Recent Advancements in *In situ* TEM Capabilities Applied to Extreme Environments Including the Synthesis of Surrogate Nuclear Fuel Nanosystems

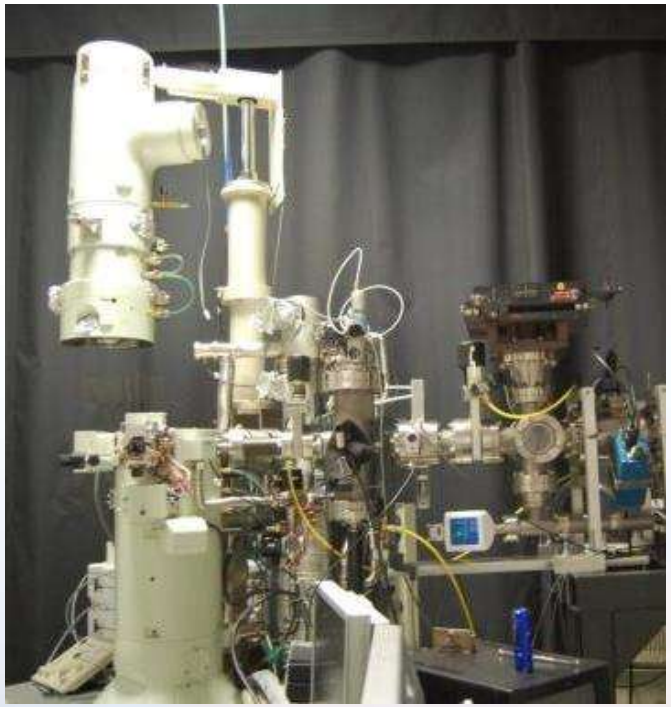
SAND2013-10155C

K. Hattar, S.H. Pratt, and T.M. Nenoff

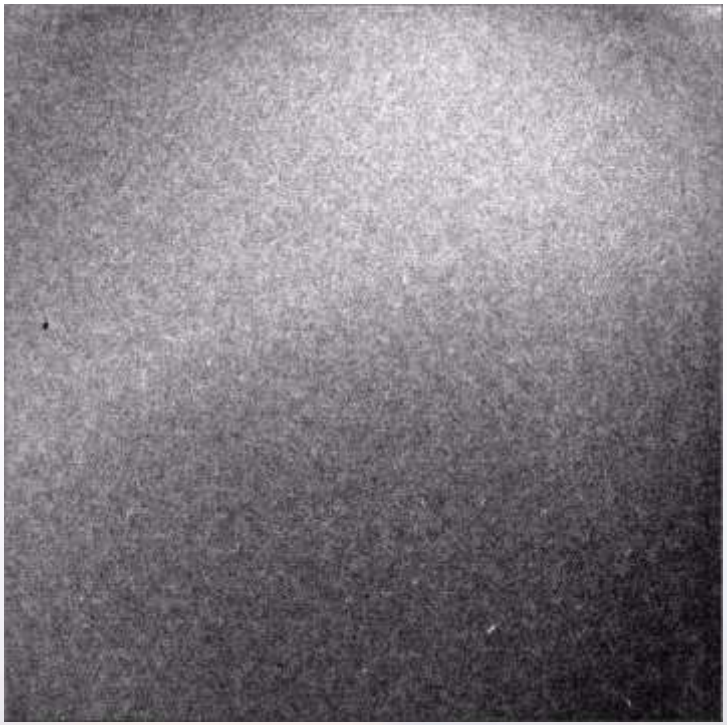
Ion Beam Lab

Sandia National Laboratories

December 5, 2013



***In situ* Ion Irradiation TEM is operational, while still in development it is providing experimental insight**



Collaborators:

D. Buller, J.A. Scott, D. Masiel, N. Li, A. Misra, B.A. Hernandez-Sanchez, T.J. Boyle, J. Sharon, B. L. Boyce, C. Chisholm, A.M. Minor, D. Sasaki, B.G. Clark, D. Gross, J. Kacher, P.J. Cappillino, B.W. Jacobs, L.R. Parent, I. Arslan, M.A. Hekmaty, D.B. Robinson, & I.M. Robertson

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.

Sandia National Laboratories

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

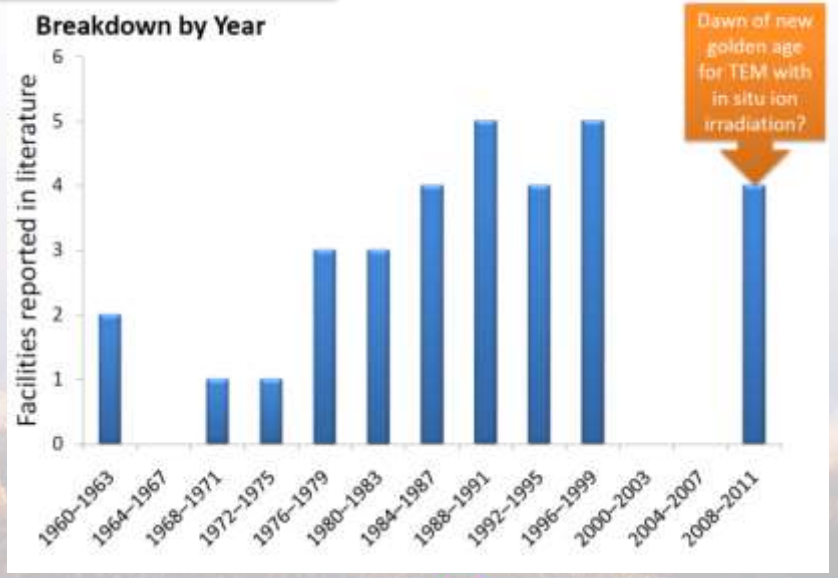
1978
First in-situ ion irradiation experiments at ANL

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

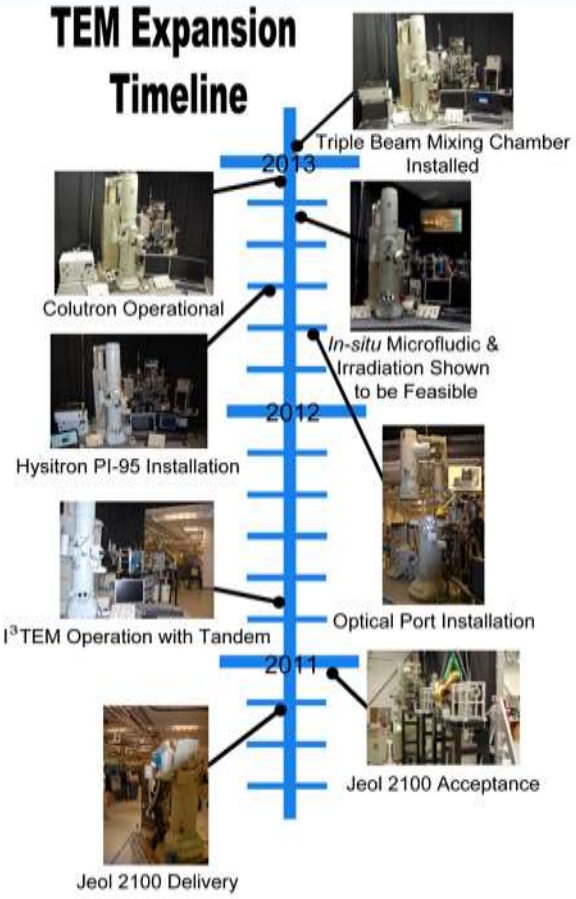
Workshop on Ion Irradiation TEM
Huddersfield, UK (2008)
Albuquerque, USA (2011)
Saporro, Japan (2013)

“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



I³TEM History at the Ion Beam Lab



Current Status of the *In situ* TEM Beamline

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

Beam burn from 14 MeV Si

10 kV Colutron

Double tilt stage needs to tilt only 12°

6 MV Tandem

Gas Heating TEM Stage Controls

Microfluidic TEM Stage Controls

Bending Magnet to Mix Beams

Quantitative Mechanical Testing

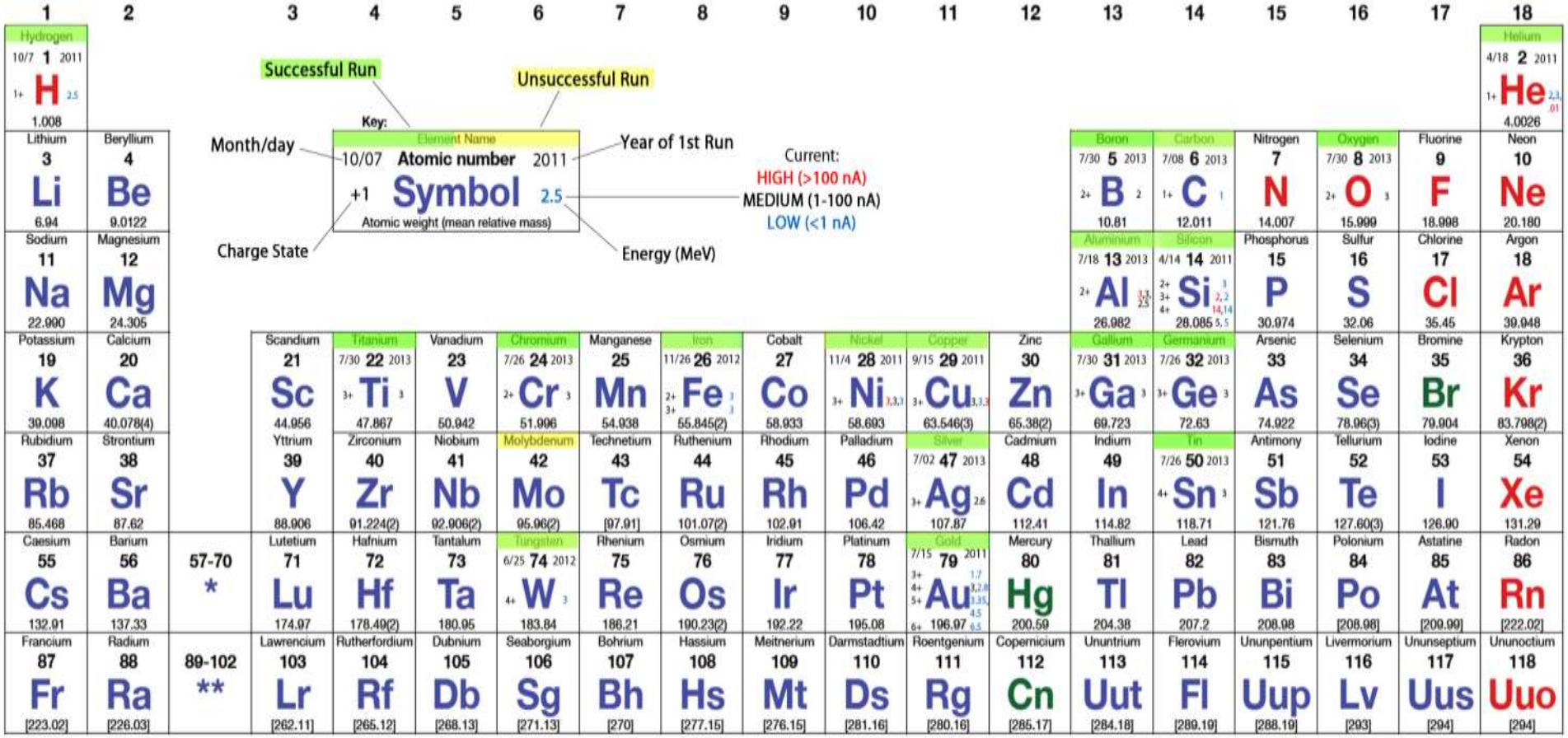
TEM Stage Controls

Pre-TEM Coupon Irradiation Chamber

I^{3}TEM is operational,

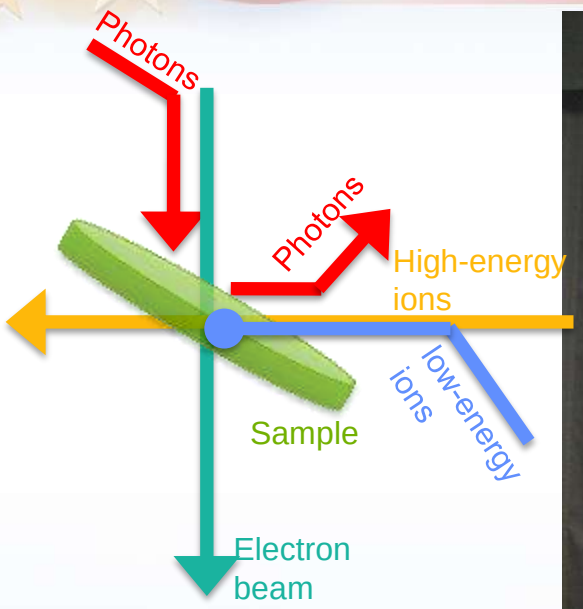
but also still in development

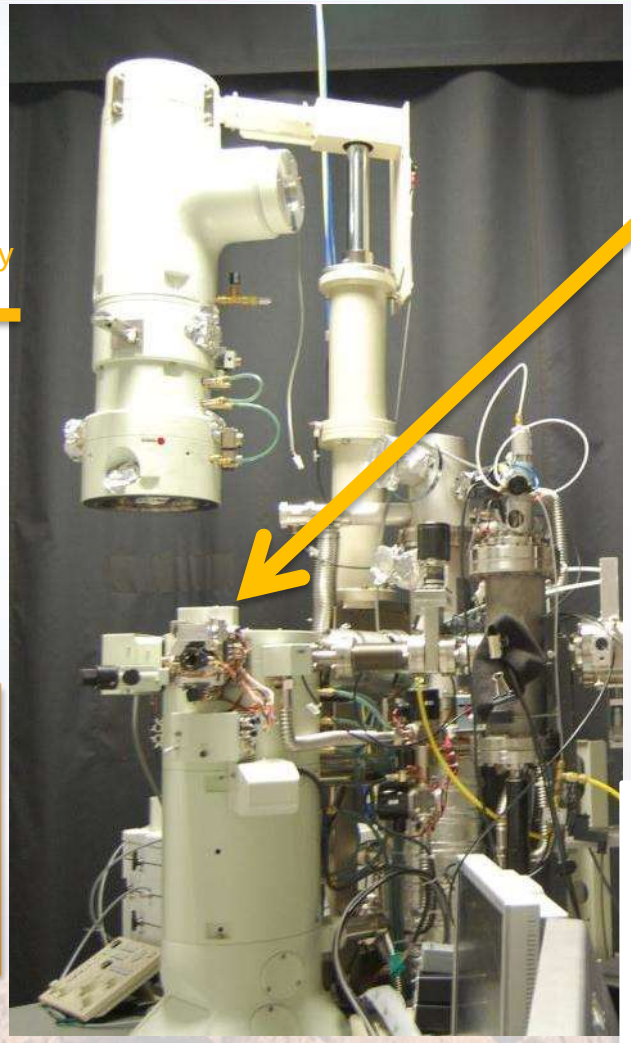
Ion Species Attempted as of 7/23/2013



In situ TEM Luminescence

Collaborators: D. Masiel, D. Buller, and C. Chisholm

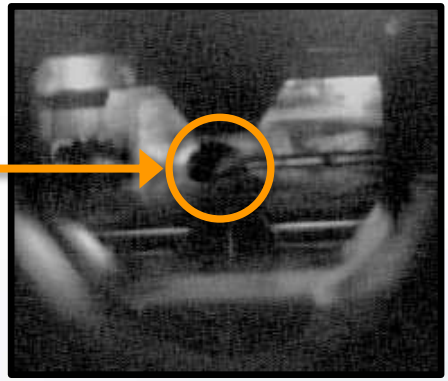




Optical Mirror in TEM

First IBIL in TEM

Sample

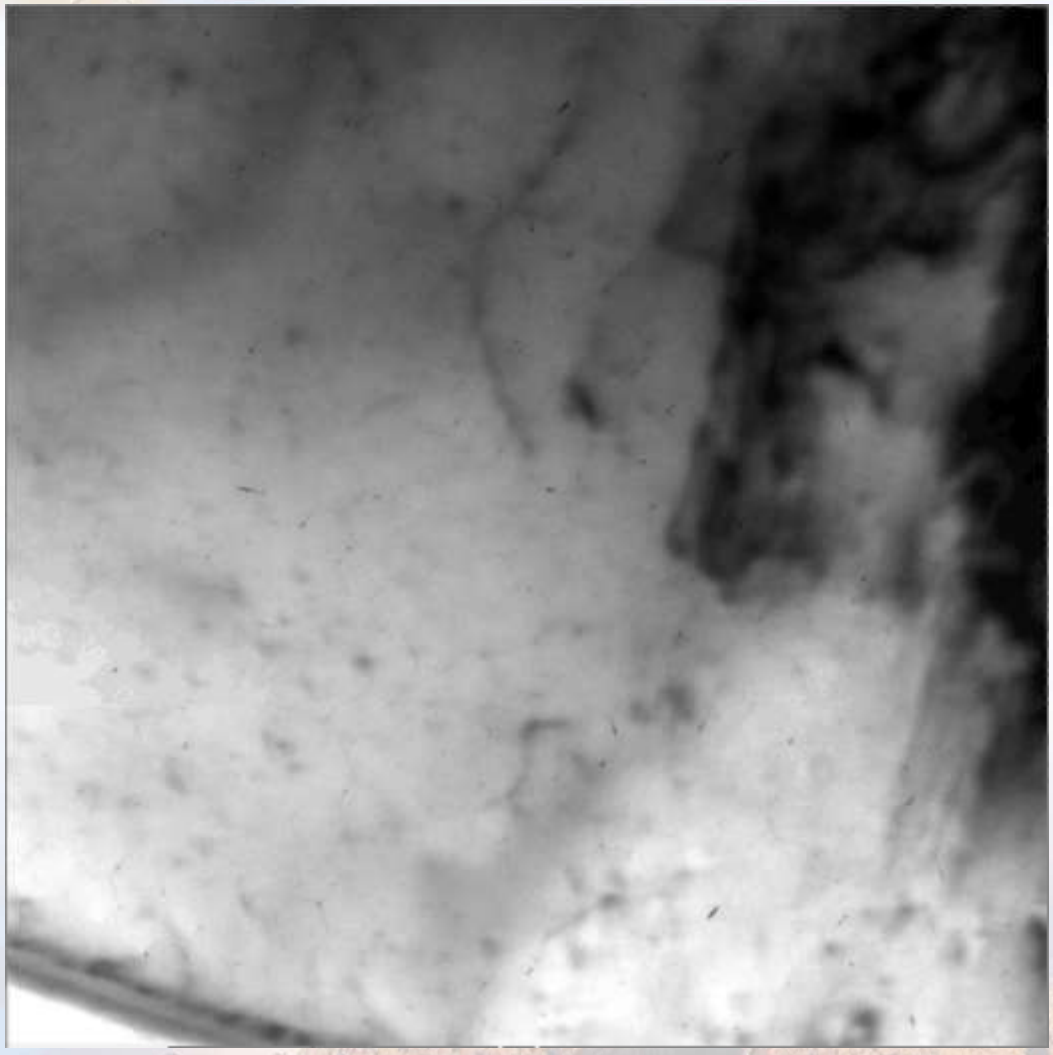


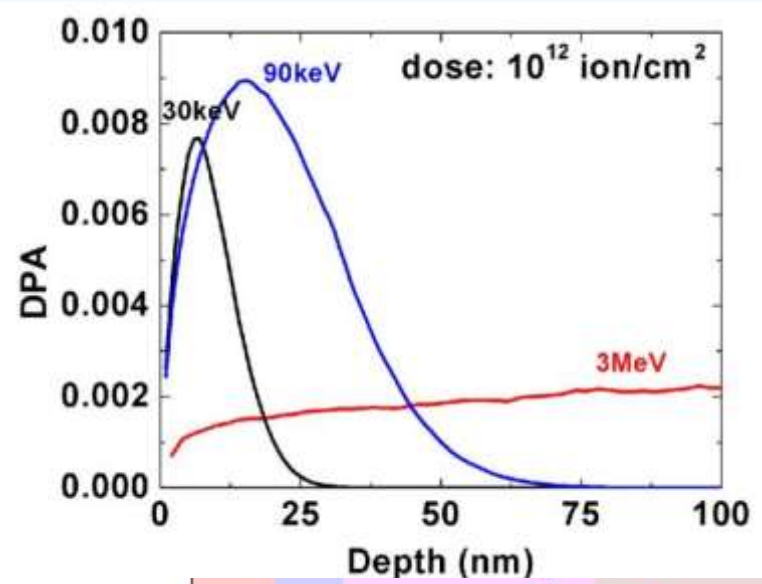
Two optical port were added to the I³TEM already containing a electron beam and two ion beams, which permits *in situ* TEM luminescence studies

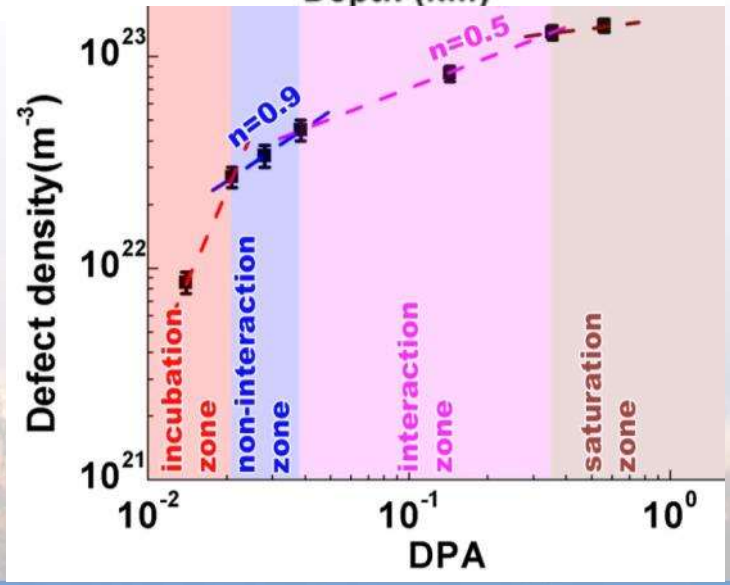
Optical Pathway in an I³TEM

- Angled mirror with bore hole for the electron path was installed above the polepiece
- Another mirror is located just above the ion beams in the beamline
- Two perspective of the sample are possible
- Permits *in situ* IBIL and CL.

Quantifying Defect Evolution



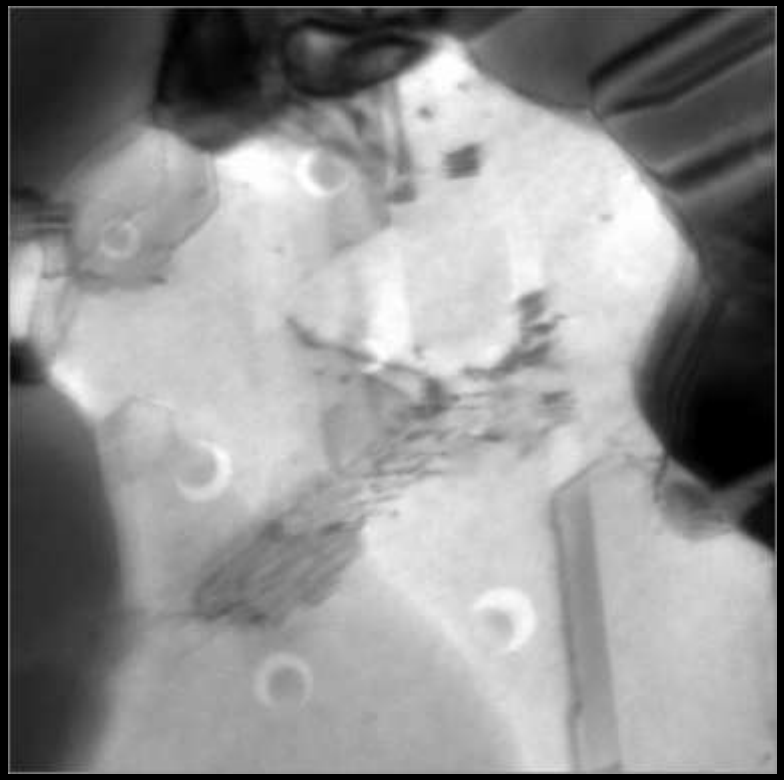




Single Ion Strikes

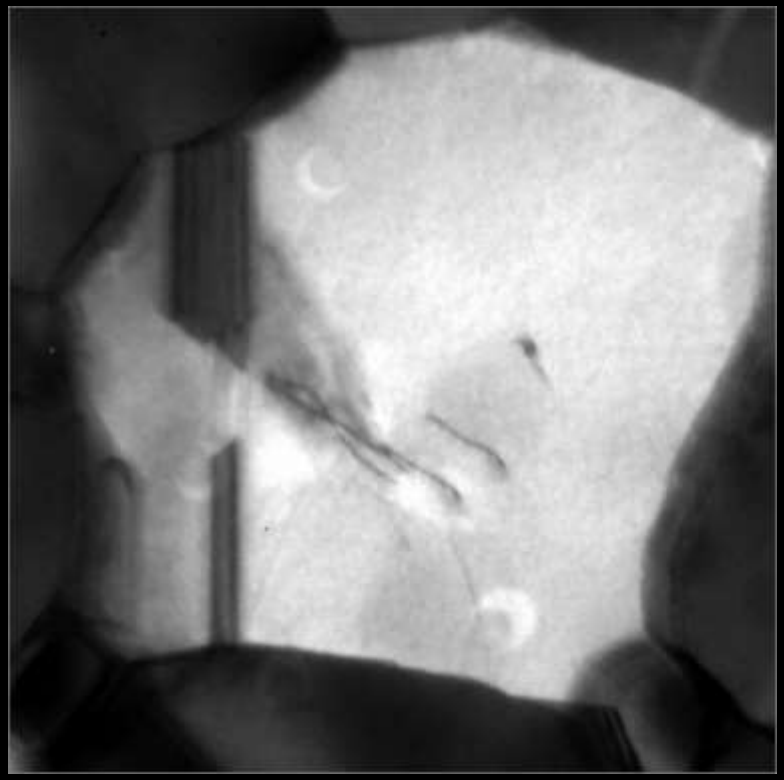
Collaborators: C. Chisholm & A. Minor

7.9×10^9 ions/cm^2/s



VS

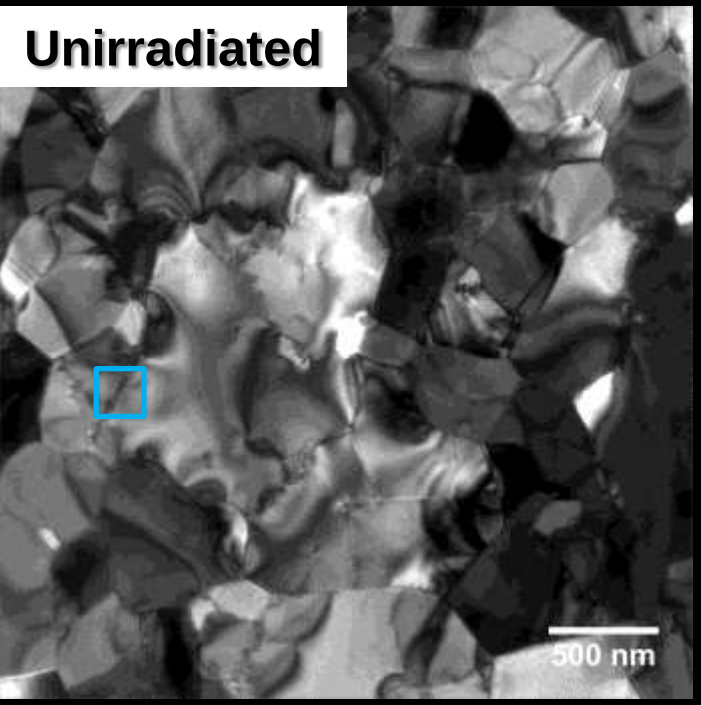
6.7×10^7 ions/cm^2/s



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

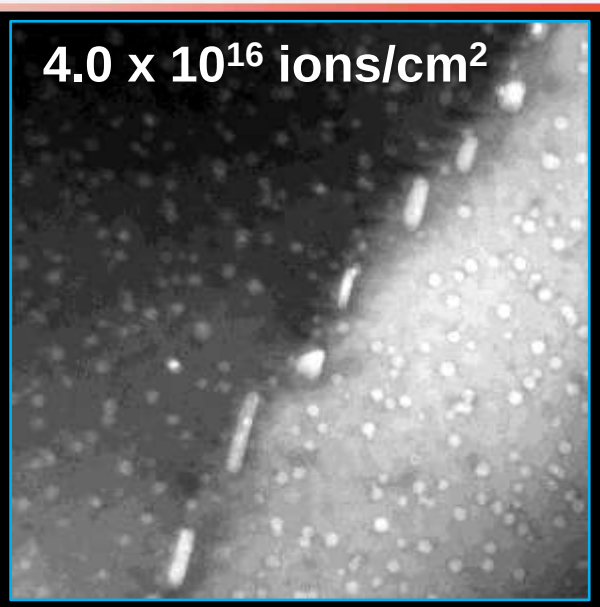
In situ Implantation

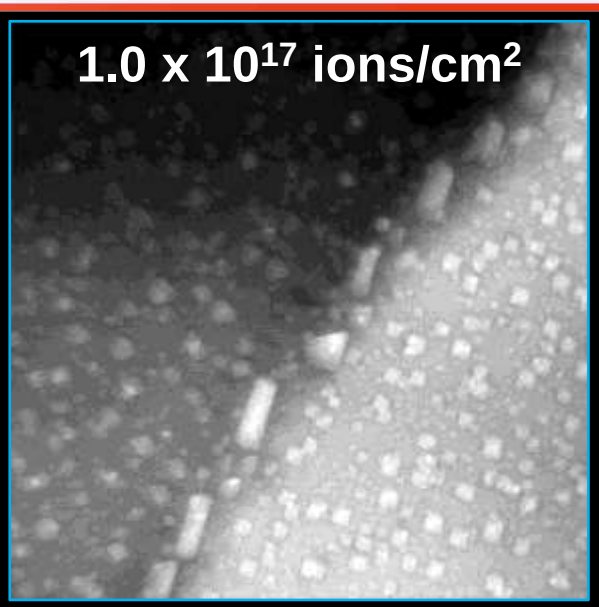
Collaborators: C. Chisholm & A. Minor

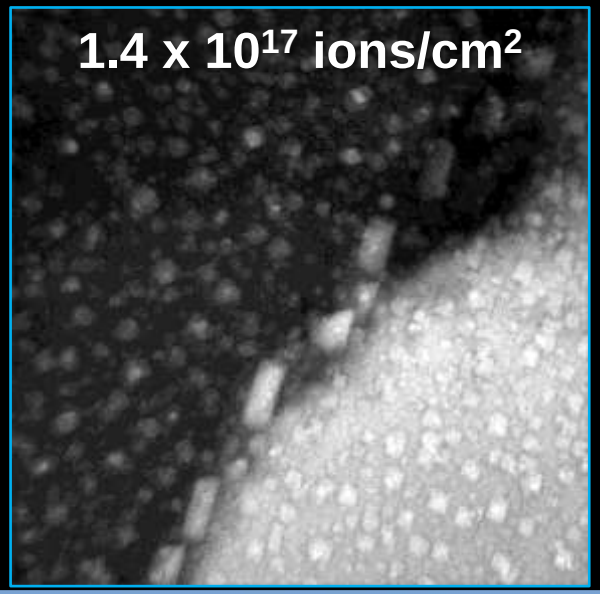


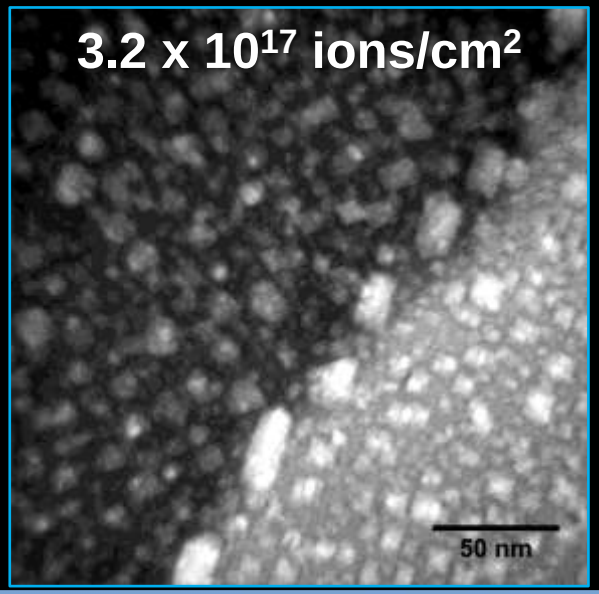
Gold thin-film implanted with 10keV He^{2+}

Result: porous microstructure









Single Ion Strikes During Concurrent Irradiation: Nucleation of Helium Cavities

Collaborators: C. Chisholm & A. Minor

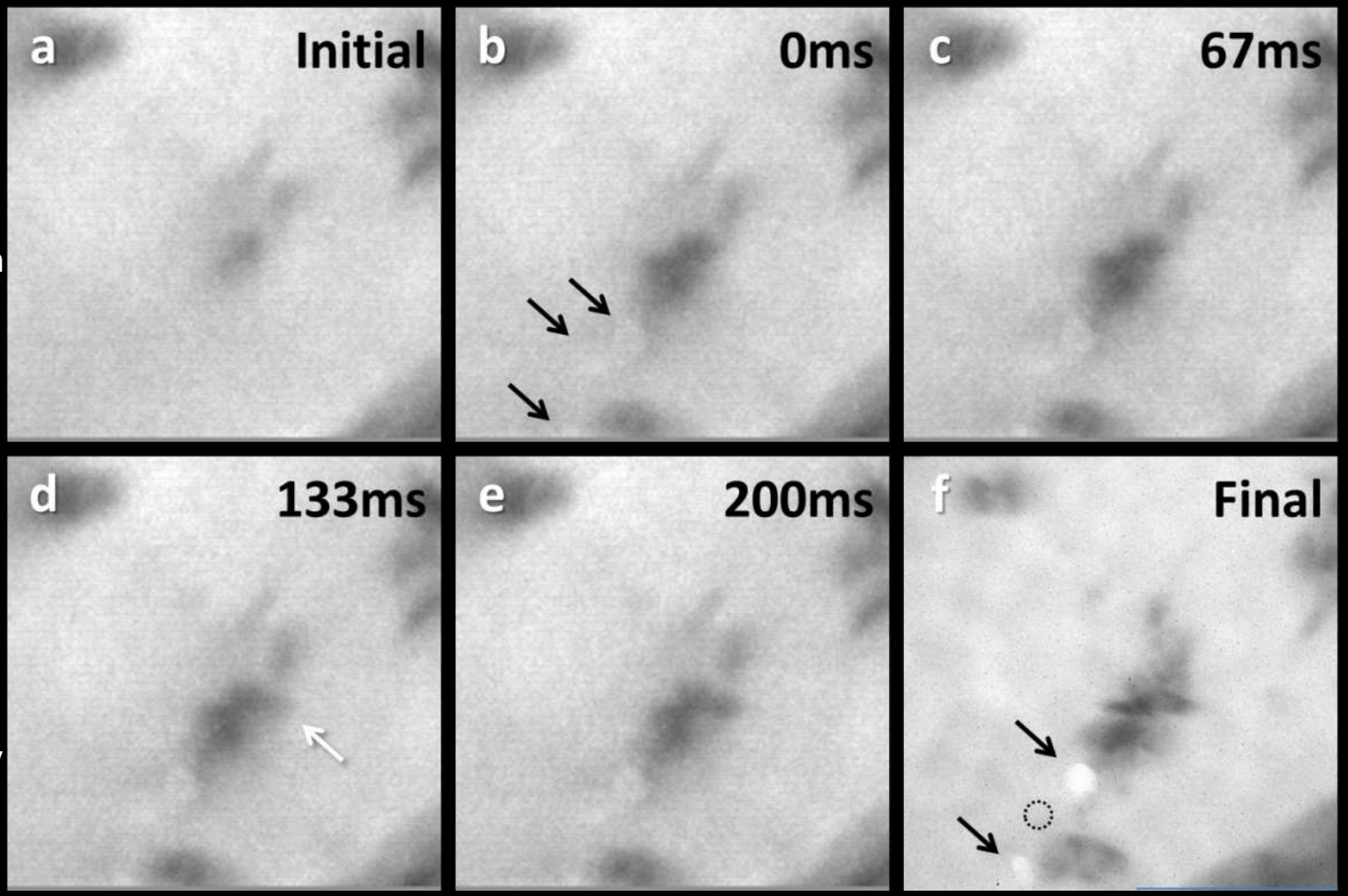
a) Initial microstructure

b) Cascade: Creation of dislocation loops, vacancy clusters, and three cavities

d) Cascade damage still evolving

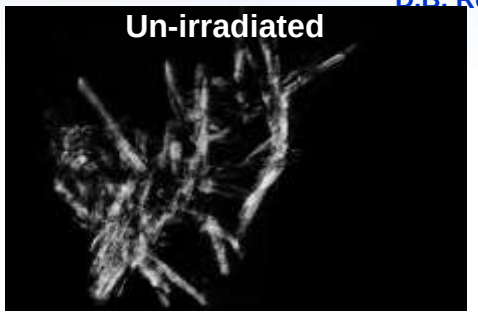
e) Apparent stability

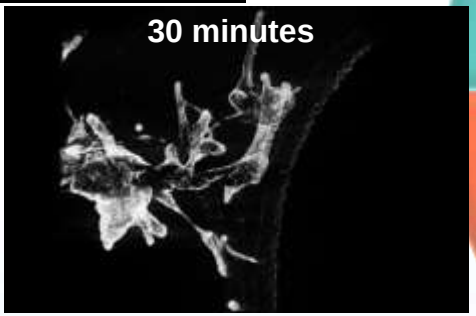
f) Final microstructure: Only two remaining cavities

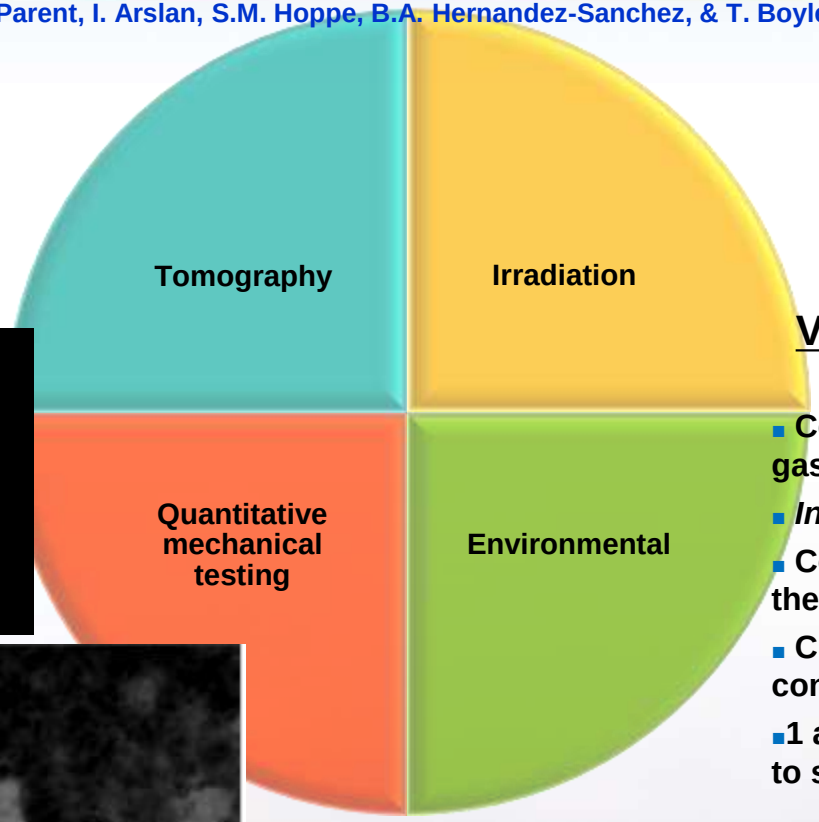


Add in-situ Capabilities from Advancements in TEM Stages

Contributors: J. Sharon, B. L. Boyce, C. Chisholm, A.M. Minor, B.G. Clark, P.J. Cappillino, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, L.R. Parent, I. Arslan, S.M. Hoppe, B.A. Hernandez-Sanchez, & T. Boyle

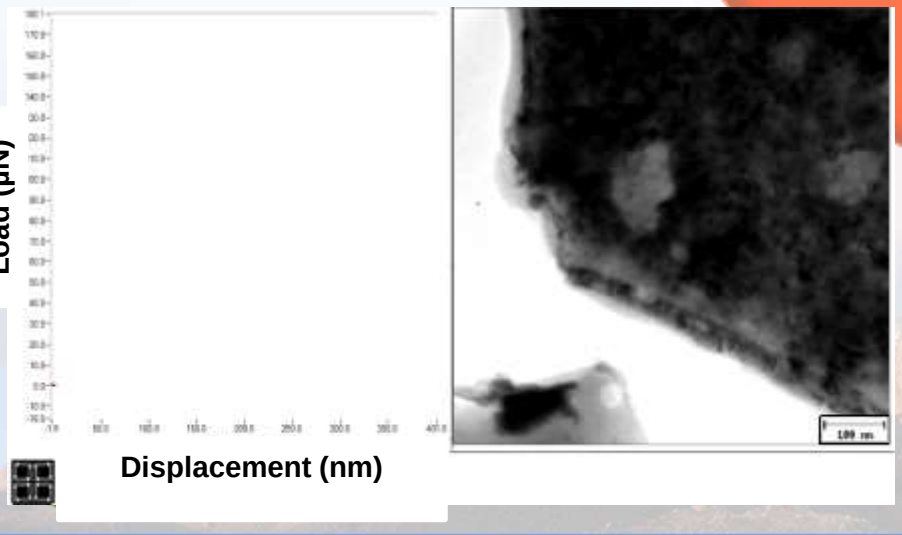


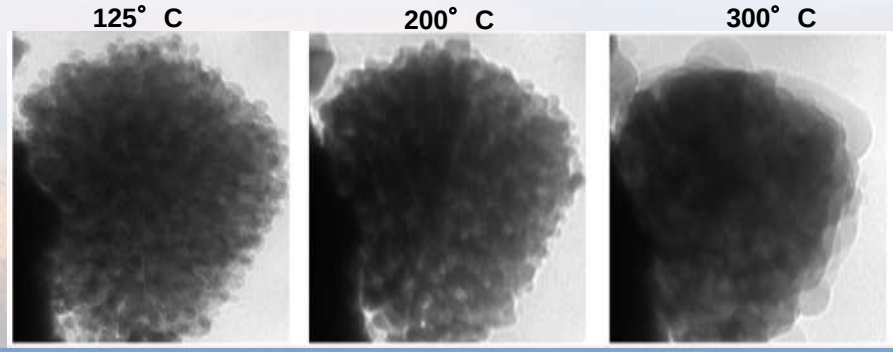




Vapor-Phase Heating TEM Stage

- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- 1 atm H_2 after several pulses to specified temp



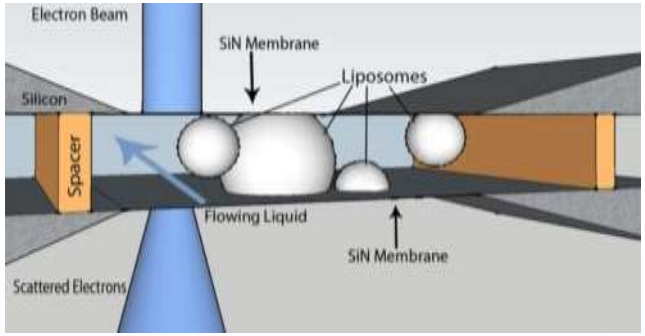


In situ TEM microfluidic Environments

Contributors: S.H. Pratt, E. Carnes, J. Brinker, D. Sasaki, D. Gross, J. Kacher, I.M. Robertson & Protochips Inc.

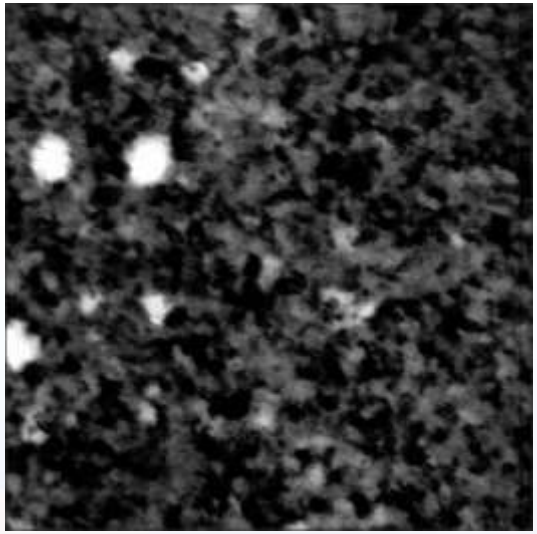
Microfluidic Stage

- Mixing of two or more channels
- Continuous observation of the reaction channel
- Chamber dimensions are controllable



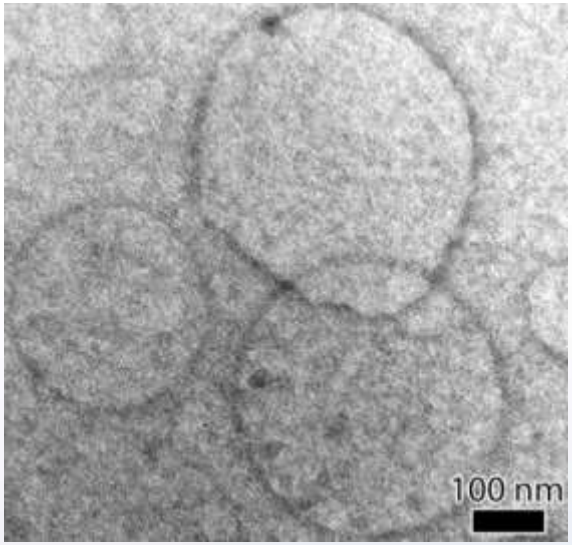
***In situ* microfluidic TEM can provide insight in events as diverse as corrosion and drug delivery**

Fe Corrosion



Dilute flow of acetic acid over 99.95% nc-PLD Fe

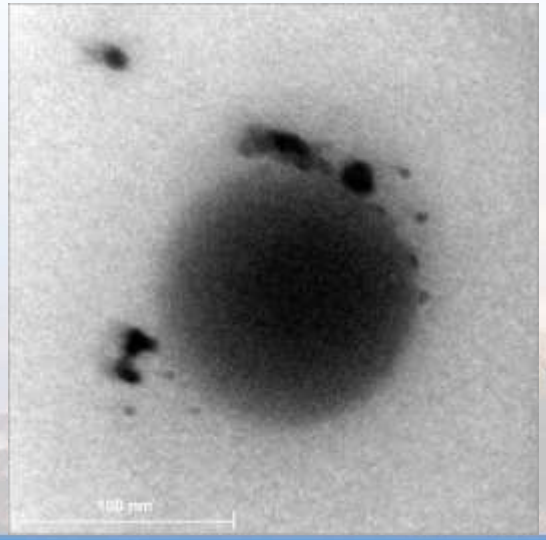
Liposomes in Water



Liposomes imaged in flowing aqueous channel

Protocell Drug Delivery

Liposome encapsulated Silica destroyed by the electron beam in aqueous environment

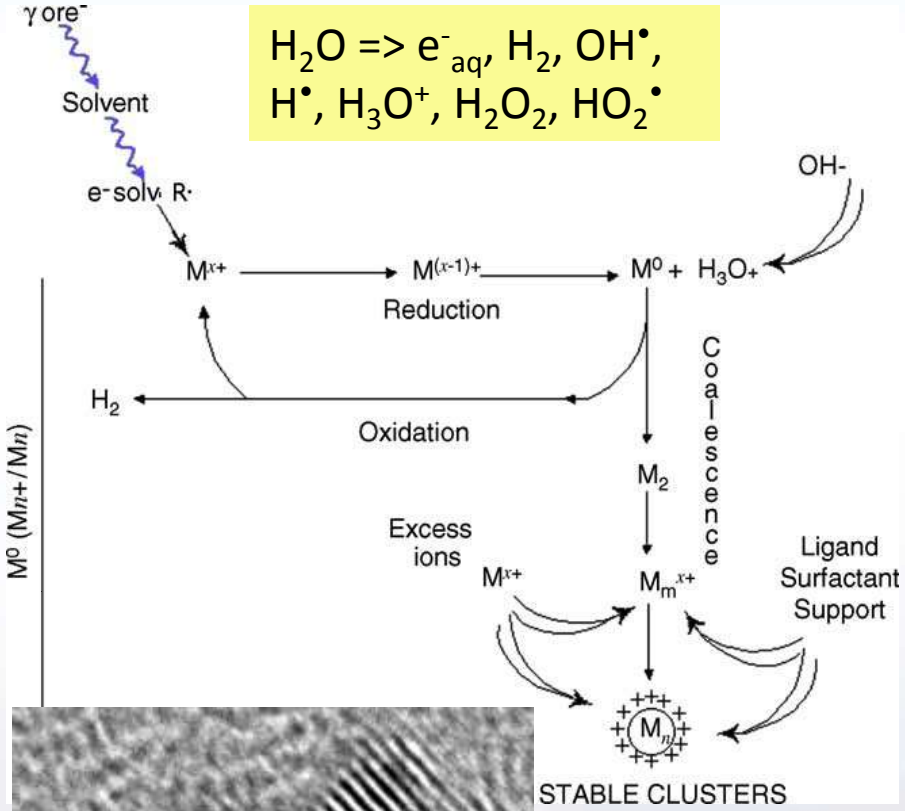


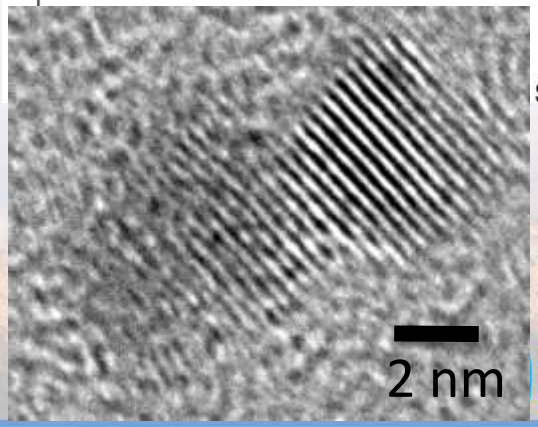
We hope to go a step further and combine it with ion implantation and irradiation environments

Sandia National Laboratories

Low Temperature Synthesis of Lanthanide and Actinide Nanoparticles

- **Fabrication of nuclear fuels at low temperatures allowing for the formation of complex Uranium metal and alloys with minimum volatility of alloy compounds in the process**
- **Potentially permits the recycling of dissolved uranium and lanthanide ions from acidic solutions (used fuel) into metals and alloys**
- **Gamma (g) irradiation is used to reduce uranium and lanthanide ions to metal in solution at room temperature**
- **result in Limits potential release to the environment and/or other ES&H issues**

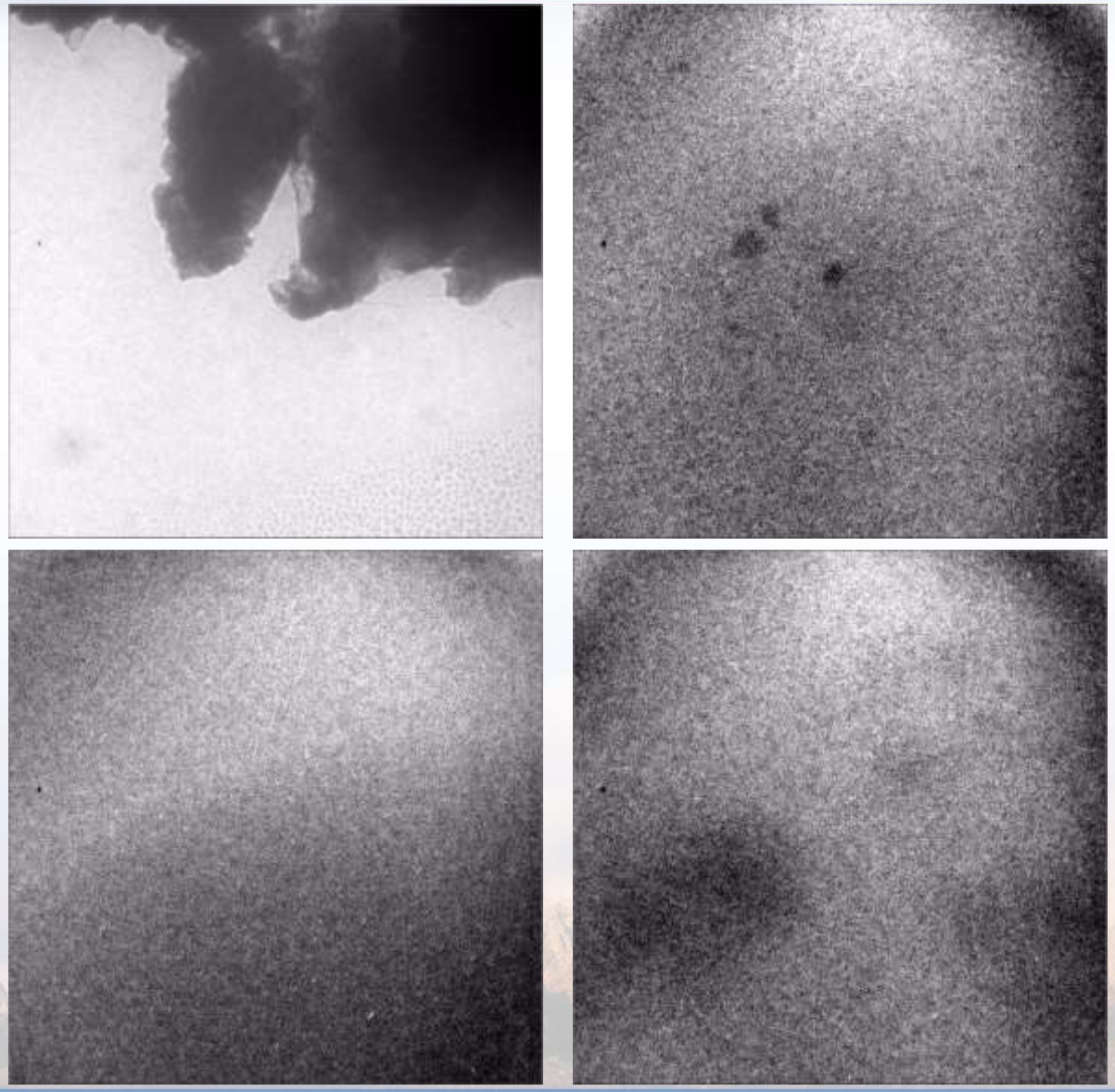




Radiolytic formation of actinide nanoparticles
RT approach for nuclear fuels synthesis reducing defect formations in bulk metals when compared to normal sintering at 1500-1700°C

T.M. Nenoff, *et al*. J. Nuc. Mater. 442 (2013)
T.M. Nenoff, *et al*.
Chem. Mater. 2011, 23,

In situ Observation of the Various Structural Evolutions

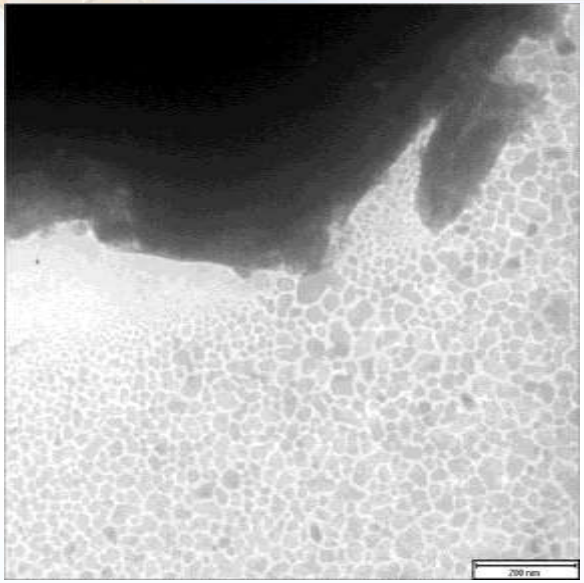


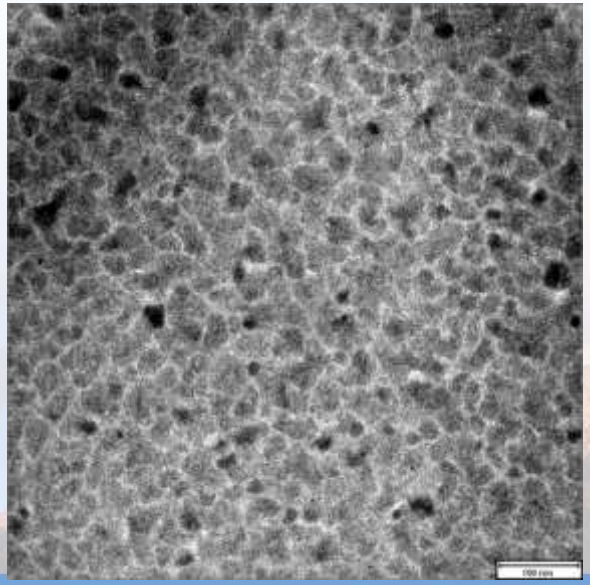
Structural evolution is highly dependent on:

- **the lanthanide salt**
- **local salt conc.**
- **flow rate**
- **proximately to other nanostructures**
- **e^- beam dose**
- **e^- beam dose rate**
- **etc.**

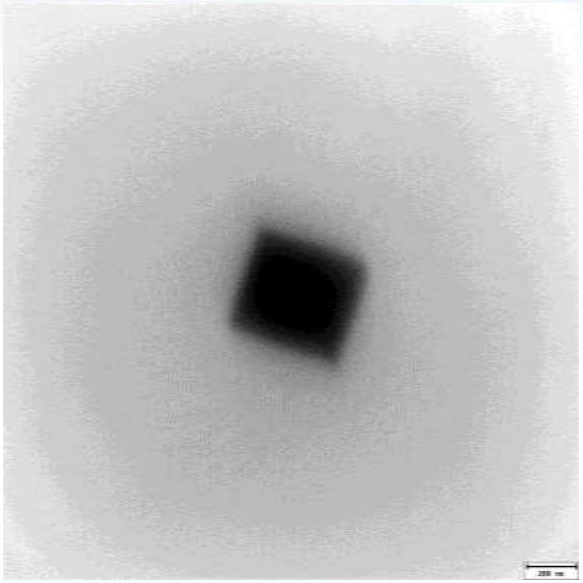
The Resulting Microstructures

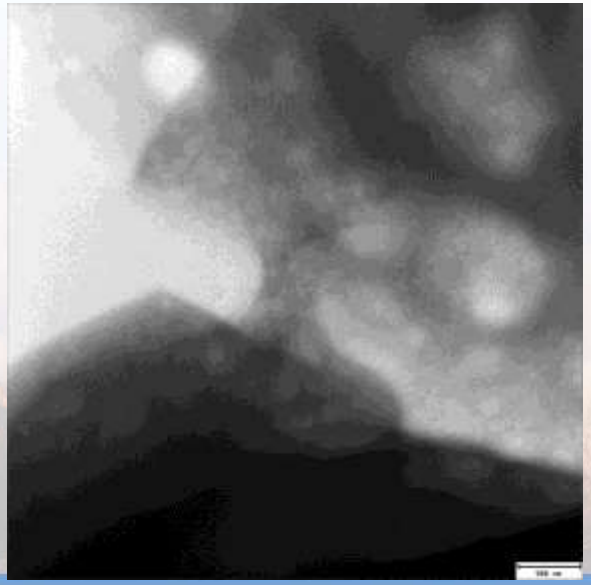
$EuCl_3$ in H_2O



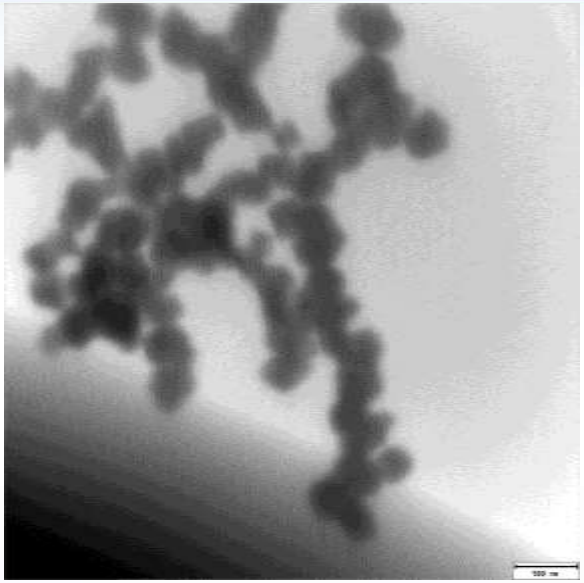


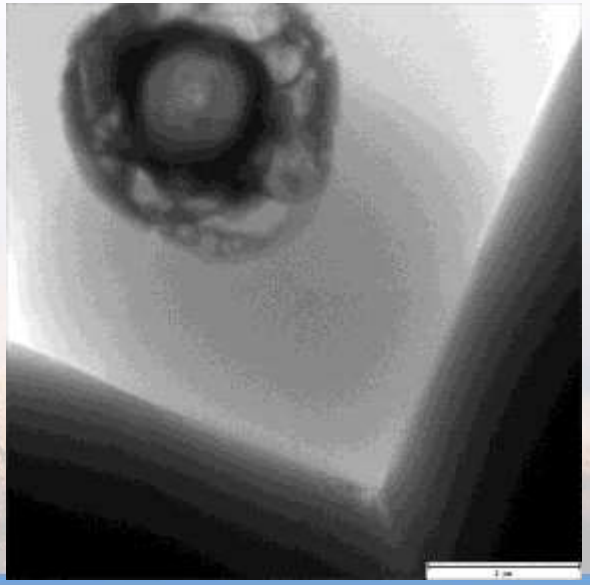
$EuCl_3$ in H_2O





$LaCl_3$ in H_2O





Summary & Future Interests

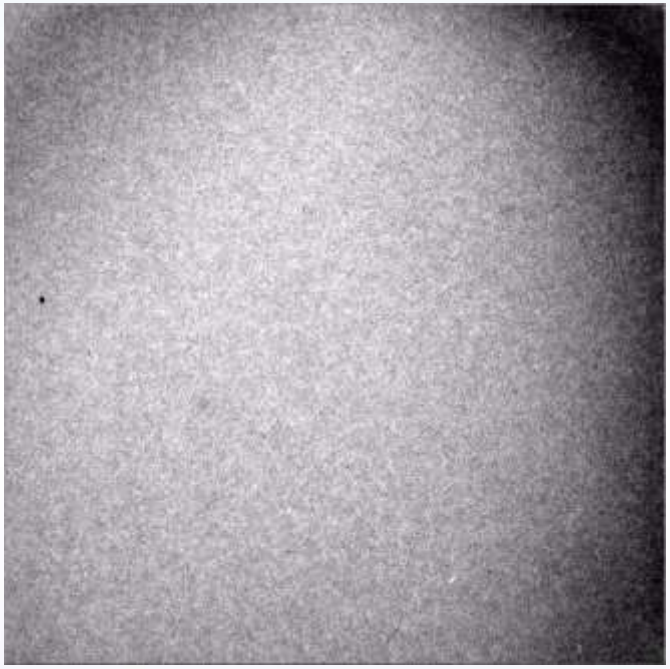
Sandia's I³TEM is one of only two facilities in the US

- ***In situ*** **irradiation from H to Au**
- ***In situ*** **gas implantation**
- **Eight TEM stages with various capabilities (two beta-testing)**

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

It has provided insight into the formation of lanthanide nanoparticles

The I³TEM capability are still being expanded



Sandia's I³TEM although still under development is providing a wealth of interesting initial observations

Collaborators:

D. Buller, J.A. Scott, D. Masiel, N. Li, A. Misra, B.A. Hernandez-Sanchez, T.J. Boyle, J. Sharon, B. L. Boyce, C. Chisholm, A.M. Minor, D. Sasaki, B.G. Clark, D. Gross, J. Kacher, P.J. Cappillino, B.W. Jacobs, L.R. Parent, I. Arslan, M.A. Hekmaty, D.B. Robinson, & I.M. Robertson

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.