Characterization of Nuclear Reactor Materials using Synchrotron Radiation

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Exploring the Pivotal Role of Next Generation X-Rays in Bridging the Scale-Gaps in Next Generation Energy Materials under Extremes
Brookhaven National Laboratory
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Acknowledgements

ANL Team

- Jonathan Almer
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- Yiren Chen
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Collaborators

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- Stu Maloy, LANL
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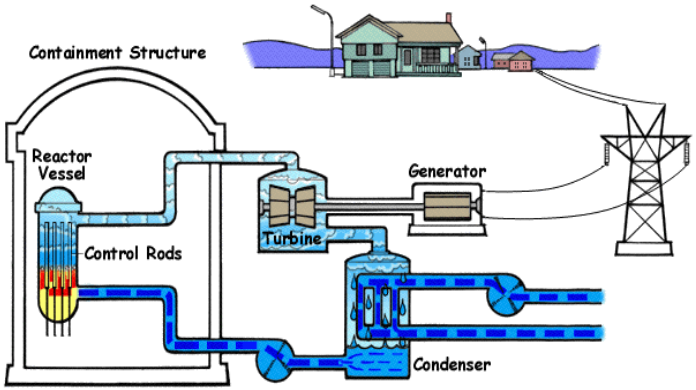
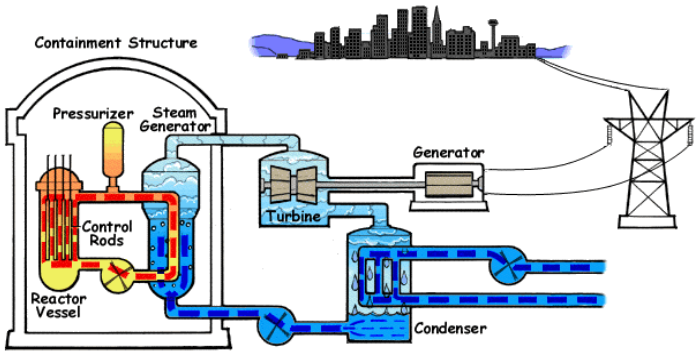
- Lizhen Tan, ORNL
- Jeff Terry, Illinois Institute of Technology
- Yong Yang, University of Florida

Overview of Nuclear Energy Systems

Current Nuclear Power Plants

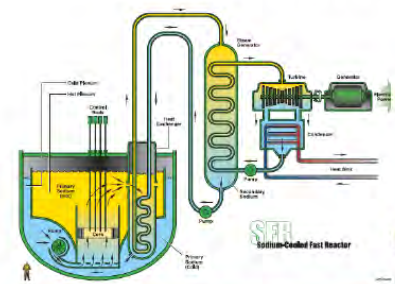


Boiling Water Reactor (BWR)

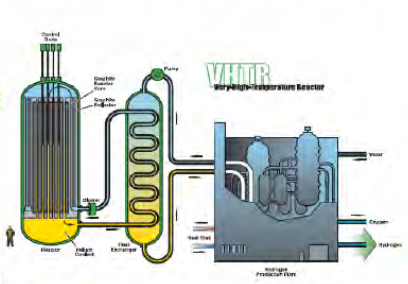


Pressurized Water Reactor (PWR)

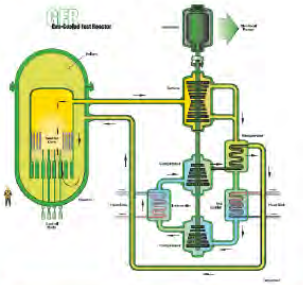
Advanced Reactor Concepts



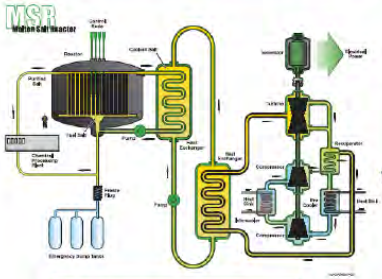
Sodium-cooled fast reactor



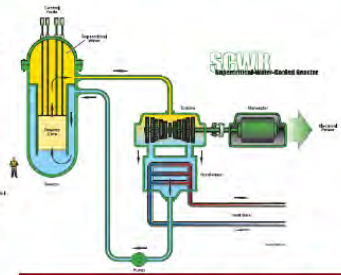
Very high temperature reactor



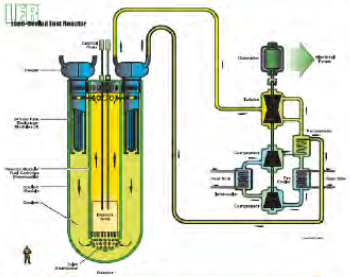
Gas-cooled fast reactor



Molten salt reactor

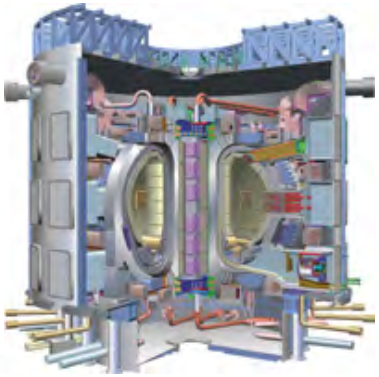


Supercritical water reactor



Lead-cooled fast reactor

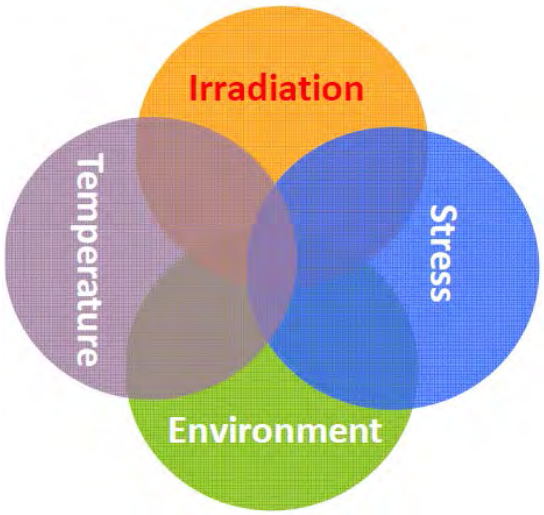
Fusion Reactors



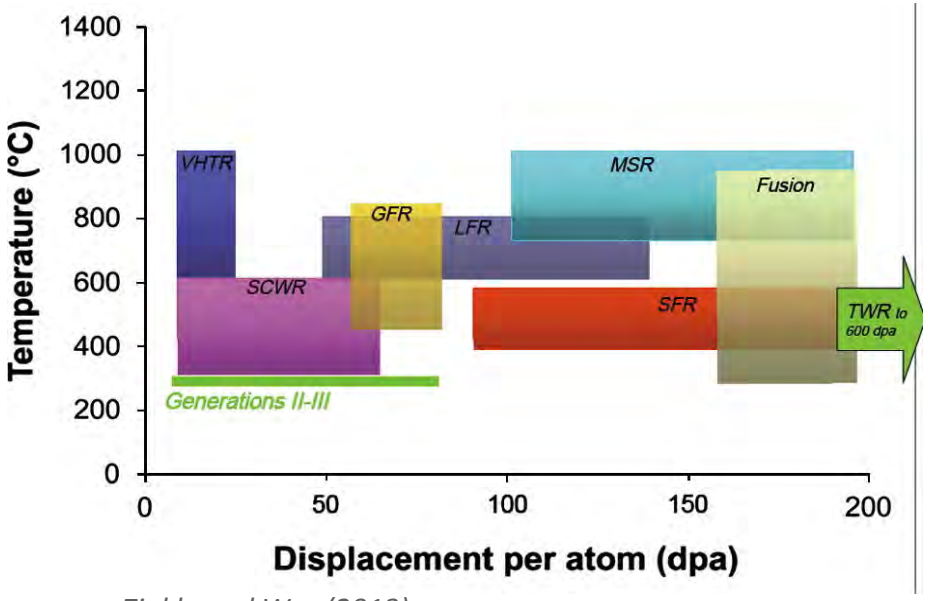
Material Challenges

- Radiation tolerance
- Heat resistance
- Corrosion resistance
- Accident tolerance
- Predicting long-term material behavior

Complex Nuclear Environments

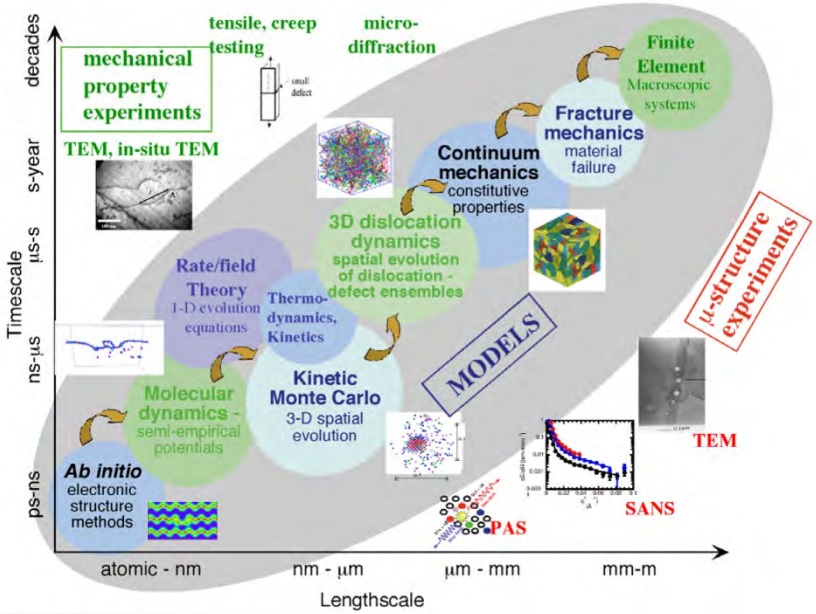


Increased Demands on Materials



Zinkle and Was (2013)

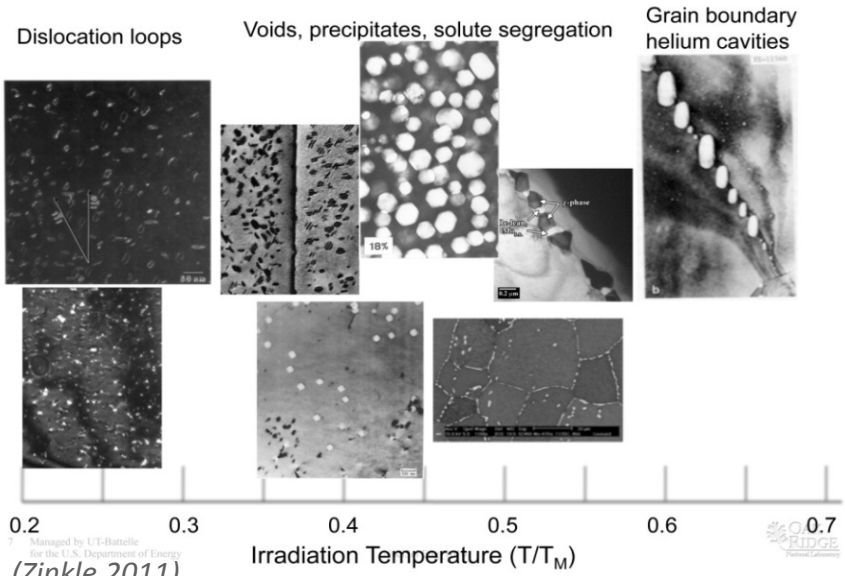
Multi-scale physical problems



B. Wirth et al (2004)

Radiation Damage and Radiation Effects

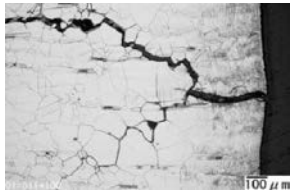
Radiation-induced defects



(Zinkle 2011)

Radiation-induced mechanical degradation

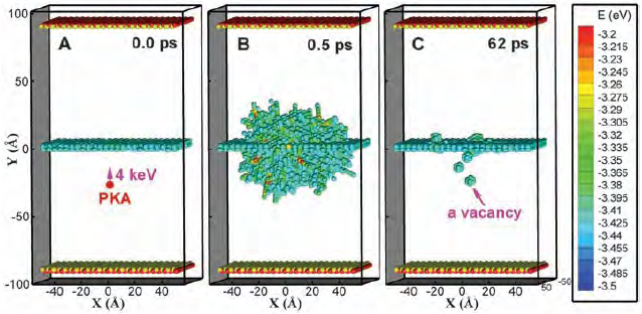
- Radiation hardening and embrittlement
- Void swelling and dimensional instability
- Irradiation creep
- Irradiation-assisted stress corrosion cracking
- High temperature helium embrittlement



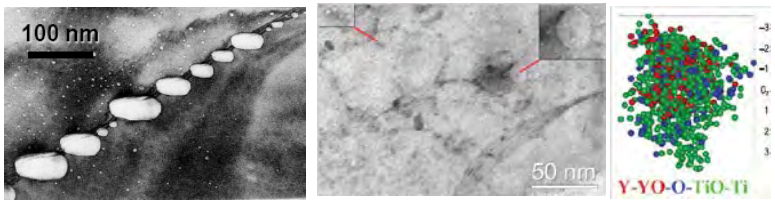
Stress Corrosion Cracking
(T. Shoji, 11th Env. Deg. Conf.)

Roles of Interfaces

Defects can be absorbed at interfaces (grain boundaries, particle interfaces, etc.), enhancing radiation tolerance.

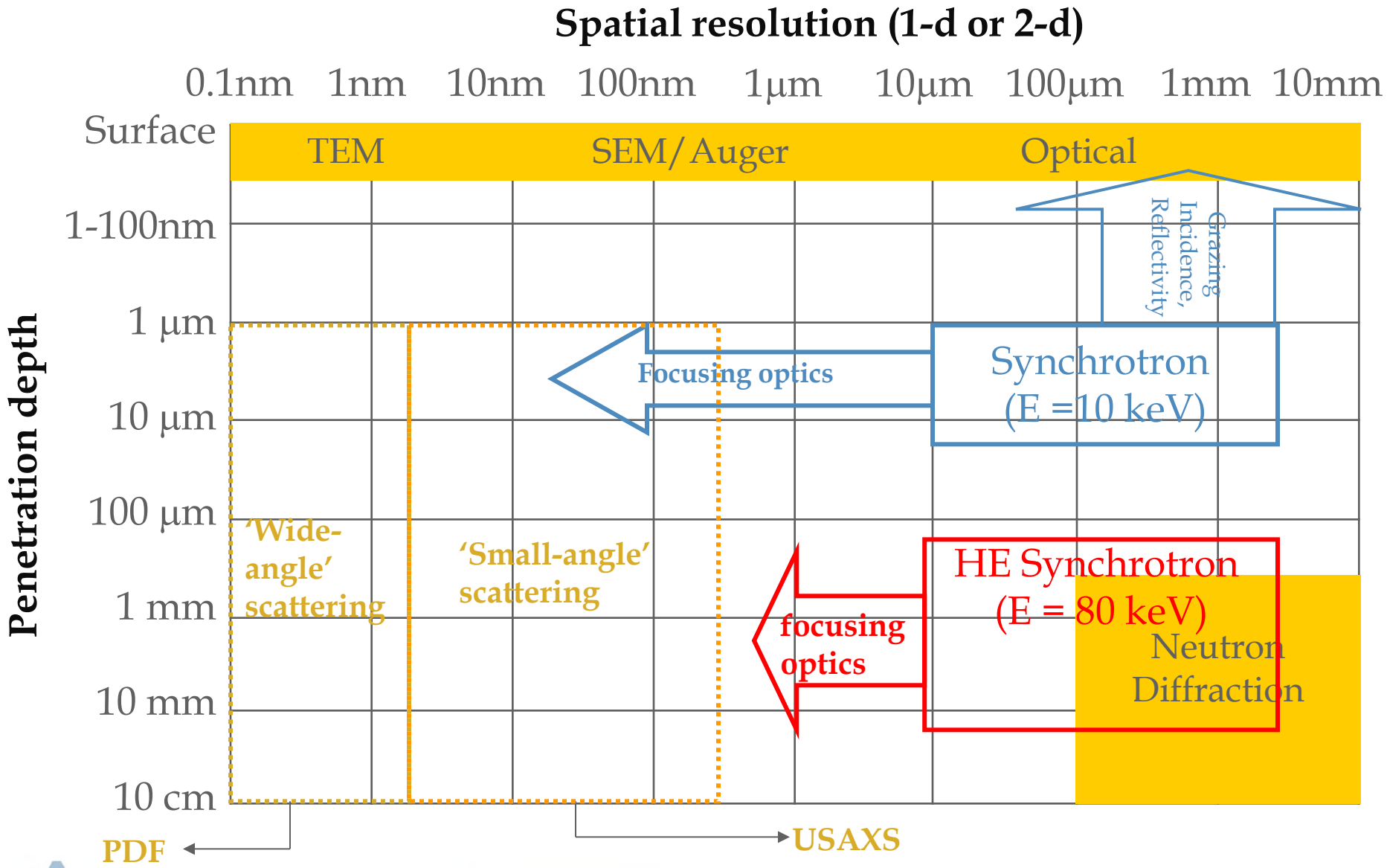


MD simulations showed defect absorption at grain boundaries. (Bai et al, 2010)



Dispersion of nanosized particles reduce He bubble formation in steel. (Odette and Hoelzer, 2008, 2010)

Comparison of Selected Characterization Techniques



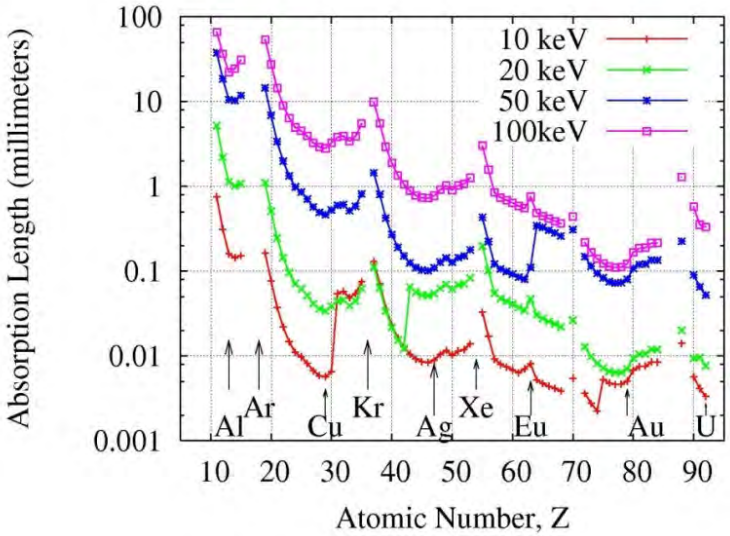
By J. Almer

Applications of Hard X-rays in Nuclear Materials

- Real Material in Real Environment in Real Time

- **Deep penetration**
  - Bulk behavior
  - Environmental chambers
- **In situ, real-time studies**
  - Dynamics
  - Structural evolution
- **Unit mechanisms and collective effects in complex engineering materials**
  - Spatially resolved (inhomogeneity)
  - Phase-specific
  - Chemistry-specific
  - Multiple probes for concurrent, multi-scale characterization
- **Minimal specimen preparation of irradiated specimens**

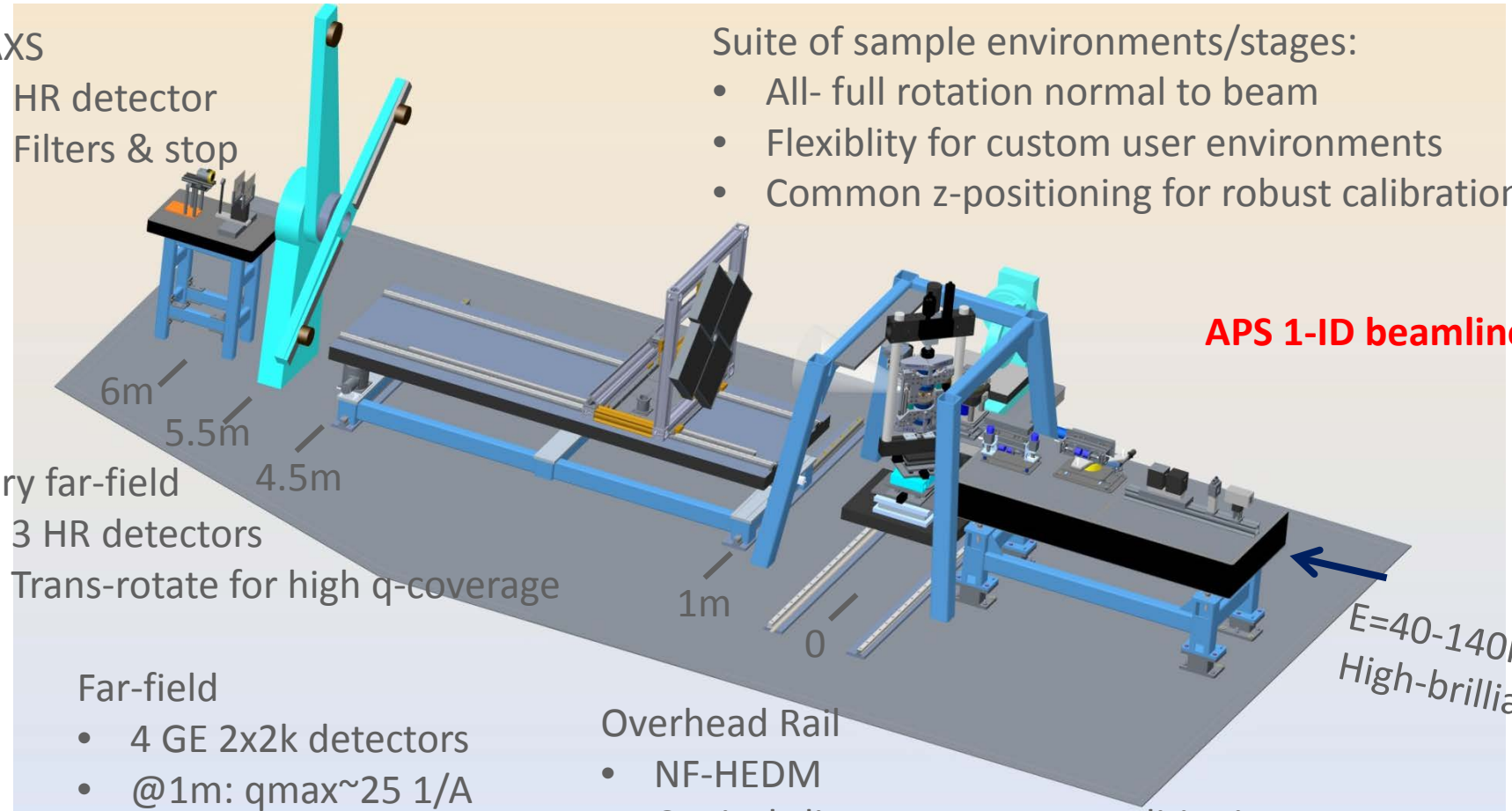
APS – Hard X-ray Synchrotron Source



Combined in-situ high-energy techniques

SAXS
- HR detector
- Filters & stop

Suite of sample environments/stages:
- All- full rotation normal to beam
- Flexiblity for custom user environments
- Common z-positioning for robust calibration



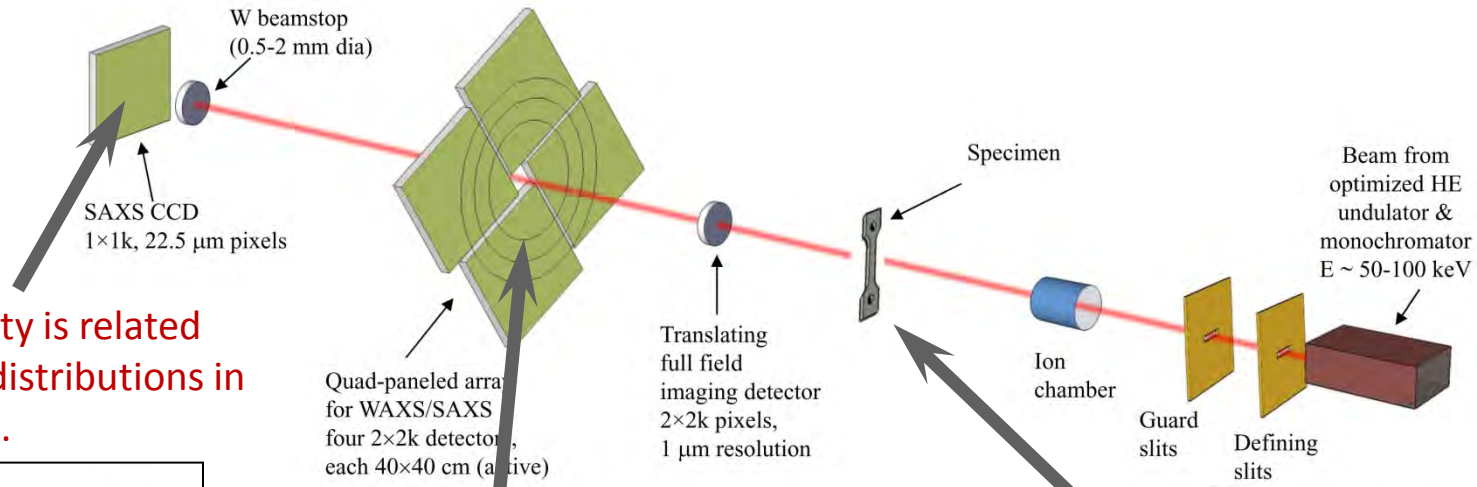
Very far-field
- 3 HR detectors
- Trans-rotate for high q-coverage

Far-field
- 4 GE 2x2k detectors
- @1m: qmax~25 1/A
- Center-hole (SAXS)

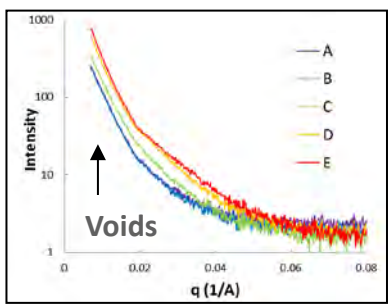
Overhead Rail
- NF-HEDM
- Conical slit
- Tomography
- Lasers

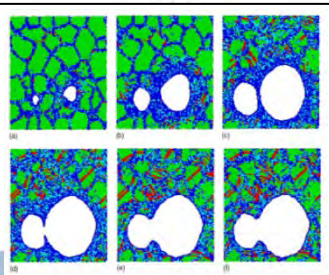
Beam-conditioning system
- Focusing optics for <um beams
- White and mono capable
- BPMs, slits, DIC, etc

Multi-probe *in situ* X-ray measurements under thermal-mechanical loading

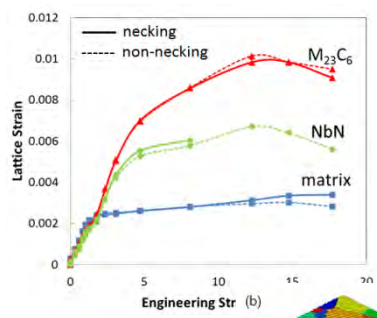


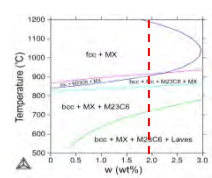
SAXS intensity is related to the void distributions in the material.

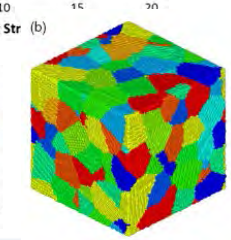


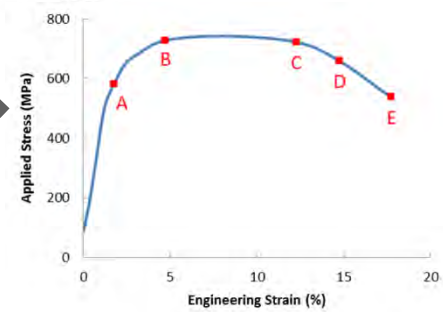


Diffraction data are used to understand deformation and strengthening mechanisms associated with microstructural features (phases, grain orientation, etc.)



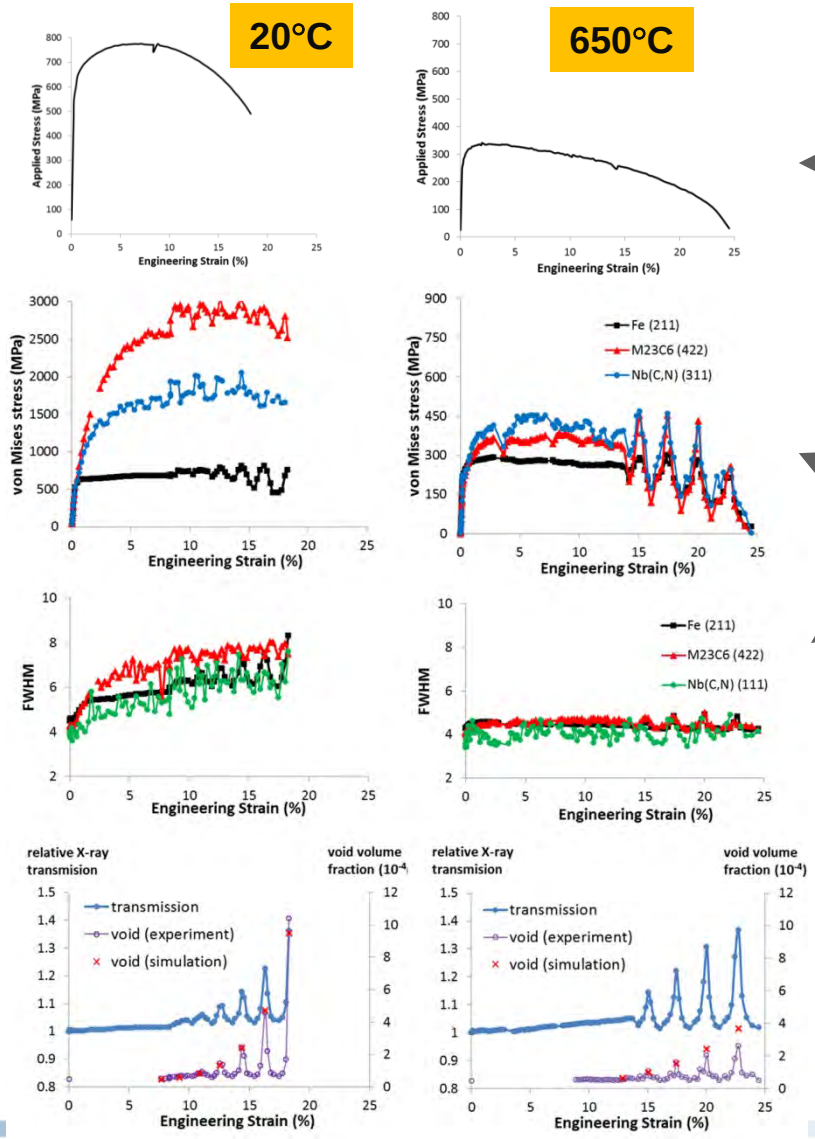






Mechanical test provides information on macroscopic mechanical behavior

Concurrent WAXS/SAXS/Radiography to Study Deformation and Failure Mechanisms in Steels

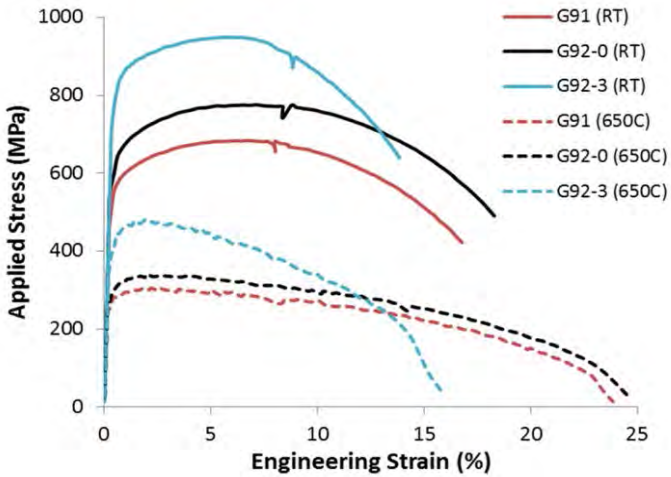


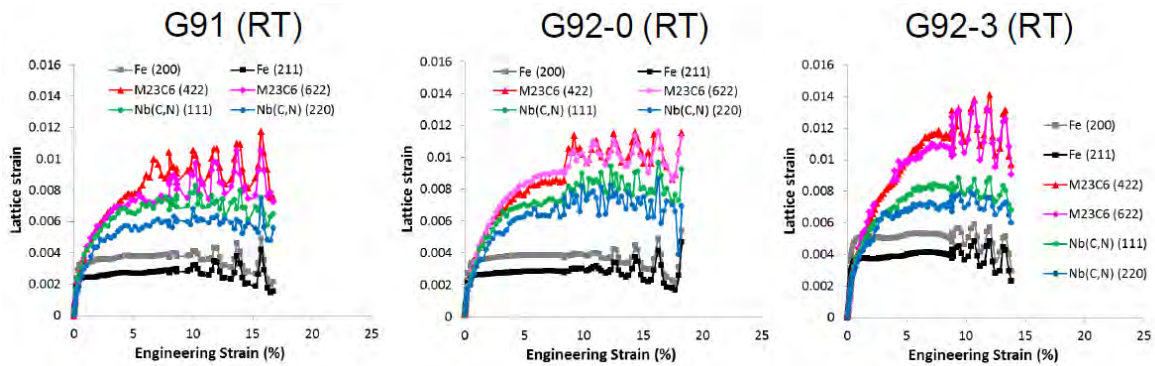
Lab-scale mechanical test provides information on macroscopic stress-strain behavior.

Diffraction data are used to understand deformation and strengthening mechanisms associated with microstructural features (phases, grain orientation, etc.).

SAXS intensity is related to void distributions in the material. Radiography track the specimen dimensional changes. These data provide important information to under failure mechanisms.

Lattice Strain – Quantifying Strengthening Effects





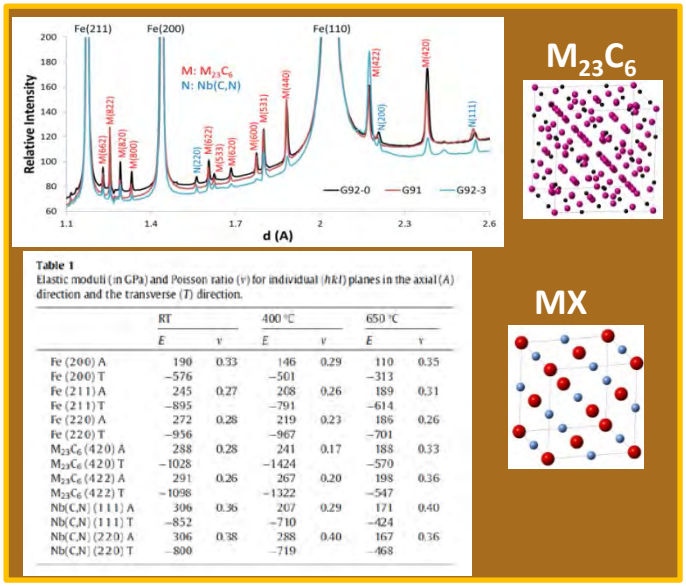


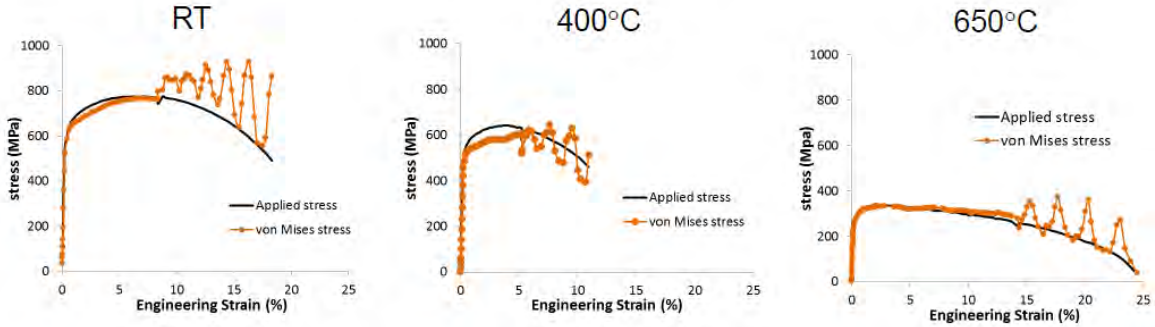
Table 1
Elastic moduli (in GPa) and Poisson ratio (ν) for individual (hkl) planes in the axial (A) direction and the transverse (T) direction.

	RT		400 °C		650 °C	
	E	ν	E	ν	E	ν
Fe (200) A	190	0.33	146	0.29	110	0.35
Fe (200) T	−576		−501		−313	
Fe (211) A	245	0.27	208	0.26	189	0.31
Fe (211) T	−895		−791		−614	
Fe (220) A	272	0.28	219	0.23	186	0.26
Fe (220) T	−956		−967		−701	
$M_{23}C_6$ (420) A	288	0.28	241	0.17	188	0.33
$M_{23}C_6$ (420) T	−1028		−1424		−570	
$M_{23}C_6$ (422) A	291	0.26	267	0.20	198	0.36
$M_{23}C_6$ (422) T	−1098		−1322		−547	
Nb(C,N) (111) A	306	0.36	207	0.29	171	0.40
Nb(C,N) (111) T	−852		−710		−424	
Nb(C,N) (220) A	306	0.38	288	0.40	167	0.36
Nb(C,N) (220) T	−800		−719		−468	

$M_{23}C_6$

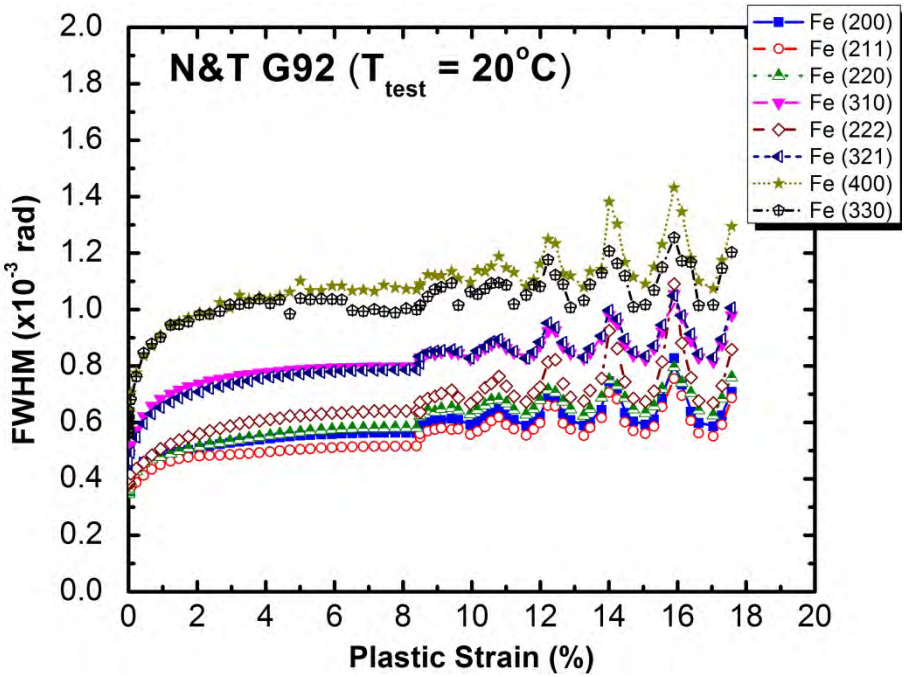
MX

$$\sigma_{VM}(material) = \sigma_{VM}^{Nb(C,N)} \times 0.004 + \sigma_{VM}^{M_{23}C_6} \times 0.02 + \sigma_{VM}^{Fe} \times 0.976$$



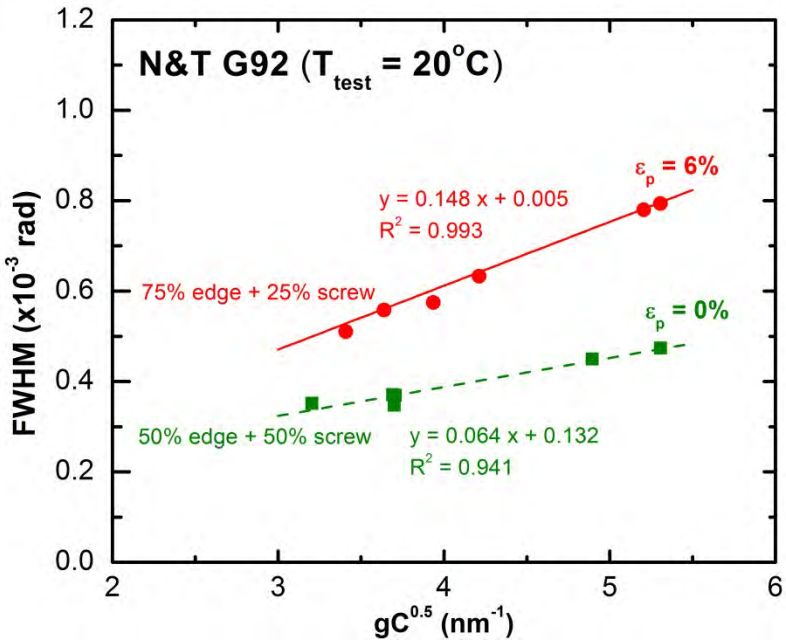
Peak Broadening – Dislocation Evolution

Dislocation density and subgrain size obtained from analysis of multiple x-ray peaks



FWHM of bcc Fe peaks was evaluated by the modified Williamson-Hall (W-H) plot

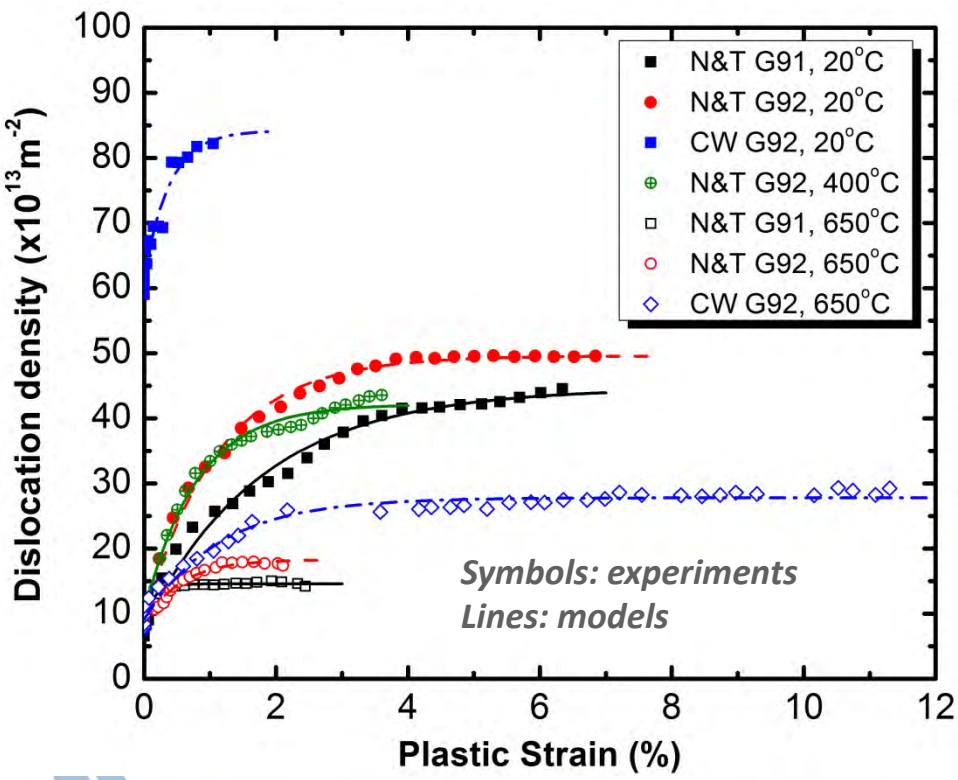
$$\Delta K \cong 0.9/D + \sqrt{\pi M^2 b^2/2}\sqrt{\rho}(KC^{0.5})$$



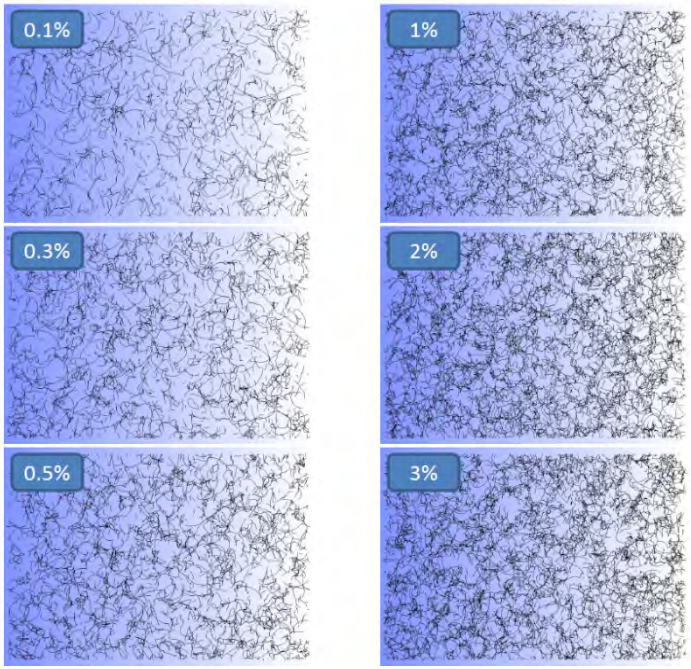
Modeling Dislocation Evolution during Deformation

Evolution of the dislocation density during deformation is described by dislocation creation/storage and dislocation annihilation

$$d\rho/d\varepsilon = \bar{m}/(b \cdot s) - \Omega \cdot \rho$$

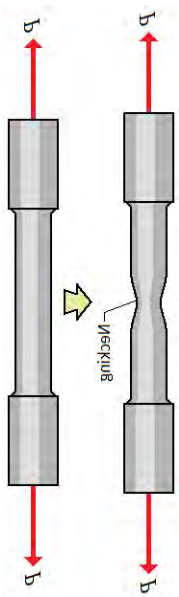


Dislocation dynamic modeling of dislocation density change as a function of strain that may be validated by in situ X-ray measurements.

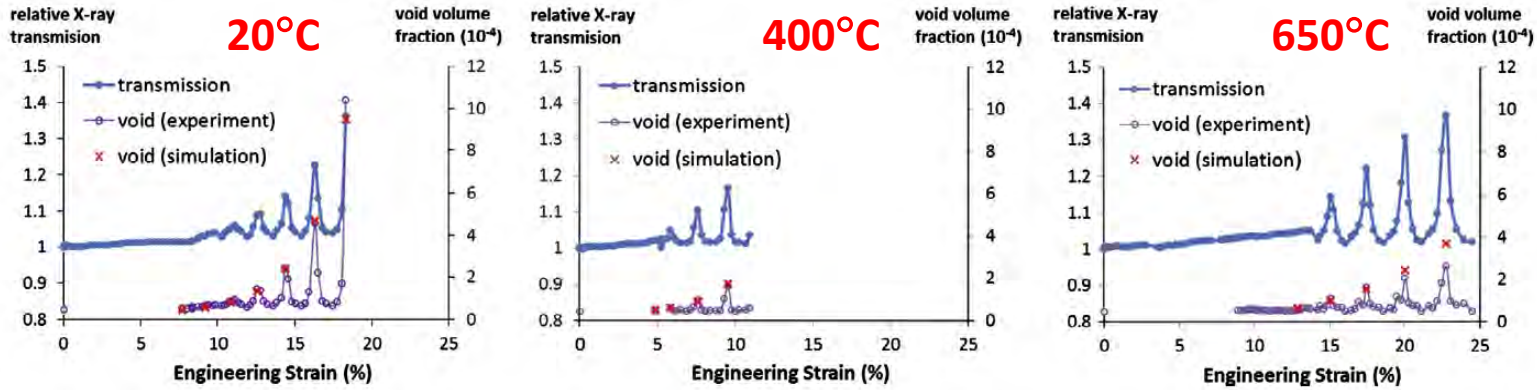


(Liu et al., 2012)

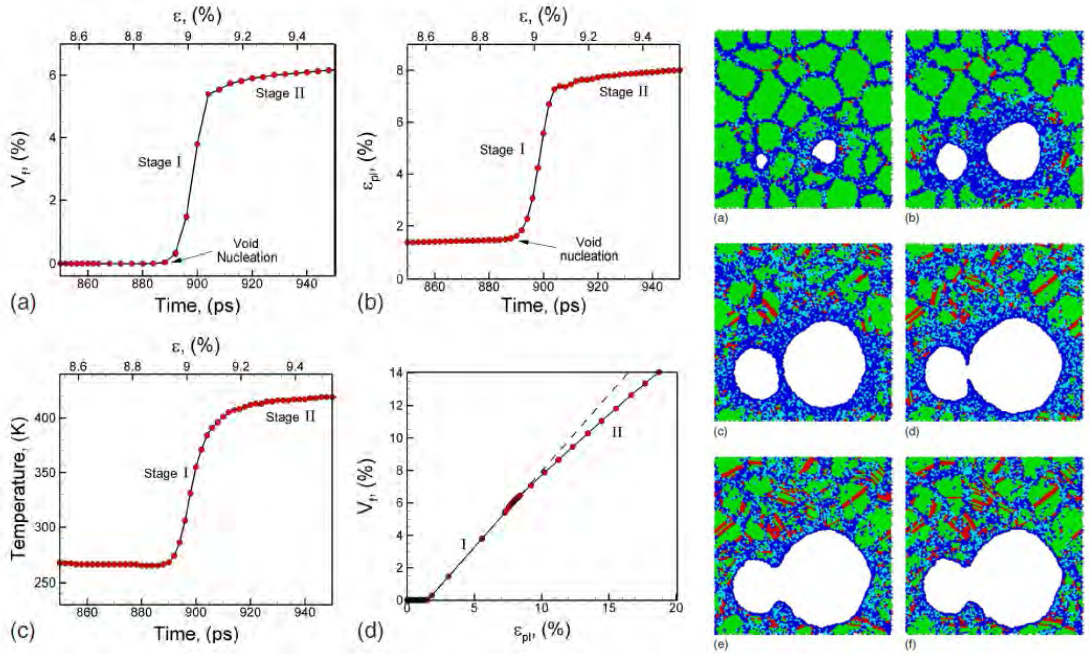
SAXS – Void Formation and Evolution



Void formation and evolution during necking captured by SAXS

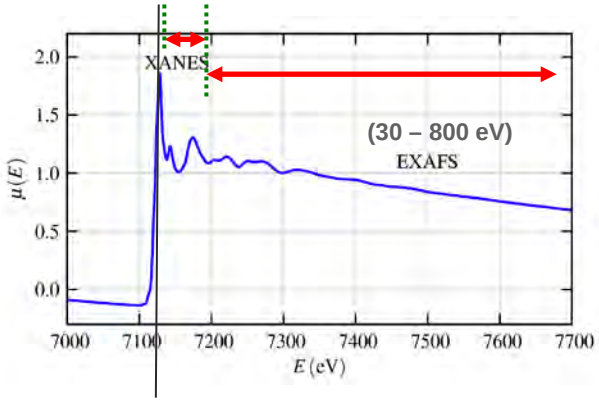


Molecular Dynamics (MD) simulations showed void evolution during tensile deformation
(Dongare et al (2009))

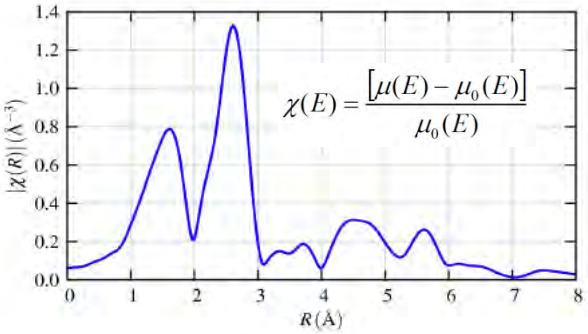


Synchrotron Extended X-ray Absorption Fine Structure (EXAFS)

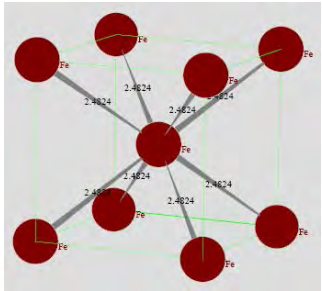
- Extended X-ray absorption fine structure (EXAFS) is the oscillations of the x-ray absorption spectrum ranging between 30 – 800 eV above an absorption edge of a specific element.
- A photoelectron is ejected from the absorbing atom by the photoelectric effect when the energy of incident X-rays is above the ionization threshold. The oscillations result from the interference of the outgoing photoelectrons with the scattered waves from surrounding atoms.
- EXAFS is a element-specific atomic probe. It characterizes the local atomic structure around a given atom including
  - Type of neighboring atoms
  - Position of neighboring atoms
  - coordination number of neighboring atoms
- Structural parameters are obtained by fitting the model function to the measured EXAFS profiles



Fe K-edge



Fourier transformed EXAFS Profile



EXAFS Measurements of Irradiated Mod.9Cr-1Mo Steel

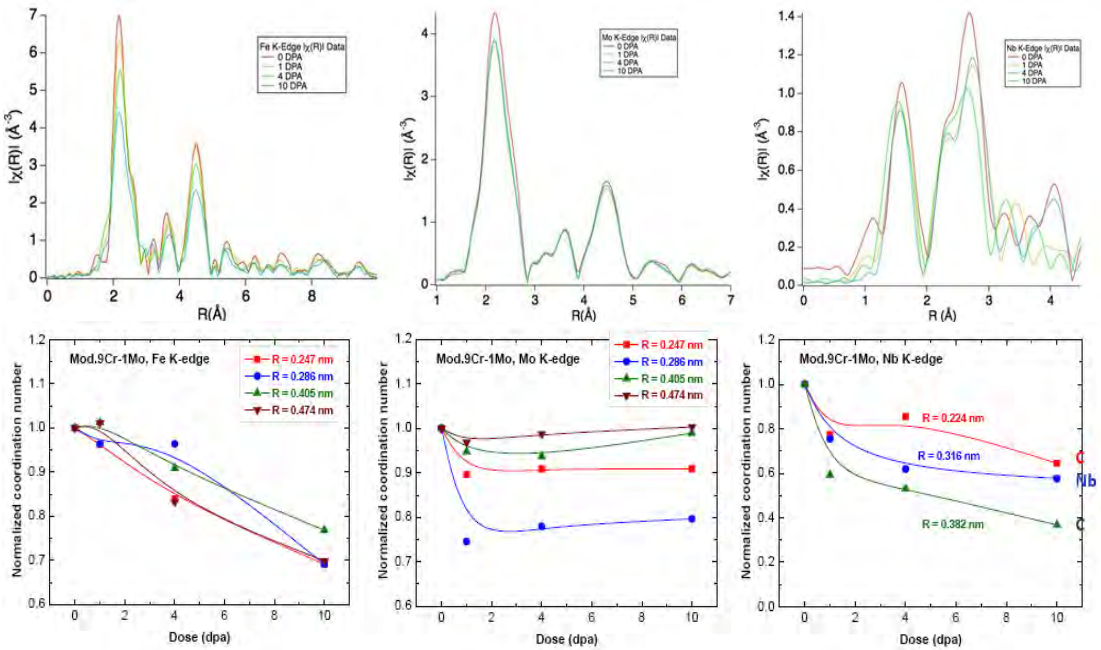
EXAFS measurements detected vacancy environments around alloying elements in an irradiated steel, providing new insight into the defect interactions with individual alloying element at the atomic level.

Experiment

- EXAFS measurements were performed at APS MR-CAT
- Specimens of mod.9Cr-1Mo steel were irradiated to 1, 2, 4 10 dpa at 40-70°C.
- Examined TEM or coupon specimens: 4x2x0.25 mm
- Irradiated specimens were sealed in a triple-contained container
- Absorption spectra were recorded in fluorescence geometry

Results

- Detected irradiation-induced local changes of atomic environments (within 6 Angstroms) associated with each alloying element.
- Vacancy environment around Fe, Mo, Nb, and Cr atoms evolved differently under irradiation, indicating strong interactions between defects and alloying elements.



The reduction in coordination number around Fe approached 30% at 10 dpa, while the coordination number was reduced by 25% around Mo and 40% around Nb irradiated to as low as 1 dpa. The coordination number loss around Mo saturated at 1 dpa, while the loss of near neighbor around Fe atoms continued with increasing dose, and the measured damage can be correlated with the dpa values.

Meimei Li, D. Olive, Y. Trenikhina, H. Ganegod, J. Terry and S. A. Maloy, J. Nucl. Mater. 441 (2013) 674.

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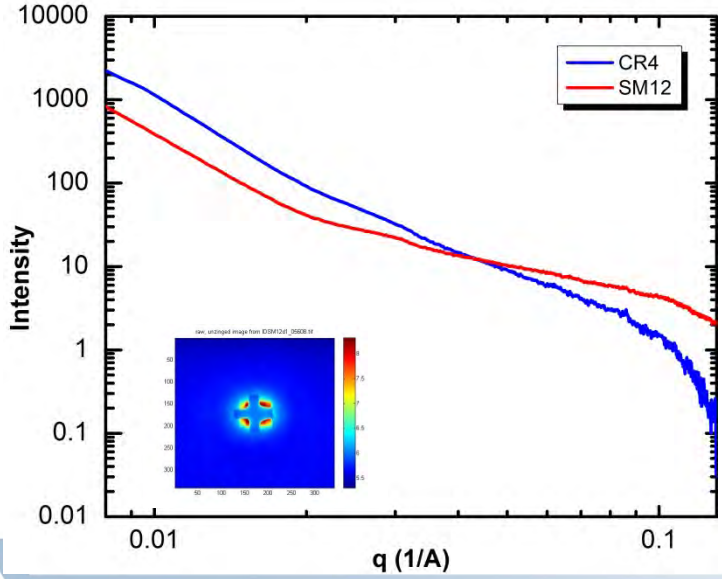
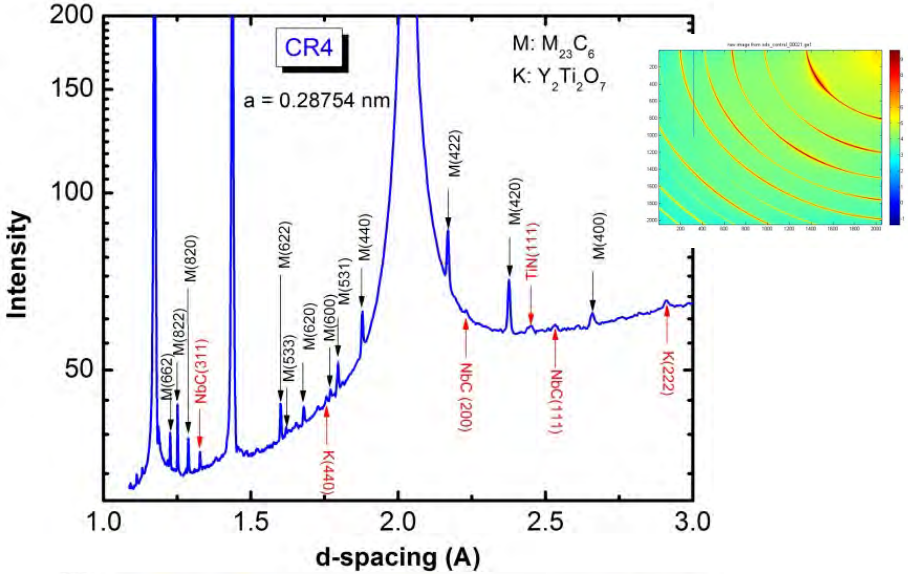
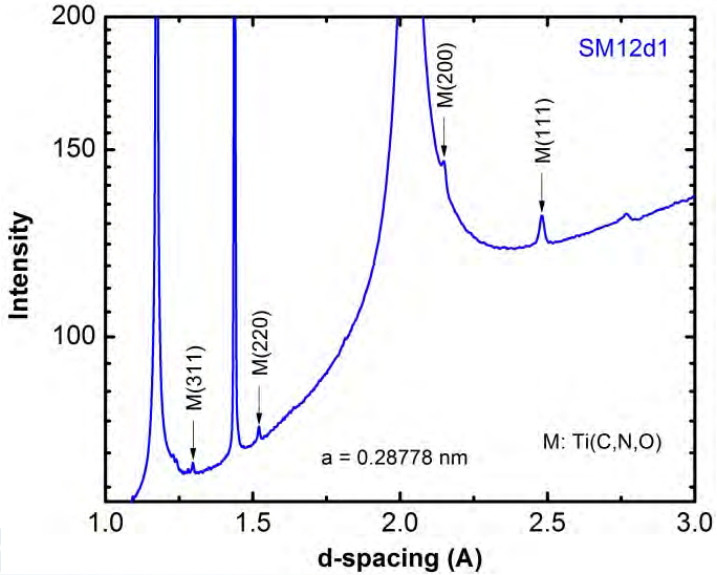
Characterization of Nano-Phases in Oxide-Dispersion-Strengthened Steels by WAXS/SAXS

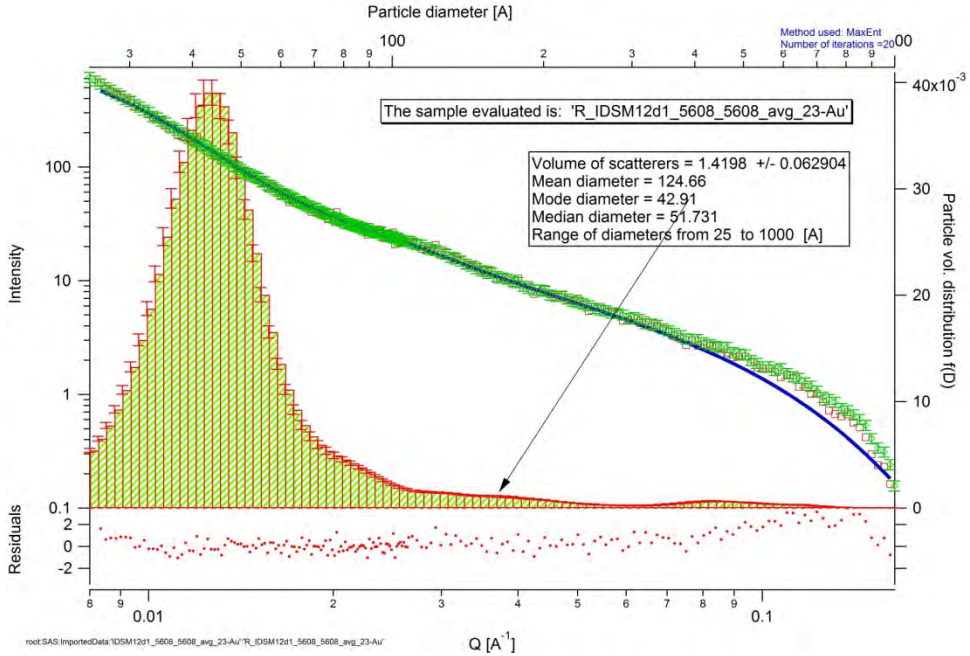
- CR4 was an early-developed heat of 14YWT with a high level of C and O, resulting in a high concentration of M23C6.
- SM12d is a new-generation 14YWT with improved chemistry and processing controls.

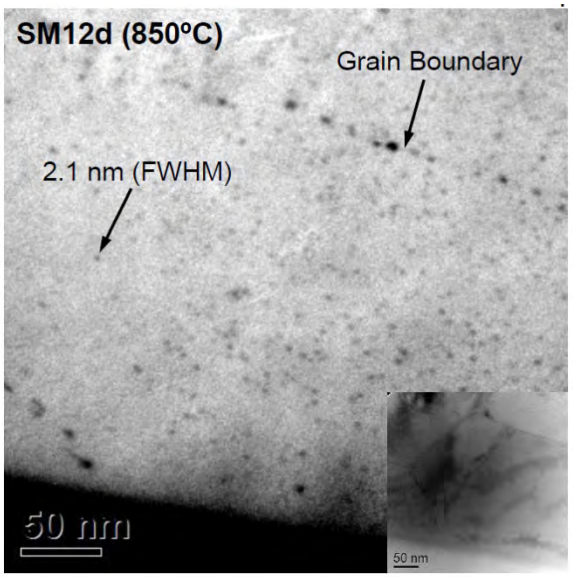


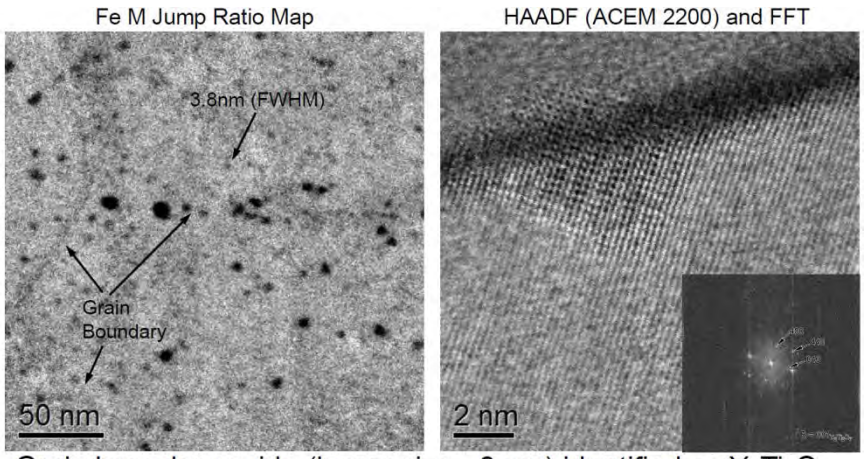




Nano-sized Oxide-clusters Revealed by SAXS in 14YWT-SM12 Heat





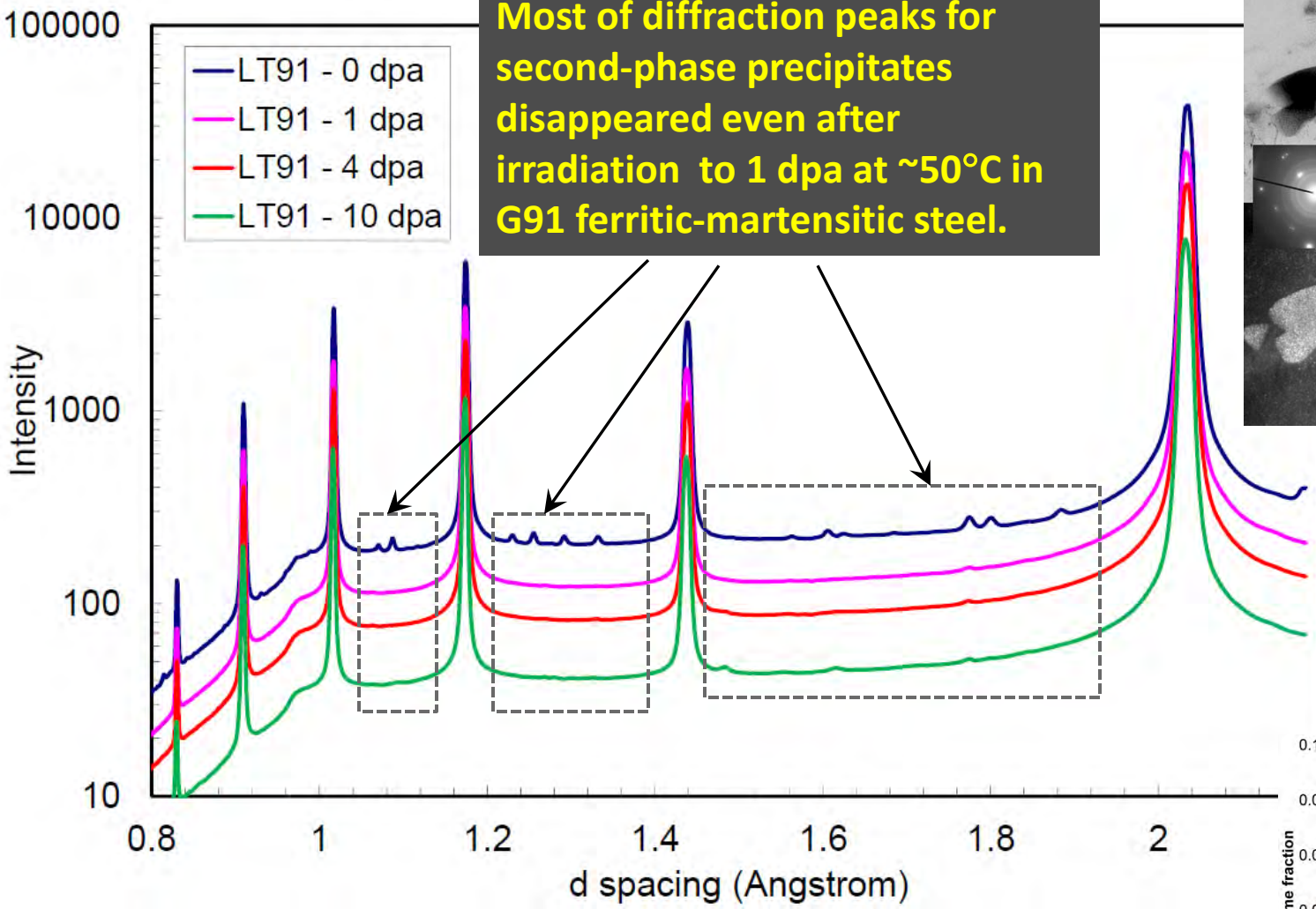


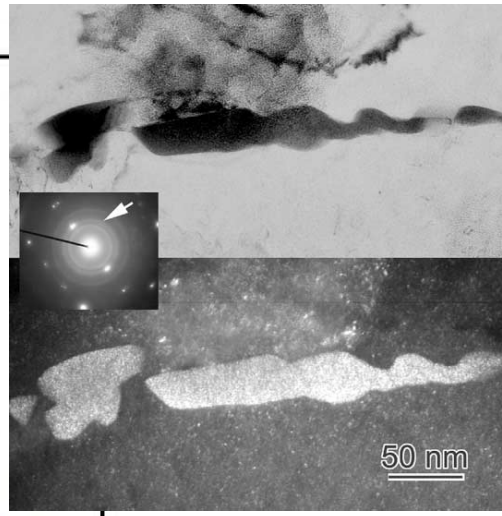
Grain boundary oxide (larger size ~8 nm) identified as $Y_2Ti_2O_7$

- SAXS detected ~4 nm oxide clusters
- SAXS results agree well with EFTEM measurements.

Phase Stability under Irradiation

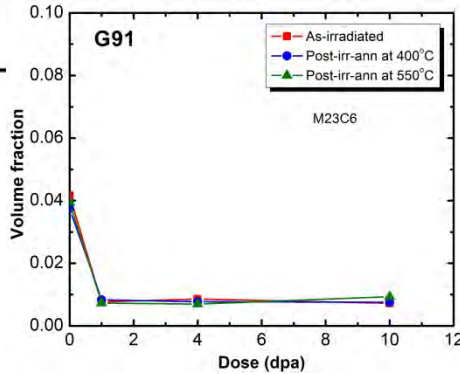
Most of diffraction peaks for second-phase precipitates disappeared even after irradiation to 1 dpa at ~50°C in G91 ferritic-martensitic steel.





TEM showed $M_{23}C_6$ became amorphous after irradiation.

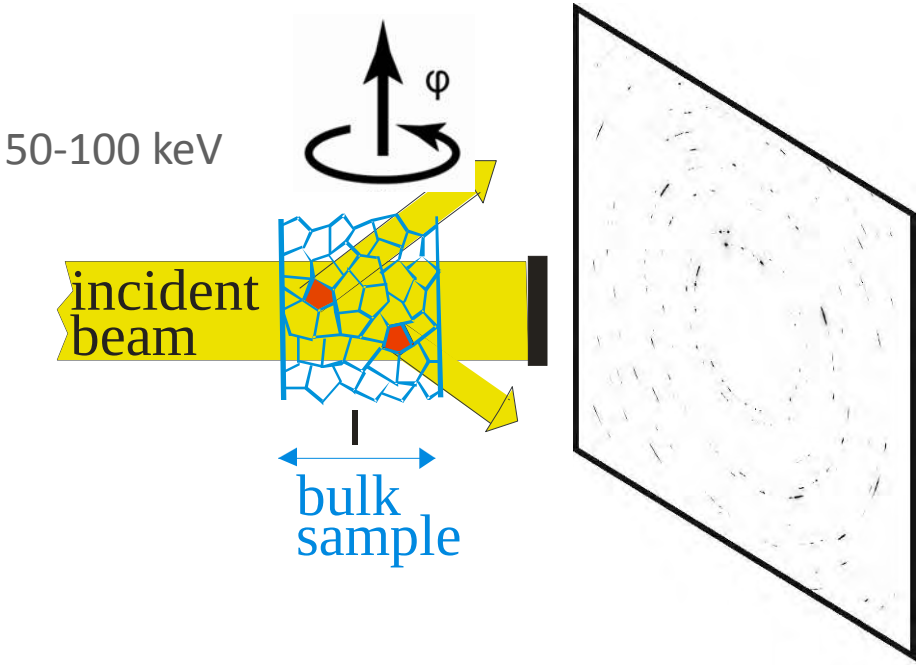
(Sencer et al (2002))

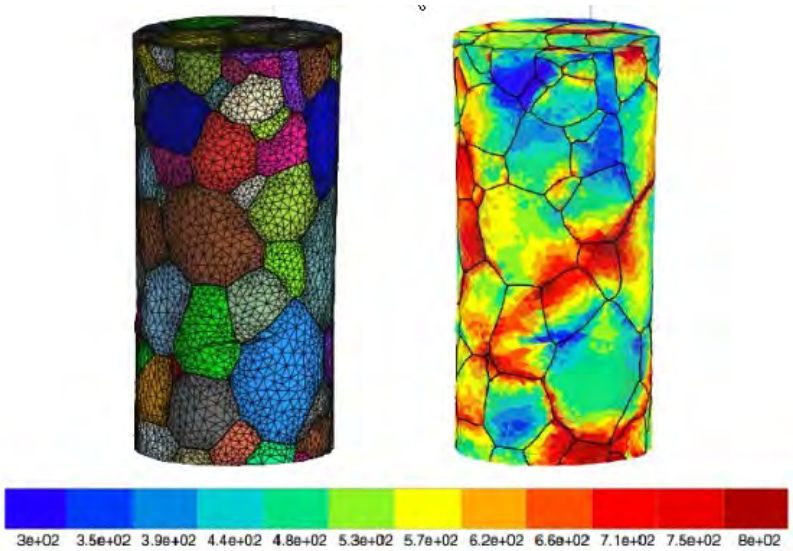


- Volume fraction of M23C6 decreased significantly after irradiation.
- MX showed significant chemistry change after irradiation

3D Characterization: High Energy Diffraction Microscopy (HEDM)

- Enable non-destructive characterization of microstructural and micromechanical response of individual grains with the bulk of a polycrystalline specimen.



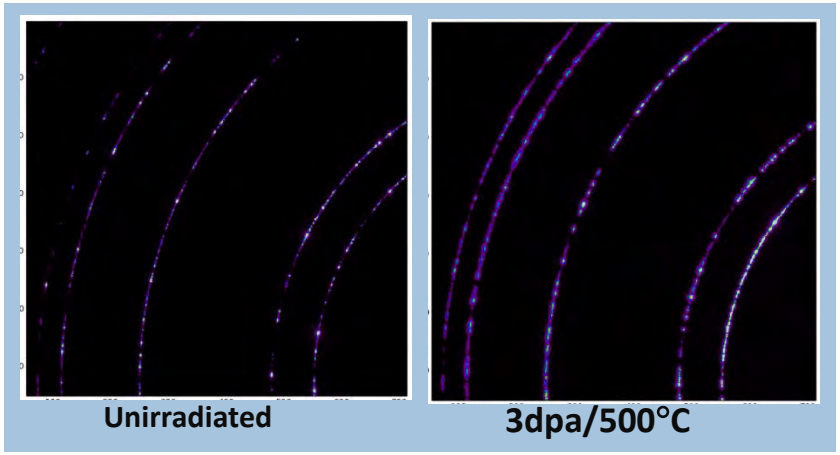


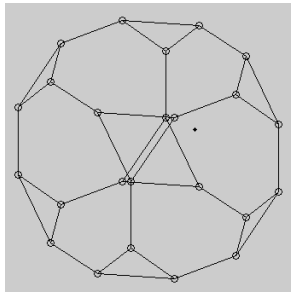
FEM simulation of von Mises stress in a Ti alloy sample loaded to 500 MPa. (Ludwig, et al, MSE A524 (2009))

- Near-field HEDM: grain shape, orientation
- Far-field HEDM: strain, orientation
- >100 grains in mm-size samples *in situ*

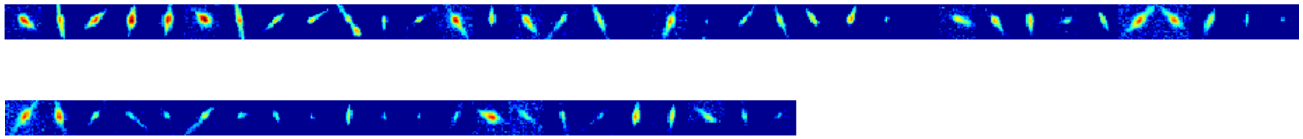
Far-Field HEDM of Neutron-Irradiated HT-UPS Steel

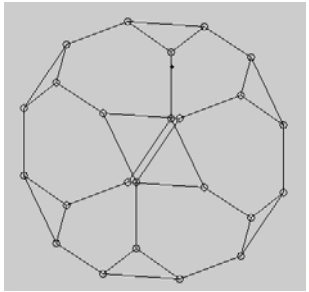
- HT-UPS (High-Temperature Ultrafine-Precipitation Strengthened) stainless steel
- Neutron irradiated to 3 dpa at 500°C
- APS 1-ID beamline, 86 keV



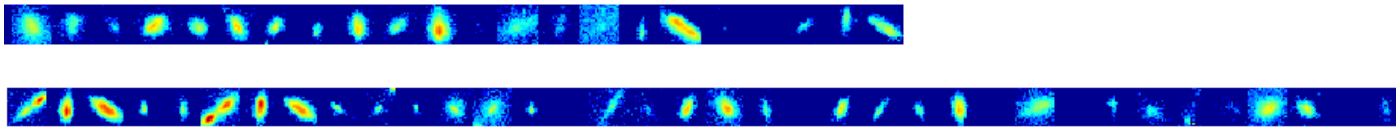


Unirradiated, a_0 (unirr) = 0.3593 nm;



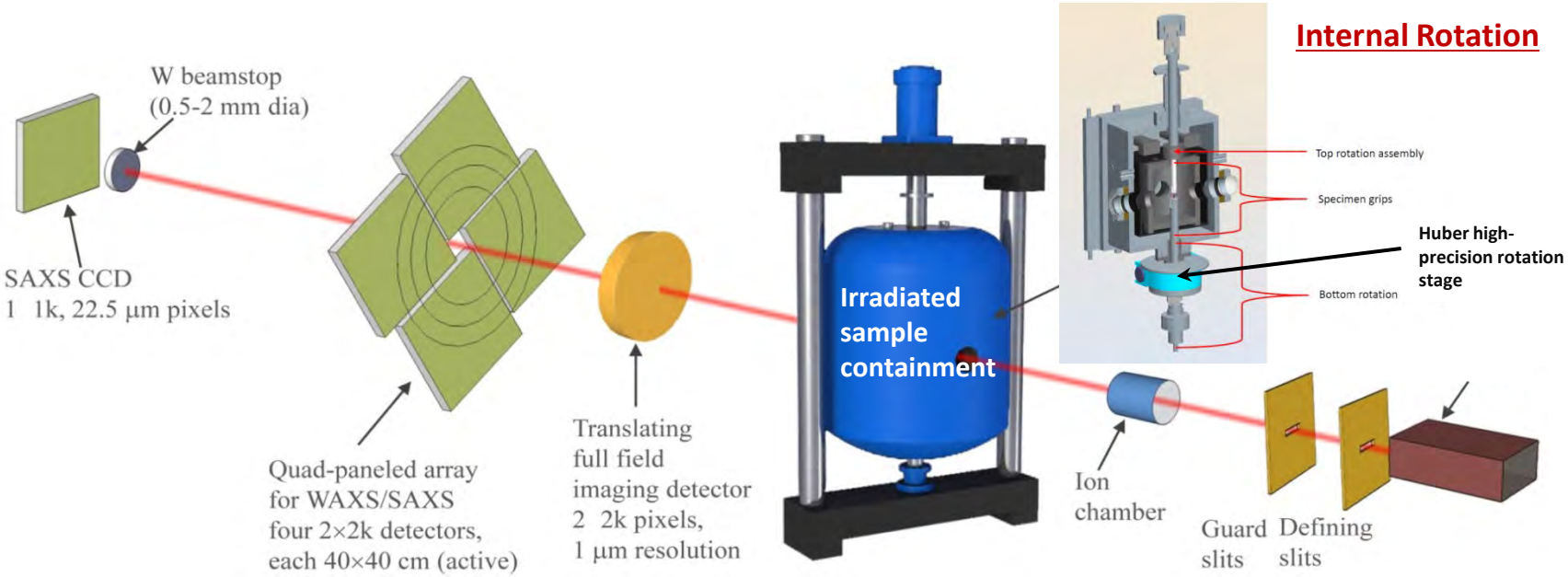


3dpa/500°C, a_0 (3dpa/500°C) = 0.3590 nm;

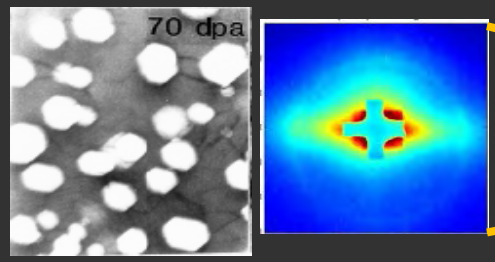


Showed negative hydrostatic strains (ε_{xx}, ε_{yy}, ε_{zz})

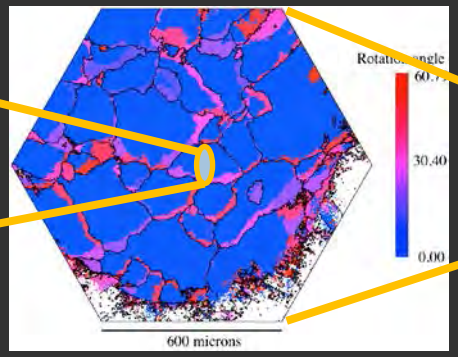
Multi-Probe *in situ* Characterization of Irradiated Materials



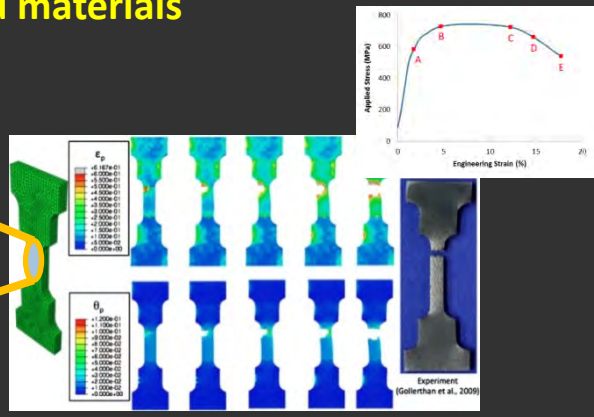
Combined techniques and intense, penetrating hard x-rays will visualize material evolution across relevant length scales under thermal-mechanical loading in irradiated materials



Nanoscale: diffraction and SAXS



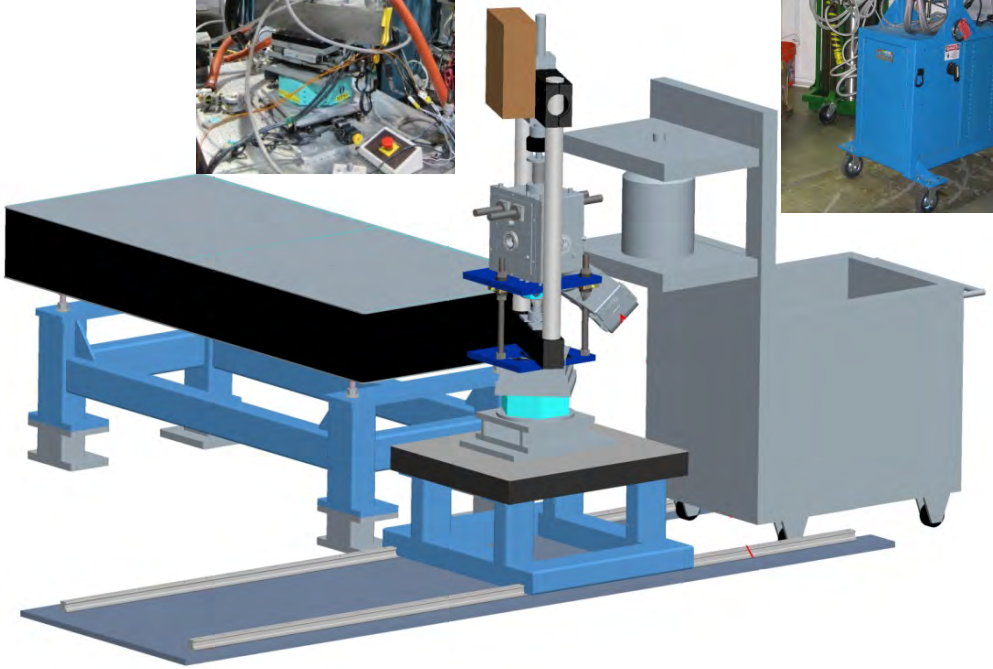
Mesoscale: diffraction microscopy



Macroscale: mechanical loading

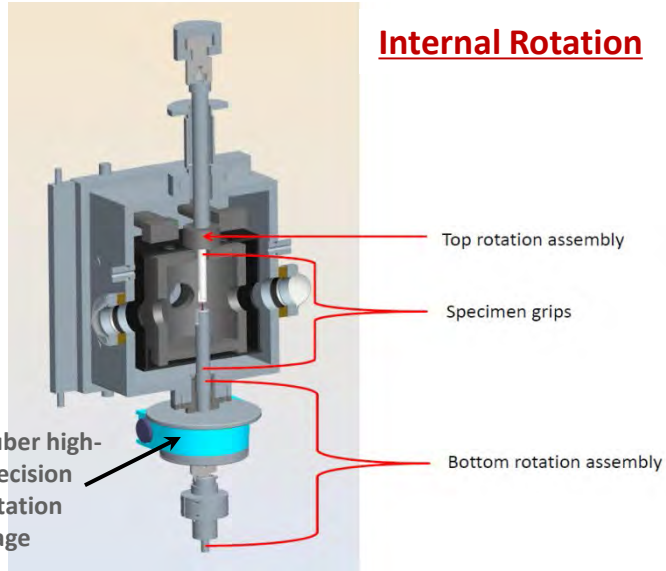
Experiment Module

Worldwide-unique x-ray sample environment
- Internal radiation shielding for activated samples
- Temperature: to 1000° C
- Vacuum: $1x10^{-5}$ Torr
- Tension, creep, fatigue loading
- In-grip rotation for tomography & diffraction microscopy



Grain-scale probes of mechanical behavior

Internal Rotation



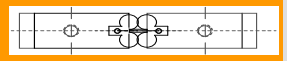
Integrated Radiation Shielding

W plates & shutters

Safe Handling of Radioactive Materials

Specimen receiving, unpackaging, survey, cleaning

Specimen cutting, treatment, loading/unloading



Load the experiment assembly into a shielded transport containment

HP survey

On-site Radiological Facility

HP survey

Unload the experiment assembly into a shielded transport containment

Transfer between Radiological Facility and APS (Special Metals)

Advanced Photon Source

Load the experiment assembly into a shielded transport containment

HP survey

Synchrotron Radiation experiments

Unload the experiment assembly into a shielded transport containment

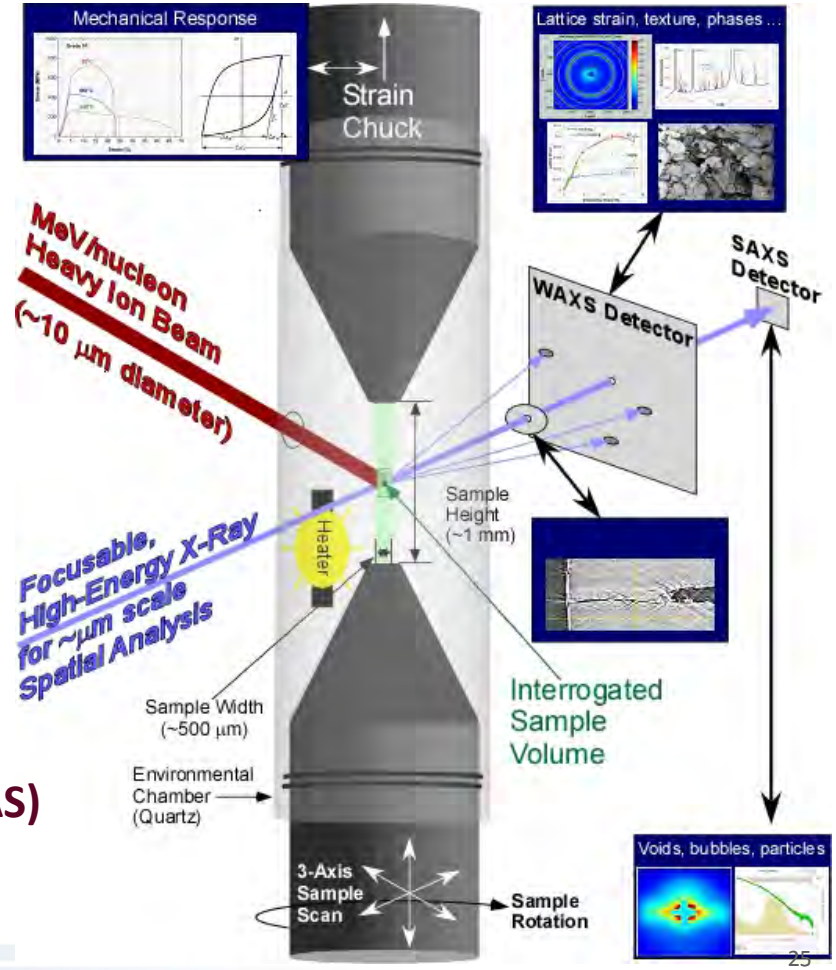
HP survey

Proposal – eXtreme MATerials beamline (XMAT)

Lead: Mike Pellin (ANL)

A new beamline at APS for *in situ* studies of nuclear energy materials under irradiation, temperature, stress, and environment.

XMAT will provide x-ray probes for *in-situ* study of materials in simulated nuclear reactor environments, enabling rapid evaluation of new materials performance under extreme service conditions including structural materials and for the first time nuclear fuels.



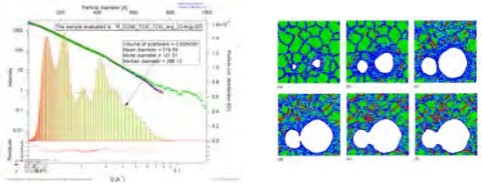
XMAT is made possible by combining two of Argonne's unique capabilities:

1. **Energetic, Heavy Ion Beams** **(ATLAS)**
2. **Focusable, High Energy X-Rays** **(APS)**

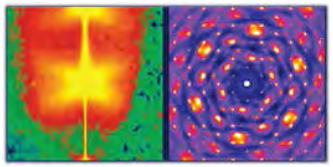
Summary

- Recent studies showed great promise of using synchrotron radiation to understand dynamic behavior and radiation damage of nuclear reactor materials over a wide range of lengh scales.
- Combination of x-ray techniques for *in situ* studies of nuclear materials allow concurrent characterization of microstructural features over wide range of length scales and direct correlation of structure and properties.
- Efforts are being made to apply these state-of-the art techniques to activated materials to better understand radiation damage and effects.

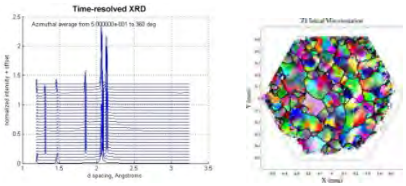
SAXS:
voids, bubbles, precipitates



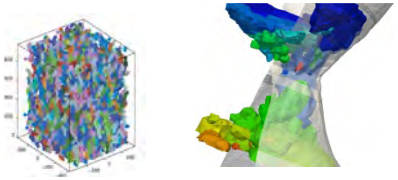
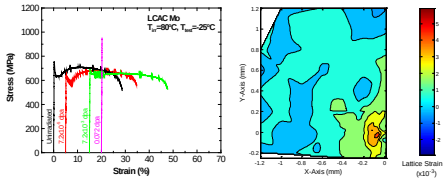
Diffuse scattering:
SIA and vacancy clusters



XRD:
Phases, grains, dislocations, lattice strain, texture



Tomography:
Cracks, voids, grain structure



mechanical loading:
macroscale stress-strain response