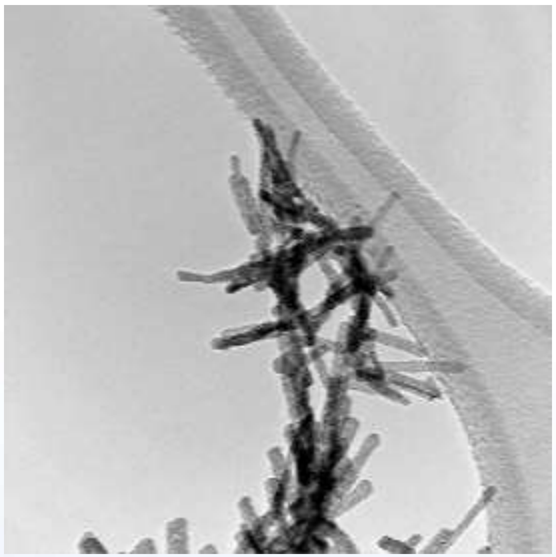
Evaluating the Radiation Stability of Advanced Scintillators and Structural Metals

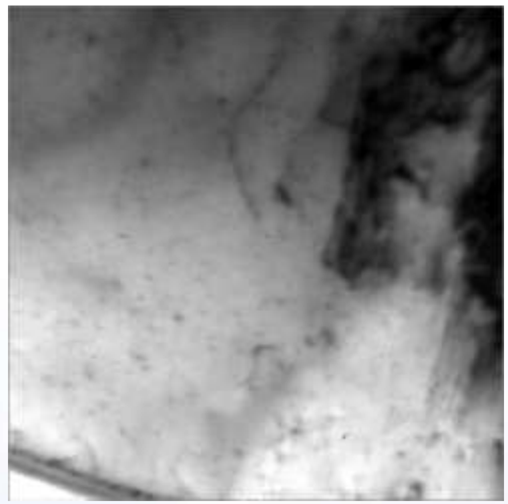
SAND2014-2962C

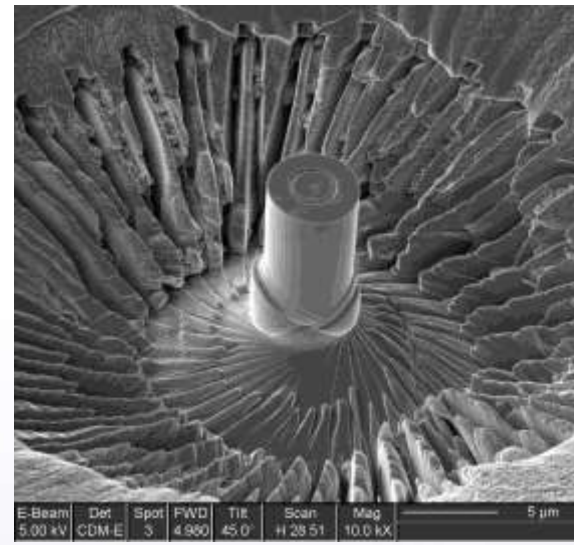
K. Hattar

Sandia National Laboratories

April 11, 2014







Collaborators:

- **IBL: D. Buller, B.G. Clark, B.L. Doyle, S. Hoppe, S. Rajasekhara, J. Villone, G. Vizkelethy**
- **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.**

What is Sandia National Laborator*ies*?

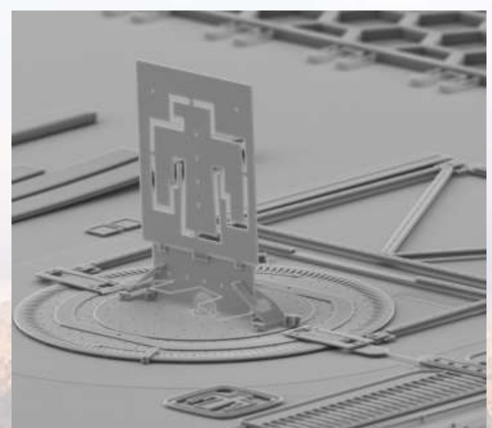
4 Mission Areas

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

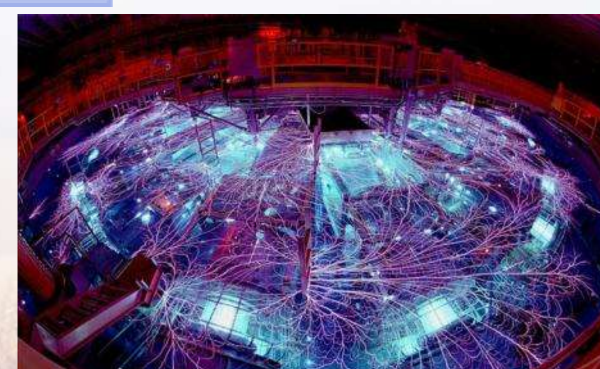
96% of total NW parts

Sled track

Renewable and alternative energy



Clean room invented at SNL in 1963



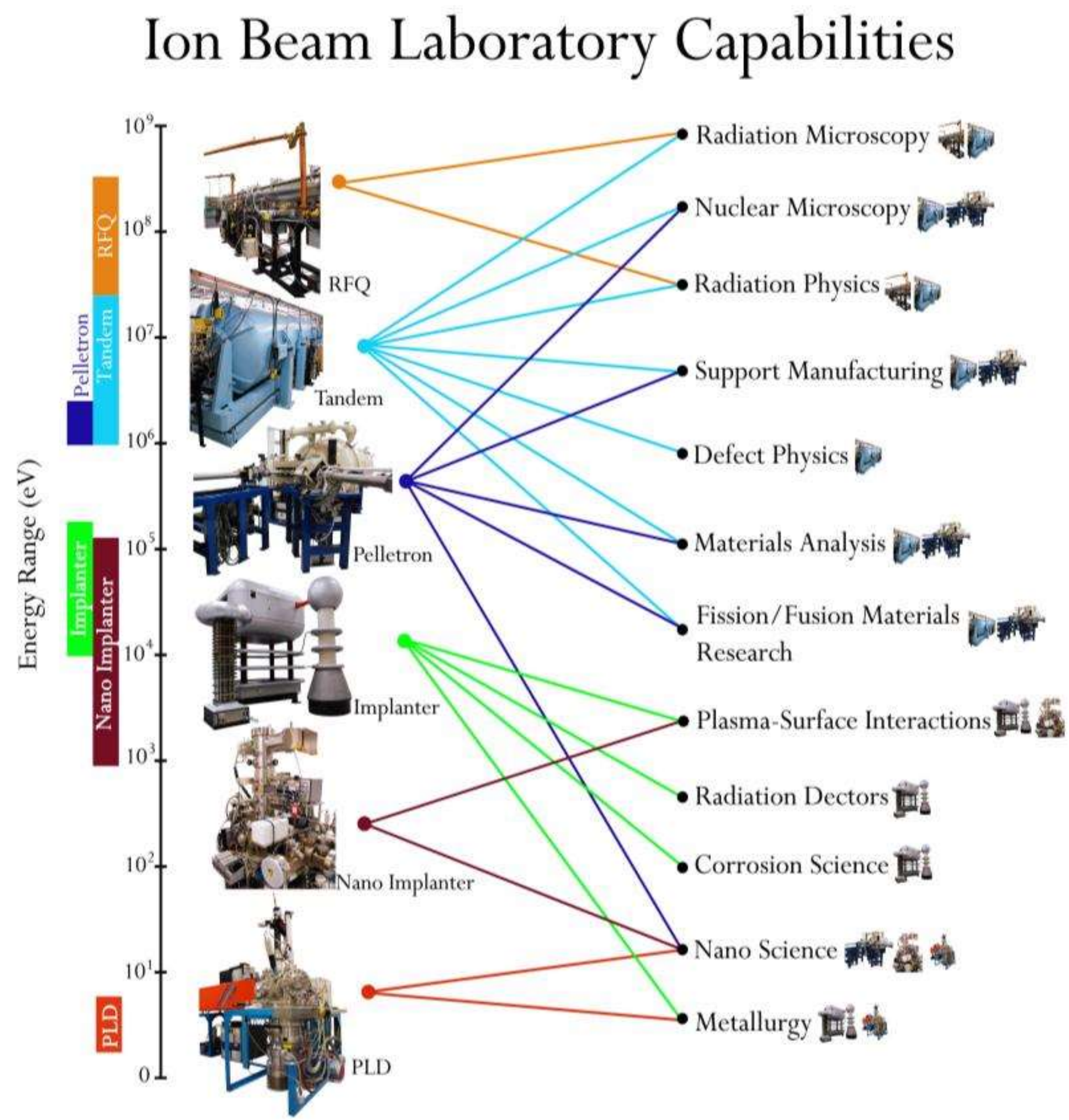
Z machine:
the world's most powerful X-ray source

Sandia's New Ion Beam Lab

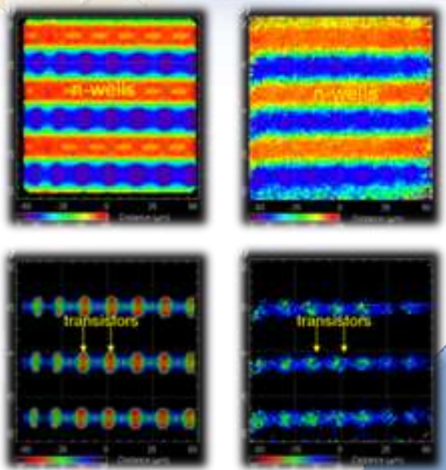
A special 72 wheeled vehicle with independent steering for each pair of wheels was used to move the Tandem accelerator

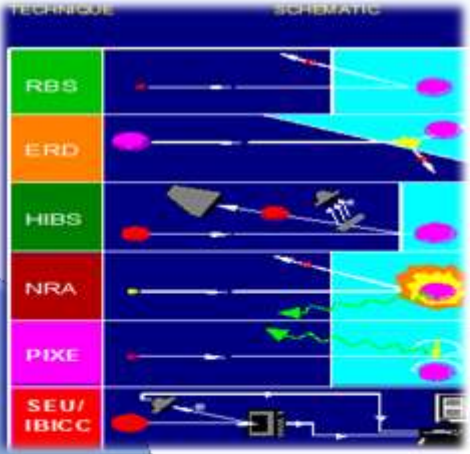
New Facility
laboratory space
1850 m^2
office space
650 m^2
Old Facility:
1300 m^2 total

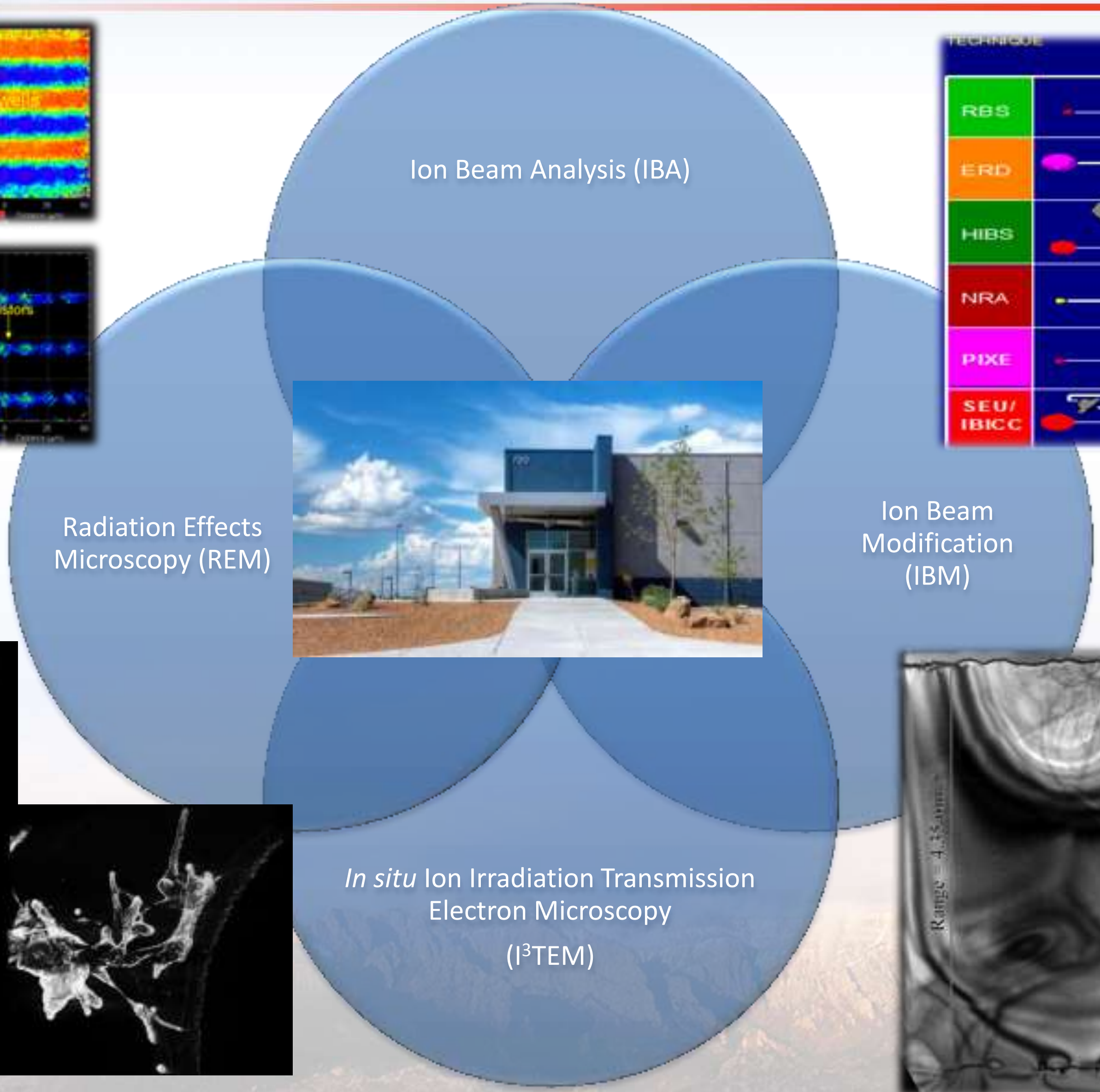
Building: $20M
Equipment: $11M
Total: $40M

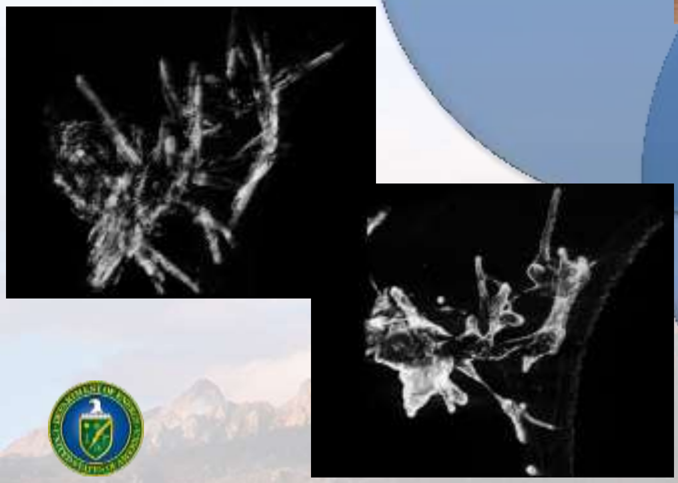


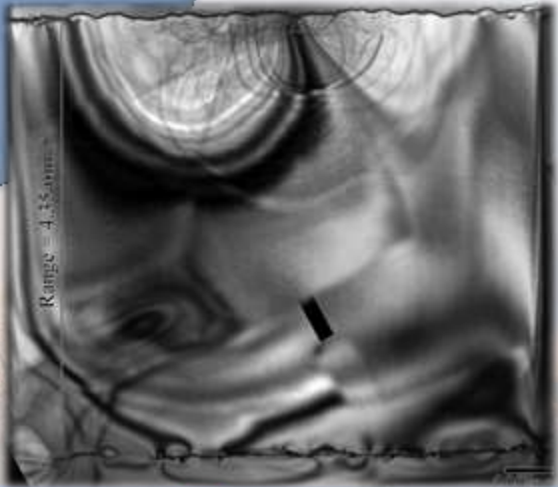
Sandia's Ion Beam Laboratory











Scintillator Applications

National Security

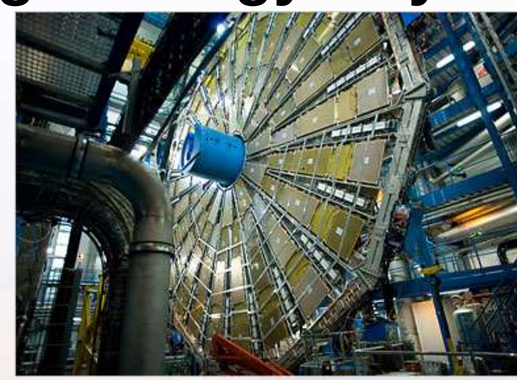
Oil & Mineral

Nuclear Power

Medical Imaging

LEDS

High Energy Physics



Current Issues

1) Long decay times
2) Crystal anisotropy
3) Low energy resolution (low luminosity & poor linearity)
4) Complicated synthesis (single crystal growth)
5) Chemical instability (hygroscopic)

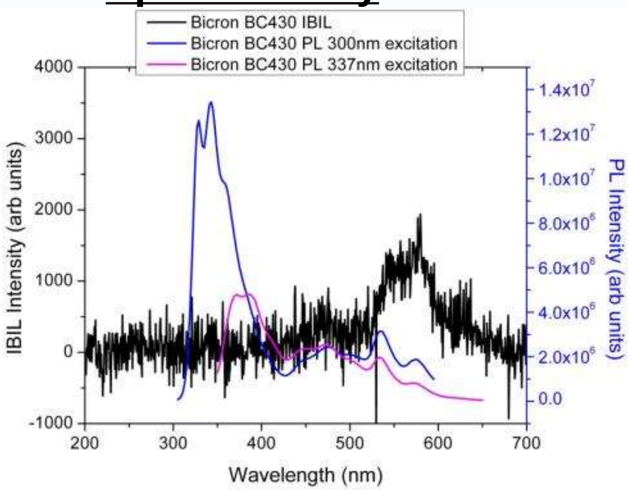
Scintillators with low energy resolution & detection efficiency cannot distinguish radiation type or quantify radiation

Discovery of new scintillating materials

IBL Capabilities for Luminescence Studies

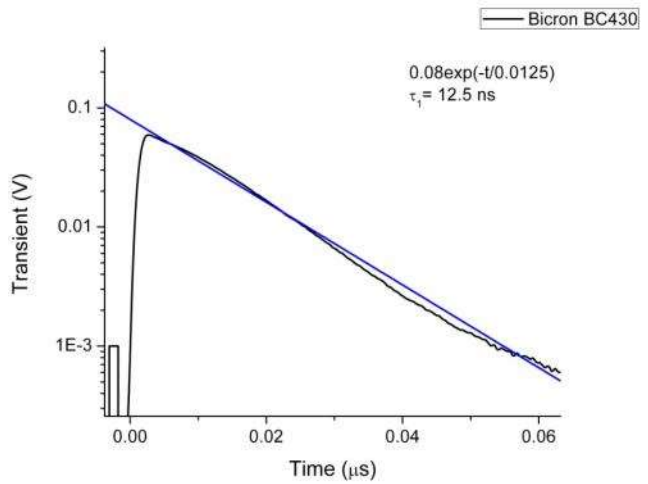
Collaborators: J. Villone and G. Vizkelethy

Spectrometry



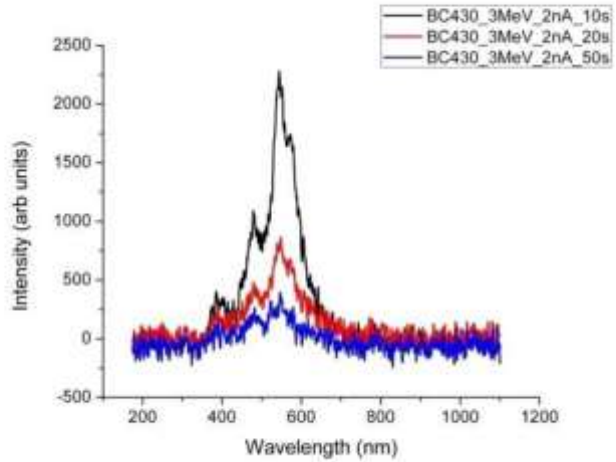
- 3 MeV H^+ beam
- Thin films of samples on PIN diodes
- Hamamatsu PMT run in photon-counting mode
- Light intensity measured as a function of time after ion strike

Decay Time



- 3 MeV H^+ beam used as excitation
- Scintillation light collected as ion beam excites sample
- Light collected with OM-40 microscope or fiber optic mounted close to sample
- Avantes AvaSpec 2048 spectrometer

Radiation Hardness



- Radiation hardness experiments performed with 3 MeV H^+ beam from tandem accelerator
- IBIL Spectra measured constantly as sample exposed to beam
- Overall decrease in emitted light observed due to radiation damage

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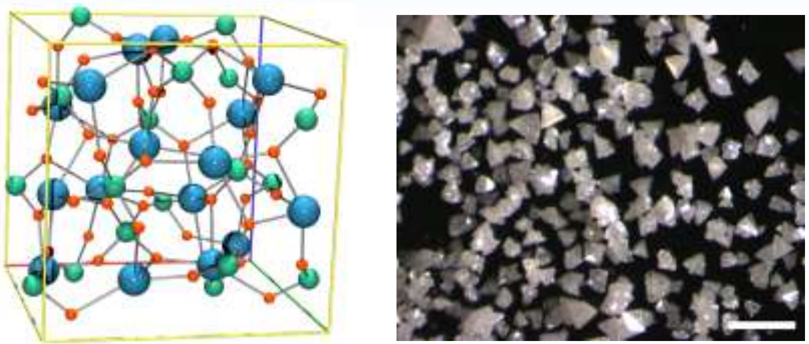
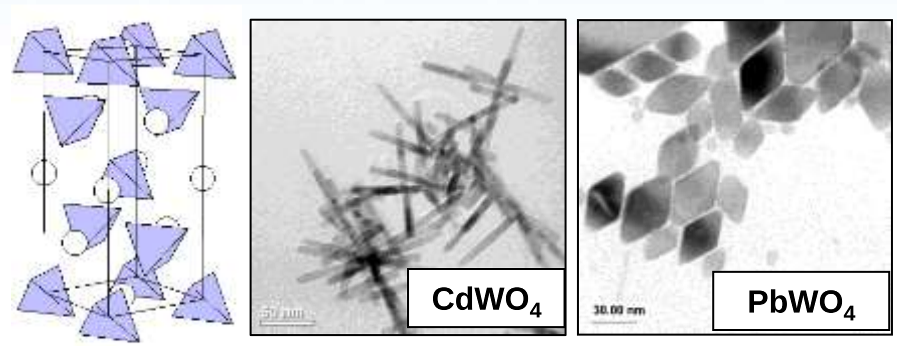
Scintillators Explored

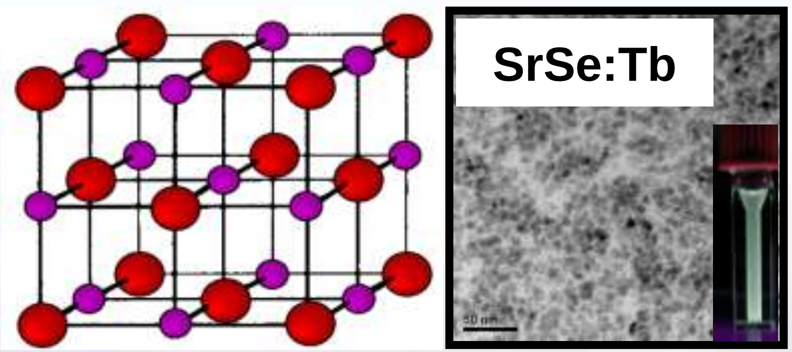
Collaborators: B. Hernandez-Sanchez, S. Hoppe, T.J. Boyle, J. Villone, P. Yang



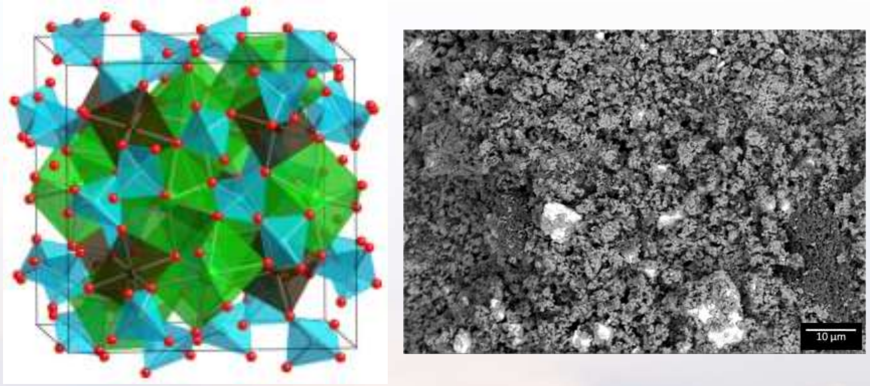
Bismuth Germanium Oxide (BGO)



Metal Tungstates (MWO_4) M = Ca, Cd, Pb



Alkaline Earth Chalcogenides (A^EE)



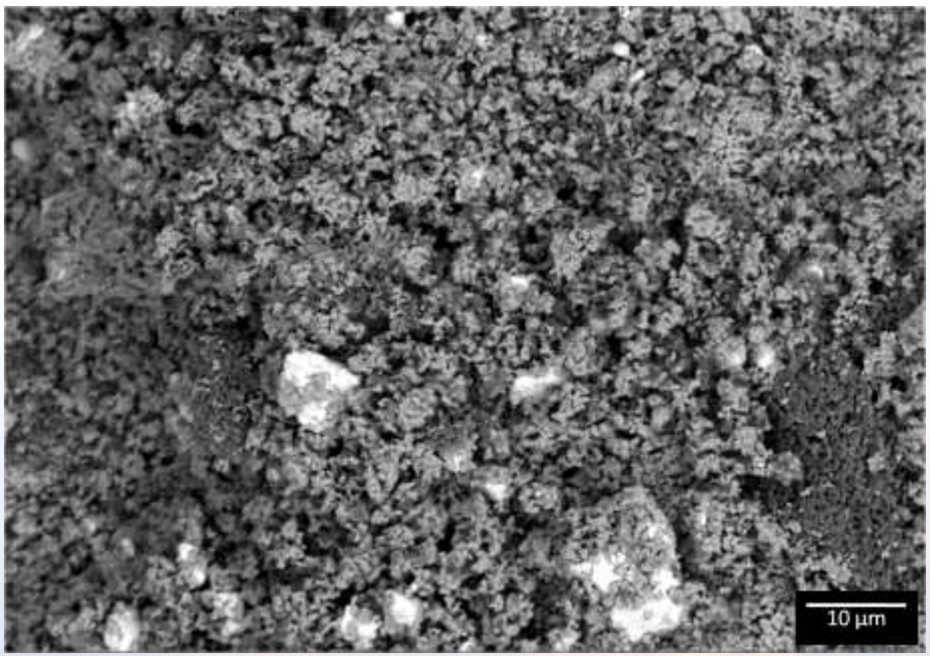
Yttrium Aluminum Garnet (YAG:Ln)

Lutetium Aluminum Garnet (LuAG:Ln)

Solution Precipitation Reaction

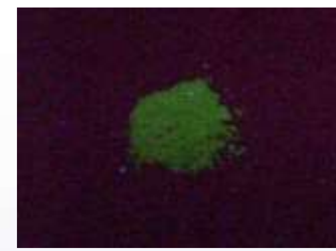
Collaborators: B. Hernandez-Sanchez, S. Hoppe, T.J. Boyle, J. Villone, P. Yang

$$3\text{-}X\ Y(NO_3)_3 + 5\ Al(NO_3)_3 \cdot 9H_2O + X Ln(NO_3)_3 \cdot 6H_2O + CH_4N_2O \xrightarrow[80°C \atop 1500°C]{NH_4OH \atop \Delta} Y_{3\text{-}x}Al_5O_{12}\text{: } XLn$$

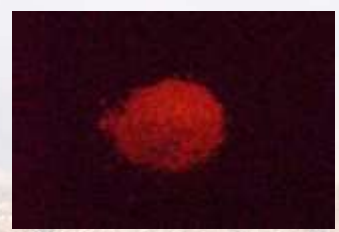


YAG: Ce

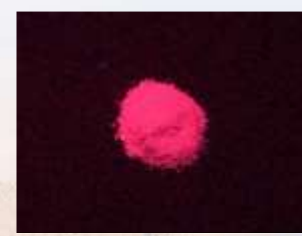
LuAG: Ce



YAG: Lu



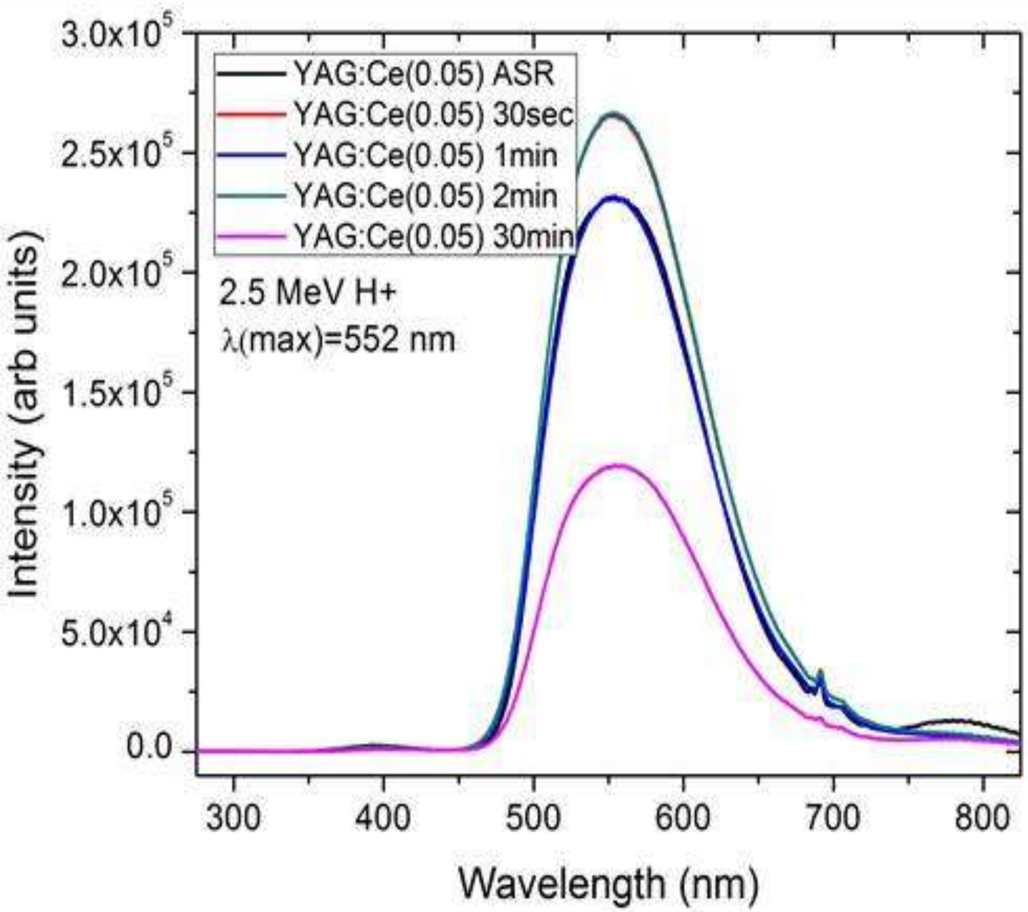
YAG: Pr

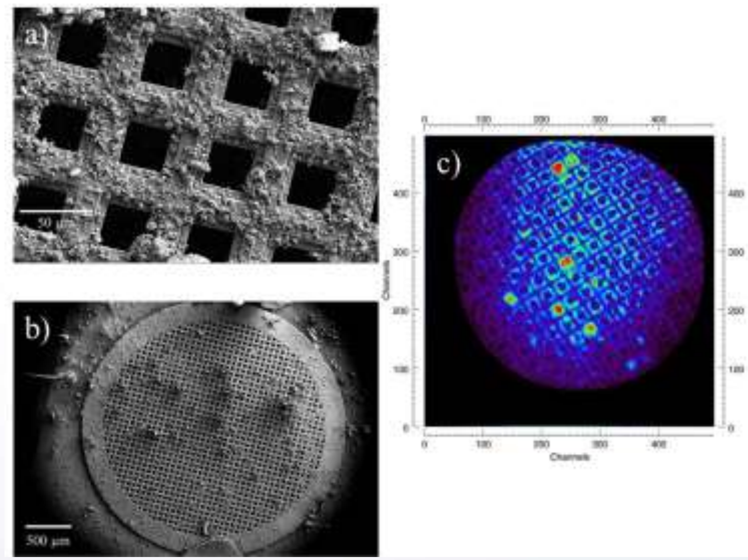


K.Y. Jung, Y.C. Kang Physica B 405 (2010) 1615-1618

Radiation Tolerance of YAG:Ce for 30 min Exposure

Collaborators: B. Hernandez-Sanchez, S. Hoppe, T.J. Boyle, J. Villone, P. Yang



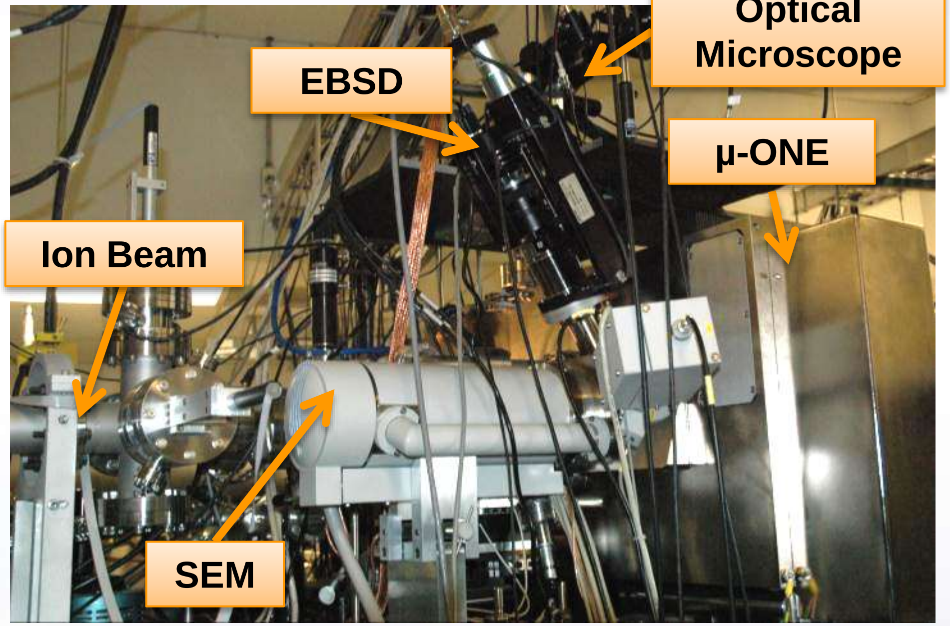


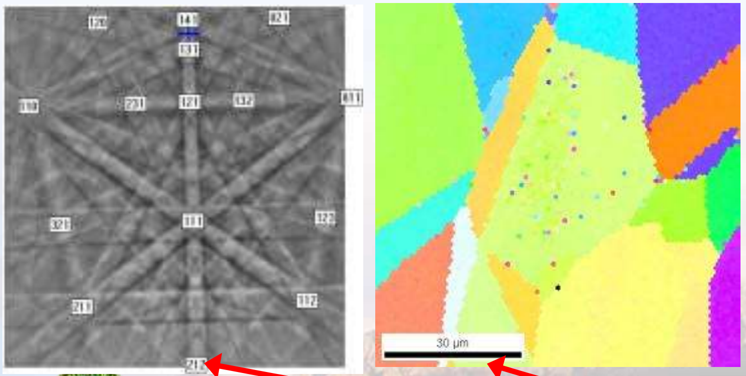
Micro-ONE Capabilities

Micro-ONE = Micrometer resolution Optical, Nuclear, and Electron Microscope

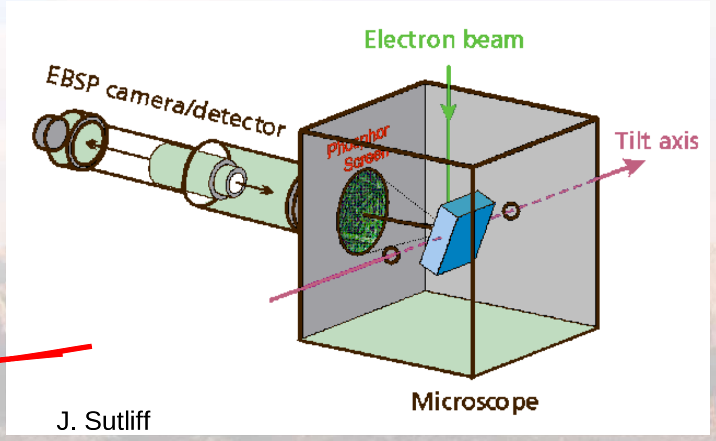
<u>Micro-ONE capabilities</u>

- Beam size: < 1µm (~ 0.5 µm)
- Current: single ions to 10,000 ions/s
- Ions: H, He, heavy ions
- Energy: (q+1)*6 MV for heavy ions
- Scan size: ~ 100x100 µm^2
- Stage position with 50 nm resolution
- Fast blanking capabilities
- Navigation based on GDS II files
- IBIC and TRIBIC capabilities
- EBSD mapping





First EBSD Pattern and Map obtained with this system

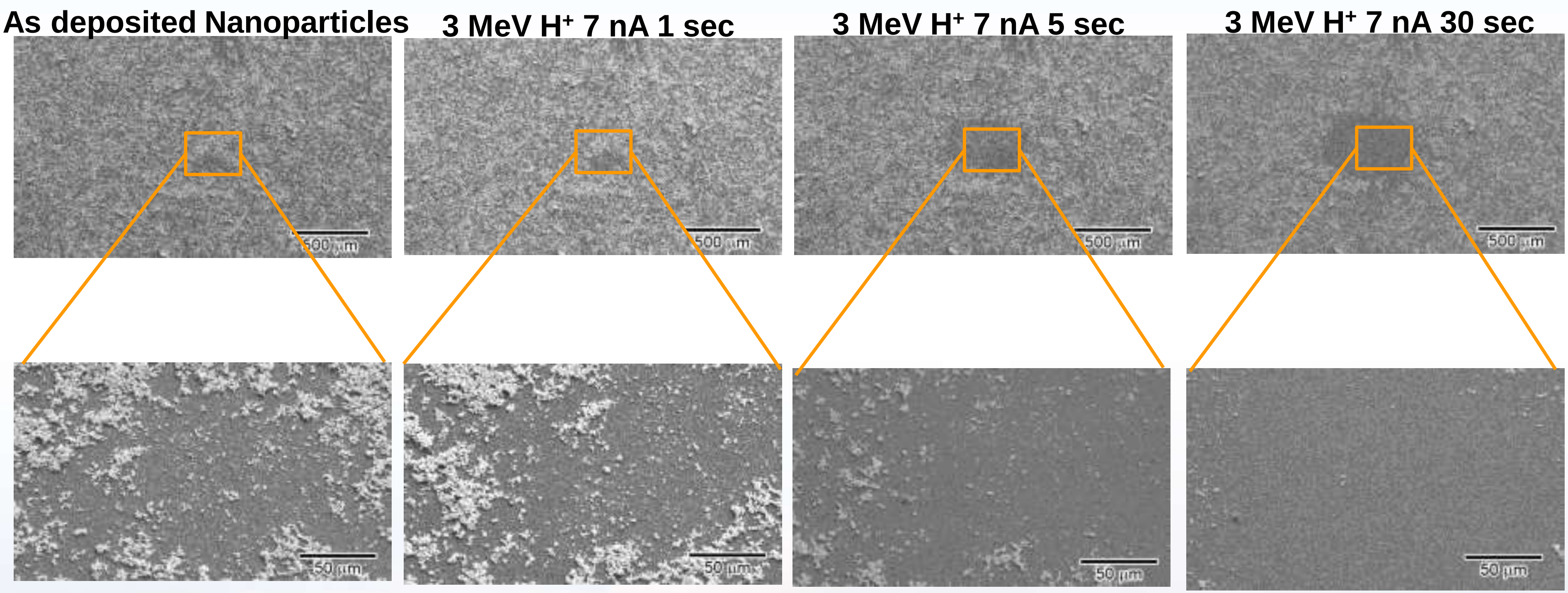


Allows parallel imaging of changes in microstructure: grain size, phase transformations.

Results - Nanoscintillators

Collaborators: B. Hernandez-Sanchez, S. Hoppe, T.J. Boyle, J. Villone, P. Yang

Nearly In-Situ SEM Ion Irradiation of Nanoscintillators



Want to understand if microstructure is affected by irradiation and how that influences optical properties

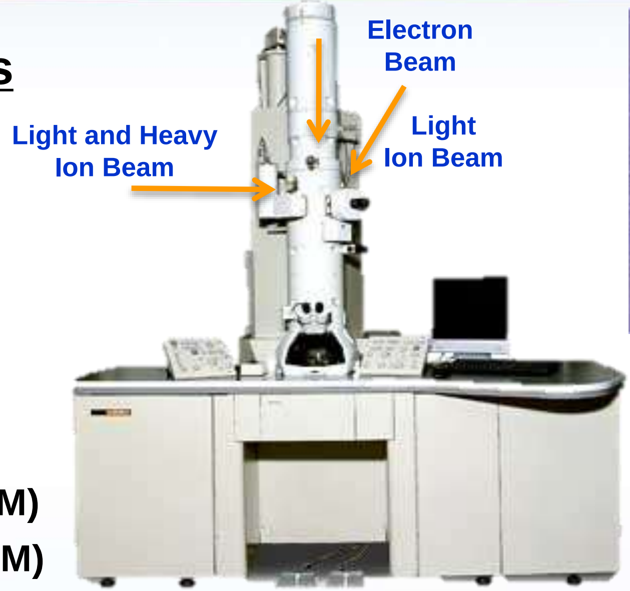
Drop cast films of $PbWO_4$ nanoscintillators irradiated with 3 MeV proton beam, then imaged with SEM

Material being ablated off of the surface – need better technique to study microstructural changes

In situ Ion Irradiation TEM

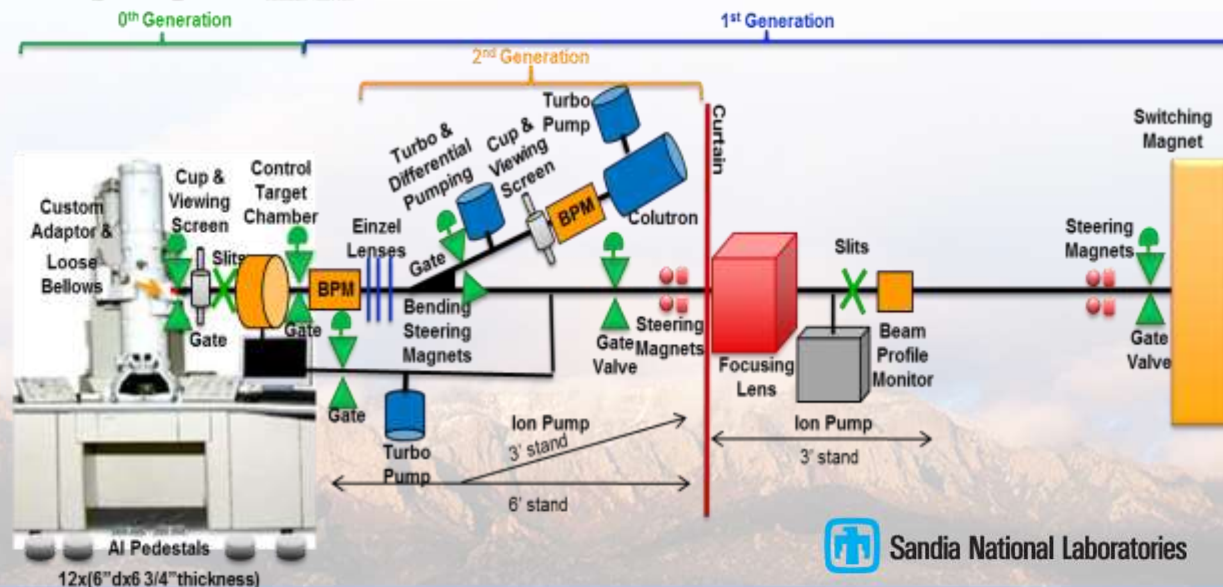
Proposed Capabilities

- 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
- *In situ* cooling stage
- *In situ* electrical bias stage
- *In situ* straining stage



TVIPS

Hummingbird



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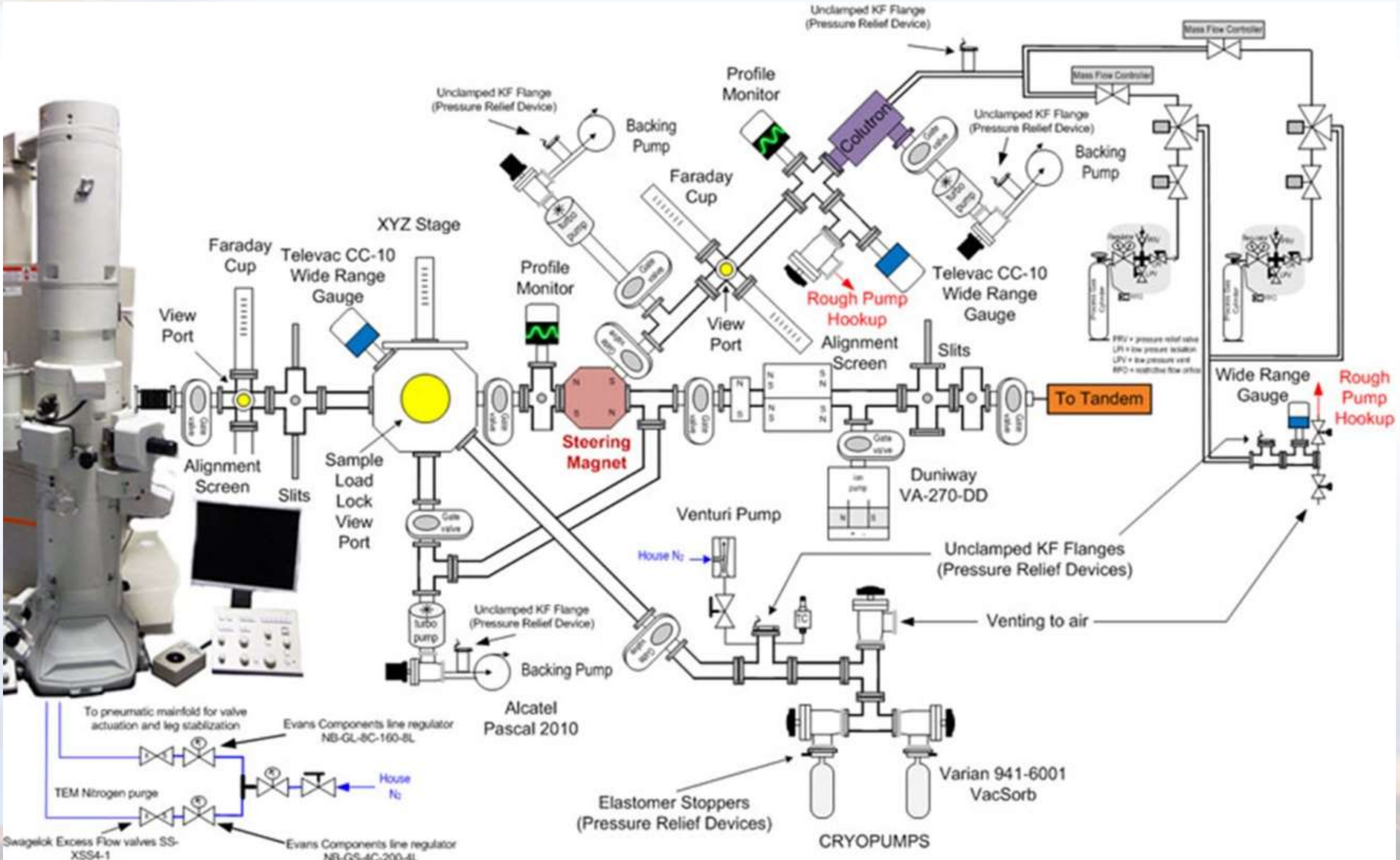
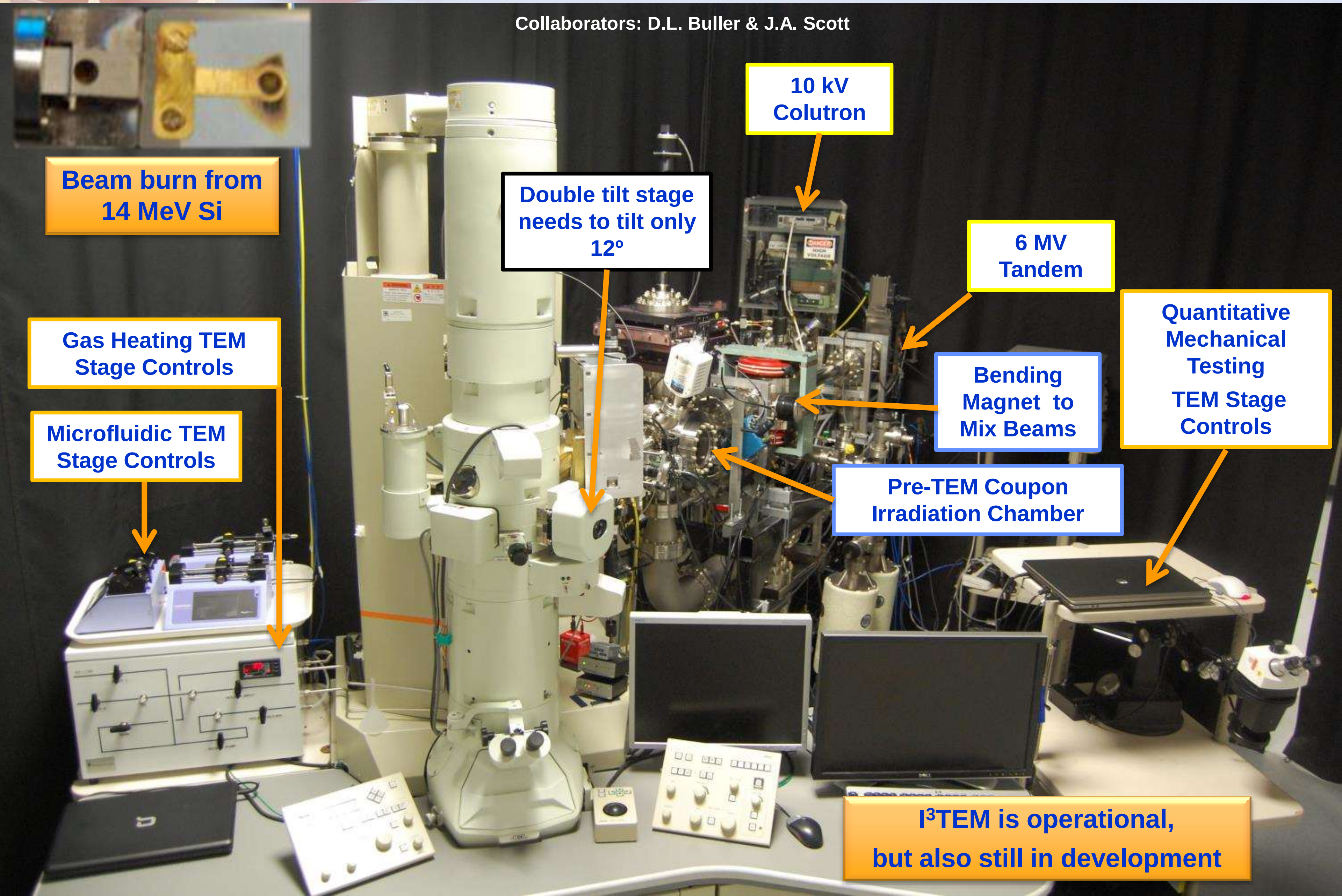
Schematic of the *In situ* TEM Beamline



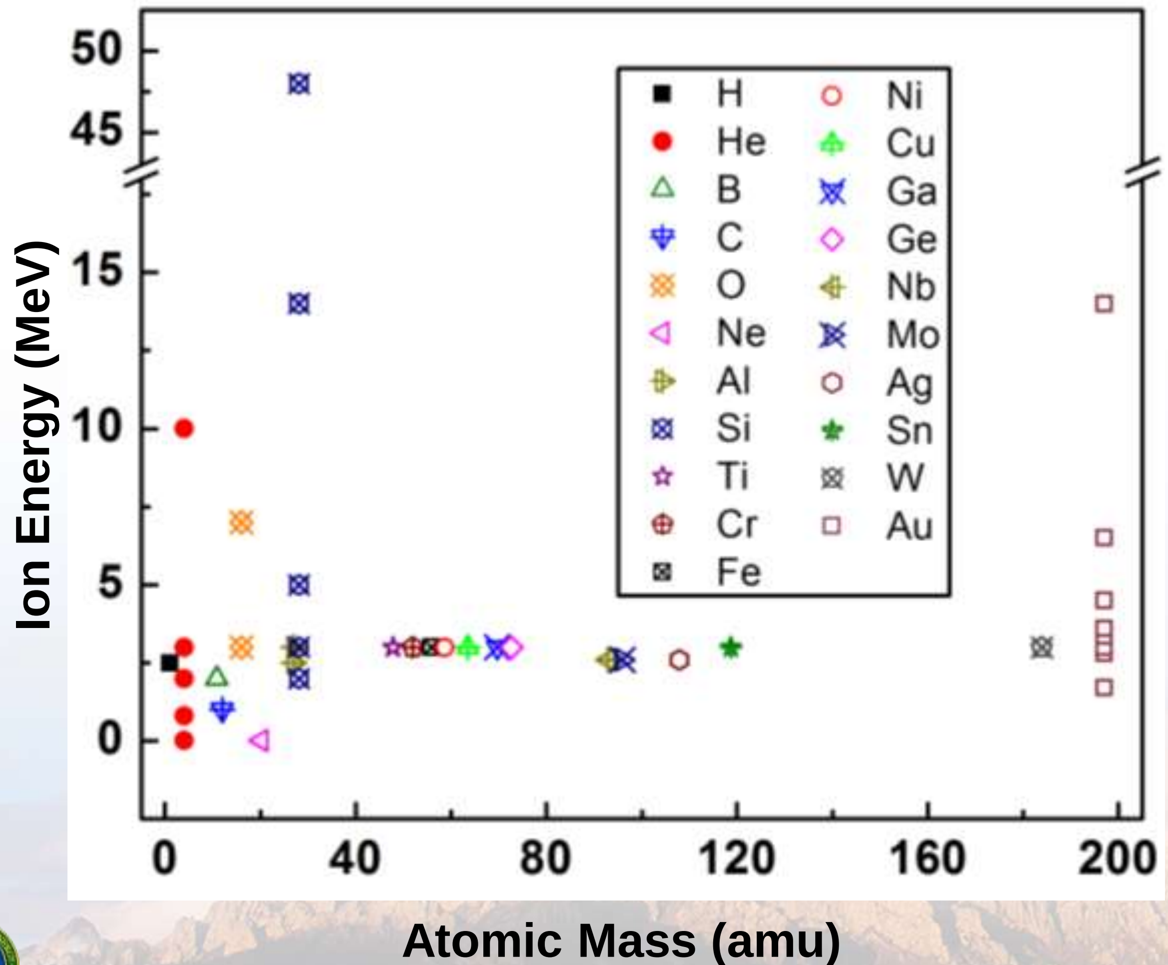
Current Status of the *In situ* TEM Beamline

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



Ion Species Recently Attempted

Collaborators: D. Buller, D.C. Bufford, M. Steckbeck



Colutron ion beams possible:

0.8-20 keV energies

Any gas species

Colutron ion beams to date:

8, 10, & 20 keV energies

He, D_2, & Ne

Tandem ion beams possible:

0.8-88 MeV energies

Any sputtered or alpha sources

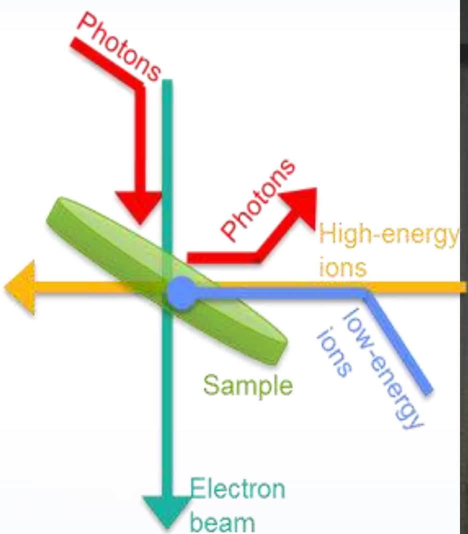
Tandem ion beams to date:

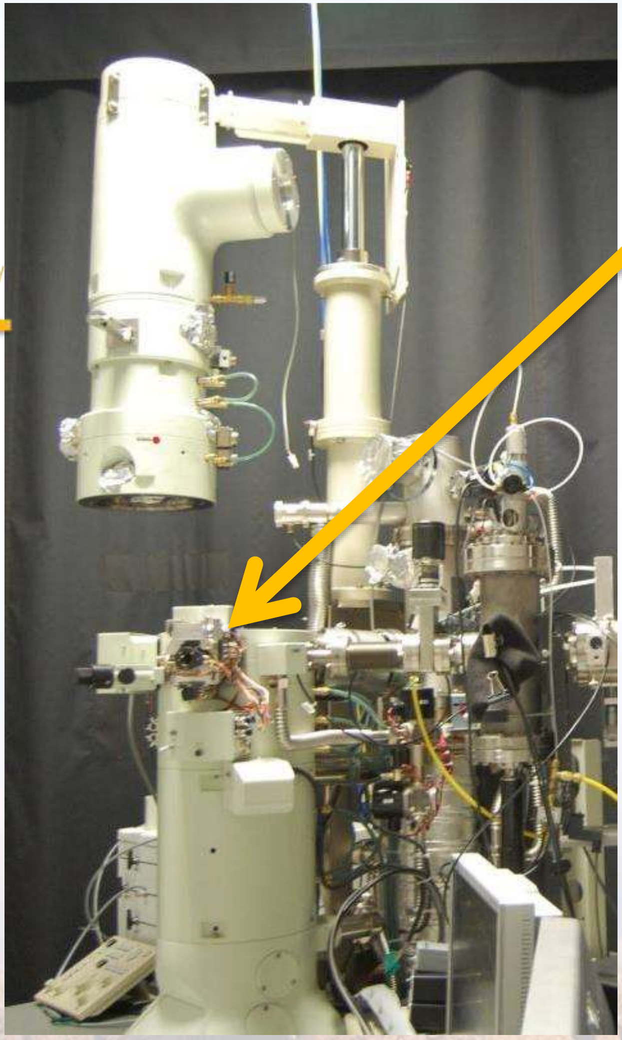
0.8-48 MeV energies

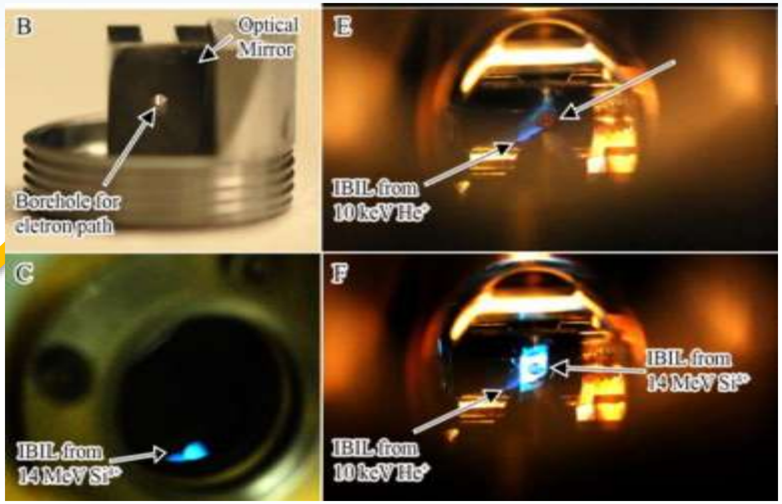
Large range

In situ TEM Luminescence

Collaborators: D. Masiel and C. Chisholm







Two optical port were added to the I^3TEM already containing a electron beam and two ion beams, which permits *in situ* TEM luminescence studies

Optical Pathway in an I^3TEM

- **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.**

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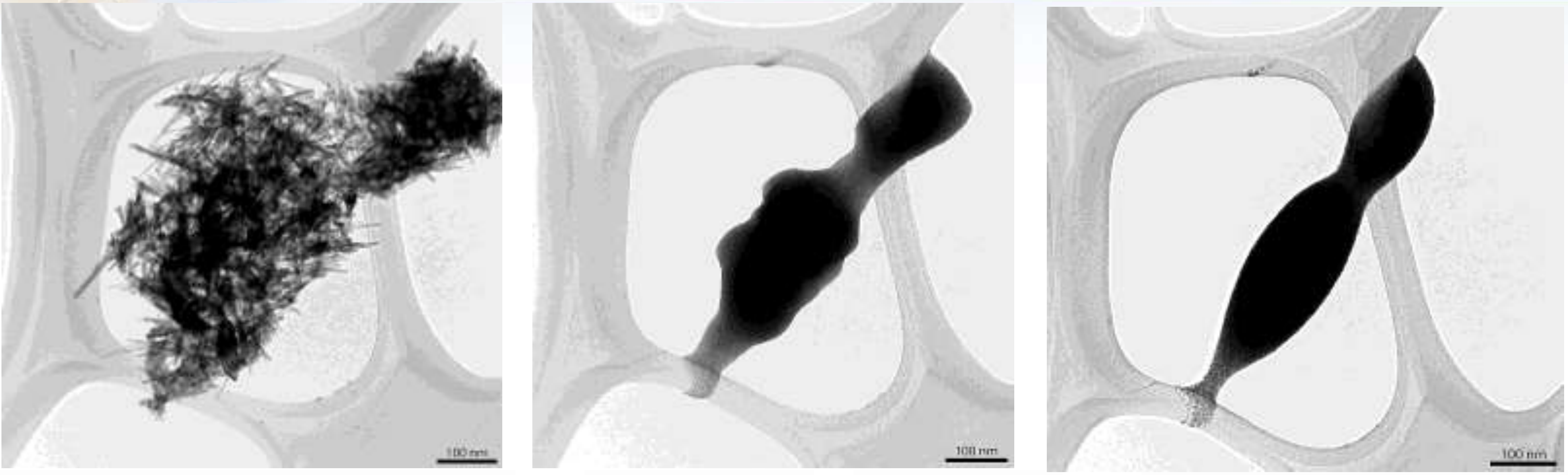
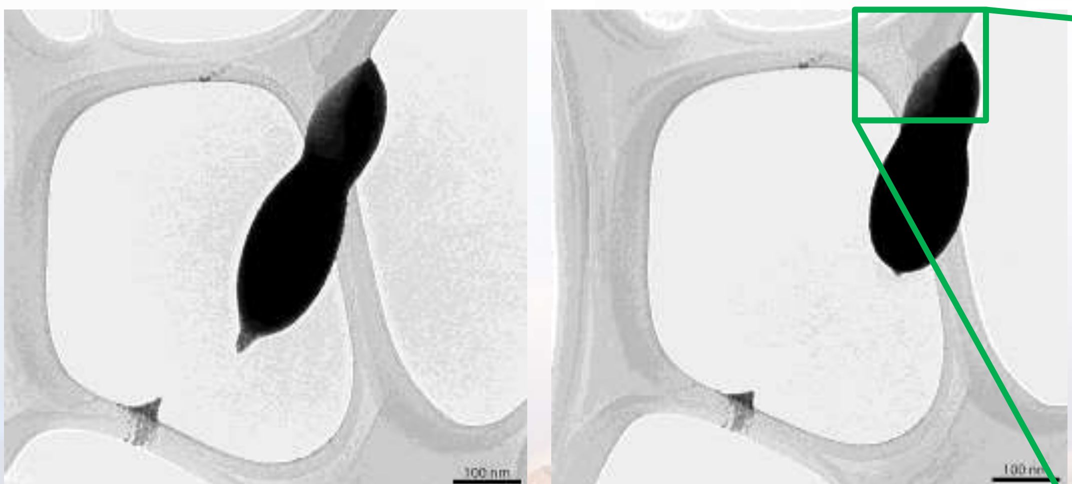
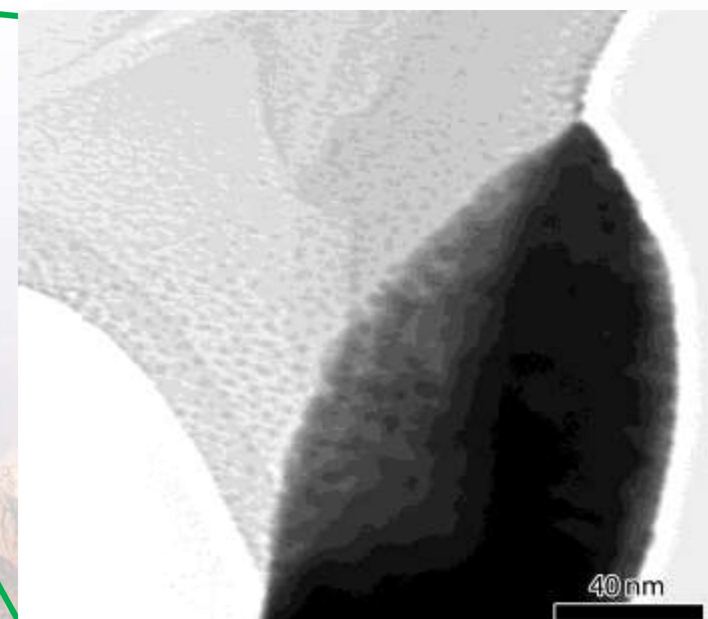
$CdWO_4$ irradiated with 50 nA of 3 MeV Cu^{3+}

Collaborators: B. Hernandez-Sanchez, S.H. Pratt, D.C. Bufford, & T.J. Boyle



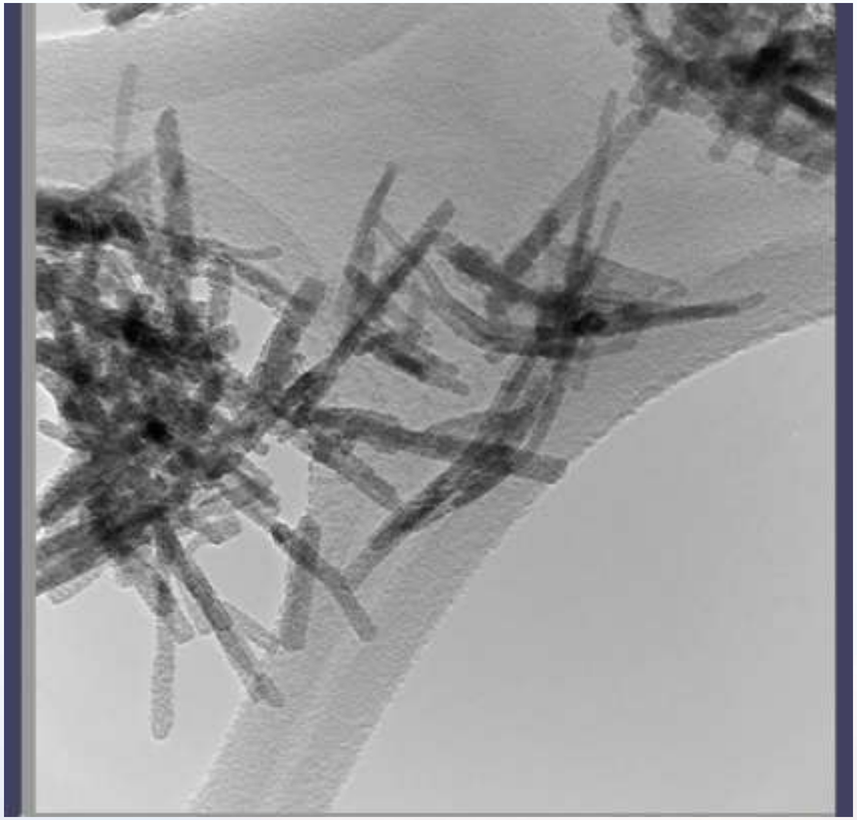


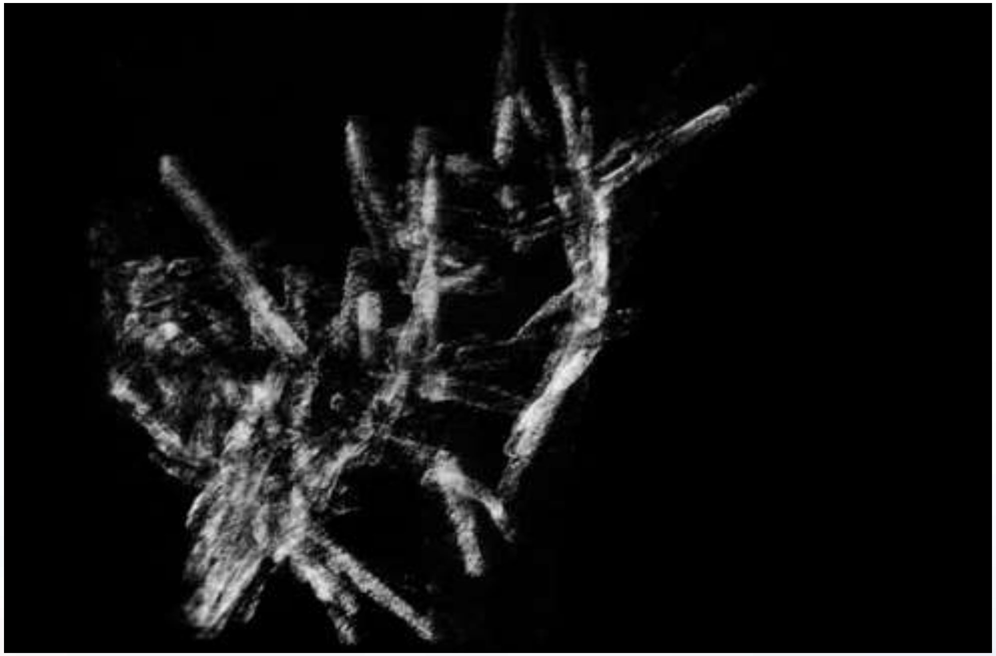


Over 1 hr, nanorods broke into small pieces and sputtered onto nearby lace.

Tomographic Reconstruction of $CdWO_4$

Collaborators: B. Hernandez-Sanchez, S.H. Pratt, D.C. Bufford, & T.J. Boyle



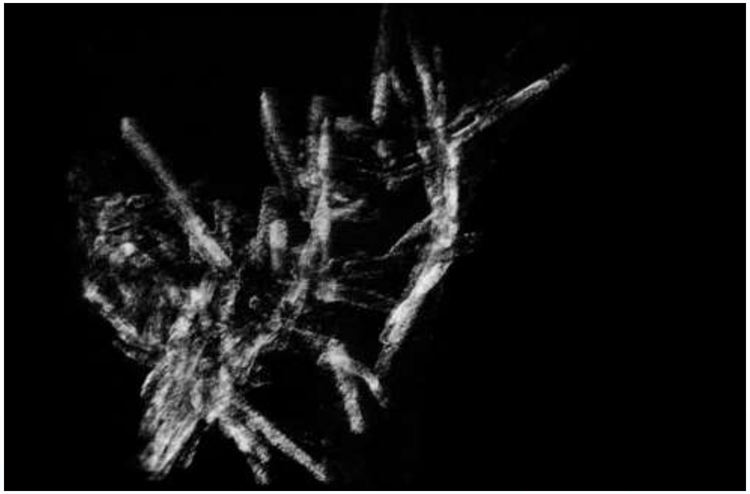


Recent advancements in TEM control and reconstruction software permit collection and production of 3D model of the "transmission" micrograph.

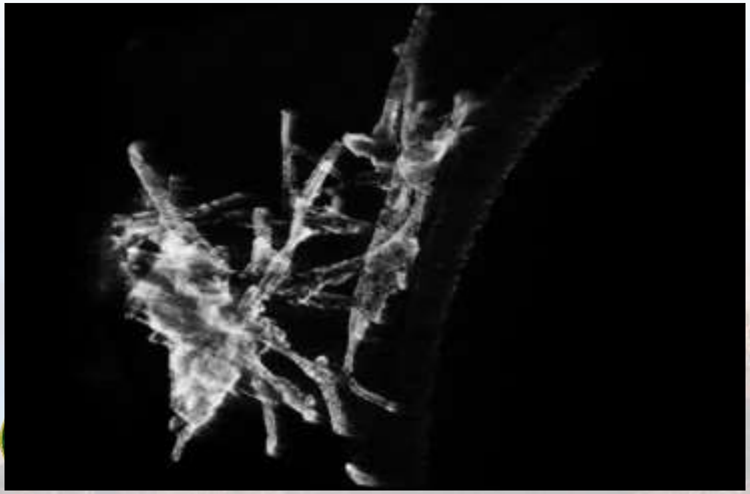
Tomography of $CdWO_4$ irradiated with 30 nA of 3 MeV Cu^{3+}

Collaborators: B. Hernandez-Sanchez, S.H. Pratt, D.C. Bufford, & T.J. Boyle

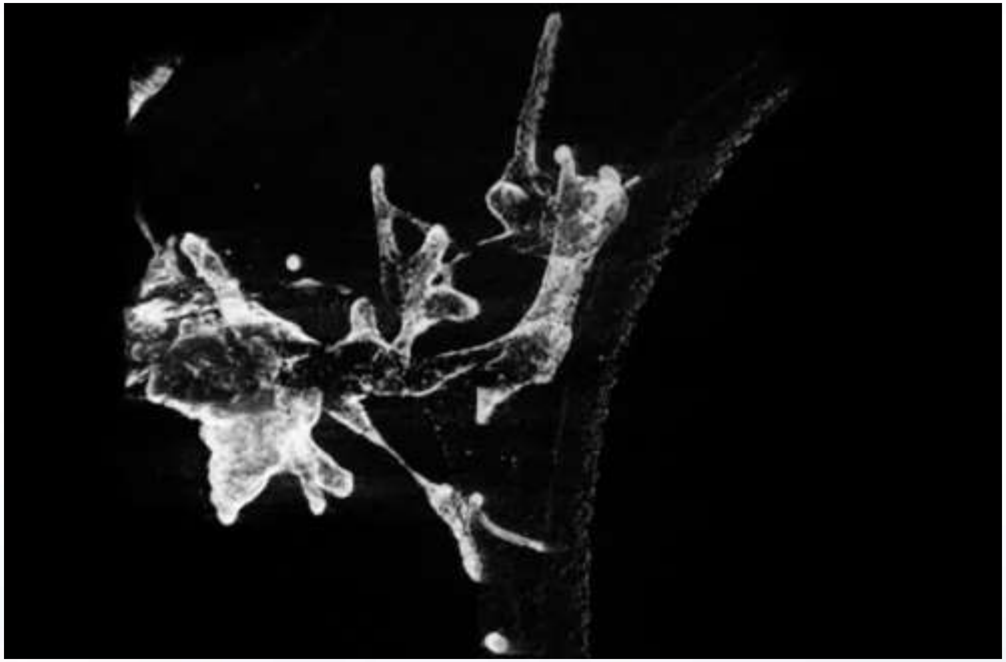
Unirradiated



5 minutes



30 minutes

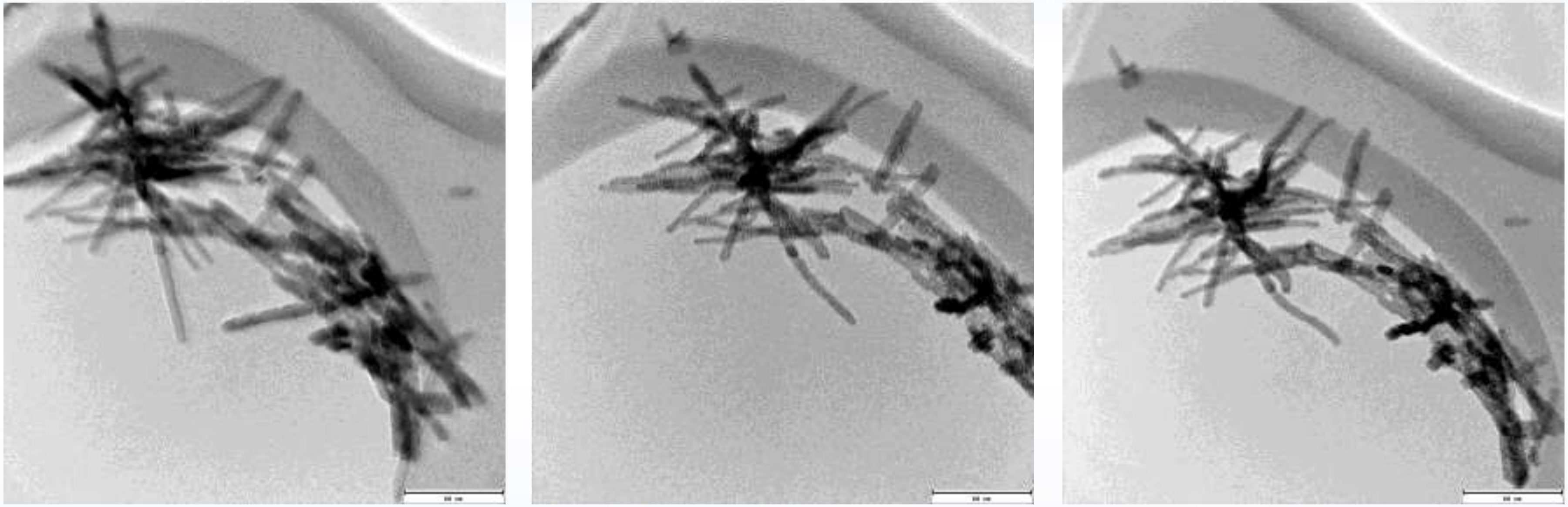


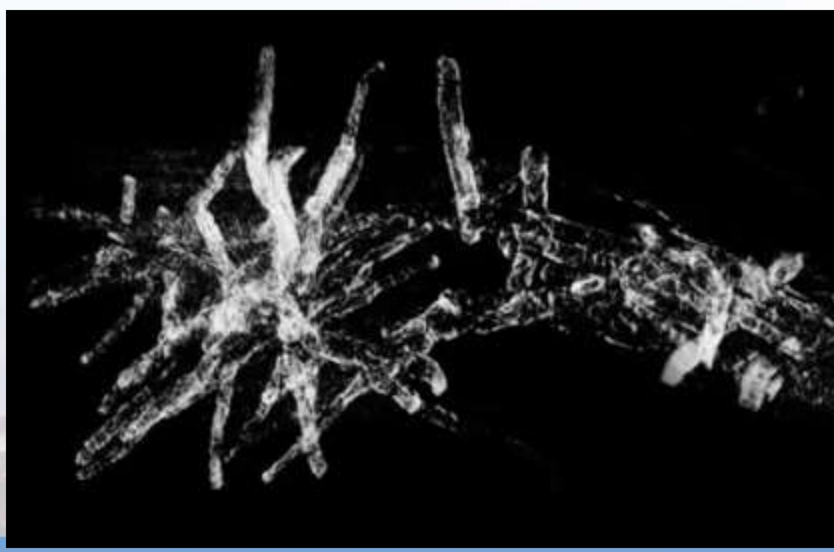
Tilt series were collected after each dose of irradiation resulting in 4D tomography with 3D reconstructions showing radiation damage over time.

Current work: *In situ* Proton Irradiation as First Order Simulation of Neutrons

Collaborators: B. Hernandez-Sanchez, S.H. Pratt, D.C. Bufford, & T.J. Boyle

Unirradiated | 15 minutes | 60 minutes





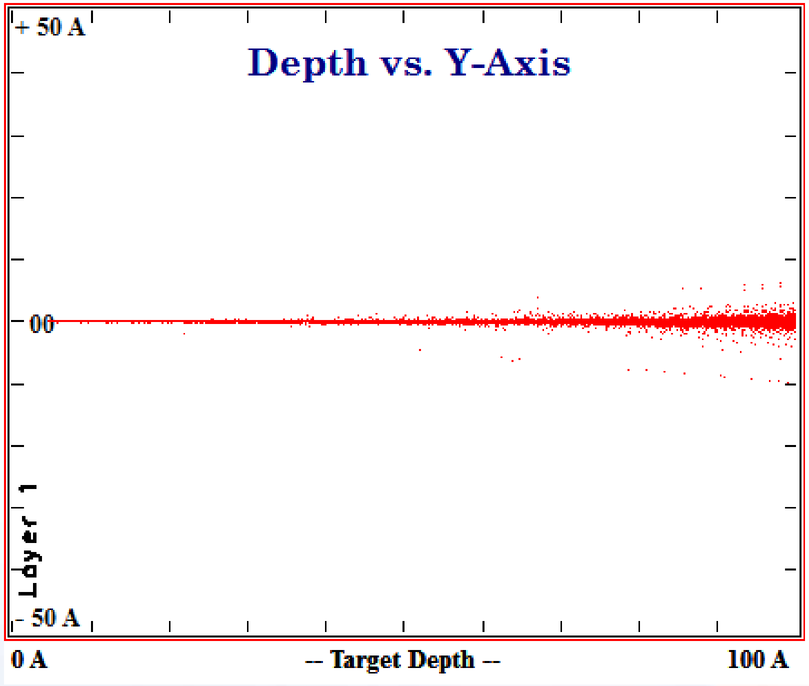
160 nA of 2.5 MeV H^+ used to simulate neutron radiation shows less change. Results suggest good radiation hardness for tungstate nanorod-composite scintillators.

Comparison of Proton and Copper Irradiation

Theoretical Comparison

Collaborators: B. Hernandez-Sanchez, S. Hoppe, T.J. Boyle, J. Villone, P. Yang

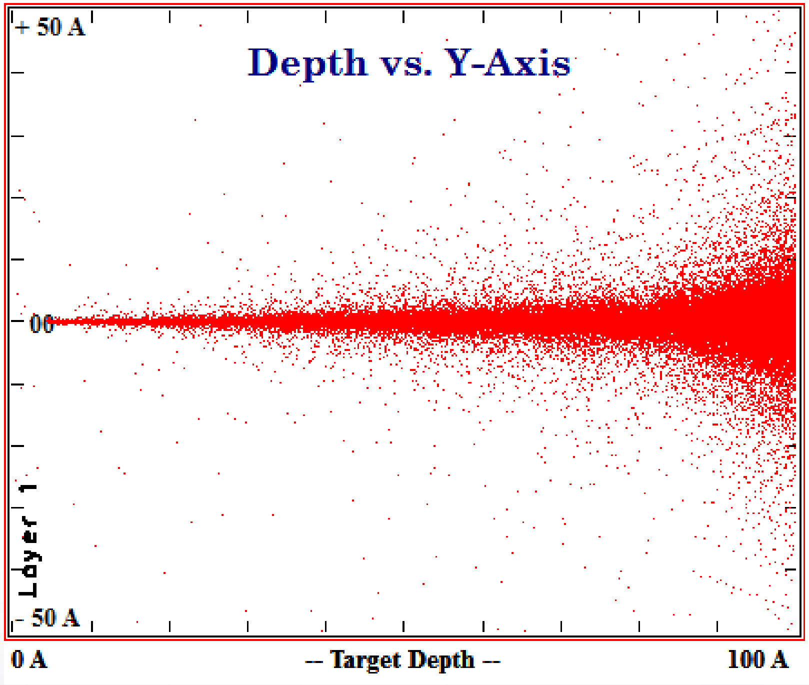
Displacement Damage from 2.5 MeV H



SRIM simulation of the Frenkel pairs created by the ion irradiation conditions used in 10 nm thick $CdWO_4$ noparticle.

Note the large number of pairs over the same simulation set, as well as the two order magnitude dE/dx elec. and four dE/dx Nuc.

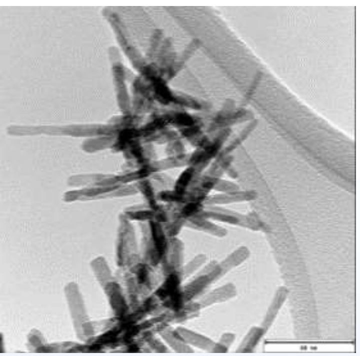
Displacement Damage from 3 MeV Cu



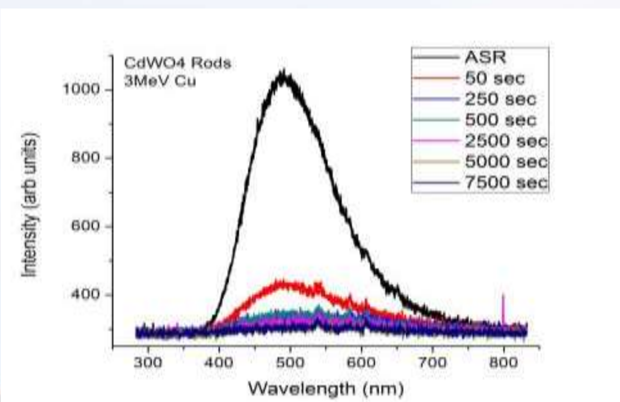
Sample	Density (g/cm3)	Species	Energy (MeV)	Current (nA)	**dE/dx Elec.**	**dE/dx Nuc**	Proj. Range (um)	Long. Straggle (um)	Lat. Struggle (um)
CdWO4	7.9	H	2.5	~100-200	**5.97E-02**	**3.80E-05**	33.62	2.2	3.59
PbWO4	8.235	H	2.5	~100-200	**5.18E-02**	**3.39E-05**	37.22	2.8	4.63
CdWO4	7.9	Cu	3	~10-30	**2.19E+00**	**5.31E-01**	1.25	0.4581	0.4096
PbWO4	8.235	Cu	3	~10-30	**2.67E+00**	**6.11E-01**	1.16	0.3632	0.3328

The Stopping and Range of Ions in Matter (SRIM) is a Monte Carlo-based simulation of the ion beam interaction with an amorphous material.

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



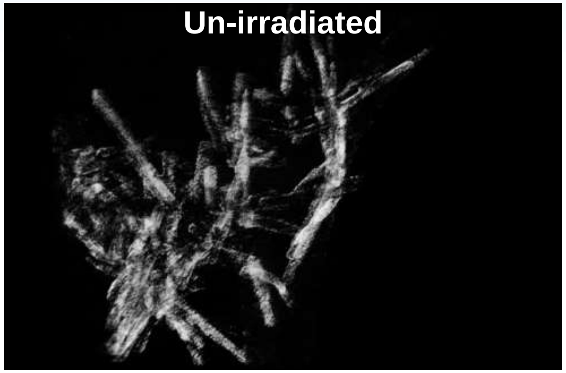
High-Z nanoparticles ($CdWO_4$) are promising, but are radiation sensitive

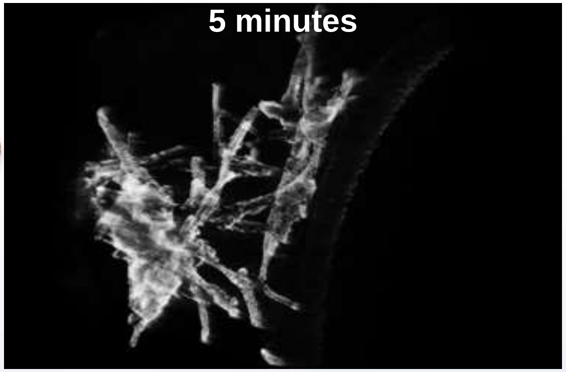


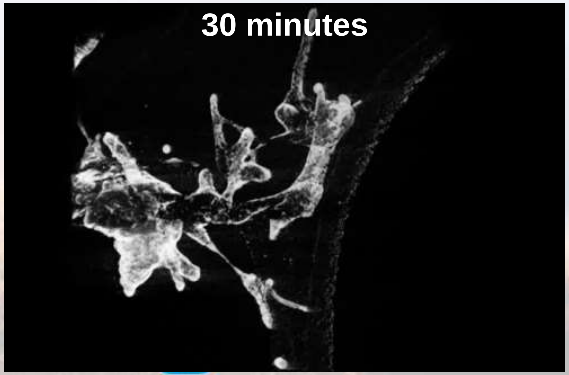
***In situ* Ion Irradiation TEM (I³TEM)**

Hummingbird tomography stage

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



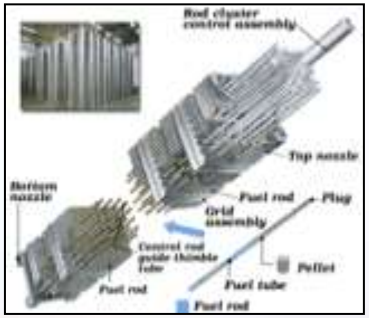


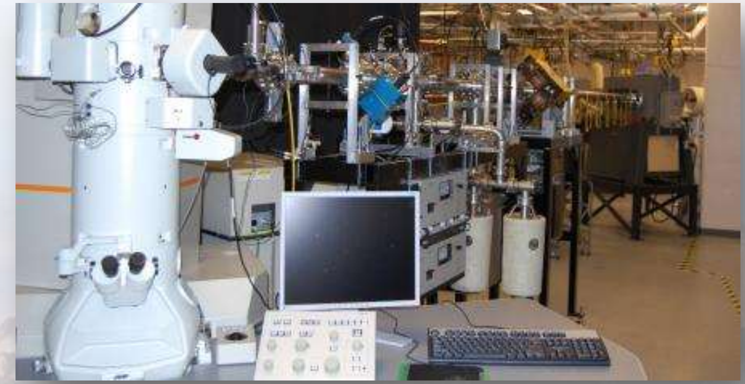


Contributors: S.M. Hoppe, B.A. Hernandez-Sanchez, T. Boyle

Investigating the nm Scale to Understand the km Scale

1 nm | 1 μm | 1 mm | 1 m | 1 km | 10^7 m





In situ Ion Irradiation TEM (I³TEM)

Ion Beam Lab (IBL)

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

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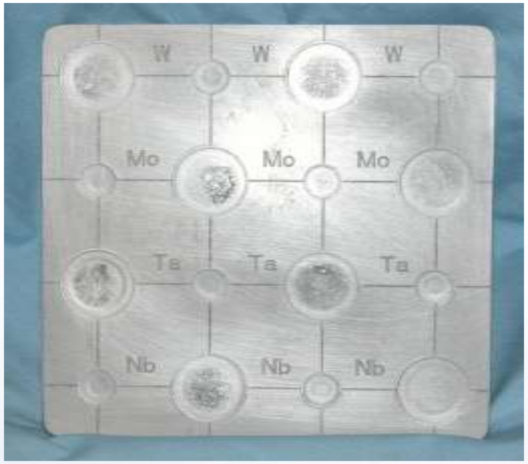
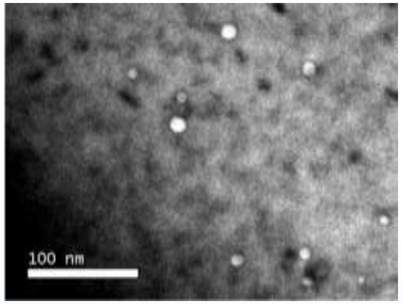
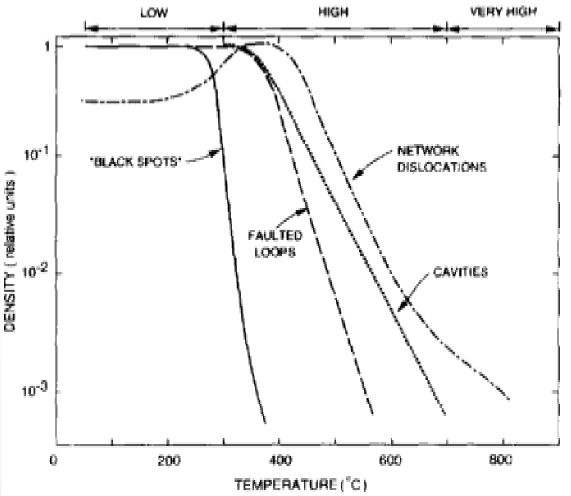




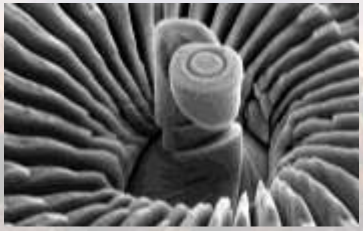


Microstructural Characterization (XTEM)

Mechanical Properties (small-scale testing)

Local Composition (Diffusion Couples) + Local Microstructural Control (Ion Irradiation)

Validating Comparison to Neutron Irradiation Experiments + Investigation into new materials



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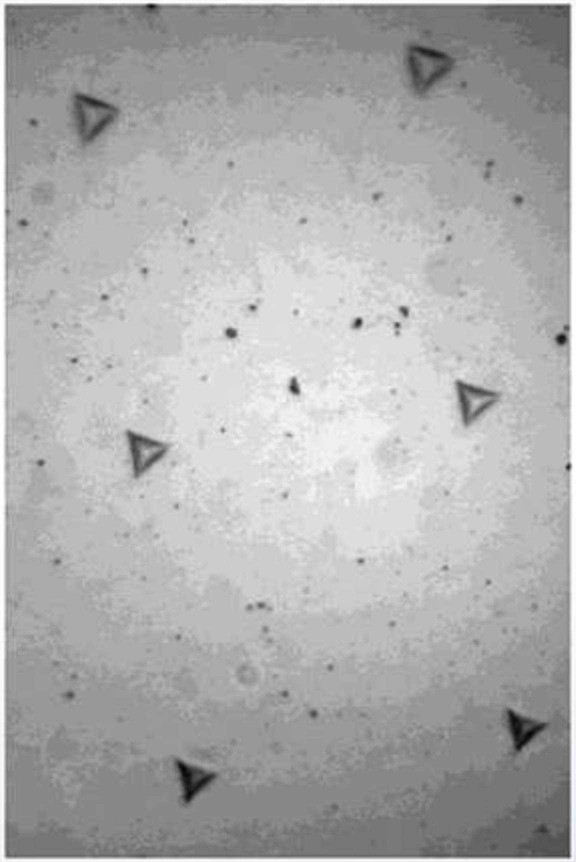
Testing of Irradiated Stainless Steels

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

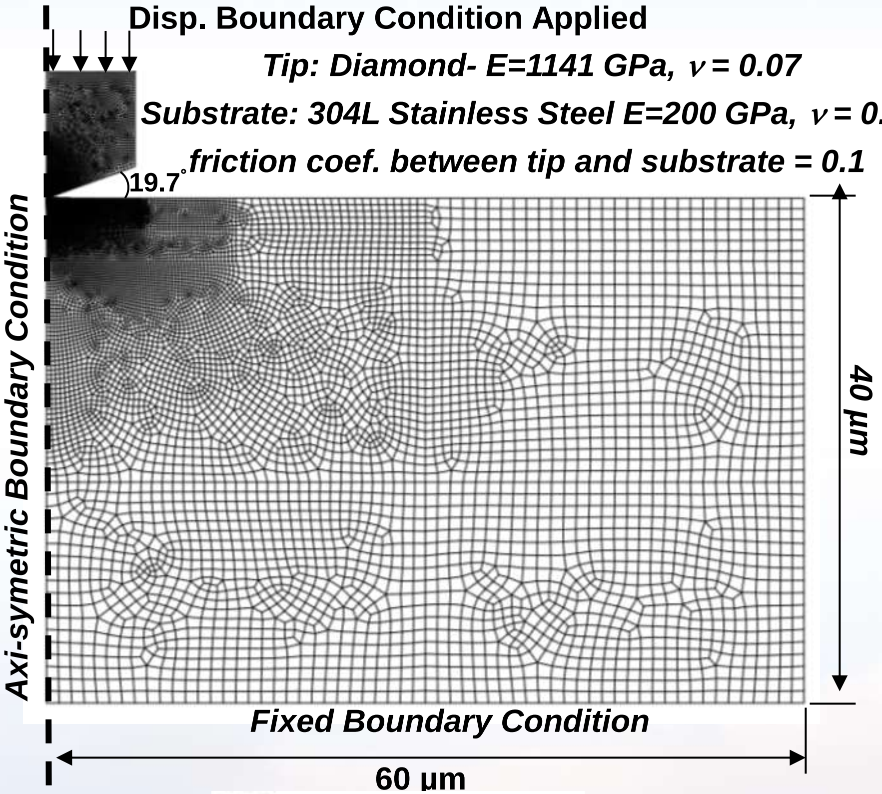
Thus, we irradiated

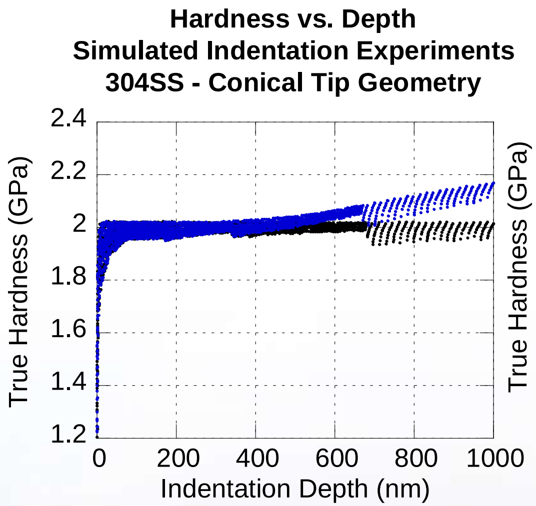
- 420, 409, and 316L SS
- Approximately 10 dpa, 40 dpa, and 100 dpa
- Temperatures of 400 ºC, 500 ºC, and 600 ºC

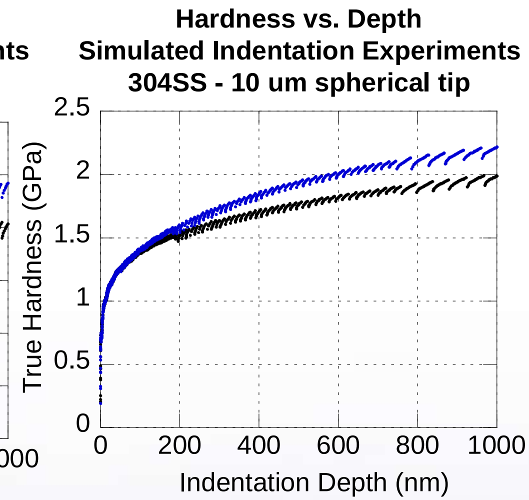
Three steel compositions were irradiated under various conditions. Nanoindentation was selected as the optimal small scale testing method.

Finite Element Simulations of Indentations into Ion Irradiated Steels

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

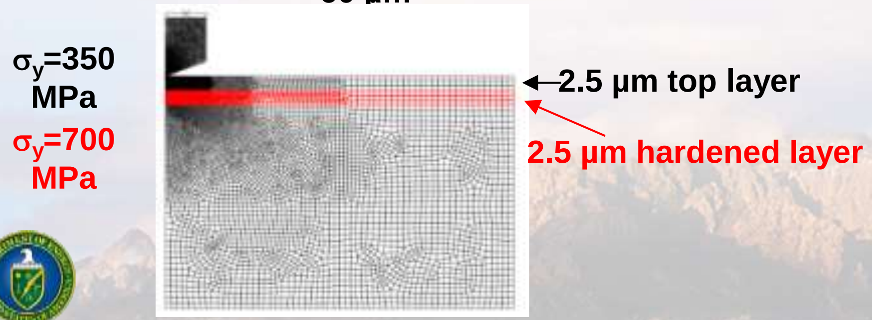






Without hardened subsurface layer

With hardened subsurface layer



Deviations due to ion irradiation are expected from both spherical and conical indentations

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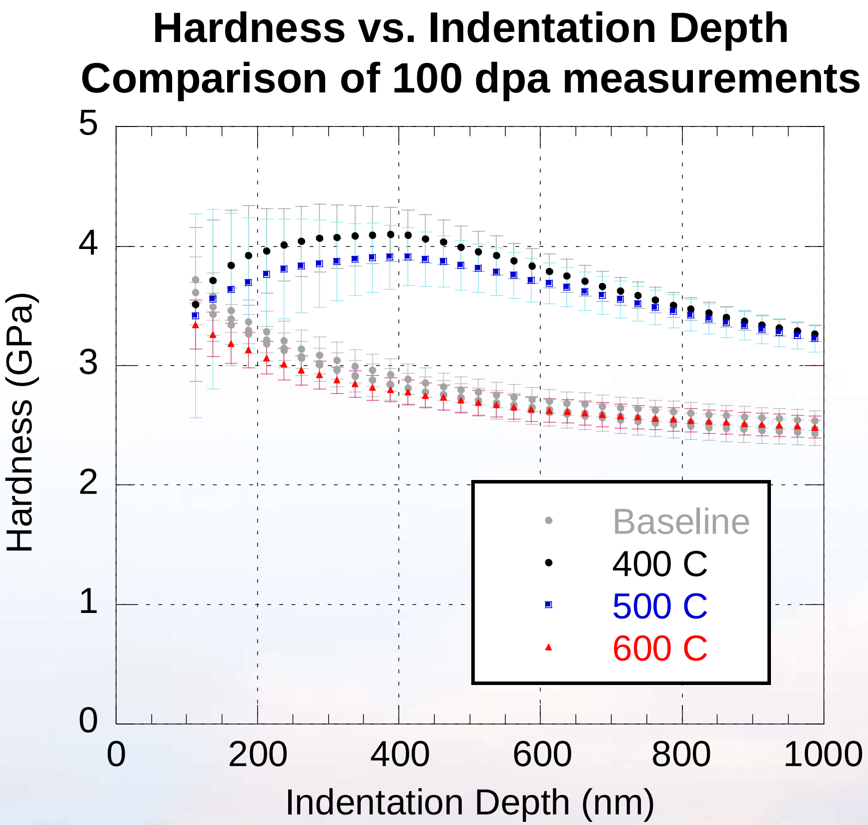
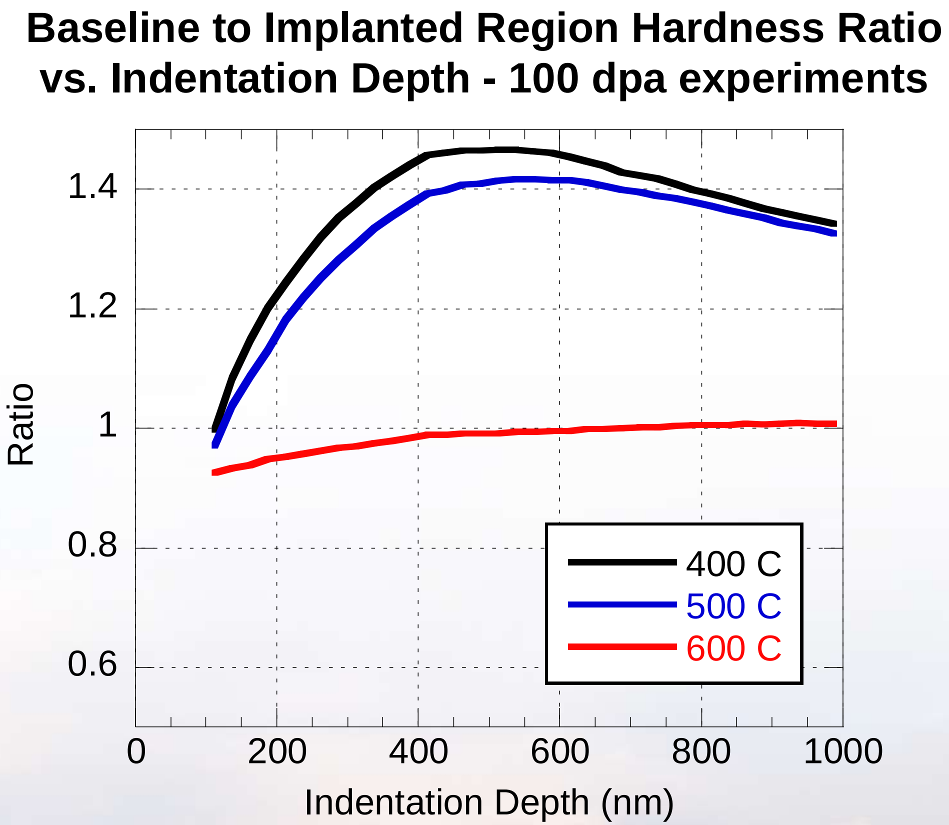
Berkovich Indentation of 100 dpa Irradiated Samples

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





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

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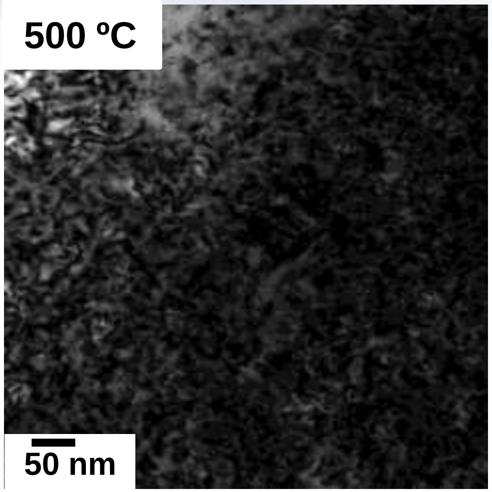
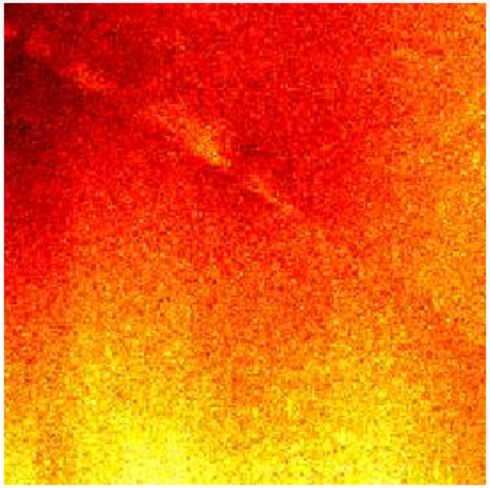
Microstructural Evolution between 500 ºC and 600 ºC

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

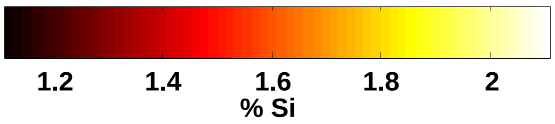
Collaborators: L.N. Brewer, A.J. Kilgo, P. Kotula

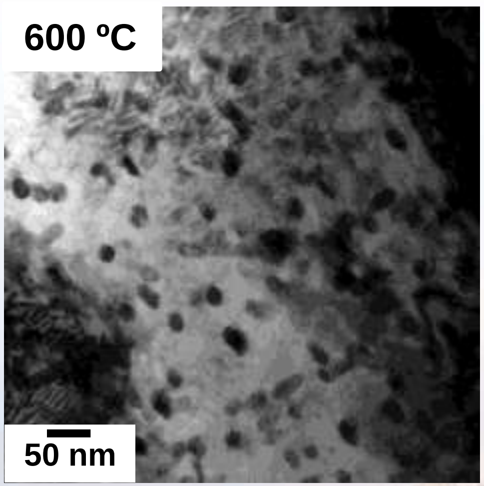


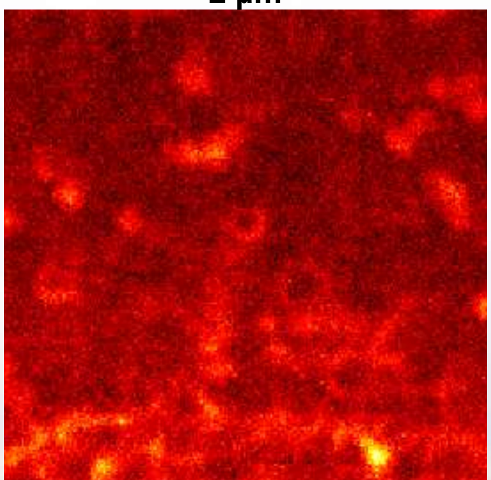


2 μm

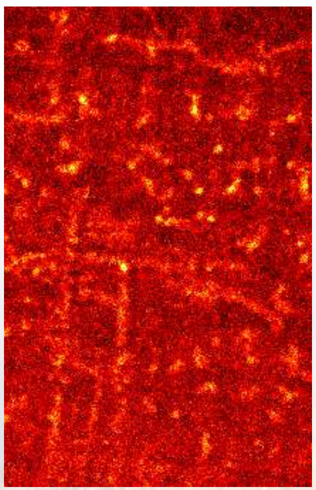
- Large number of small defects present in the irradiated region
- No significant segregation of either the Ni or Si constituents





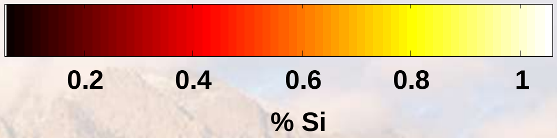


500 nm



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

Micropillar Compression Experiments

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

Sample Preparation:

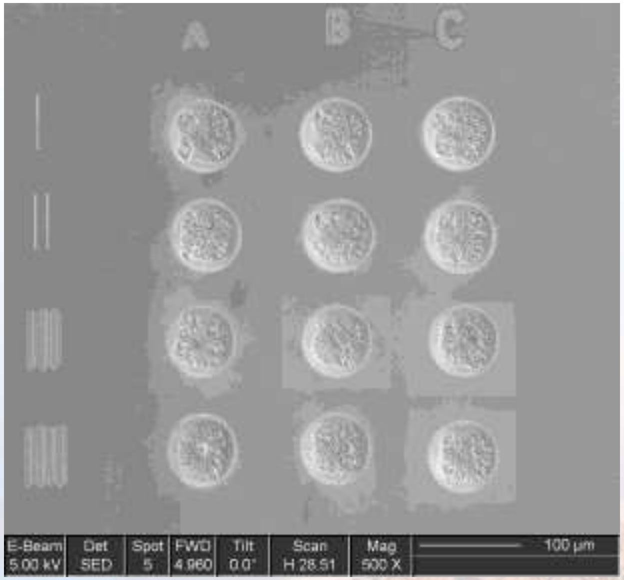
- Copper single crystals (FCC)
- Different crystallographic orientations: (100), (110), and (111)
- Self-ion Implants at 30 MeV to 0 (control), 50 dpa, and 100 dpa.

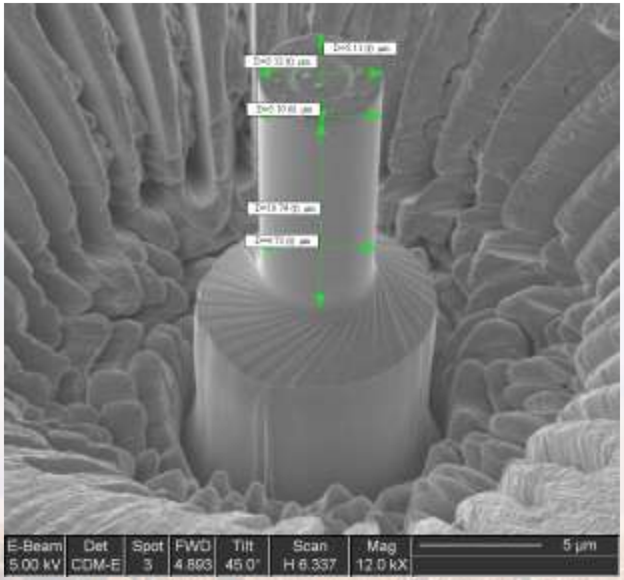
Pillar Manufacturing:

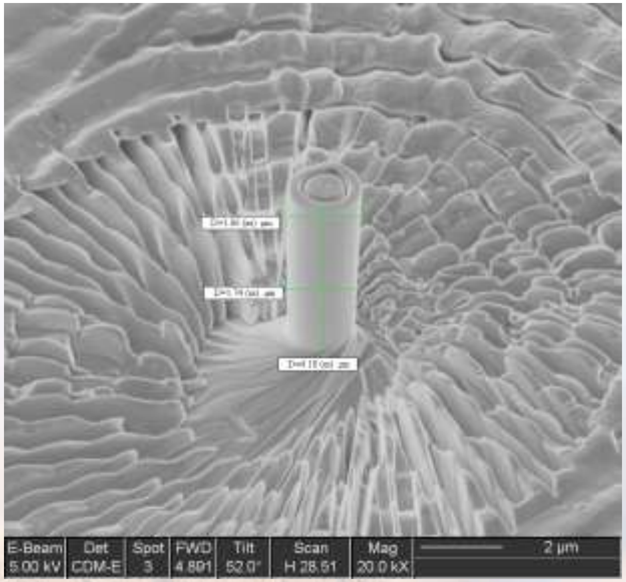
- We employed Uchic's FIB lathe machining process for straight-walled cylinders.
- Array of at least 9 nominally identical pillars tested per condition to assess statistical variability.
- Height varies from 4 µm to 10 µm

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 $0.025\ s^{-1}$.

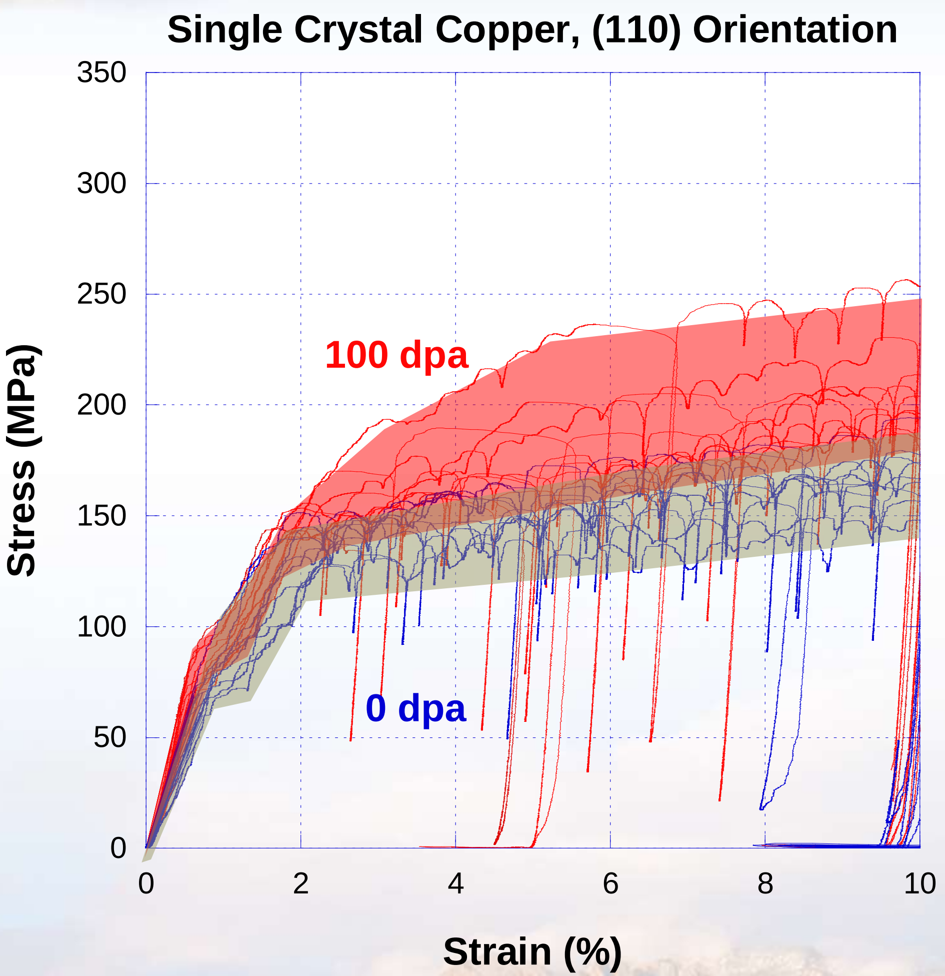


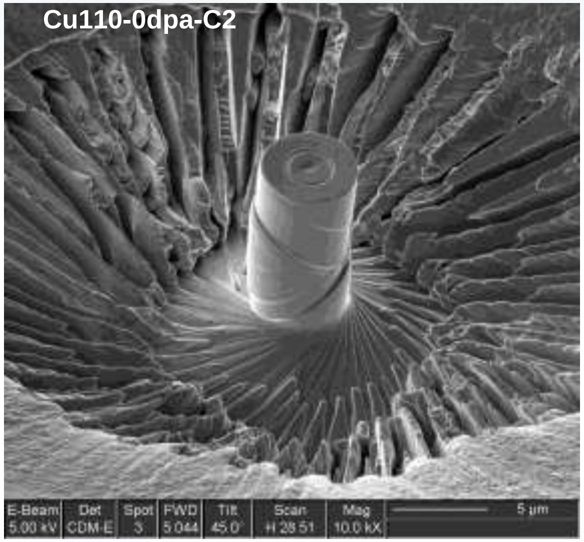


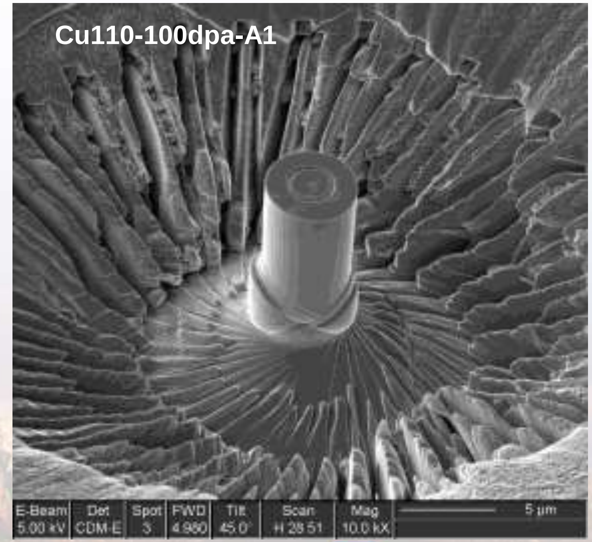


Large Micropillar Compression

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



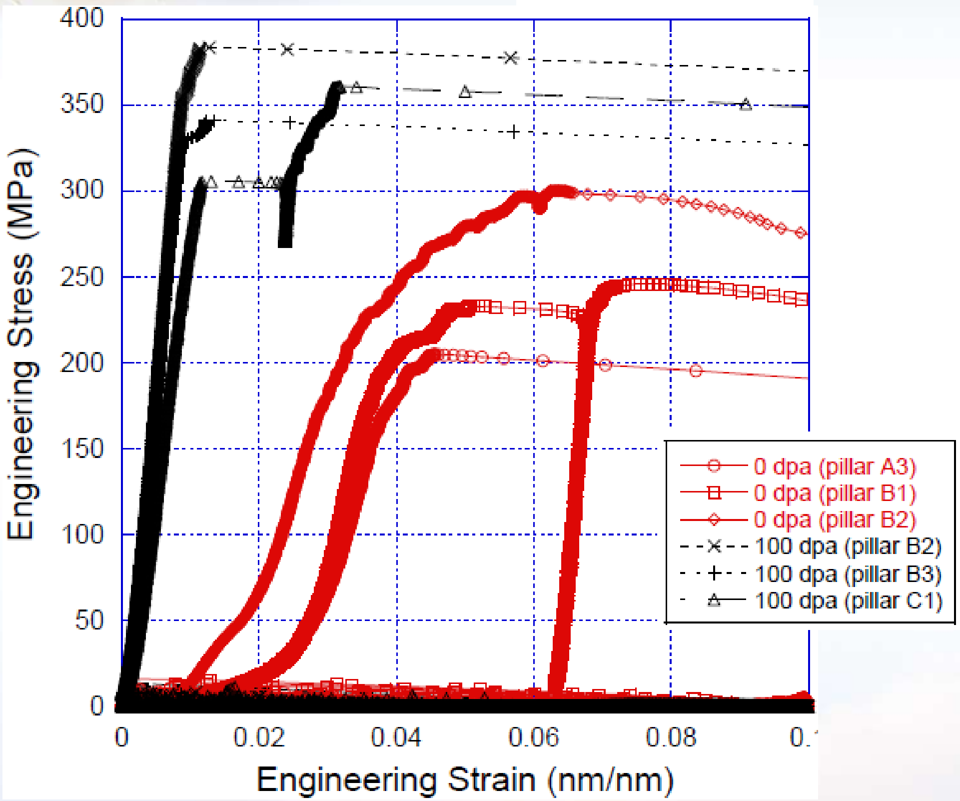


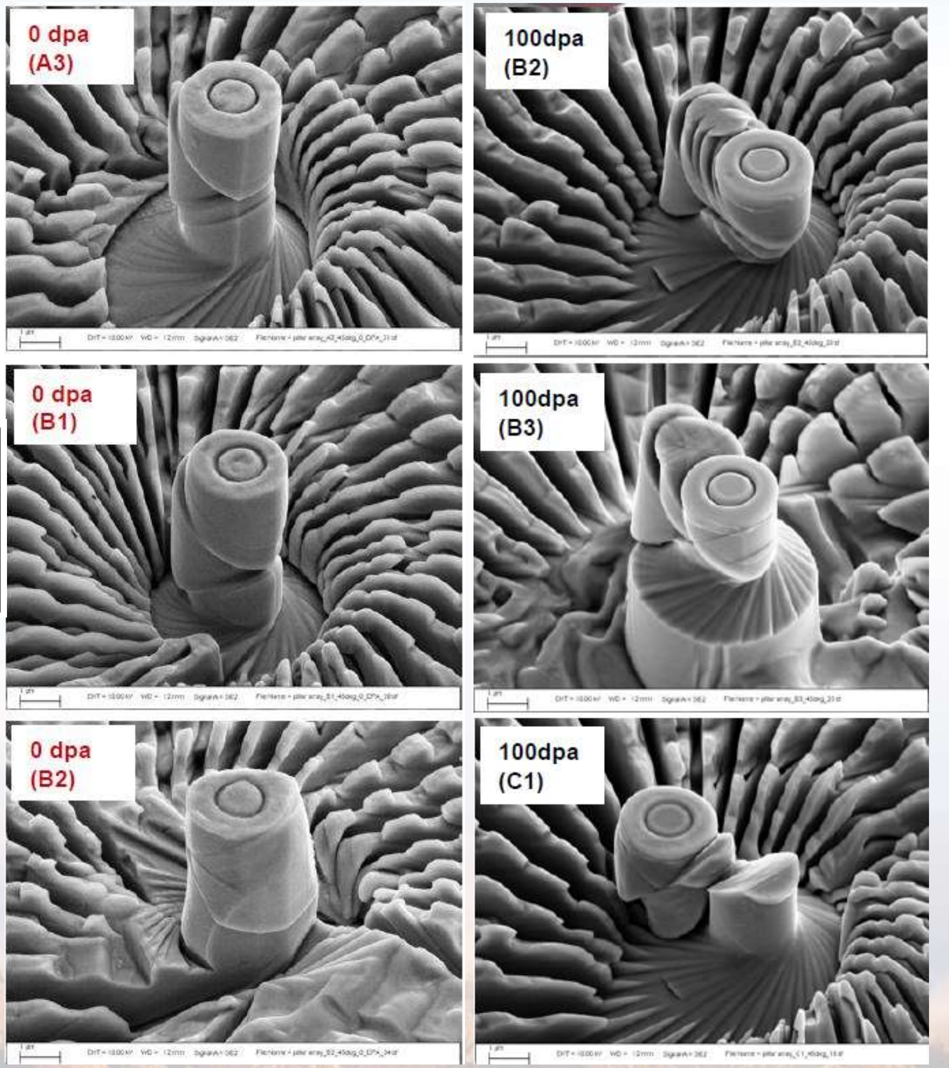


Minimal difference between the control and irradiated 10 µm-tall pillars. Slip occurred in the bottom fraction of the pillars.

Intermediate Micropillar Compression

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





5 µm-tall pillars show greater distinction with catastrophic failure

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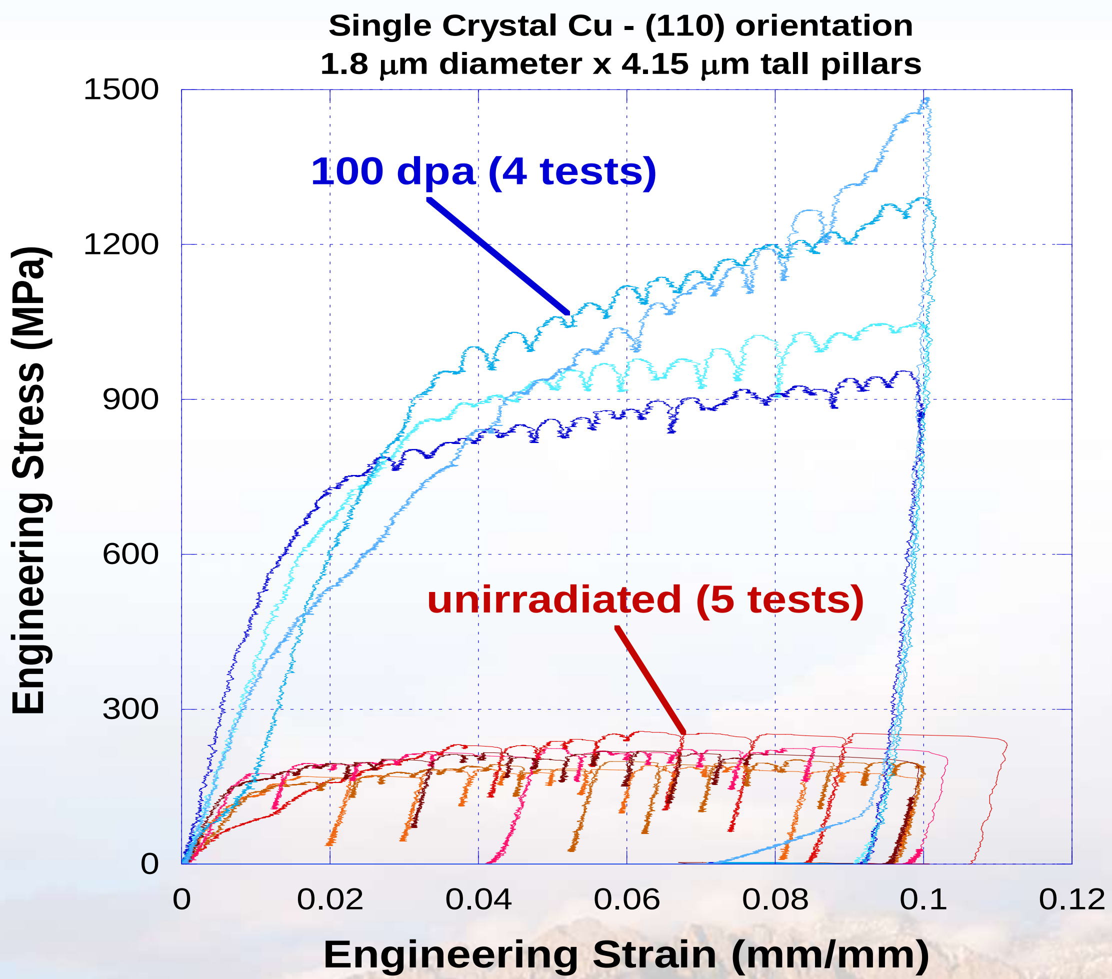
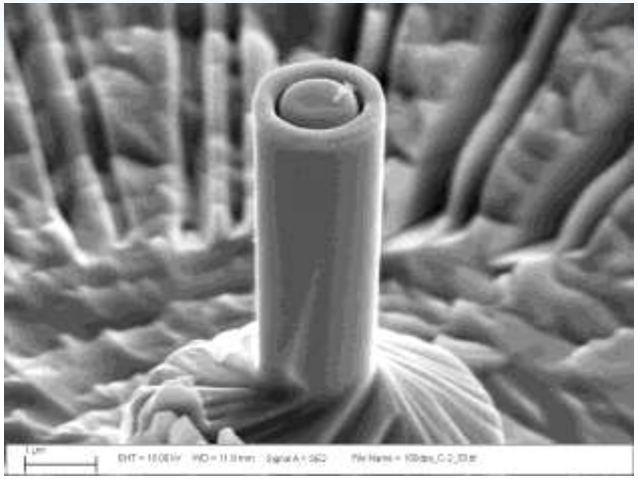
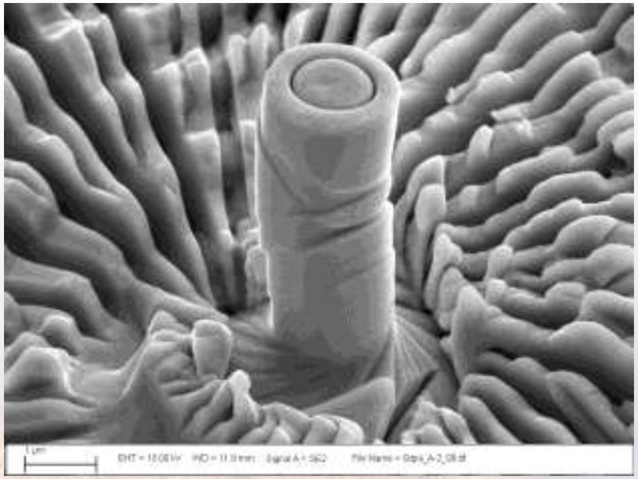
Small Micropillar Compression

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



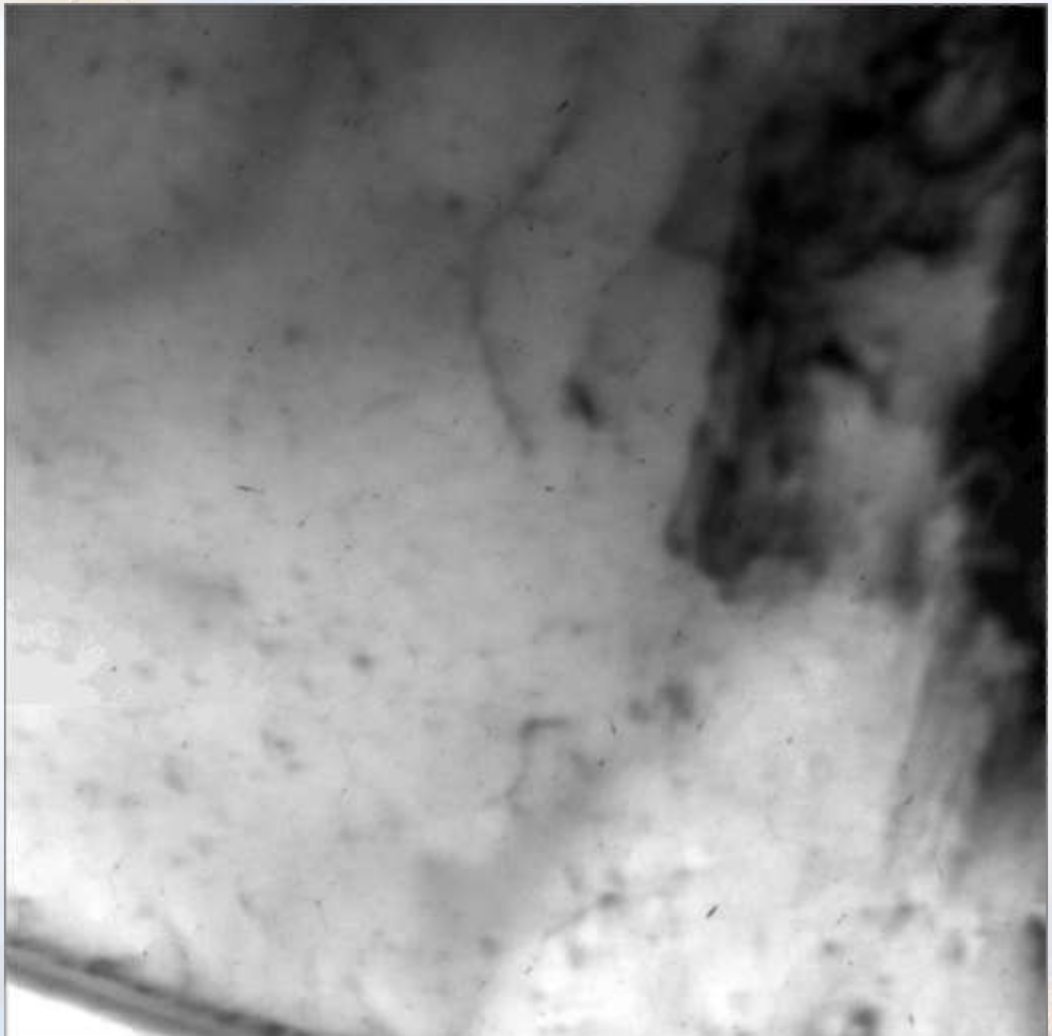


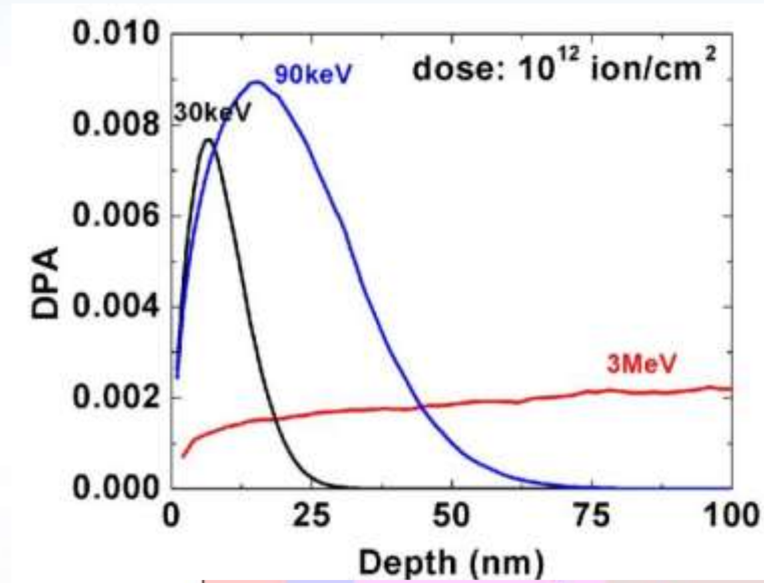


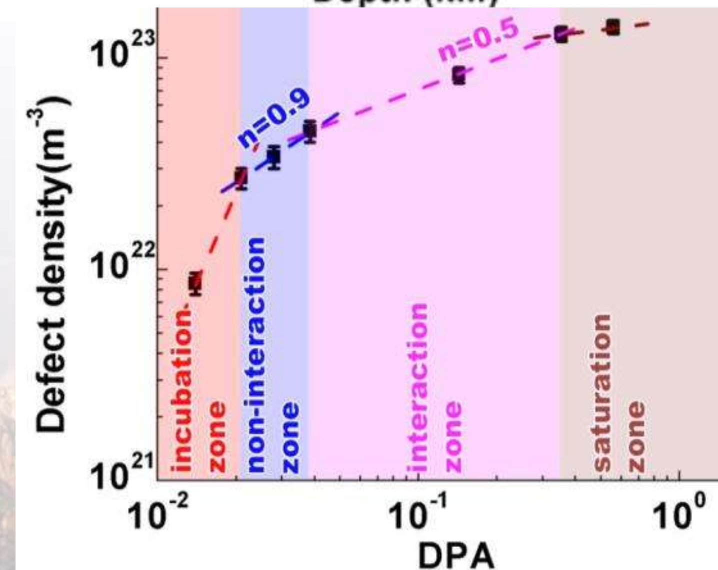
Initial tests indicate that the 4 μm-tall pillars are 5 times stronger and show no signs of slip band formation

Quantifying Defect Evolution

Collaborators: N. Li & A. Misra

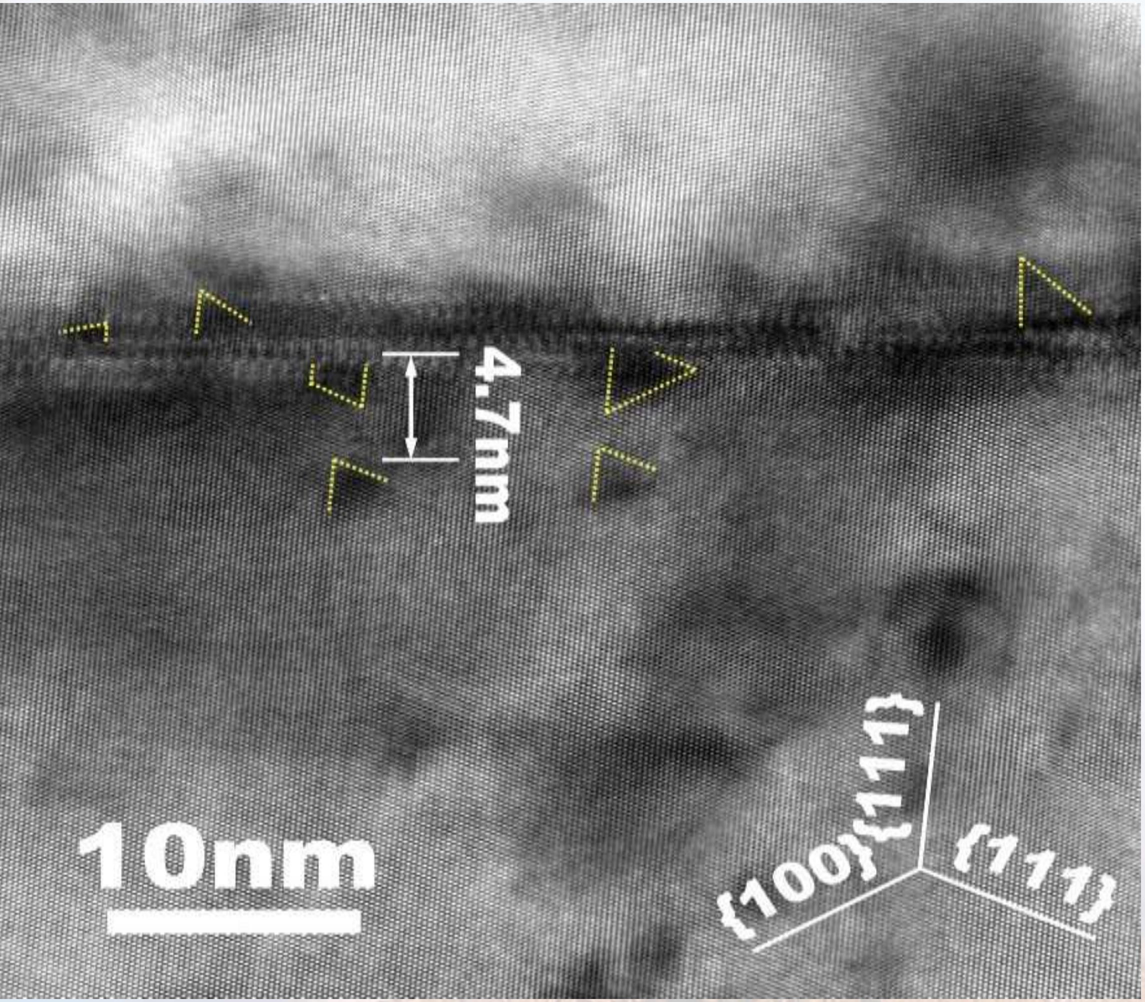


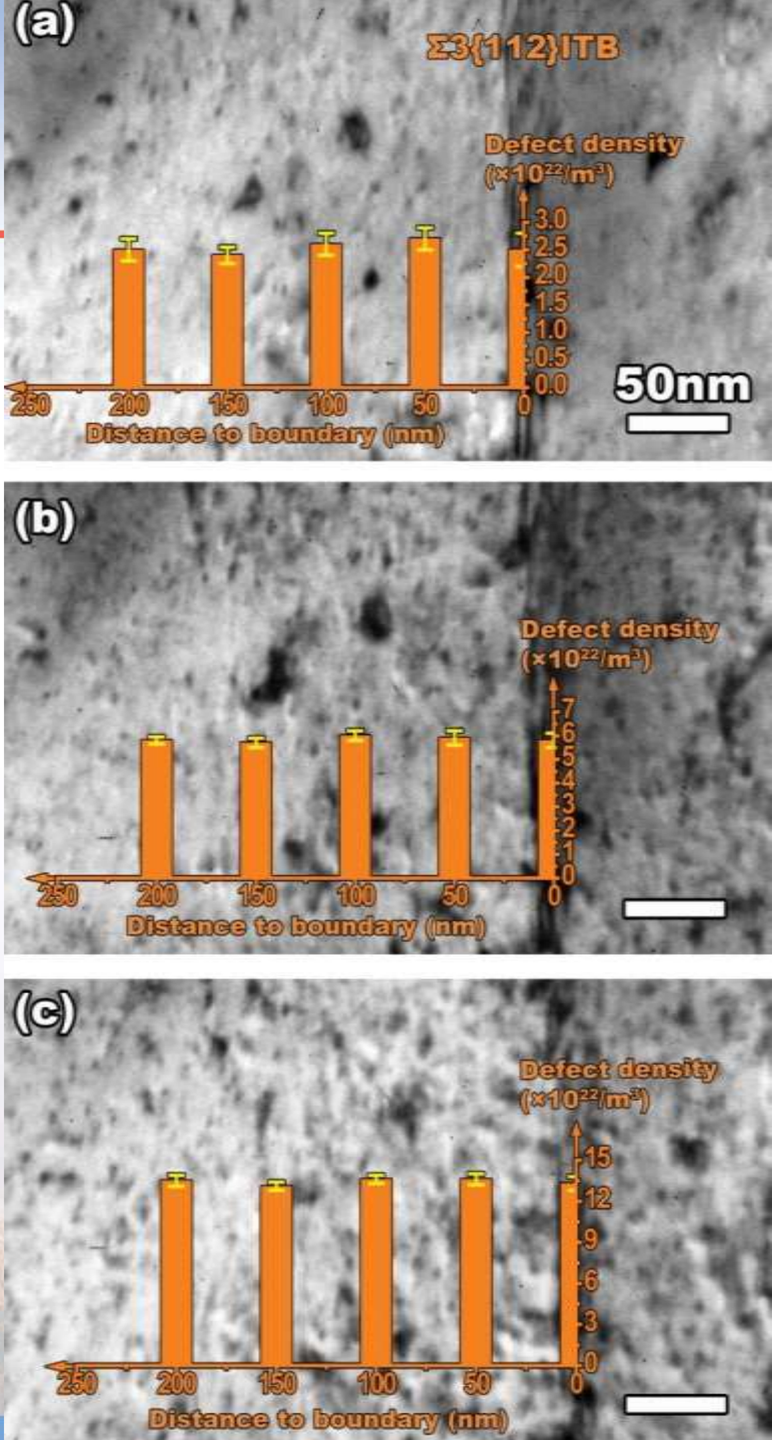




Defects are Altered Little by the Presence of Grain Boundaries

Collaborators: N. Li & A. Misra





SFT appear to be directly at GB

No change in defect density is observed near GB

Summary

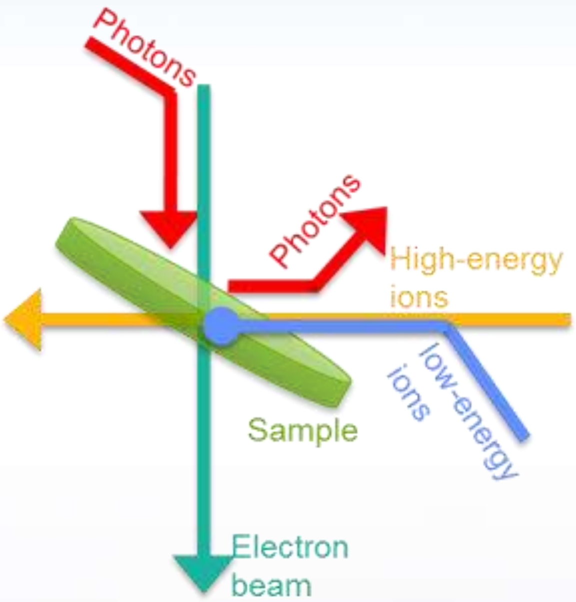
The Ion Beam Lab at Sandia National Laboratories applies a variety of tools to a wealth of national problems

Sandia's I[3]TEM is one of a few in the world

- *In situ* irradiation from H to Au
- *In situ* gas implantation

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



<u>**Collaborators:**</u>

- **IBL: D. Buller, B.G. Clark, B.L. Doyle, S. Hoppe, S. Rajasekhara, J. Villone, & G. Vizkelethy**
- **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**

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