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Understanding irradiation behaviors of ultrawide bandgap Ga_2O_3 high temperature sensor materials for advanced nuclear reactor systems

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NSUF Nuclear Science User Facilities

Nuclear Science User Facilities (NSUF)
Annual Program Review

Project Overview

Project Team/Participants

- **PI and Associated Institution:** Dr. Ge Yang (Department of Nuclear Engineering, North Carolina State University)
- **Co-PI(s)/Collaborators and Associated Institutions:** Dr. Cheng Sun (Idaho National Laboratory), Dr. Ayman Hawari (North Carolina State University), Dr. Yaqiao Wu (CAES/Boise State University)
- **Identification of NSUF Facilities:** (1) North Carolina State University, PULSTAR Nuclear Reactor; (2) Center for Advanced Energy Studies (CAES)
- **NSUF Facility Technical Lead:** Dr. Ayman Hawari (North Carolina State University, PULSTAR Reactor Program); Dr. Yaqiao Wu (Center for Advanced Energy Studies, CAES)
- **Post-Doc/Students and Facility Staff Members:** Robert McRobie, Lucia Rebeca Gomez, Da Cao, Colby Fleming, Ming Liu, Ching-Heng Shiau

Motivation – A strong need for (U)WBG sensor materials

- Radiation-hard sensor materials and devices are key components for developing advanced nuclear energy systems

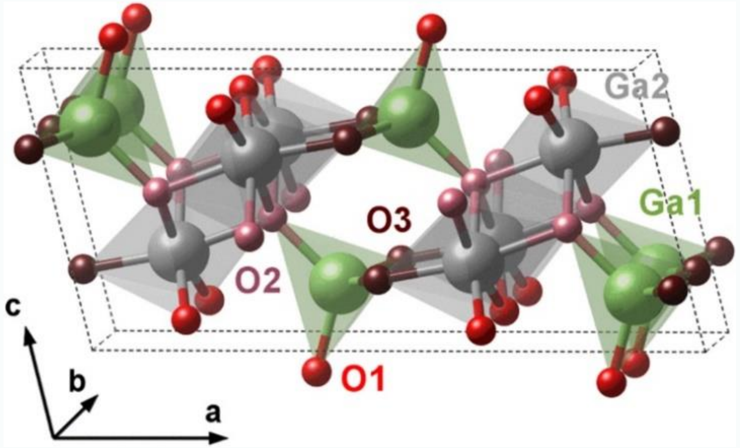
- Many traditional sensors have certain limitations for harsh environment nuclear applications due to their intrinsic material properties.
  - ☐ Survivability in radiation field
  - ☐ High temperature tolerance

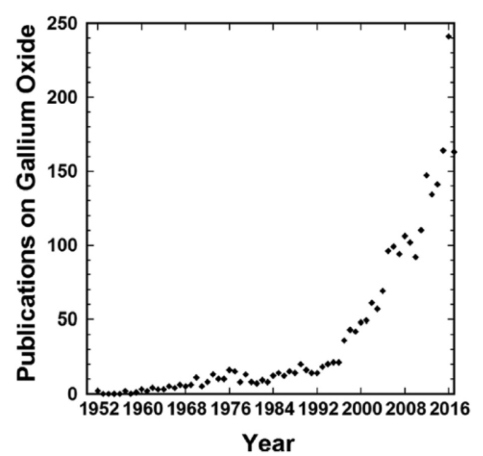
- Wide and ultrawide bandgap (U)WBG semiconductors are much less susceptible to displacement damage by irradiation than elemental and narrow bandgap compound semiconductors

Motivation – Ga_2O_3 for nuclear sensors and instrumentation

- **β-Ga_2O_3 is an emerging ultrawide bandgap compound that has many desired material advantages for extreme conditions**
  - Thermal stability (M. P. > 1800 ^{o}C)
  - The most recent ultrawide bandgap material (4.5 – 5.1 eV)
  - Very high breakdown electric field (8 MV/m)
  - High quality bulk single crystals from melt
  - Reasonable availability of Ga_2O_3 materials

- **β-Ga_2O_3 holds high promise for fitting into many nuclear-related application scenarios with the performance that are not met by currently used materials**
  - Harsh environment applicability
  - High sensing performance
  - Versatile and cost-effective synthesis and fabrication





S. J. Pearton et al., “A review of Ga_2O_3 materials, processing, and devices,” Appl. Phys. Rev., vol. 5, no. 1, p. 011301, 2018.

Systematic irradiation research is urgently needed to study and deploy the emerging Ga_2O_3 nuclear sensor material!

Objective of this Project

Understand fundamental irradiation behaviors of emerging ultrawide bandgap Ga_2O_3 high temperature sensor materials through a series of well-designed irradiation experiments and post-irradiation examination (PIE) tests

Meeting the urgent need of the nuclear community!

Use of Two NSUF Partner Facilities with Complementary Capabilities

(1) North Carolina State University (NCSU) Nuclear Reactor Program PULSTAR User Facility (for neutron irradiation and positron measurements)

(2) Microscopy and Characterization Suite (MaCS) at Center for Advanced Energy Studies (CAES) (for PIE microstructural, compositional and cathodoluminescence examination)

Objective: Establish fundamental understanding of irradiation behaviors of ultrawide bandgap Ga_2O_3 sensor materials through targeted irradiation and PIE experiments

Task 1:
Conduct room temperature irradiation experiments of Ga_2O_3 samples at NCSU's PULSTAR Nuclear Reactor

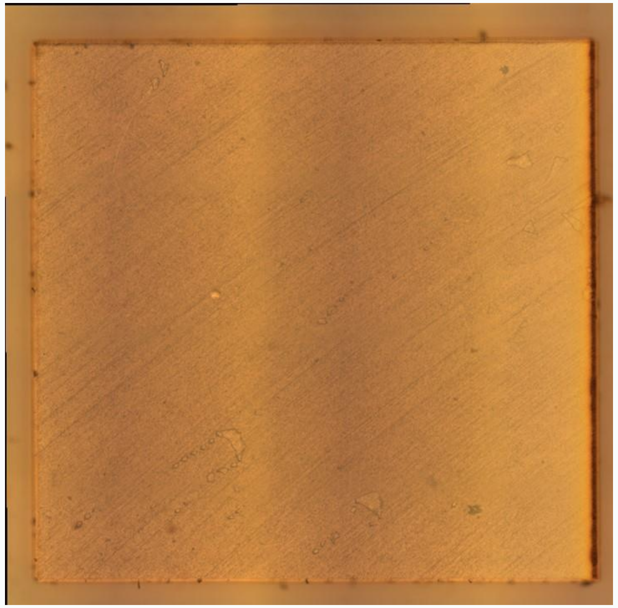
Task 2:
Perform systematic post-irradiation examination at NCSU and CAES to analyze Ga_2O_3 samples that have been irradiated at room temperature and compare with unirradiated Ga_2O_3 reference samples

Task 3:
Conduct high temperature irradiation experiments of Ga_2O_3 samples using a newly built in-pool high temperature furnace facility at NCSU's PULSTAR Nuclear Reactor

Task 4:
Perform systematic post-irradiation examination at NCSU and CAES to analyze Ga_2O_3 samples that have been irradiated at high temperature and compare with unirradiated Ga_2O_3 reference samples

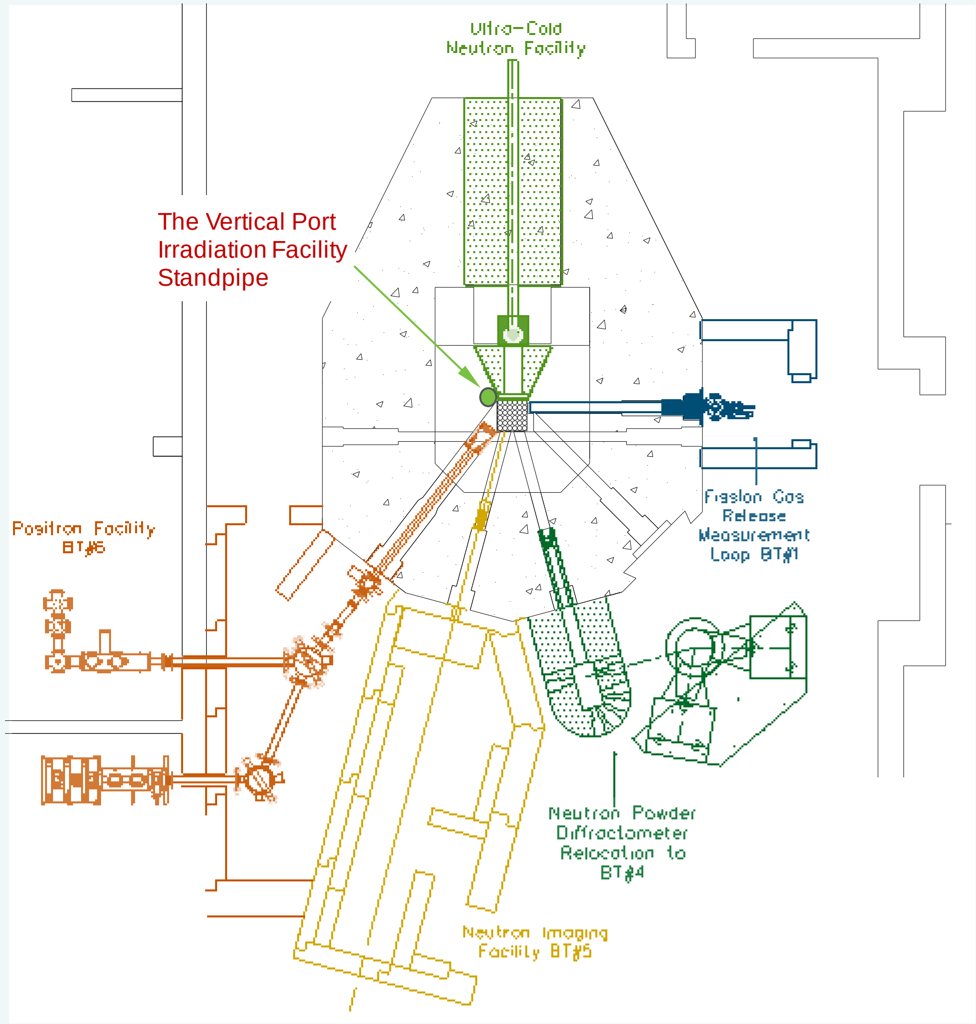
A series of Ga_2O_3 samples have been carefully prepared

Comprehensive chemical cleaning work have been conducted



Polarized light microscopy image of Ga_2O_3

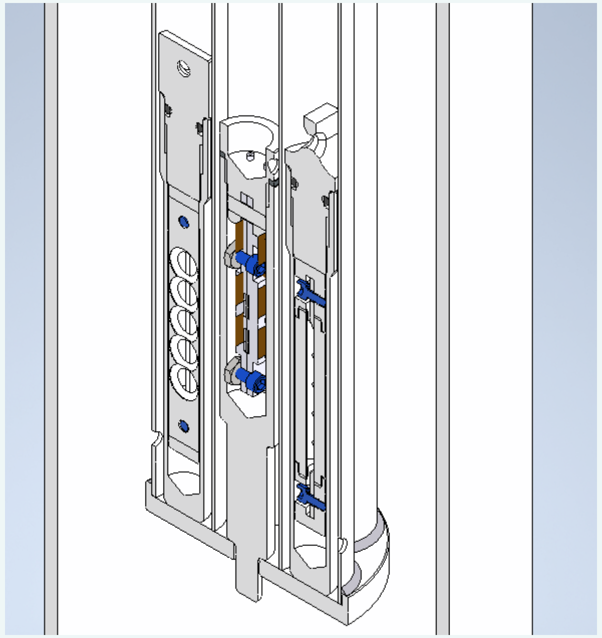
Location of the Irradiation Experiment



Location of the Vertical Port Irradiation Facility

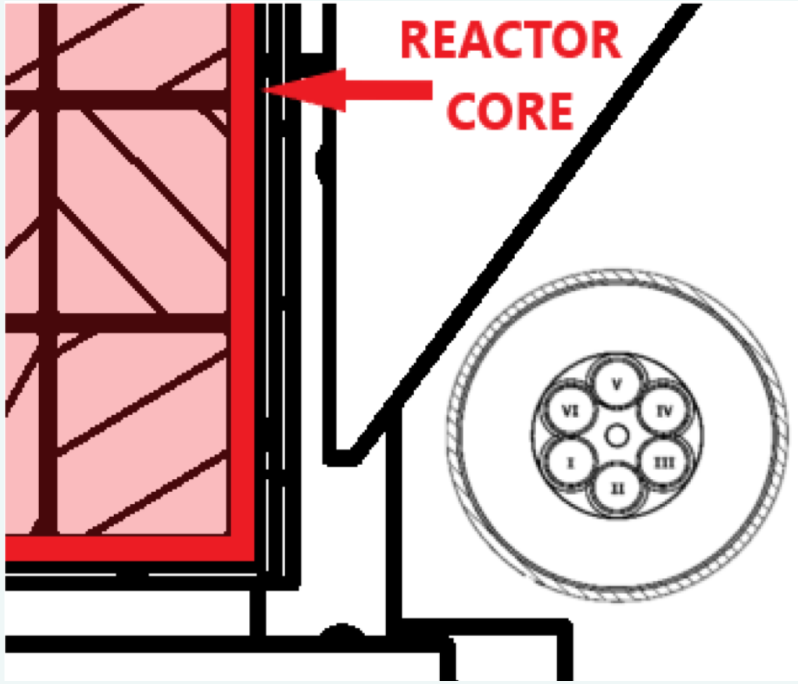
- 8”ID vertical dry well (standpipe) between BT#6 and UCN column
- In-Situ Measurement stations at the pool top level
- Flexible umbilical at the pool top connecting measurement stations and the standpipe

Ga_2O_3 Samples and Irradiation Instrument



- Sample capsules are made of Aluminum to minimize activation
- Each capsule is sealed and contains 5 samples
- Doses are calibrated using counting wires

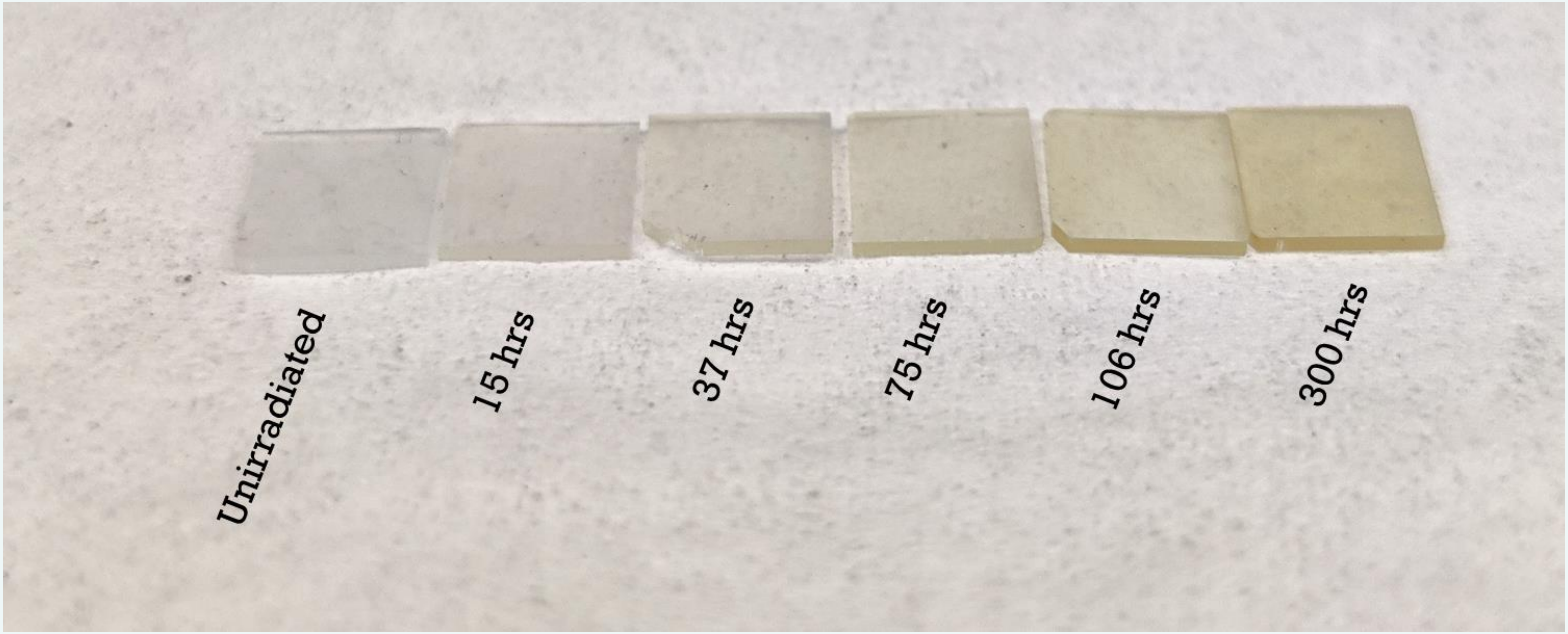
Room Temperature Irradiation Experiment



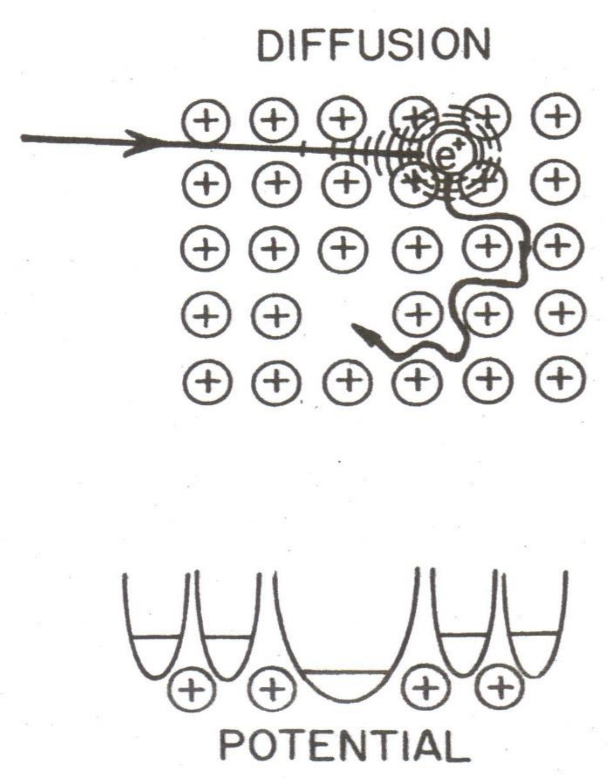
Sample Set	Integrated Exposure (MW-hrs)	Thermal Fluence (n/cm²/s)	Fast Fluence (n/cm²/s)	Total Fluence (n/cm²/s)
1	300.43	1.31E+18	3.12E+17	1.63E+18
2	105.69	6.45E+17	2.83E+17	9.28E+17
3	74.95	3.61E+17	1.14E+17	4.75E+17
4	37.08	1.84E+17	4.09E+16	2.25E+17
5	14.93	7.49E+16	1.75E+16	9.24E+16
6	7.52	3.31E+16	7.40E+15	4.05E+16

- The 6 sample capsules, each containing 5 samples, were irradiated to different doses
- Doses are calibrated using counting wires

Samples after room temperature irradiation experiments



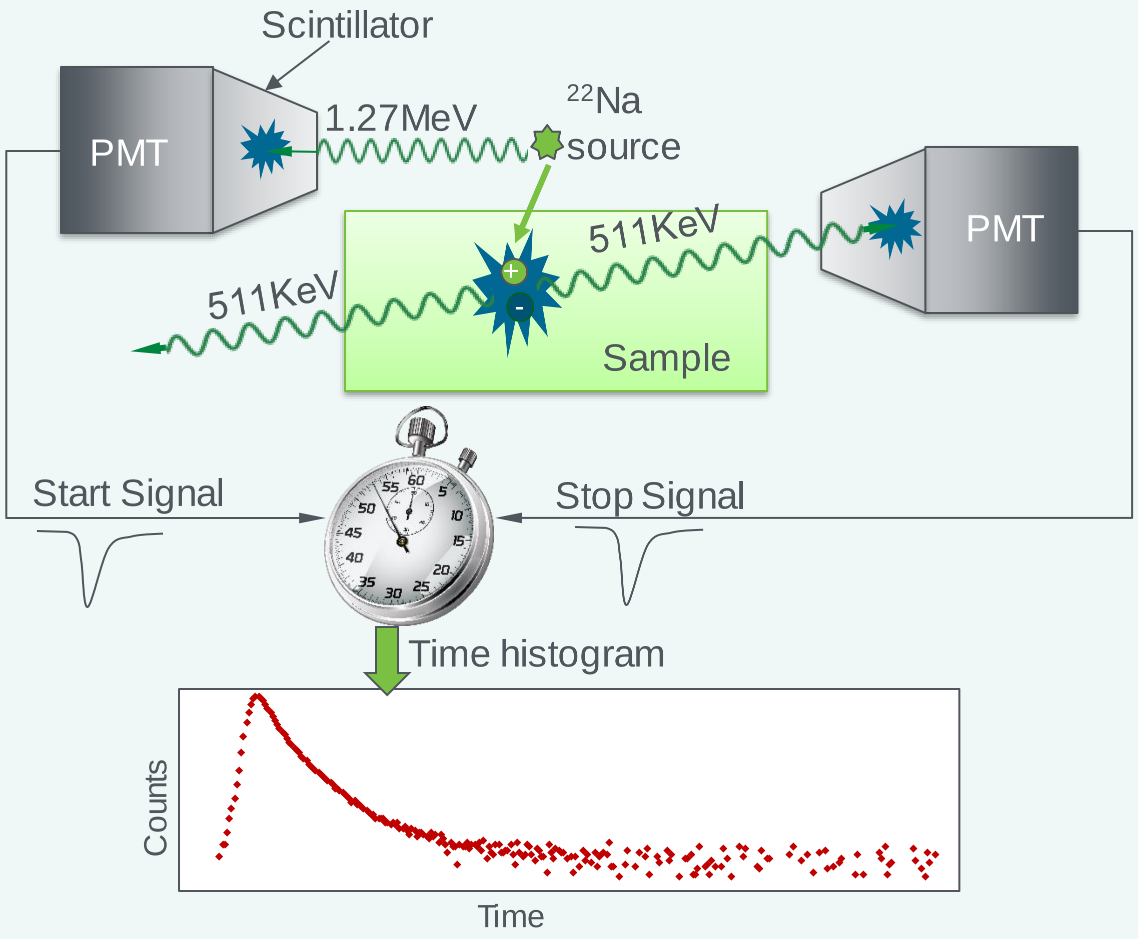
Positron – an Intrinsic Probe of Nanoscale



- Automatically seeks out vacancy-type defects – positrons “sense” certain types of defects acting as positron traps e.g. vacancies or impurities
- Annihilation takes place at defect site, yields information of defects
  - Lifetime -> electron density
  - DB spectrum -> electron momentum
  - Intensity -> defect concentration
- Provide quantitative nanoscale information averaged over macroscopic area/volume

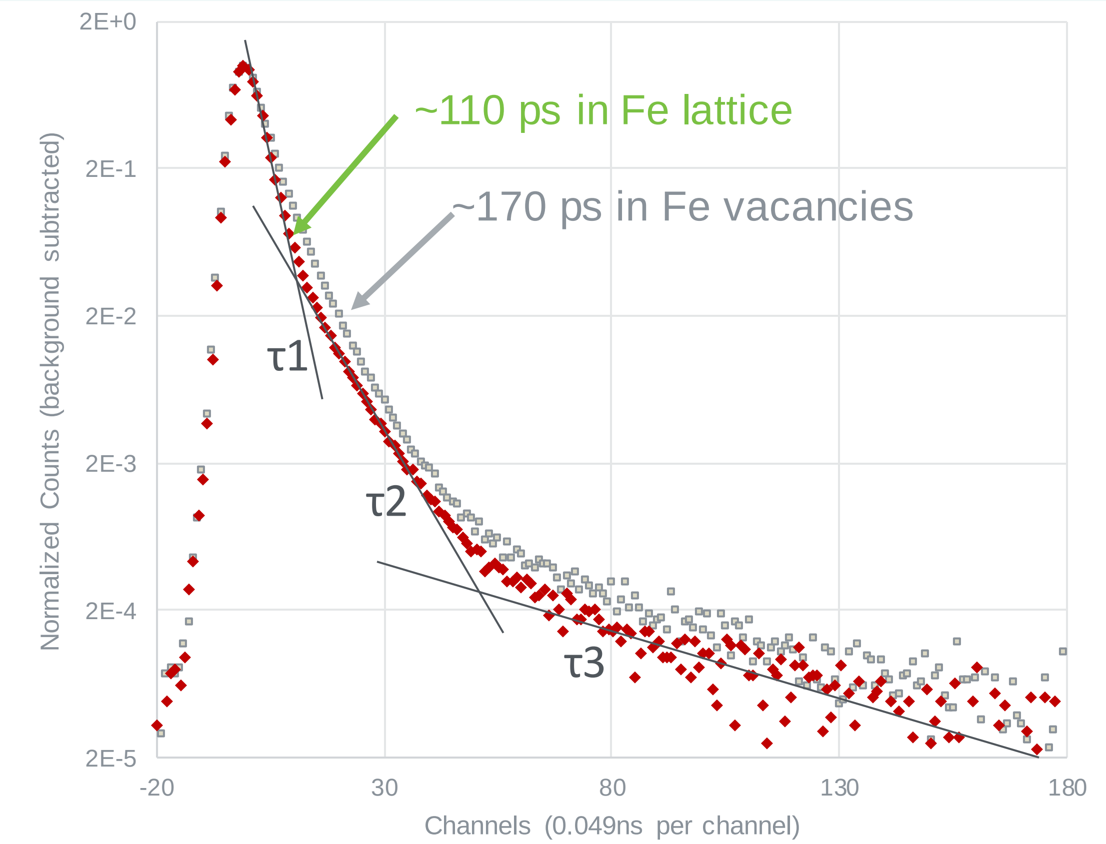
Missile vs. Carpet Bombing

Positron Annihilation Lifetime Spectroscopy



- Start signals – the coincident 1.27MeV gamma when a positron is born
- Stop signals – any of the 511keV annihilation gammas when a positron is dead
- PALS spectrum – a histogram of timing intervals between the birth and death of positrons
- Positron sources
  - Radioisotopes
  - Positron beams

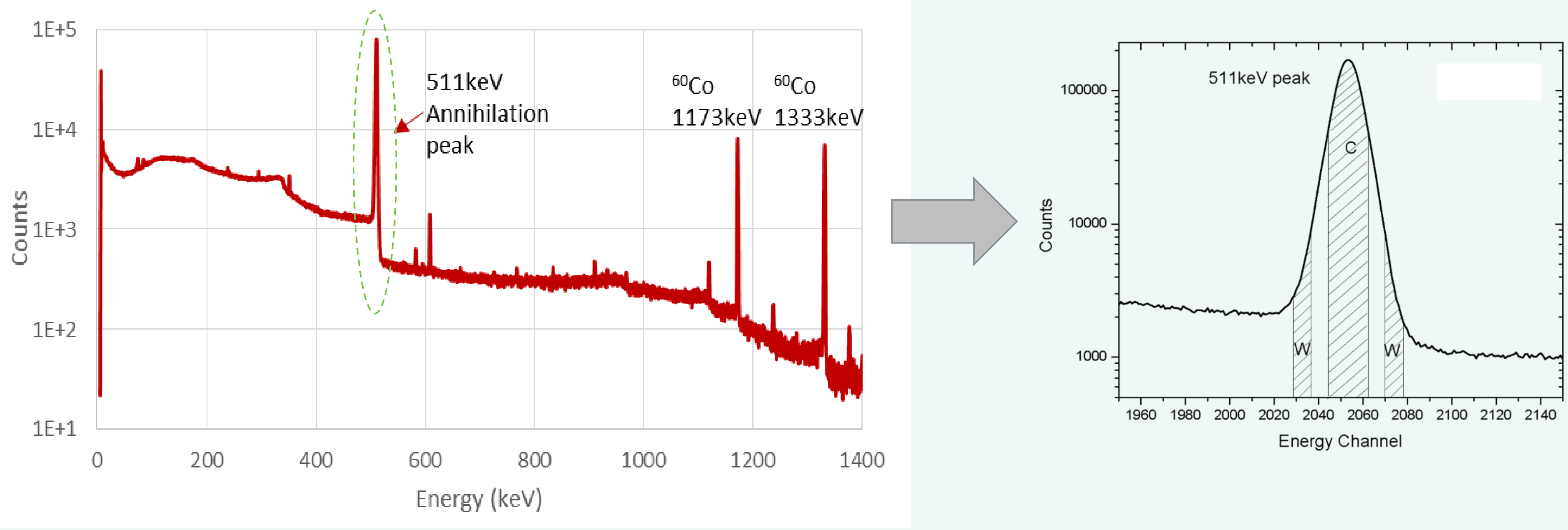
Positron Annihilation Lifetime Spectroscopy



$$N(t) = \sum_{i=1}^{n} \frac{I_i}{\tau_i} exp\left(-\frac{t}{\tau_i}\right)$$

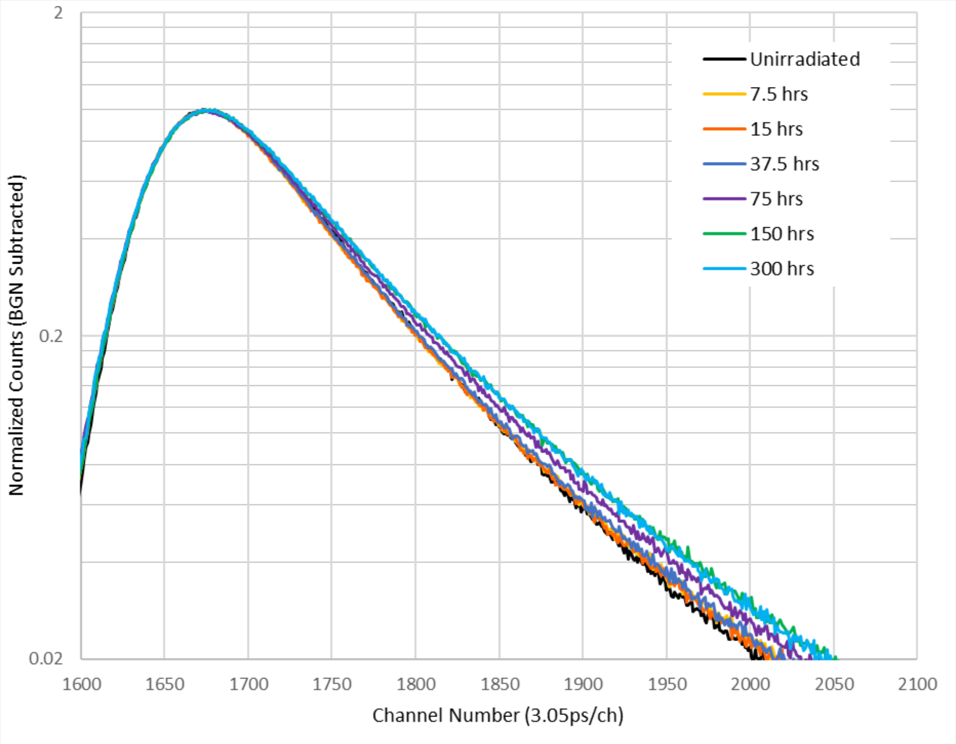
- Positron exponential decay lifetimes shown as lines of various slopes in the semi-log histogram and fitted by POSFIT, LT10, or PALSfit
  - Short positron lifetimes typically related to vacancy-type defects
  - Intermediate lifetimes typically related to vacancy clusters and/or grain boundaries
  - Long lifetimes with shallow slopes typically related to positronium annihilation in free volume voids

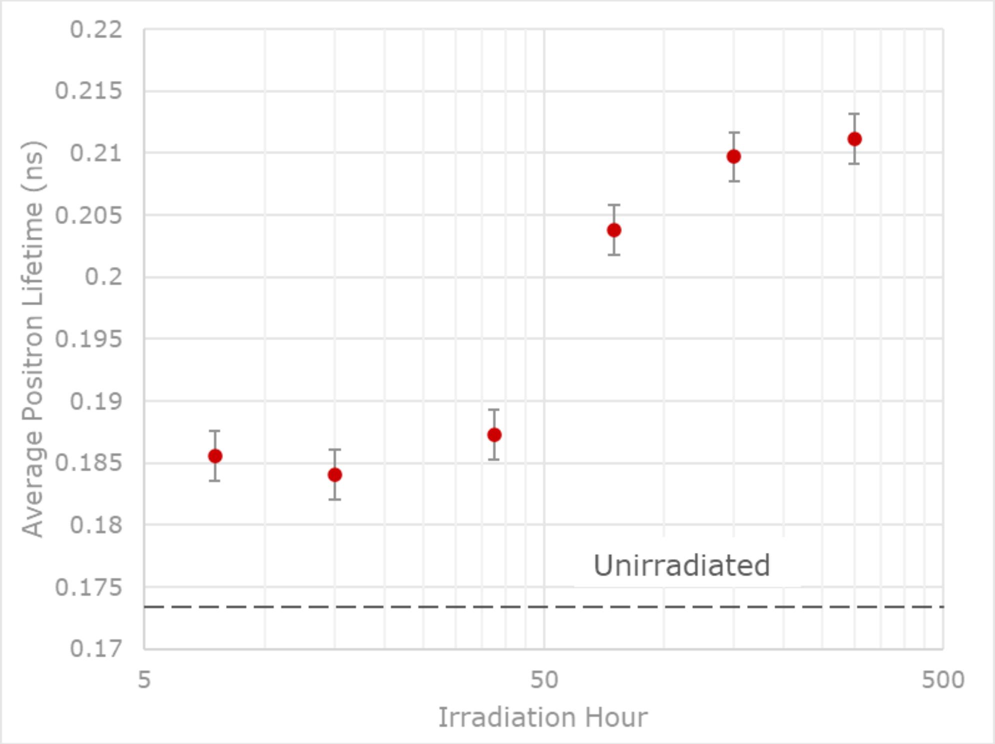
Doppler Broadening Spectroscopy (DBS)



- DBS examines the energy spectrum of the annihilation
- The width of the 511 keV peak is related to property of the defects
- S-parameter is defined as the ratio of the center region to the total counts in the whole peak

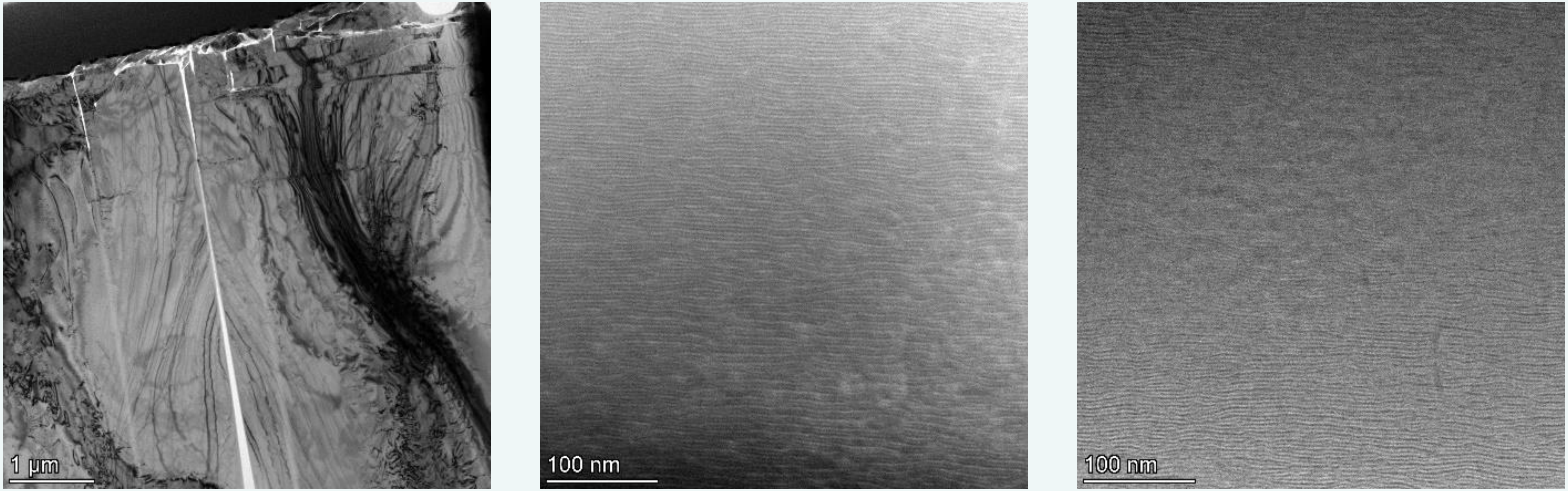
PALS Results





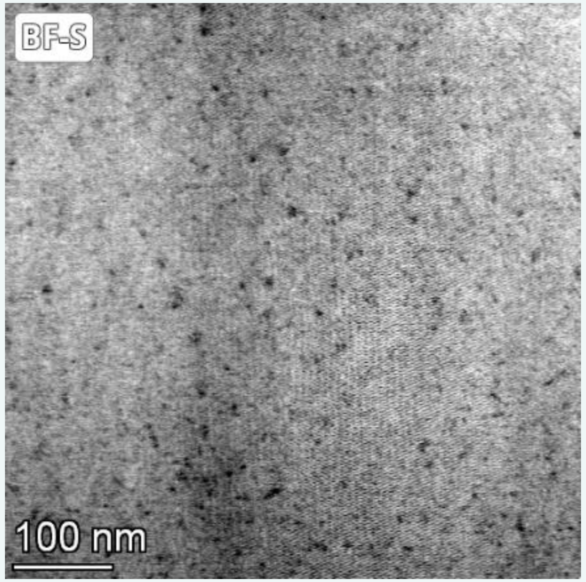
- ☐ Average positron lifetime increases vs irradiation dose
- ☐ Extra vacancy-type defects are introduced by radiation
- ☐ A possible transition occurs at ~50 hrs of irradiation (~3E+17 n/cm^2/s)

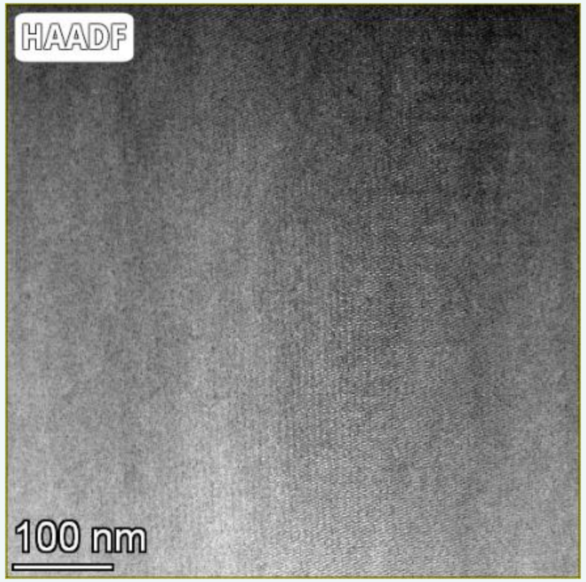
Transmission Electron Microscopy

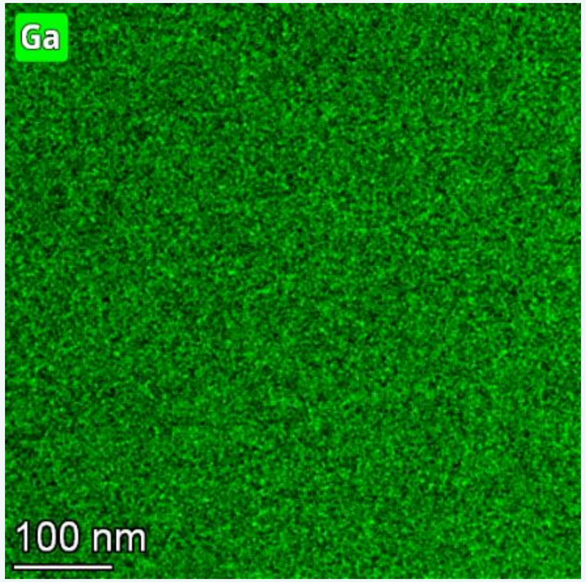


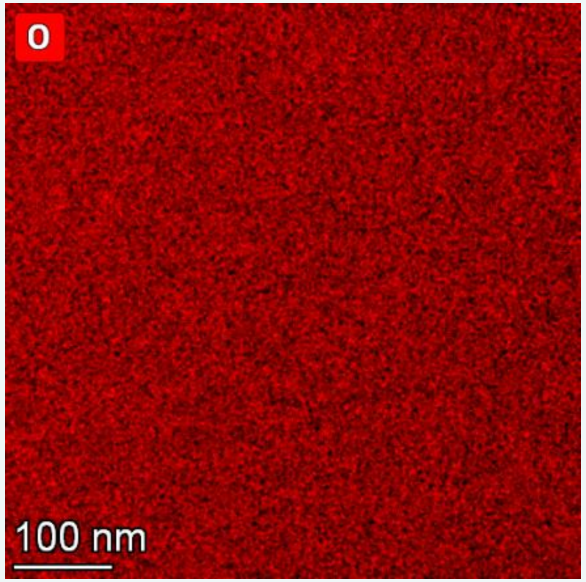
(left) Bright-field, (middle) HAADF, (right)STEM-BF

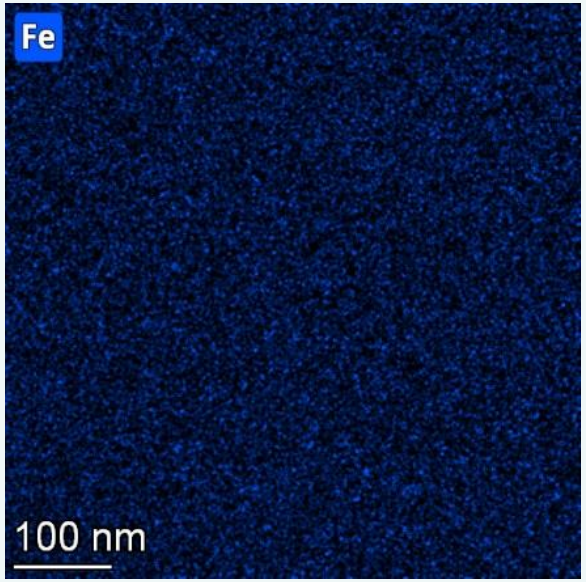
EDS measurements



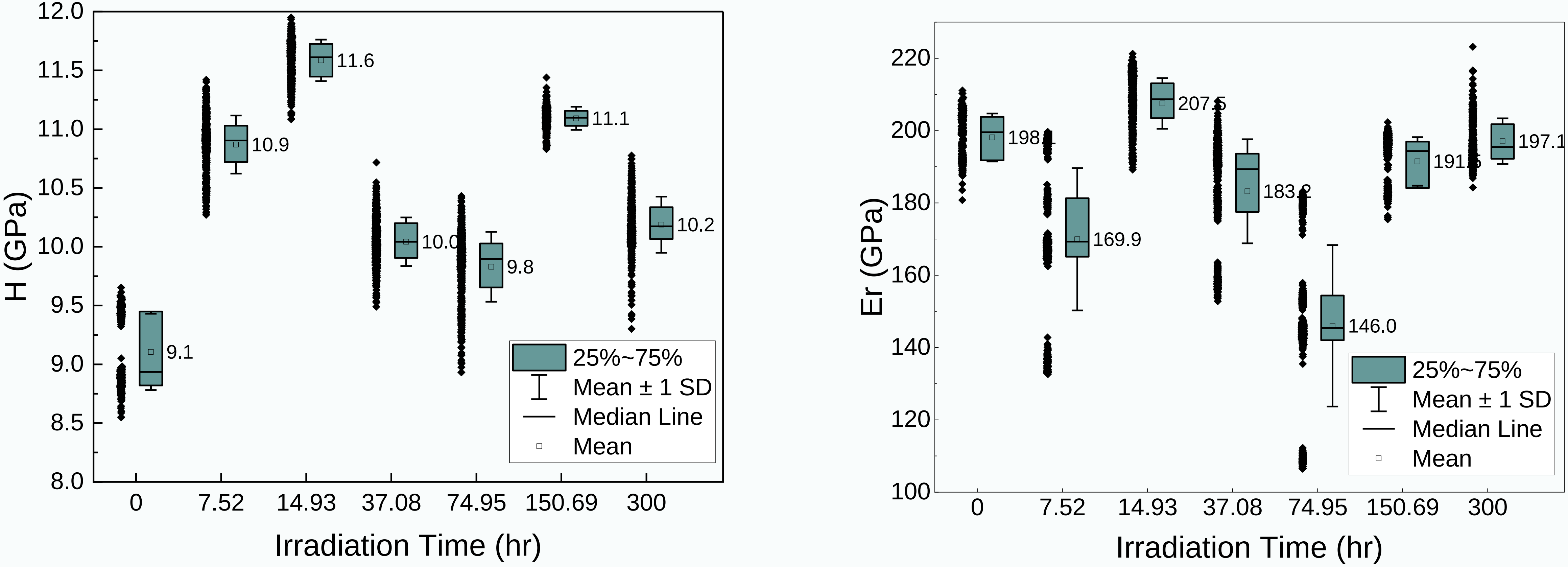






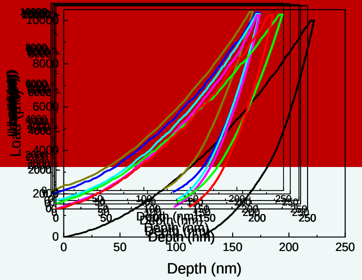


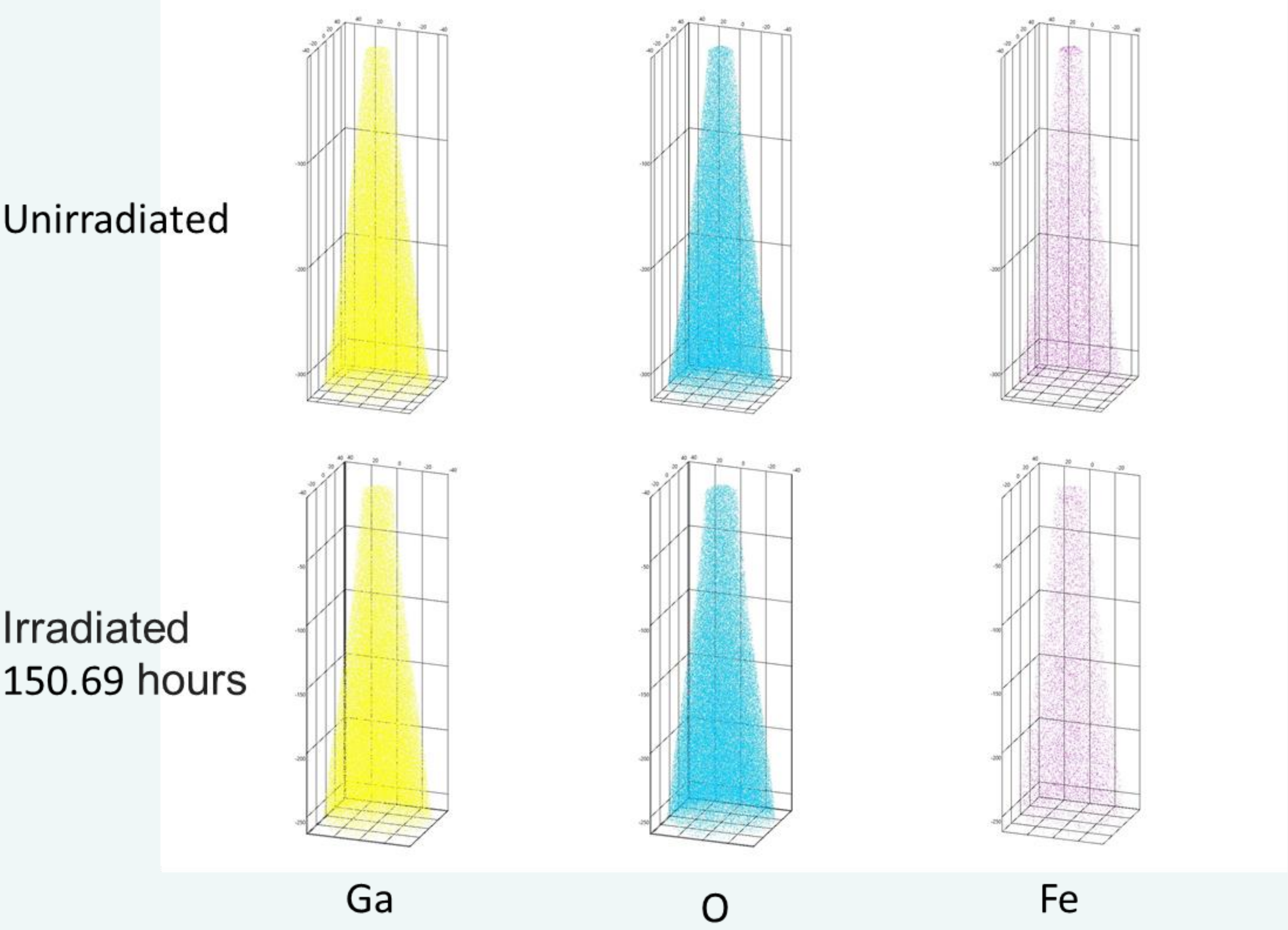
Nanoindentation



(Left) Hardness. (Right) Reduced Modulus

Atom probe tomography (APT)





Chemical composition of the LEAP tips (at. %)

	O	Ga	Fe
Unirradiated	56.86	43.127	0.013
7.52 hours	56.773	43.211	0.015
14.93 hours	56.764	43.225	0.011
37.08 hours	57.031	42.959	0.01
74.95 hours	56.982	43.006	0.01
150.69 hours	56.756	43.208	0.036
300 hours	57.145	42.815	0.041

Presentations/publications

(1) L. R. Gomez-Hurtado, R. McRobie, M. Liu, C. Fleming, A. I. Hawari, G. Yang, “Investig
the Evolution of Defects in Wide Bandgap β-Gallium Oxide”, 2023 American Nuclear Soci
Winter Meeting, November 15, 2023

(2) L. R. Gomez-Hurtado, R. McRobie, C-H. Shiau, Y. Q. Wu, M. Liu, C. Fleming, A. I. Hav
C. Sun, G. Yang, “Microstructure and Defect Study of Wide Bandgap β-Gallium Oxide”, 2
American Nuclear Society (ANS) Annual Meeting, June 12, 2023

(3) Robert Mcrobie, Ming Liu, Ayman Hawari and Ge Yang, “Radiation Induced Vacancy F
Gallium Oxide Nuclear Sensors,” NSUF Users Organization Meeting, July 12, 2022

Two more manuscripts are being prepared for submission.

Concluding Remarks

- All room temperature irradiation experiments of Ga_2O_3 samples have been done.
- A series of positron measurements are continuing at the PULSTAR reactor.
- Systematic microstructural characterization of Ga_2O_3 crystals at CAES are being conducted.
- 2 PhD students, 1 undergraduate student, and 1 post-doc researcher have been trained.
- We are working to prepare two manuscripts for publication to report recent scientific discoveries.

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Thanks for your attention!