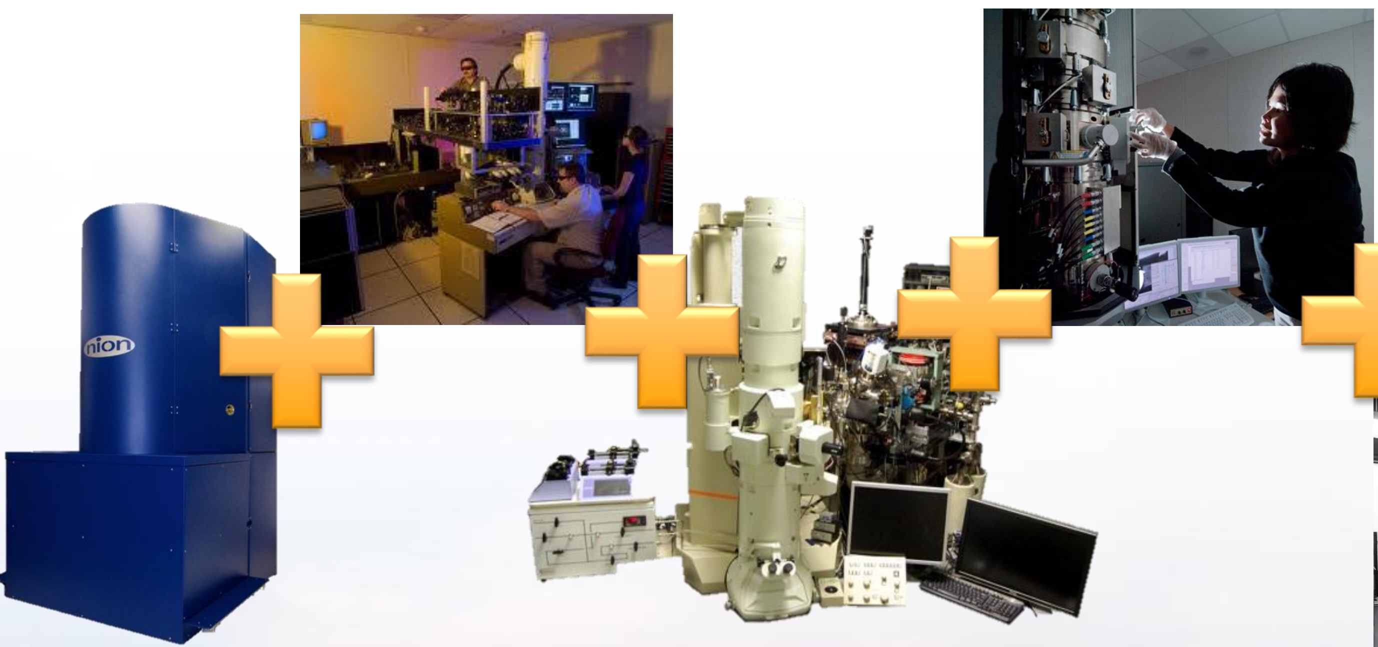
Integrative In-situ TEM

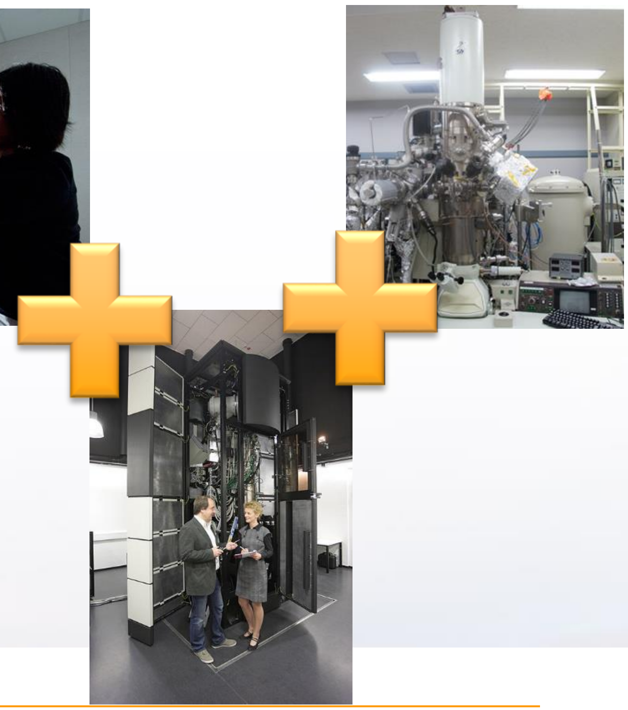
SAND2017-12312C

K. Hattar, B. Muntifering, P. Price, C. Barr, S. Briggs, C. Taylor, D. Bufford, & K. Jungjohann

Sandia National Laboratories

October 9, 2017





Collaborators:

- IBLD. Buller, C. Chisholm, B.G. Clark, M.T. Marshall, B. Muntifering, & S.H. Pratt
- Sandia: M. Abere, B. Boyce, T.J. Boyle, R. Dingreville, R.F. Hess, A.C. Kilgo, B.E. Klamm, W.M. Mook, J.D. Puskar, J.A. Scott, & J.A. Sharon
- External: A. Aitkaliyeva, H. Bei, P.J. Ferreira, K.J. Ganesh, E.P. George, D. Gross, P. Hosemann, J. Kacher, S. Maloy, A. Minor, J. Qu, S. Rajesekhara, I.M. Robertson, D. Stauffer, & Hysitron Inc.

This work was partially funded by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Evolution of Transmission Electron Microscopy

Benefits & Limitations of *in situ* TEM

Benefits

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

Limitations

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

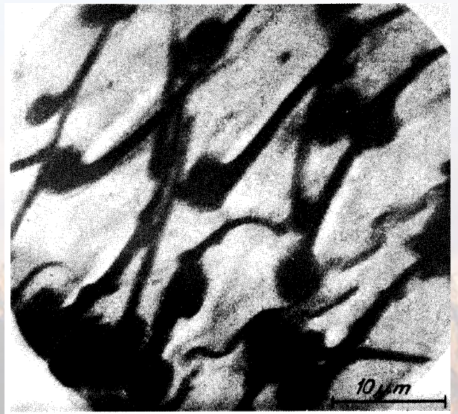


Fig. 6: Wing surface of the house fly.
(First internal photography, U = 60 kV, M_{el} = 2200)
(Driest, E., and Müller, H.O.: Z. Wiss. Mikroskopie 52, 53-57 (1935)

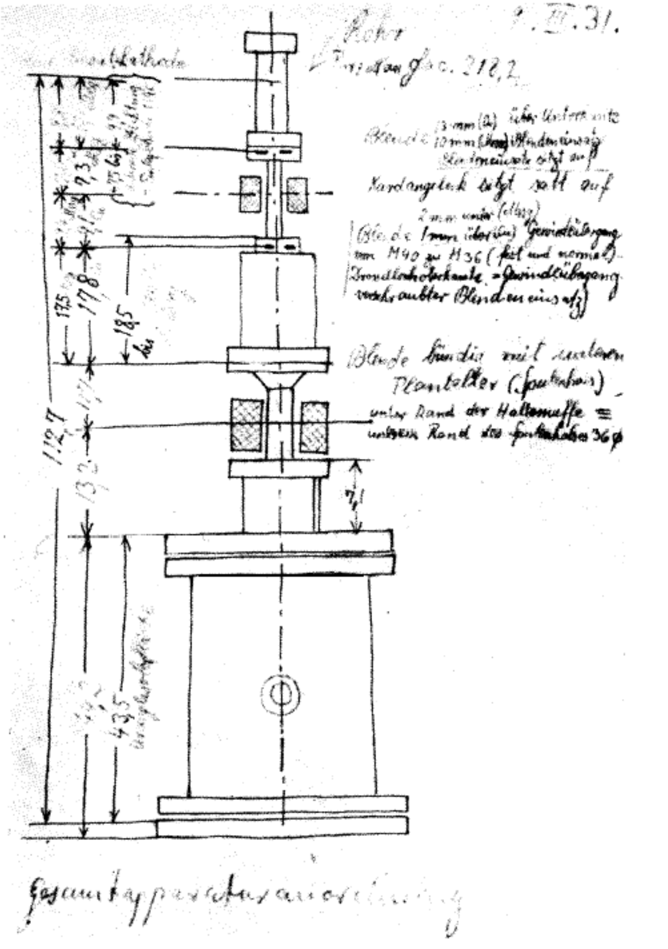
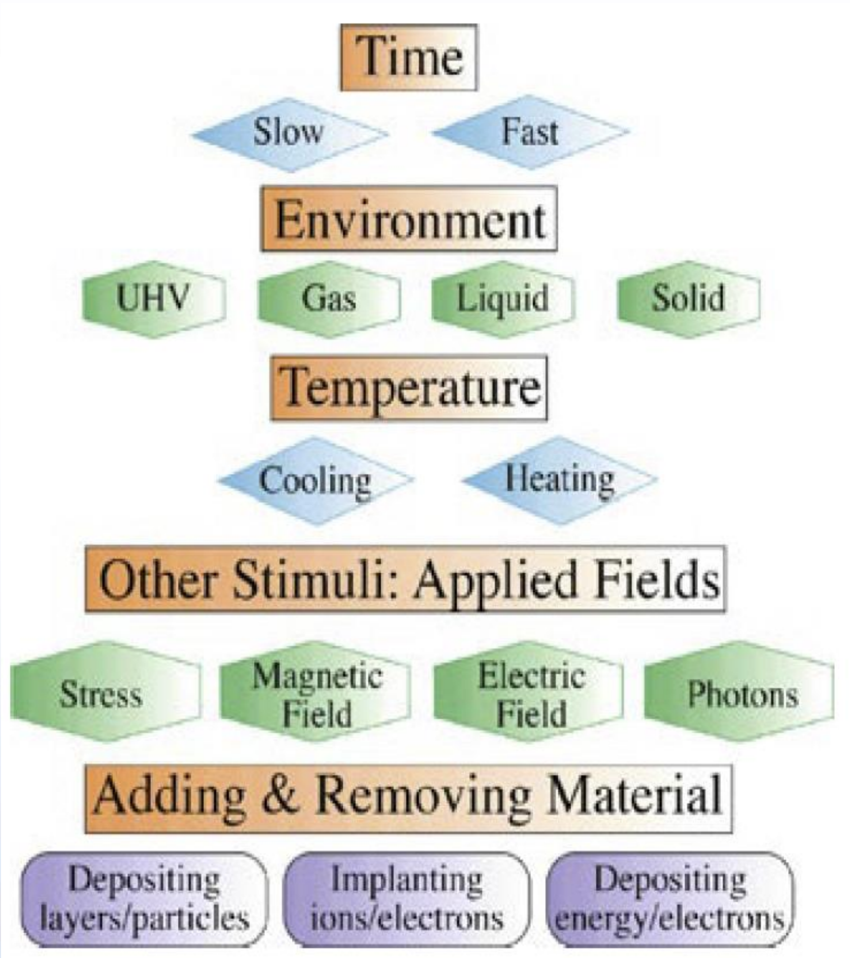
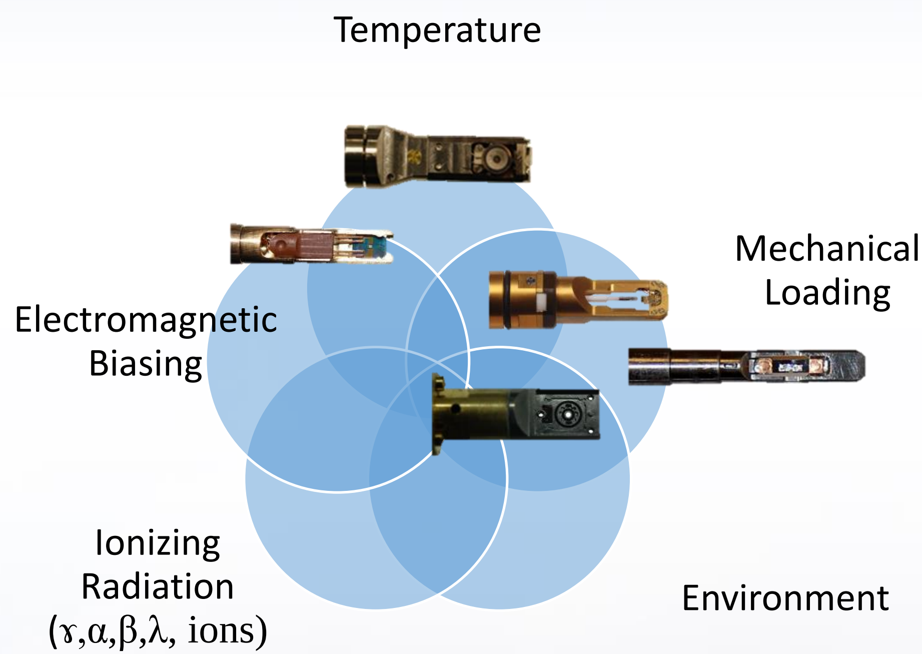


Fig. 2: Sketch by the author (9 March 1931) of the cathode ray tube for testing one-stage and two-stage electron-optical imaging by means of two magnetic electron lenses (electron microscope) [8].

E. Ruska Nobel lecture (1986)

Can TEM Capabilities be Better Integrated?





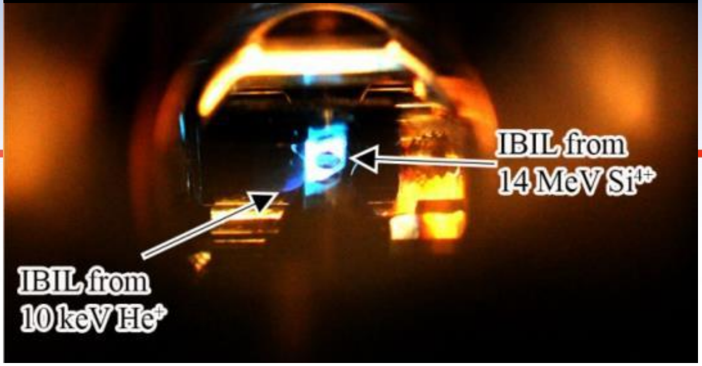
"Towards an integrated materials characterization toolbox"

- I.M. Robertson et al. JMR 26(11) 2011

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

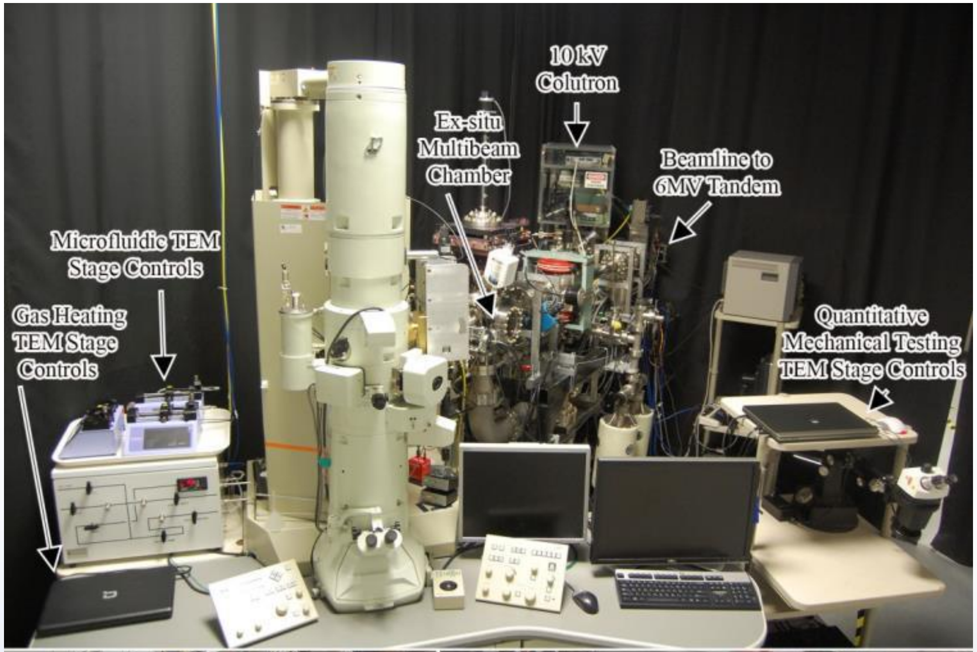
Collaborator: D.L. Buller

IBIL from a quartz stage inside the TEM

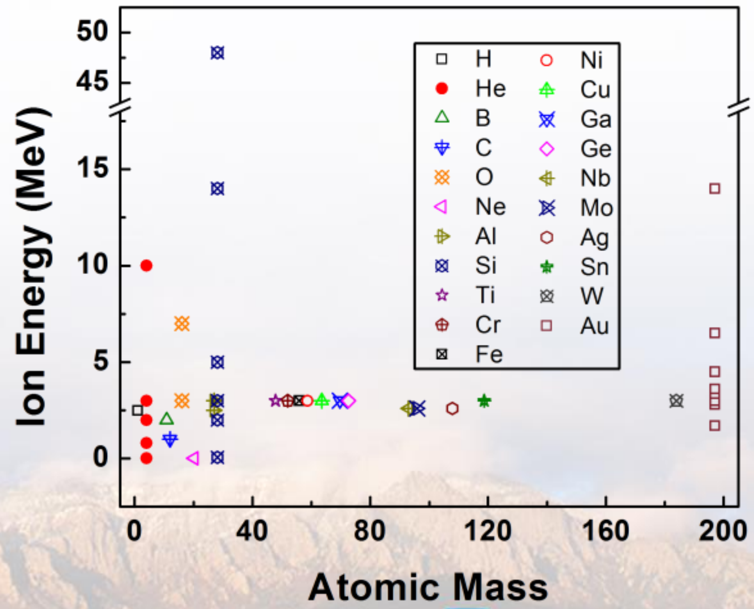


Direct real time observation of ion irradiation, ion implantation, or both with nanometer resolution

10 kV Colutron - 200 kV TEM - 6 MV Tandem



Ion species & energy introduced into the TEM



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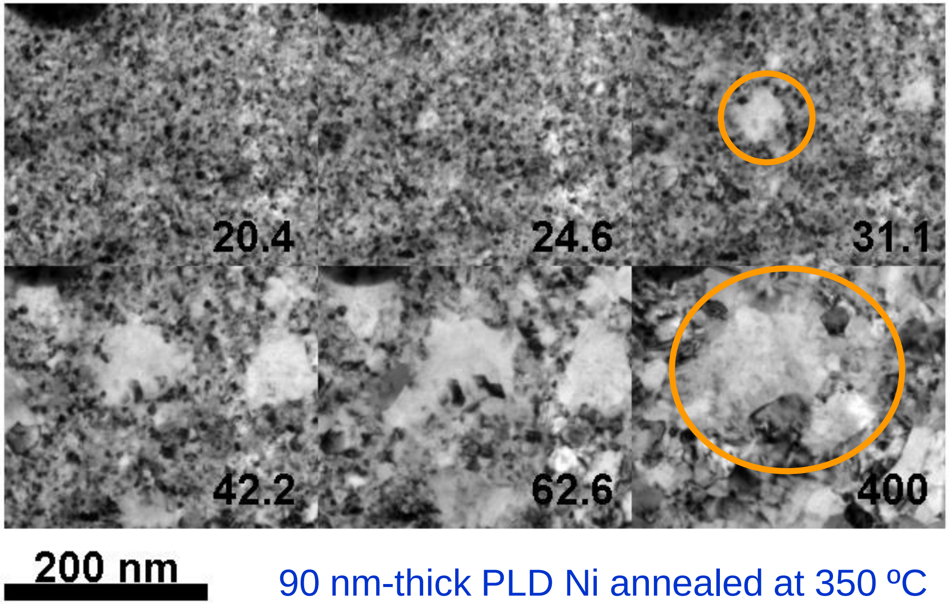
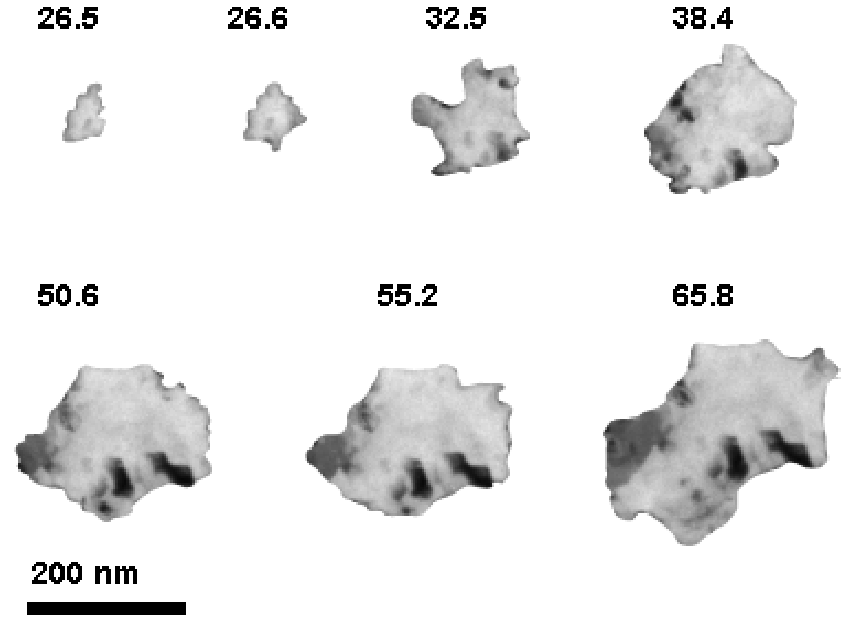
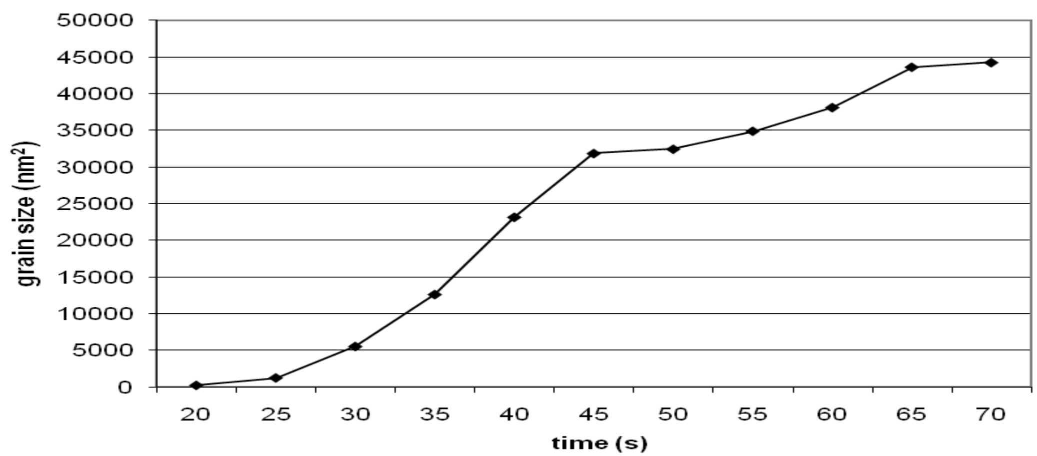
Dynamics of Individual Grain Boundaries



90 nm-thick PLD Ni annealed at 350 ºC





Grain curvature does not appear to play a significant role

Kacher, Josh, et al. "Thermal stability of Ni/NiO multilayers." MaterialsScience and Engineering: A 568 (2013): 49-60.

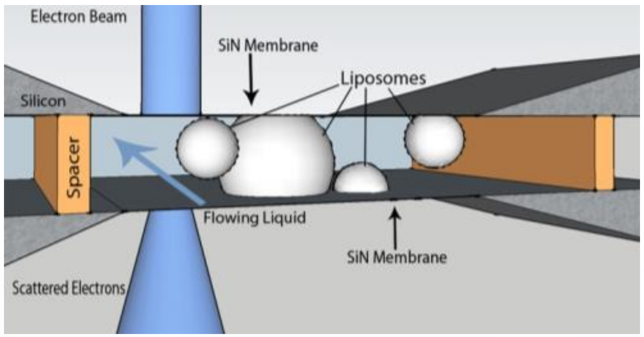
In situ TEM Microfluidic Environments

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

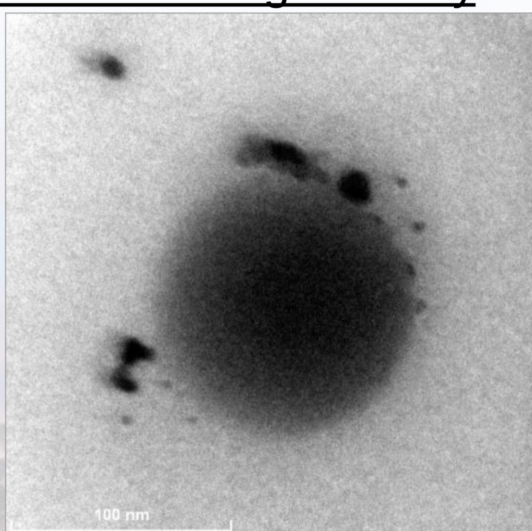
Microfluidic Stage

- 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

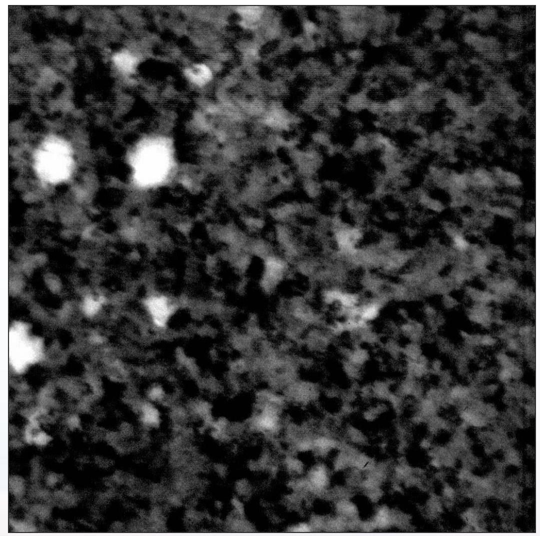


Protocell Drug Delivery



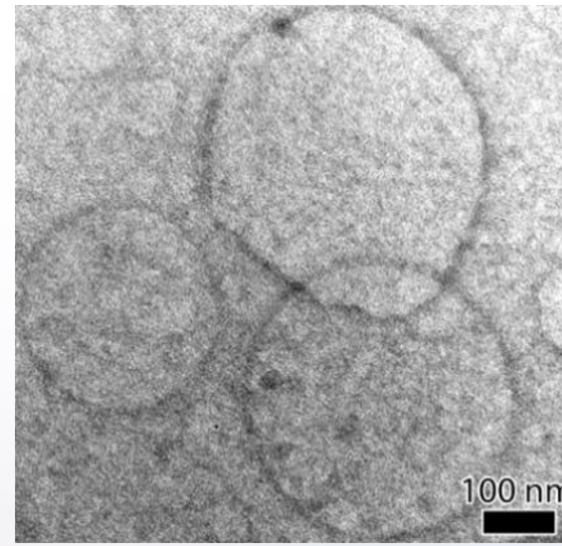
Liposome encapsulated Silica destroyed by the electron

Fe Corrosion



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

Liposomes in Water



Liposomes imaged in flowing aqueous channel

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

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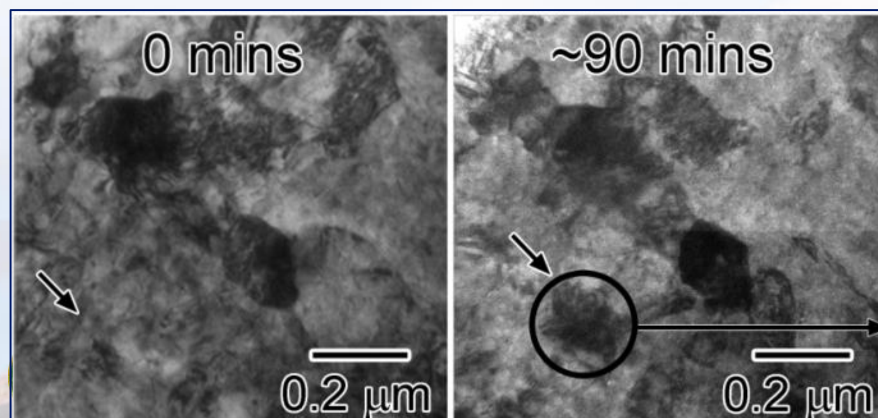
In situ TEM Gas Environments

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

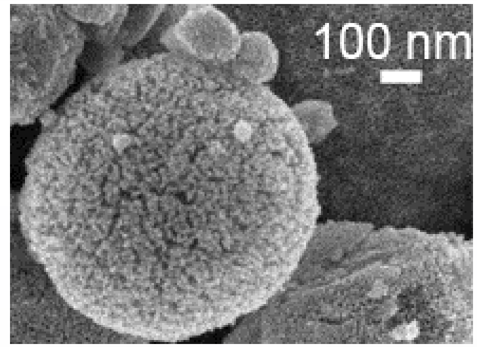
Vapor-Phase Heating TEM Stage

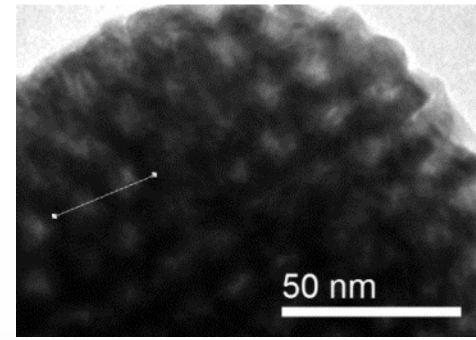
- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Compatible with MS and other analytical tools

Absolute hydrogen pressure: 327 torr,
Ramp rate: 1 ^{o}C/s, Final temperature: ~ 400 C,

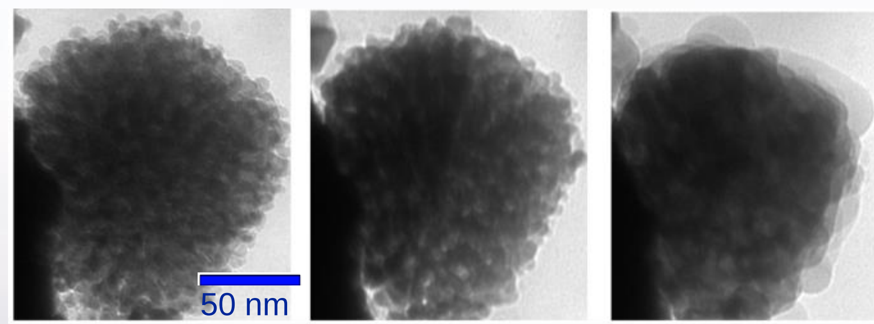


New *in situ* atmospheric heating experiments provide great insight into range of environmental studies





- 1 atm H_2 after several pulses to specified temp.



125° 200° 300°

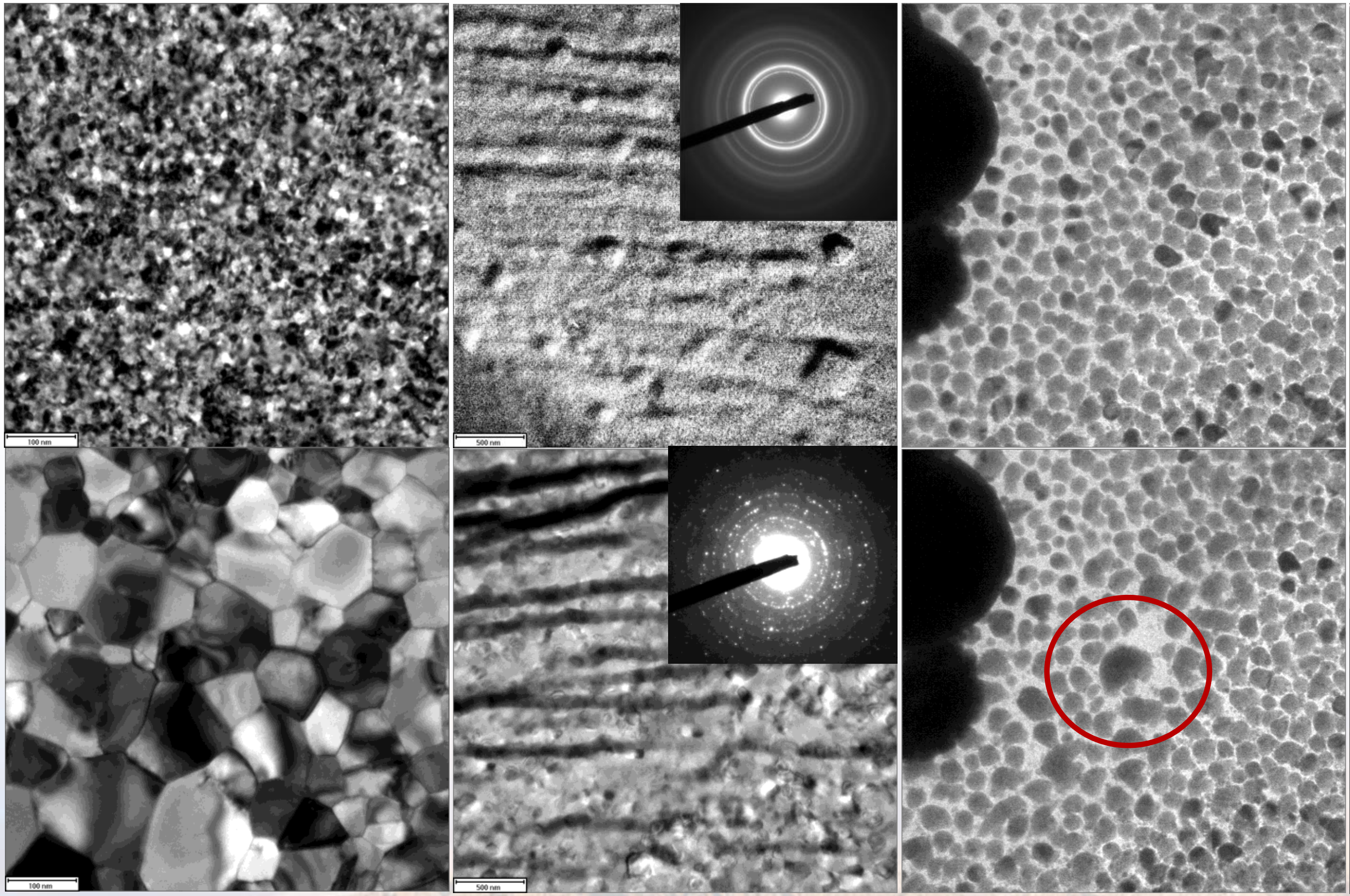
Even greater potential is envisioned when it is combined with *in situ* ion irradiation.

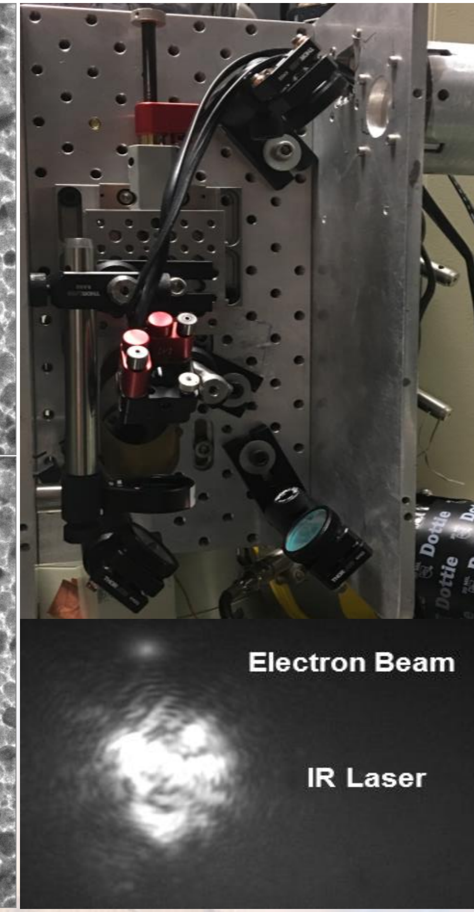
Laser Initiation Heating, Induced Reactions, and Sintering

Collaborator: P. Price, C.M. Barr, D. Adams, M. Abere

Pt Grain Growth | **Reactive Multilayer Films** | **Nanoparticle Sintering**

Before / **After**

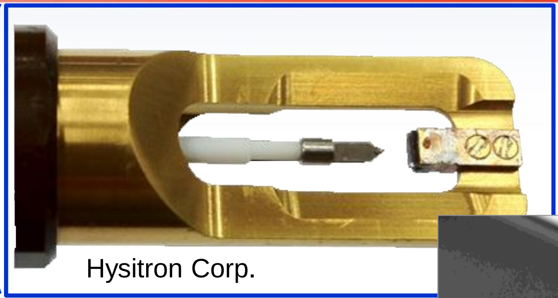




In-situ Laser Modification and Characterization of Materials in the TEM
Wednesday 10/11/ 2017 at 4:40 PM in Convention Center 303

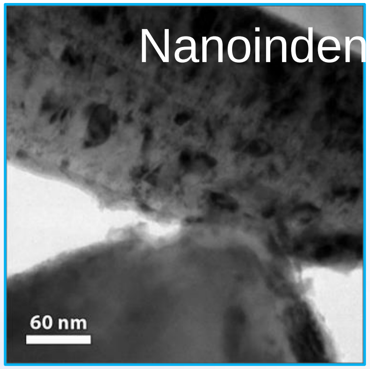
In situ Quantitative Mechanical Testing

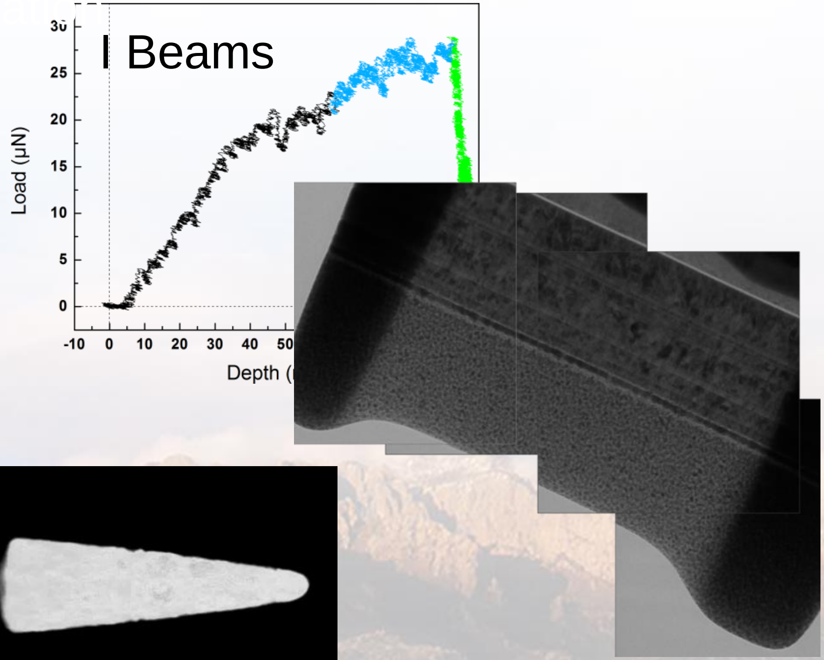
Contributors: J. Sharon, B. L. Boyce, C. Chisholm, H. Bei, E.P. George, P. Hosemann, A.M. Minor, & Hysitron Inc.

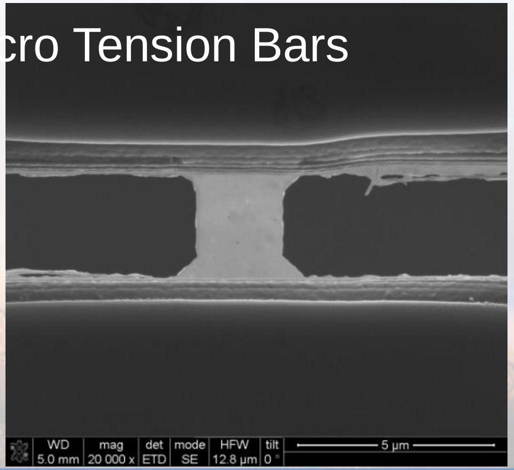


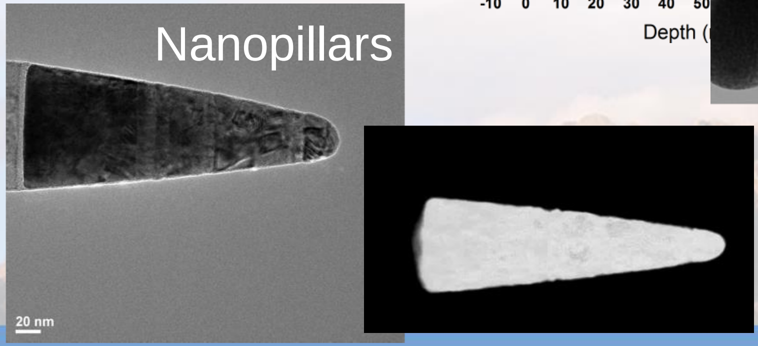
Hysitron PI95 *In Situ* Nanoindentation TEM Holder

- Sub nanometer displacement resolution
- Quantitative force information with μN resolution
- **Concurrent real-time imaging by TEM**





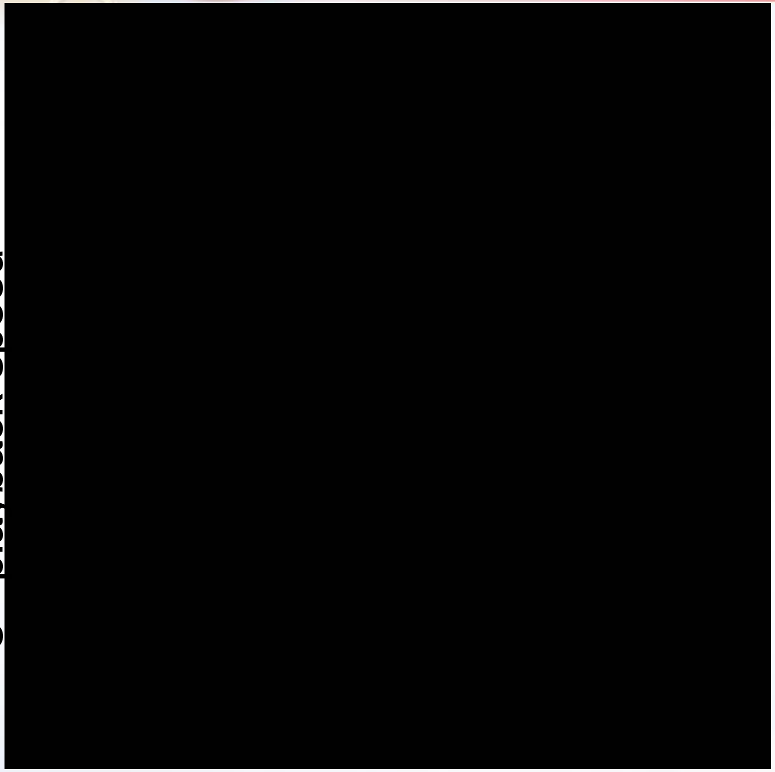


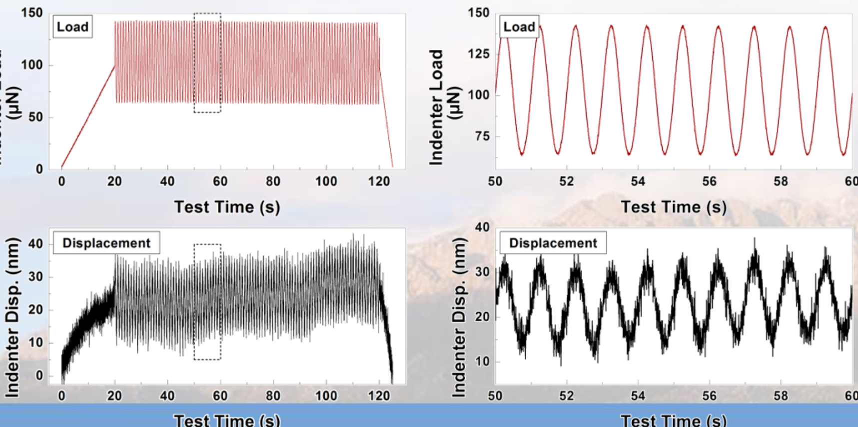


In situ TEM Quantitative Fatigue Testing

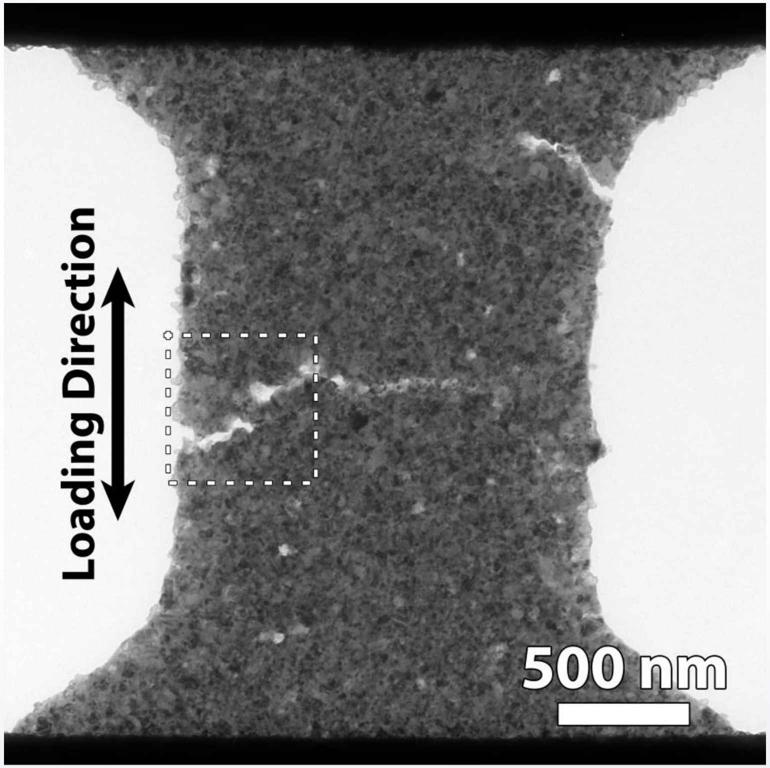
Contributors: D.C. Bufford, D. Stauffer, W. Mook

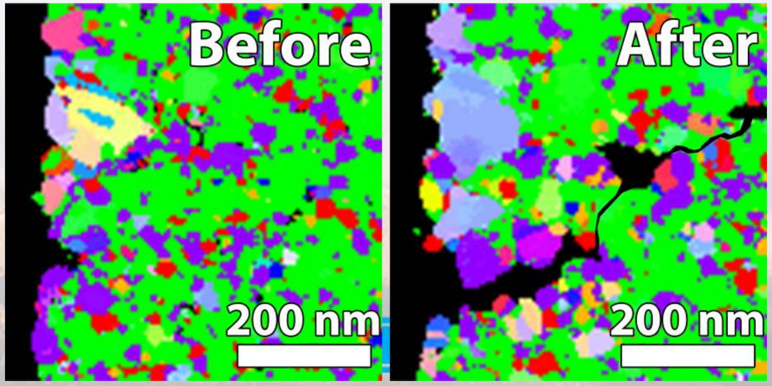
3× playback speed





High cycle fatigue in real time with nanometer resolution

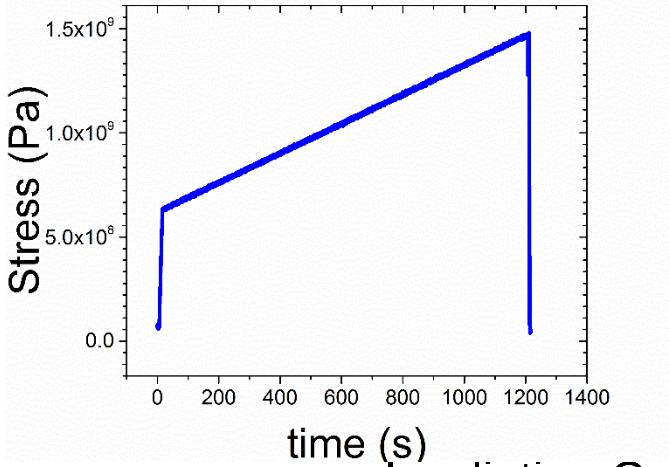




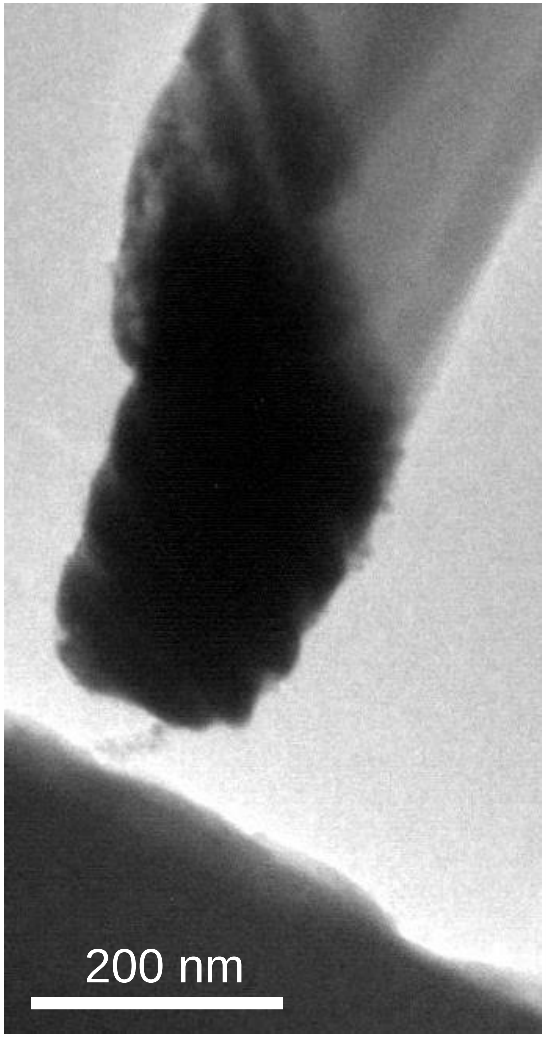
Irradiation Creep (4 MeV Cu^{3+} 10^{-2} DPA/s)

Contributors: S. Dillon & R.S. Averback

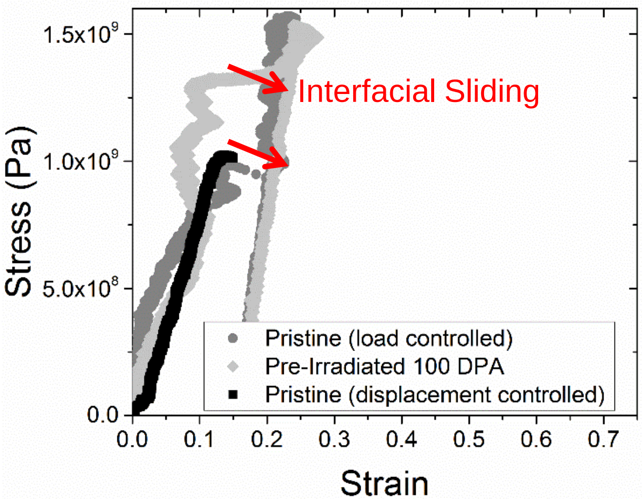
Controlled Loading Rate Experiments



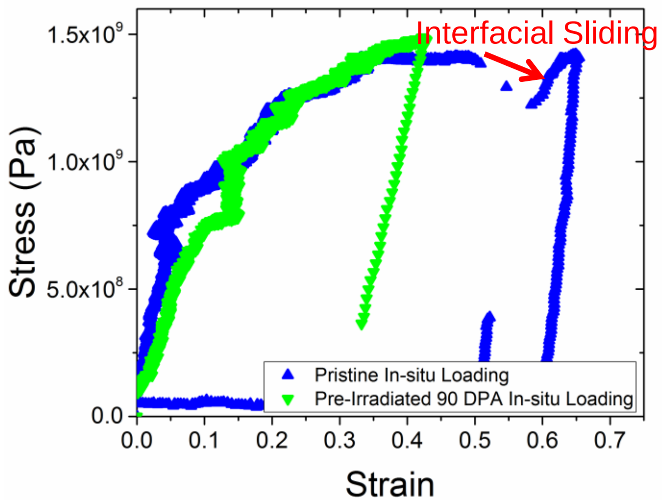
50 nm Cu-W multilayer
20 Min



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

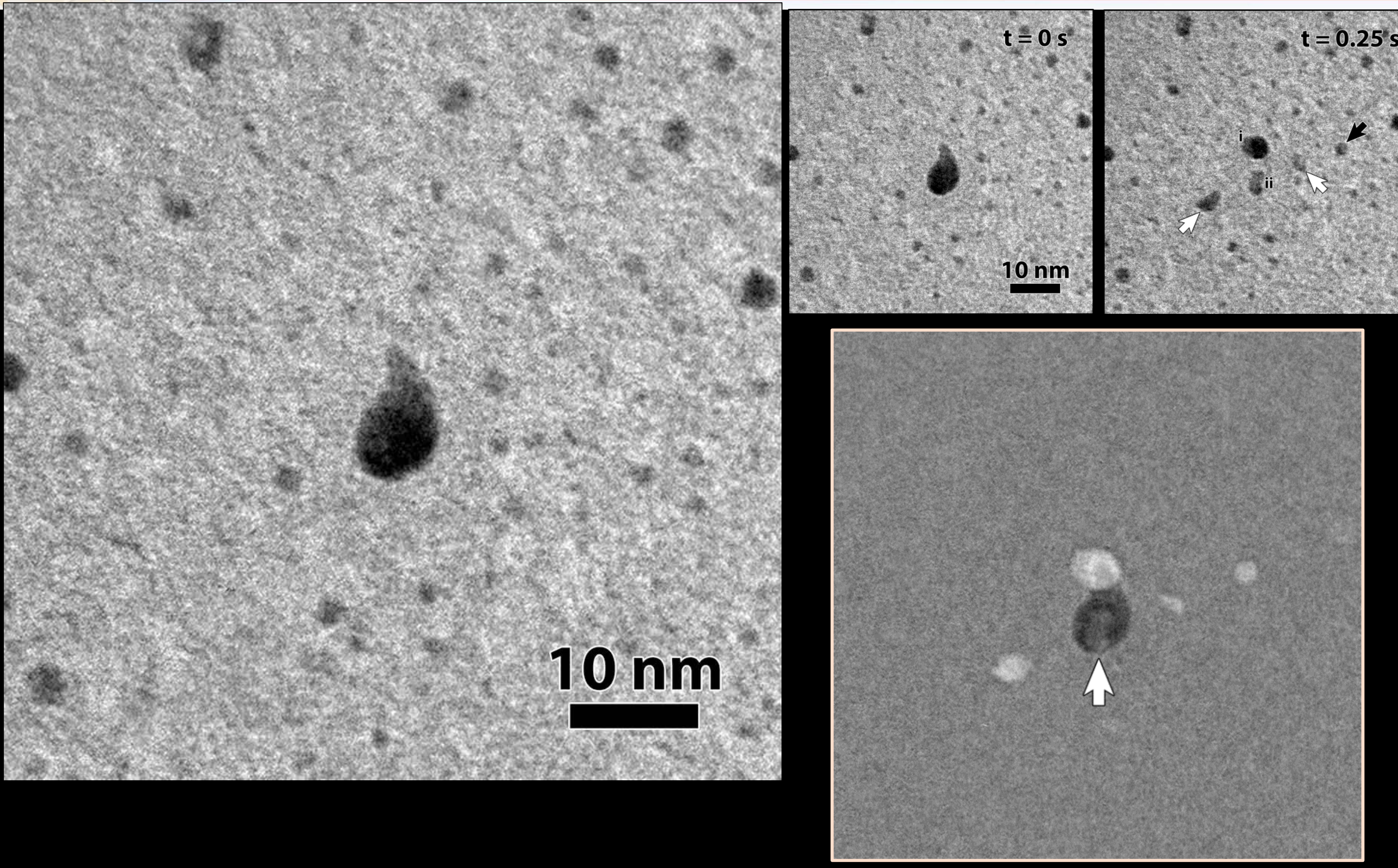


Irradiation Creep
(Loading rate 0.6 Mpa s^{-1})

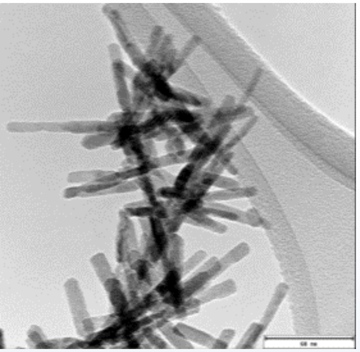


Single Ion Strikes:
46 keV Au^{1-} ions into 5 nm Au nanoparticles

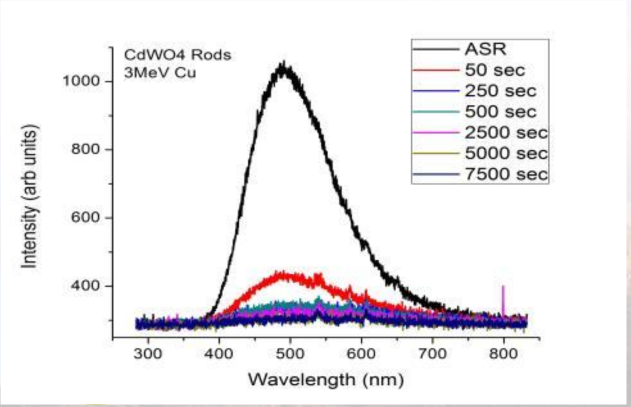
Collaborator: D.C. Bufford



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

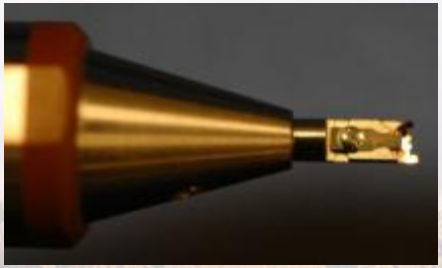


High-Z nanoparticles ($CdWO_4$) are promising, but are radiation sensitive

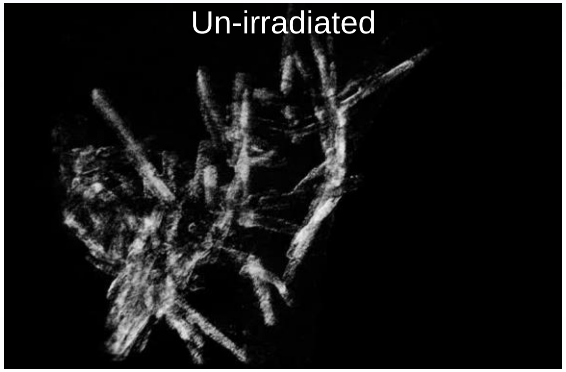


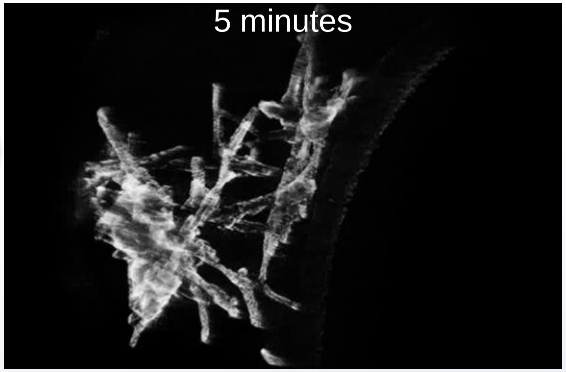
In situ Ion Irradiation TEM (I^3TEM)

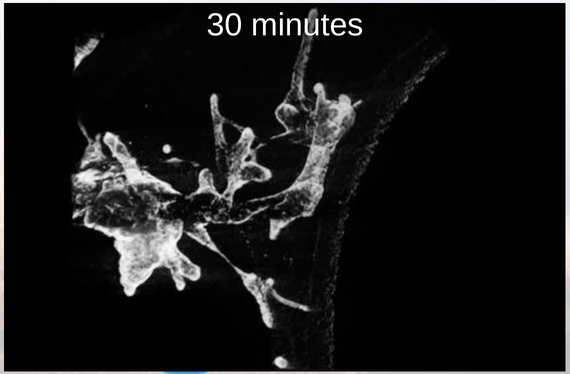
Hummingbird tomography stage



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



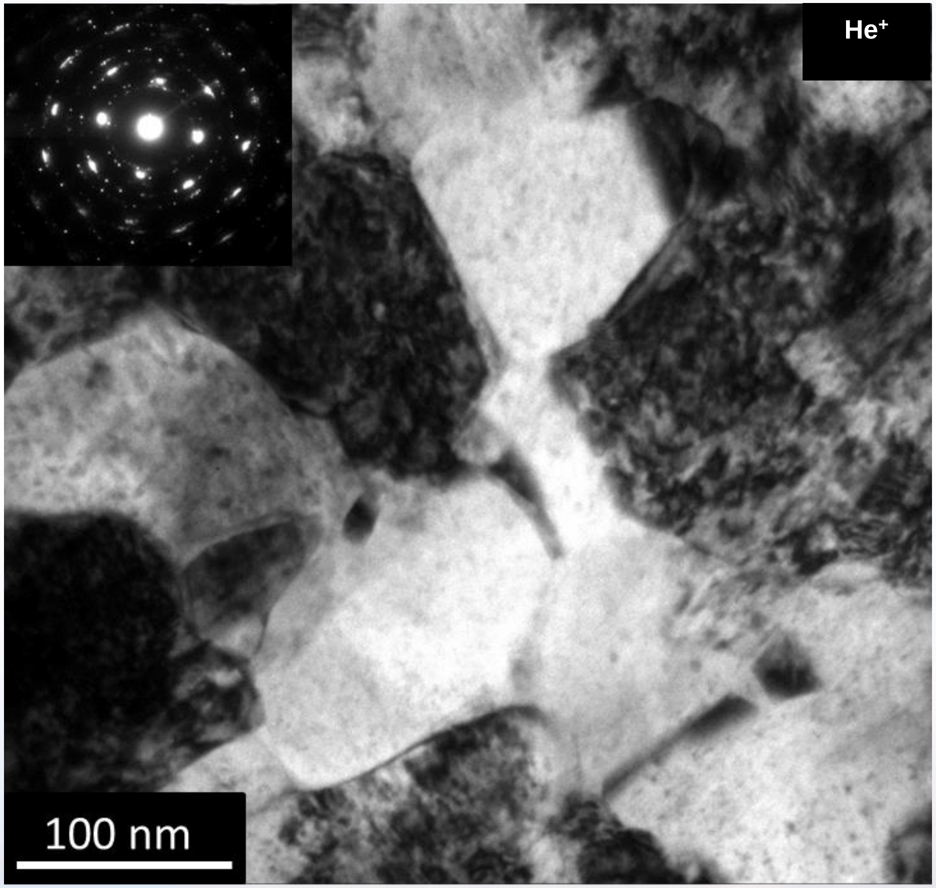


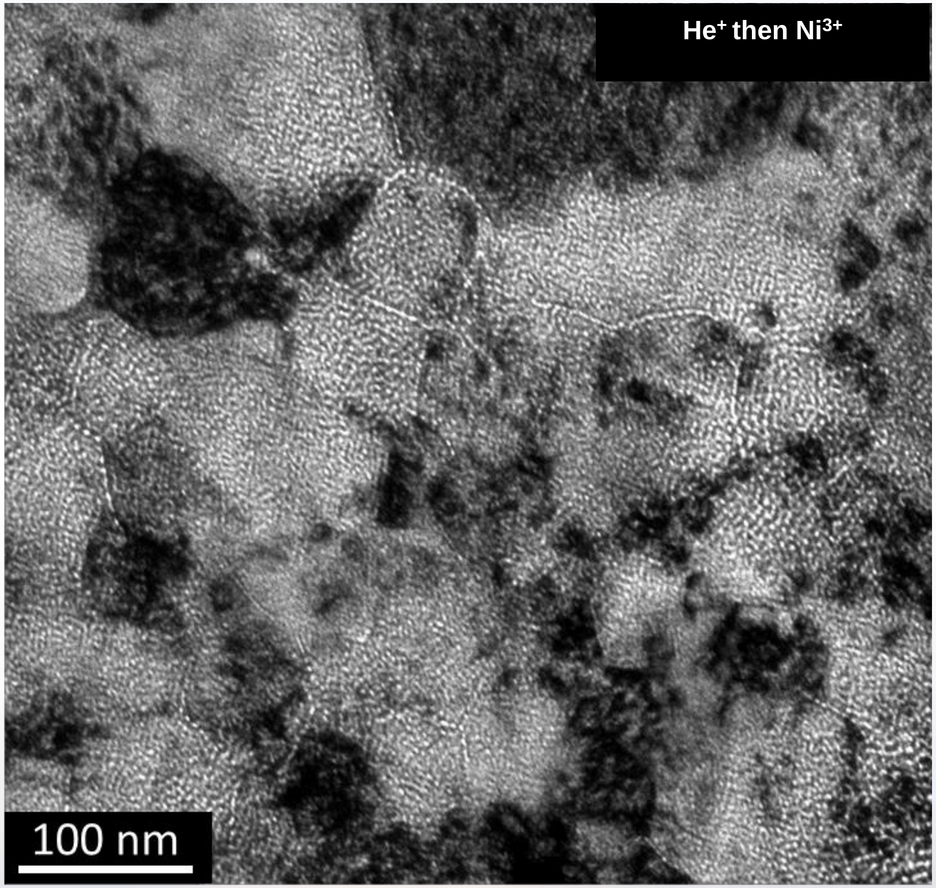


Contributors: S.M. Hoppe, P.A. Hernandez-Sanchez, T. Boyle

10 keV He^+ Implantation followed by 3 MeV Ni^{3+} Irradiation

Collaborator: B. Muntifering & J. Qu





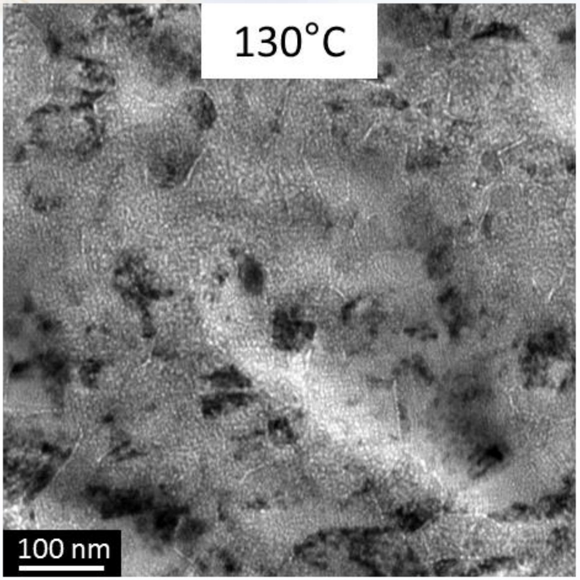
10^{17} He^+/cm^2

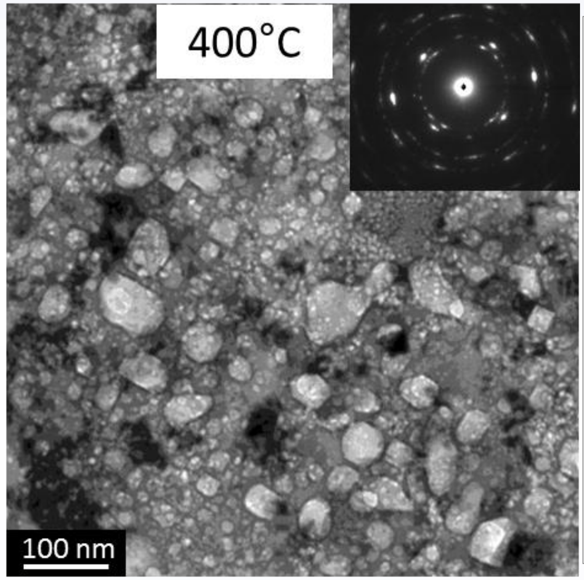
Visible damage to the sample

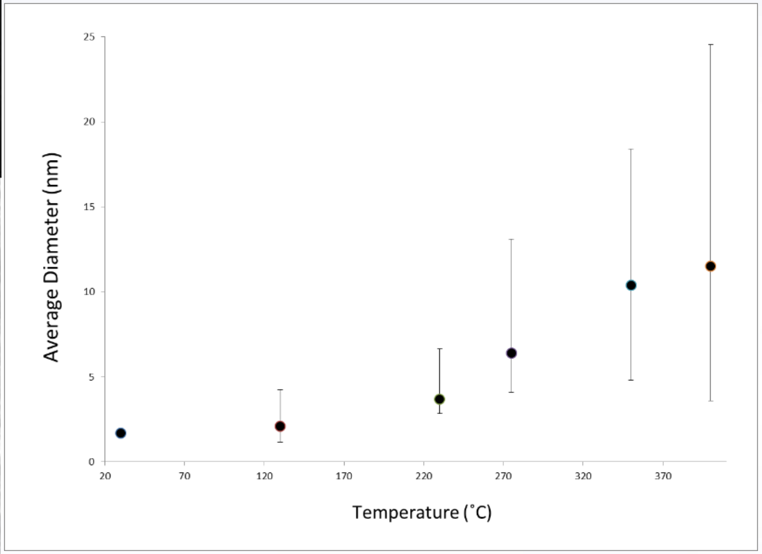
0.7 dpa Ni^{3+} irradiation

High concentration of cavities along grain boundaries

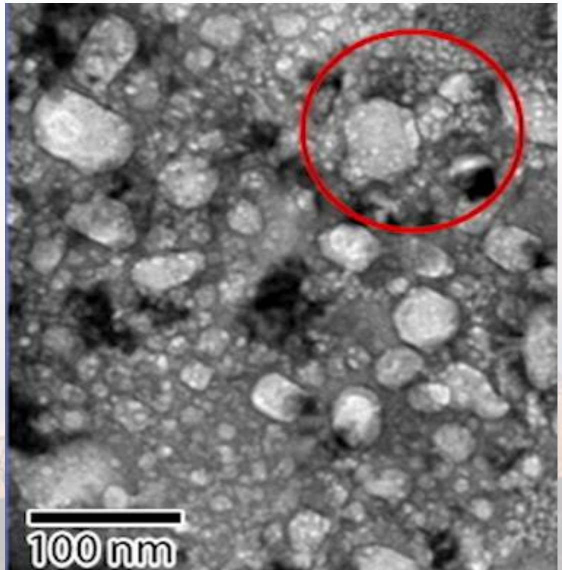
Cavity Growth during In-situ Annealing of 10 keV He^{+} Implanted and then 3 MeV Irradiated Ni^{3+}

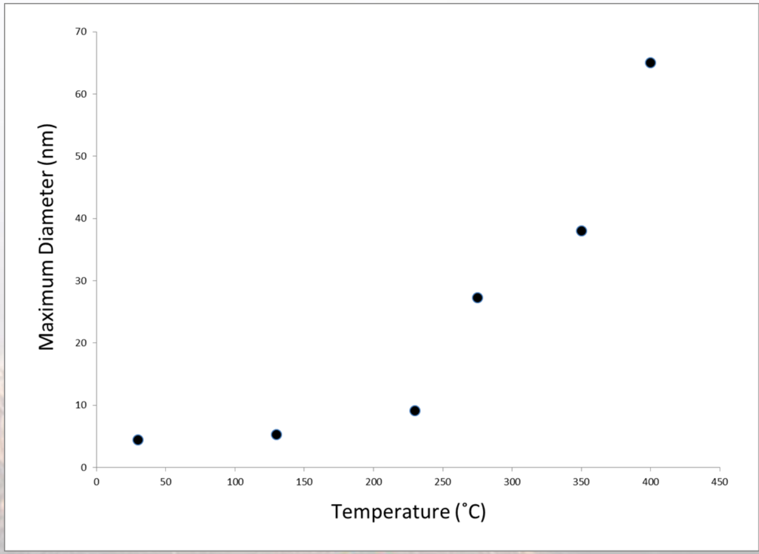






Bubble to cavity transition and cavity evolution can be directly studied

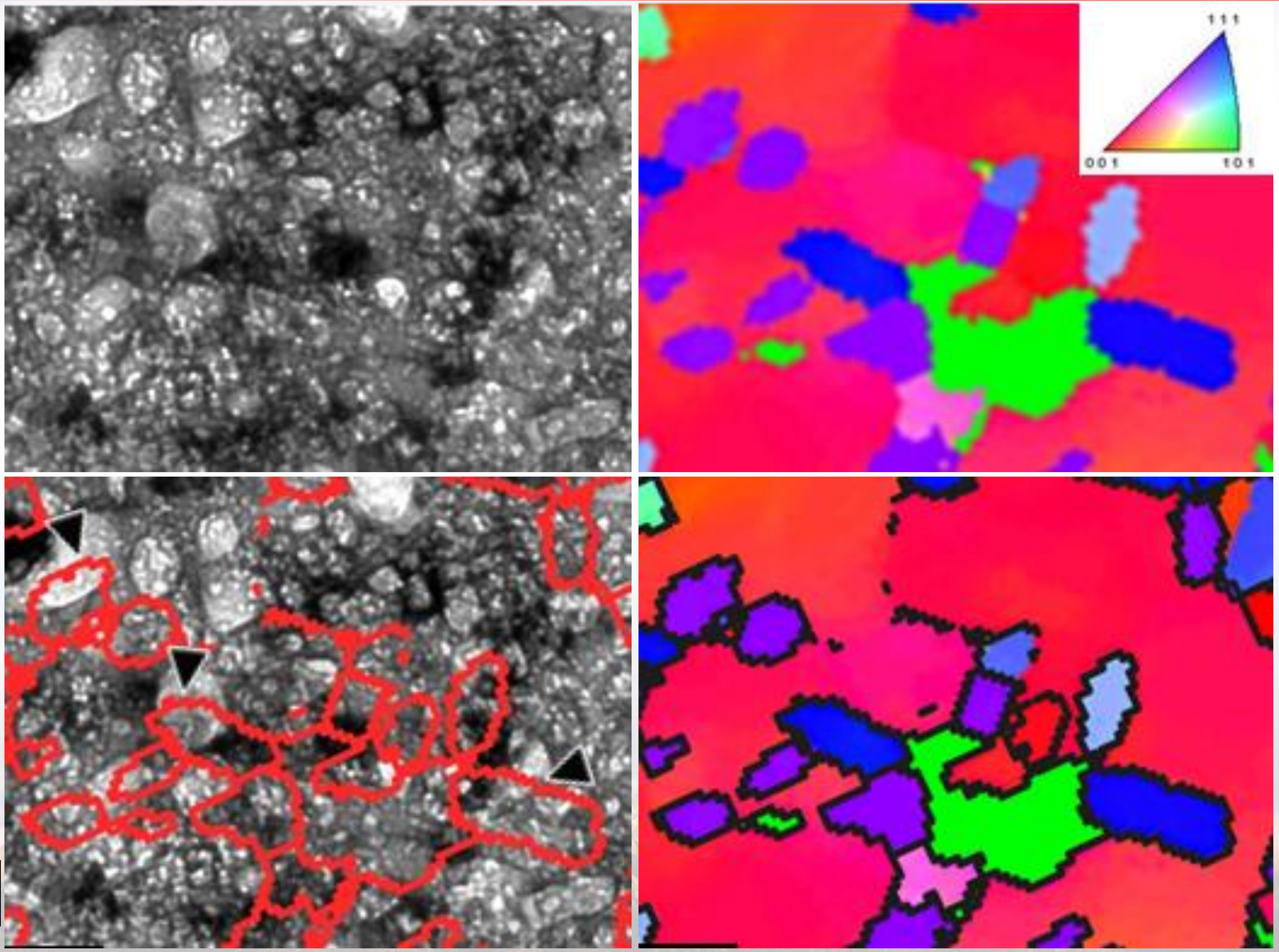




Precession Electron Diffraction Reveals Hidden Grain Structure

Cavities in helium implanted, self-ion irradiated,

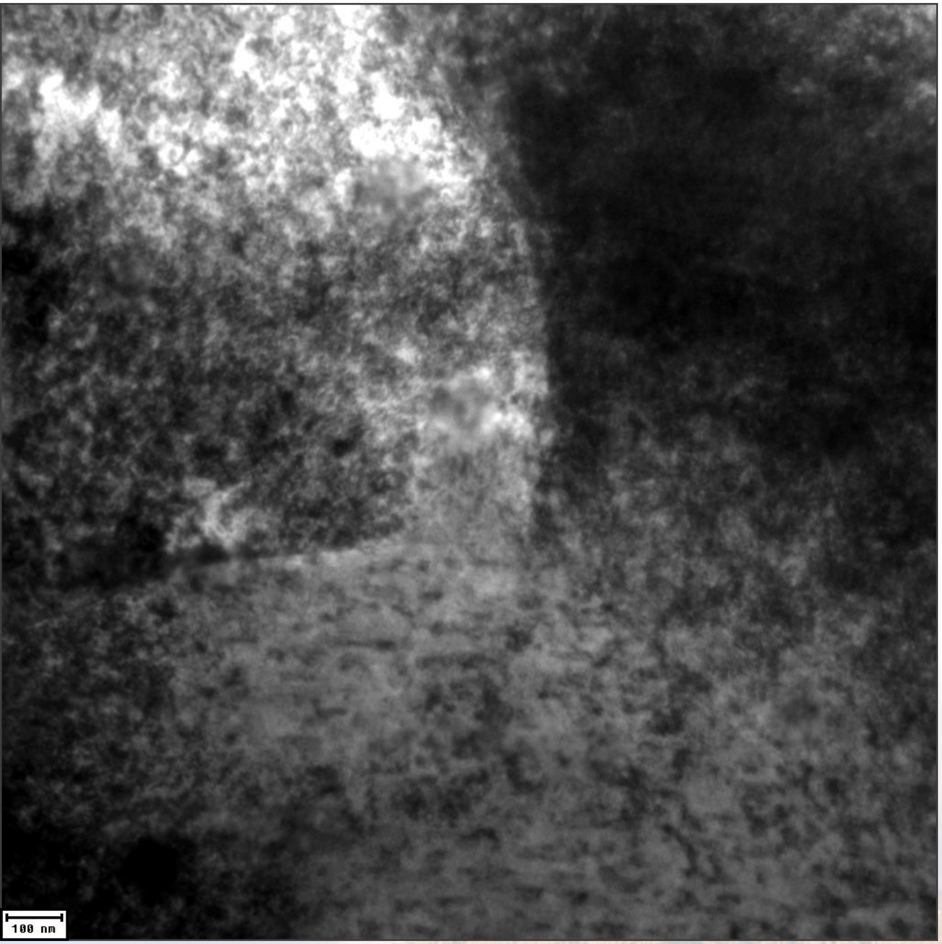
nc nickel film annealed to 400 ˚C

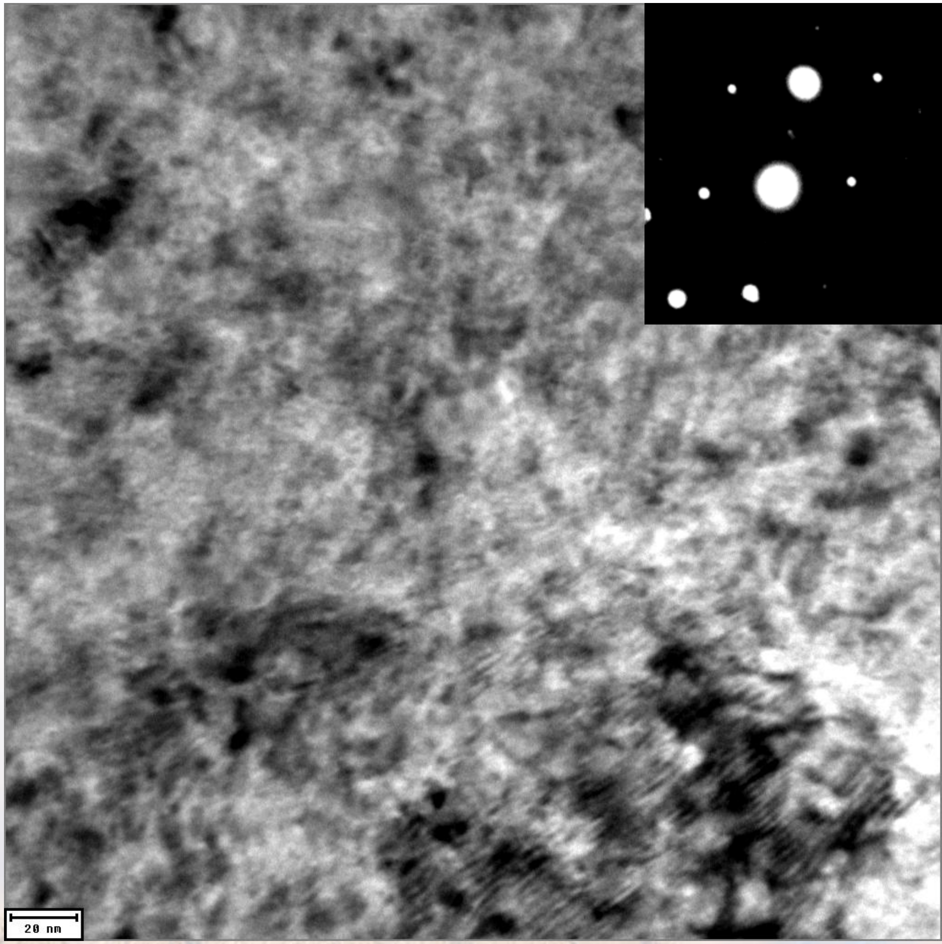


Cavities span multiple grains at identified grain boundaries

100 nm

Concurrent D & He Implantation & Zr Irradiation into Zr-4 at 310 C

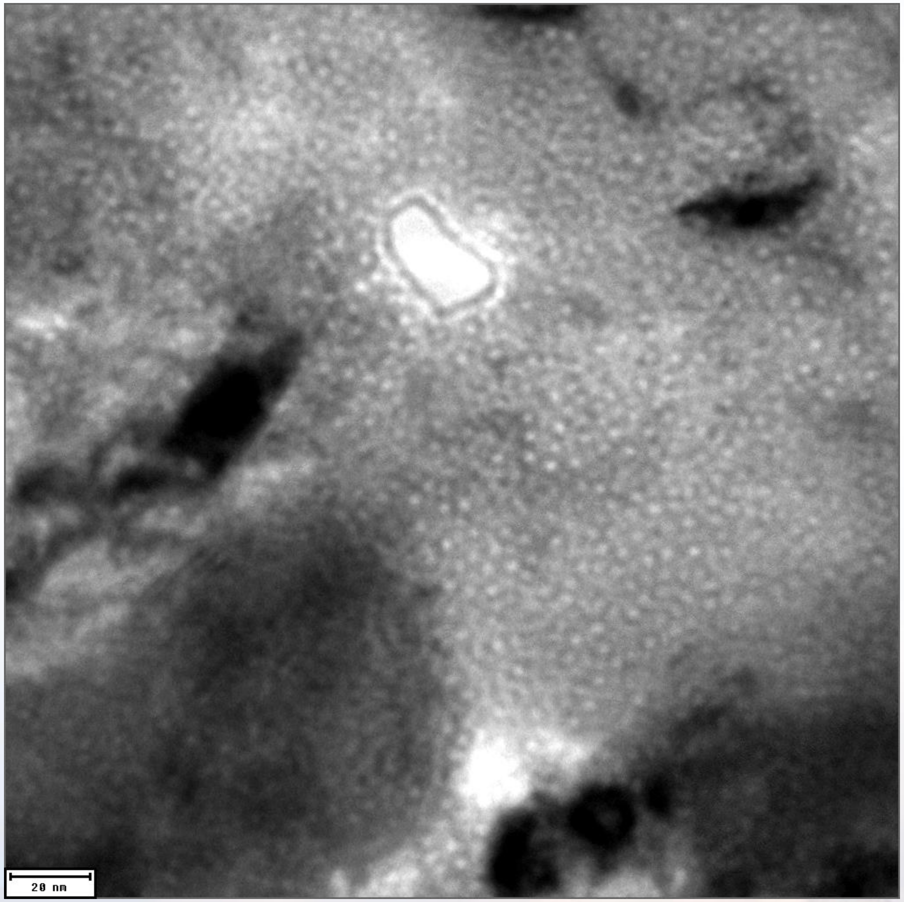


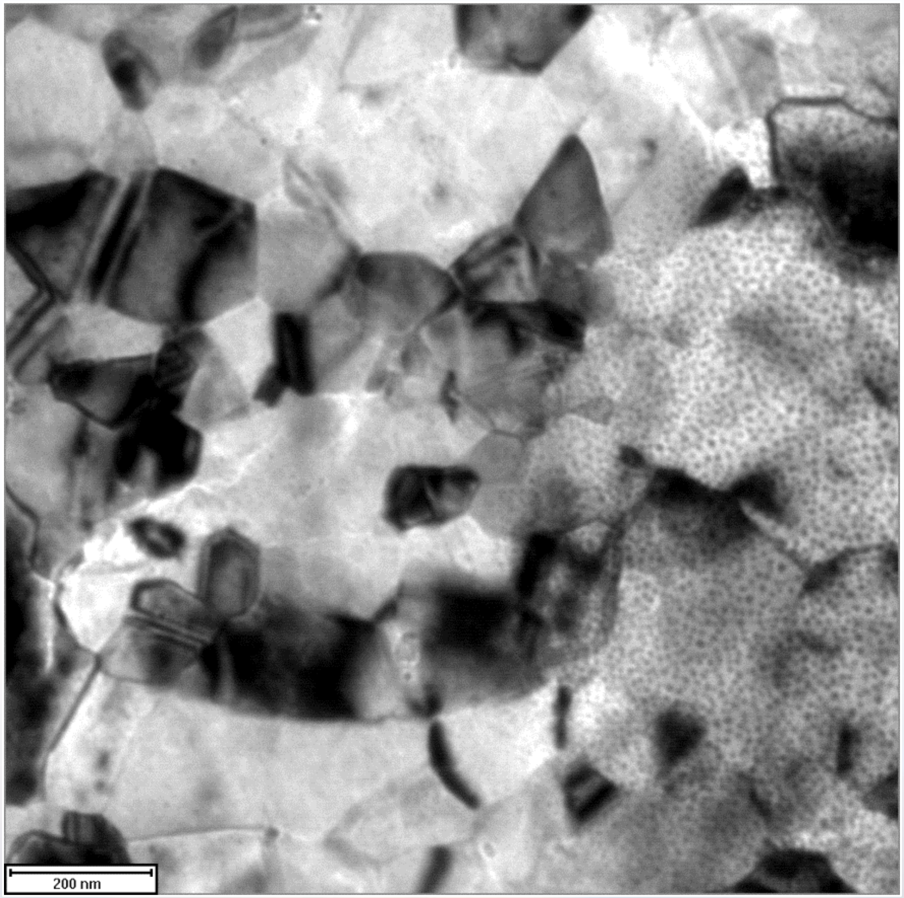


After Implantation/Irradiation
Significant Damage, but No Cavities

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

Cautionary Tale:
In situ Effects Should Never Be Assumed Negligible



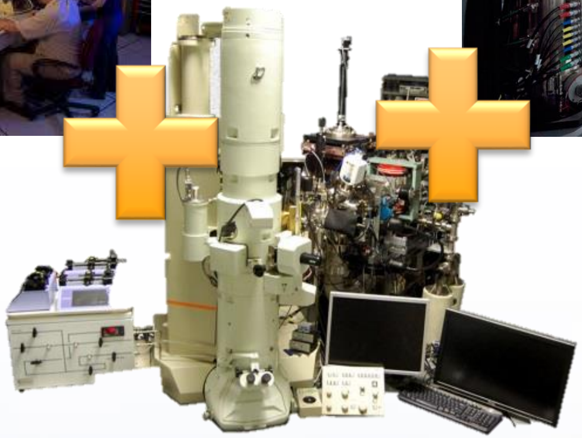


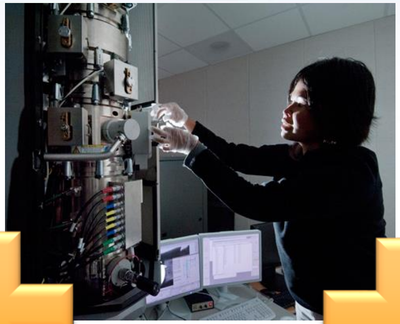
Post Examination: Through Focus Images 30 Days After the Concurrent *In situ* D & He Implantation & Zr Irradiation into Zr-4 at 310 C

Post Examination: Low Magnification After Annealing 400 °C and He^{+} + Ni^{3+}

Cautionary Tale:
“Johannes Factotum”

- 1592 Robert Greene *Groats-Worth of Wit*





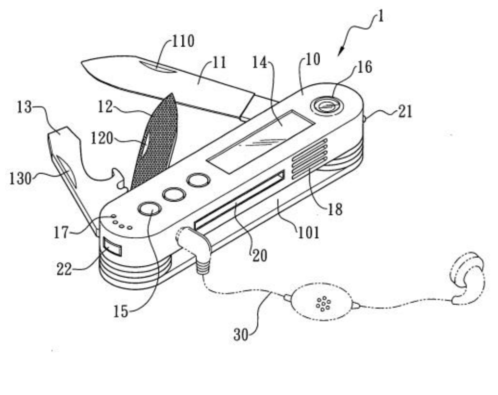


FIG. 3

M.-H. Yeh, “Knife Strucutre” US20060087845 A1

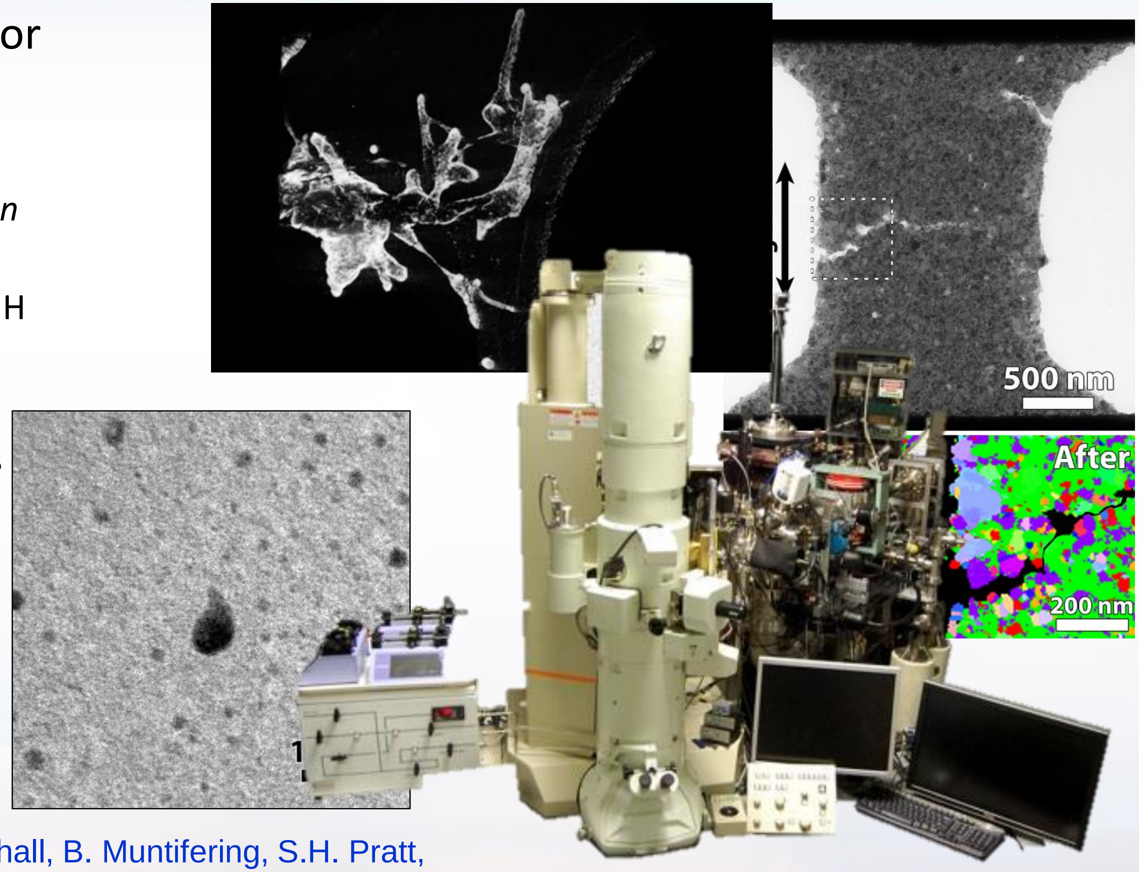
Chimera, Bellerophon and Pegasus, Athenian black-figure siana cup C6th B.C., Musée du Louvre

Can a tool do everything & Do anything well?

Summary

- Sandia's I^{3}TEM is a unique facility for understanding mechanisms at the nanoscale
  - A facility with a wealth of overlapping *in situ ion irradiation* capabilities
  - *In situ* high energy ion irradiation from H to Au
  - *In situ* gas implantation
  - 15 TEM stages with various capabilities
    - Liquid, gas, high temperature, cryo, tomography, straining

What is the limit to integrating in-situ and analytical TEM techniques?



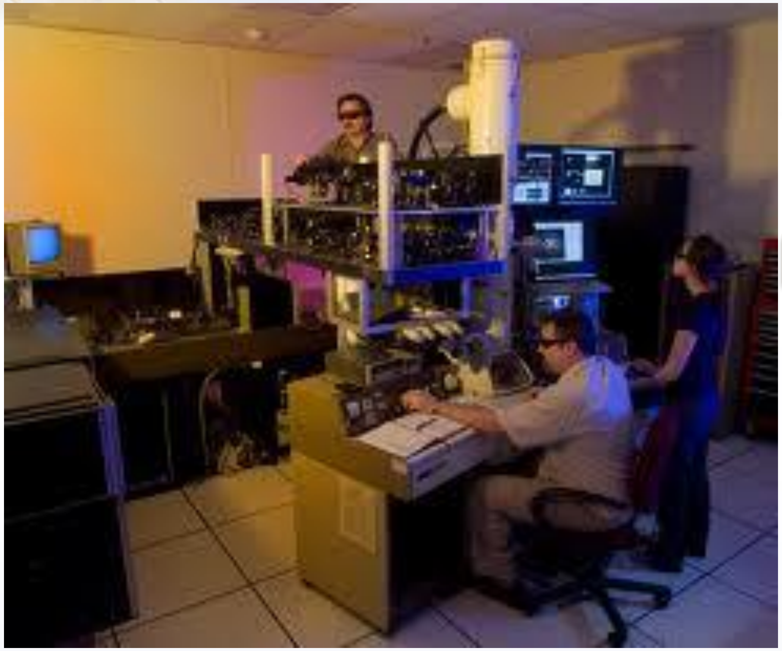
Collaborators:

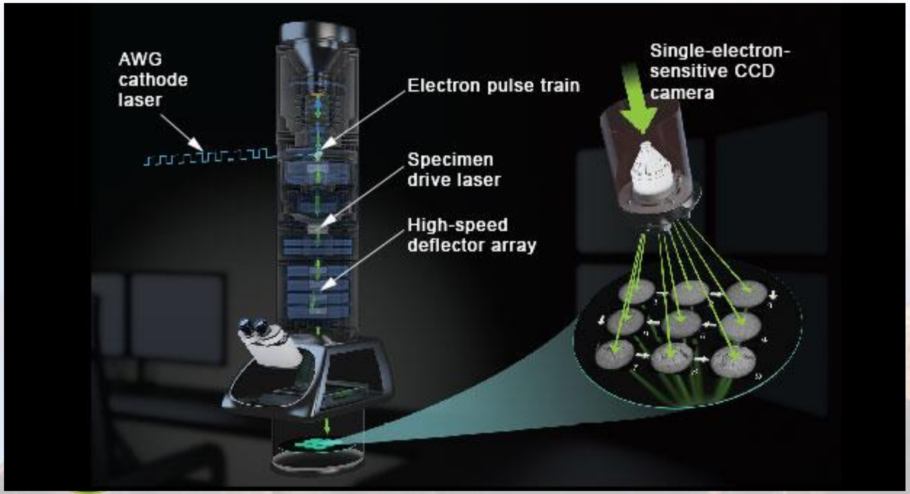
- IBL: D. Buller, C. Chisholm, B.G. Clark, M.T. Marshall, B. Muntifering, S.H. Pratt,
- Sandia: M. Abere, B. Boyce, T.J. Boyle, R. Dingreville, R.F. Hess, A.C. Kilgo, B.E. Klamm, W.M. Mook, J.D. Puskar, J.A. Scott, & J.A. Sharon
- External: A. Aitkaliyeva, H. Bei, P.J. Ferreira, K.J. Ganesh, E.P. George, D. Gross, P. Hosemann, J. Kacher, S. Maloy, A. Minor, J. Qu, S. Rajesekhara, I.M. Robertson, D. Stauffer, & Hysitron Inc.

This work was partially funded by the Division of Materials Science and Engineering, Office of Basic Energy Sciences, U.S. Department of Energy.
Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Back-up Slides

Can I^3TEM and DTEM systems be combined?



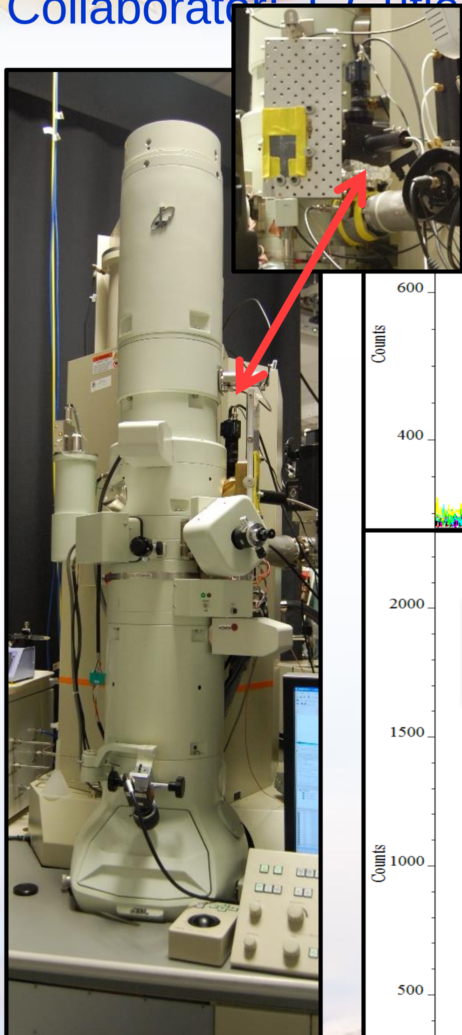


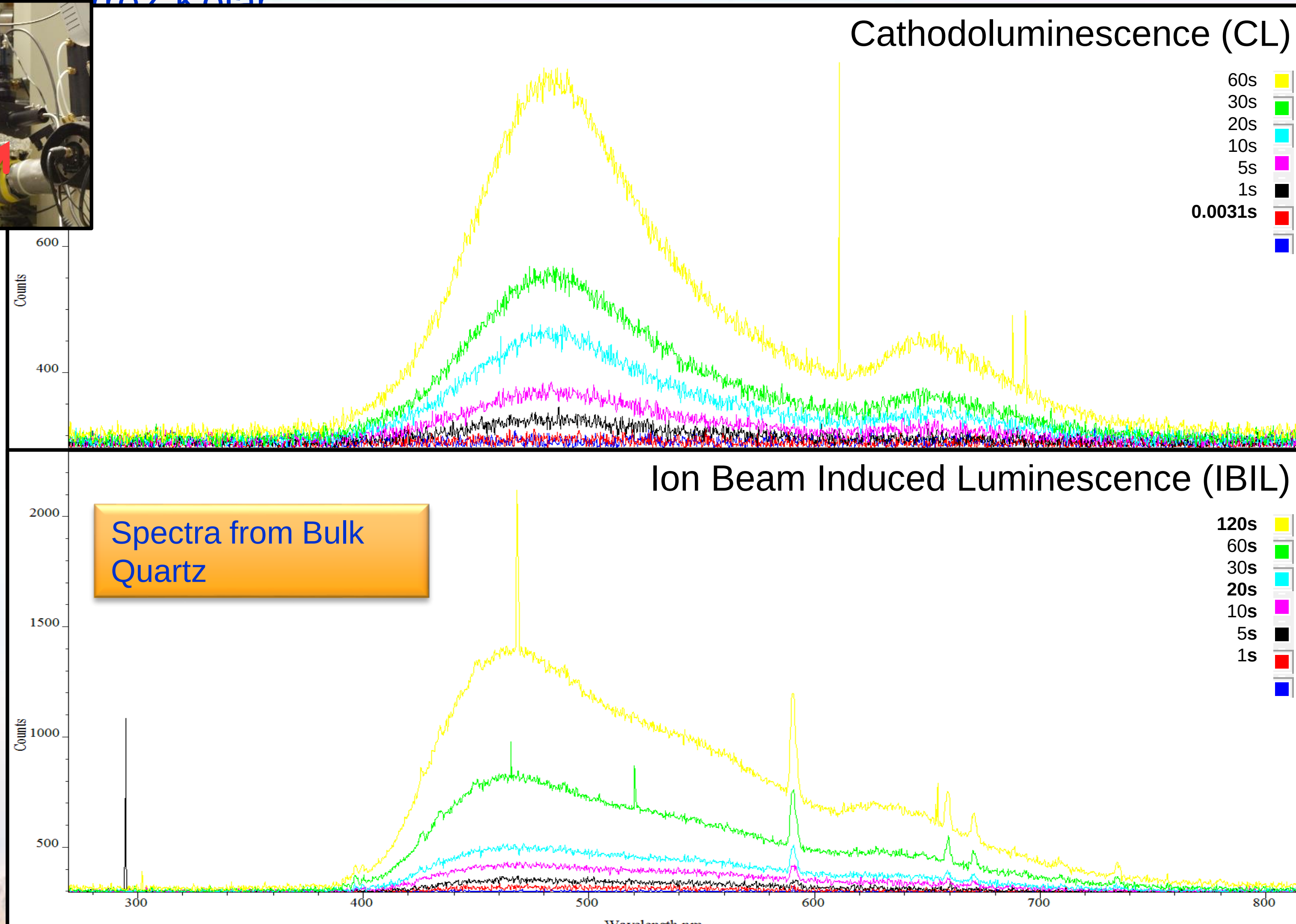
Goal:

To combine the state-of-the-art in microscopy of DTEM and I^3TEM to elucidate the response of single radiation events and other extreme overlapping environments with adequate spatial and temporal resolution.

Future Direction: *In situ* TEM Ion beam Induced Luminescence (IBIL)

Collaborator: J. Gutierrez-Kolar

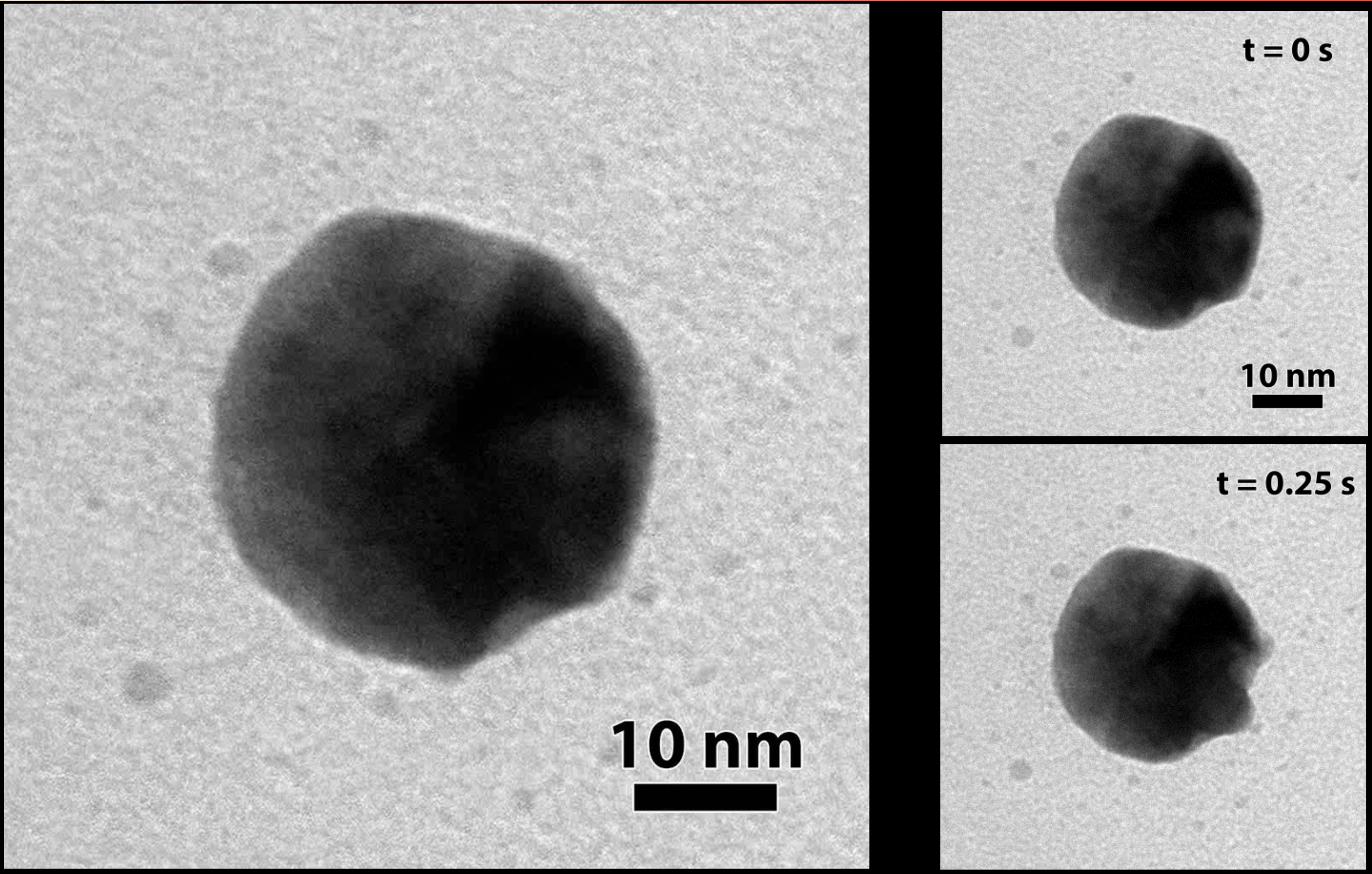




Significant optimization is still needed; potential is promising

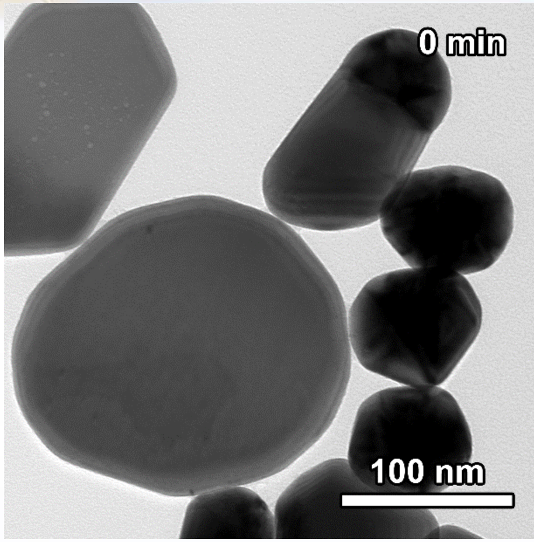
Single Ion Effects with 46 keV Au^{1-} ions: 20 nm

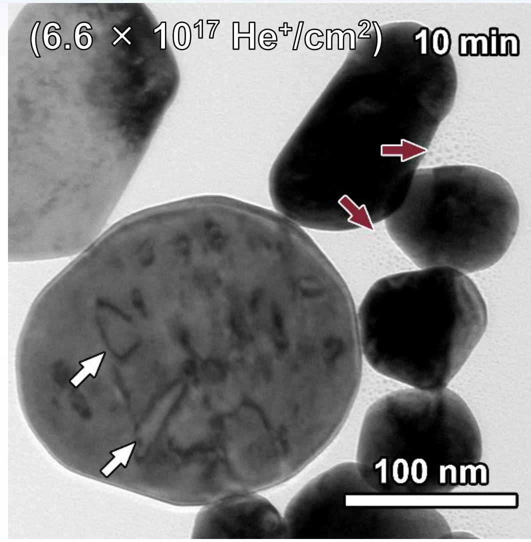
Collaborator: D.C. Bufford

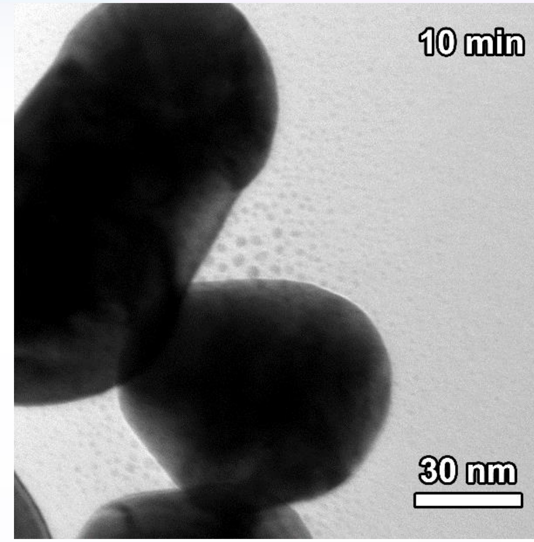


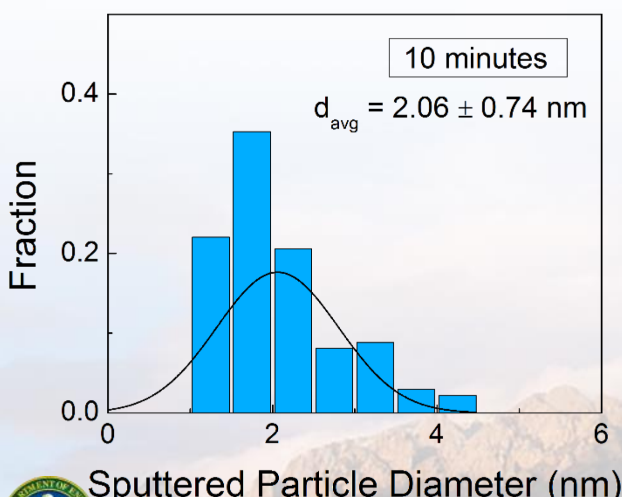
Formation of Dislocation Loops & Sputtered Particles due to He implantation

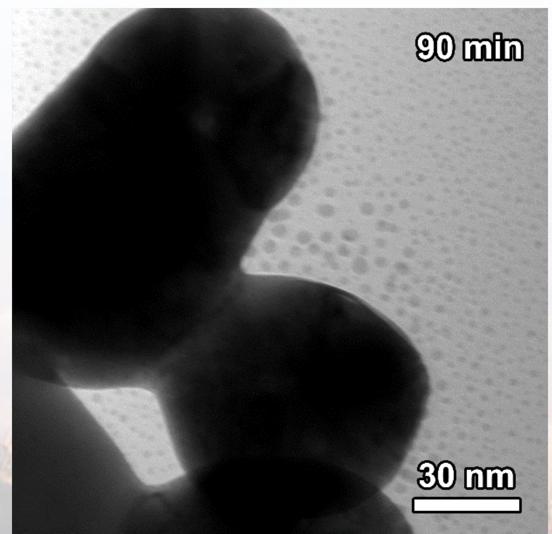
Collaborators: D.C. Bufford, S.H. Pratt & T.J. Boyle

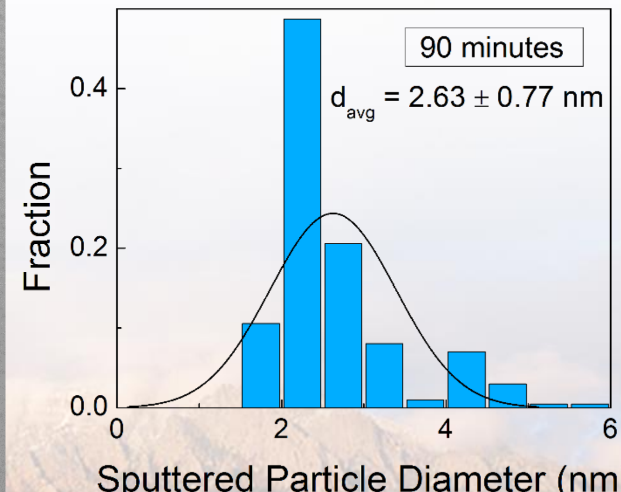








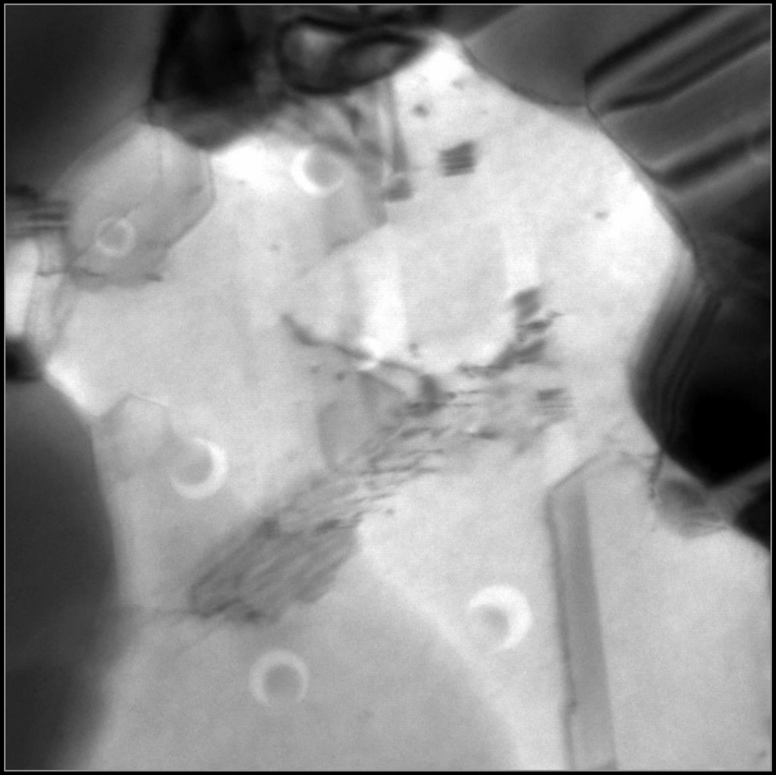




Dose Rate Effects

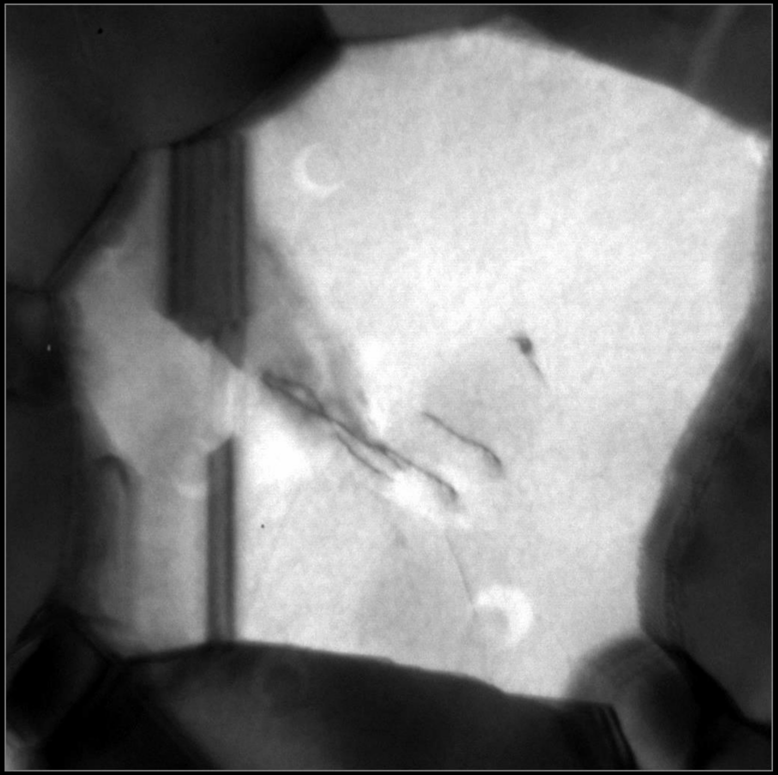
Collaborators: C. Chisholm , P. Hosemann, & A. Minor

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



VS

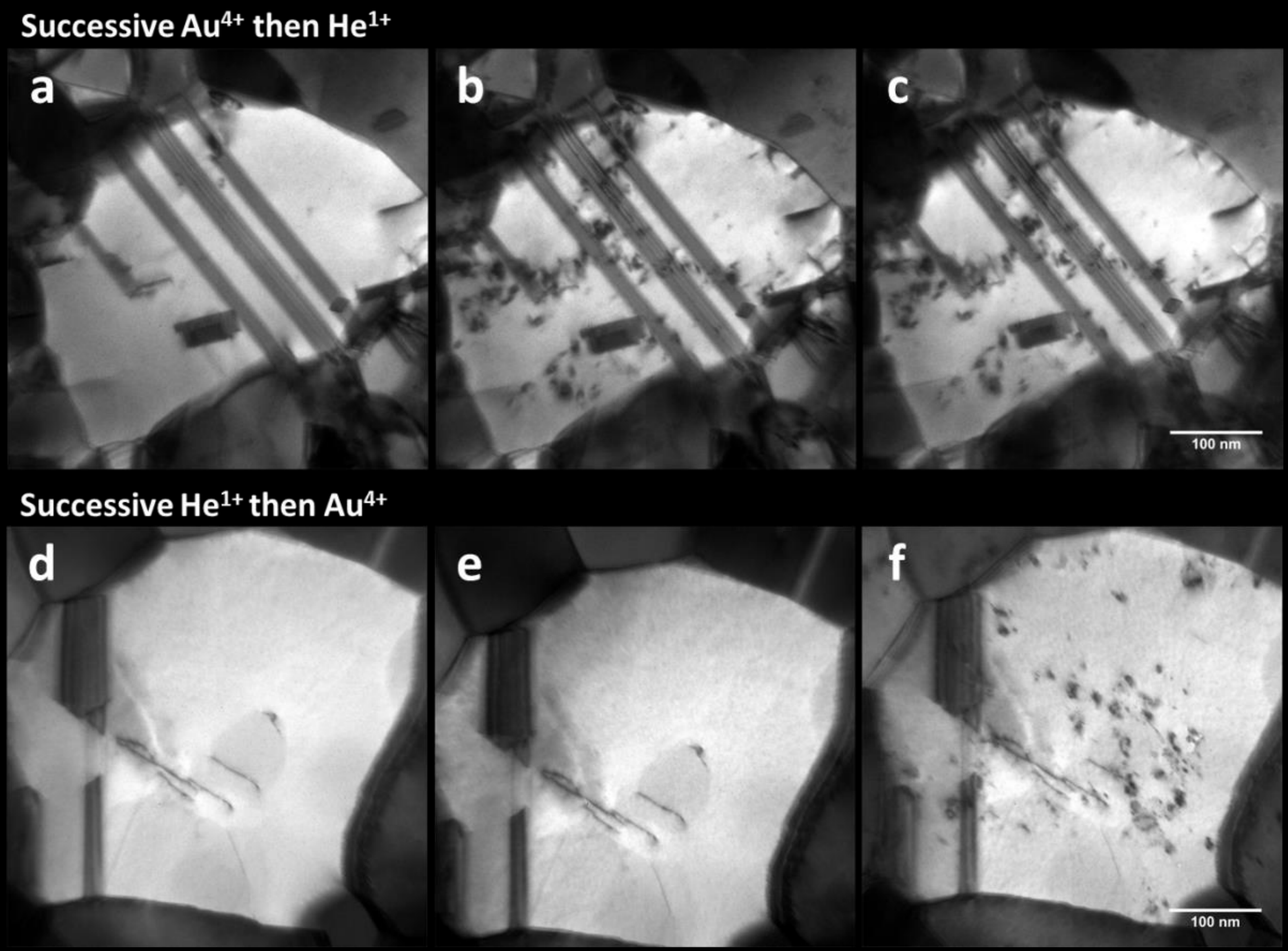
6.7 x 10^7 ions/cm^2/s



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

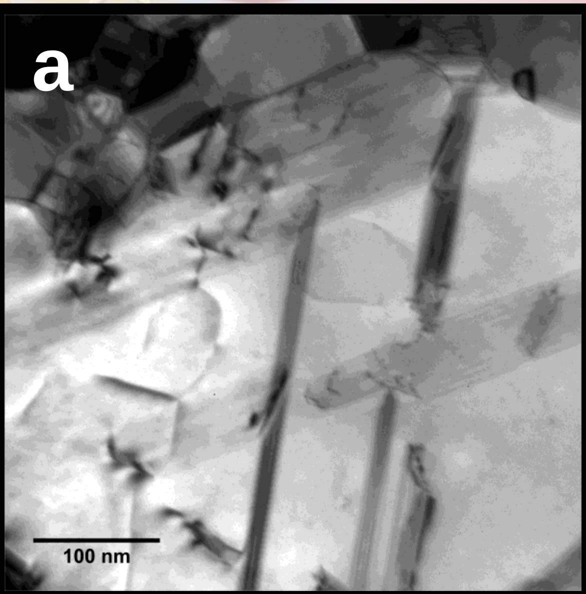
In situ Successive Implantation & Irradiation

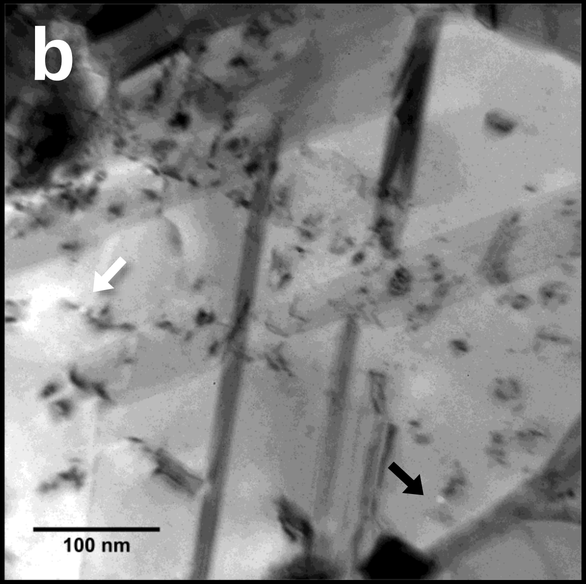
Collaborators: C. Chisholm & A. Minor



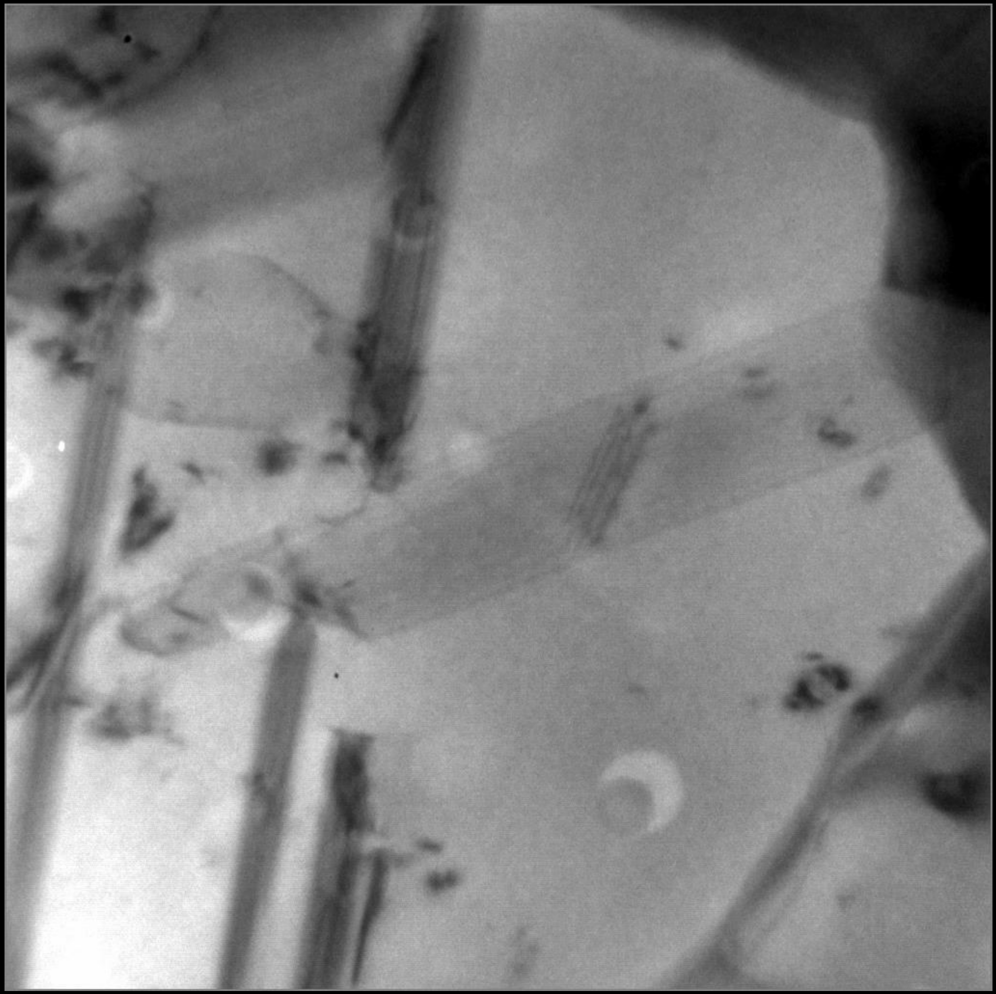
In situ Concurrent Implantation & Irradiation

Collaborators: C. Chisholm & A. Minor





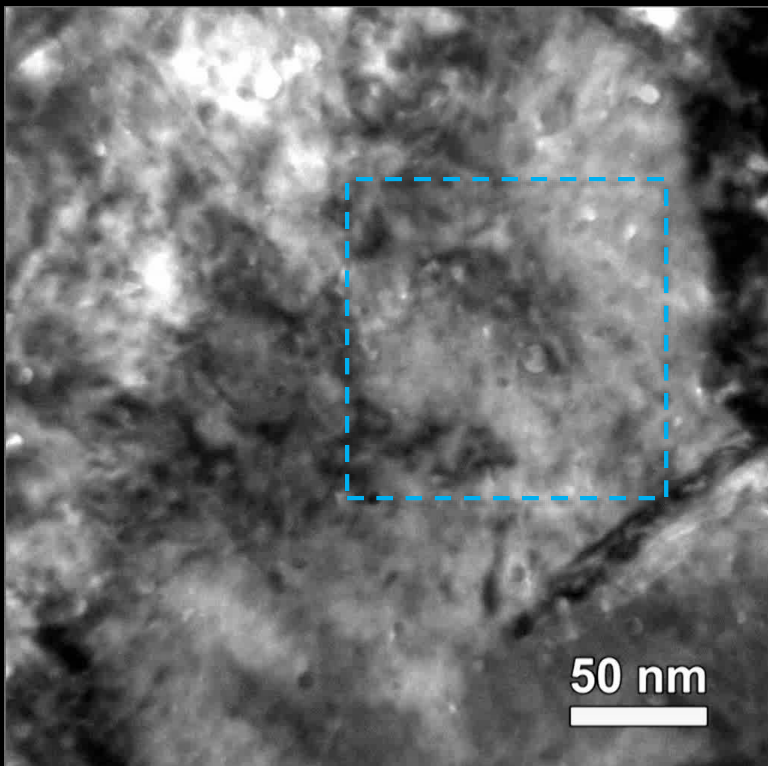
He^{1+} implantation and Au^{4+} irradiation of a gold thin film

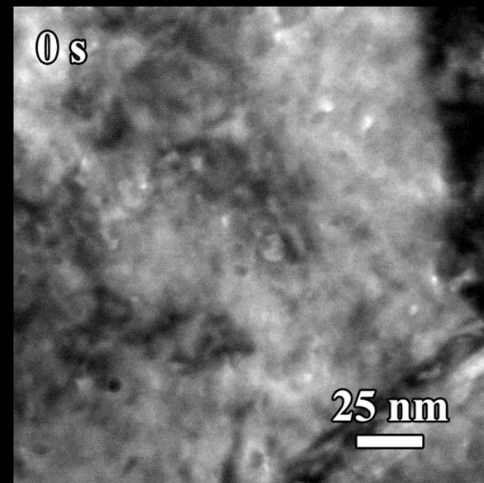


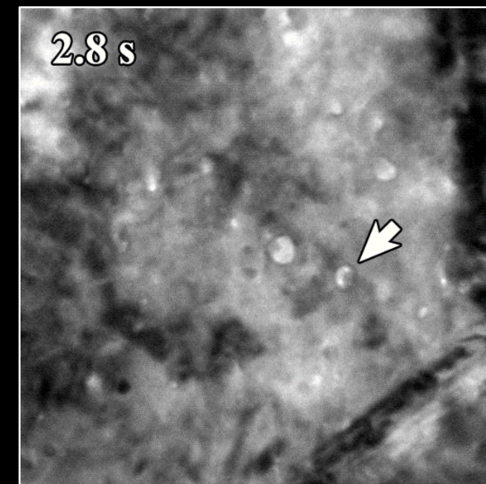
Simultaneous *In situ* TEM Triple Beam: 2.8 MeV Au^{4+} + 10 keV He^{+}/D_2^{+}

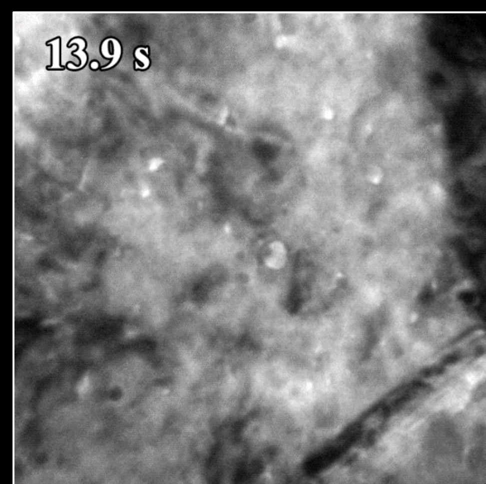
Collaborator: D.C. Bufford

Video playback speed x1.5.







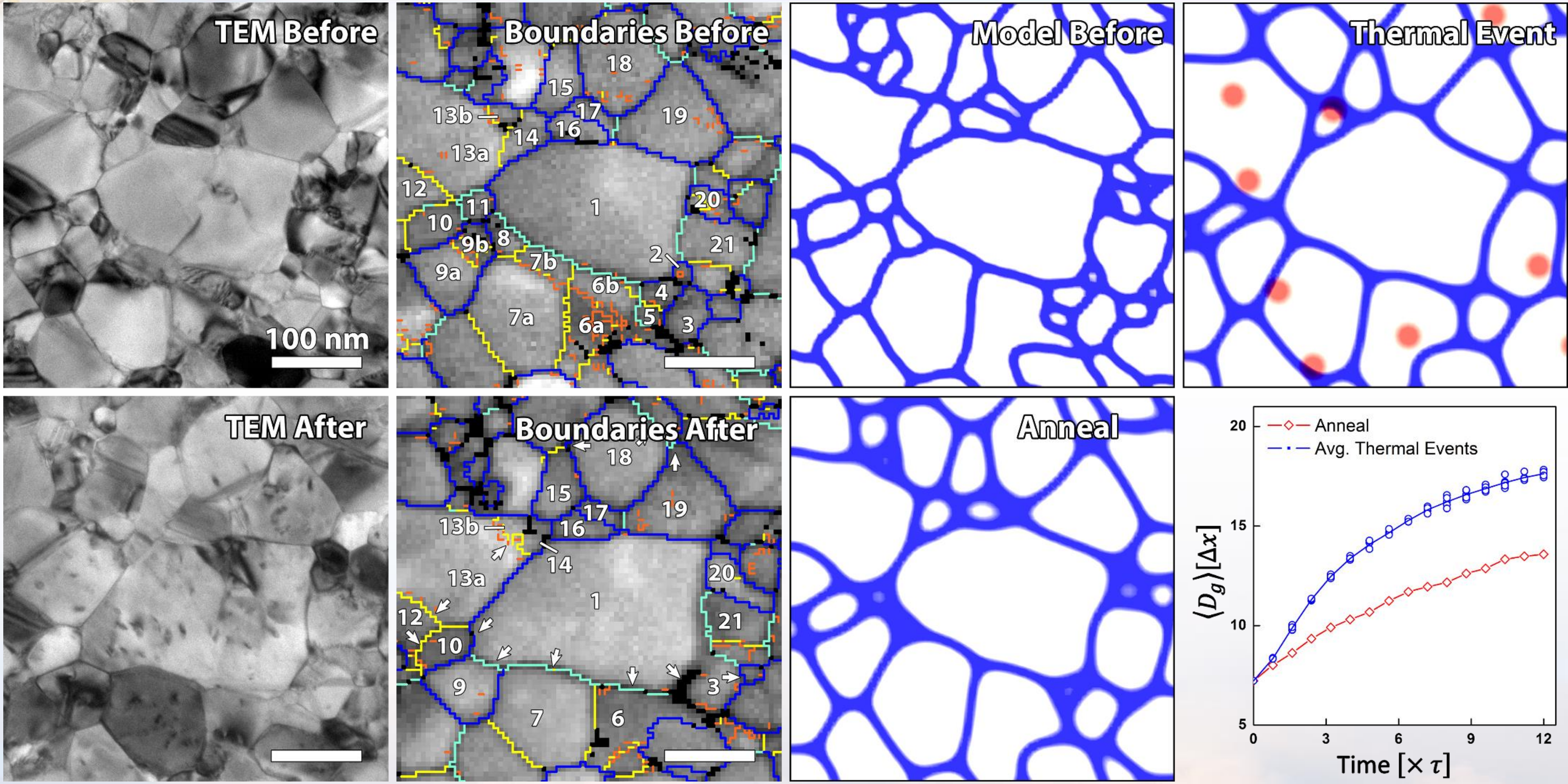


- **Approximate fluence:**
  - Au 1.2 $\times 10^{13}$ ions/cm^2
  - He 1.3 $\times 10^{15}$ ions/cm^2
  - D 2.2 $\times 10^{15}$ ions/cm^2
- **Cavity nucleation and disappearance**

In-situ triple beam He, D_2, and Au beam irradiation has been demonstrated on Sandia's I^3TEM! Intensive work is still needed to understand the defect structure evolution that has been observed.

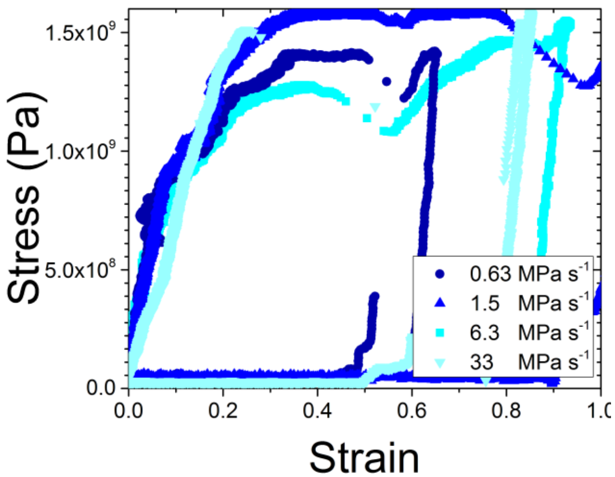
Direct Comparison to Mesoscale Modeling

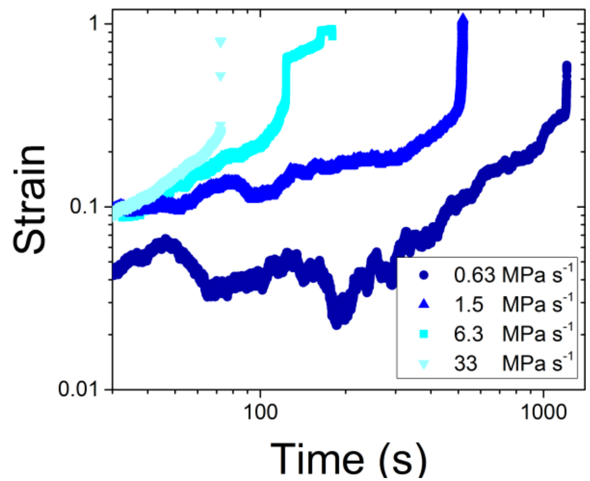
Collaborators: D.C. Bufford, F. Abdeljawad, & S.M. Foiles

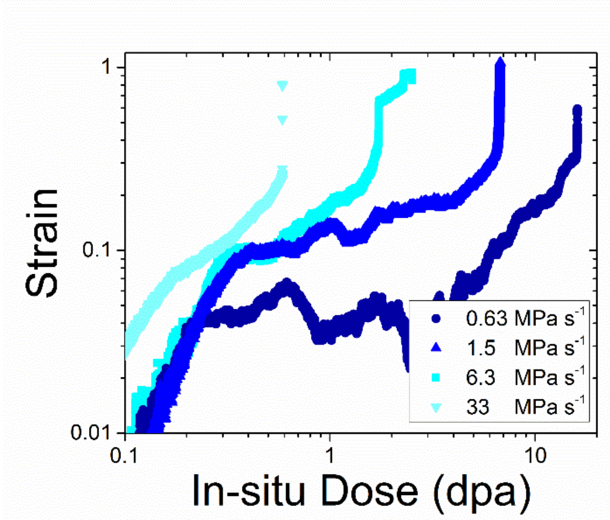


Because of the matching length scale, the initial microstructure can serve as direct input to either MD of mesoscale models & subsequent structural evolution can be directly compared.

Creep Response at Different Loading Rates



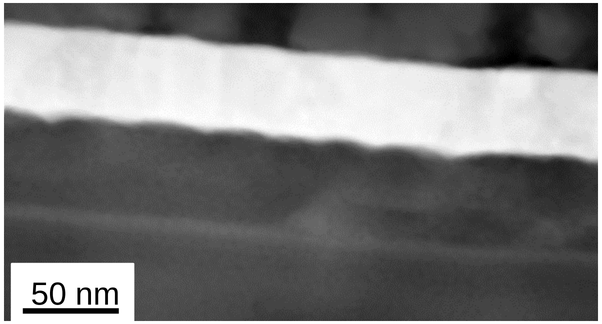




Significant creep observed at a fraction of the bulk yield strength

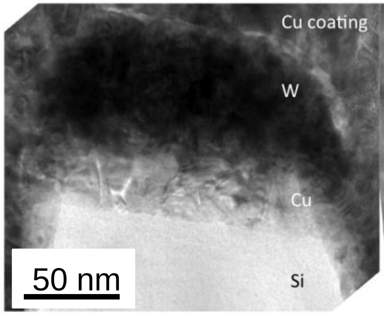
As-deposited Sample

ADF-STEM

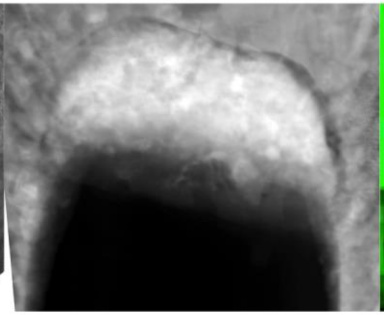


Post Creep Characterization

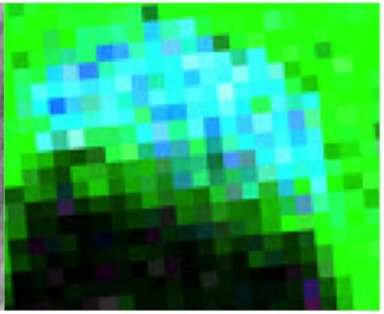
BF-TEM



ADF-STEM



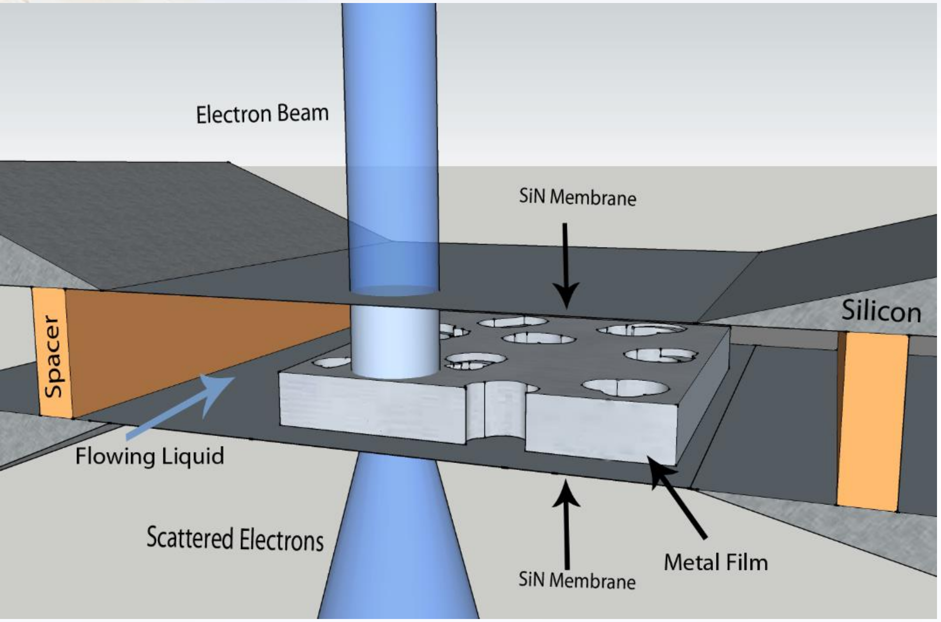
EDS Map (W, Cu, Si)

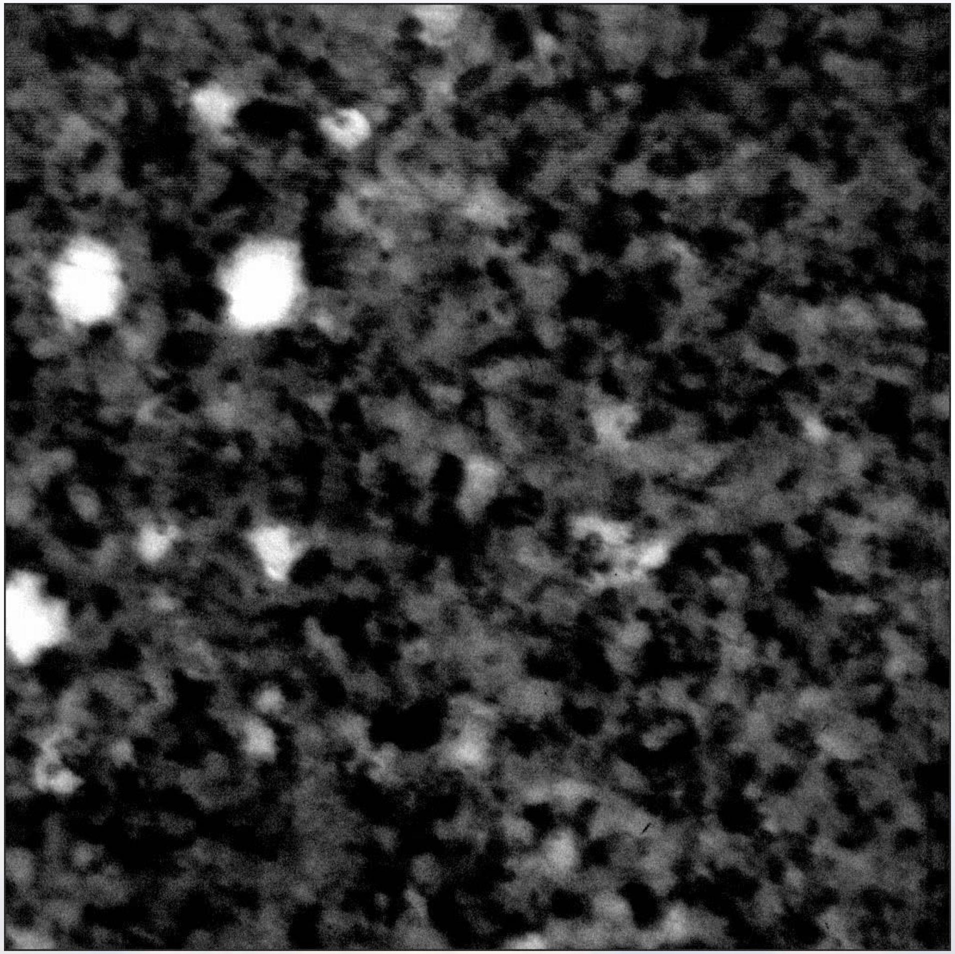


Compression (creep) only observed in Cu layer

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

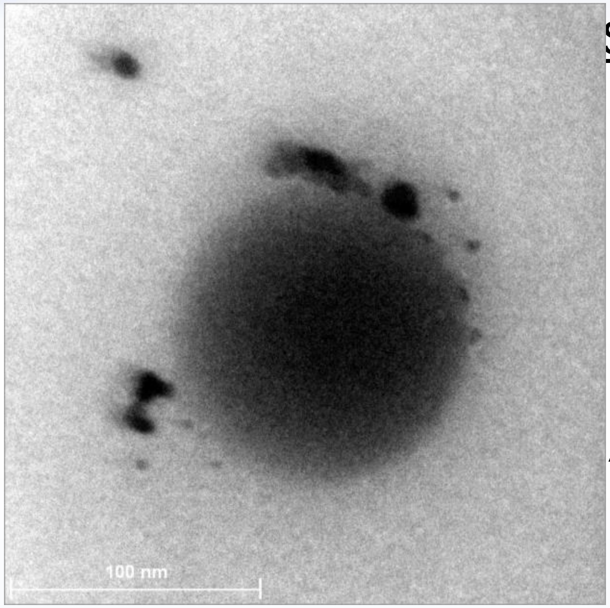
Pitting mechanisms during dilute flow of acetic acid over 99.95% nc-PLD Fe involves many grains.

Other Fun Uses of Microfluidic Cell

<u>Protocell Drug Delivery</u>

S. Hoppe, E. Carnes, J. Brinker

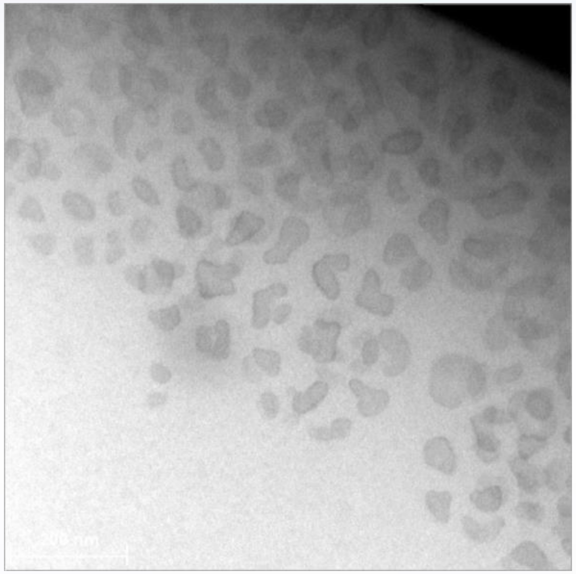
Liposome encapsulated Silica destroyed by the electron beam



<u>SA Crystallization</u>

S. Hoppe

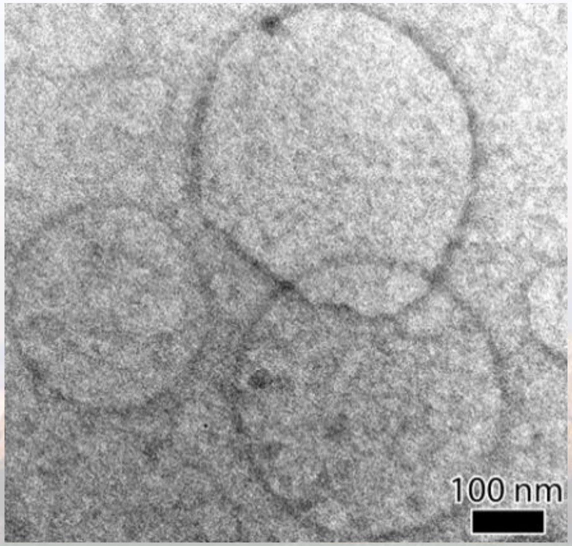
Crystallization of excess Bovine Serum Albumen during flow



<u>Liposomes in Water</u>

S. Hoppe, D. Sasaki

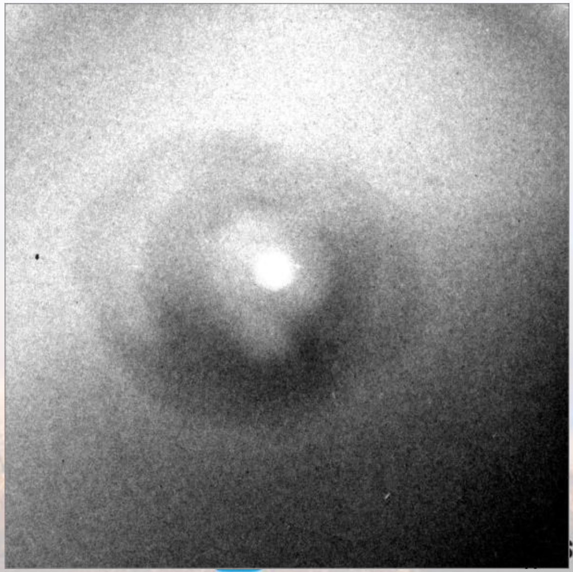
Liposomes imaged in flowing aqueous



<u>La Structure Formation</u>

S. Hoppe, T. Nenoff

La Nanostructure

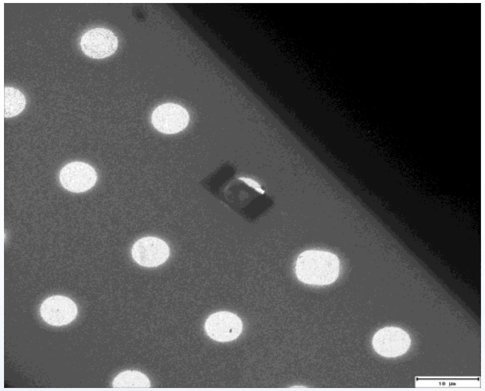


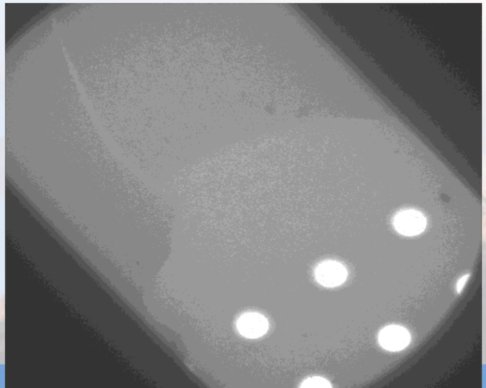
Feasibility of Studying Zircaloy 2 at Nominally 1 atm

Collaborators: S. Rajasekhara and B.G. Clark

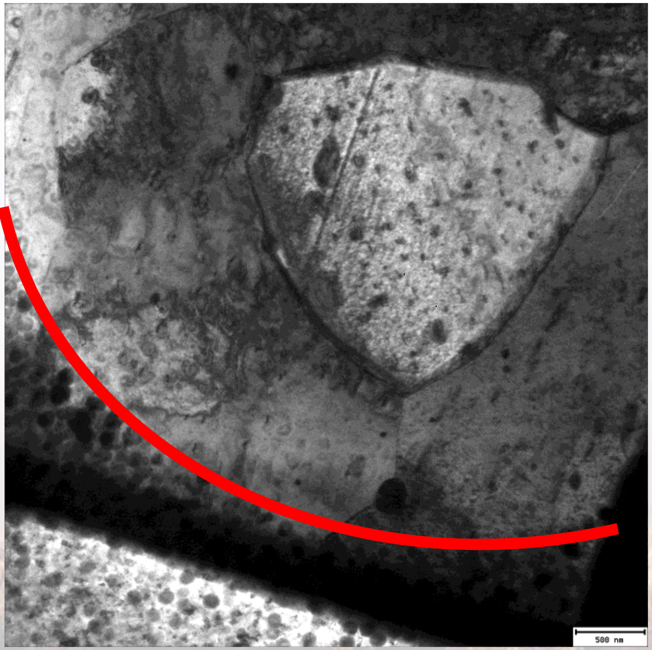
Vapor-Phase Heating TEM Stage

- 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

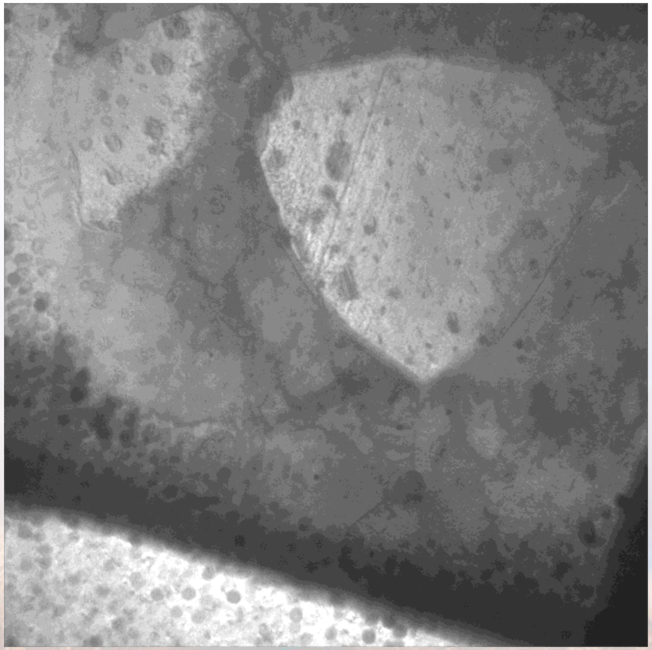




Vacuum & Single Window



Nominally 1 atm H_2 & Two Windows



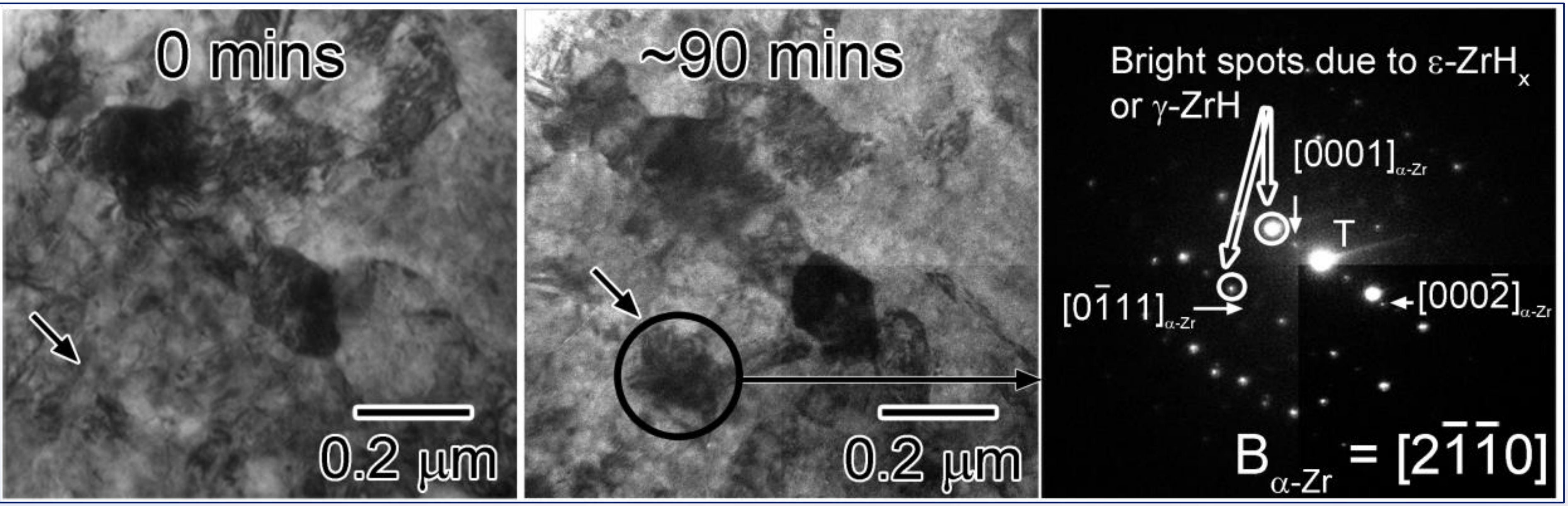
Most features are observed in both despite the decreased resolution resulting from the additional SiN window and 5 µm of air

In situ Observation of Hydride Formation in Zirlo

Collaborators: S. Rajasekhara and B.G. Clark

Absolute hydrogen pressure: 327 torr (~ 0.5 atm),

Ramp rate: 1 ^{o}C/s, Final temperature: ~ 400 C, Dwell time: ~ 90 mins



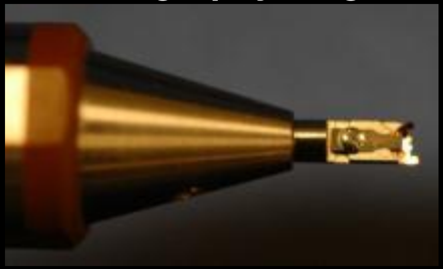
Hydride formation shown, for the first time by use of a novel TEM gas-cell stage, at elevated temperature and hydrogen pressure

Electron Tomography Provides 3D Insight

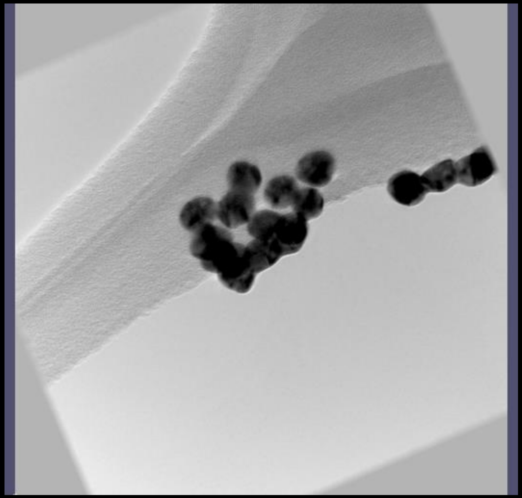
Collaborators: S.H. Pratt & T.J. Boyle

In situ Ion Irradiation TEM (I[3]TEM)

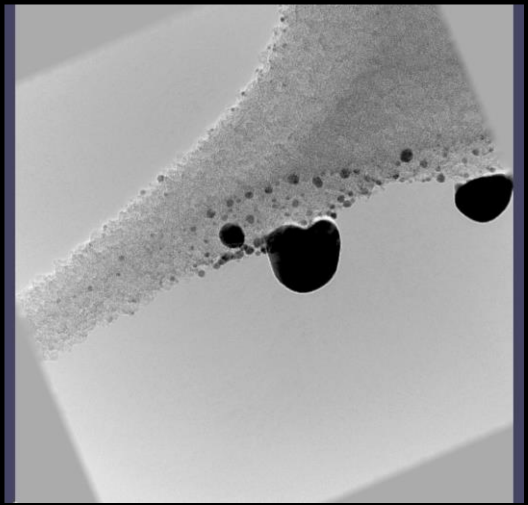
Hummingbird tomography stage



Aligned Au NP tilt series - unirradiated

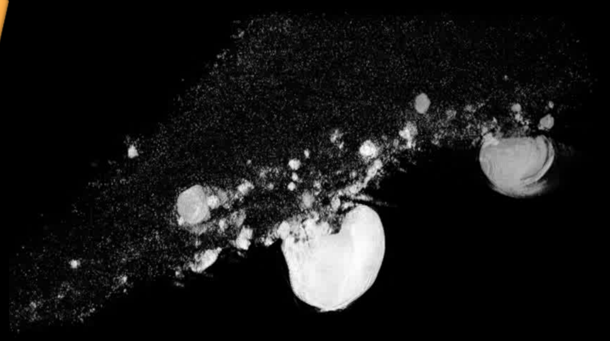


Aligned Au NP tilt series - irradiated



Unirradiated Au NP model

Irradiated Au NP model



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