TEM with *in situ* Ion Irradiation of Nuclear Materials: The IVEM-Tandem User Facility

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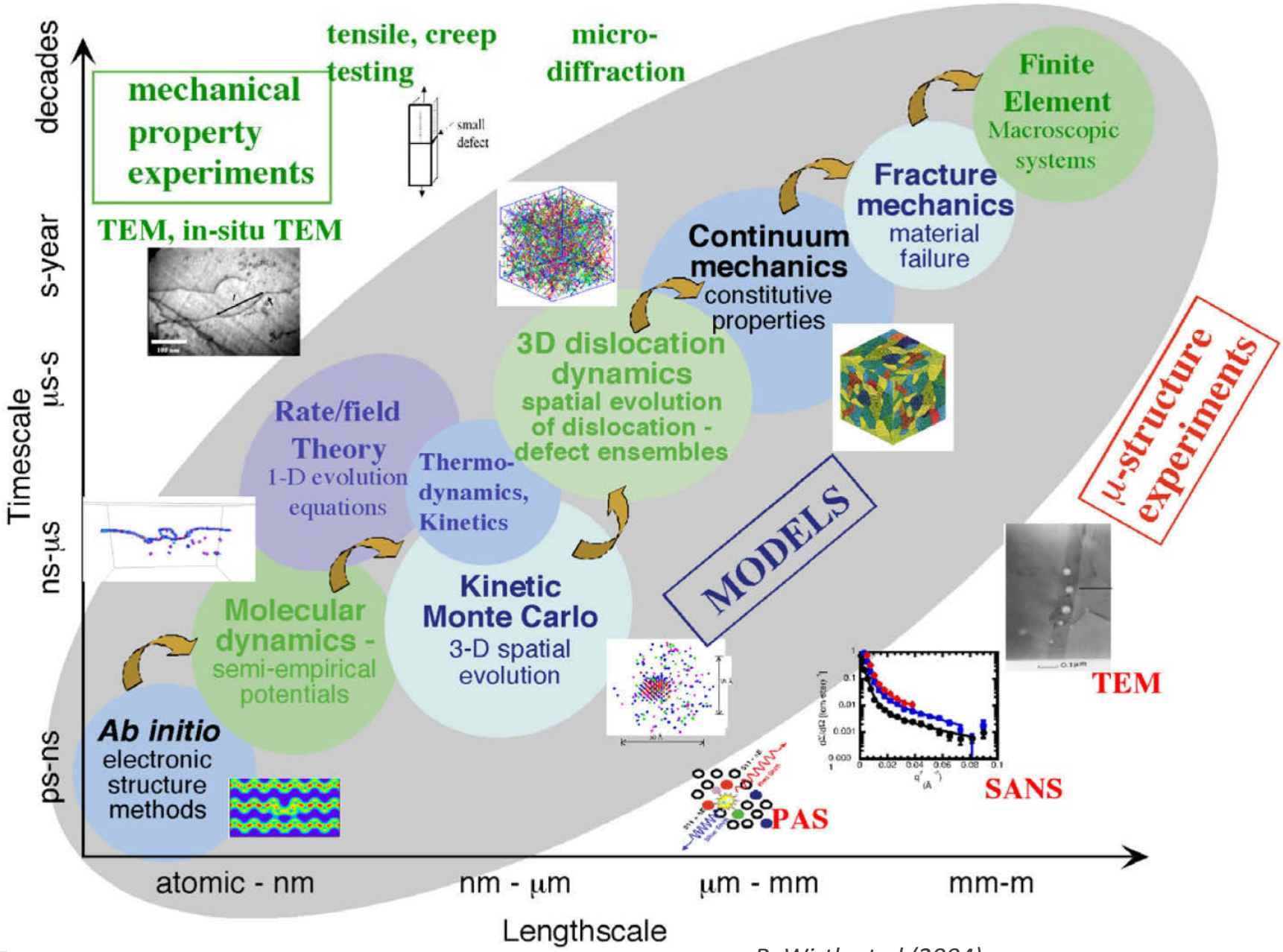
Argonne National Laboratory

Nuclear Science User Facilities (NSUF) Annual Review

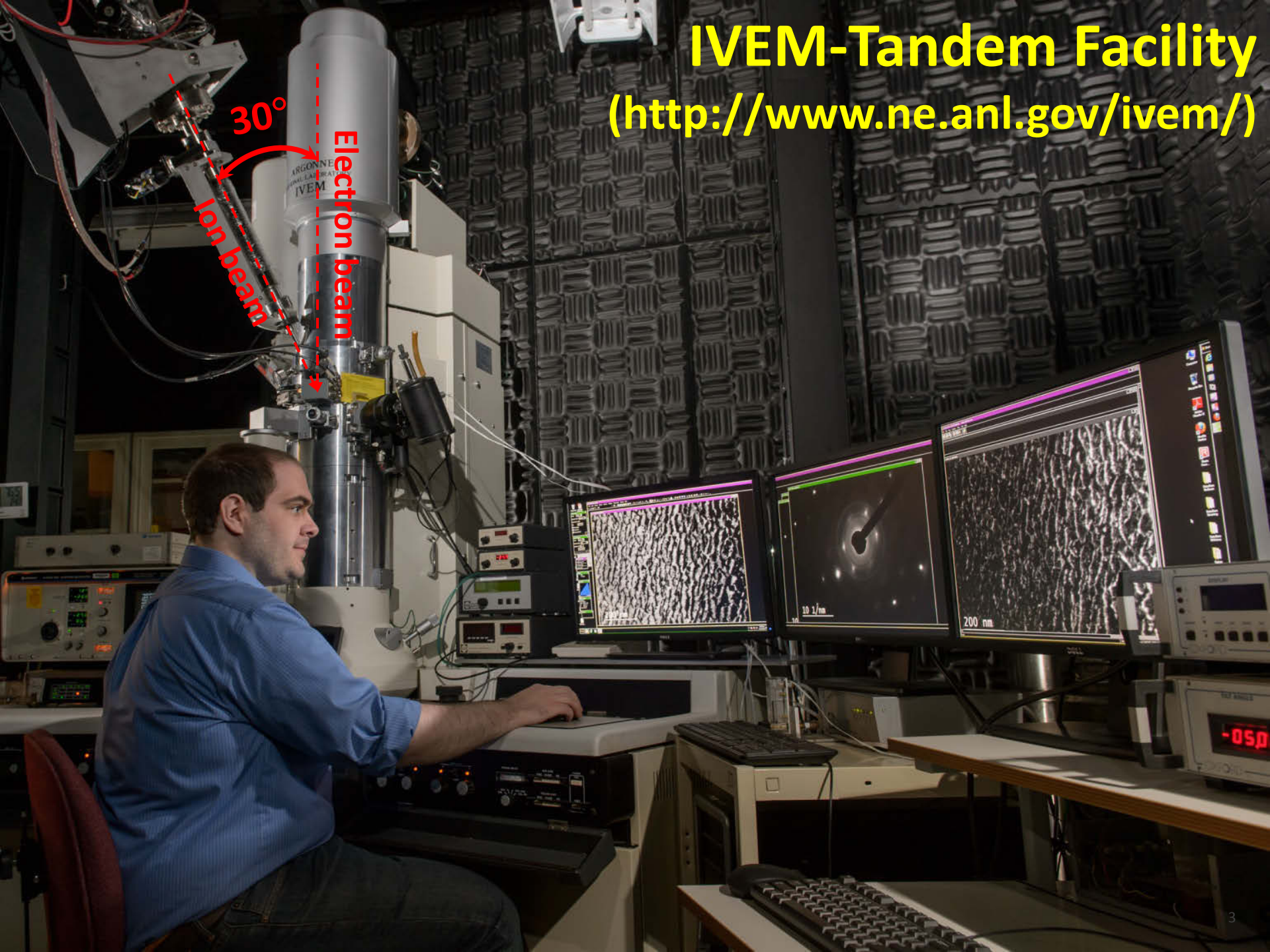
Department of Energy, Germantown, MD

March 1-3, 2016

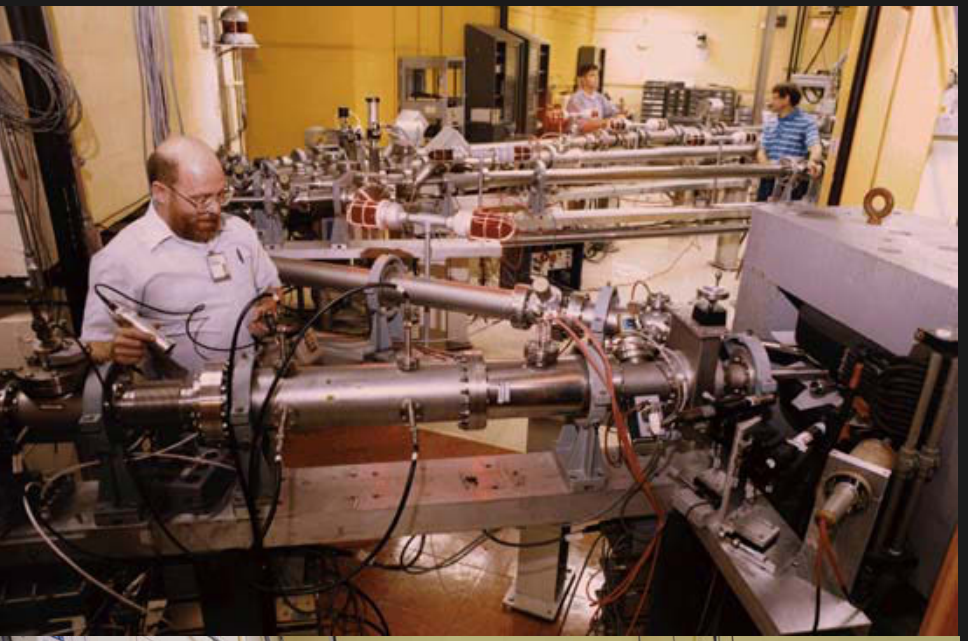


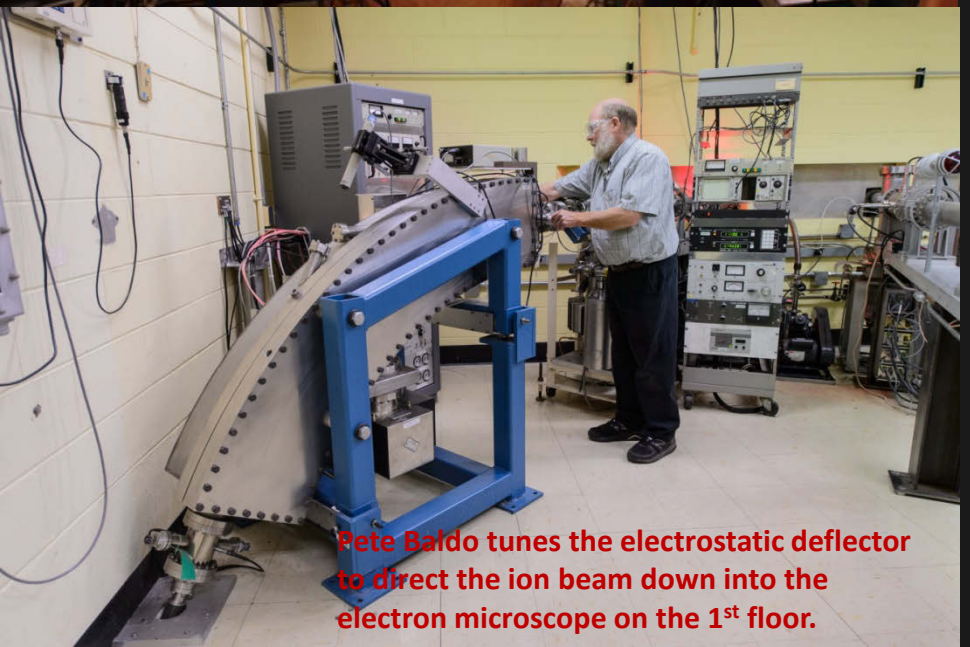
B. Wirth et al (2004)

IVEM-Tandem Facility
(http://www.ne.anl.gov/ivem/)



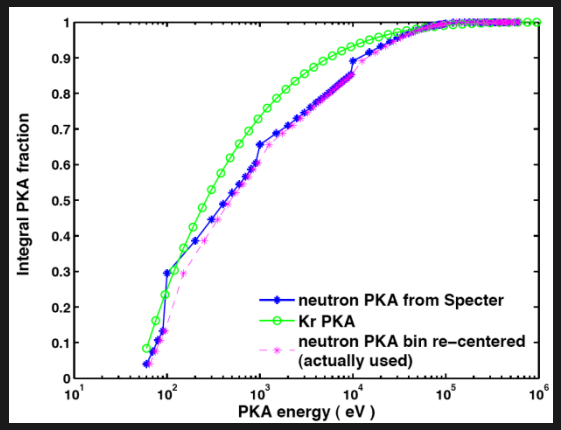
Ion Irradiation Capability





Pete Baldo tunes the electrostatic deflector to direct the ion beam down into the electron microscope on the 1st floor.

Beam Energy	50 keV – 1 MeV
Ion Types	H, He, Kr, Xe, and many elements from Al to Au
Ion Flux	10^{10} – 10^{12} ions/cm^2/s (10^{-6}–10^{-4} dpa/s)
High dose	~100 dpa/day
Beam size	Uniform beam of 1.5 mm in diameter on sample center
Accurate Dosimetry	Real-time dosimetry with Faraday cup in the microscope column

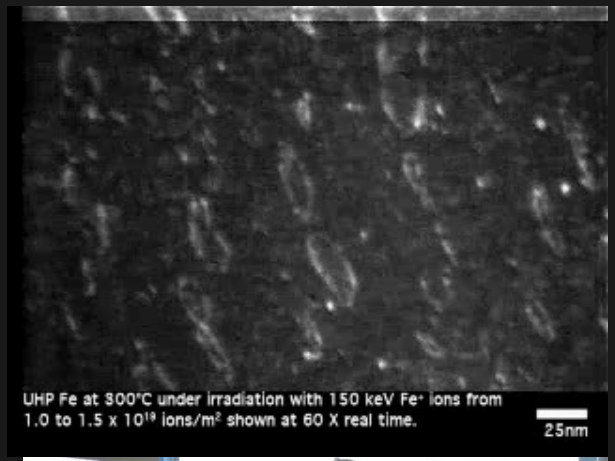


1 MeV Kr ions create similar primary knock-on atom (PKA) spectrum to neutron irradiation in HFIR

What Distinguishes IVEM-Tandem from Other Ion Irradiation Facilities:

In situ TEM during Irradiation

Real-time imaging	• **Spatial resolution: 2.5 Å point resolution** (nanoscale defect imaging • **Time resolution: 5 ms** (*in situ* image recording, 200 frames/sec)
***In situ* heating/cooling**	**-253°C (20 K) to 1300°C** • Double-tilt LT stage(-253-20°C) • Double-tilt HT stage (20-900°C) • Single-tilt HT stage (20-1300°C)
***In situ* straining**	• **Straining stage (20-600°C)**
3D Characterization	• **Triple axis specimen holder for tomography**
Precision control of experiment conditions	• **Controlled metallurgical variables** – Constant specimen area – Crystal orientation – Single microstructural feature • **Controlled irradiation variables** – Ion type, energy – dose rate, dose • **Controlled irradiation temperature**



TEM Specimen Holder Vacuum Storage Station

Supporting Infrastructure

- ***In situ*** **ion irradiation at the IVEM-Tandem does not activate samples, which is an advantage.**
- **Supporting infrastructure for handling radioactive samples is available for *in situ* ion irradiation of radioactive samples**
  - Neutron-irradiated steels
  - Nuclear fuels: U, U-Mo, UO_2, etc.
- **Irradiated Materials Laboratory (IML)**
  - Radiological Facility in the same building
  - Argonne Nuclear Engineering Division operates both IVEM-Tandem and IML
- **Radioactive sample preparation**
  - Electro-polishing (Tenupol-5) of radioactive TEM specimens in IML
- **Irradiated Material Characterization at the Advanced Photon Source**

Irradiated Materials Laboratory

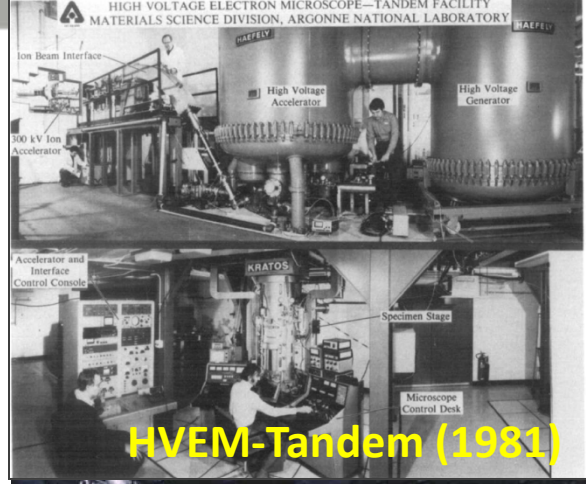
Electro-polishing of radioactive specimens in IML

Advanced Photon Source

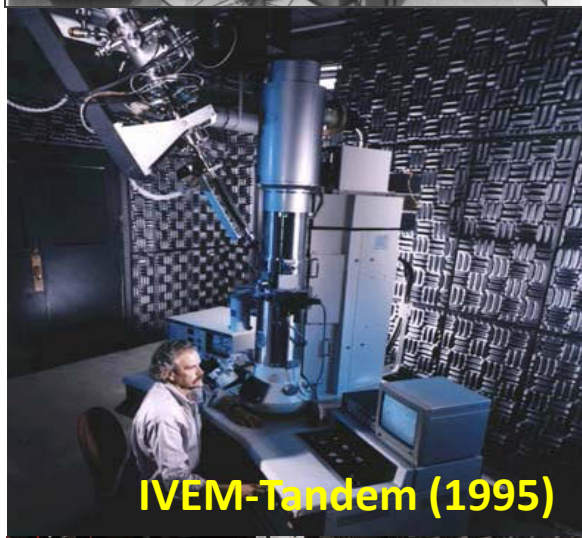
Facility History and Current Status

Has been a User Facility since 1981.

- The 1st generation, **HVEM-Tandem** user facility was commissioned in 1981 in Materials Science Division, ANL, supported by DOE BES.
  - A high-voltage electron microscope (HVEM) interfaced with two accelerators (2 MV tandem and 650 kV ion implanter).
- The 2nd generation, **IVEM-Tandem** was commissioned in 1995, and was part of Argonne's Electron Microscopy Center supported by DOE BES until 2014.
  - Increased imaging spatial resolution by nearly one order of magnitude
- DOE NE and ANL signed Memorandum of Agreement in May 2014 for full time operation of IVEM-Tandem to support nuclear energy research.
- The Facility was transitioned to Nuclear Engineering Division, ANL in June 2014.
- In FY 2016, the IVEM-Tandem received 50% DOE NE support through NSUF.



HVEM-Tandem (1981)


IVEM-Tandem (1995)

IVEM-Tandem (2014)

User Projects in FY15 - FY16

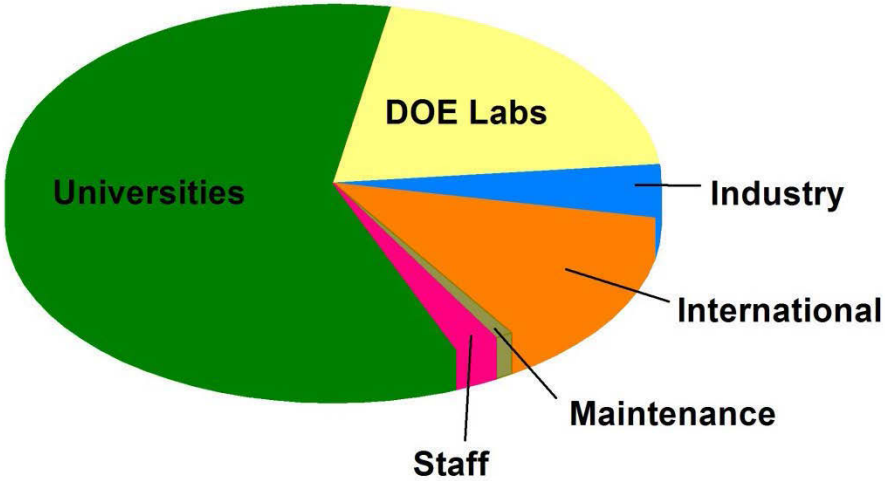
- Areva
- Argonne National Lab
- Australian Nuclear Science and Technology Organization (ANSTO)
- CEA
- China University of Petroleum-Beijing
- Drexel University
- Idaho National Lab
- Los Alamos National Lab
- Louisiana State University
- Massachusetts Institute of Technology
- Michigan State University
- Northwestern University
- Oak Ridge National Lab
- Peking University, China
- Penn State University
- Queen's University
- Rensselaer Polytechnic Institute
- Standford University
- TerraPower
- Texas A&M University
- University of California - Berkeley
- University of Florida
- University of Illinois - Urbana
- University of Nebraska-Lincoln
- University of Oxford
- University of Pittsburgh
- University of South Carolina
- University of Tennessee
- University of Wisconsin - Madison

- **40 IVEM user proposals**
- **FY16 NSUF RTEs:**
  - **1 awarded, 3 under review**
- **Universities, national labs, nuclear industry, international**
- **~80 users**
- **>50% over-subscription**

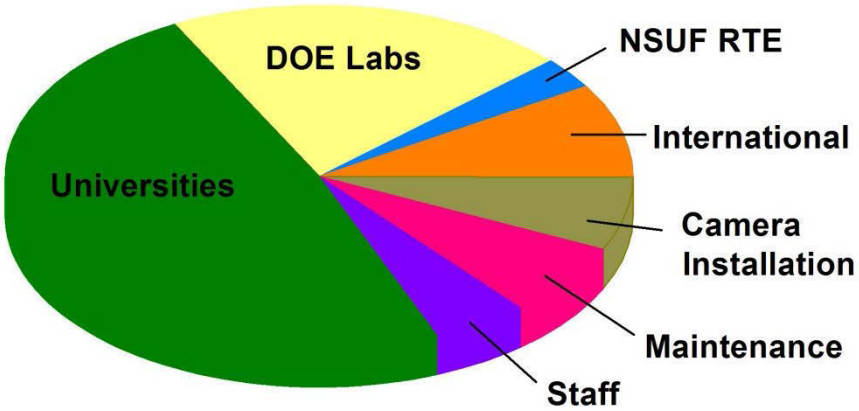
Facility Usage and Productivity

- **Strong Demand**
  - Fully booked for FY2015 and FY2016; >50% over-subscription
- **High Productivity**
  - over **100** publications in the past four years; many of them are published in premier scientific journals with high impact, e.g. Nature, Science
- **Education and Training**
  - In the last four years about **30** PhD student theses were generated using data obtained at the IVEM-Tandem.

FY 2015 (229 operation days)



FY 2016 Cycle 1 (101 operation days)

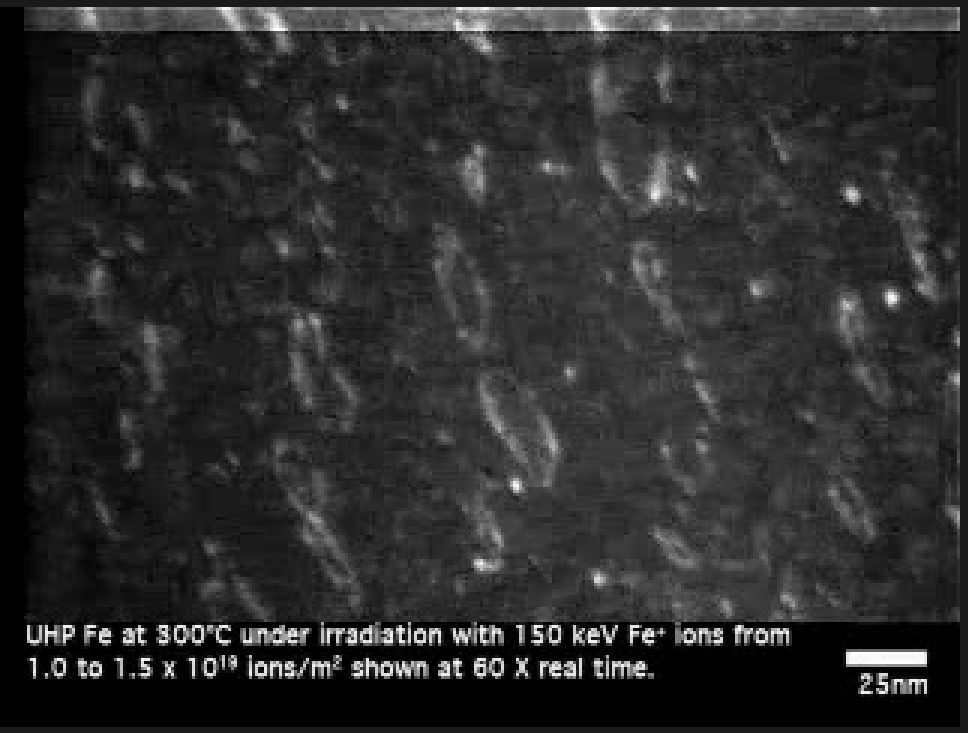


User Research

- **100% of user research is nuclear related, supporting multiple NE programs and nuclear industry needs**
- **Research areas**
  - Radiation-Resistant Nuclear Fuels, Cladding and Structural Alloys, and Waste Storage Materials
  - Advanced accident tolerant materials and fuels
  - New material design concepts (nanostructured materials, high-entropy alloys, etc.)
  - Validation and verification of computer modeling and simulations.
- **Materials studied**
  - Cladding and Structural Materials
    - Ferritic model alloys and commercial ferritic-martensitic steels
    - Austenitic Fe-Cr-Ni model alloys and austenitic stainless steels
    - Ni-based alloys
    - Refractory metals
    - SiC composites
    - Zirconium alloys
    - High entropy alloys
    - Nanostructured materials (ODS steels, Nano-grained Fe, Ni, Multilayers, nanovoids, nano-twins)
  - Nuclear fuels: U, UO_2, U alloys
  - Waste forms: complex oxides, apatite, porochlore, etc.

Unique Experiment – Radiation Defect Dynamics

In situ movie reveals irradiation defect formation, motion, and coalescence to form extended dislocation structure



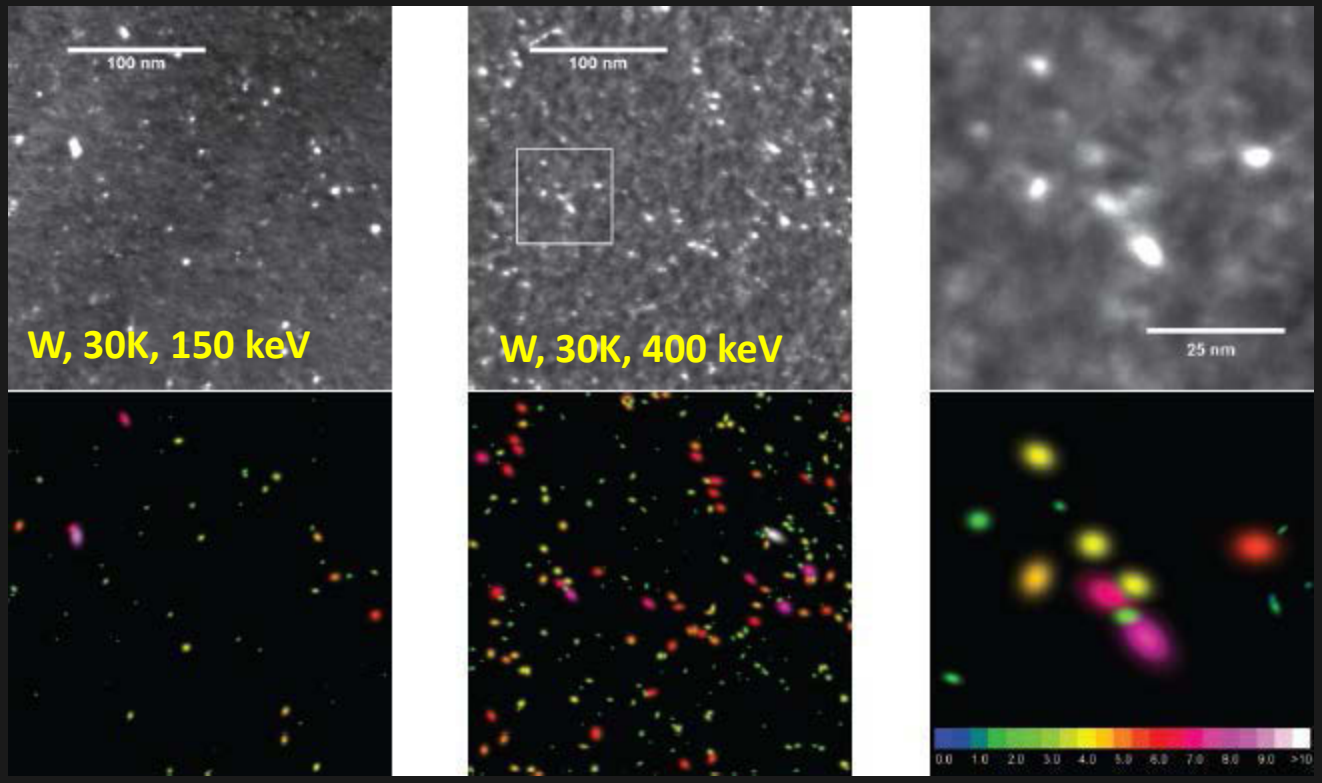
11 video files were included in the publication:

- One-dimensional hopping of small dislocation loops
- Dislocation loops with BV= <111> highly mobile in direction of BV.
- Dislocation loops with BV <100> sessile.
- Loop hopping was much less common in Fe-Cr alloys
- Some formed over 0.2 sec consistent with cascade overlap mechanism.
- Dislocation loop coalescence in string alignment.
- Temperature dependence, only <100> loops at ≥500°C.

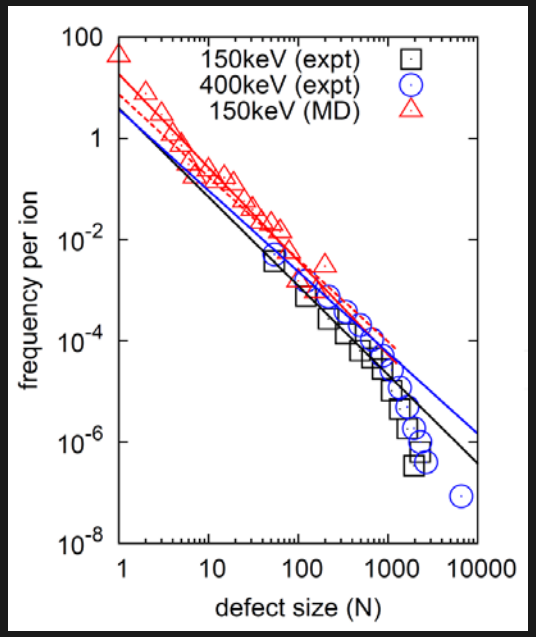
M.Hernandez-Mayoral, Z. Yao, M. Jenkins, M. Kirk, "Heavy-ion irradiations of Fe and Fe-Cr model alloys Part 2: Damage evolution in thin-foils at higher doses," Phil Mag 88(21), 2881 (2008).

Unique Experiment - Cascade Damage Events

- The cryogenic temperature irradiation makes it possible to observe the primary cascade events by suppressing defect diffusion.
- The *in situ* TEM observations can be directly compared with MD simulations of cascade damage events.

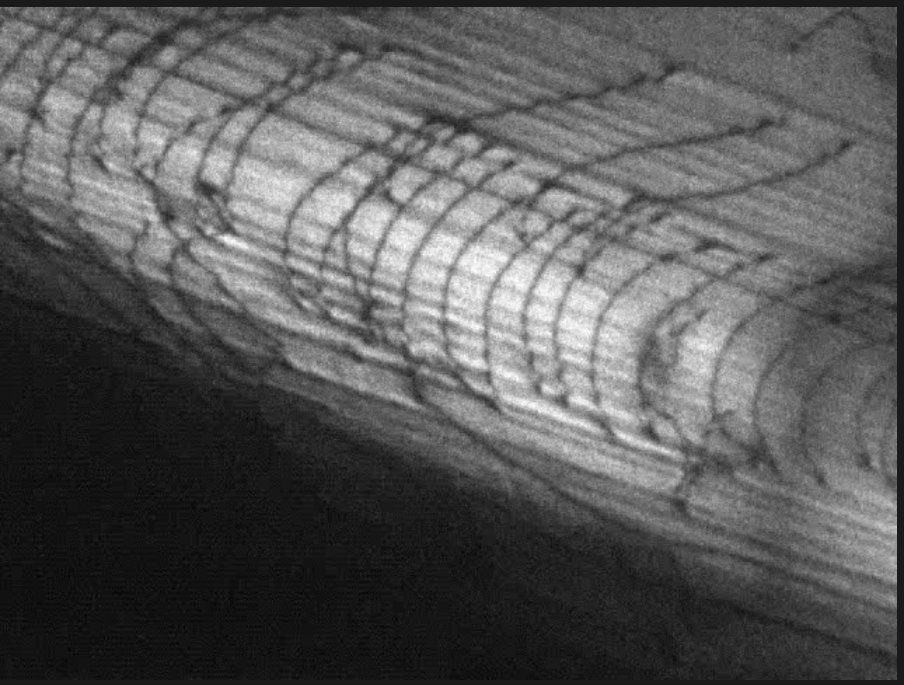


Experiment – MD simulation comparison



(X. Yi, et al, Europhysics Letters 110 (2015) 110.)

Unique Experiment – Defect and Dislocation Interactions under Irradiation+Temperature+Load



304 SS irradiated *in situ* at 400°Cwith 1 MeV Kr ions to dose of $3x10^{13}$ ions/cm^2.

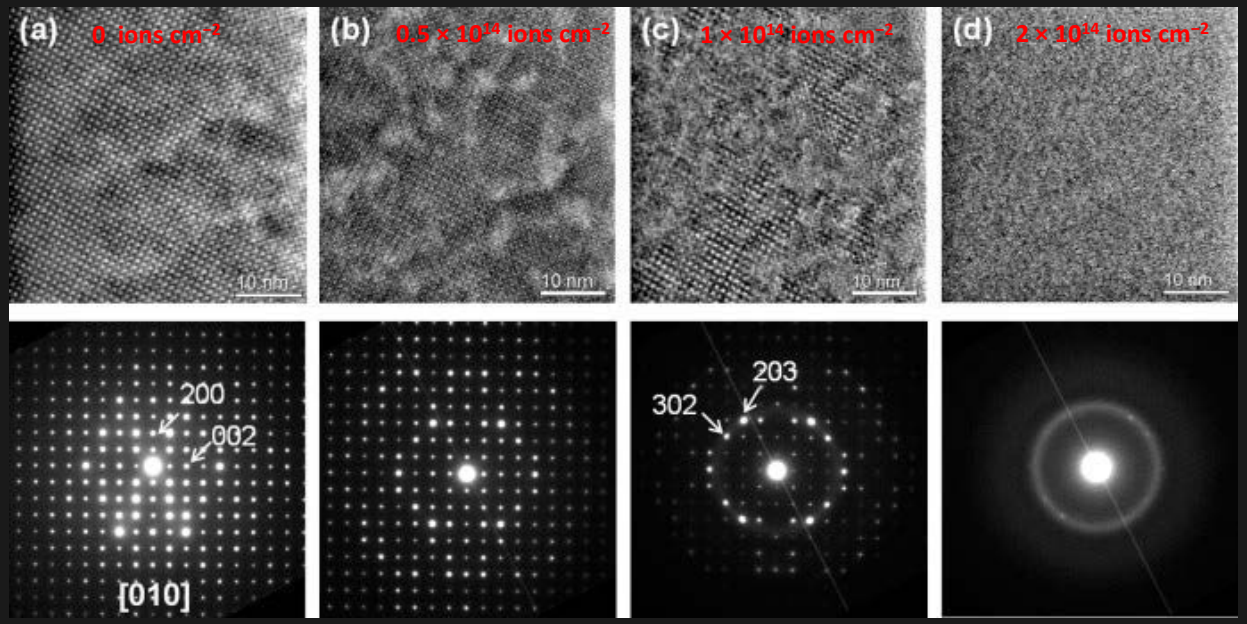
Without ion irradiation **With ion irradiation**

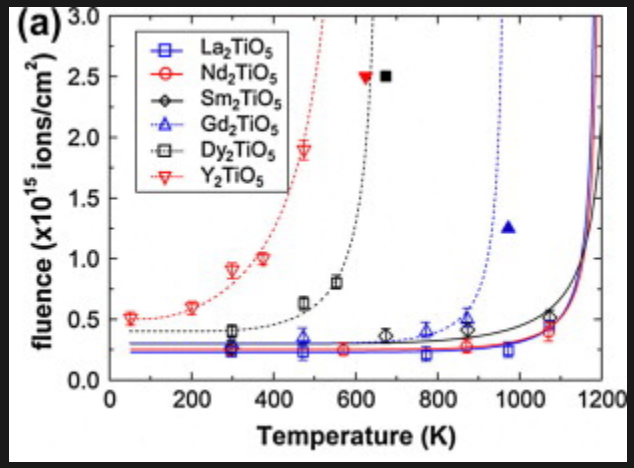
- Direct insight to the mechanisms by which dislocations interact with and annihilate radiation defects to create channels, dispelling some of the common held beliefs about the processes and mechanisms.
- New insight as to how strain is transferred across grain boundaries in irradiated metals, the results of this effort have identified the deterministic step in irradiation stress corrosion cracking.

J. Kacher, I. M. Robertson, Acta Mater. 60 (2012) 6657. M. Briceño et al., J. Nucl. Mater. 409 (2011) 18.

Unique Experiment - Mapping Temperature-Dependent Critical Doses for Amorphization of Wasteforms

- The temperature dependence of the critical amorphization dose was determined *in situ* for a wide temperature range of 50 – 1073 K under well-controlled experimental conditions at the IVEM-Tandem
- This information can only be obtained *in situ* at the temperature because of spontaneous crystallization during warm-up of the specimen.





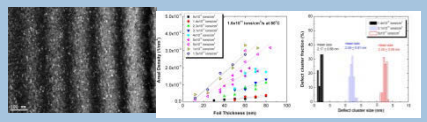
Temperature dependence of critical amorphization doses (ions cm^{-2}) for Ln_2TiO_5.

J. Zhang, F. Zhang, M. Lang, F. Lu, J. Lina, R. Ewing, Acta Mater. 61 (2013) 4191.

Predicting Neutron Irradiation Damage from *in situ* Ion Irradiation through Computer Modeling

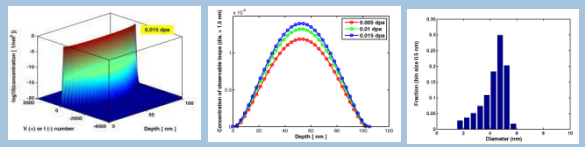
In situ Ion Irradiation Experiments

Well-controlled TEM with *in situ* ion irradiation experiments of thin foils were designed to improve and validate computer models. Experimental data provide a complete set of high-quality, quantitative information, and described the defect behavior at a level of detail unavailable before.



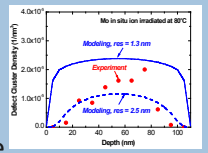
Computer Simulations

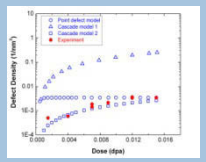
Multiscale modeling to simulate defect evolution from atomic-scale, pico-second events to nanometer-scale, hour evolution of defect structures.



Experiment-Simulation Comparison

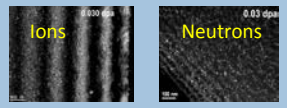
Quantitative, absolute comparisons between experiments and modeling at the same spatial and time scales have led to the establishment of accurate, reliable computer models.

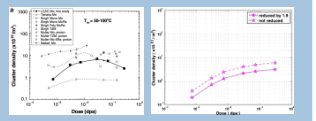




Prediction of Neutron Damage in Reactors

The experimentally-validated model for ion irradiated thin foils is used to predict neutron damage in Mo irradiated in a reactor, and validated by neutron irradiation data





Neutron experiment

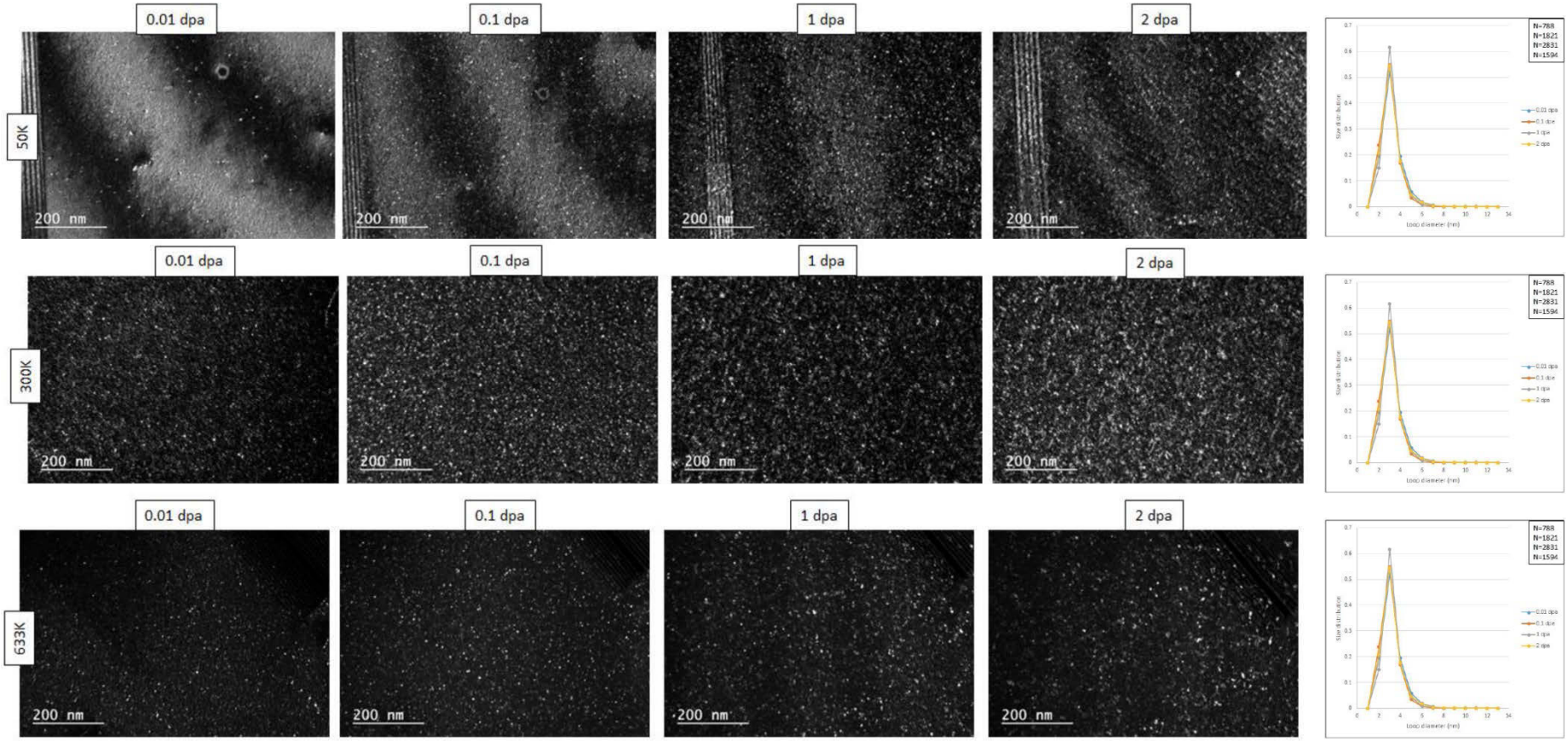
Model

Meimei Li, M.A. Kirk, P.M. Baldo, Donghua Xu, and B. D. Wirth, Phil Mag. 92 (2012) 2048.
D. Xu, B. D. Wirth, M. Li, and M. Kirk, Acta Mater. 60 (2012) 4286.

NEUP IRP:

High Fidelity Ion Beam Simulation of High Dose Neutron Irradiation (U Michigan, PI: Gas Was)

- Conducted *in situ* ion irradiations at IVEM-Tandem to study the temporal evolution of the microstructure in the low dose regime (Penn State U), and to serve as benchmarks for the modeling activity (U Tennessee).



Fe21Cr32Ni alloy

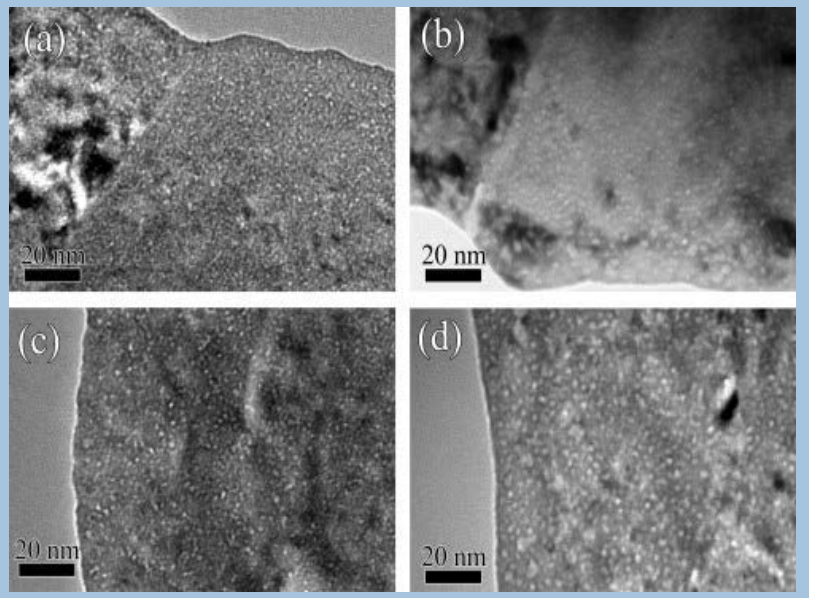
Microstructural Evolution in UO_2 under Irradiation (INL)

- **Understand the nucleation and evolution of inert gas bubbles in UO_2**

In situ Ion Irradiation with TEM

Polycrystal UO_2 150 keV Kr at 25°C

Polycrystal UO_2 1 MeV Kr at 800°C

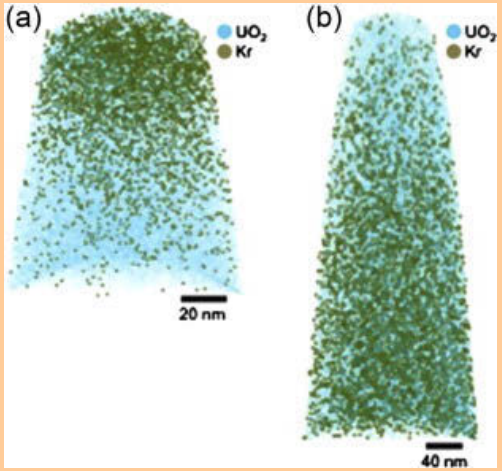


Single crystal UO_2 150 keV Kr at 600°C

Single crystal UO_2 1 MeV Kr at 800°C

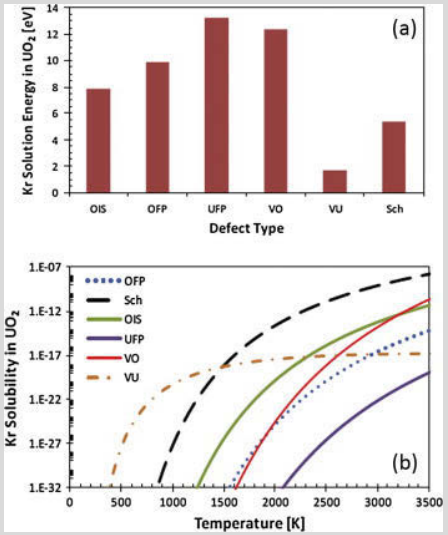
TEM images showing bubbles in UO_2 irradiated with 150 keV Kr and with 1 MeV Kr up to a final dose of 5×10^{15} ions/cm^2.

Atom Probe Tomography



Atom probe tomography reconstruction of Kr-irradiated UO2 representing the distribution of Kr (gold dots) for (a) 150 keV at 600 °C and (b) 1 MeV at 800 °C.

Density Function Theory Calculations



(a) Kr solution energy at 0 K under oxygen-rich condition from DFT calculations in UO_2 and (b) Kr solubility in UO_2 as a function of temperature contacting with 1 atm air or oxygen with 0.21 atm partial pressure.

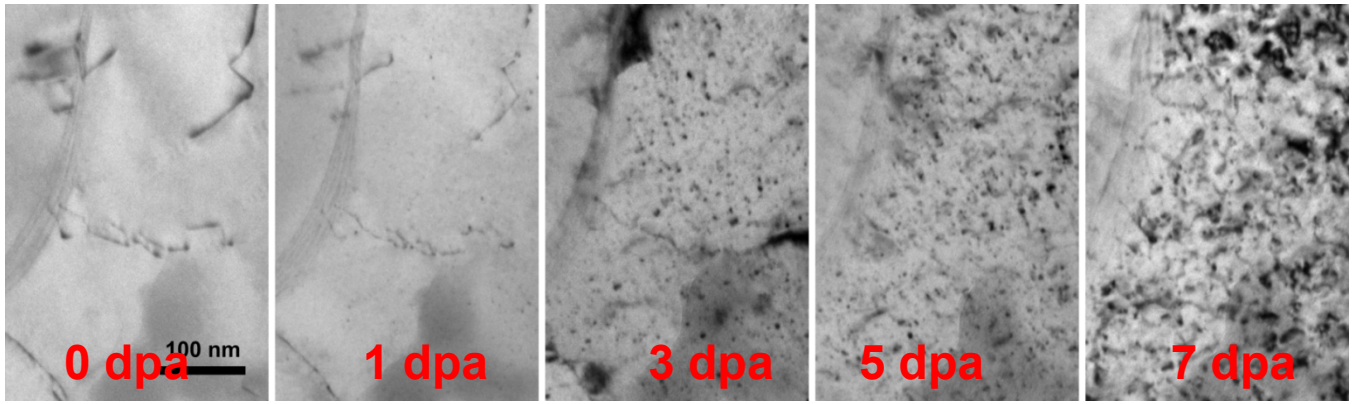
L.F. He, B. Valderrama, A.-R. Hassan, J. Yu, M. Gupta, J. Pakarinen, H.B. Henderson, J. Gan, M.A. Kirk, A.T. Nelson, M.V. Manuel, A. El-Azab, T.R. Allen, J. Nucl. Mater. 456 (2015) 125.

NEUP (Project 14-6762):

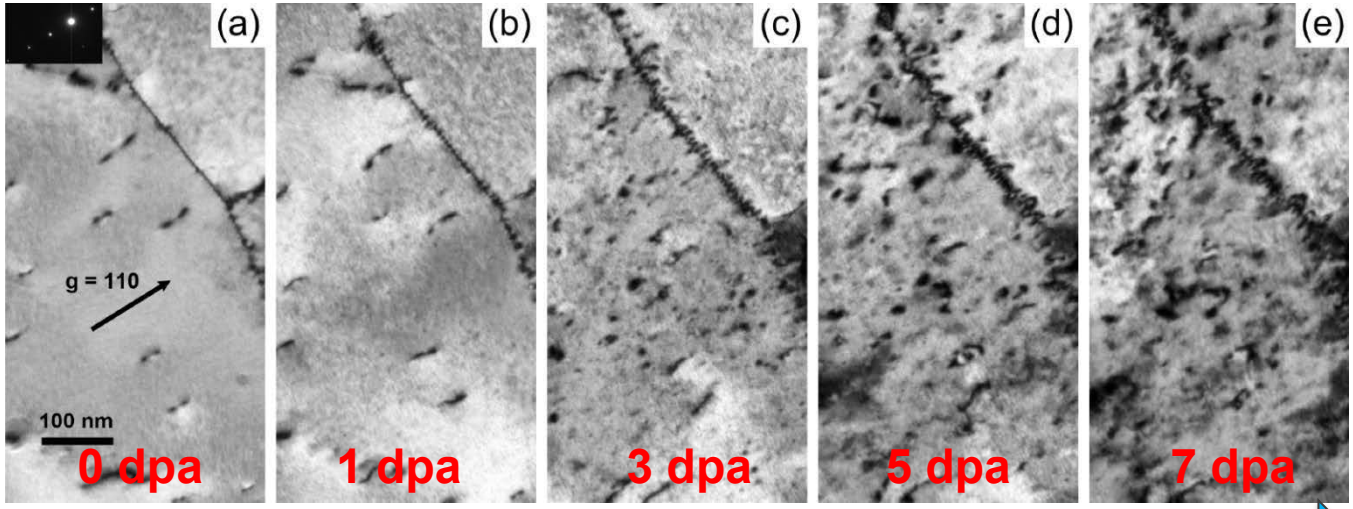
Fundamental Studies of Irradiation-Induced Modifications in Microstructural Evolution and Mechanical Properties of Advanced Alloys (U. Illinois, PI: James Stubbins)

RC-3.1:ADVANCED HIGH TEMPERATURE ALLOY SCOPING IRRADIATIONS

Advanced G92 ferritic-martensitic steel with optimized chemistry



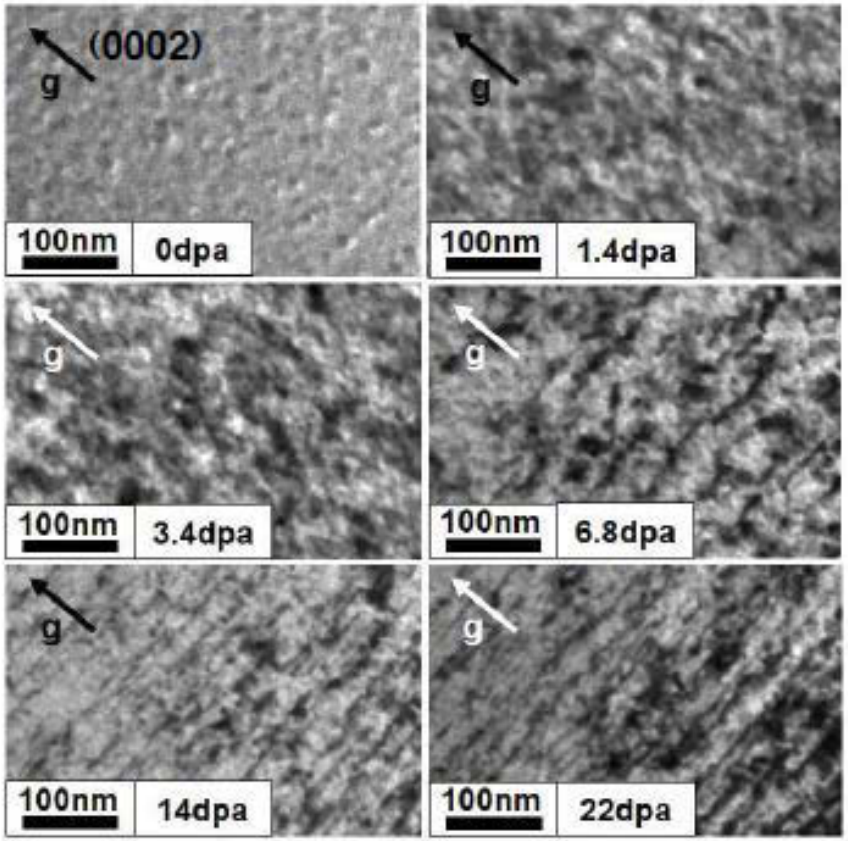
Traditional G91 ferritic-martensitic steel



1 MeV Kr irradiation at 300-400°C with increased irradiation dose

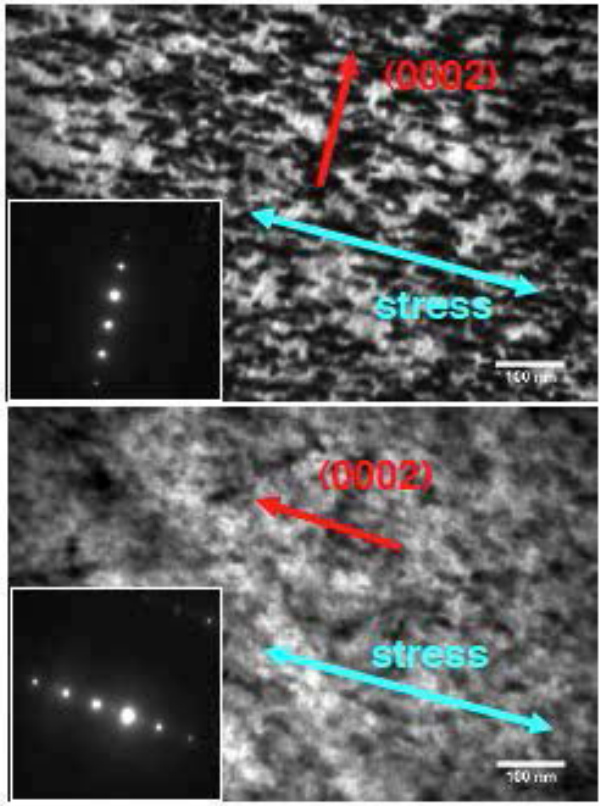
Areva Project:
Effect of stress on Loop formation in Pressurized Water Reactor Guide Tube

Dose-dependent <c>-loop formation in M5



(J. Nucl. Mat. 423, 170-182 (2012))

Influence of tensile stress on <c>-loop formation in adjacent grains, Zry-4

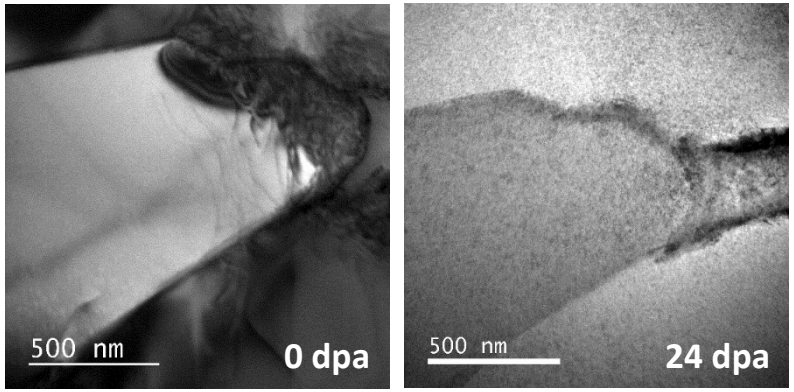


(to be published)

R.M. Hengstler-Eger and W. Petry (Technische Universität München, Germany)
P. Baldo and M.A. Kirk (Argonne National Laboratory, EMC)
P.B. Hoffmann (AREVA NP GmbH, Germany)

NSUF RTE -
In Situ Study of Defect Accumulation in Ti-6Al-4V under Heavy Ion Irradiation (Michigan State U, PI: Carl Boehlert)

- The first NSUF RTE proposal awarded in FY2016. IVEM experiments conducted during Feb. 22 – 24, 2016.
- Investigate the effects of thermo-mechanical processing on radiation resistance of Ti-6Al-4V alloy
  - Powder metallurgy rolled
  - Direct metal laser sintering (DMLS)
  - DMLS + hot isostatic pressing
  - DMLS + heat treatment (1020°C/2h)
- 1 MeV Kr ions were used to irradiate Ti-6Al-4V TEM samples

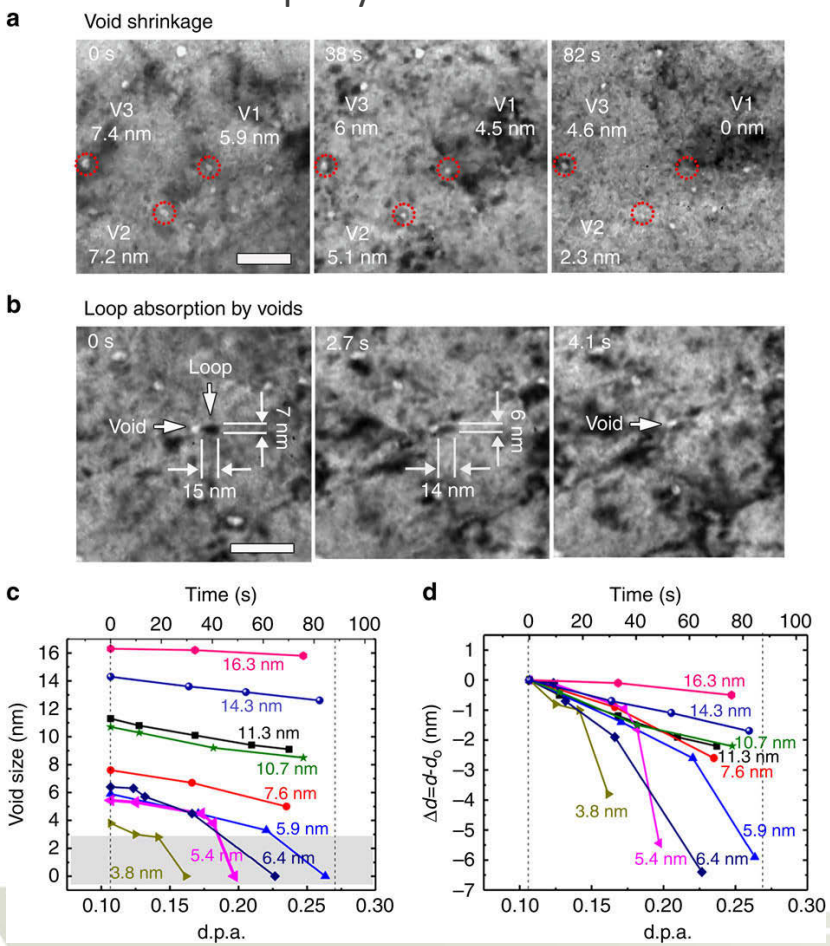


The material exhibited a lath-type morphology and greatest resistance to irradiation damage up to 24 dpa.

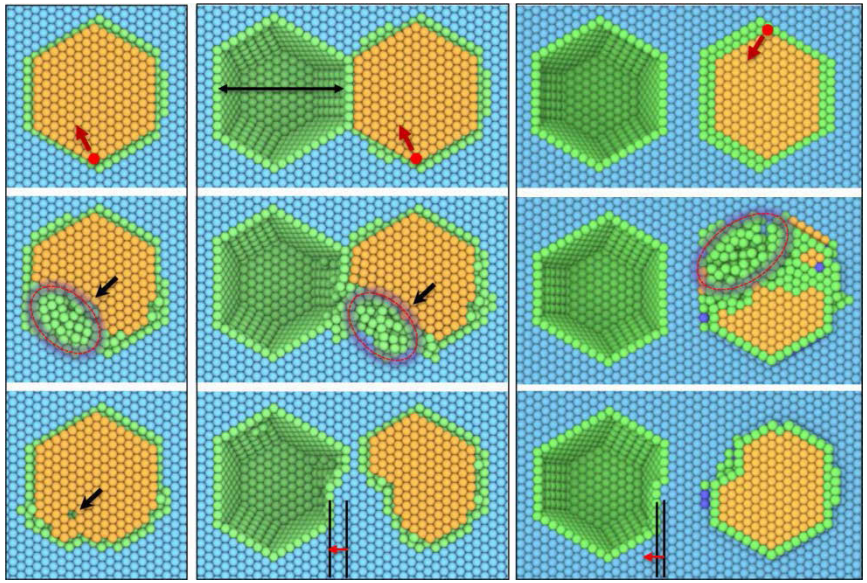
1 MeV Kr irradiation of 3D HIPed Ti-6Al-4V alloy

Exploring New Alloy Design Concept: Radiation-tolerant nanotwinned metals with nanovoids

In situ Kr ion irradiation studies of nv-nt Cu unravelling continuous shrinkage of nanovoids and absorption of mobile dislocation loops by nanovoids.



MD simulation reveals dynamic process through which a void absorbs a neighboring dislocation loop.



Y. Chen, et al., Nature Communications 6 (2015), 7036.

Facility Organization

- **Staff**
  - Director: Meimei Li (5-10%)
  - Associate Director: Mark Kirk (retiree, 50%)
  - Accelerator Scientist: Pete Baldo (90%)
  - TEM specialist: Ed Ryan (retiree, 50%)
  - Administrative assistant: Denise Moores
- **Scientific Advisory Committee (11 members)**
  - Arthur Motta (Chair), Ian Robertson, James Stubbins, Brian Wirth, Rod Ewing, Mitra Taheri, Xinhang Zhang, Hussein Khalil, Todd Allen, Stephen Donnelly, Gary Was
- **Proposal Review Committee (6 members)**
- **Other Supports**
  - Nuclear Engineering (NE) Division ESH, administrative support
  - Health Physics and Radioactive Material Shipping, Handling, and Inventory
  - NE division Irradiated Materials Laboratory
  - Argonne User Facility Agreements through the Advanced Photon Source (APS) User Office

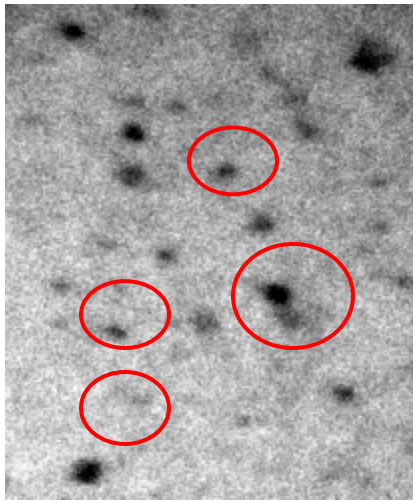
User Proposal Process

- As a user facility, IVEM-Tandem provides free facility access for non-proprietary work through a peer review proposal process
- ANL researchers are treated as general users through the same peer review proposal process
- **User proposals submitted directly to IVEM-Tandem**
  - Before submitting a proposal, users discuss with staff about their proposed experiments
  - User can submit a proposals for review at any time, and we maintain a continuous review process
  - IVEM staff review for program relevance, feasibility, and safety; Proposal review committee review for scientific merit.
  - A proposal remains active for a maximum period of two years following the approval date;
  - An active proposal is required to submit a progress summary for evaluation when requesting for facility time
  - Three calls for requests for facility time in FY 2016:
    - Cycle 1: Nov. 1, 2015 to Feb. 29, 2016;
    - Cycle 2: Mar. 1, 2016 to June 30, 2016;
    - Cycle 3: July 1, 2016 to Sept. 30, 2016;
- **NSUF Rapid Turnaround Experiment (RTE) Proposals for rapid access**
  - Started in FY2016
  - 1 NSUF RTE proposal was awarded in FY16 Cycle 1
  - 3 NSUF RTE proposals are under review

Recent Upgrade & Proposed Capability Enhancement

Recent upgrade

- Installed a high-speed, high-resolution OneView camera in December 2015
  - 25 frames/sec at 4kx4k resolution
  - 200 frames/sec at 1kx1k resolution

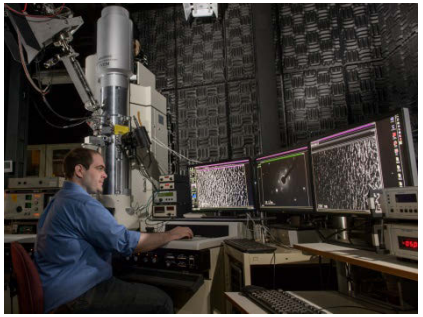


Proposed Enhancement

- Dual-beam capability to study combined effect of helium and displacement damage
  - Acquired a low energy ion gun

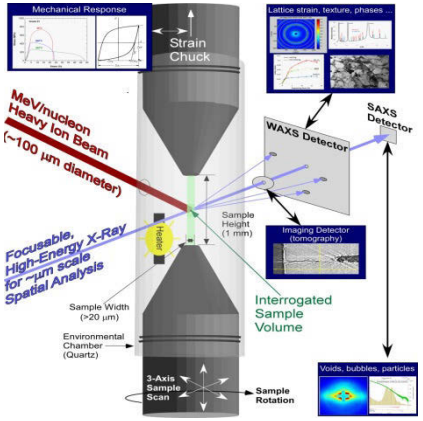
In situ Multiscale Characterization Facility

In situ ion irradiation with electron beams (IVEM-Tandem)



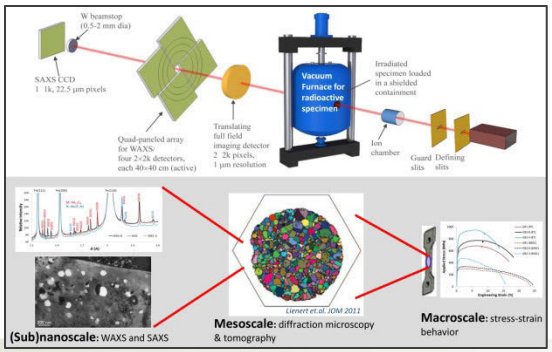
100-200 nm

In situ ion irradiation with X-rays (XMAT Proposal)



Microns

In situ characterization of neutron damage with high-energy X-rays (*i*RadMat)



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