SAND2022-6167C

Utilizing *In Situ* TEM to Decipher the Nanomechanical Properties of Helium Implanted Metals

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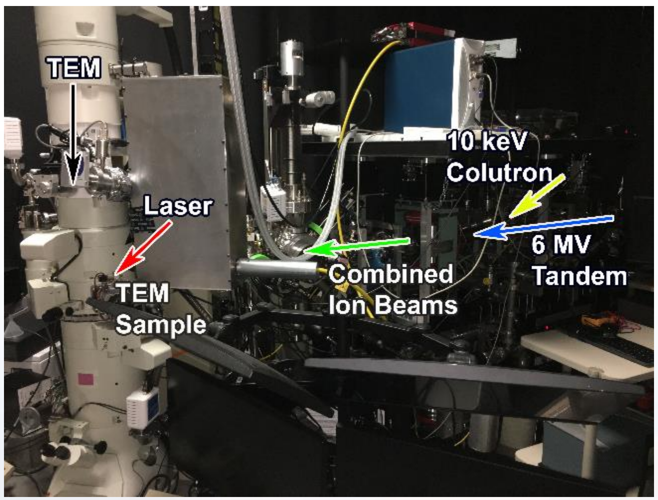
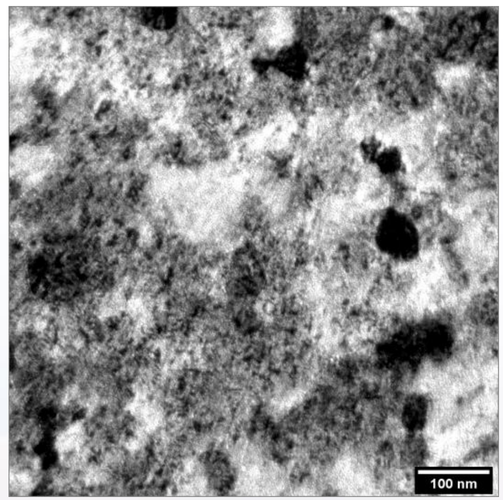
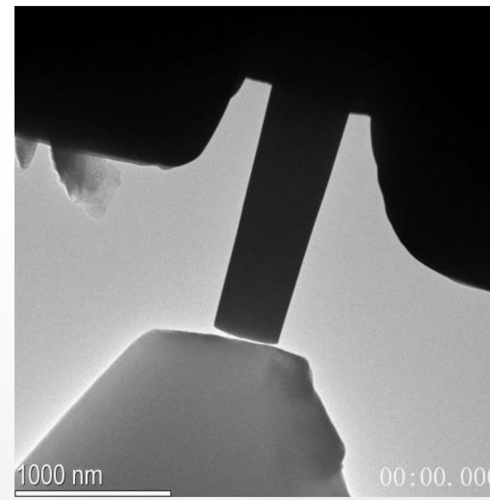
Sandia National Laboratories

May 9, 2022



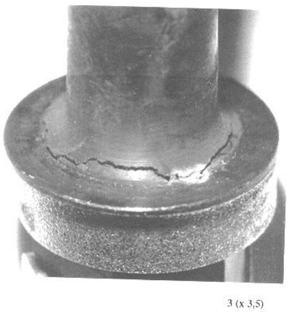




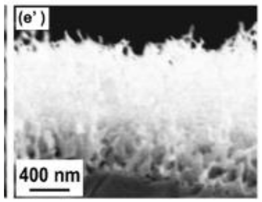
Utilizing *in situ* TEM mechanical testing to elucidate nanoscale mechanisms dictating mechanical properties

Macroscopic and Microscopic Helium Effects

Reactor Steel Embrittlement

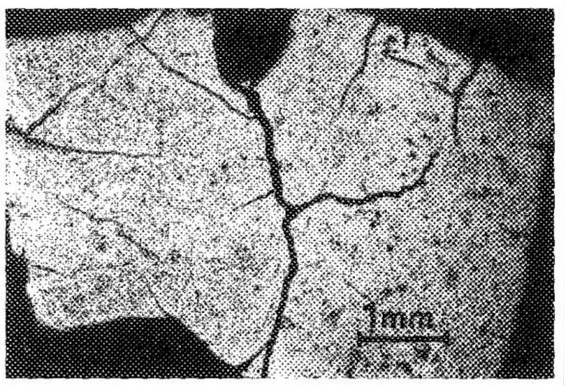


W Fuzz

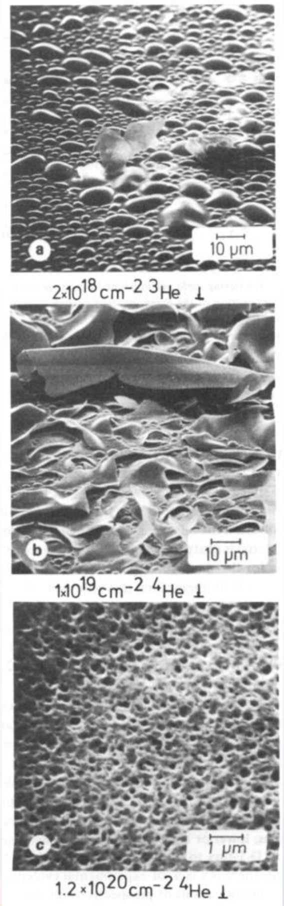


Kajita *et al* Nucl Fusion 49 (2009)

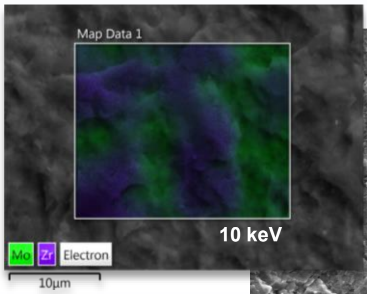
Fast Neutron Irradiation-induced swelling (3-6% He) in Boron Carbide



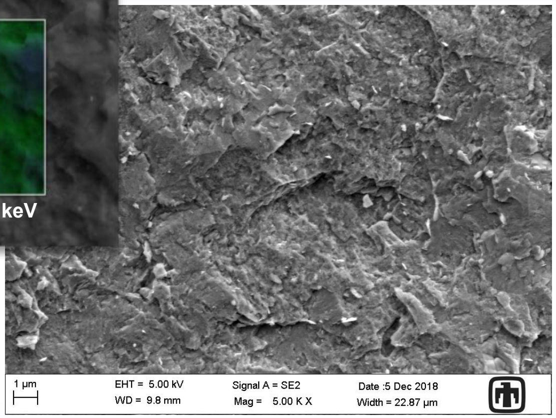
Stoto *et al* J. Appl. Phys. 68 (1990)



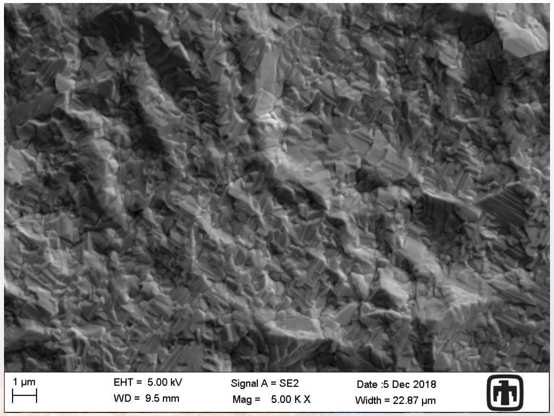
Ullmaier *Nucl Fusion* 24 (1984) 1039



ZrT_2, ~5 y 9 m old

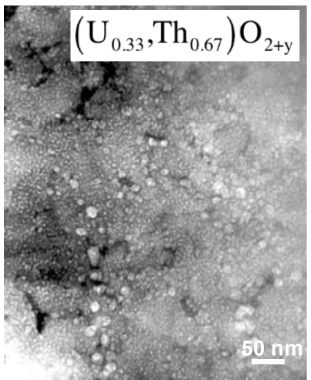


ZrT_2, ~1 y 10m old

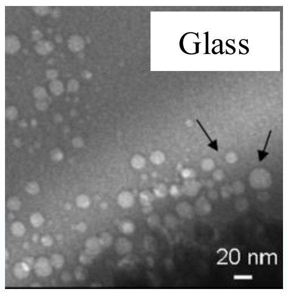


Nanoscale Helium Bubbles

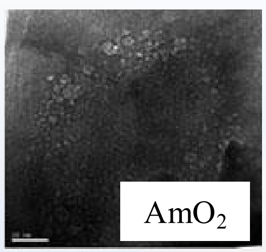
Ceramics



T. Wiss *et al.*, JNM (2014)



G. Gutierrez *et al.*, JNM (2014)



T. Wiss *et al.*, JNM (2015)

Metals

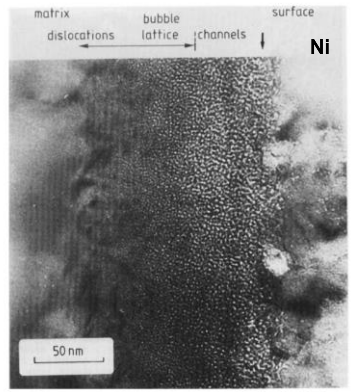
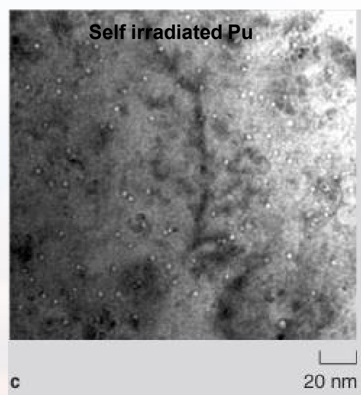
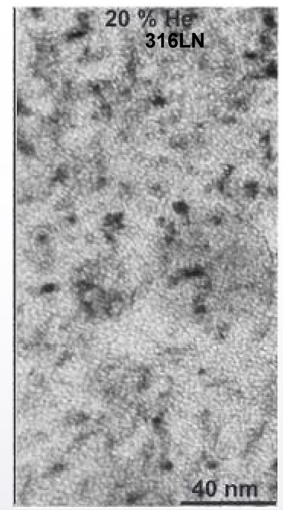


FIGURE 8 Transverse section of a high dose He-implanted surface layer of Ni showing interconnected channels, bubble lattice and dislocations (TEM micrograph by Jäger, 1980).

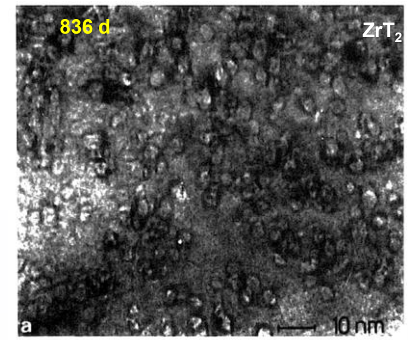


Zocco and Schwartz JOM (2003)

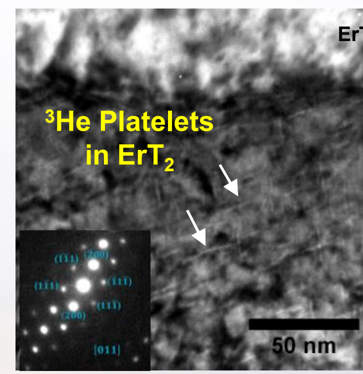


Hunn *et al* JNM 282 (2000) 131-136

Metal Hydrides



Schober, Trinkaus, Lasser, JNM 141-143 (1986) 453-457



Bubbles evolution at temperature can elucidate physical mechanisms

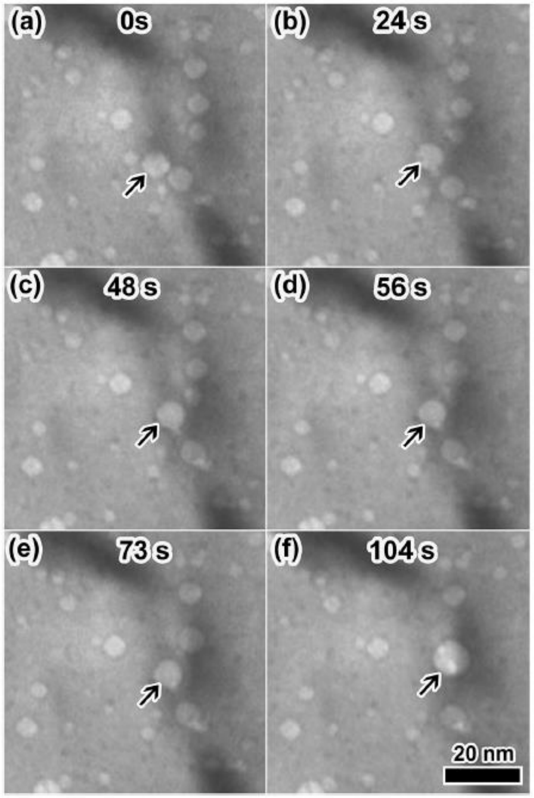
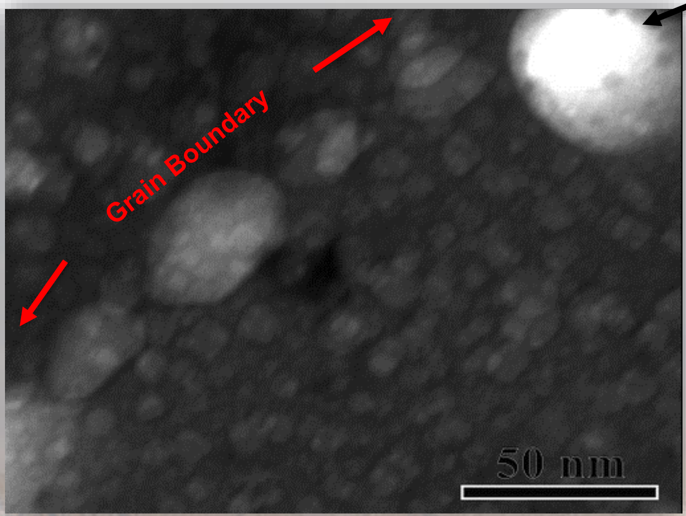


Fig. 6. BF TEM micrographs showing the mechanism of cavity migration and coalescence which drives cavity growth during

Taylor, C. A., et al. (2020). "Using In Situ TEM Helium Implantation and Annealing to Study Cavity Nucleation and Growth." JOM 72(5): 2032-2041.

- Blisters form at boundaries by absorbing nearby cavities
- Large faceted cavities form inside the grains by absorbing smaller bubbles and possibly He from the matrix
- Blisters eventually burst, leaving behind a denuded zone at the boundary



1x10^{17} He implantation, 450 C hold with in-situ resistive heating

C. Taylor, TMS, 2019.

How does heating rate affect bubble evolution?

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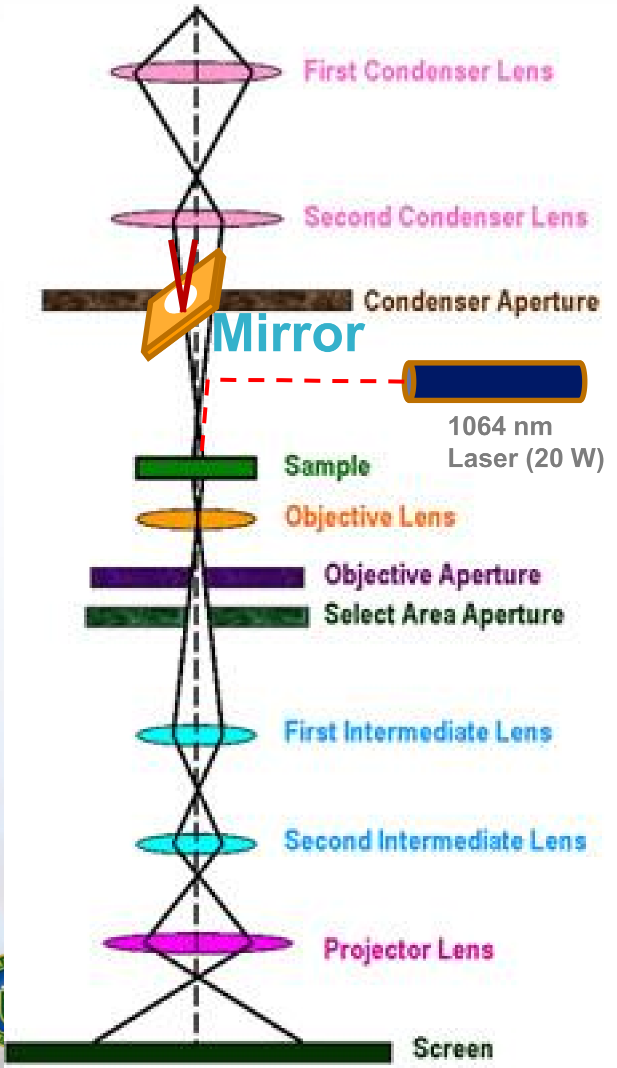
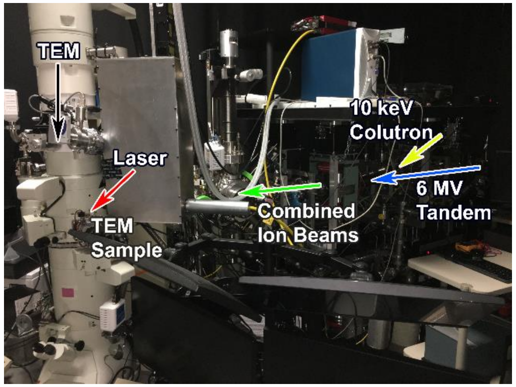
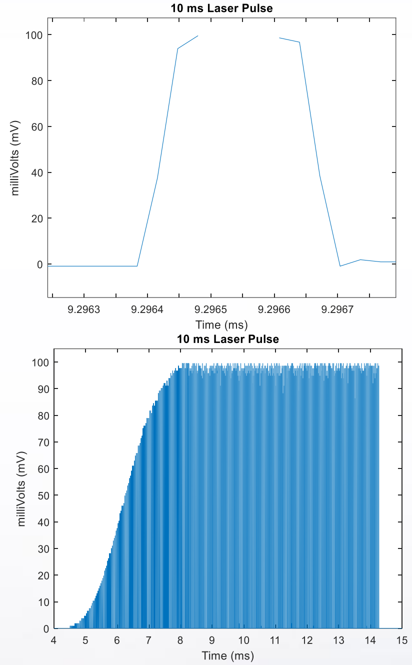
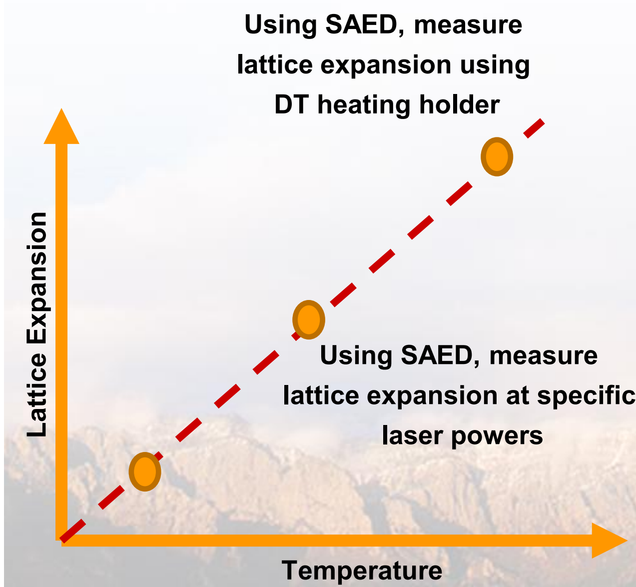
Realizing rapid, in-situ TEM heating with 1064 nm laser





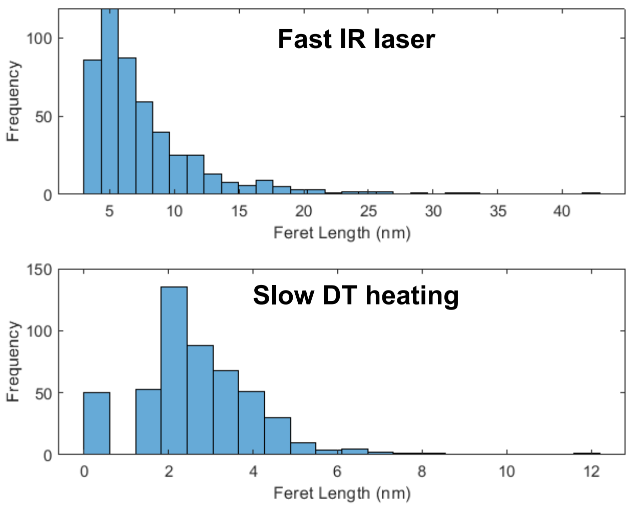


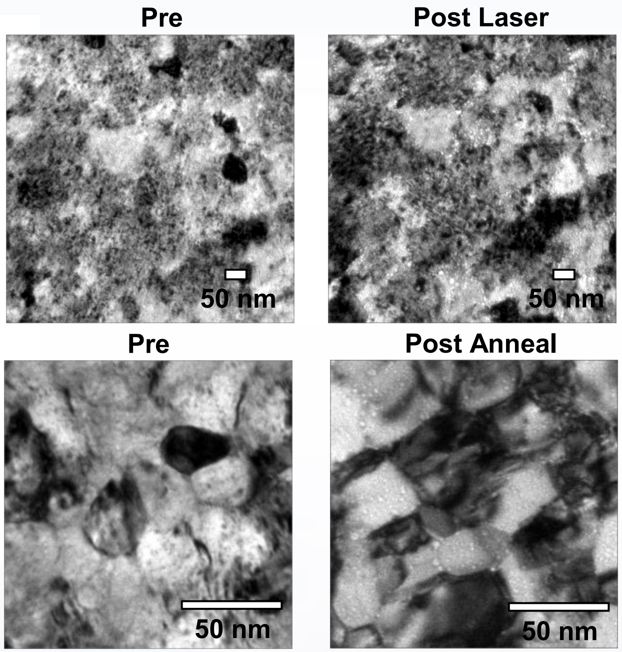
Rise time and pulse characteristics of 10ms laser pulse

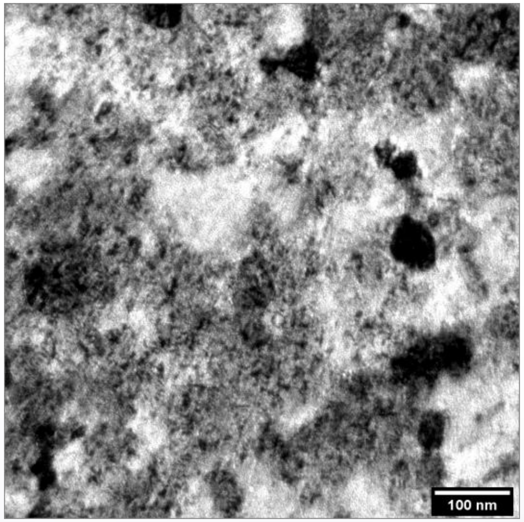


Compare laser heating with standard in-situ resistive heating holder

Heating rate affects He bubble size







- **Fast IR Laser: 3 W, 3sec pulse**
- **Slow DT Heating: 10C/min to 450 C**
- **The final temperature was attempted to be same: 450 C**
- **Pd thin film (50 nm) ex-situ implanted to a fluence of 6x10^{16} ions/cm^2 with 10 keV He**

Higher heating rate yields larger bubbles

Future work involving direct-detection camera will track bubble motion during heating transients at up to 500 fps

Direct Electron DE-64 Camera

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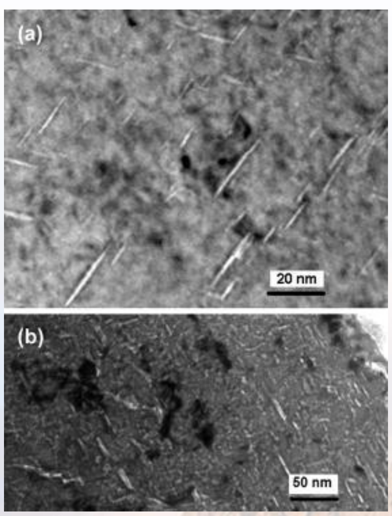
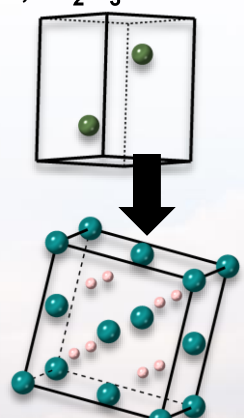
Emulating erbium hydride aging through ion irradiation

- Er undergoes a phase transformation from a hexagonal to a fcc structure under hydriding, forming ErH_2
- Tritium β-decays to ^3He, which models predict to remain in the tetrahedral site. Diffusion may occur through the octahedral site
- ^3He in ErT_2 tends to form platelet structures instead of bubbles
- ErH_2 usually contains some oxide, Er_2O_3

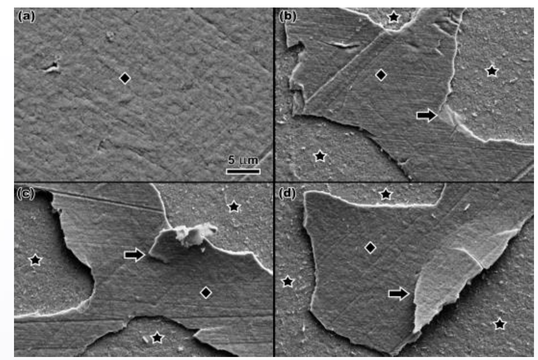


J.A. Knapp, et al. *J. Appl. Phys.*, 105 2009.

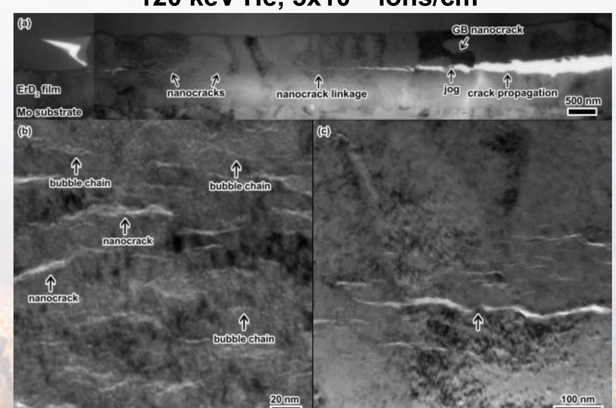


10% volume expansion

He implantation of ErD_2 causes surface flaking through bubble linkage and crack growth

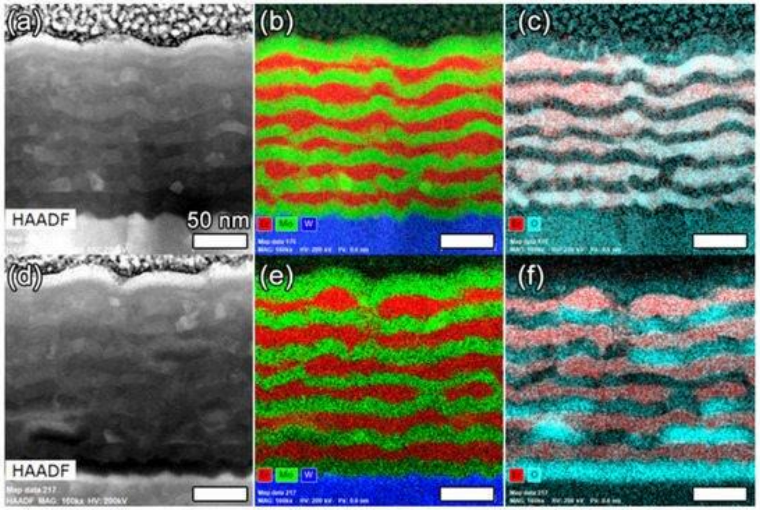


120 keV He, $5x10^{17}$ ions/cm^2



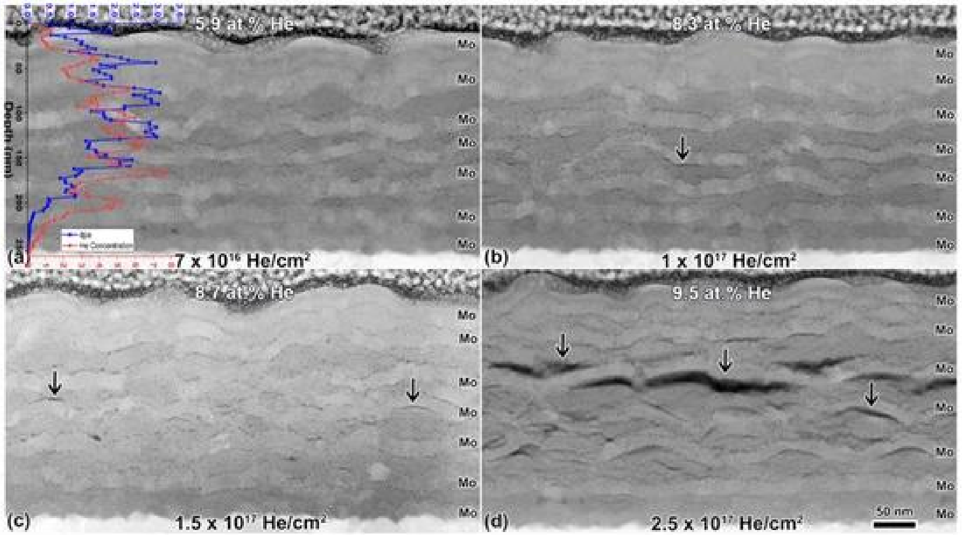
C.A. Taylor, et al. Materialia, 2021.

Multilayered Er composites to limit He bubble impacts

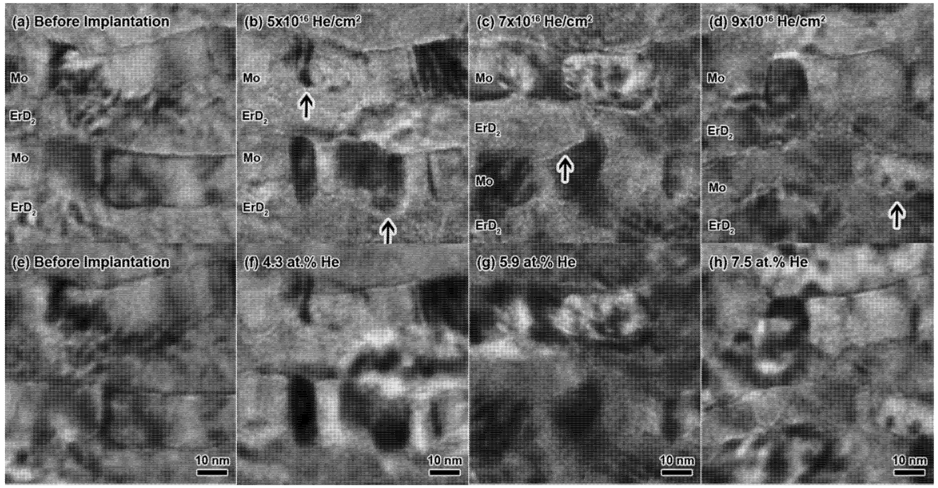


- Er/Mo multilayered samples fabricated via e-beam deposition
- Deuterated without formation of intermetallic phases
- He implantation to investigate He bubble nucleation

Er/Mo multilayered composites show He bubble accumulation at interfaces



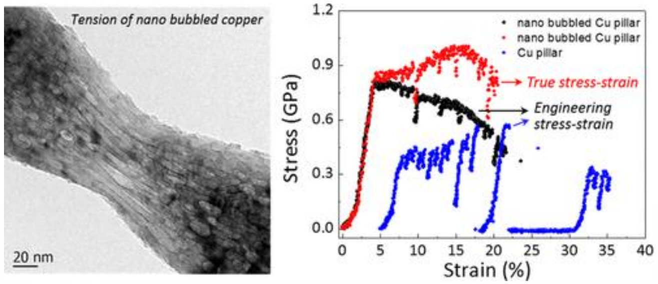
30 keV He *ex situ* implantation at Los Alamos National Lab

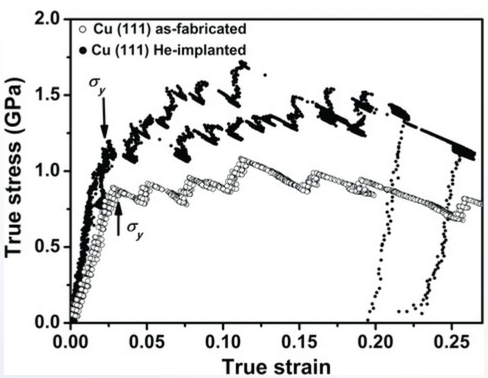


10 keV He *in situ* implantation at SNL

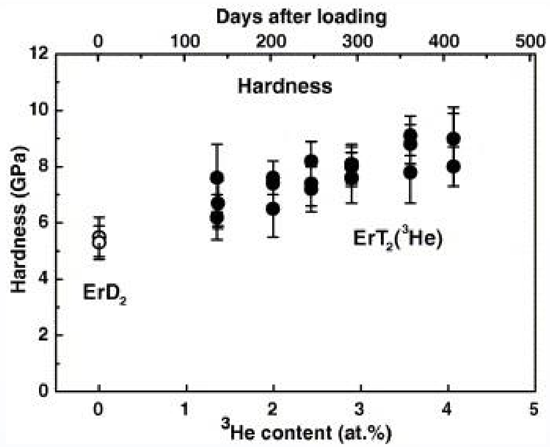
C.A. Taylor, et al. *Materials* 14 2021.

D/T and He accumulation changes mechanical properties of metals



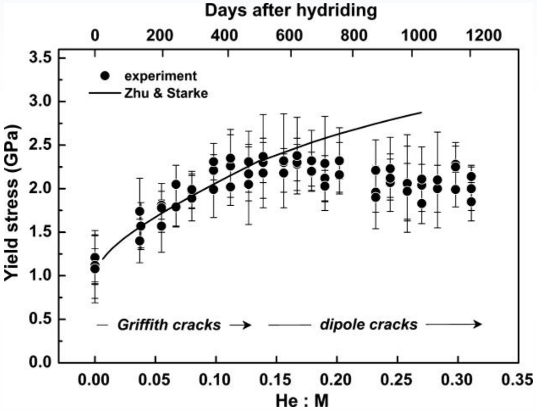


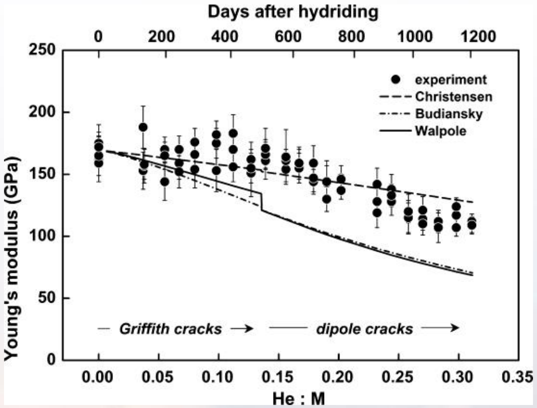
He accumulation changes mechanical properties of nanoscale specimens



ErD$_2$ shows similar hardness as un-aged ErT$_2$, yet properties of ErT$_2$ changes with time

How are the mechanical properties altered via accelerated aging?





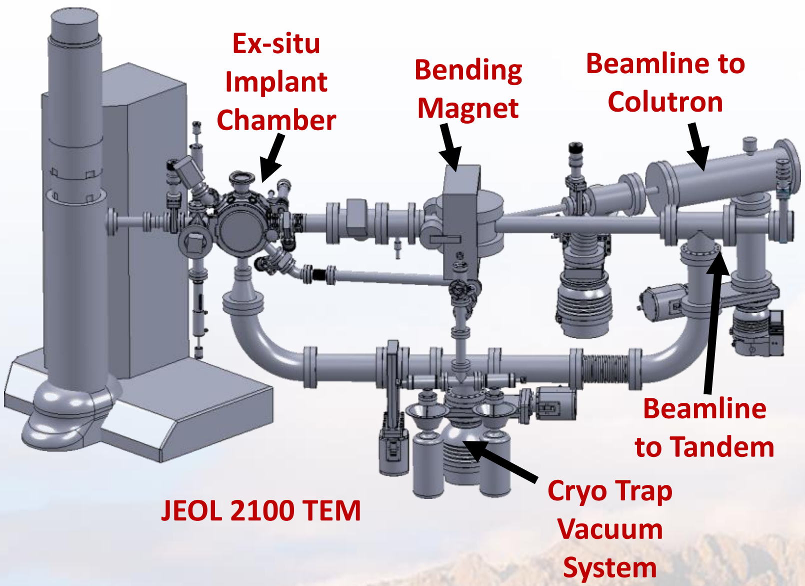
Yield stress and modulus of aged ErT$_2$

M-S. Ding, et al. *Nano Lett*. 2016.
Q. Guo, et al. *Small,* 2012.

J.A. Knapp, et al. *JNM*, 350 2006.
J.A. Knapp, et al. *J. Appl. Phys.,* 105 2009.

Rapid Evaluation of Helium in Materials using Sandia's I^{3}TEM

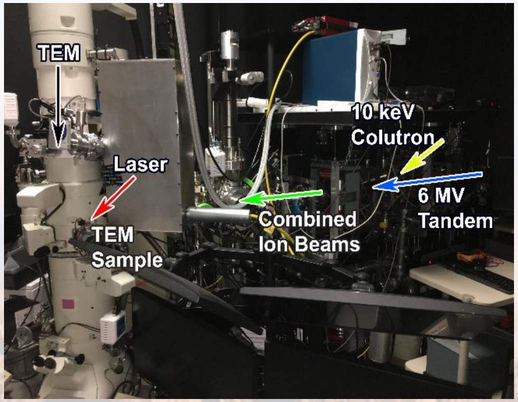
- **In-situ implantation** only takes a few hours – tritium aging takes several months and rad work
- **In-situ annealing** with the Gatan DT Heating stage or 1064 nm laser used to quickly assess the stability of bubbles
- **Hysitron PI-95 PicoIndenter** In-situ TEM nanomechanical testing



Quantitative Mechanical Testing

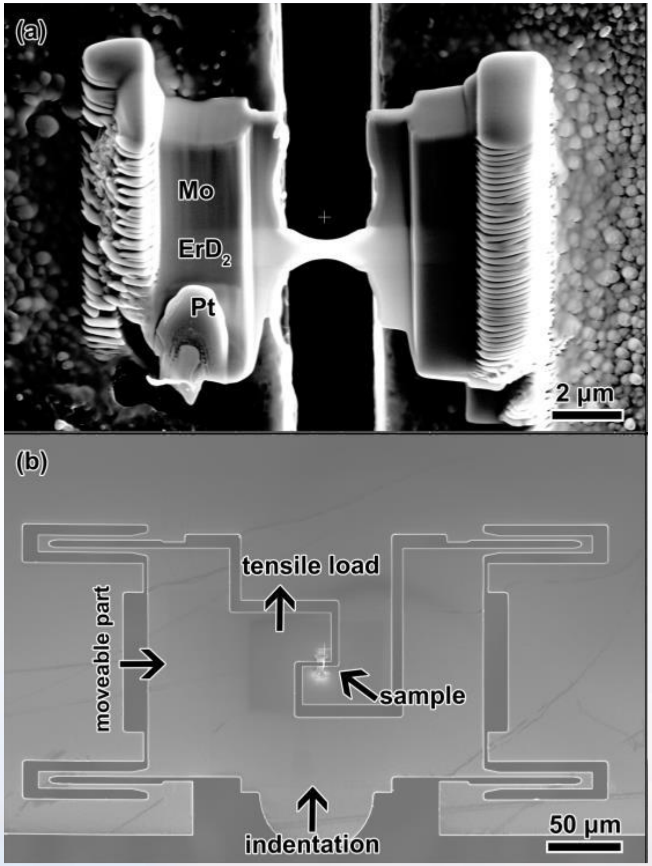
Minimal control over displacement and no "out-of-box" force information

- **Sub nanometer displacement resolution**
- **Quantitative force information with μN resolution**



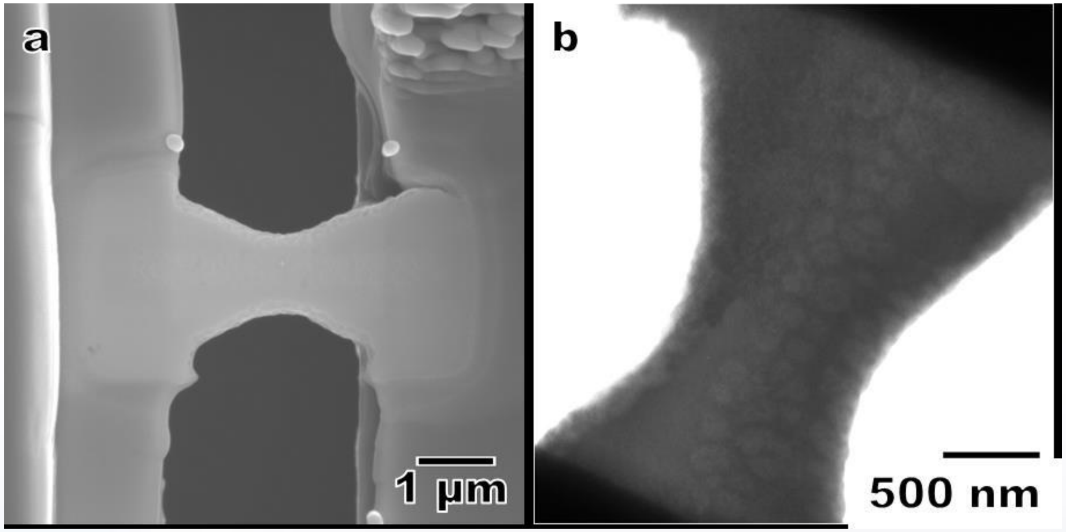
1) Indentation
2) Tension
3) Fatigue
4) Creep
5) Compression
6) Bend

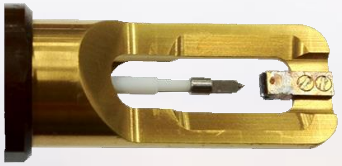
Utilizing in-situ TEM push-to-pull device for tensile testing



Nano-tensile bars for *in situ* TEM tensile testing successfully fabricated via FIB liftout

FIB liftout micro-machining to fabricate tensile bars for TEM testing

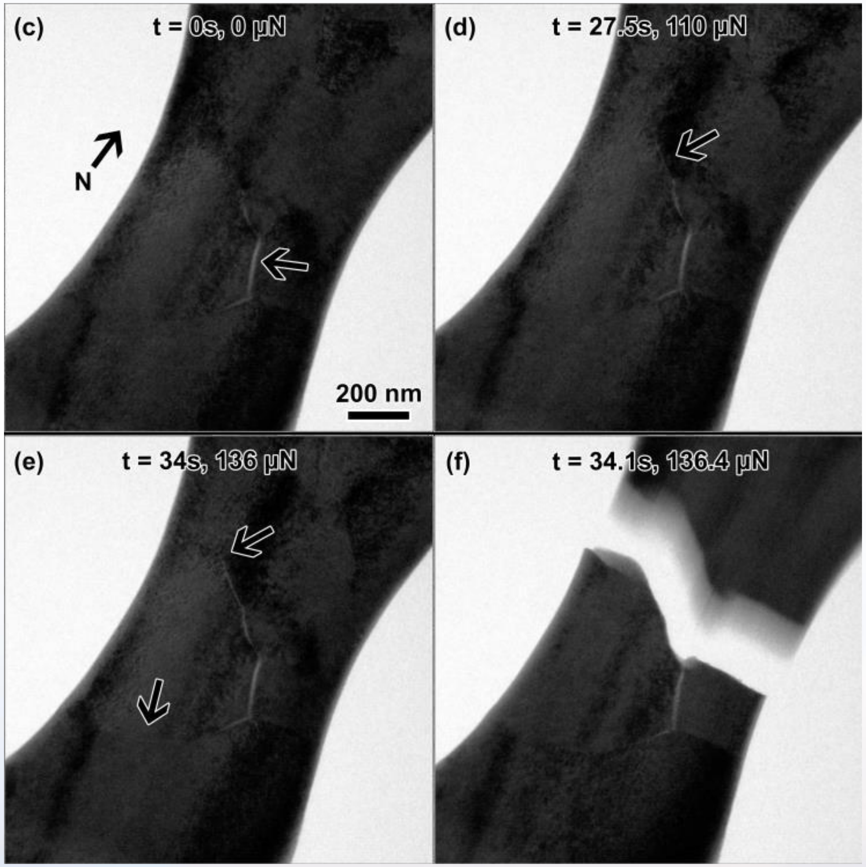




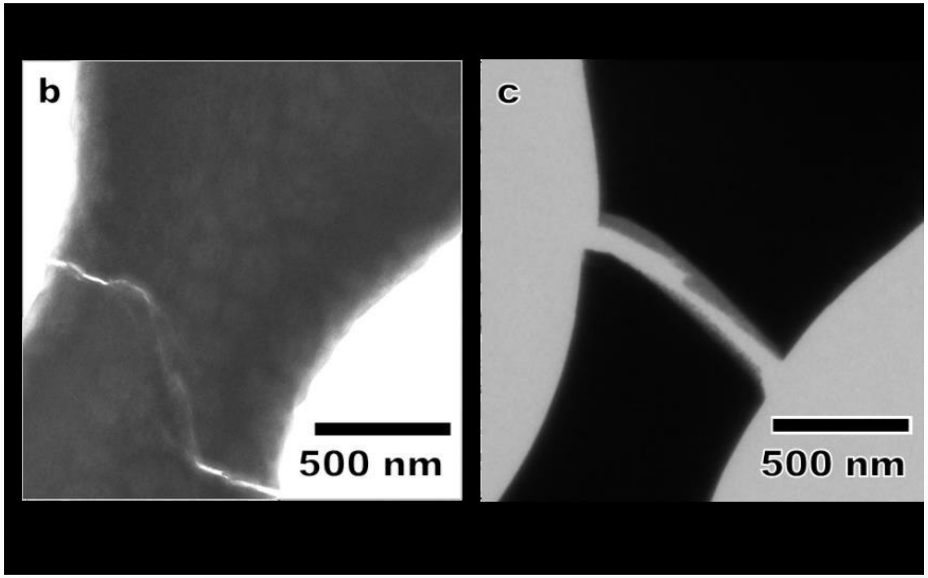
Displacement controlled test, 20 nm/s

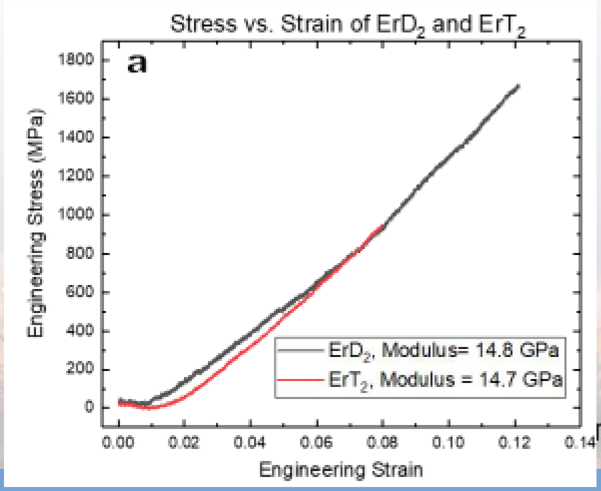
Push-to-pull testing using Hysitron PI-95

In-situ TEM tension tests resulted in brittle failure



No necking observed, brittle failure

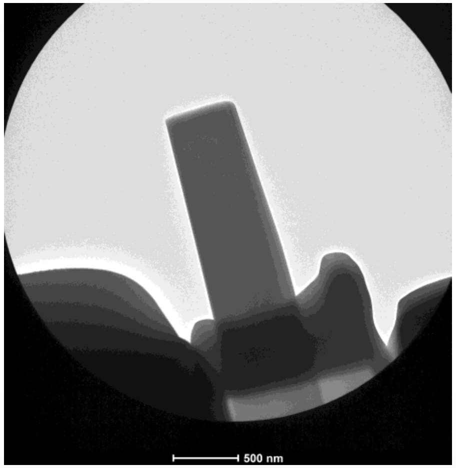


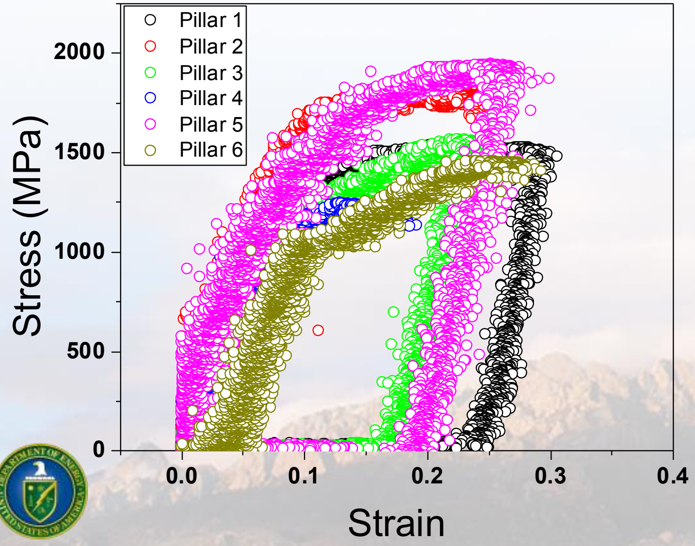


Similar moduli for ErD_2 and ErT_2, though lower stress needed for failure of ErT_2

Nanopillar compression likely more elucidating for brittle material like ErD_2

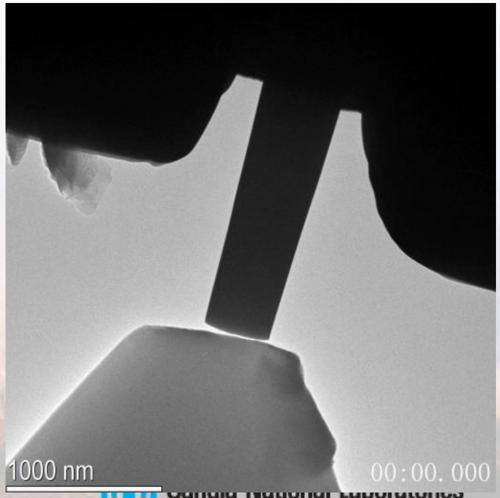
Pillar	Yield Strength [GPa]	Modulus [GPa]
1	1.5	2.5×10^1
2	1.8	1.2×10^1
3	1.6	0.5×10^1
4	1.3	-
5	1.9	1.4×10^1
6	1.5	1.0×10^1



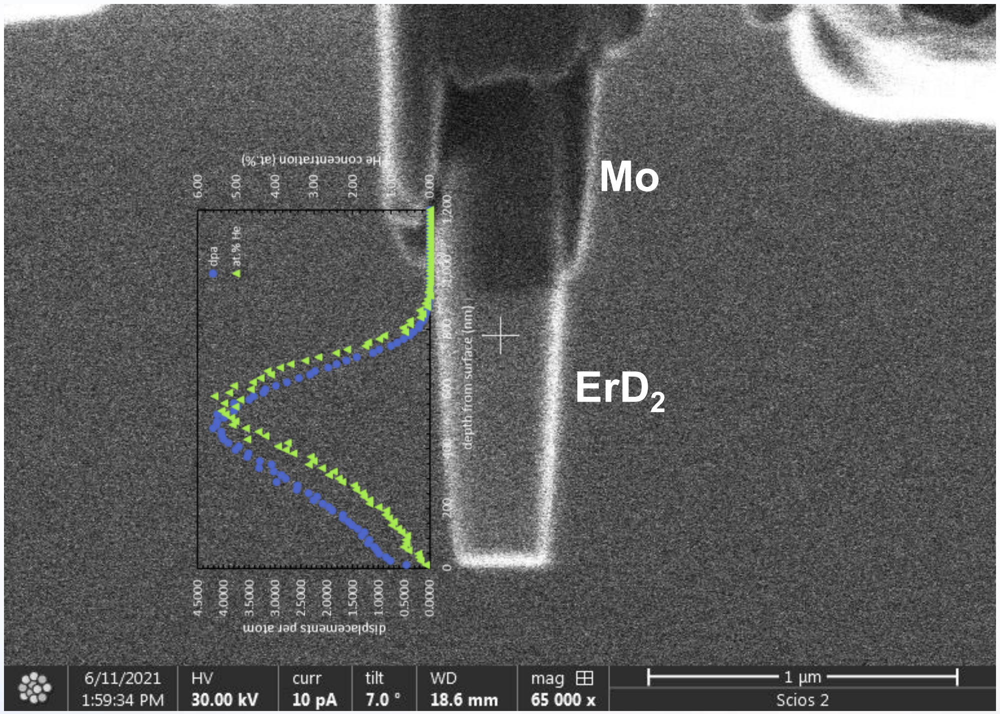


Testing the nanoscale mechanical properties of ErD_2 as an analogue to ErT_2

Thanks to Nan Li for assistance with experiments



Nanopillar compression likely more elucidating for brittle material



FIB-milled nanopillars of Er on Mo substrate

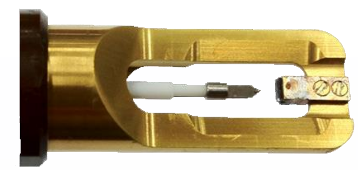
120 keV He implantation profile shows He peak in center of pillar

Peak He concentration: ~5 at.%

Utilizing *in situ* TEM nanopillar compression of ErD_2 and He-implanted ErD_2 thin film

In-situ TEM nanopillar compression tests

Hysitron PI-95 indenter 1 um flat tip



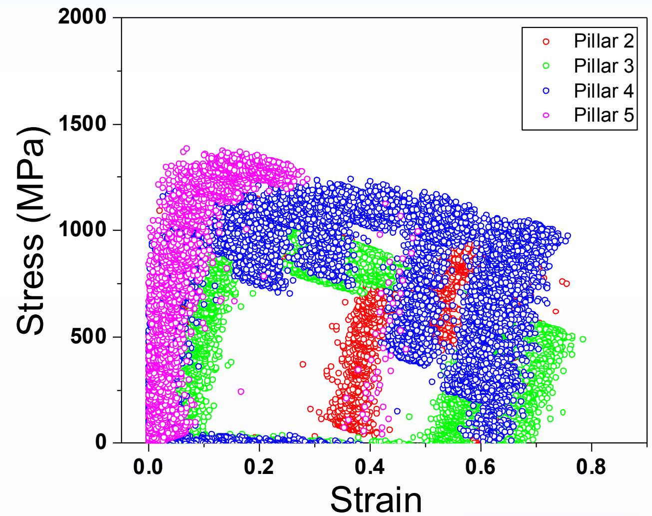
Displacement controlled test

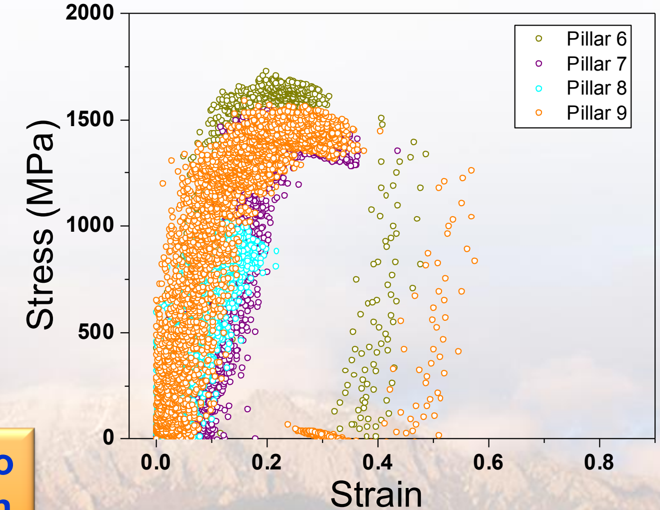
Load: 0-200 nm at 2nm/s

Unload: 100-0 nm at 2nm/s

He implantation affects pillar response, leading to softening, no strain hardening

Pillar	Yield Strength [GPa]	Modulus [GPa]
2	1.2	5.9
3	1.1	1.1
4	1.2	5.1
5	1.3	4.1
6	1.7	3.4
7	1.7	-
8	1.1	-
9	1.5	-





Aged ErT_2 :

He concentration of 5 at.%

From nanoindentation: Yield Strength - ~1.5 GPa

Young's Modulus - ~165 GPa

Low fluence He implantation leads to hardening, high fluence implantation can lead to softening

Increasing strain rate increases the measured strength of He-implanted ErD_2 pillars

Aged ErT_2 :
He concentration of 5 at.%

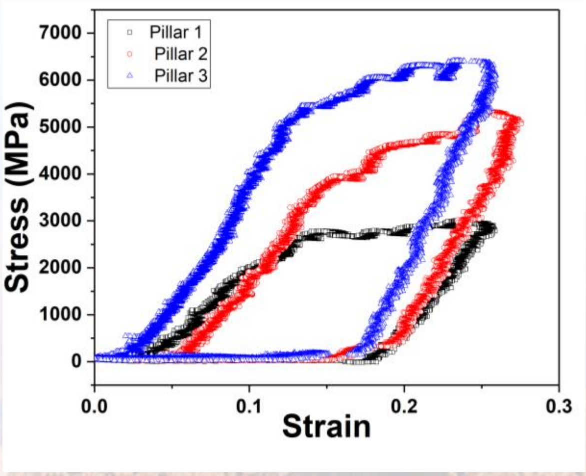
From nanoindentation:
Yield Strength - ~1.5 GPa
Young's Modulus - ~165 GPa

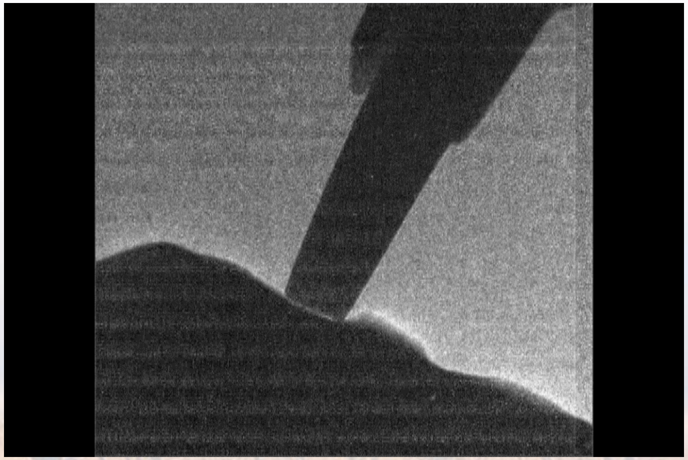
Pillar	Yield Strength [GPa]	Modulus [GPa]
1	2.68	$4.2x10^1$
2	4.03	$5.5x10^1$
3	5.44	$7.4x10^1$
4	4.98	$9.3x10^1$

Compliance of device needs to be accounted for in future

Increased strain rate increases strength observed in other material systems

J.A. Knapp, et al. *J. Appl. Phys.*, 105 2009.

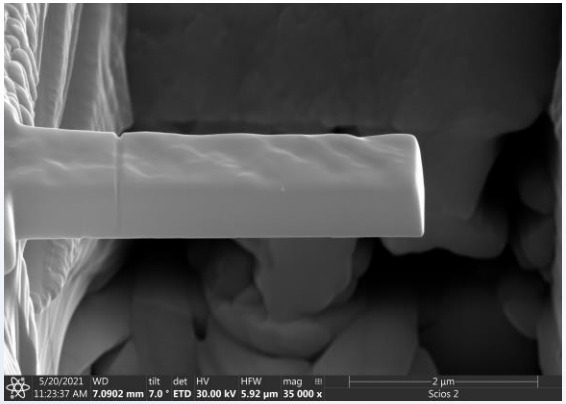




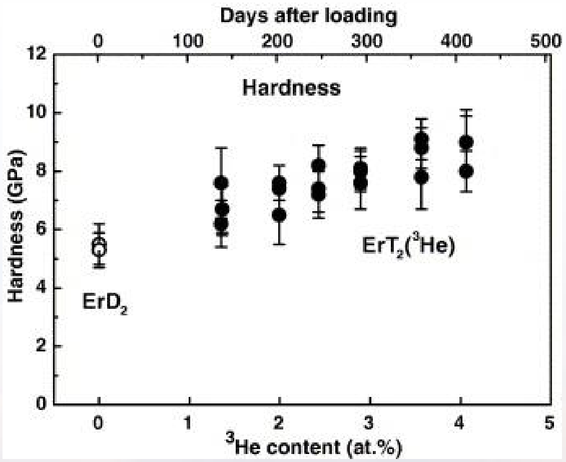
Nanoindentation can be used to test effects of He implantation on accelerated-aged ErD_2, scalable to ErT_2

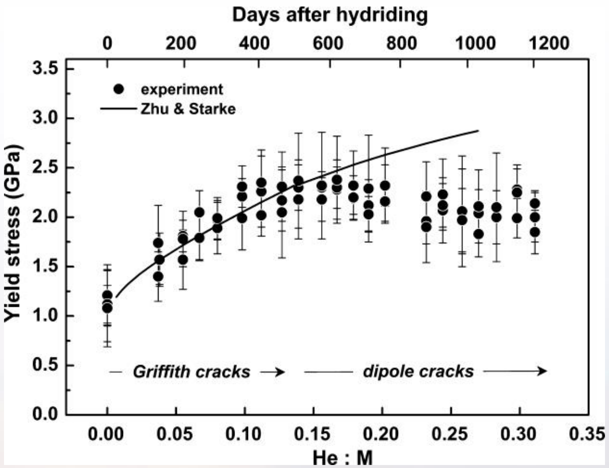
Further testing to decipher impacts of D and He loading

- **He implantation to $1x10^{17}$ He fluence slightly softens nanopillars**
  - Change in yield strength: 1.59 vs. 1.35 GPa
  - Intermediate He implantation concentrations
- **Will test ErT_2 pillars different aging times (days after loading)**
- **Micro-cantilevers fabricated for in-situ SEM fracture tests**



Micro-cantilevers of He-implanted ErD_2 for in-situ SEM testing



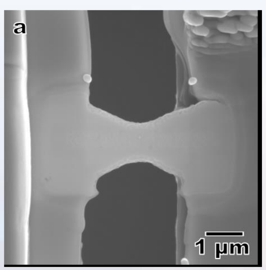


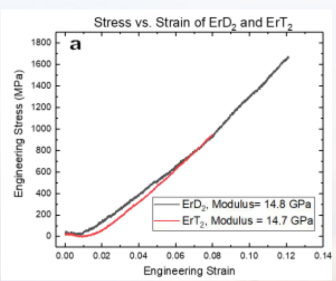
Utilizing *in situ* TEM mechanical testing to qualify the accelerated aging techniques in Er

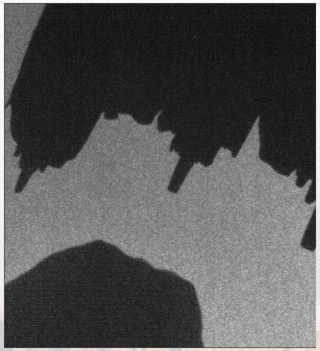
In-situ TEM techniques to elucidate helium effects on metals and metal hydrides

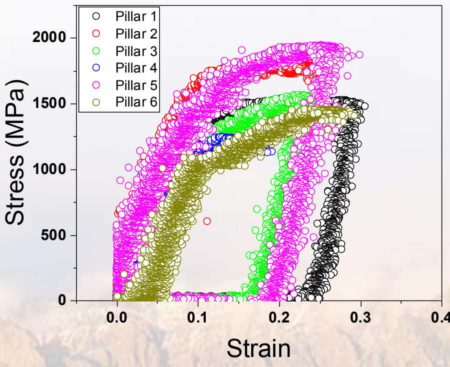
- Tritiated metals have mechanical properties that change as they age
- Can we simulate their aging through ion implantation?
- He implantation of deuterated metals used to simulate aging of tritiated metals
- In-situ TEM tensile and compression tests important for determining mechanical properties of aged and implanted hydrides

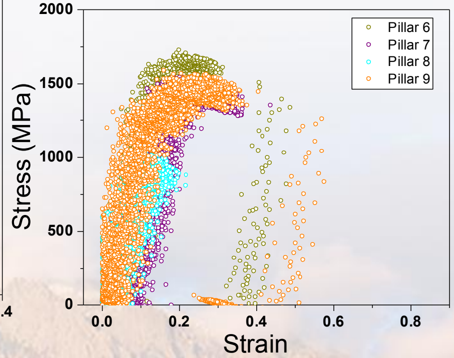
Preliminary results show accelerated aging useful to qualifying mechanical properties of aged films





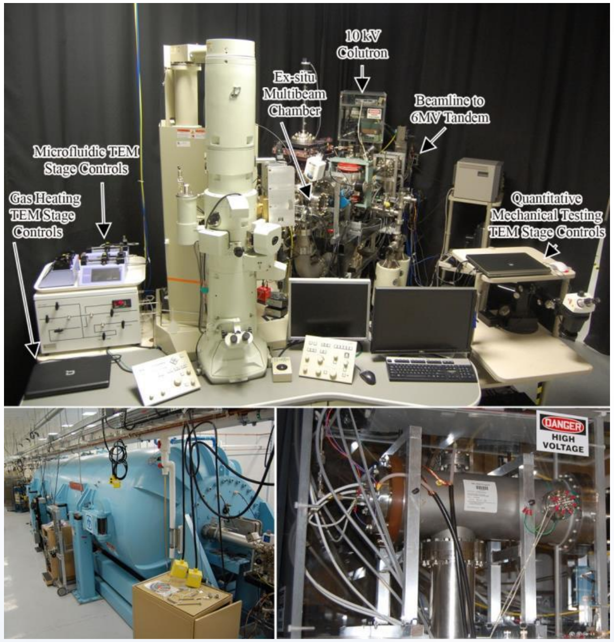


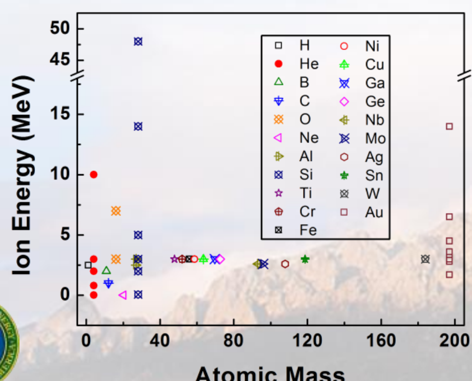




Sandia's Concurrent *In situ* Ion Irradiation TEM (I3TEM) Facility to study material evolution

10 kV Colutron - 200 kV TEM - 6 MV Tandem





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

Effect of He implantation on tensile straining behavior of Nb and W

Rapid heating of He-implanted NC metals to examine grain growth

Rapid heating of He-implanted NC metals to examine bubble growth

Environmental heating to examine effect of atmospheric exposure on grain growth

Heavy Ion Irradiation + Gaseous Implantation

Control ratio of dpa and gas species implantation and characterize coupling effects

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 (3x)

Acknowledgements

- Film Deposition: Ron Goeke (SNL)
- STEM/EDS: Paul Kotula (SNL)
- Bulk He Implantation: Yong Wang (LANL)
- In-situ Tensile Tests: Riley Parrish (SNL), Patrick Price (SNL), Caitlin Taylor (LANL), Khalid Hattar (SNL)
- In-situ Compression Tests: Nathan Madden (SNL), Khalid Hattar (SNL)

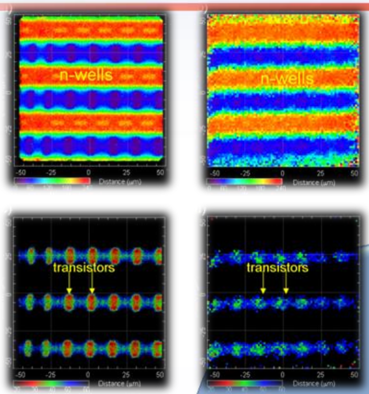
Access to the I^3TEM and associated facilities is now available through both the Nuclear Science User Facilities (NSUF) and the Center for Integrated Nanotechnologies (CINT).

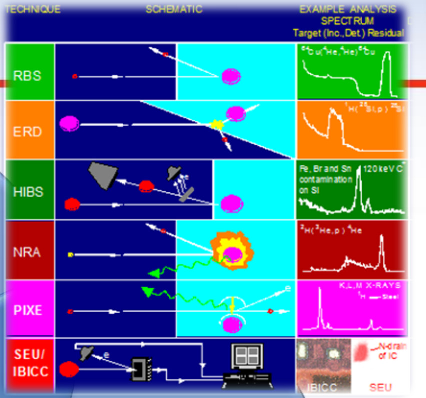
https://nsuf.inl.gov

http://cint.lanl.gov

This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc., for the U.S. DOE's National Nuclear Security Administration under contract DE-NA-0003525. The views expressed in the article do not necessarily represent the views of the U.S. DOE or the United States Government.

Sandia's USER Capabilities



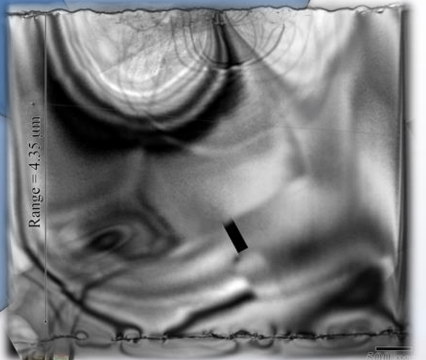


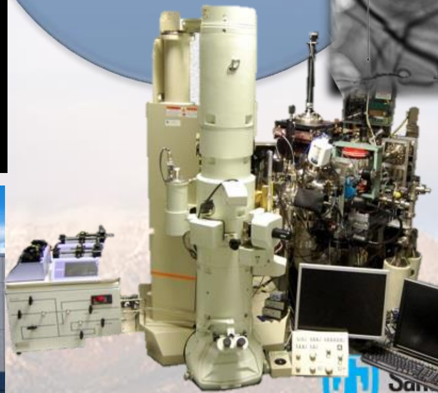
Ion Beam Analysis (IBA)

Radiation Effects Microscopy (REM)

Ion Beam Modification (IBM)

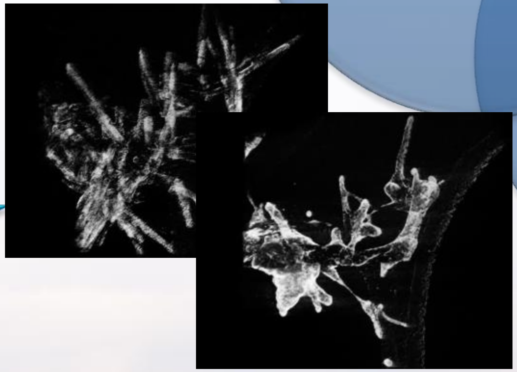
In situ Ion Irradiation Transmission Electron Microscopy (I³TEM)





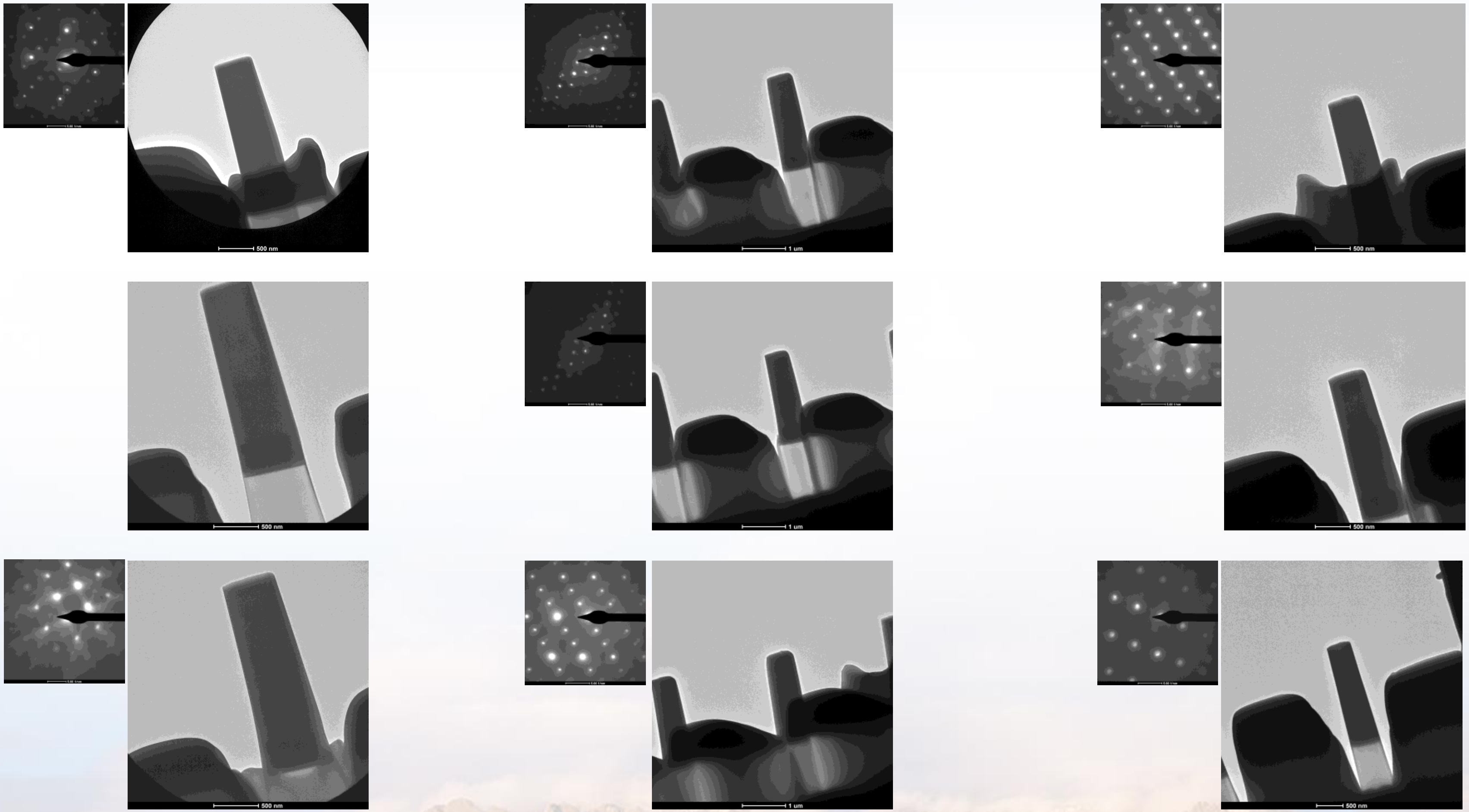
Core Facility - SNL

- Nanophotonics & Optical Nanomaterials
- Soft- Biological & Composite Nanomaterials
- Quantum Materials
- In-situ Characterization and Nanomechanics



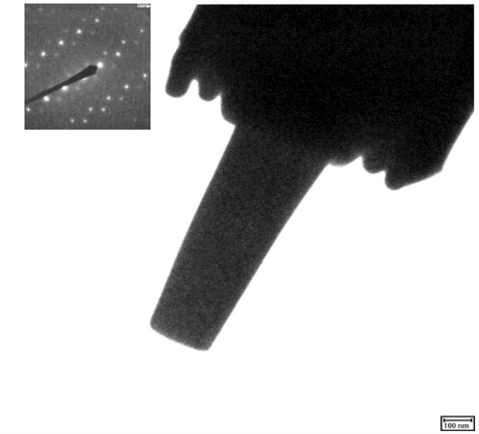
Gateway Facility - LANL

ErD_2 Pillars

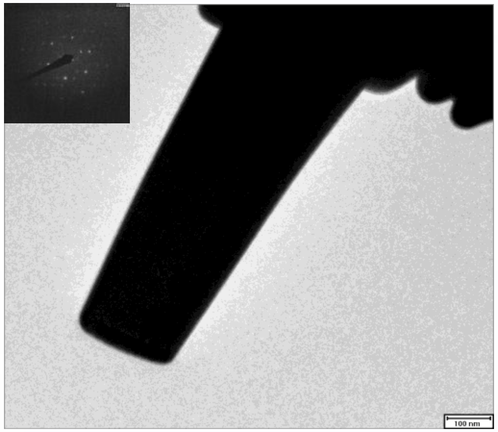


He-Implanted ErD_2 Pillars

He-implanted ErD_2 Pillars



Pillar 1



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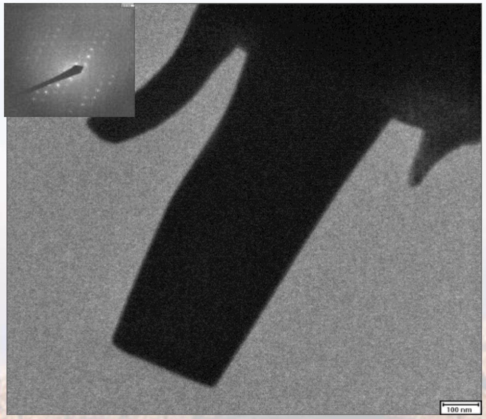
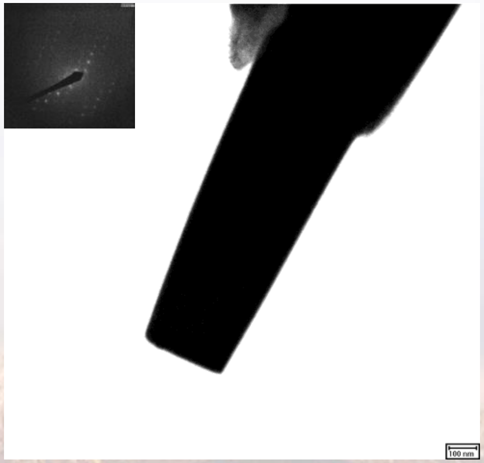


Pillar 3



Pillar 4