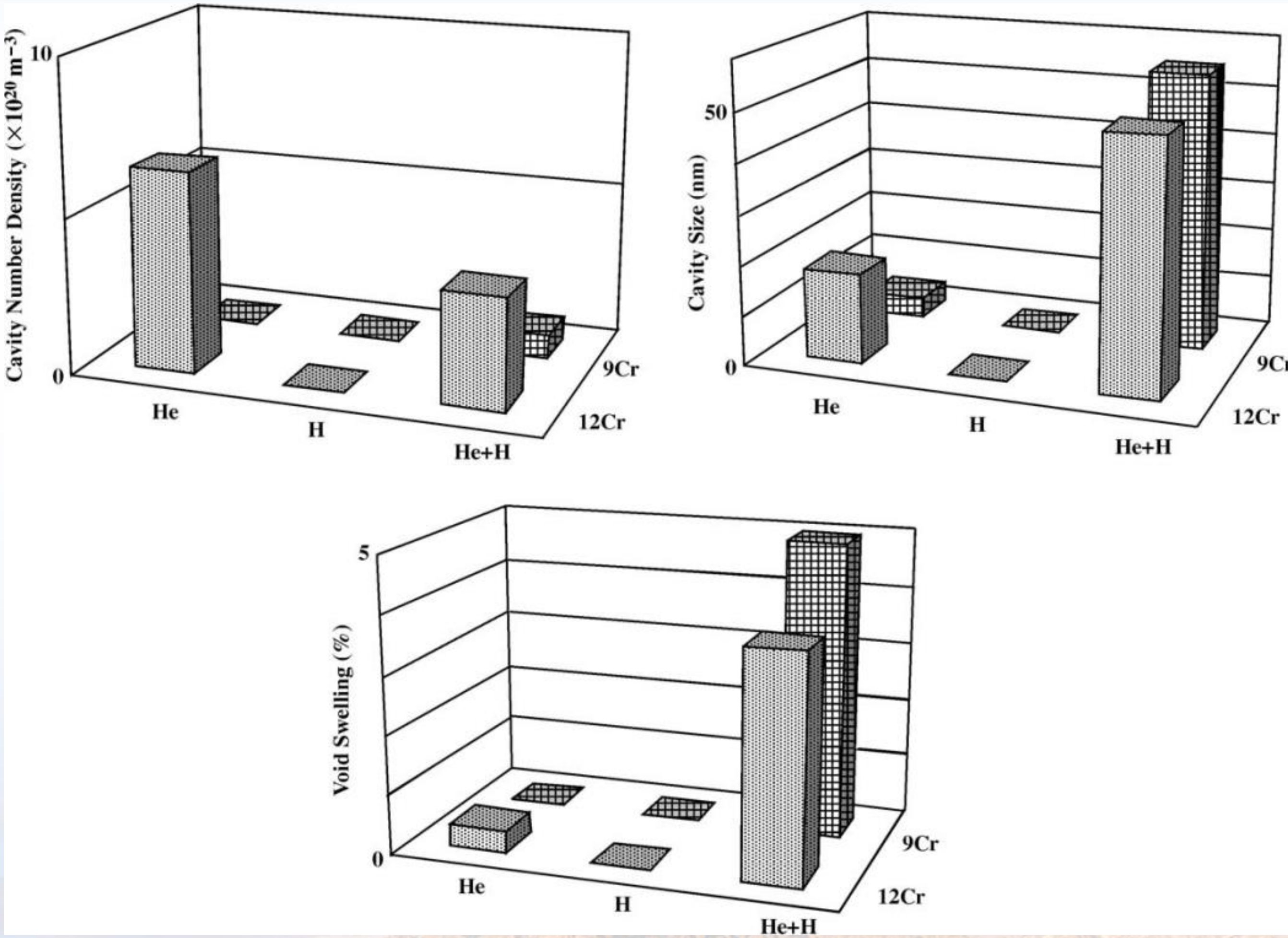




Gold thin-film implanted with 10keV He^{2+}

Result: porous microstructure

H, He, and Displacement Damage Synergy



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

Coupling Effect

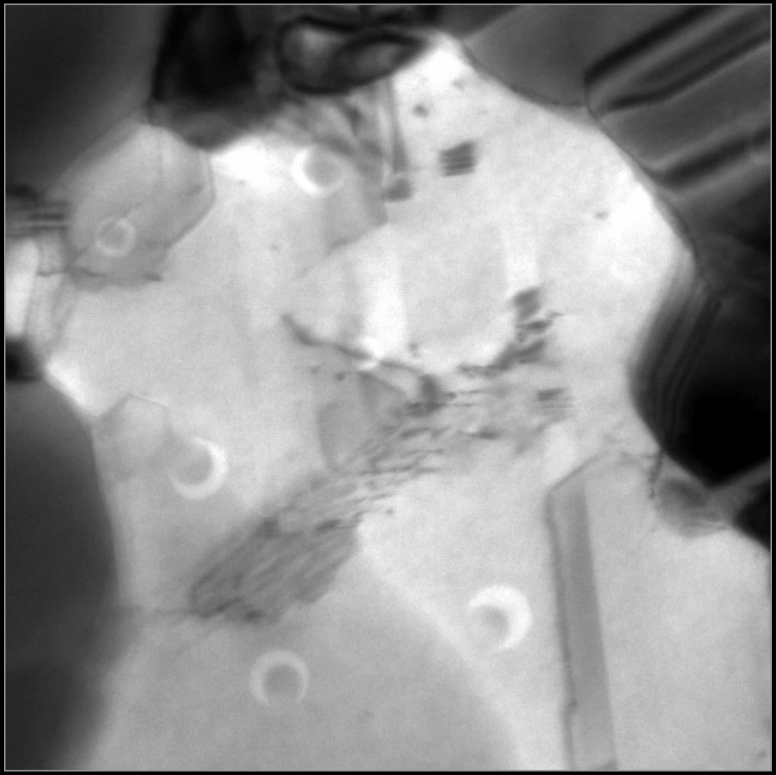
- **H and He are produced as decay products**
- **The relationship between the point defects present, the interstitial hydrogen, and the He bubbles in the system that results in the increased void swelling has only been theorized.**
- **The mechanisms which governs the increased void swelling under the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

No capability currently exist for triple beam irradiation in the U.S. and No capability for tripple beam TEM ion irradiation exists in the world

Single Ion Strikes

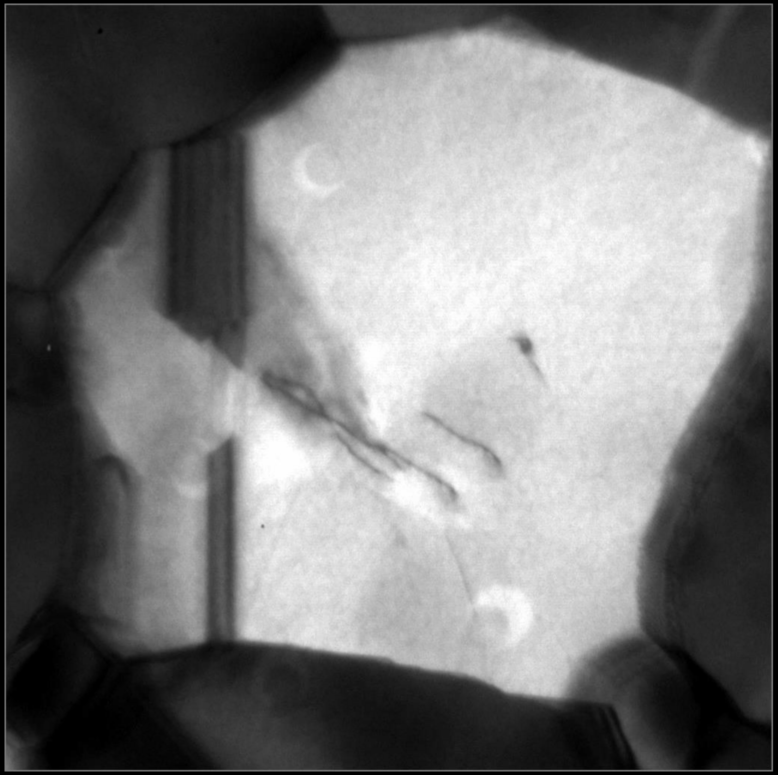
Collaborators: C. Chisholm & A. Minor

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



VS

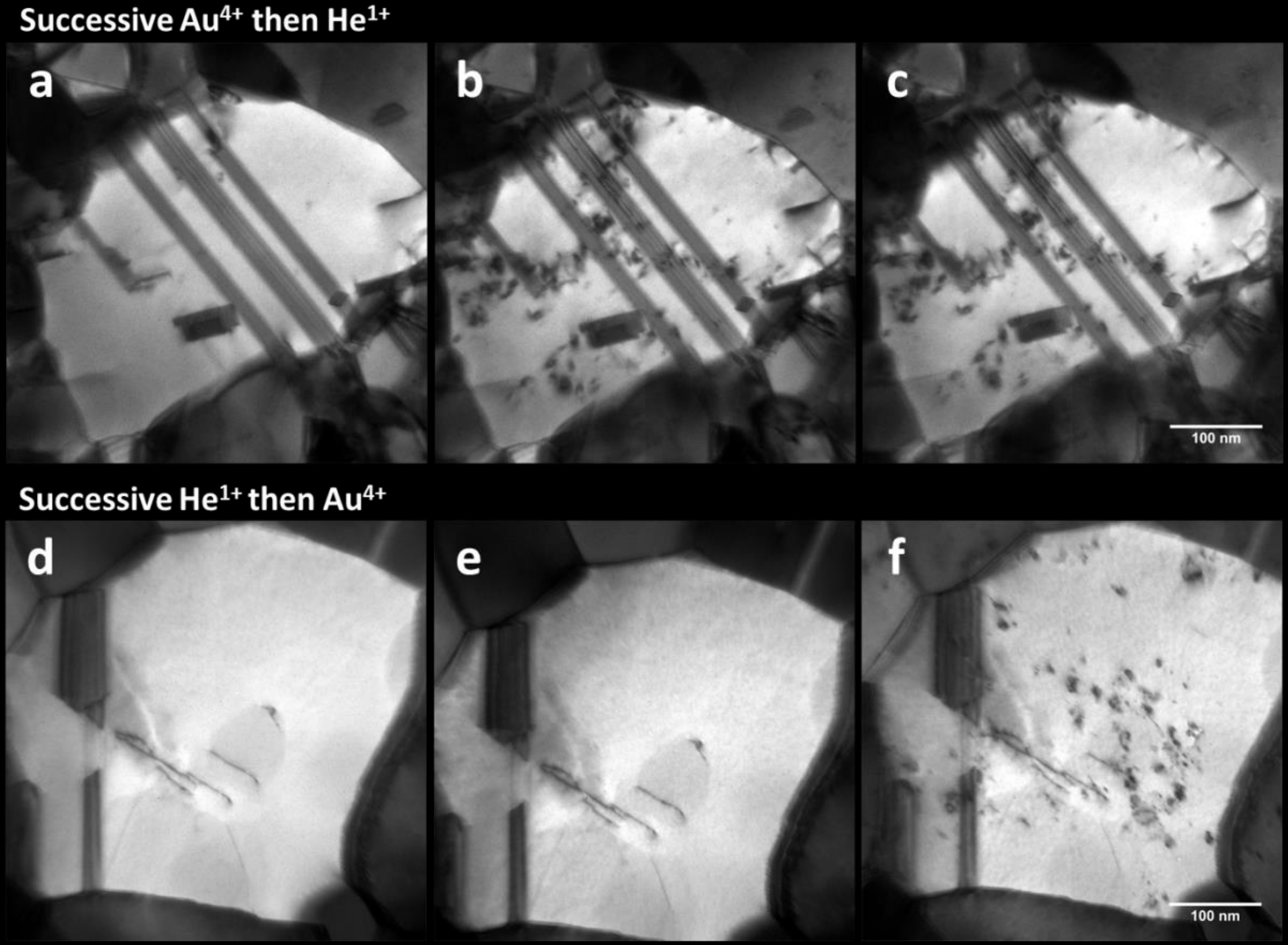
6.7 x 10^7 ions/cm^2/s



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

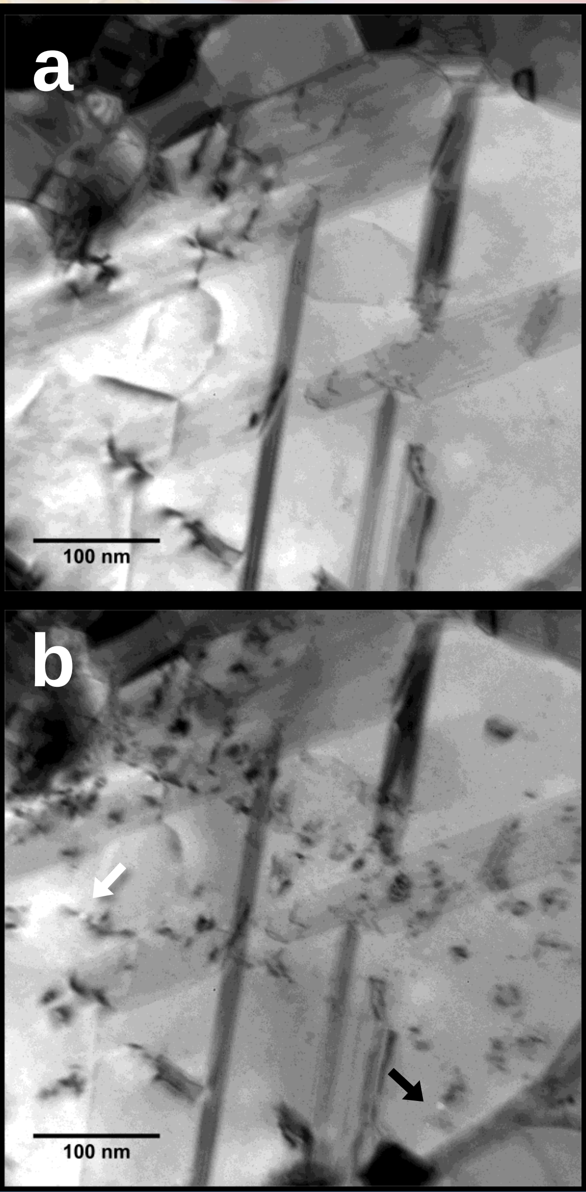
In situ Successive Implantation & Irradiation

Collaborators: C. Chisholm & A. Minor

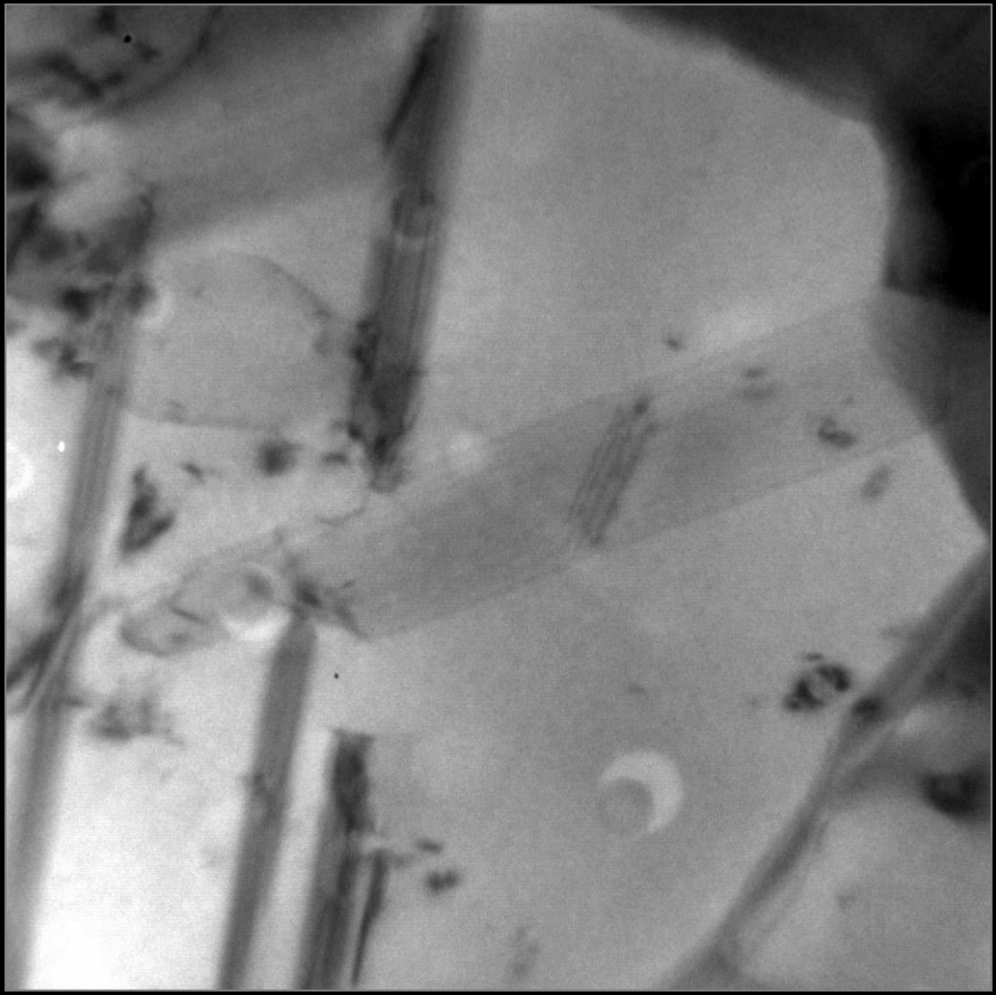


In situ Concurrent Implantation & Irradiation

Collaborators: C. Chisholm & A. Minor



He^{1+} implantation and Au^{4+} irradiation of a gold thin film



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

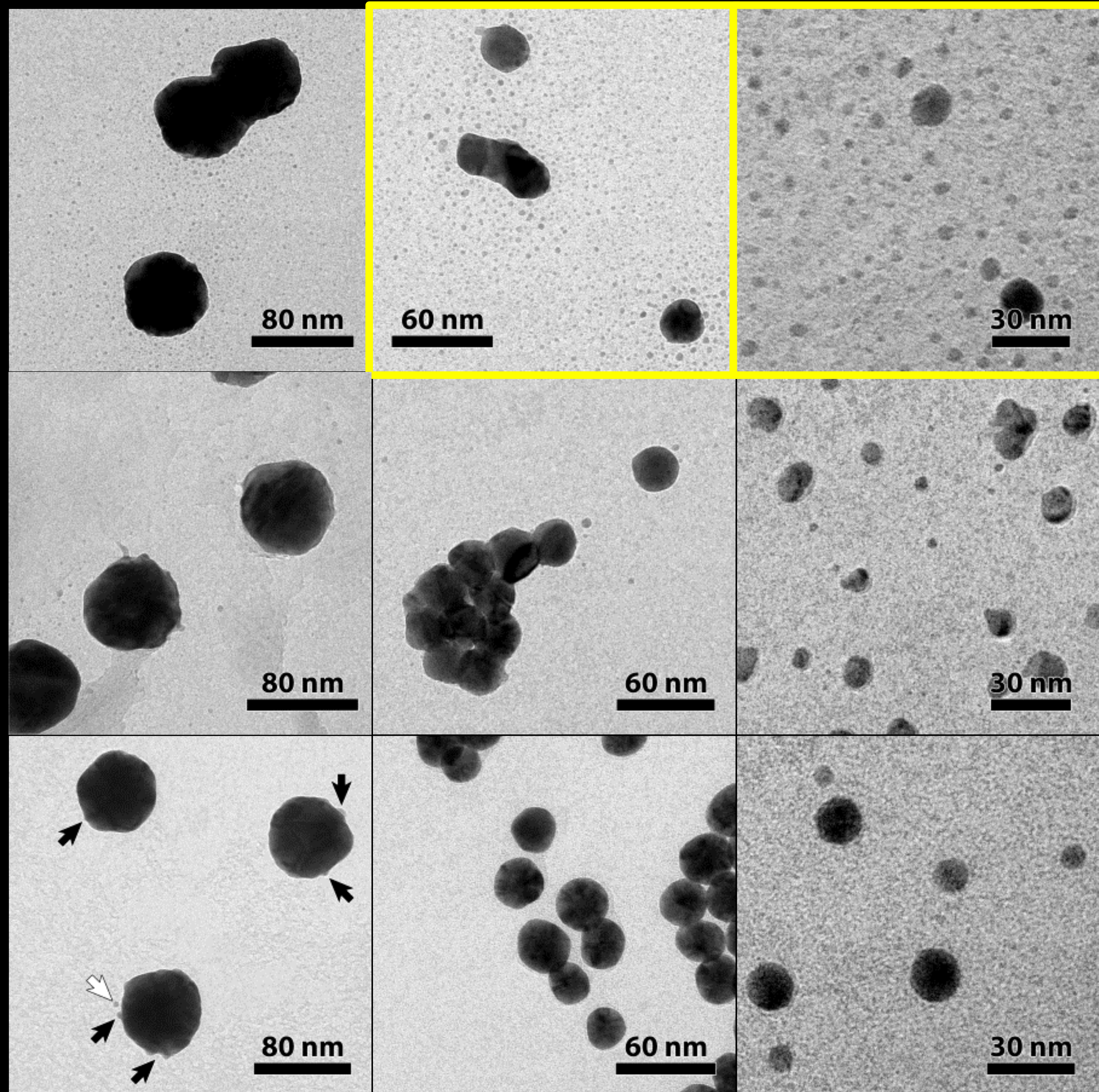
Collaborator: D.C. Bufford

60 nm | 20 nm | 5 nm

46 keV Au^{1-}
3.4×10^{14} /cm²

2.8 MeV Au^{4+}
4×10^{13} /cm²

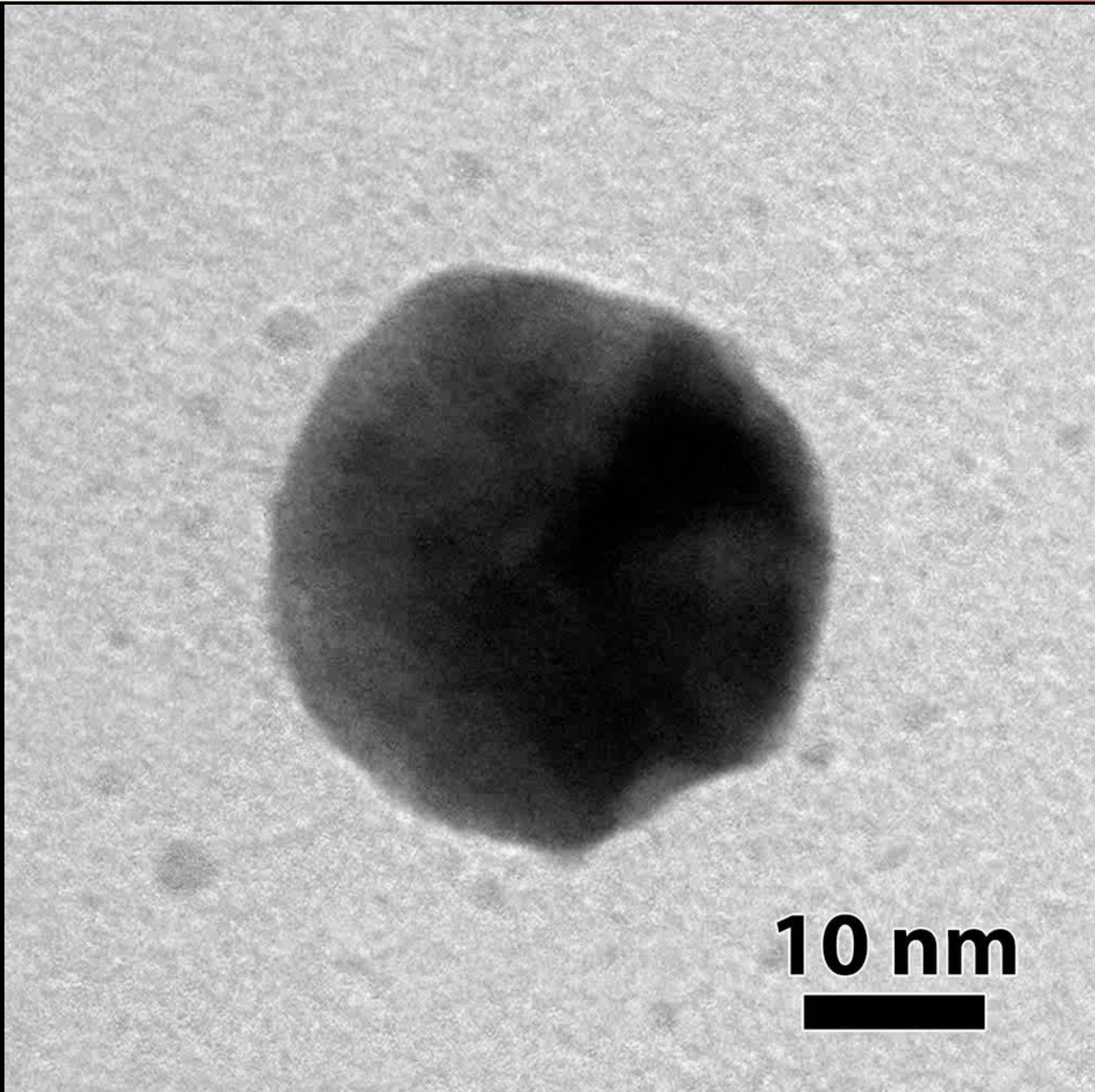
10 MeV Au^{8+}
1.3×10^{12} /cm²

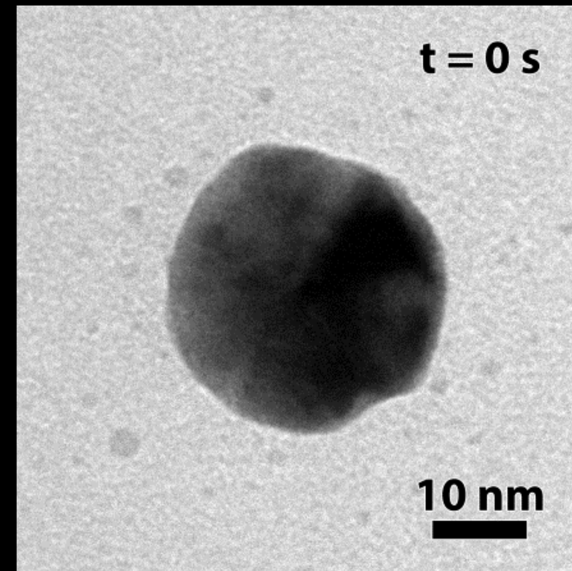


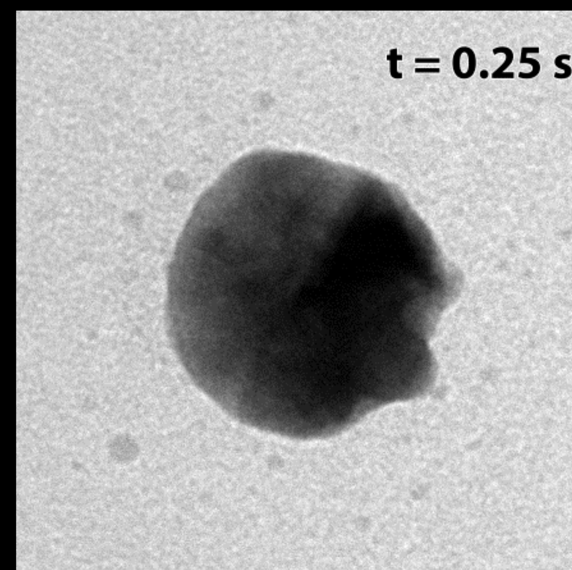
Particle and ion energy dictate the ratio of sputtering, particle motion, particle agglomeration, and <u>other active mechanisms</u>

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

Collaborator: D.C. Bufford

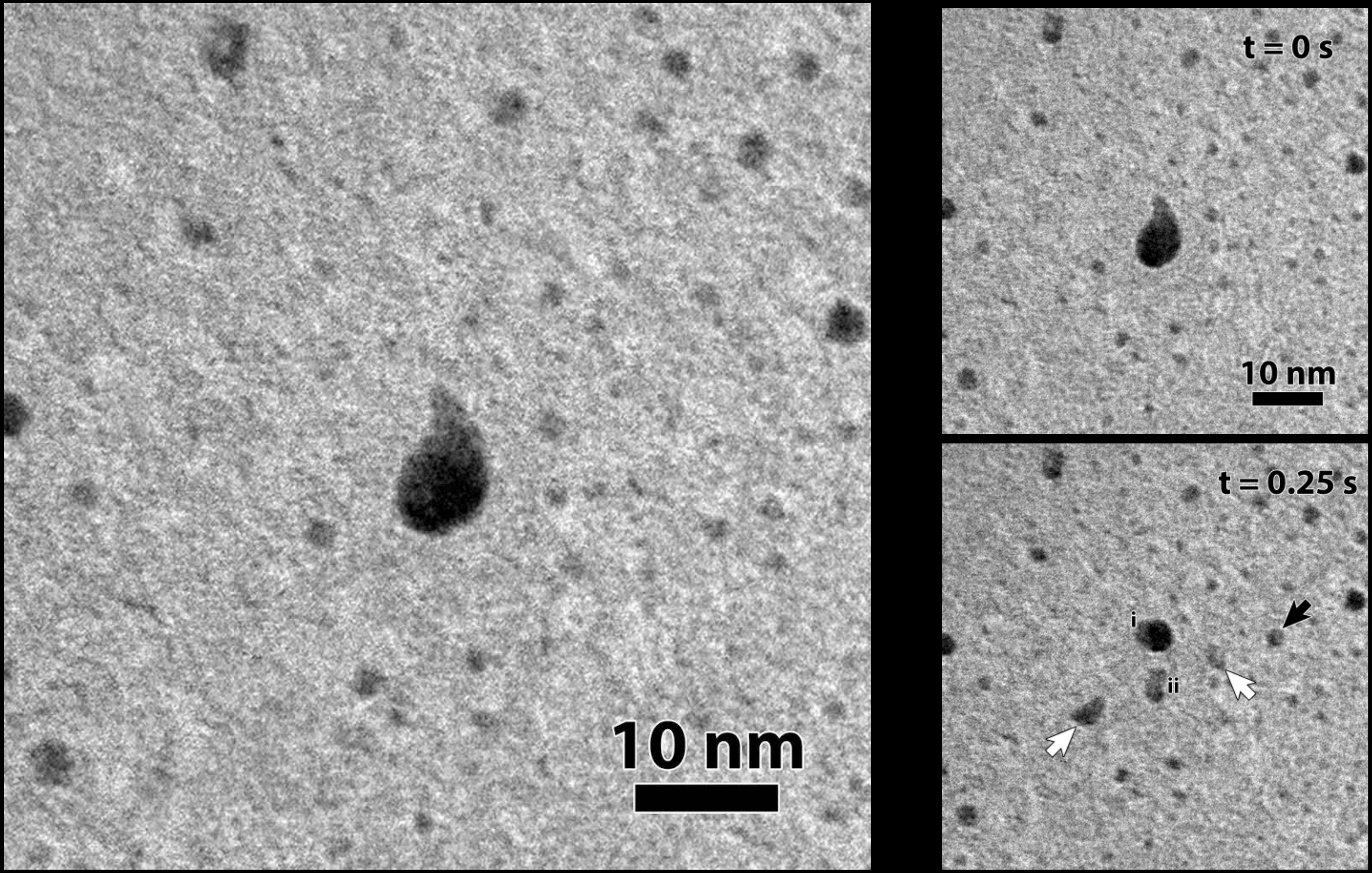






Single Ion Effects with 46 keV Au^{1-} ions: 5 nm

Collaborator: D.C. Bufford



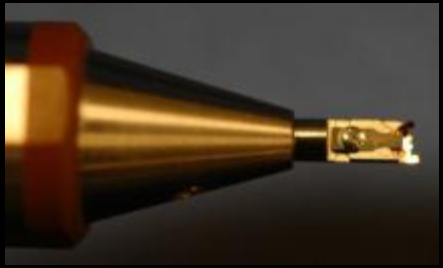
Advanced Microscopy Techniques Applied to Nanoparticles in Radiation Environments

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

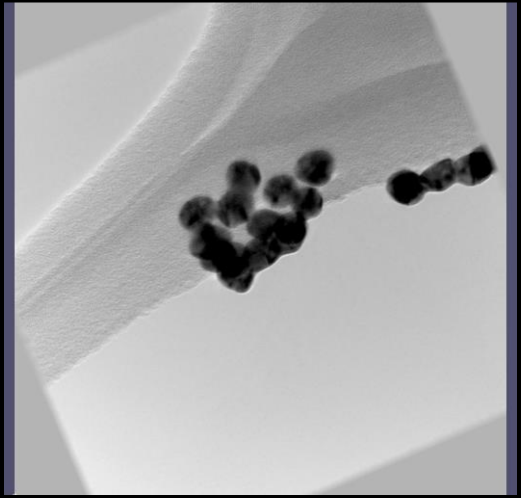
***In situ* Ion Irradiation TEM (I³TEM)**

+

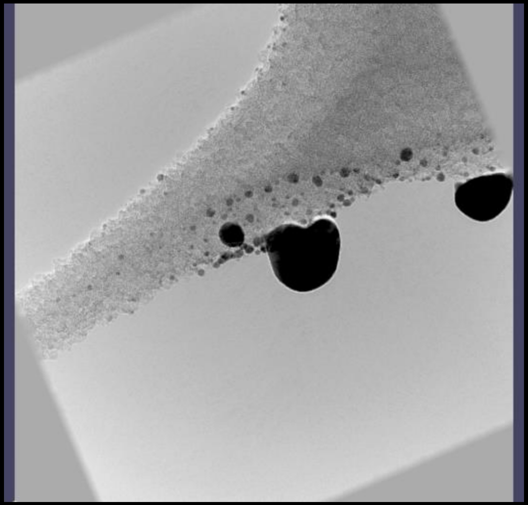
Hummingbird tomography stage



Aligned Au NP tilt series - unirradiated

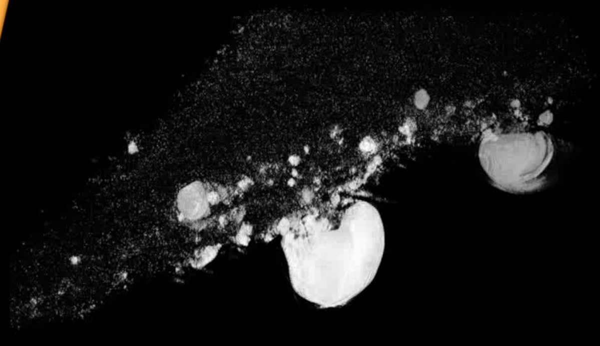


Aligned Au NP tilt series - irradiated



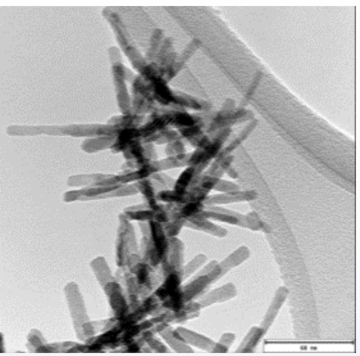
Unirradiated Au NP model

Irradiated Au NP model

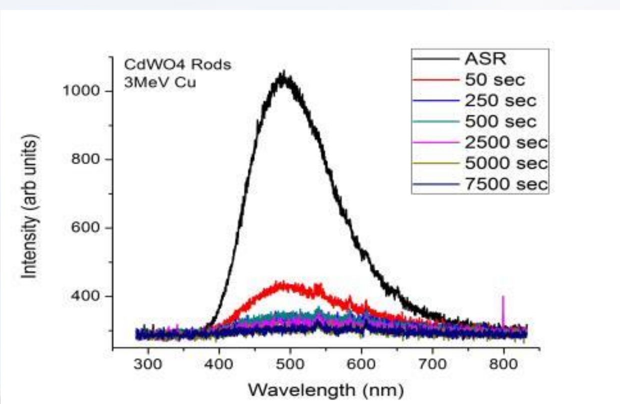


The application of advanced microscopy techniques to extreme environments provides exciting new research directions

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

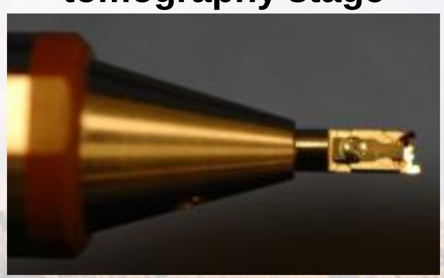


High-Z nanoparticles ($CdWO_4$) are promising, but are radiation sensitive

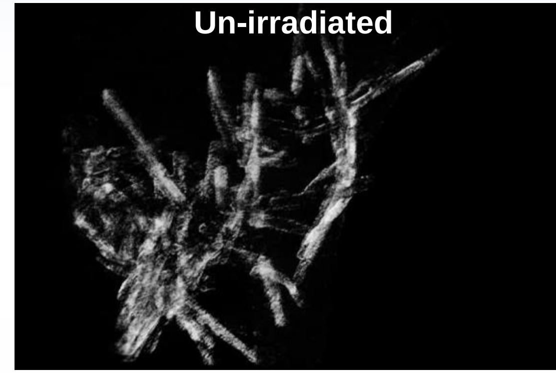


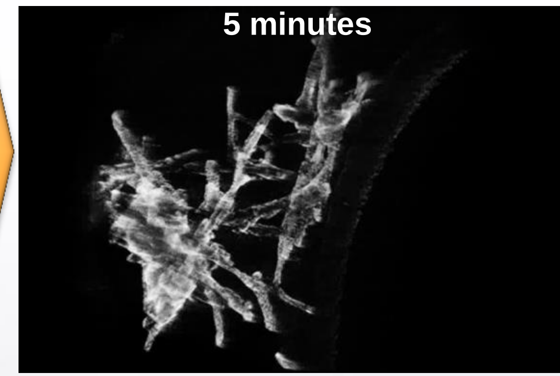
In situ Ion Irradiation TEM (I^3TEM)

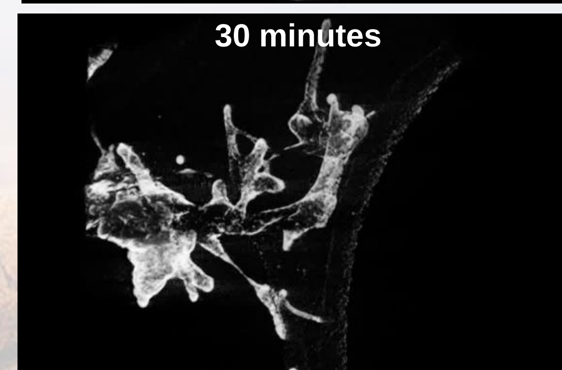
Hummingbird tomography stage



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



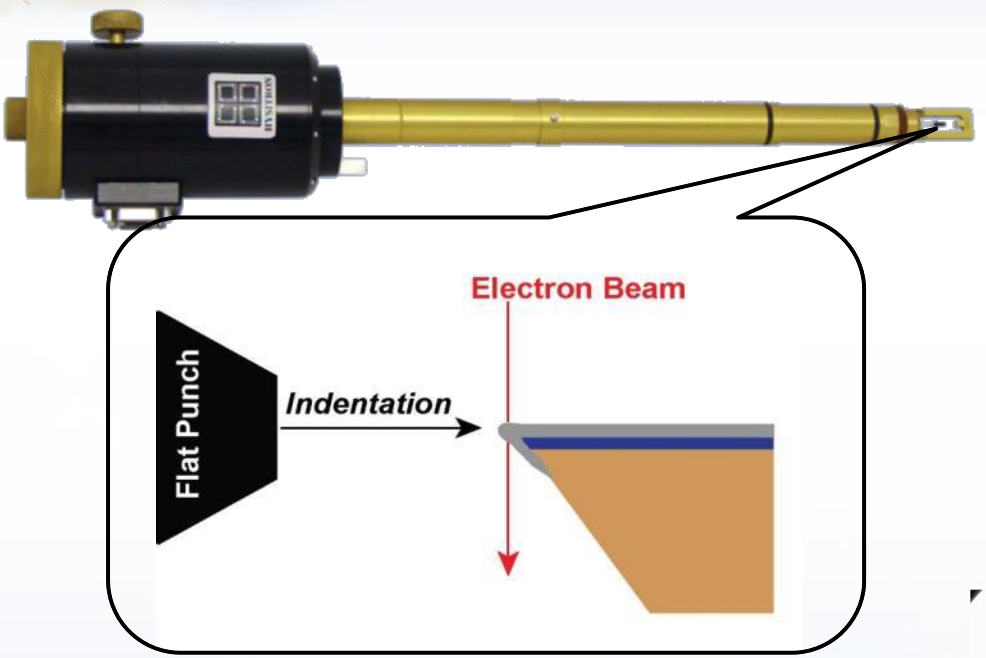


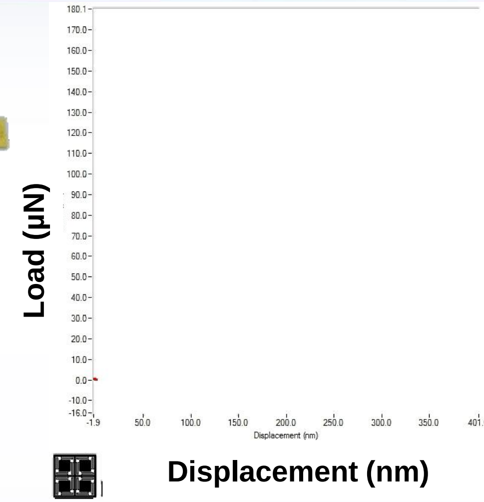


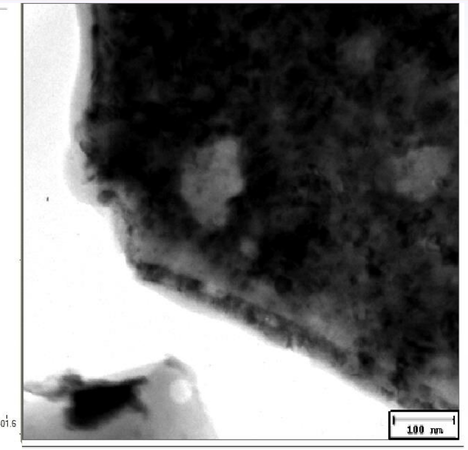
Contributors: S.M. Hoppe, B.A. Hernandez-Sanchez, T. Boyle

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.



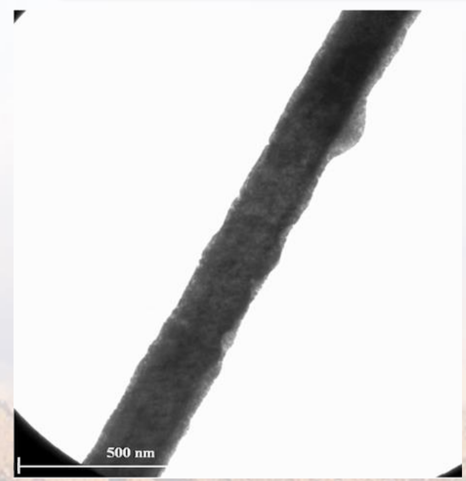


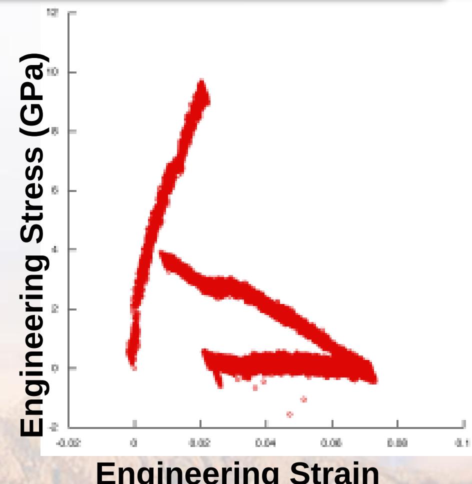


Fundamentals of Mechanical Properties

Range of Mechanical Testing Techniques

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

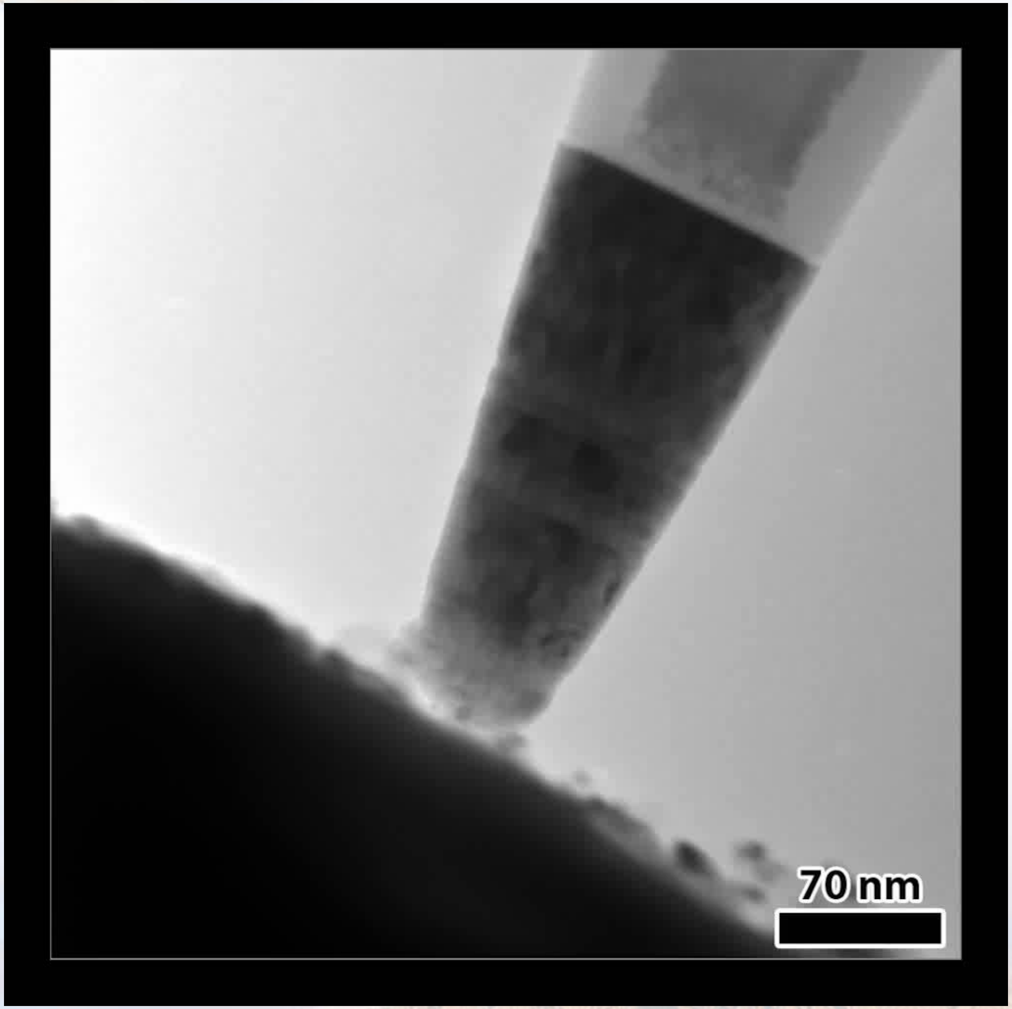




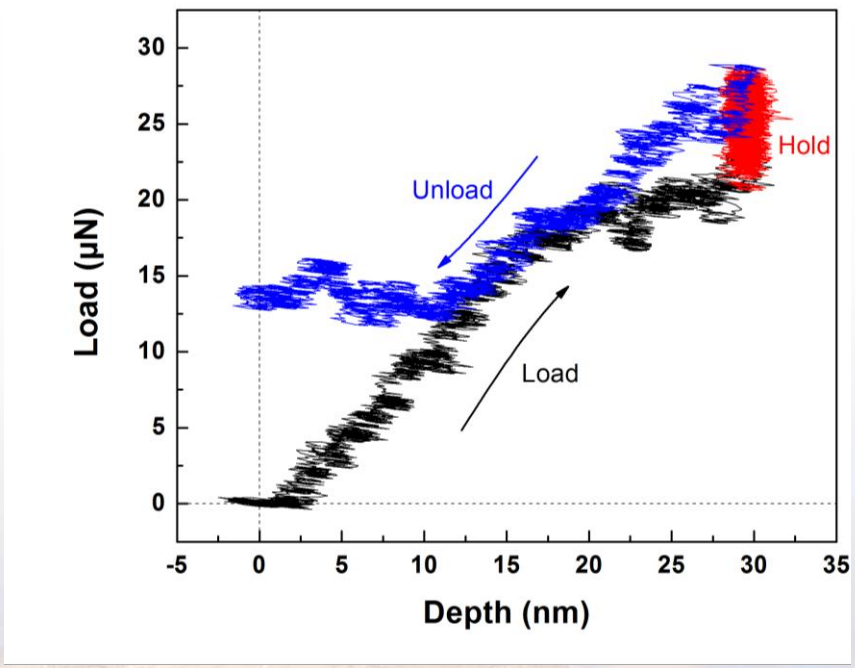
We have started looking at the effects of ion irradiation on mechanical properties

NC Ni Pillar Indentation

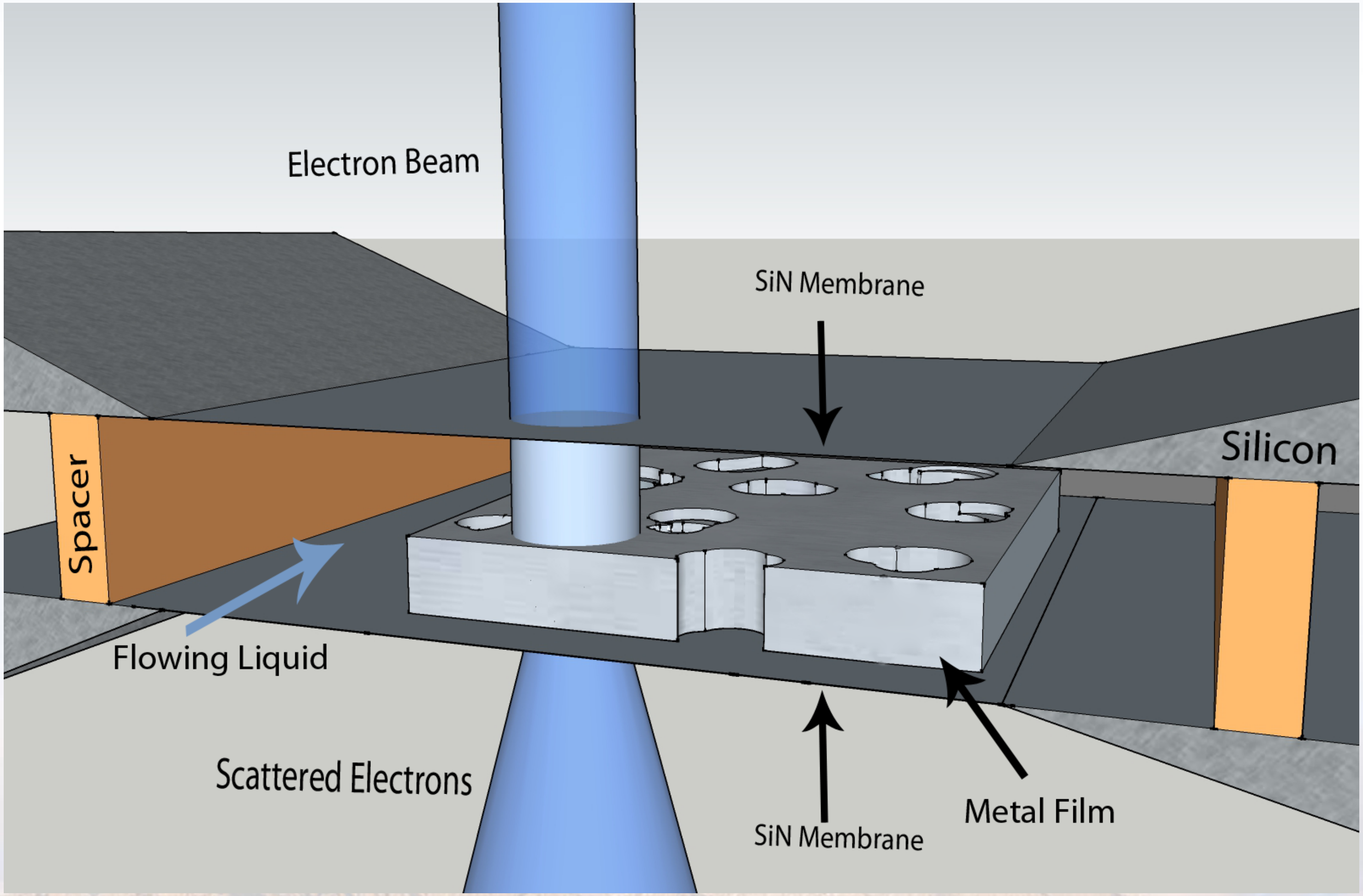
Collaborator: D.C. Bufford & W.M. Mook



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



Can We Gain Insight into the Corrosion Process through *In situ* TEM?

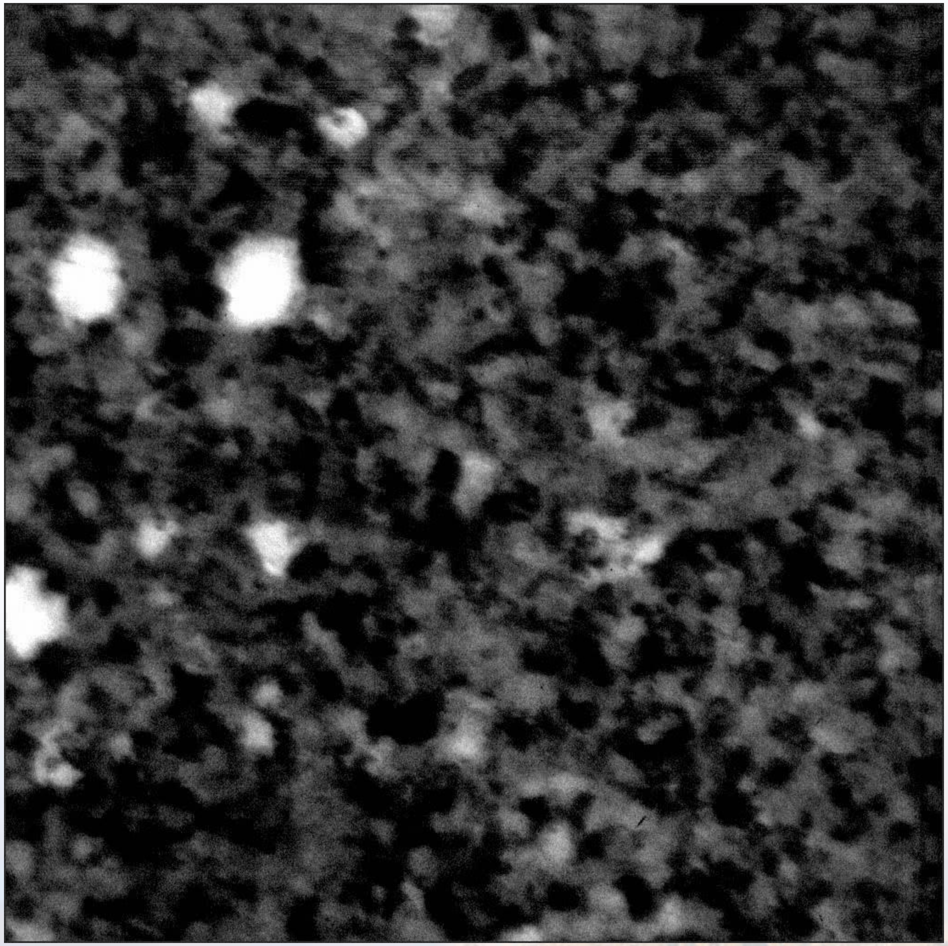


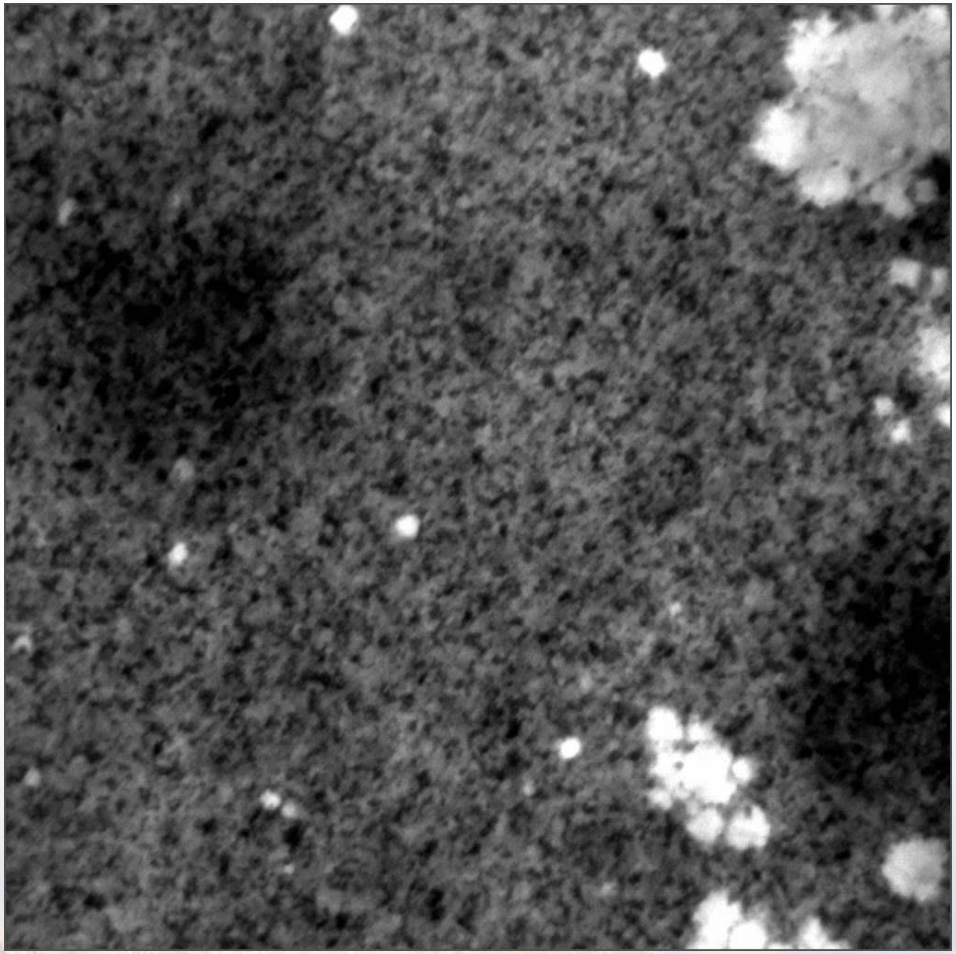
<u>Microfluidic Stage</u>

- **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**

Acetic Acid Corroding Nanograined Iron

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

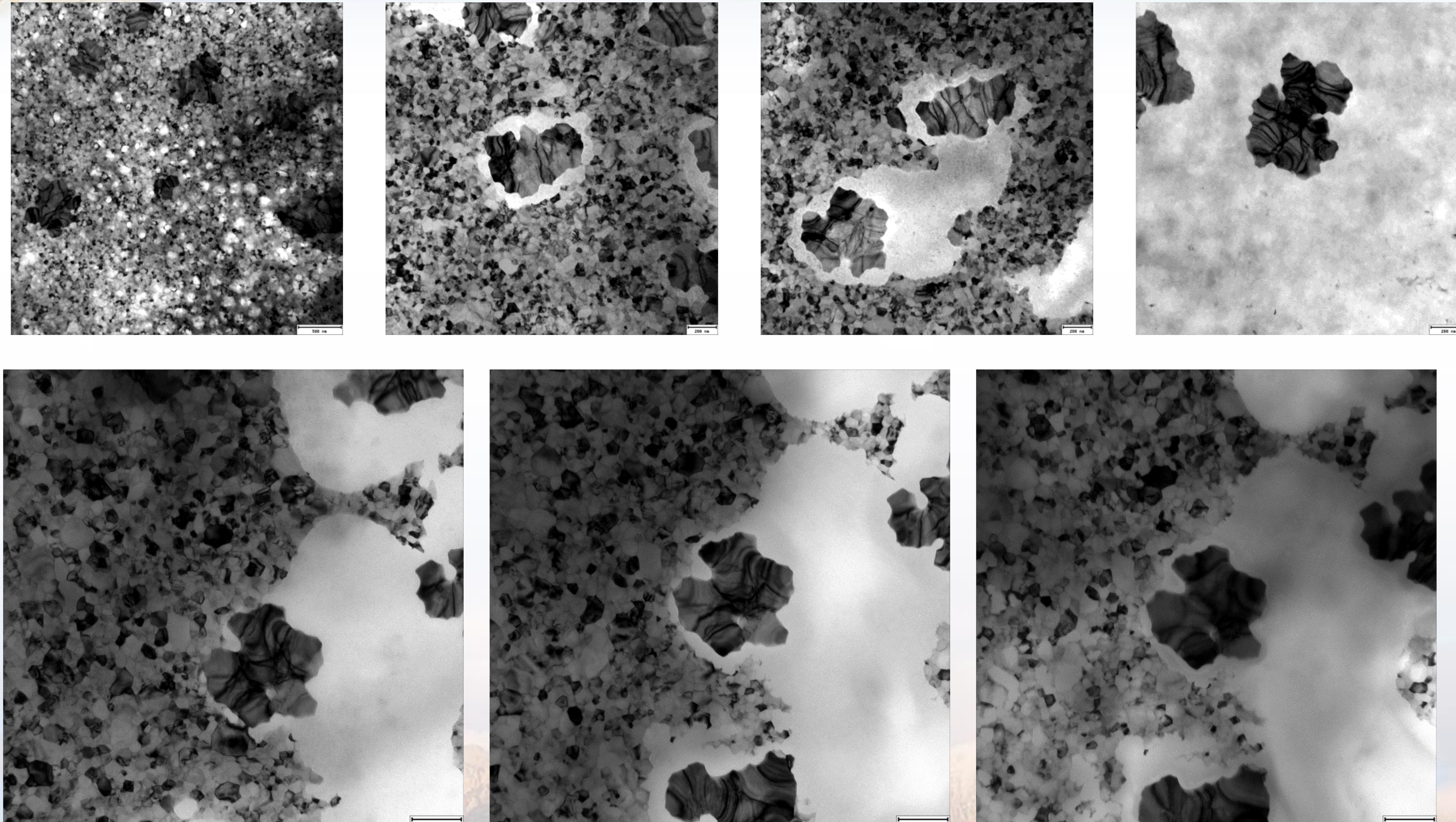




Pitting mechanisms during dilute flow of acetic acid over 99.95% nc-PLD Fe involves many grains.

Acetic Acid Corroding in Annealed Nanograined Iron

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



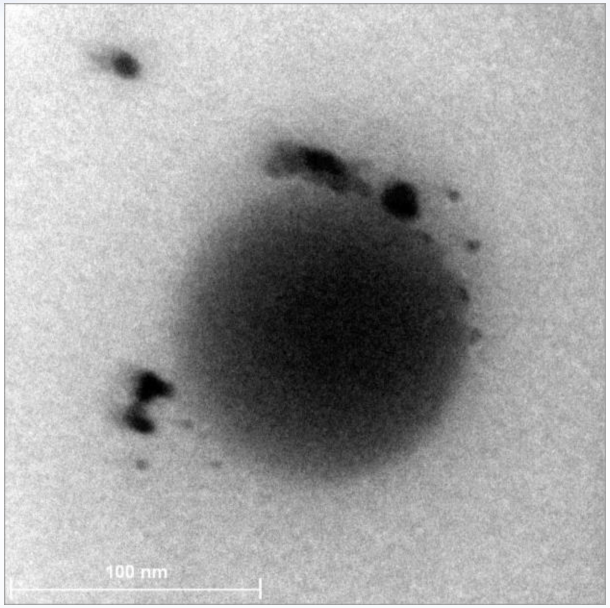
Large grains resulting from annealing appear more corrosion tolerant

Other Fun Uses of Microfluidic Cell

Protocell Drug Delivery

S. Hoppe,
E. Carnes,
J. Brinker

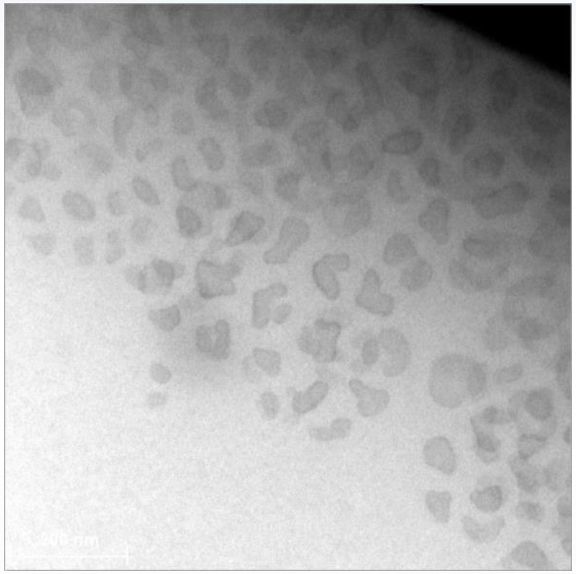
Liposome encapsulated Silica destroyed by the electron beam



BSA Crystallization

S. Hoppe

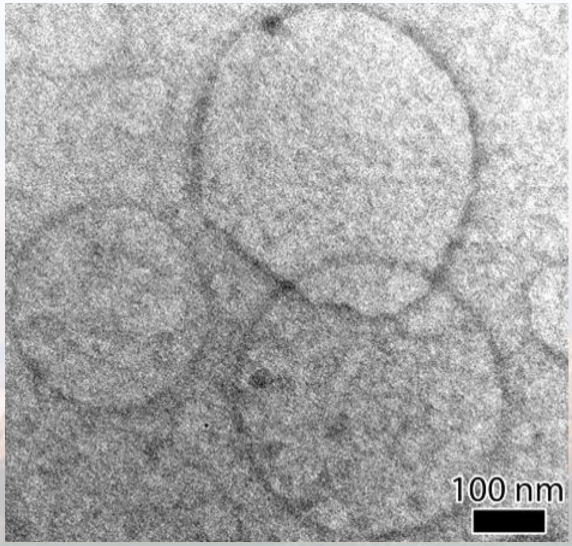
Crystallization of excess Bovine Serum Albumen during flow



Liposomes in Water

S. Hoppe,
D. Sasaki

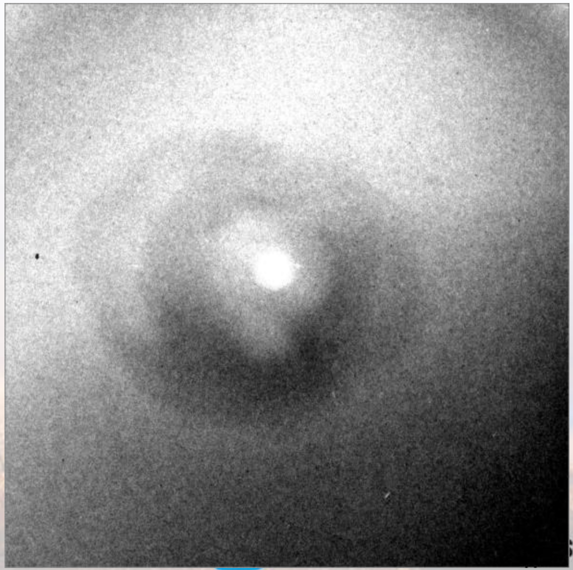
Liposomes imaged in flowing aqueous channel



La Structure Formation

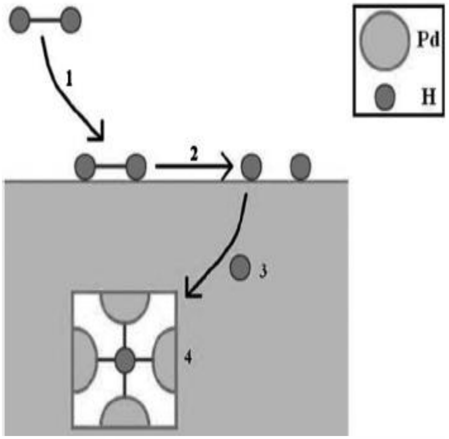
S. Hoppe,
T. Nenoff

La Nanostructure form from $LaCl_3$ H_2O in wet cell due to beam effects

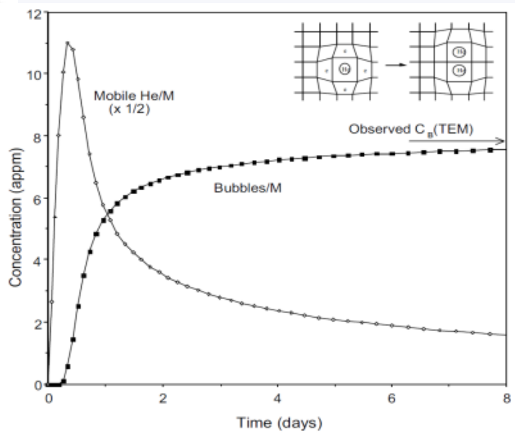


Can *In situ* TEM Address Hydrogen Storage Concerns in Extreme Environments?

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

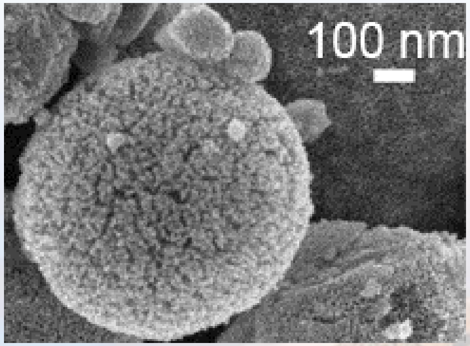


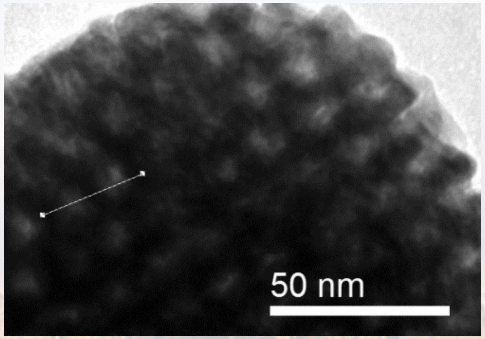
Cowgill, D., Fusion Sci. & Tech., 28 (2005) p. 539

Trinkaus, H. *et al*., JNM (2003) p. 229

Thiebaut, S. *et al*. JNM (2000) p. 217

Harmful effects may be mitigated in nanoporous Pd

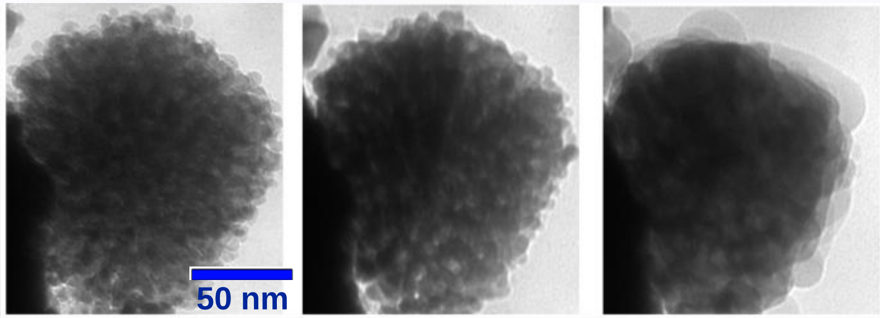




Vapor-Phase Heating TEM Stage

- 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.

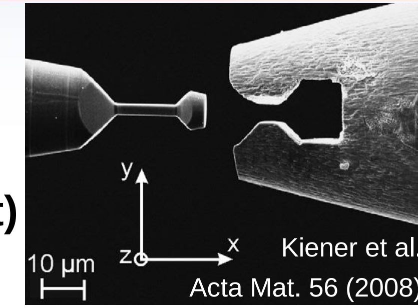


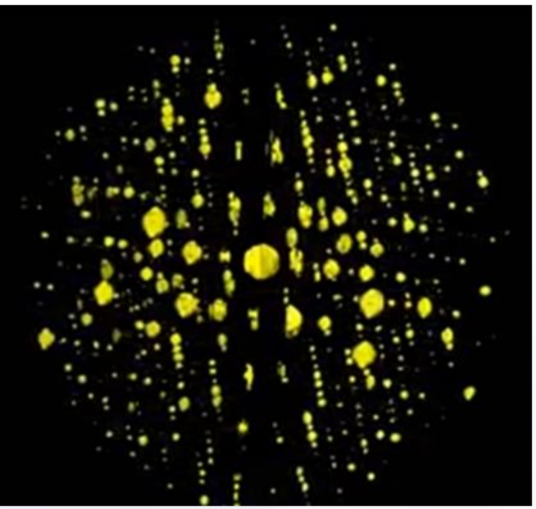
125° C 200° C 300° C

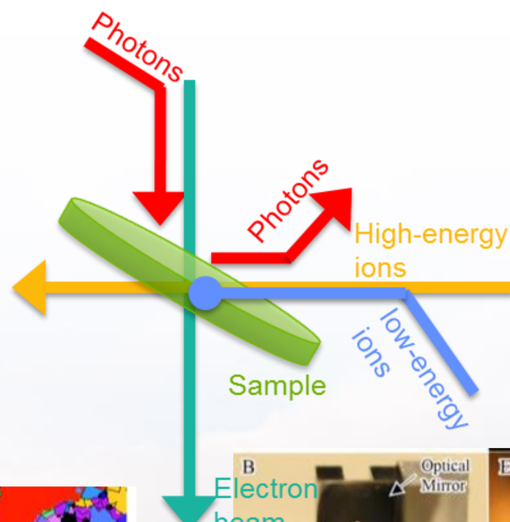
New *in situ* atmospheric heating experiments provide great insight into nanoporous Pd stability

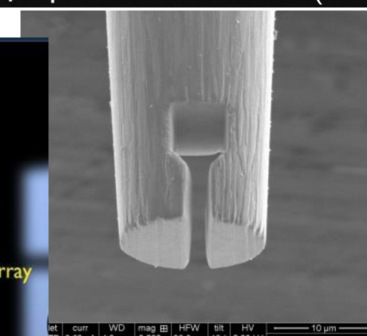
Future Directions Under Pursuit

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 (arriving FY15)
4. Quantative in-situ tensile/creep experiments (Sample in development)
5. DTEM: Nanosecond resolution (laser optics needed)

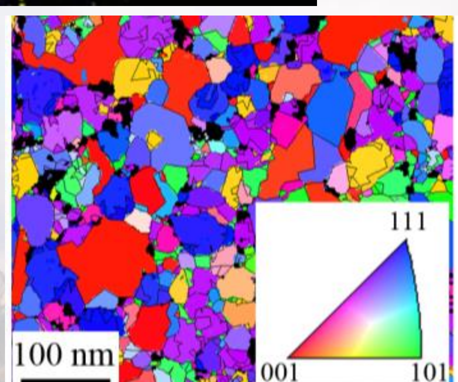


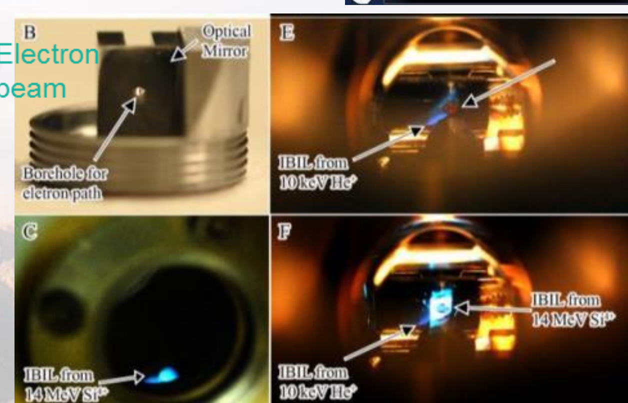


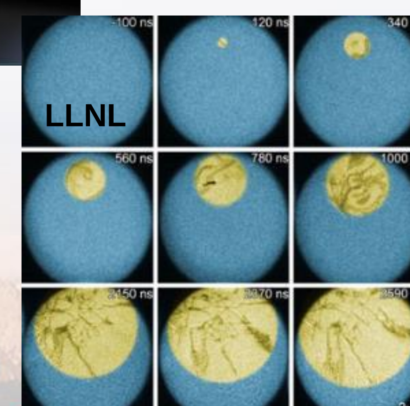




AppFive
NanoMegas

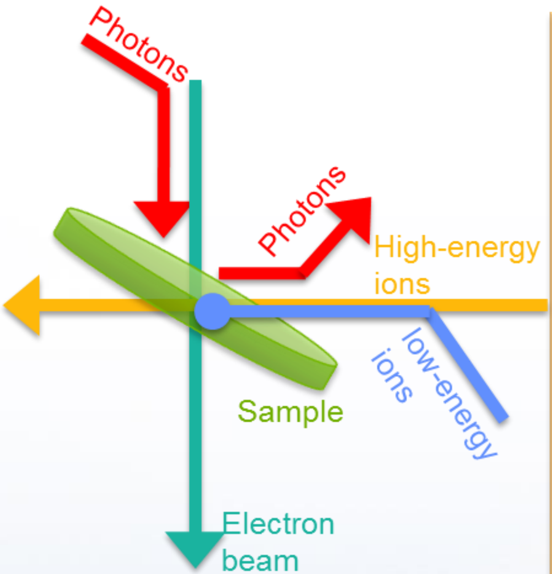






Sandia National Laboratories

Summary



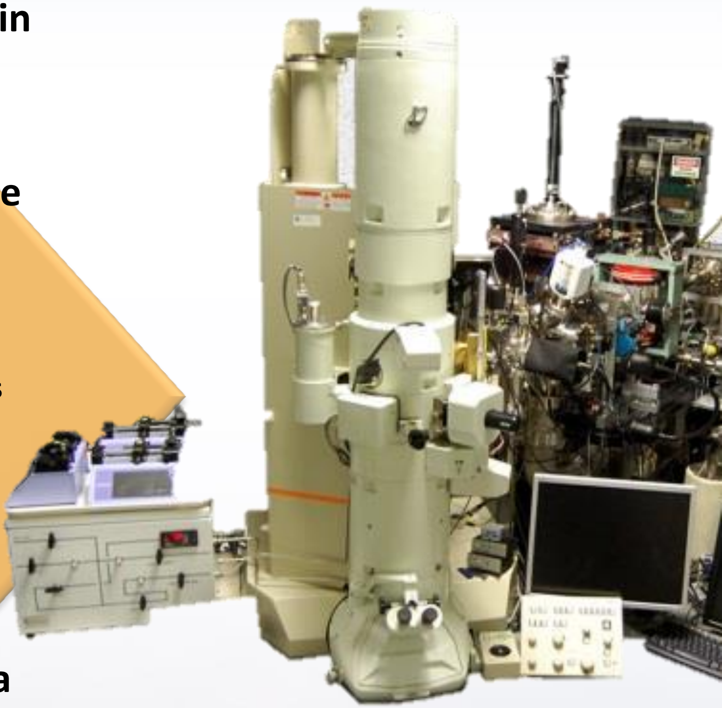
I³TEM can provide fundamental understanding to key mechanisms in a variety of extreme conditions

Sandia's I³TEM is one of a few in the world

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

The I³TEM capability are still being expanded…

We are still a long way away from a complete design process that goes from fundamental physics to system engineering



Collaborators:

- **IBL: D.C. Bufford, D. Buller, C. Chisholm, B.G. Clark, 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**
- **External: A. Minor, L.R. Parent, I. Arslan, H. Bei, E.P. George, P. Hosemann, D. Gross, J. Kacher, & I.M. Robertson**