Application of Accelerators in Nanomaterials Research

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LA-UR-22-24755

International Conference on Accelerators for Research and Sustainable Development: From Good Practices Towards Socioeconomic Impact
May 23-27, 2022, Vienna, Austria

Los Alamos: ~7500 ft, >90% sunny days
~35 miles NW Santa Fe
Los Alamos County: ~20,000 populations
Los Alamos Lab: ~13,000 work force
~40 sq. miles, est. 1943

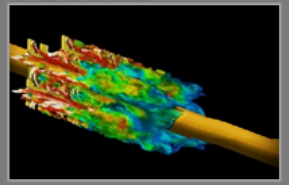
LANL capability pillars define six key areas of science, technology & engineering to support its missions

Materials for the Future

Defects and Interfaces

Extreme Environments

Emergent Phenomena

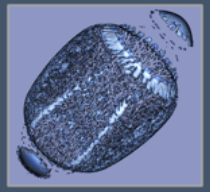


Science of Signatures

Nuclear Detonation

Nuclear Processing, Movement, Weaponization

Natural and Anthropogenic Phenomena



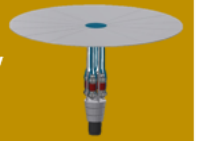
Nuclear and Particle Futures

High Energy Density Physics & Fluid Dynamics

Nucl. & Particle Physics, Astrophysics & Cosmology

Applied Nuclear Science & Engineering

Accelerator Science & Technology

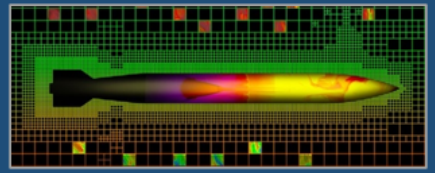


Weapons Systems

Design

Manufacturing

Analysis

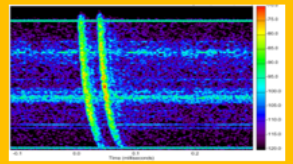


Integrating Information, Science, and Technology for Prediction

Complex Networks

Computational Co-Design

Data Science at Scale



Complex Natural and Engineered Systems

Human–Natural System Interactions: Nuclear

Engineered Systems

Human–Natural System Interactions: Non-Nuclear

The LANSCE facility has a diverse set of capabilities — many are essential for the Materials Pillar

- **Operations began in 1972**
  - Risk mitigation project completed in 2015; other efforts underway for sustainability
- **800-MeV (1 MW) proton beam**
- **Highly capable/flexible facility**
  - 100-800 MeV proton energies
  - 6 target stations
  - 3 neutron spallation targets
  - 16 beam lines
  - Time structure of beam allows for a large dynamic range of experiments
- **Dynamic proton radiography (pRad)**
- **Neutron radiography (Lujan)**
- **Structural material properties (Lujan)**
- **Nuclear properties of materials (WNR)**
- **Fundamental physics (WNR)**
- **Isotope production (IPF)**

DOE Nuclear Energy's Nuclear Science User Facility

Accelerator Applications in Nuclear Materials and Nuclear Energy: Radiation Damage with ions and neutrons; Defects Characterization with ions, neutrons, electrons, positrons, and X-ray photons etc.

BEAMLINE

A wide variety of beamline facilities are available, including ion, neutron, positron, and x-ray.

GAMMA IRRADIATION

The NSUF offers researchers access to best-in-class gamma irradiation facilities.

REACTORS

The NSUF offers ten reactor facilities to users whose proposals are awarded access.

POST-IRRADIATION EXAMINATION

Users can be awarded access to a broad range of PIE facilities.

COMPUTING RESOURCES

Providing researchers with advanced modeling and simulation capabilities to support a wide range of activities in harsh environments.

NUCLEAR ENERGY INFRASTRUCTURE DATABASE

Search nuclear energy R&D infrastructure at national labs, universities and industry.

NUCLEAR FUELS & MATERIALS LIBRARY

Proposals may be submitted for these samples, which were irradiated in ATR

PUBLICATIONS

Search the database of NSUF publications and presentations since its inception.

SEARCH R&D DATABASES

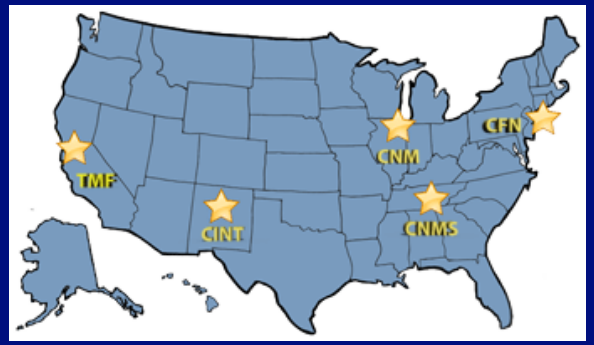
Search projects, samples, facilities and capabilities.

DOE Office of Science Nanoscale Science Research Centers

(Established in 2006)

The Molecular Foundry
Lawrence Berkeley National Laboratory

Center for Integrated Nanotechnologies
Los Alamos and Sandia National Labs



- Established in response to the National Nanotechnology Initiative
- FREE access to staff expertise and equipment for open science
- The NSRCs work with ~3500 researchers
  - High school, undergraduates, graduates, postdocs, staff
  - Academic institutions, industry, national labs, consortia, …
  - Domestic and international users

Center for Functional Nanomaterials
Brookhaven National Laboratory

Center for Nanophase Materials Science
Oak Ridge National Laboratory

Center for Nanoscale Materials
Argonne National Laboratory

CINT has four scientific thrusts that steward capabilities

www.cint.lanl.gov

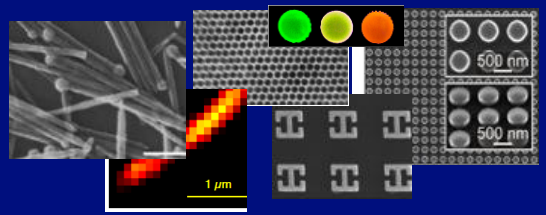
CINT is a LANL/SNL partnership to create a national resource for nanomaterials integration

CINT Today:

- 4 Science Thrusts, 1 Leadership Team
- 2 Primary Facilities (total 160,000 gsf)
- 60 Scientists & Technologists
- 19 Affiliate Scientists
  - Unique SNL/LANL capabilities & expertise
- 40+ Post-Docs & Students (SNL-LANL)
- Two open calls (Mar and Sept)
- Rapid Access Call Anytime

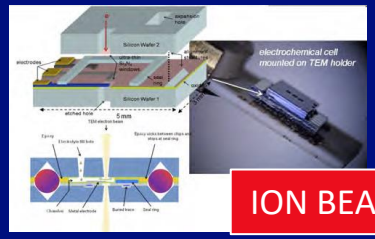
Nanophotonics & Optical Nanomaterials (NPON)

Synthesis, excitation, energy transformations of optically active nanomaterials



In-situ Characterization and Nanomechanics (ICNM)

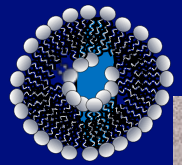
Developing capabilities to study dynamic response of materials and nanosystems to mechanical, electrical, or other stimuli

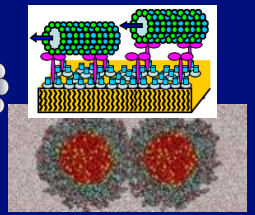


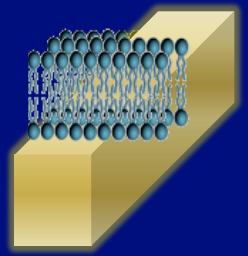
ION BEAM RESEARCH

Soft, Biological, & Composite Nanomaterials (SBCN)

Solution-based nanomaterials synthesis and assembly of soft, composite, and artificial biomimetic nanosystems

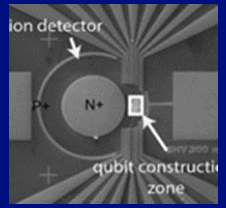






Quantum Materials Systems (QMS)

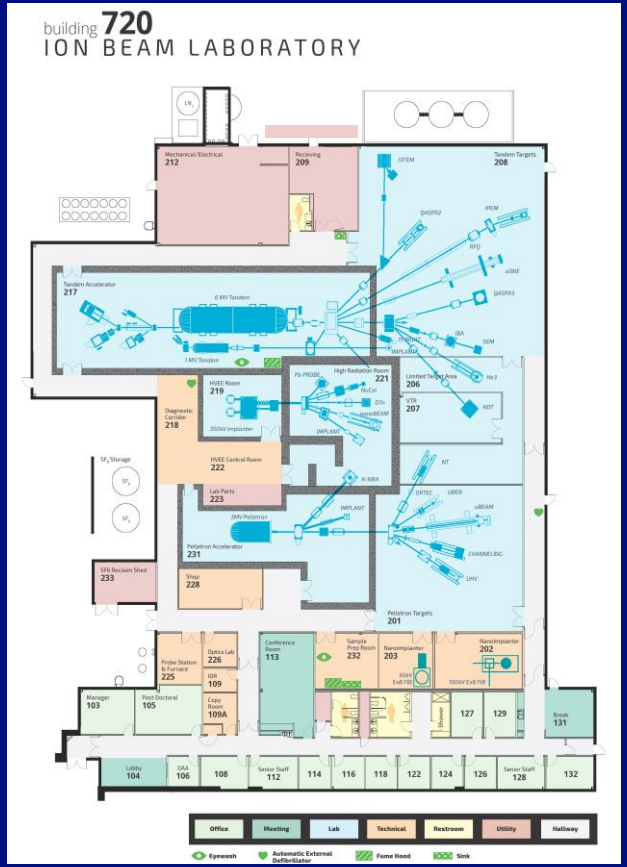
Understanding and designing nanomaterials to create new functionalities based on quantum effects that span multiple length scales (from nm to mm)



Los Alamos NATIONAL LABORATORY

CINT users have access to unique host NNSA laboratory facilities

Sandia Ion Beam Laboratory (IBL)



(Ed Bielejec: esbiele@sandia.gov)

8 Accelerators and
>25 end-stations
(including *in-situ* DLTS, PL, TEM, SEM, 1200°C heating, etc)

Deterministic Ion implantation and defect generation in Quantum Materials Research

Operational

(1) 6 MV Tandem Accelerator
(2) 3 MV Pelletron Accelerator
(3) 1 MV Tandem Accelerator
(4) 350 kV HVEE Implanter

High energy focused micobeams 1 μm to mm

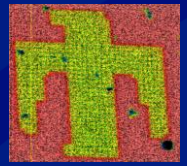
(5) 100 kV ExB FIB nanoImplanter
(6) 35 kV ExB FIB Raith Velion
(7) 35 kV Zeiss HeIM

Low energy focused nanobeams <1 to 20 nm

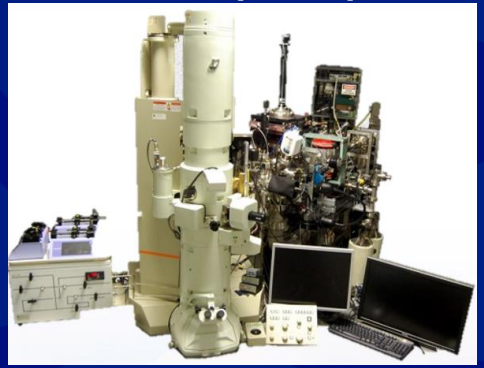
Installing

(8) 35 kV Plasma FIB

Focused Ion Beams



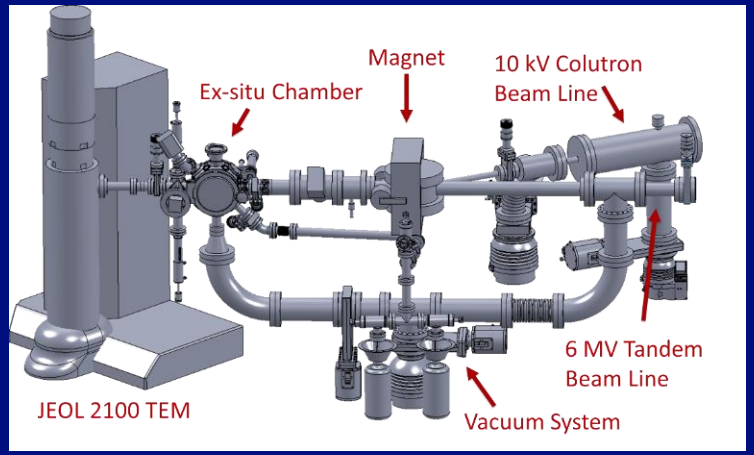
In situ Ion Irradiation TEM (I^3TEM)



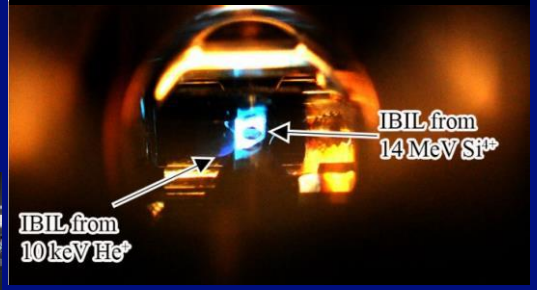
Sandia's Concurrent *In situ* Ion Irradiation TEM Facility

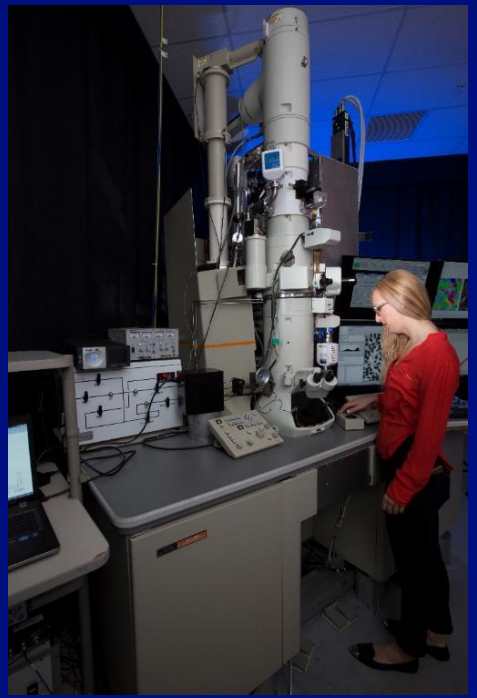
10 kV Colutron - 200 kV TEM - 6 MV Tandem

Contact: Khalid Hattar
khattar@sandia.gov



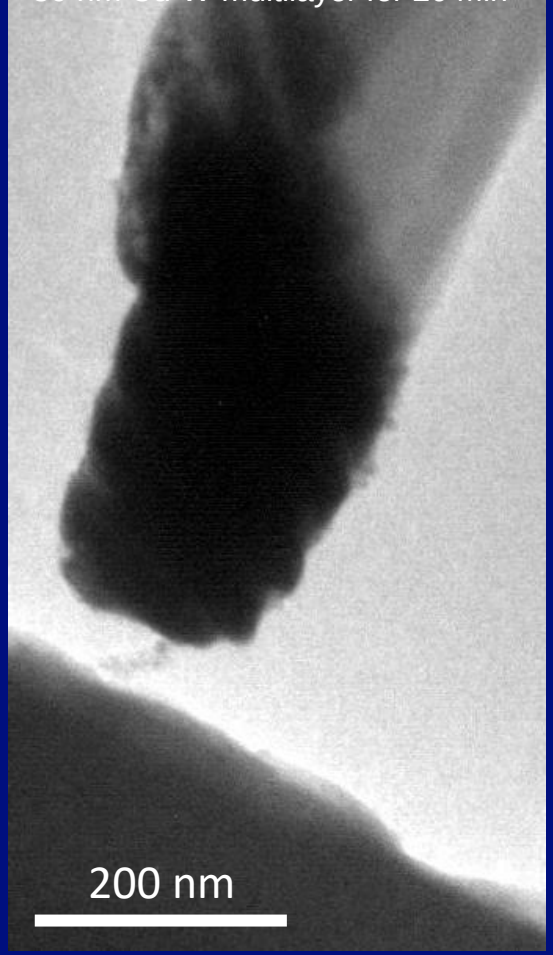
IBIL from a quartz stage inside the TEM





In Situ TEM Irradiation Creep

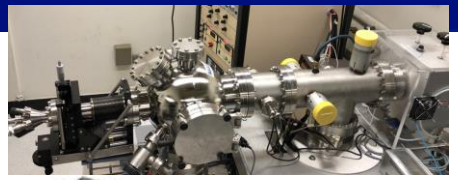
50 nm Cu-W multilayer for 20 Min



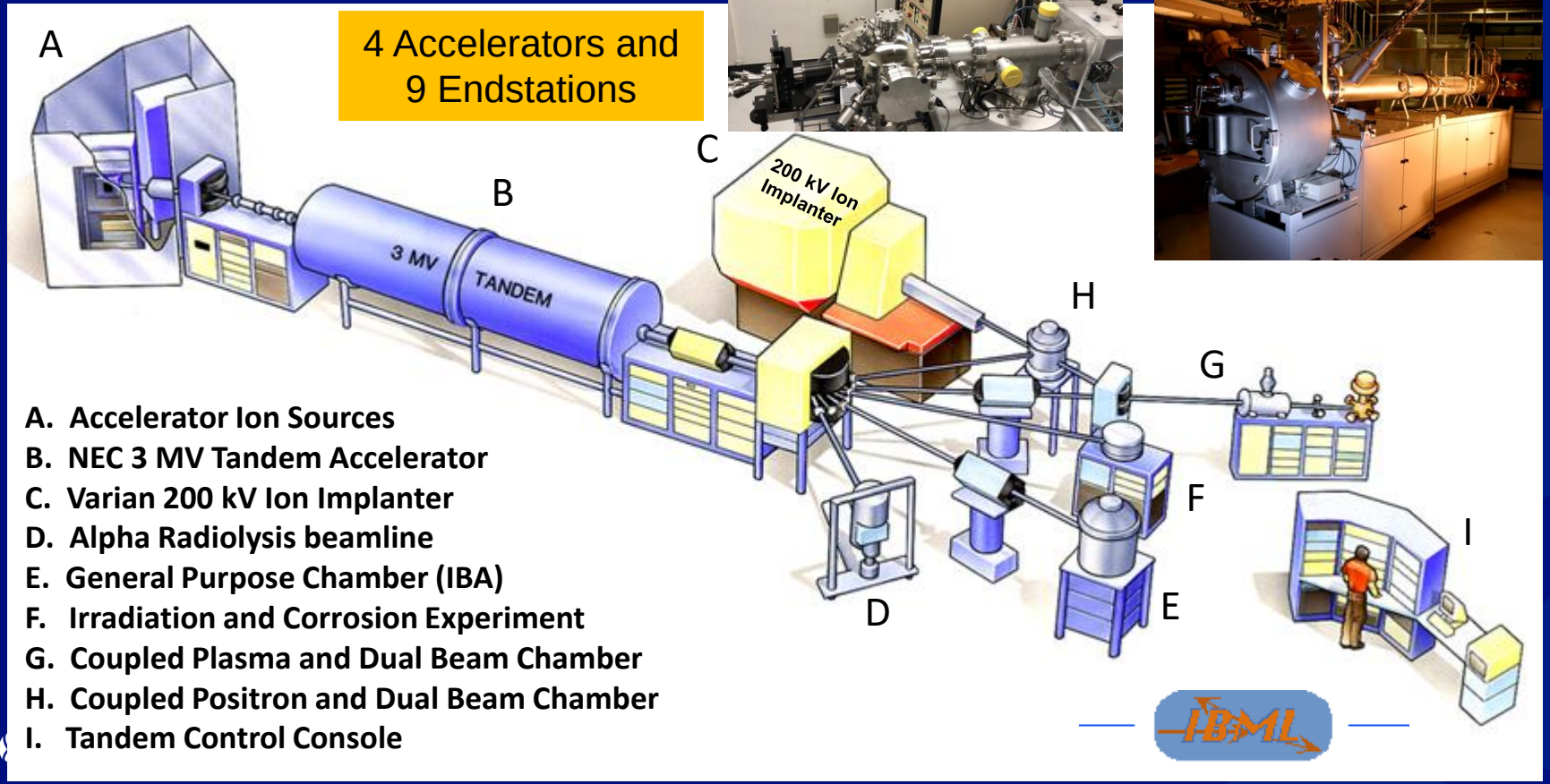
CINT users have access to unique host NNSA laboratory facilities

LANL Ion Beam Materials Laboratory (IBML)

Colutron 20 kV Implanter



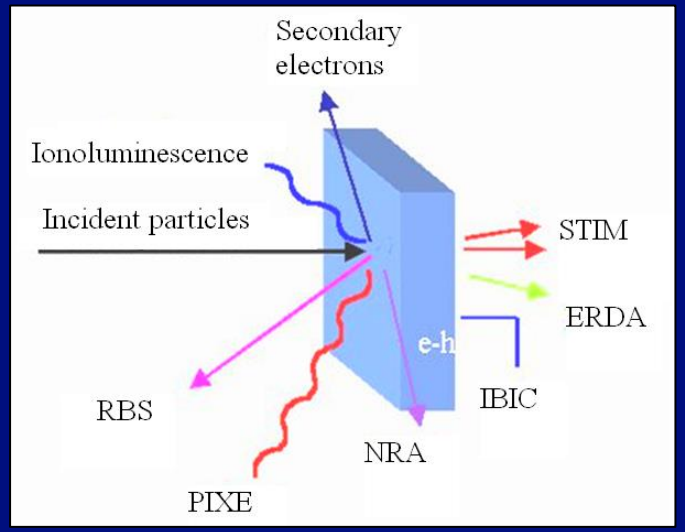
Danfysik 200 kV Ion Implanter



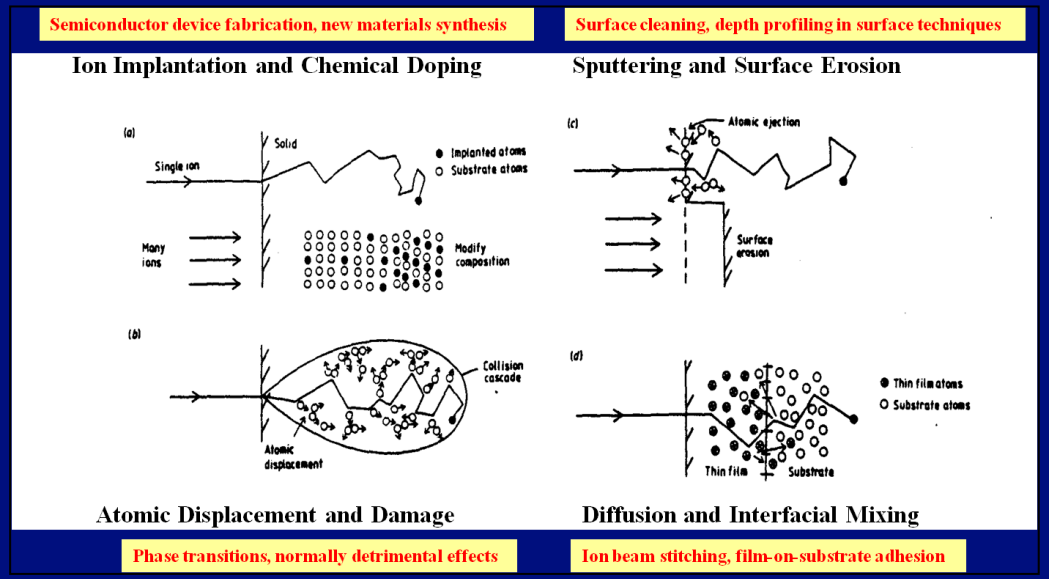
A. Accelerator Ion Sources
B. NEC 3 MV Tandem Accelerator
C. Varian 200 kV Ion Implanter
D. Alpha Radiolysis beamline
E. General Purpose Chamber (IBA)
F. Irradiation and Corrosion Experiment
G. Coupled Plasma and Dual Beam Chamber
H. Coupled Positron and Dual Beam Chamber
I. Tandem Control Console

Application of Ion Beams: Ion – Solid Interactions

Materials Analysis



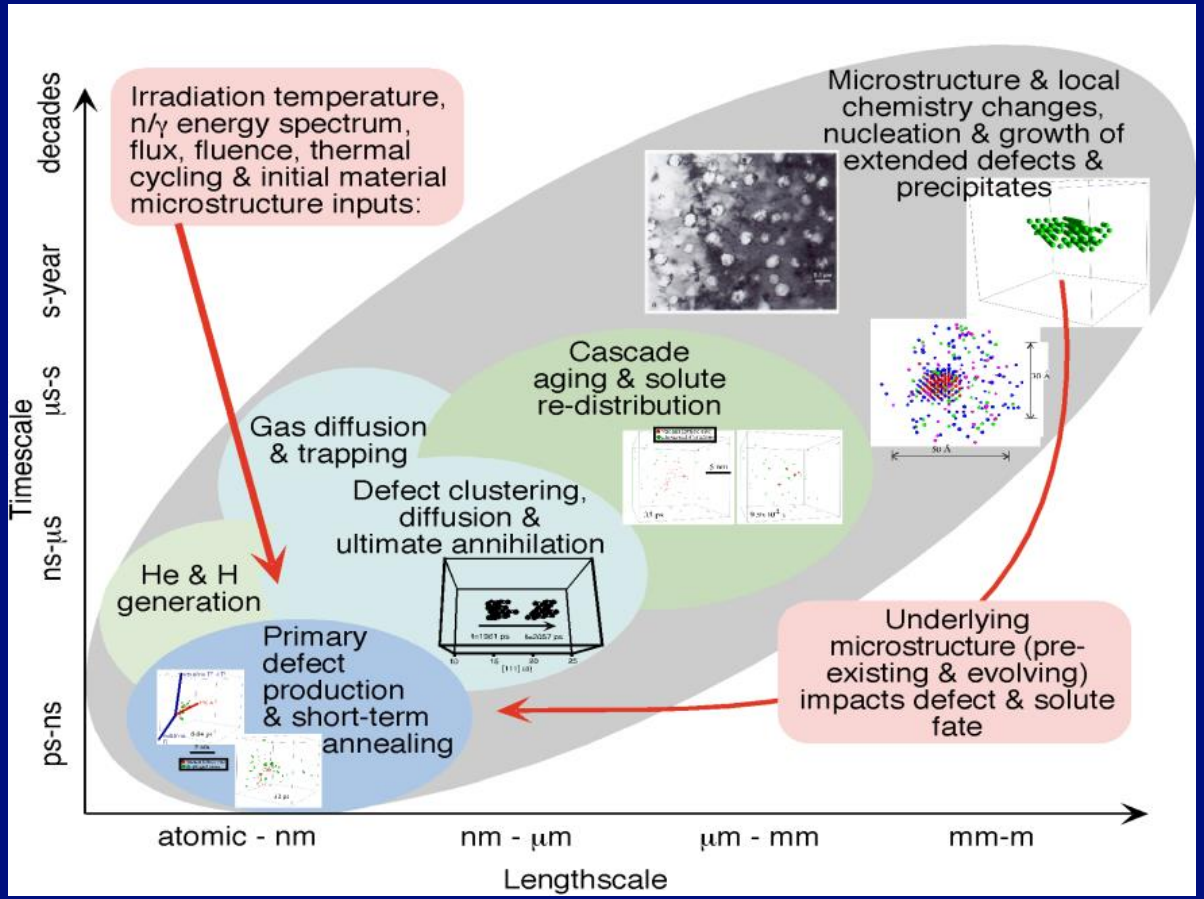
Materials Modification and Radiation Damage

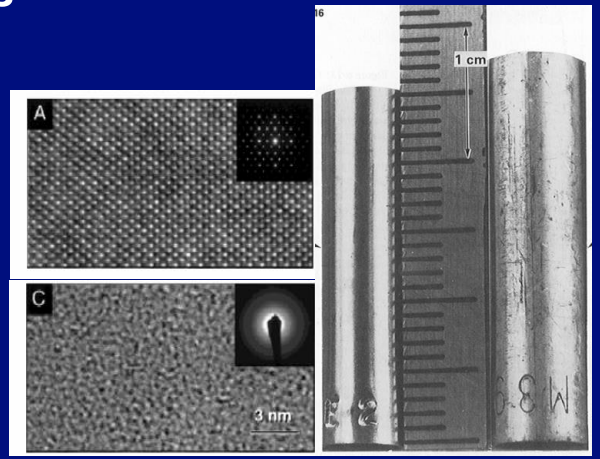


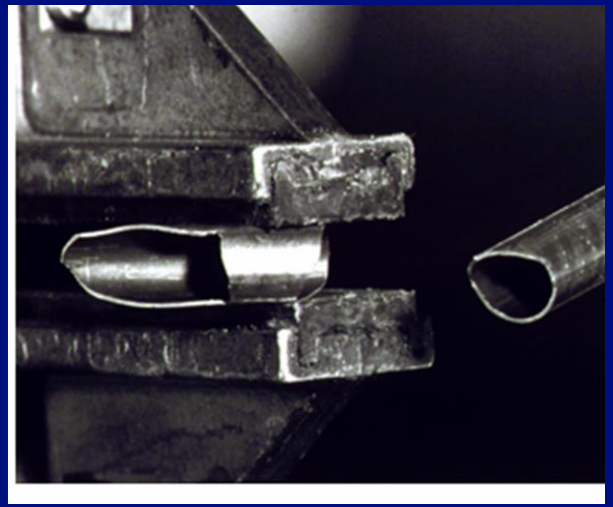
Ion Beams Contribute to Materials Research in Three Ways:

- ✓ Materials Characterization with Ion Beam Analysis Techniques (<u>as an analytical probe</u>)
- ✓ Materials Modification and Synthesis through Ion Implantation (<u>as a chemical dopant</u>)
- ✓ Radiation Damage Effects in Materials by Ion Bombardment (<u>as an irradiation source</u>)

Complex materials science under irradiation extremes



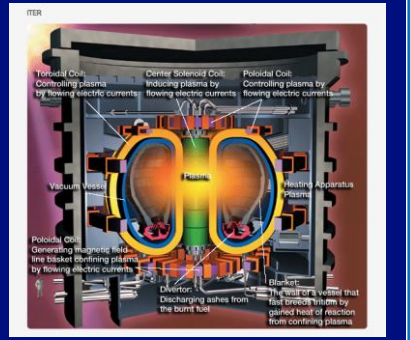




Example 1: Helium Damage Mitigation in Novel Plasma Facing Materials

The ultimate energy source: Nuclear Fusion Energy

ITER



ITER future timeline

Official esitmate (2016)

2015 1st components arrive on site

2018 Assembly of machine starts

2025 1st plasma. Beginning of shakedown operations in prep for fusion tests

2035 1st use of D (deuterium) and T (tritium) in the discharge. After this, repairs and replacements only by robots.

Personal (*unauthorized*) esitmate for achievement of facility goals. Assumes

(1) ITER has no unexpected component failures

(2) Plasma flow operates exactly as anticipated.

2040 Ignited plasma achieved – self sustaining fusion "burn."

2045 Q=10 (= P_{Fusion} / P_{Input}) achieved

Fusion Power Plant

Fusion Plasma - First Wall Material Interactions (PMI)

(D + T → He + n + 17.6 MeV ~1.5 x 10^8 K)

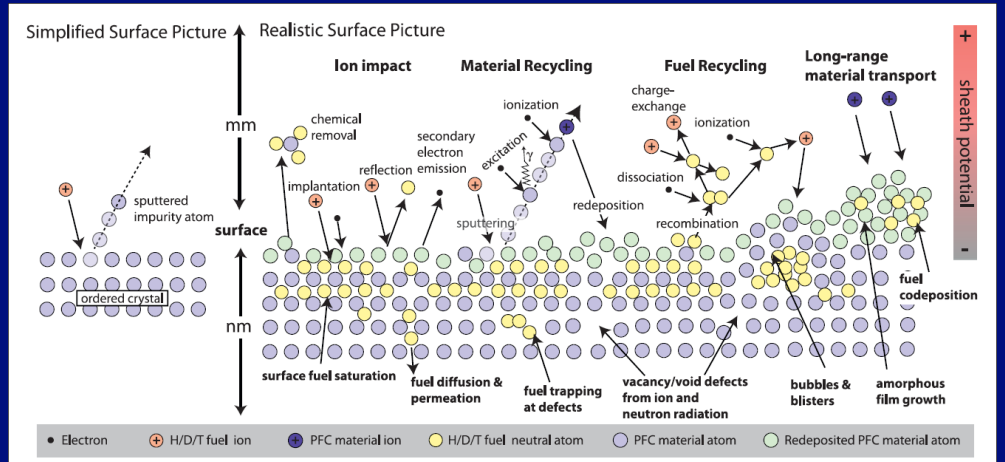


Figure 1. Schematic illustration of the complex, synergistic, and inherently multiscale surface interactions occurring at the material surface in a realistic magnetic fusion plasma environment. H, hydrogen; D, deuterium; T, tritium; PFC, plasma facing component; γ, gamma ray.

(BD Wirth, K Nordlund, DG Whyte, and D Xu, MRS Bulletin, 36, 2011)

Plasma facing materials face unprecedented harsh environments:

- High particle flux (>10^{22} $m^{-2}s^{-1}$) of D, T, He, etc.: Tritium recycling and He induced "Fuzz" in W
- Intense heat flux (~20 MW/m^2 steady-state, ~10 GW/m^2 if plasma control is lost): Thermal stability
- High neutron irradiation (~10^{18} $m^{-2}s^{-1}$) of 14.1 MeV neutrons: radiation damage resistance
- Unknown synergies among these effects

Tungsten: the leading candidate for plasma facing components

W is the choice for ITER Divertor and the lead candidate for DEMO PFM:

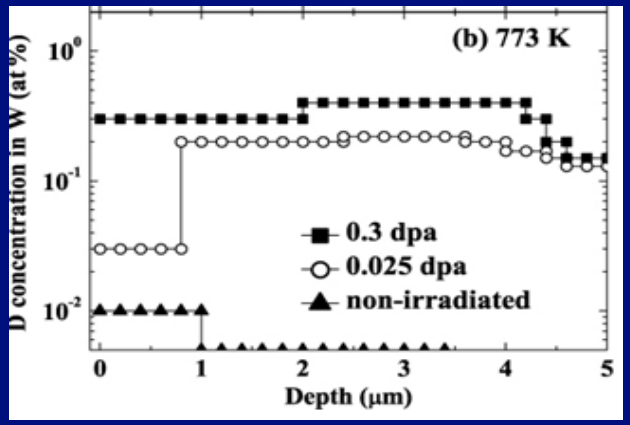
- Highest melting point (3695 K)
- Good thermal conductivity (173 W/m/K)
- Low tritium retention
- Low sputter rate
- Low activation
- …

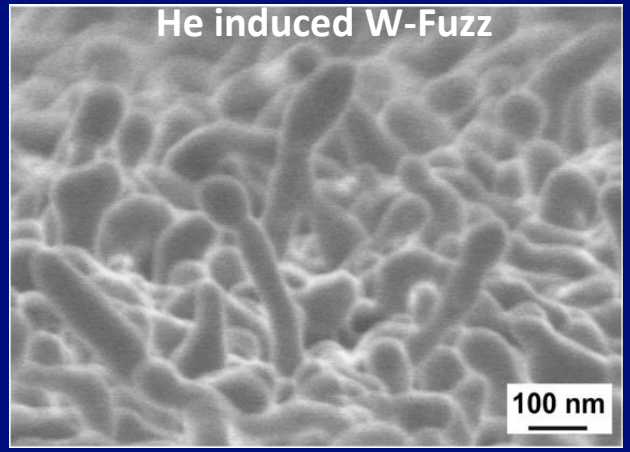
M. Baldwin and R. Dorener, J. Nucl. Mater. **404**, 165 (2010).

A. Hasegawa et al. Materials Transactions, **54,** 466 (2013).

Tungsten: Some uncertainties

T-retention in W

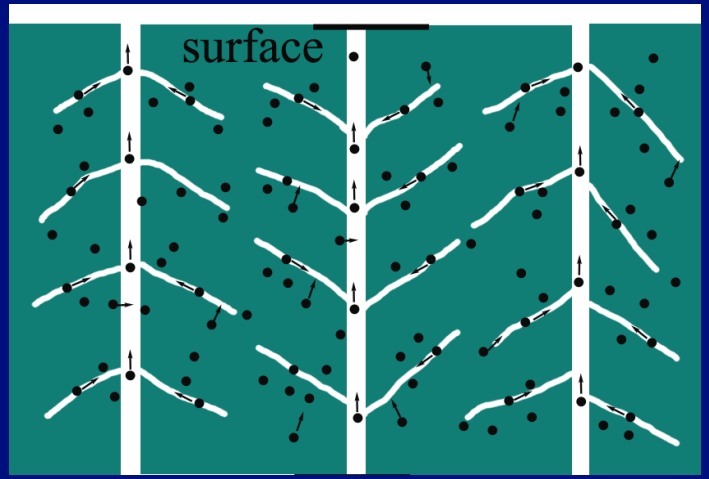




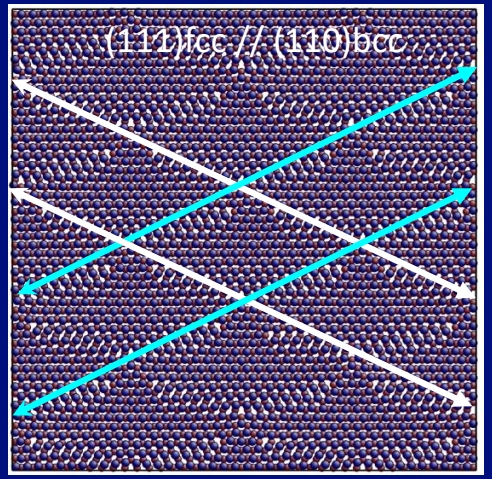
How to manage He release in a more controlled way?

- Root cause: very low solubility of He in metals and alloys
- Reducing net He flux “seen” by the plasma facing material
- Releasing He outside of the material to block the formation of large He bubbles and surface blisters
- Preserving surface morphology and mechanical cohesiveness

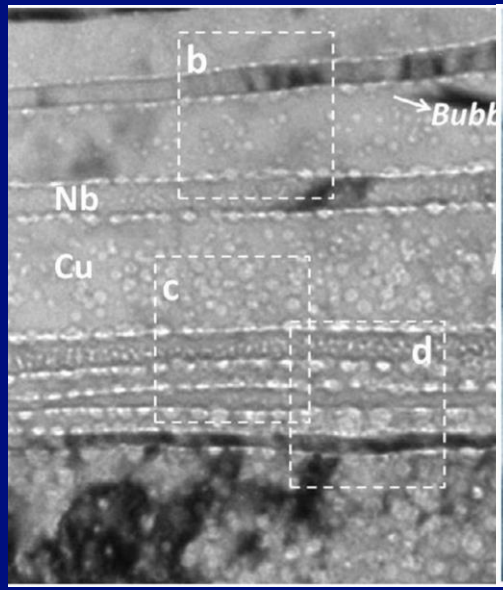
“Vein-like” W-nanochannels are designed and formed during film growth

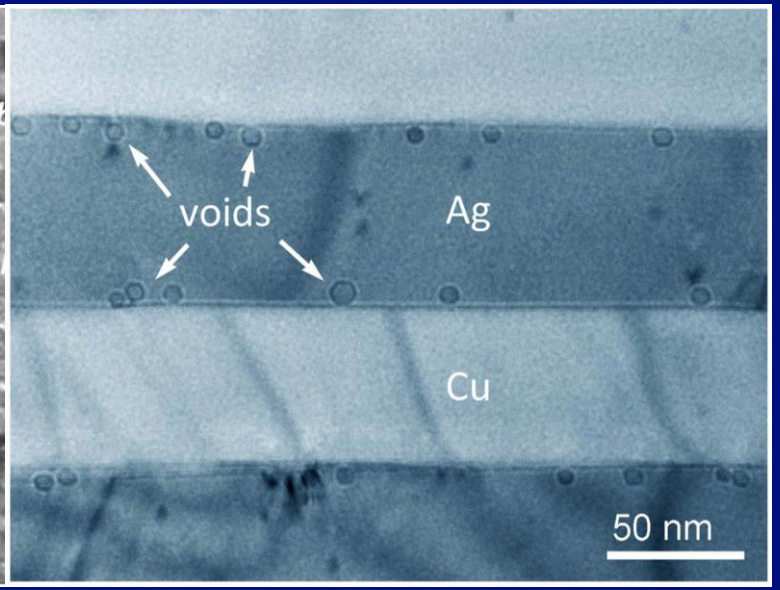


“Vascular” helium nanochannels are formed in operando He exposure



Physical vapor deposited metal multilayers - He bubble distributions are non-uniform





W. Z. Han *et al.*, J. Nucl. Mater. 452, 57 (2014)

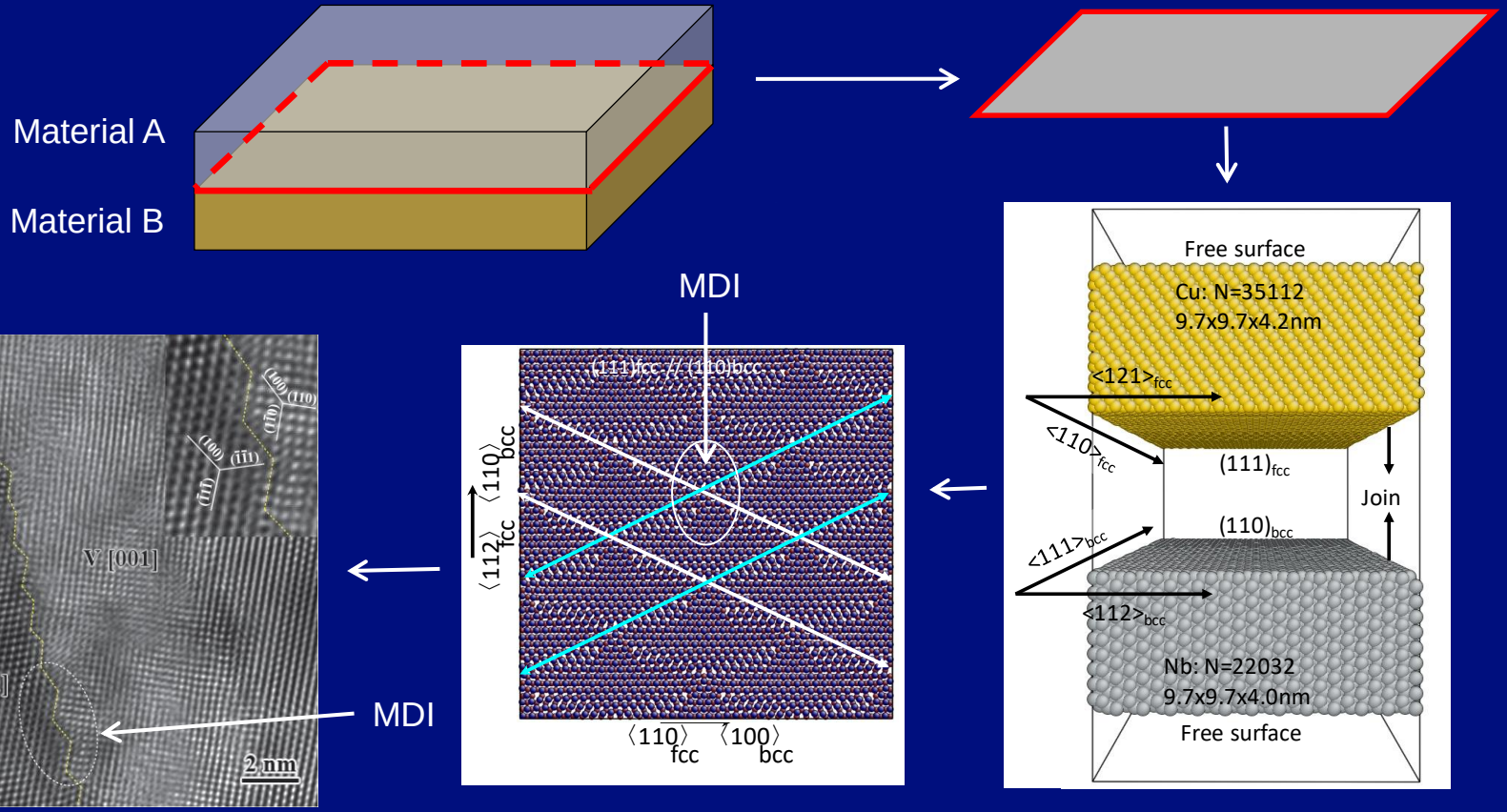
S. Zhang *et al.*, Sci. Rept. 5, 15428 (2015)

Bubbles (and voids) preferentially grow:

- along interfaces;
- into metals with lower vacancy formation energies;
- typically He bubbles are confined in the fcc layer by bcc interfaces;
- typical shape: spherical or multifaceted

Preferential precipitation at specific locations within interfaces

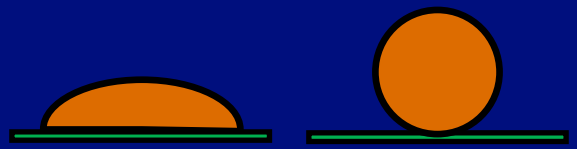
(Courtesy of M. Demkowicz, TAMU)



Interface wetting controls He-cluster growth

Wetting Coefficient:

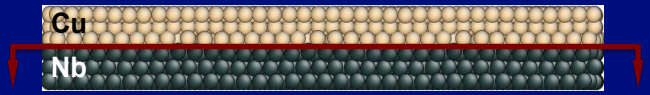
$$W = \gamma_{Cu\text{-}Nb} + \gamma_{He\text{-}Cu} - \gamma_{He\text{-}Nb}$$

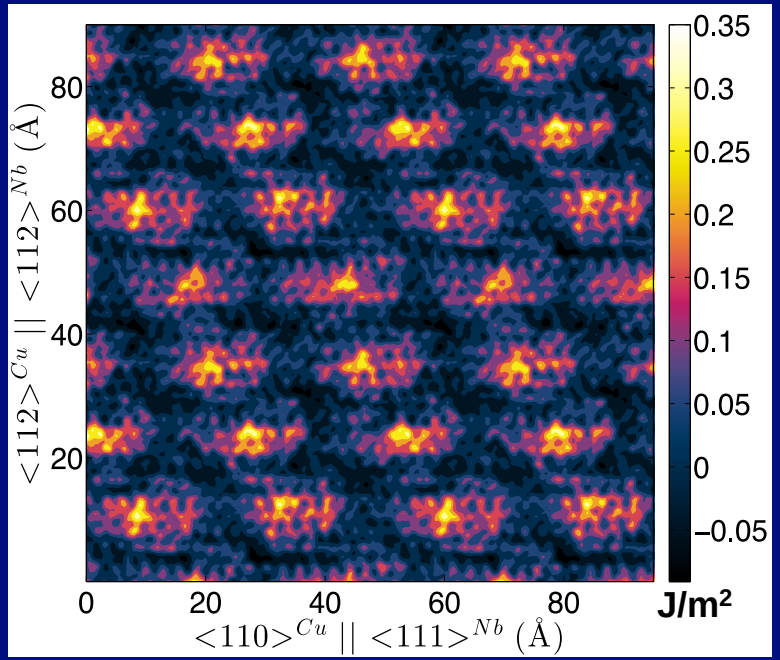


Wetting
$W > 0$

Non-wetting
$W < 0$

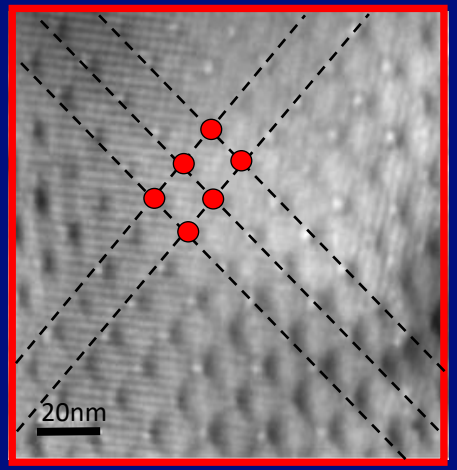
	Interface Energy (J/m^2)
$\gamma_{He\text{-}Cu}$	1.93
$\gamma_{He\text{-}Nb}$	2.40
$\gamma_{Cu\text{-}Nb}$	Depends on location in the interface plane





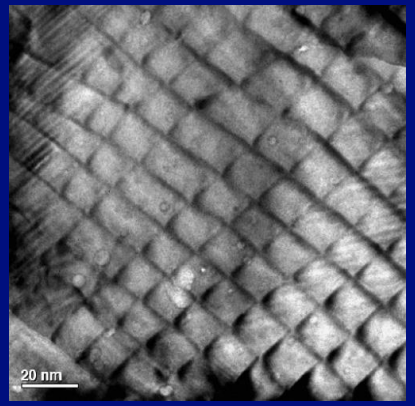
He-V clusters grow by wetting regions of high interface energy

Misfit Dislocation Intersections (MDI) can be designed and fabricated (50 nm Au film)

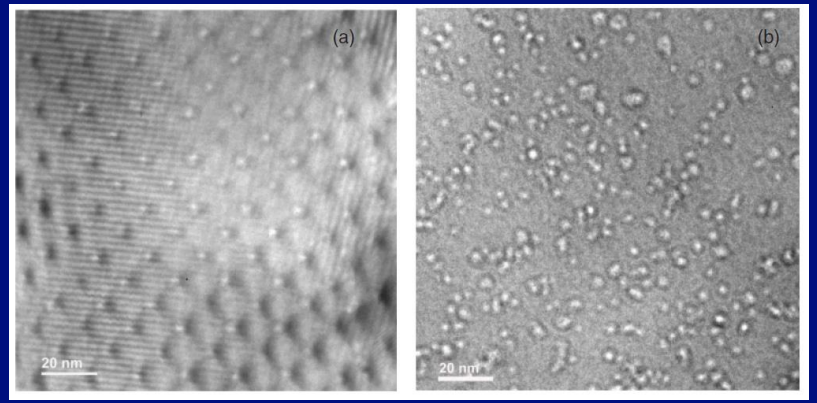


Z. F. Di *et al.*, PRB **84**, 052101 (2011)

Plane view of Au bicrystral: He precipitates at MDIs

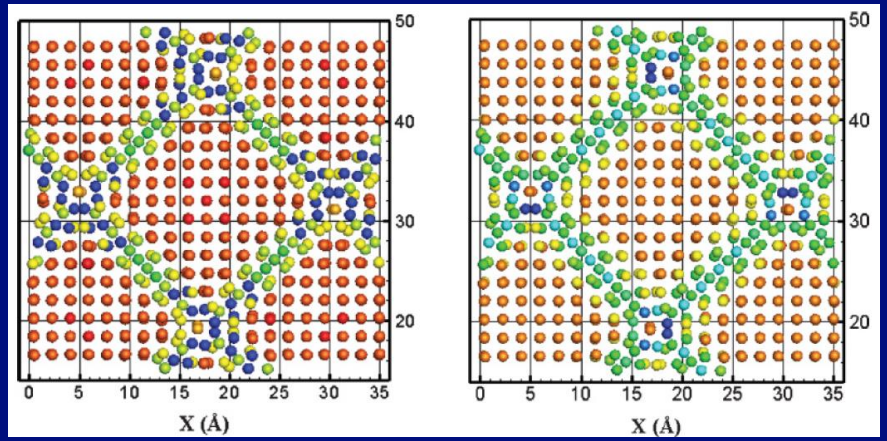


Screw dislocation network and nodes



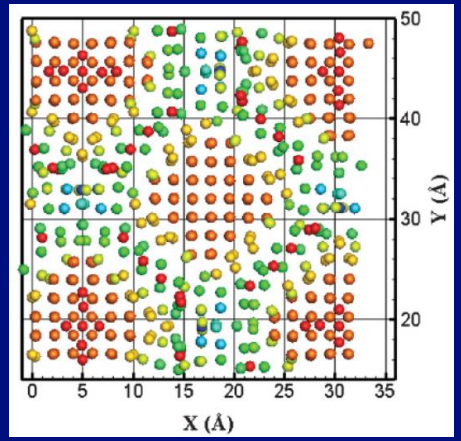
He bubbles formation at nodes

Random He bubbles in Au foil



Vacancy formation energy

He substitutional solution energy



He interstitial solution energy

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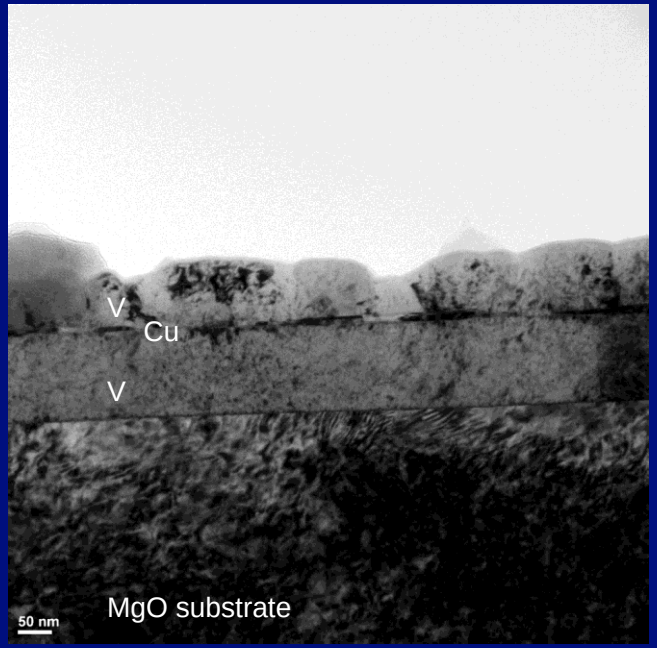
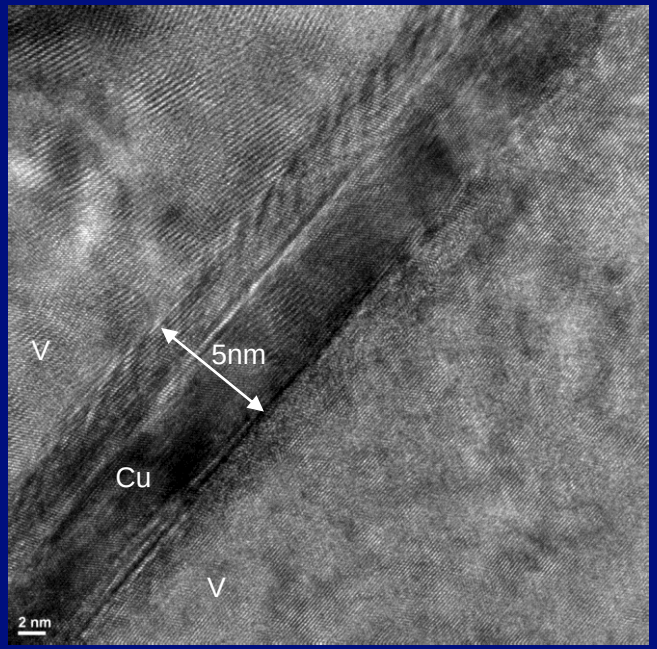
Model material: 5 nm Cu layer confined by two adjacent V layers

Edge-on view



As deposited

Edge-on view, high resolution

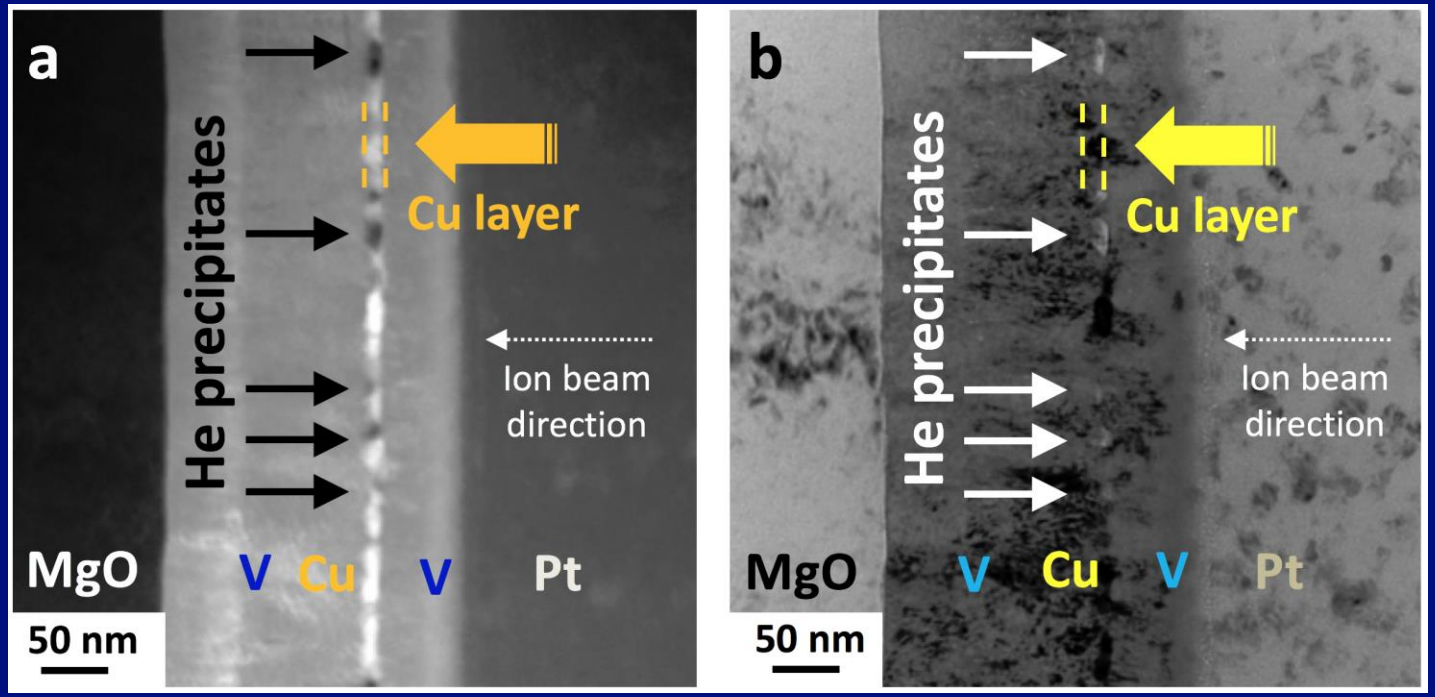


As deposited

Sample is designed to enable plan-view imaging of He precipitates

Preferential He bubble growth in the Cu layer

Edge-on view



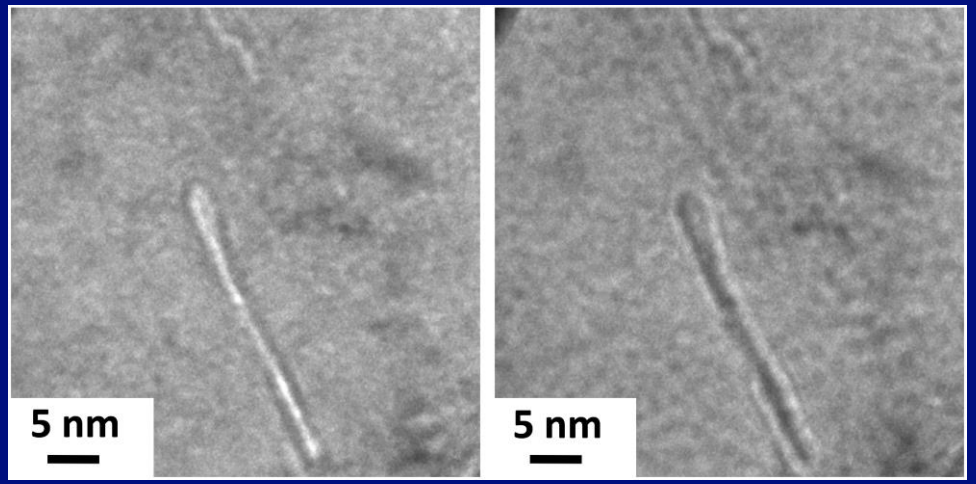
Dark field STEM

Bright field TEM

20 keV He^{+},10^{16} ions/cm^{2}

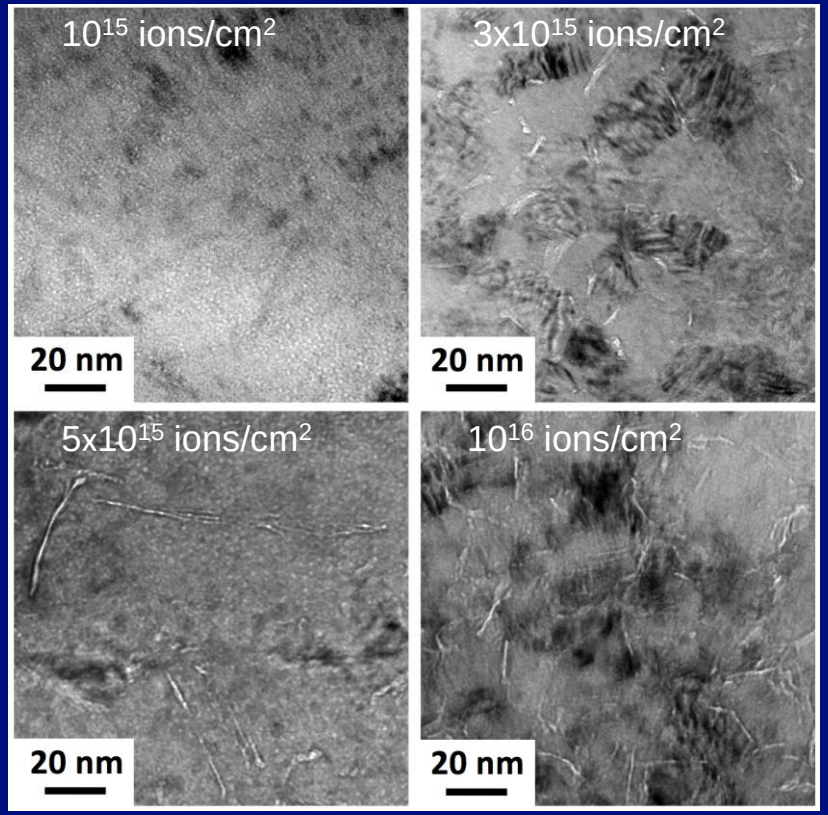
Formation of elongated He "channels": Departure from traditional equaxial bubble formation

The channels really are He precipitates



400 nm under-focus 400 nm over-focus

D. Chen *et al.*, Science Advances 3, eaao2710 (2017)



All images are taken in "plan" view, i.e. looking perpendicular to the Cu layer

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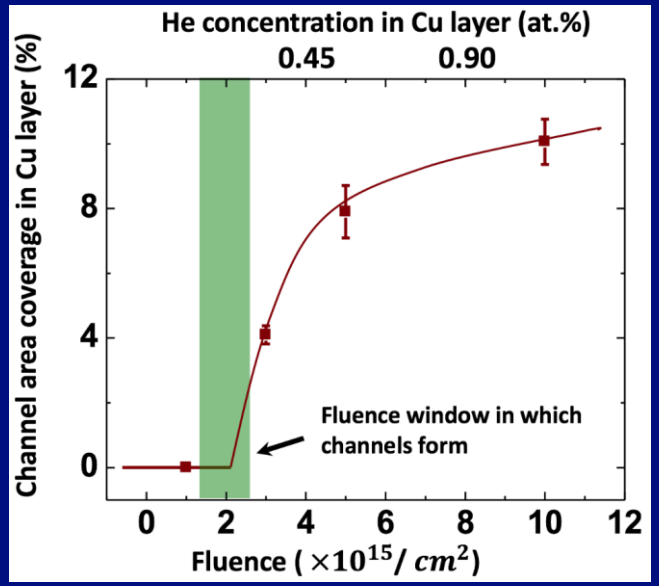
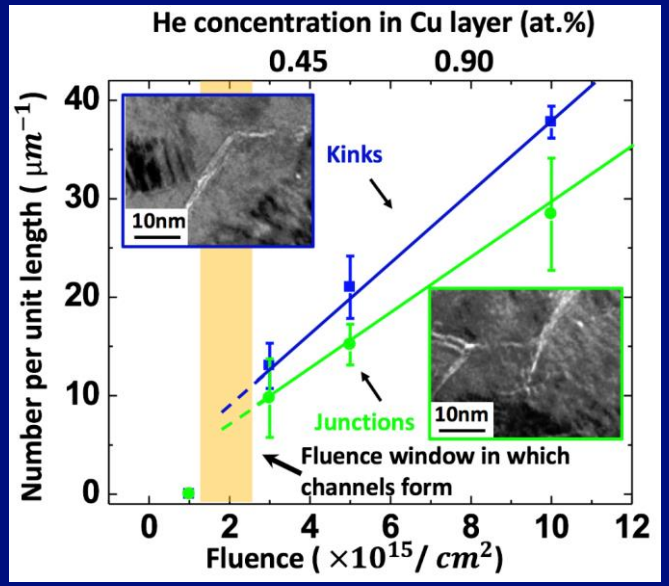
The connectivity of He channels increases with fluence



Asymptotic convergence with increasing fluence to ~12%: bond percolation threshold in 2-D?



Increasing interconnectivity of ligaments leads to the formation of a ”vascular network”?

Does He outgassing in multilayers occur through a vascular network of nanochannels?

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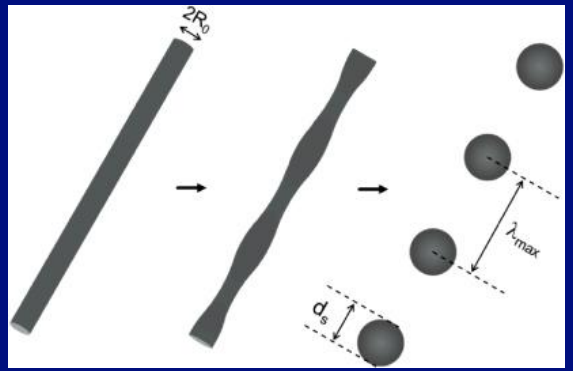
D. Chen *et al.*, Science Advances 3, eaao2710 (2017)

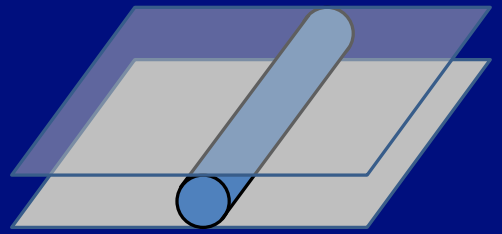
Confinement by adjacent V layers stabilizes He nanochannels

Unconfined cylinders decompose into spheres by Plateau-Rayleigh instability



Decomposition is spontaneous because it has no energy barrier

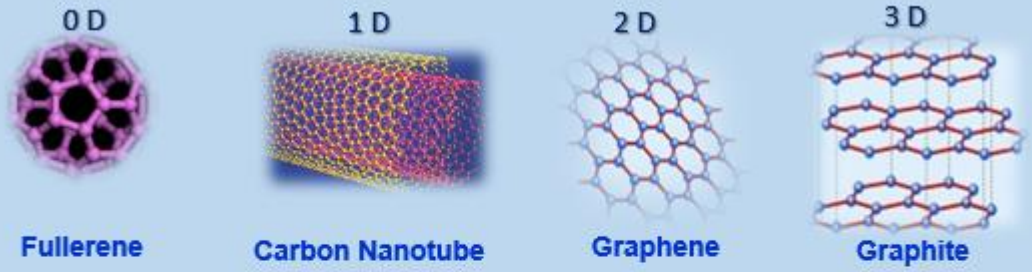
Cylinders pinched between two layers do not undergo the P-R instability



Even through there are lower energy shapes, decomposition is prevented by an energy barrier

Example 2: Ion Beam Synthesis of Layer – Tunable Graphene

Dimensionality of Carbon



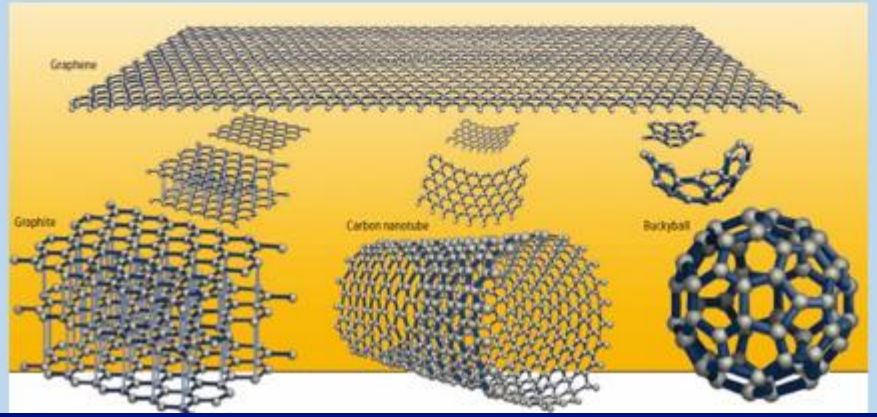
1985 at Rice Univ by Kroto, Curl and Smalley shared Nobel Prize in 1996.

1991 in NEC by Iijima Sumio.

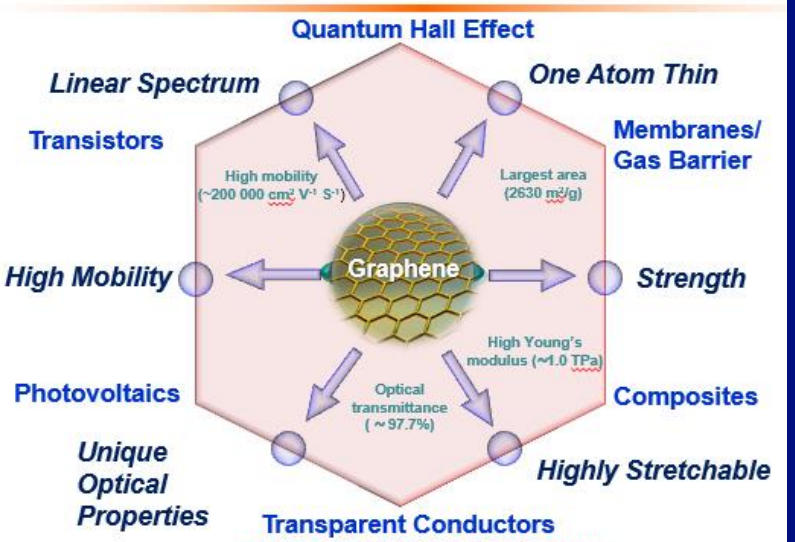
2004 at Manchester Univ. by Geim and Novoselov. Nobel Prize in 2010.

Discovered A.D. 200 in China

K. S. Novoselov, et al, *Science* 306 (2004) 666.



A rich lode of novel fundamental physics



Allen et al. Chemical Reviews 2010, 110, 132-145

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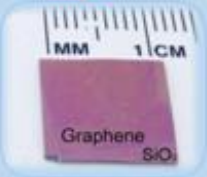
Dr. Z. Di, Shanghai Institute of Microsystems
Mr. K. Baldwin, Los Alamos National Laboratory

Rapid evolution of graphene production by CVD

Continuous film

1*1cm^2 (on Cu)



Science 2009, 324, 1312.

1.5*1cm^2 (on Ni)

Nature 2009, 457, 706 .

20 inch (Roll to roll)

Nat Nanotechnol 2010, 5, 574.

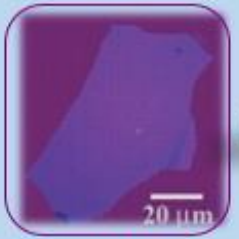
294 mm width (low temp)

Carbon 2012, 50, 2615.

100-m-long(Roll to roll)

Appl.Phys. Lett, 2013, 102, 023112.

Mechanical Exfoliation

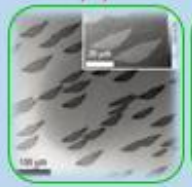


Science 2004,306, 666.

2004…… 2008 2009 2010 2011 2012 2013 2014

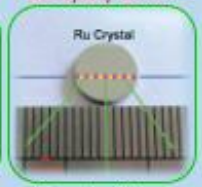
Single crystal

Nat Mater 2008, 7, 406.



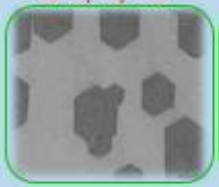
Micrometer-size (on Ru)

Adv. Mater, 2009, 21, 2777.



Diameter 8 mm (on Ru)

Nat Mater 2011, 10, 443.

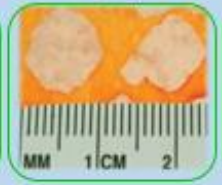


Micrometer-size (on Cu)

Nat Commun, 2012, 3, 699.

Millimetre-size (on Pt)

Science 2013, 342, 720.



Centimeter-size (surface oxygen)

Science 2014, 344, 286.

Wafer-scale (on Ge)

CVD Challenges

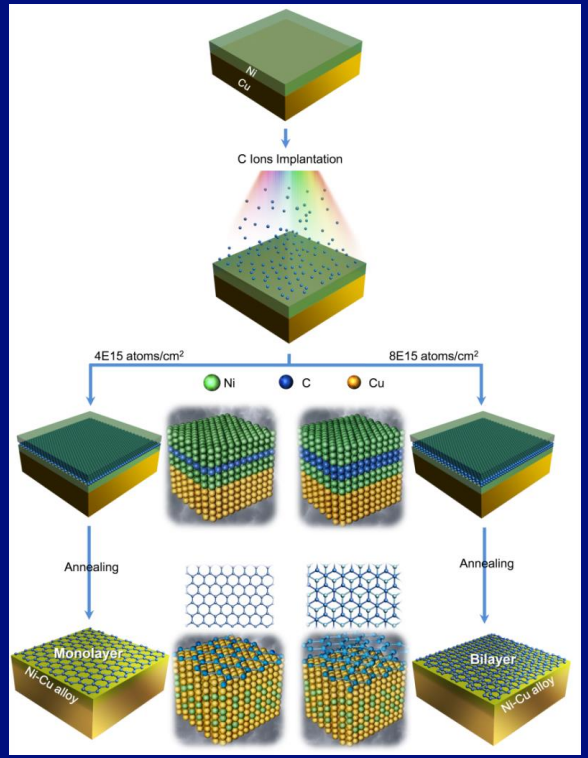
Layer-tunable?

Layer-Transfer?

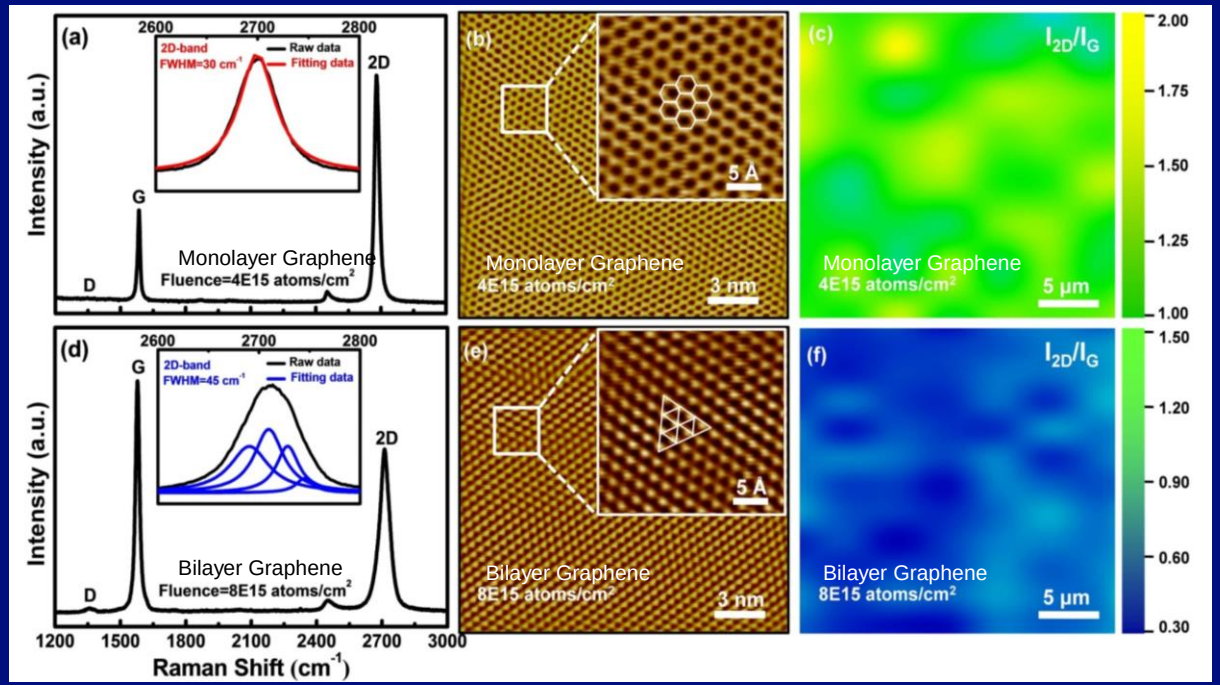
Ion Beam Synthesis of Layer-Tunable Graphene:

A Combined Kinetic Implantation and Thermal Ejection Approach

Smart Janus Substrate: Ni-Cu
Finite C-solubility in Ni
Very Low C-solubility in Cu



G. Wang et al. Adv. Funct. Mater. 2015, 25, 3666.

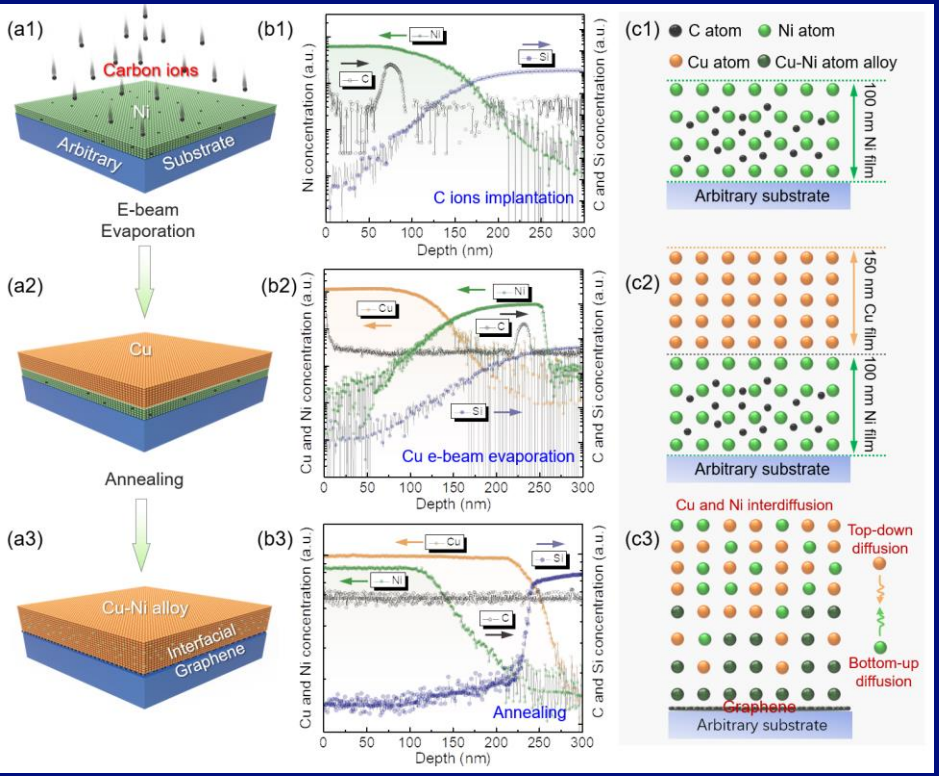


Excellent quality of monolayer and bilayer graphene as characterized by Raman Spectroscopy and Scanning Tunneling Microscopy (STM)

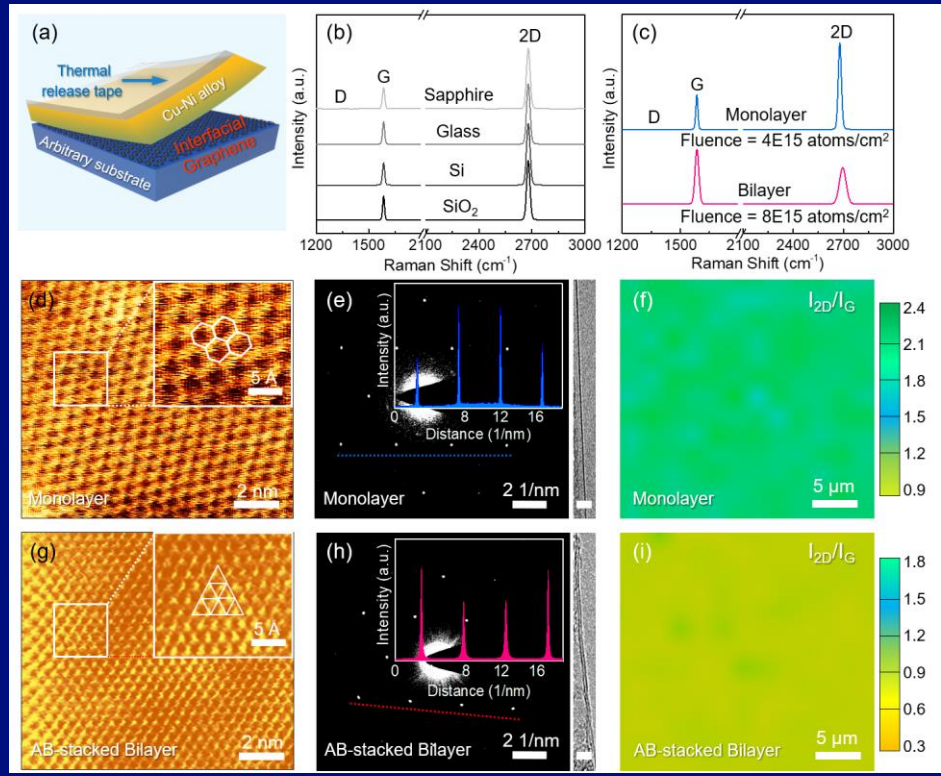
Synthesis of Layer-Tunable and Transfer-Free Graphene on Arbitrary Substrates

Smart Janus structure: Ni-Cu-Sub to Cu-Ni-Sub

Formation mechanism of graphene



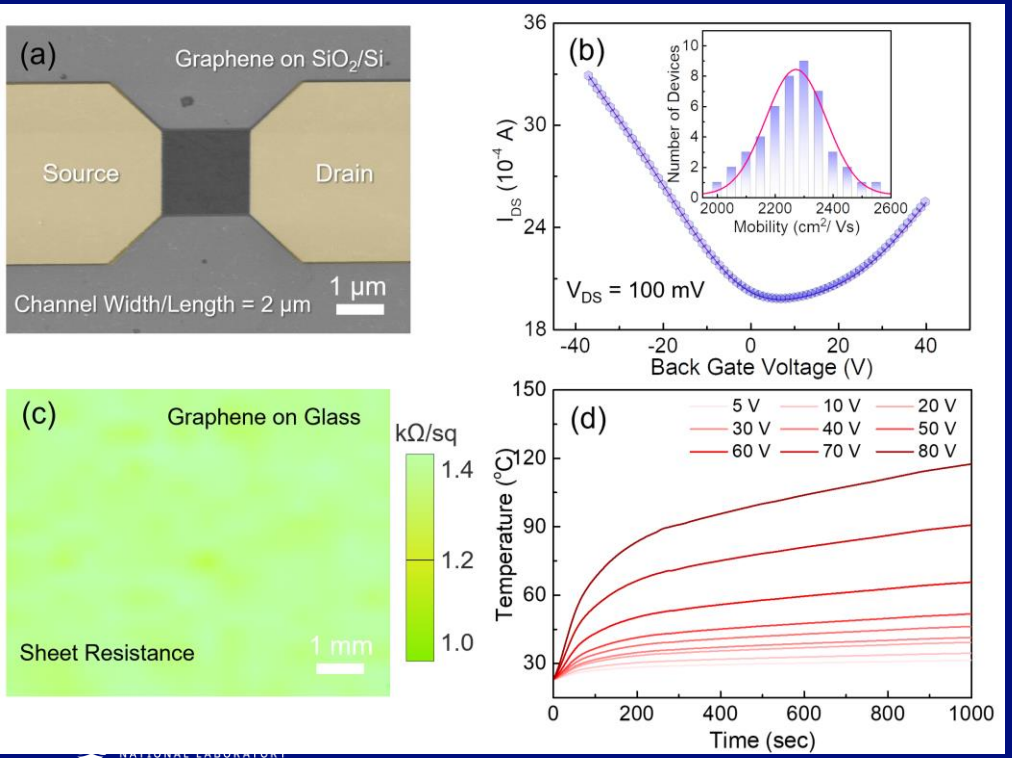
Graphene characterization

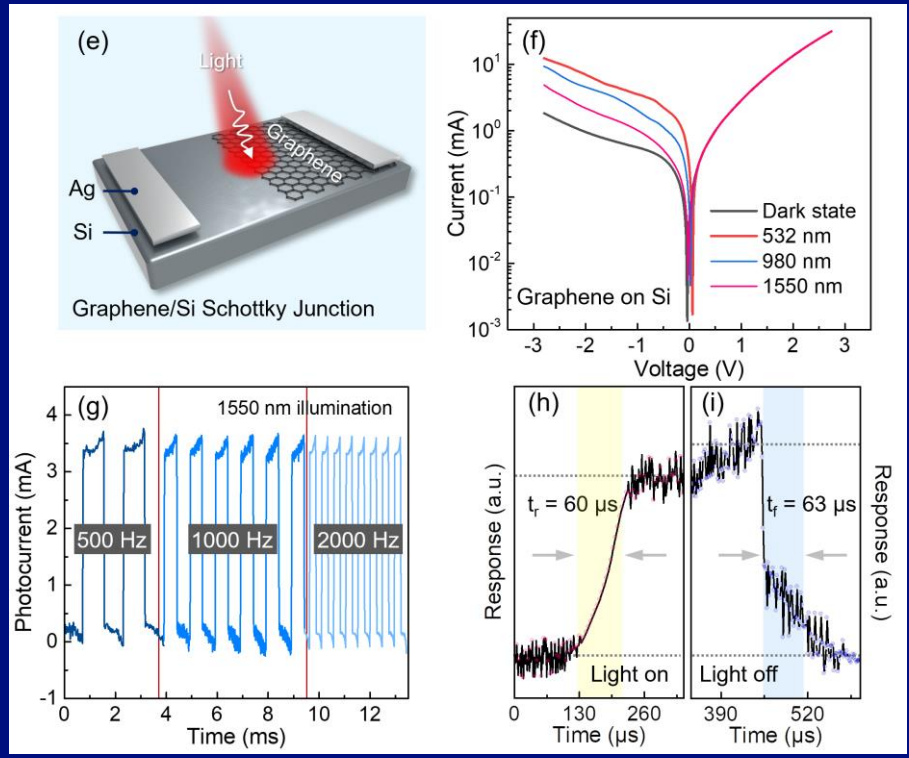


Excellent Properties and Versatile Applications

(a-b) High quality field-effect transistors
(c-d) Large area heating devices

(e-i) Near Infrared photodetectors with superb detection sensitivity

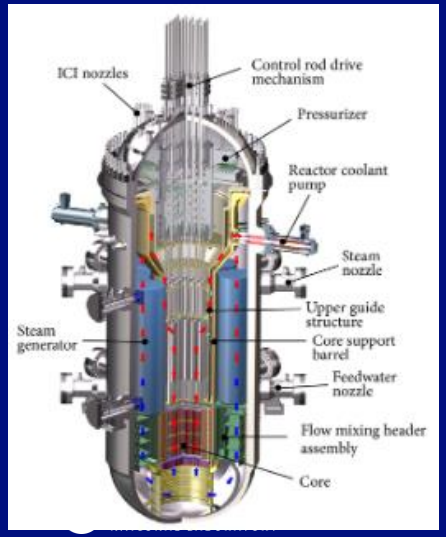


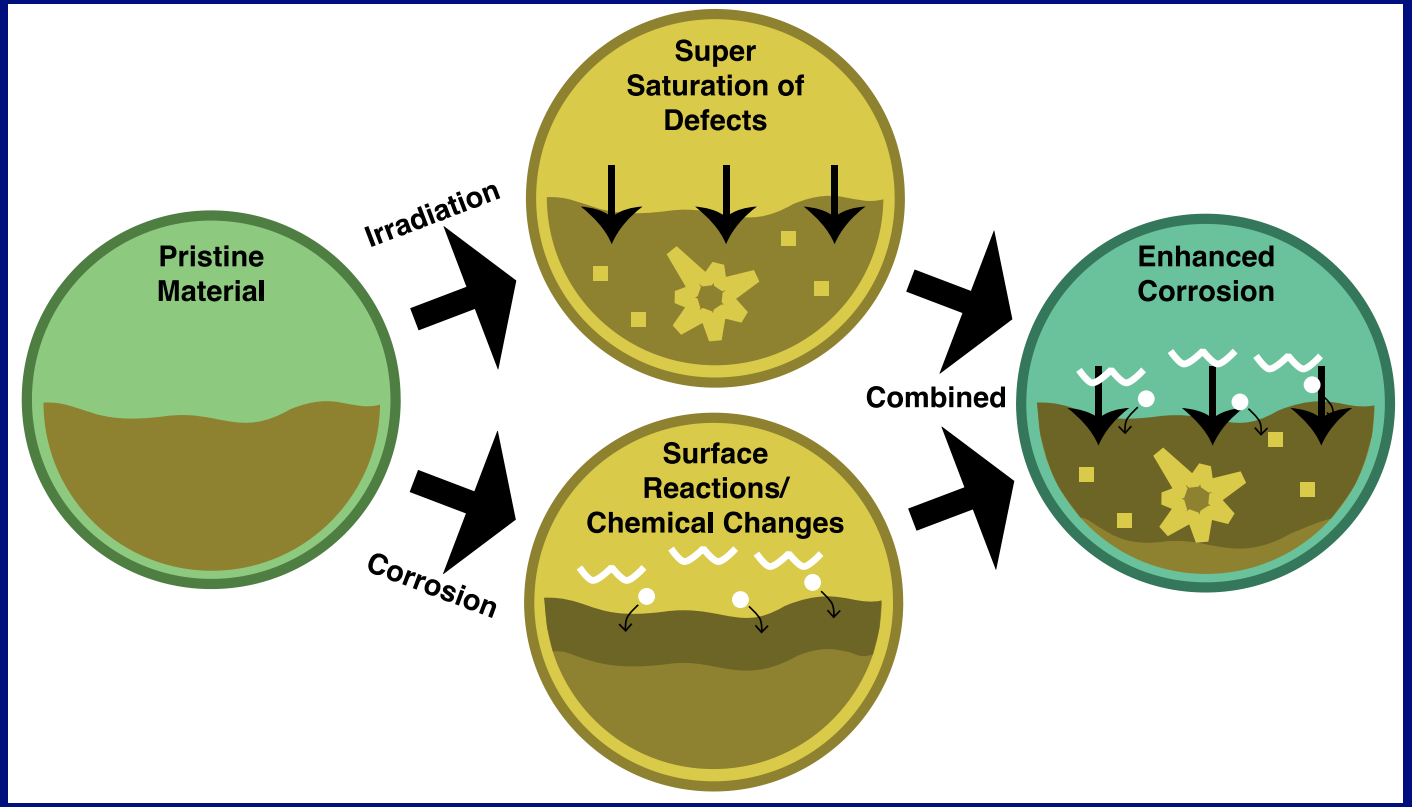


Example 3: In Situ PIXE Monitoring of Corrosion under Irradiation

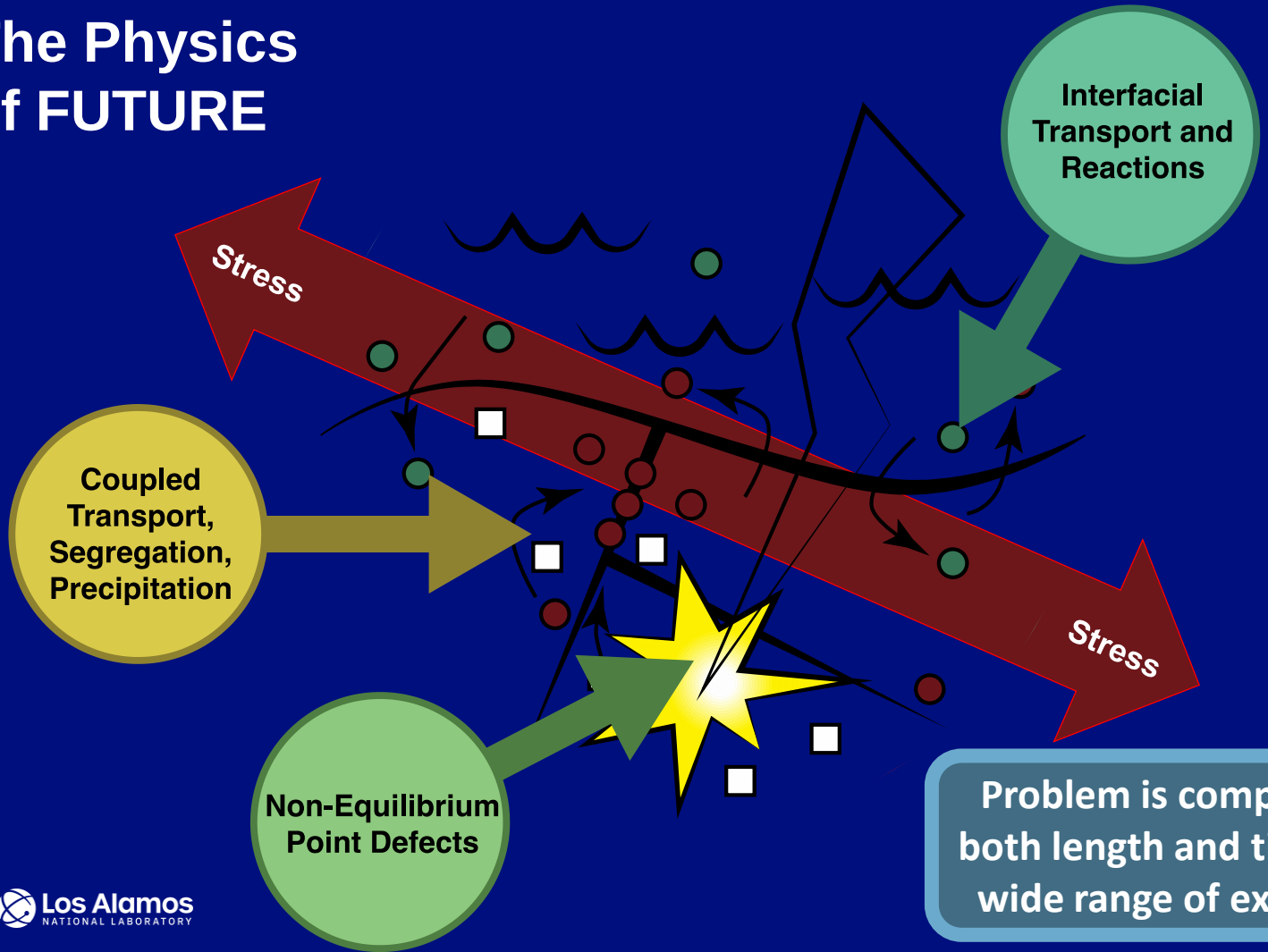
Our Goal: To understand how the coupled extremes of irradiation and corrosion work in concert to modify the fundamental mechanisms on the evolution of materials by coupling experiments and modeling

Nuclear Reactor Core



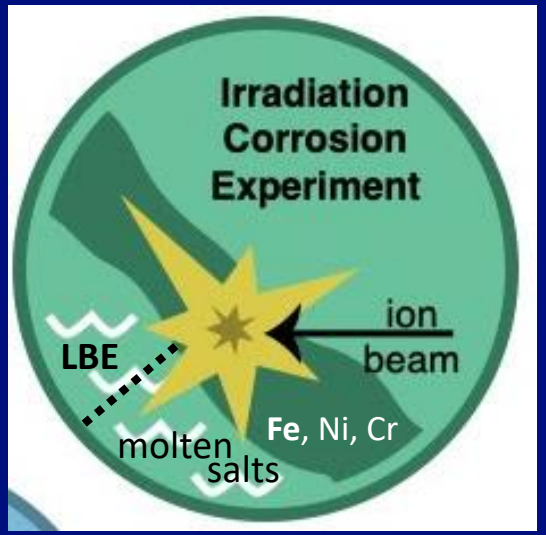


The Physics of FUTURE



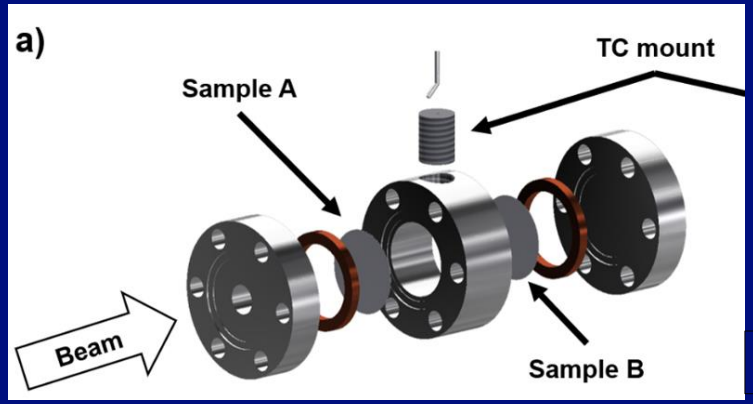
Problem is complex, multi-scale in both length and time, and requires a wide range of expertise to address

The Irradiation-Corrosion Experiment (ICE): 4 MeV protons to $3x10^{23}$ m^{-2}@450 C

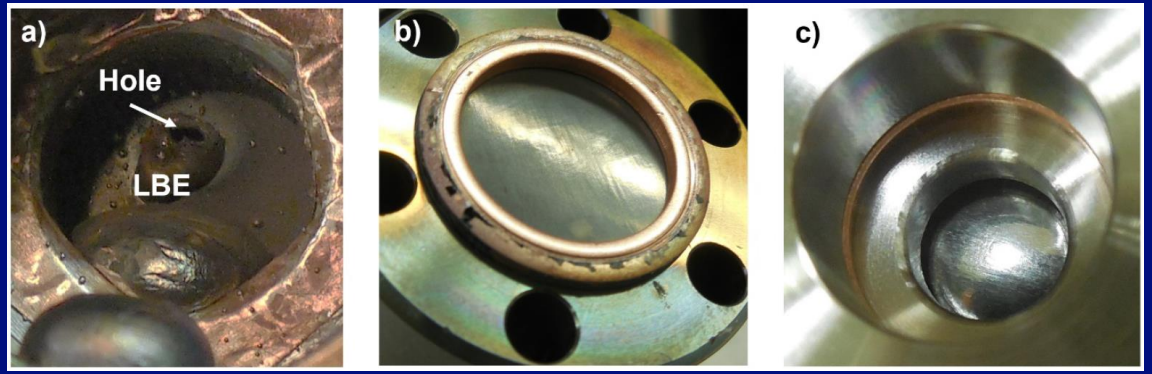


Goals:

- Produce thin (<50 μm) simultaneously irradiated and corroded samples for fast analysis
- Implement in situ corrosion monitoring capability to provide early warning of long hour ICE experiment
- Improve understanding of irradiation-corrosion interactions in model materials



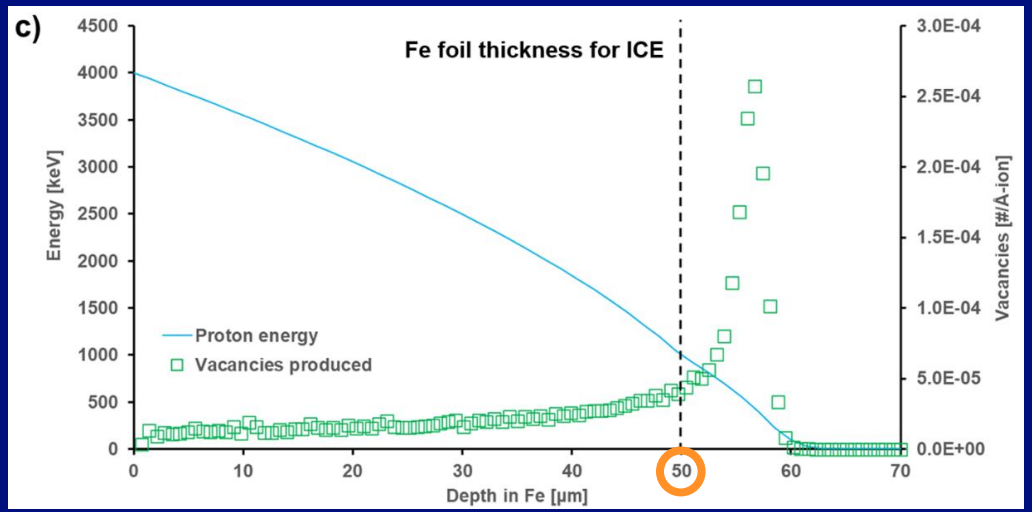
a) Sample A after 56 hrs, b) Sample B after 56 hrs, c) Sample A after 22 hrs.

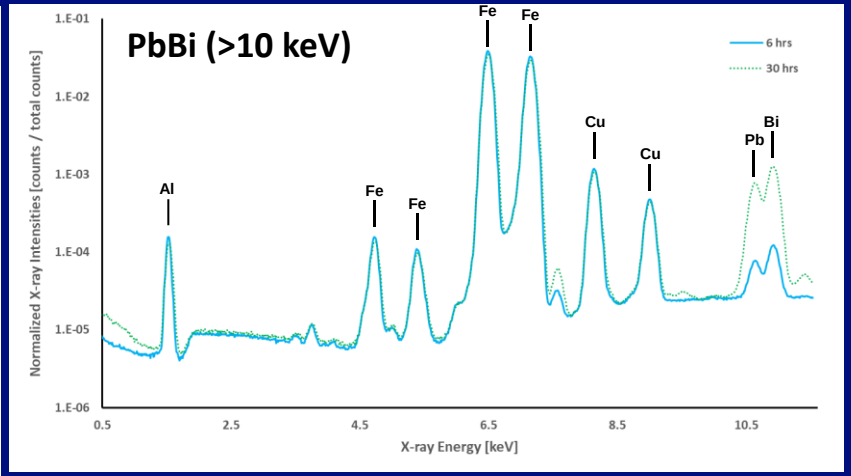


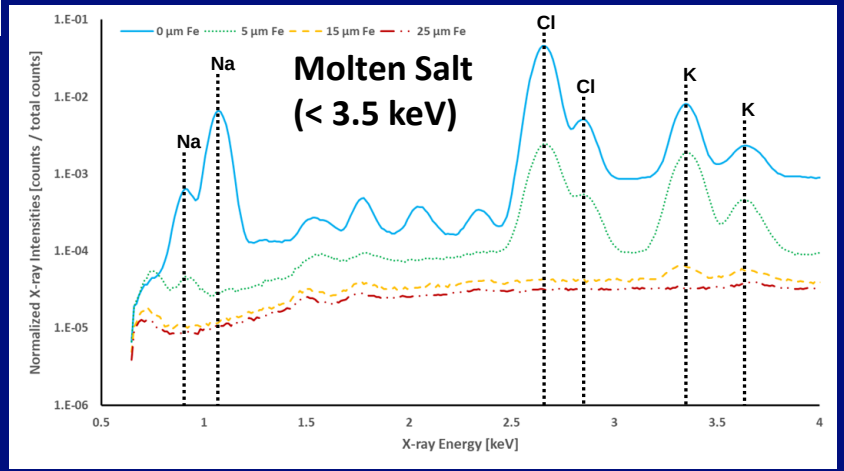
Corrosion-only experiments at 450 °C up to 200 hours showed intact foils

Proton energy loss and damage profile

F. Schmidt et al. JOM, 12 (2021) 4041.







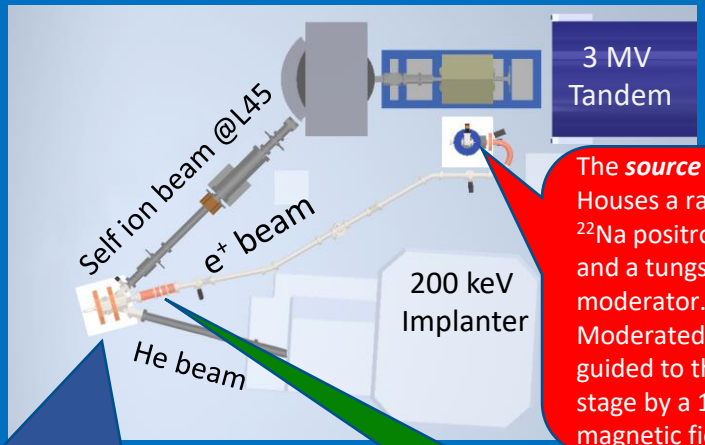
Protons have ~1 MeV left when entering LBE
→ can excite Pb and Bi X-rays

PbBi X-rays have 10-15 keV
→ 0.1-10% can pass through 50 µm of Fe
→ Proton-induced X-ray emissions (PIXE) should show relative quantity of Fe and PbBi atoms in beam path

Outlook: New Capabilities

In Situ Positron Annihilation Spectroscopy to characterize radiation damage during irradiation

Dual Beams Coupled to Positron Beam for PAS



The ***source stage***: Houses a radioactive ^{22}Na positron (e^+) source and a tungsten foil moderator. Moderated positrons are guided to the target stage by a 100 G axial magnetic field.

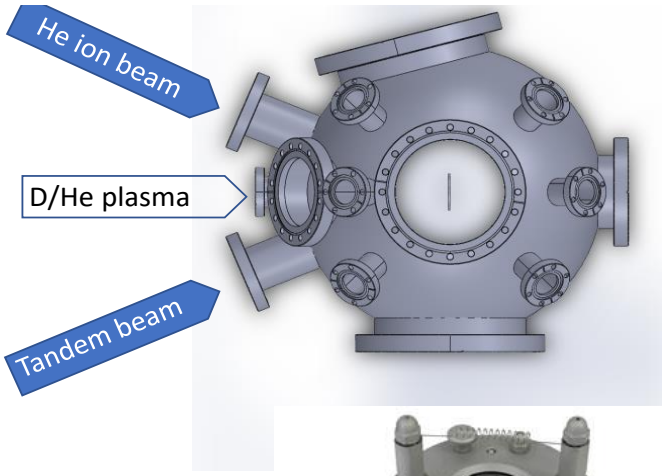
A ***target chamber*** for Positron Annihilation Spectroscopy (PAS) with capability to study a target material under single or dual ion irradiation.

A HV bias on the target accelerates e+ beam to varying energies, providing a depth resolved probe.

The moderated positron beam will be fed into a ***RF bunching system*** to form pulses with sub-nanosecond widths for lifetime spectroscopy.

Tribeam to mimic fusion prototypic condition for PMI research

Dual Beams Coupled to Plasma Gun



EH400 Plasma Source:
Energy: 40 – 300 eV He
Flux: 1.5×10^{21} ions/m^2/s

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Edwards Bielejec and Khalid Hattar

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Z.F. Di, Shanghai Institute of Microsystems
N. Li, Los Alamos National Laboratory
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P. Hosemann, University of California - Berkeley

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26th International Conference on the Application of Accelerators in Research & Industry
and
53rd Symposium of Northeastern Accelerator Personnel

October 30 to November 3, 2022

Hilton Embassy Suites in Denton, Texas, USA