Radiation And Nuclear Energy

NRRCAT/ATR NSUF Workshop

June 3, 2009

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Illinois Institute of Technology

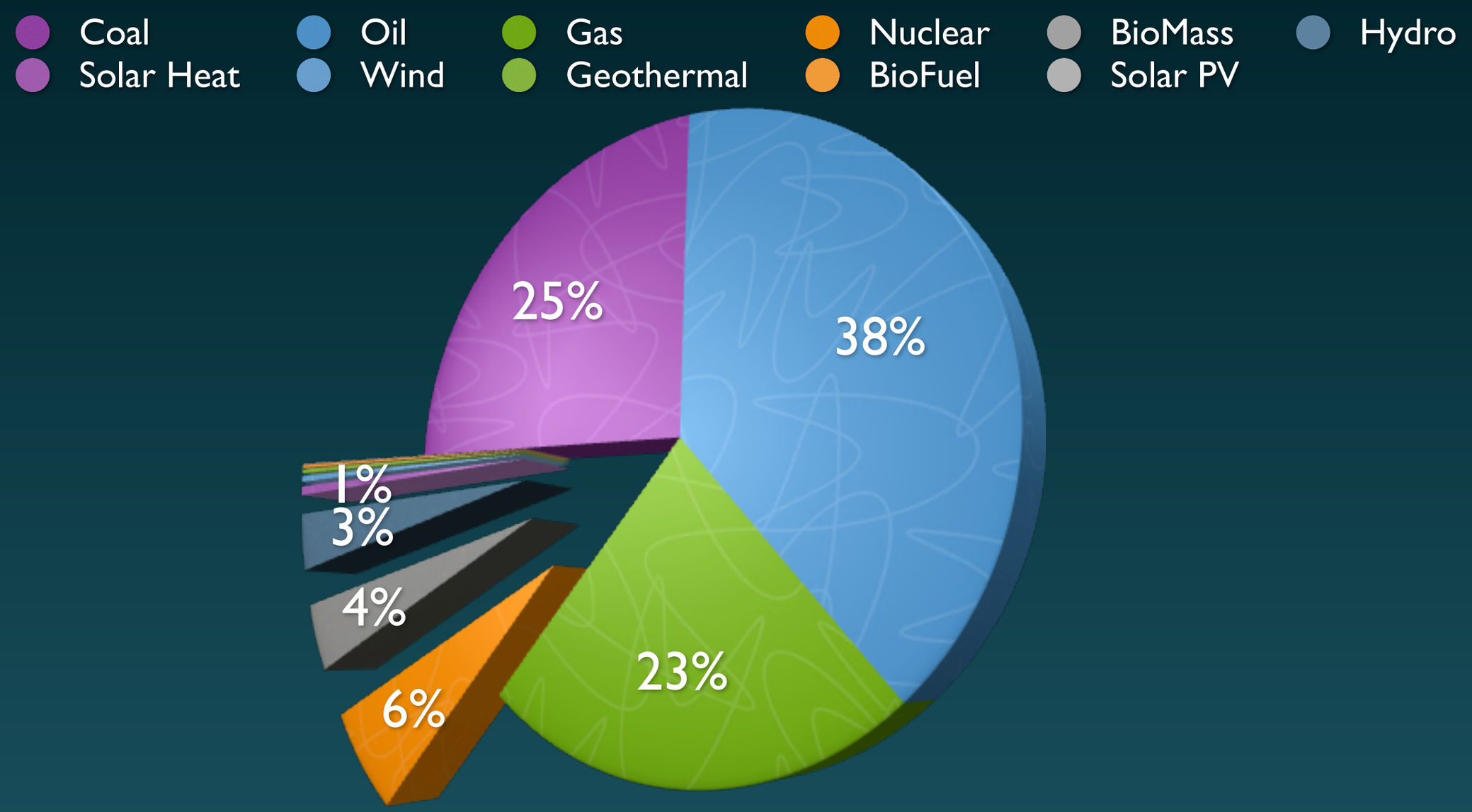
Nuclear and Radiological Research
Collaborative Access Team

Chicago IL 60616

Collaborators

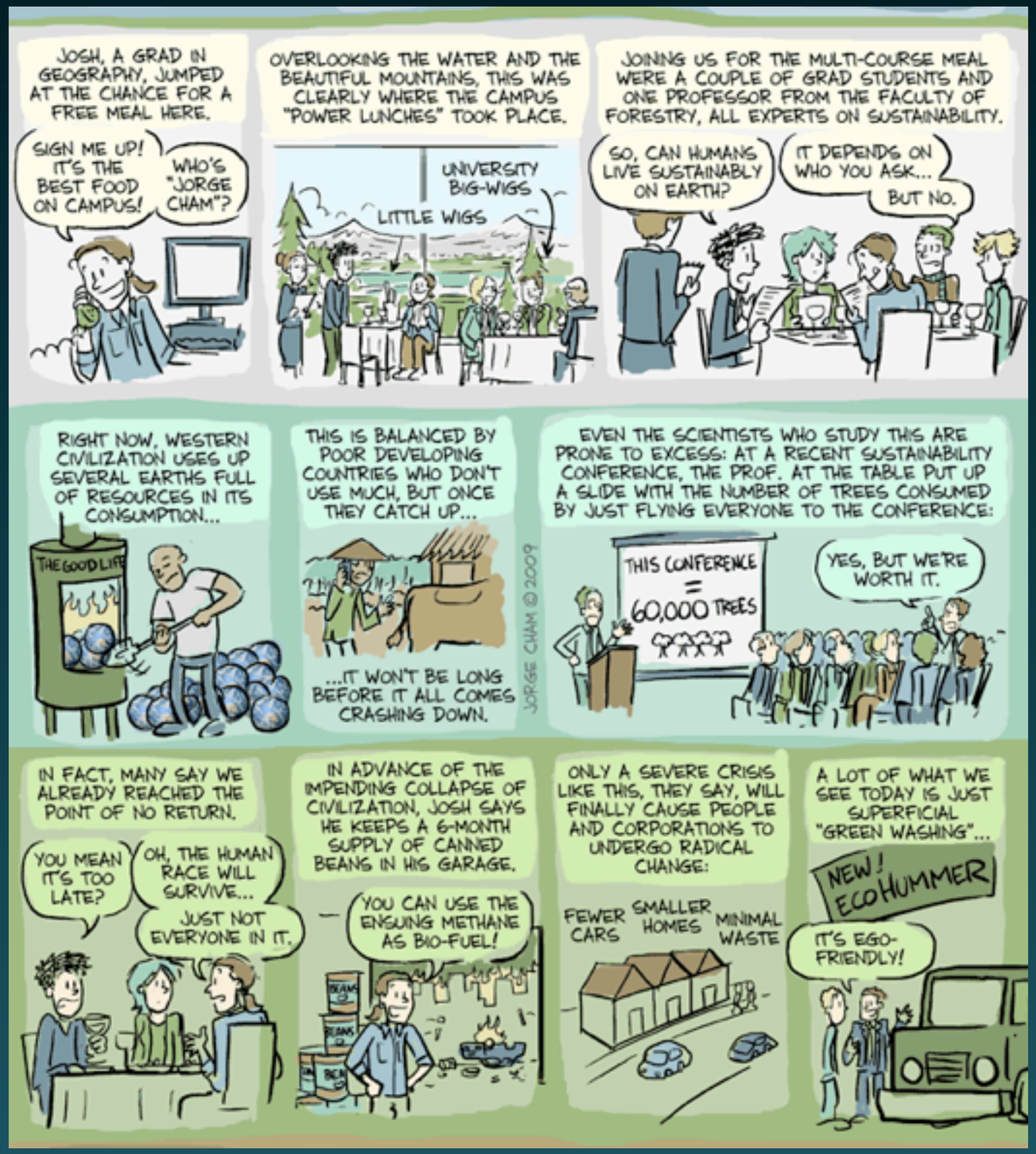
- Dan Olive, Yewon Gim, Hasetha Ganegoda, Shanshan Liu, Carlo Segre - Illinois Institute of Technology
- Yongjian Liu, Bin Hua, Silvia Jurisson, and Baolin Deng, Zhimang Gu - University of Missouri, Columbia
- Don Reed, Jef Lucchini, Marian Borkowski, Mike Richmann, Roland Schulze, Kurt Sickafus, Jason Lashley - Los Alamos National Laboratory
- Jim Tobin, Dario Arena - Livermore National Laboratory
- David Shuh - Lawrence Berkeley National Laboratory
- Meimei Li, Yiren Chen - Argonne National Laboratory

Global Energy Usage



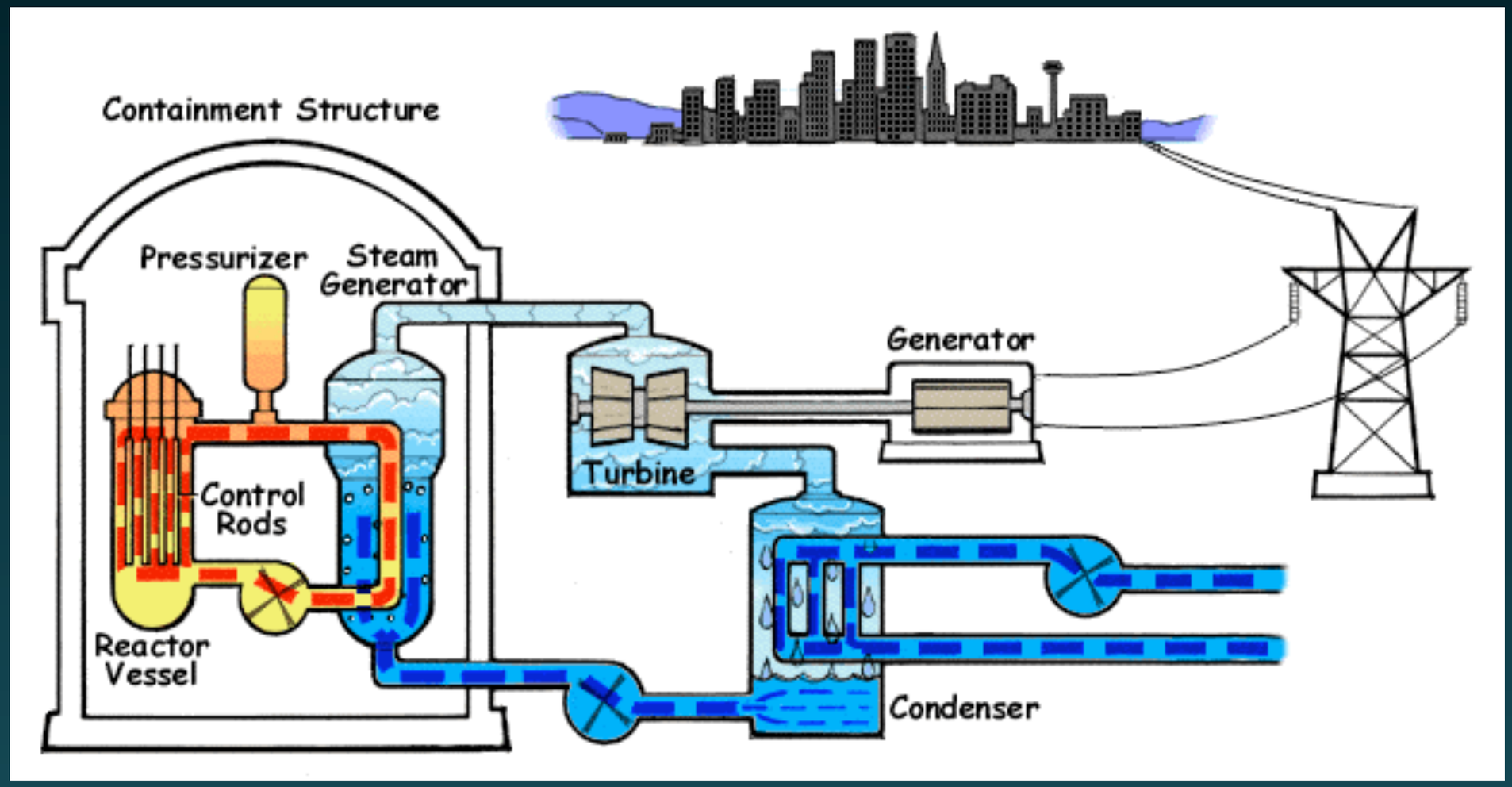
- 15 Terawatts - 2006 BP Survey
- Still Expanding

Sustainability



PhD Comics

Nuclear Power



- Big Part Of The Solution Over The Next 100 Years

Nuclear Issues

- PWR Sustainability
  - Average Age of 439 Operating Nuclear Reactors Is 24 Years
  - Average Age of 119 Shutdown Reactors 22 Years
  - The Bulletin Of The Atomic Scientists 2008 Status Report
- Push Lifetime To 40 Years
  - Aging
  - Stress
  - Corrosion
  - Fatigue
  - Irradiation Effects

Nuclear Issues

- Gen IV
  - Reactor Design
  - Materials Issues
  - Corrosion
  - Neutronics
  - High Temperature
- Fuel
  - Corrosion
  - Pellet Cladding Interaction
  - Fission gas release and swelling
  - Hydrogen induced embrittlement of cladding alloys

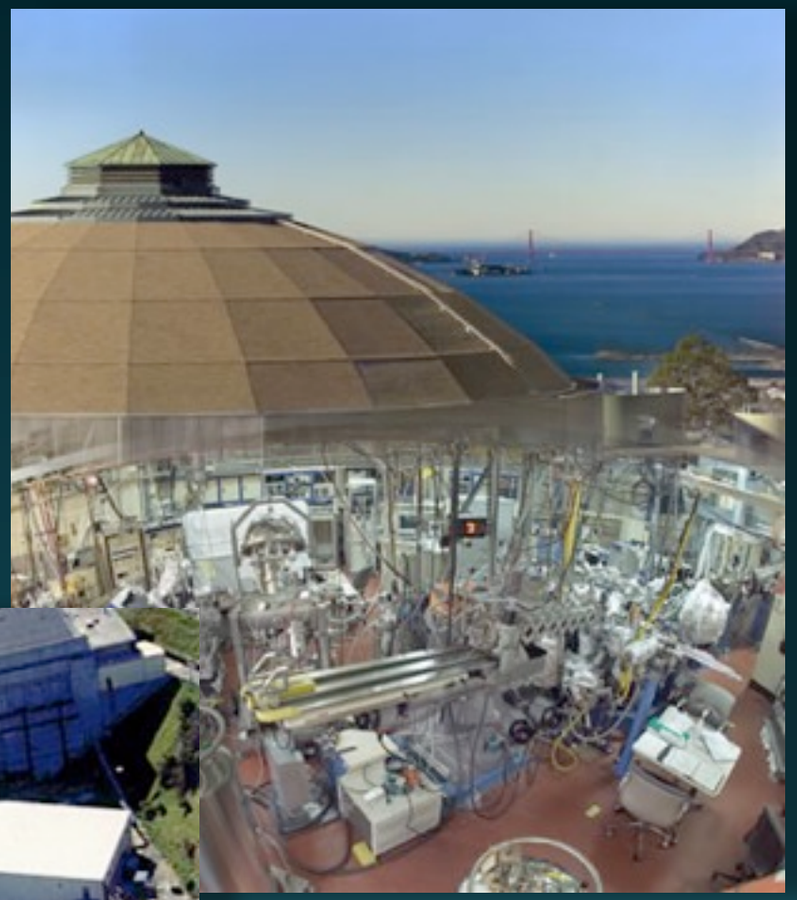
Nuclear Issues

- Nuclear Waste
  - Fate
  - Transport
  - Geochemistry
  - Biochemistry
  - Repository Science
- Fundamental Science
  - 5f Electron Behavior
    - Highly Correlated Electrons
  - 5f Chemistry

Synchrotron Radiation

- In The Late 1960s, Electron Storage Rings Began To Be Used As Intense, Forward Focused Sources Of Radiation
- Since That Time, These Sources Have Been Used To Determine The Properties Of Semiconductors, Proteins, Superconductors, Multilayers, Highly Correlated Electron Materials, And Anything Else That People Have Thought To Put In The Photon Beam
- The Synchrotron Radiation Sources Have Not Been Well Utilized In the Nuclear Engineering Community

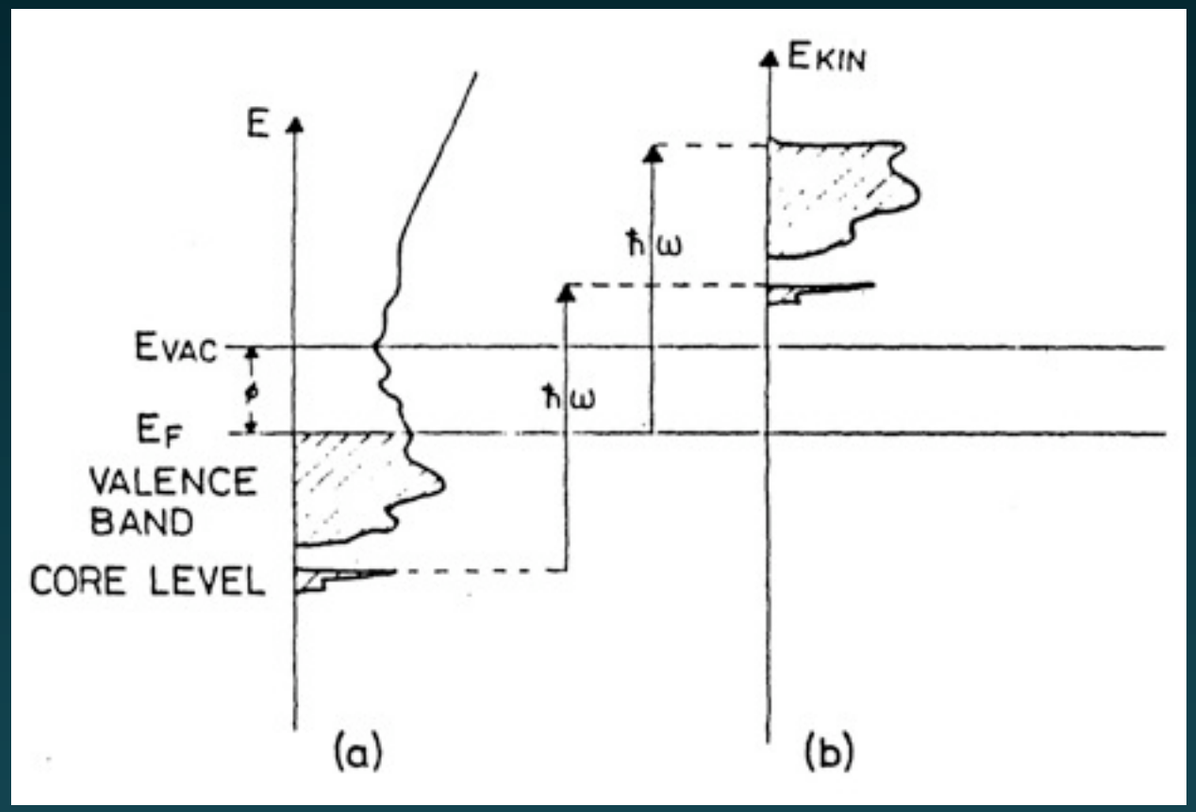
Nuclear Synchrotrons In US

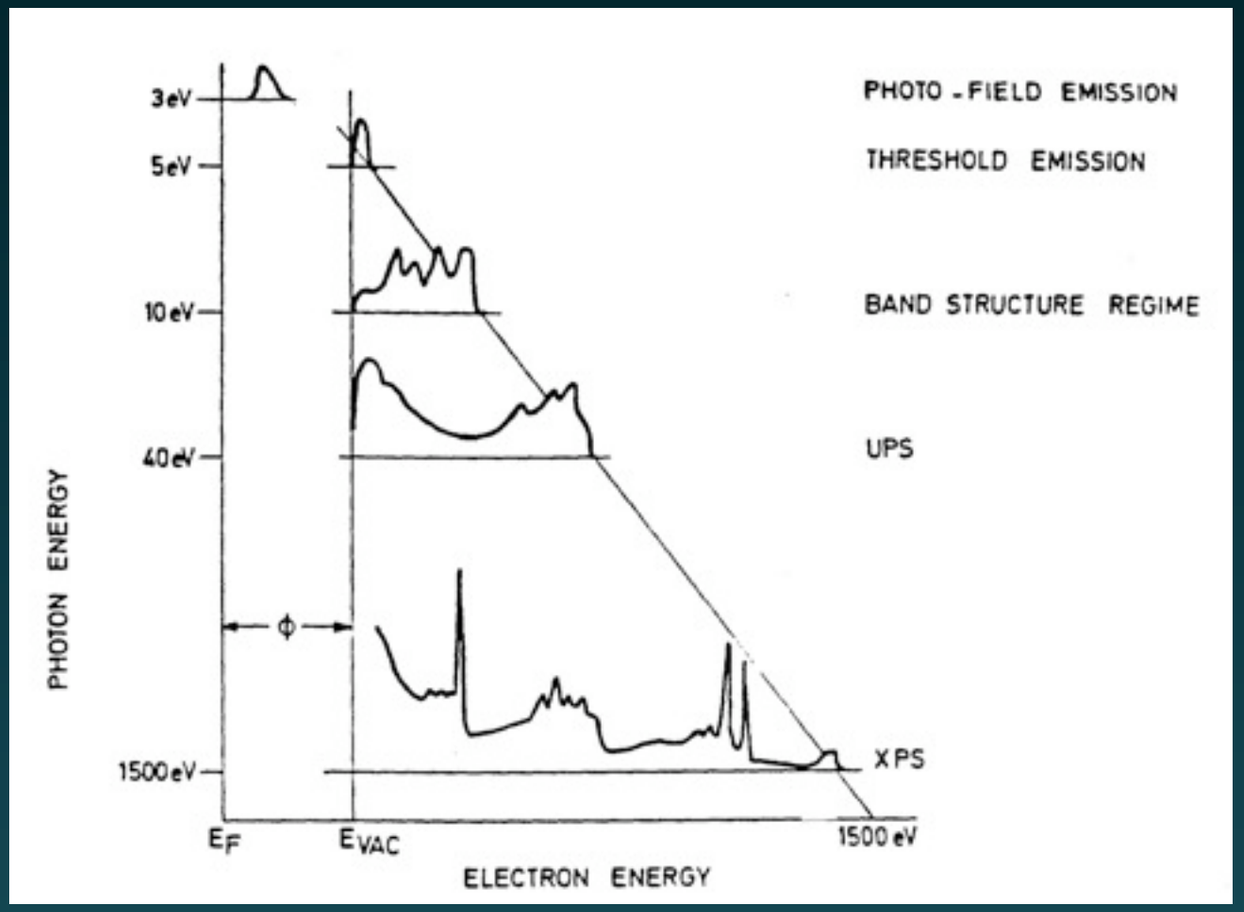


Synchrotron Techniques

- Electronic Structure
  - Photoelectron Spectroscopy
  - X-ray Absorption Near Edge Spectroscopy
- Geometric Structure
  - Extended X-ray Absorption Fine Structure
  - X-ray Scattering

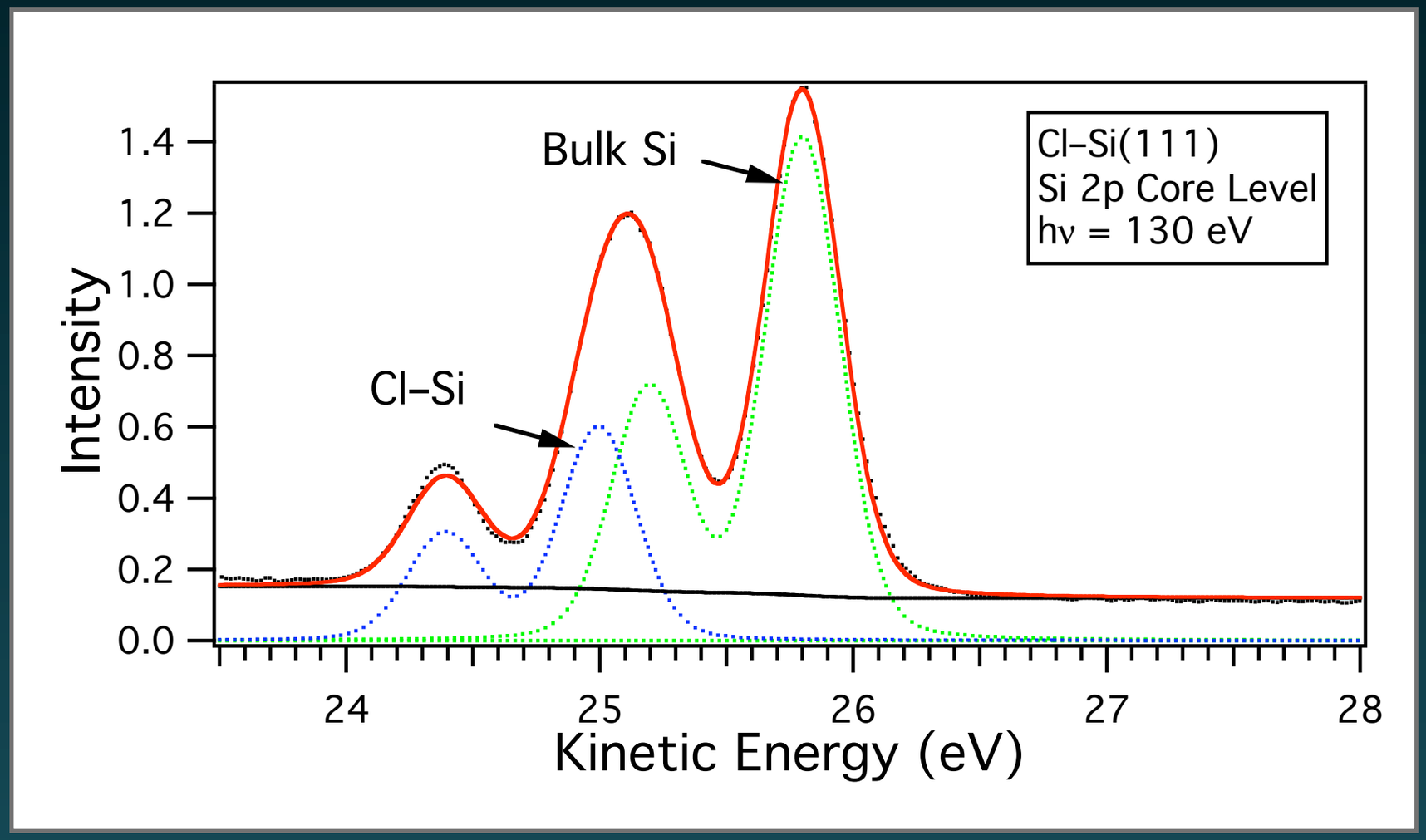
Photoelectron Spectroscopy





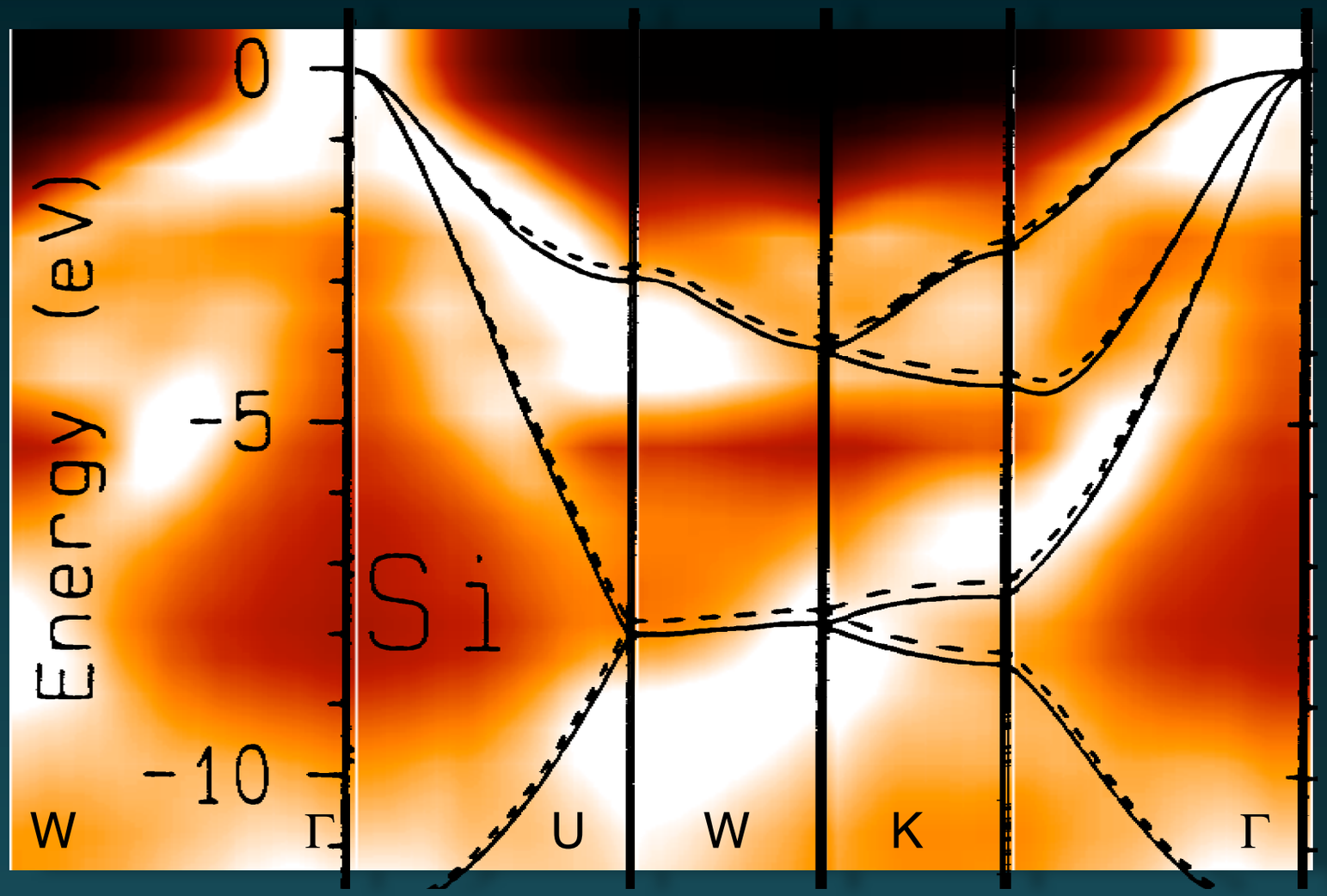
- Electronic Structure
  - Core Level Spectroscopy
  - Valence Band Spectroscopy

Photoelectron Spectroscopy



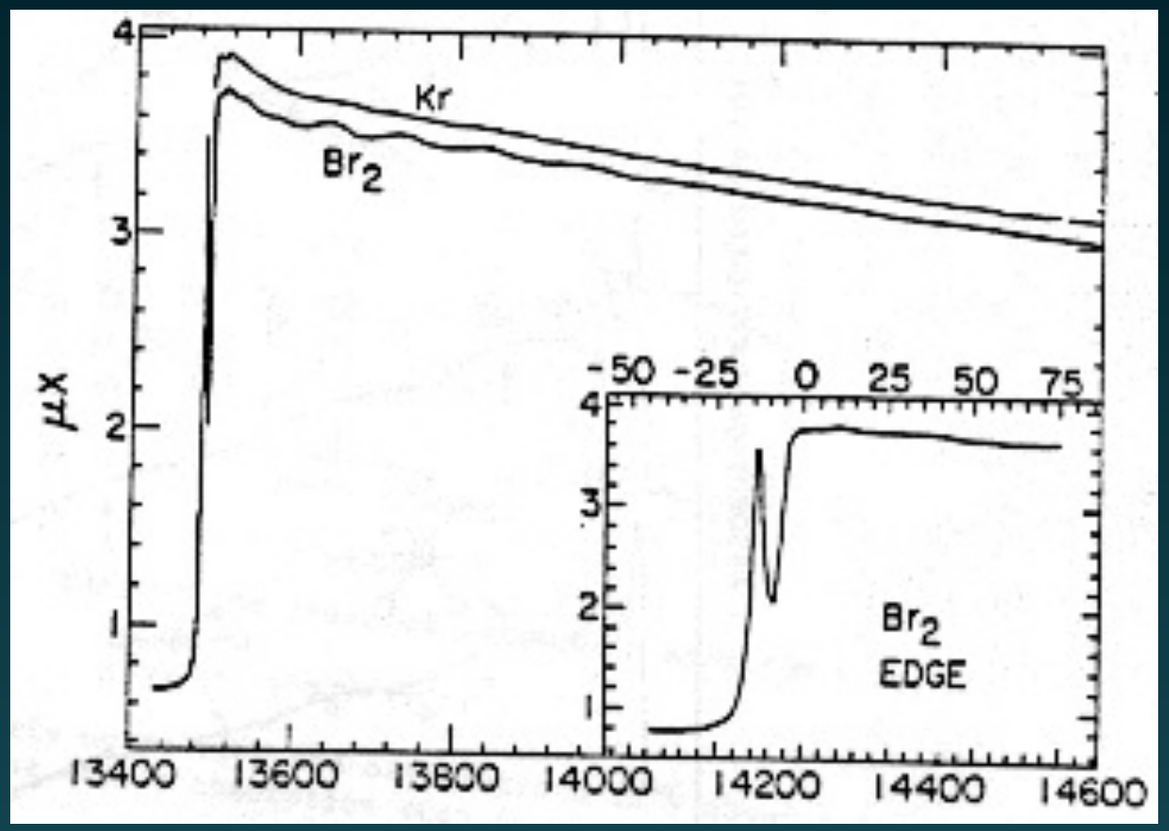
- Core Level Spectroscopy
  - Oxidation State
  - Charge Transfer

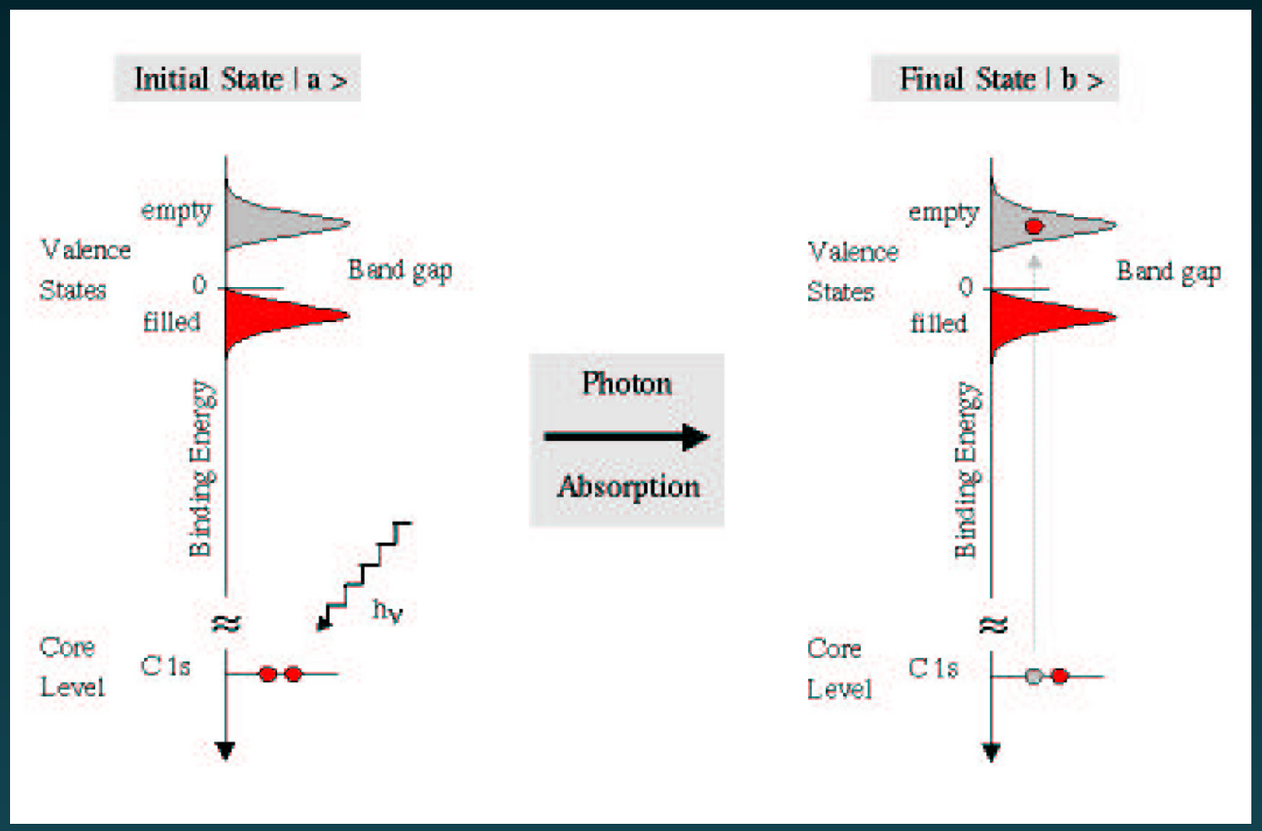
Photoelectron Spectroscopy



- Valence Band Spectroscopy
  - Band Structure
  - Bonding Electrons

X-ray Absorption

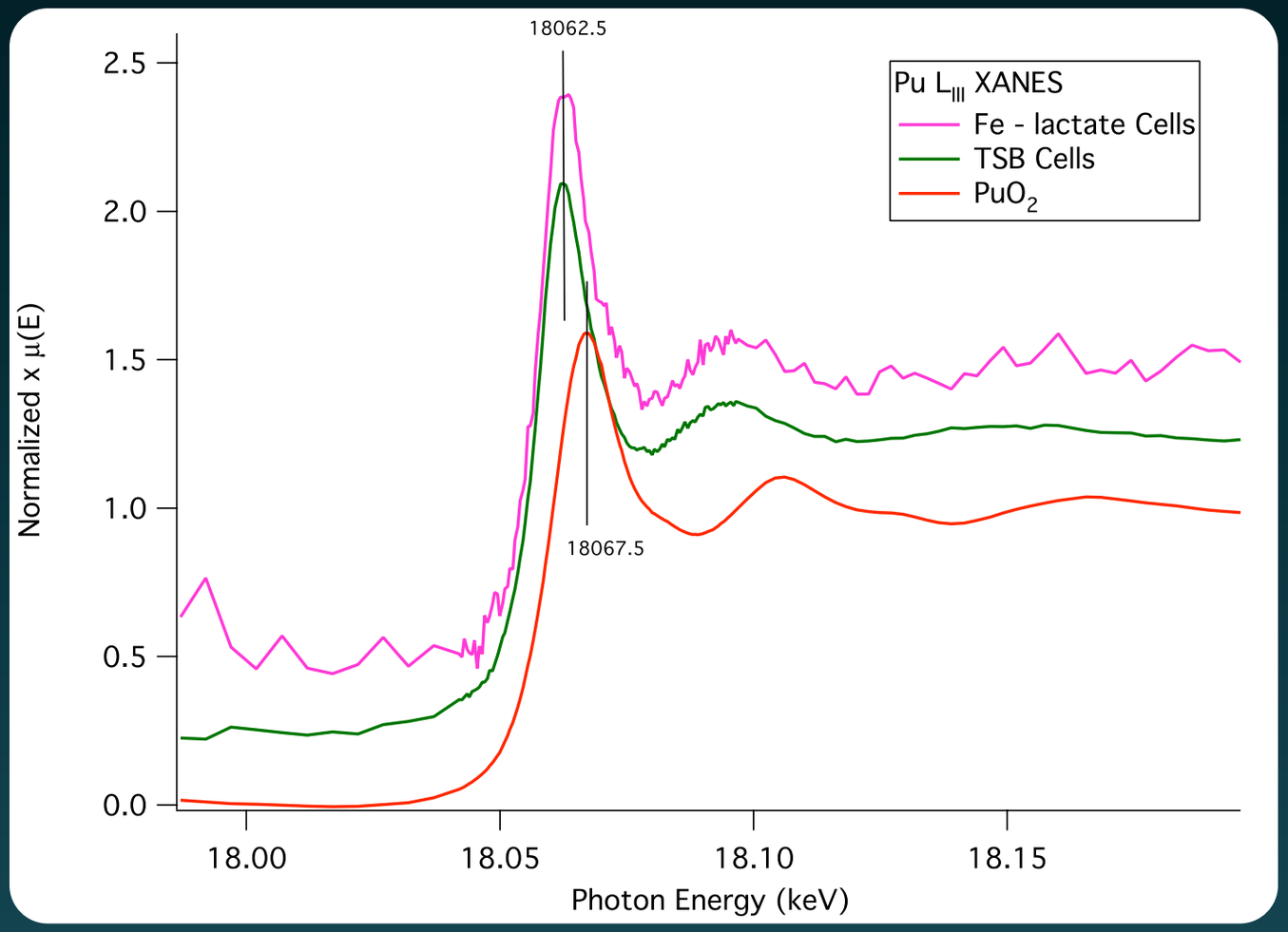




J. Stohr NEXAFS Spectroscopy

- Electronic Structure
  - XANES
  - Overlapping Electronics States
    - Edge Position

X-ray Absorption



- Electronic Structure
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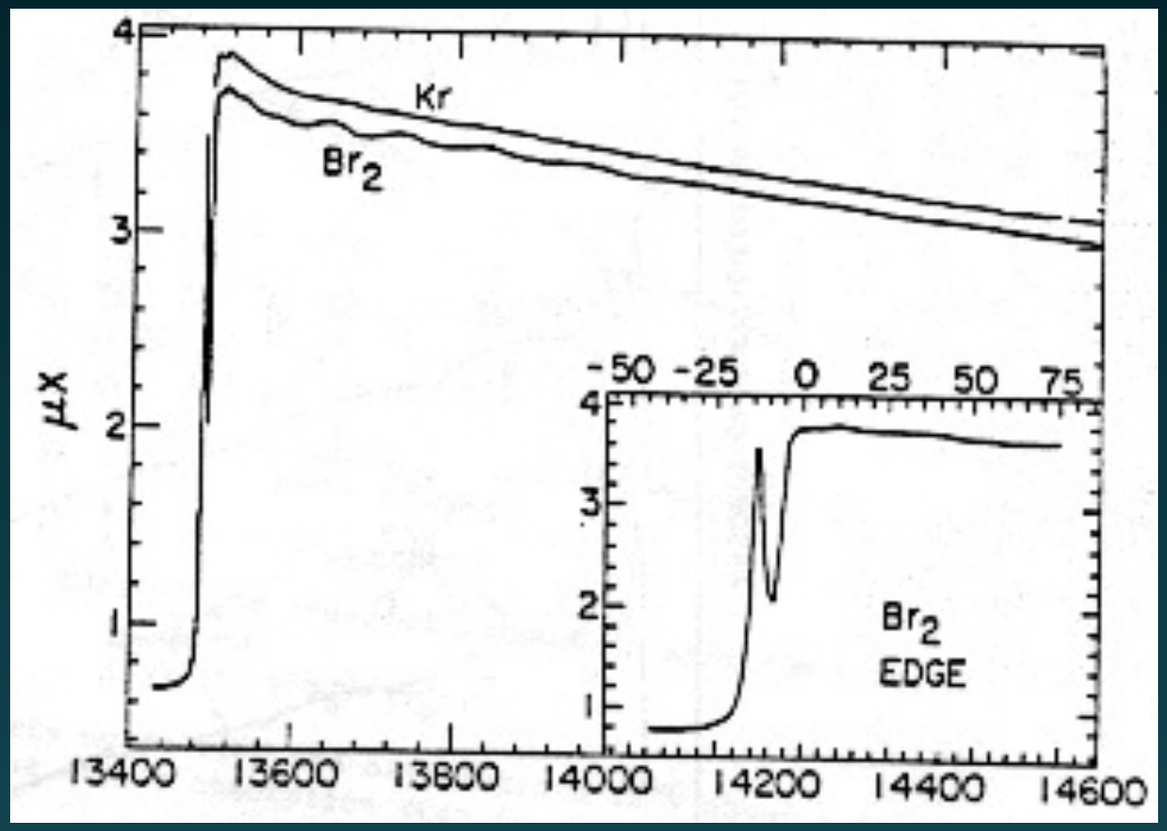
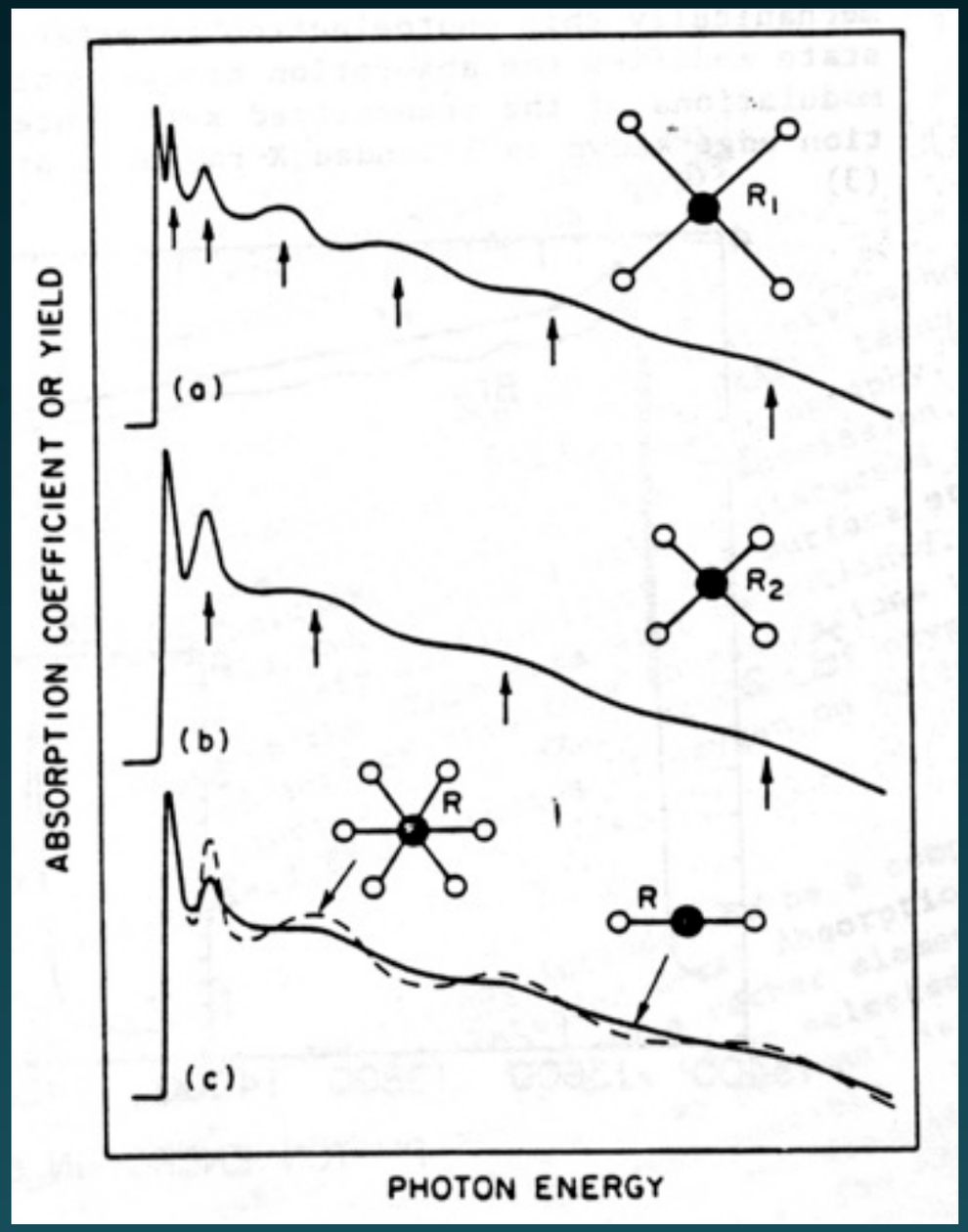
X-ray Absorption

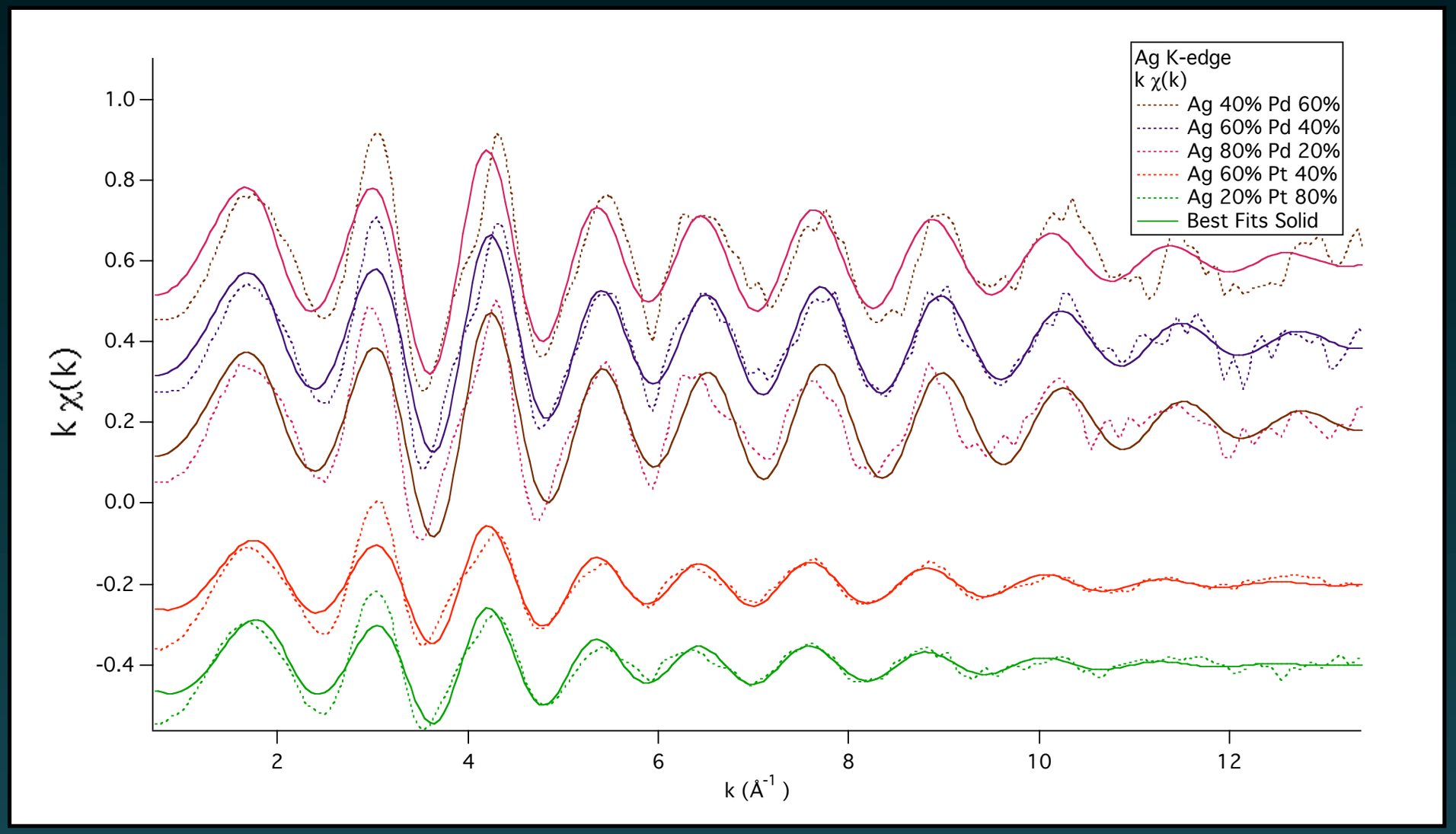


J. Stohr
NEXAFS
Spectroscopy



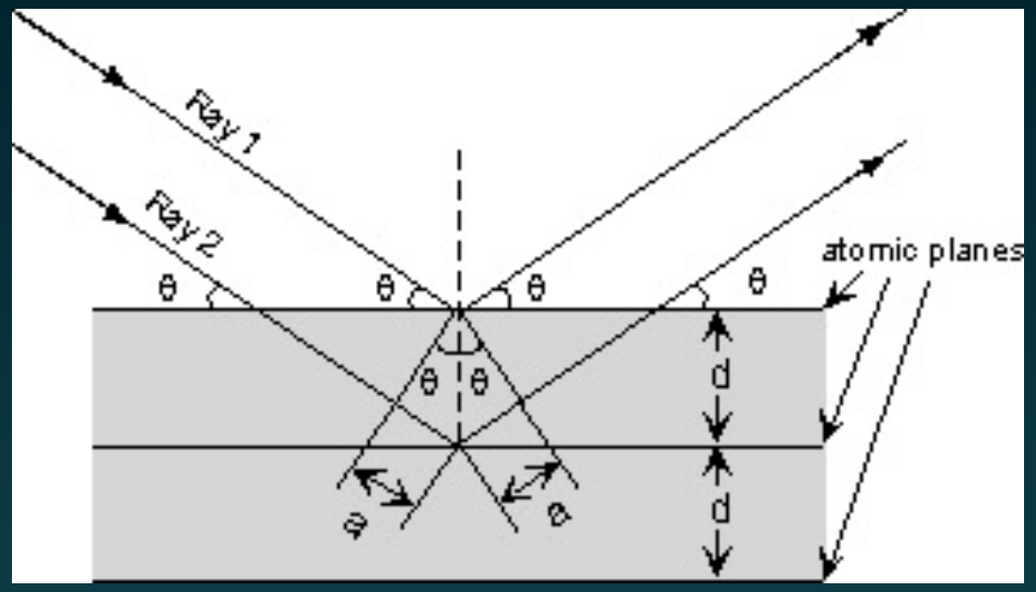
- Geometric Structure
  - EXAFS
  - Interference Between Emitted And Scattered Electron Waves

X-ray Absorption



- Geometric Structure
  - EXAFS
  - Interference Between Emitted And Scattered Electron Waves

X-ray Scattering



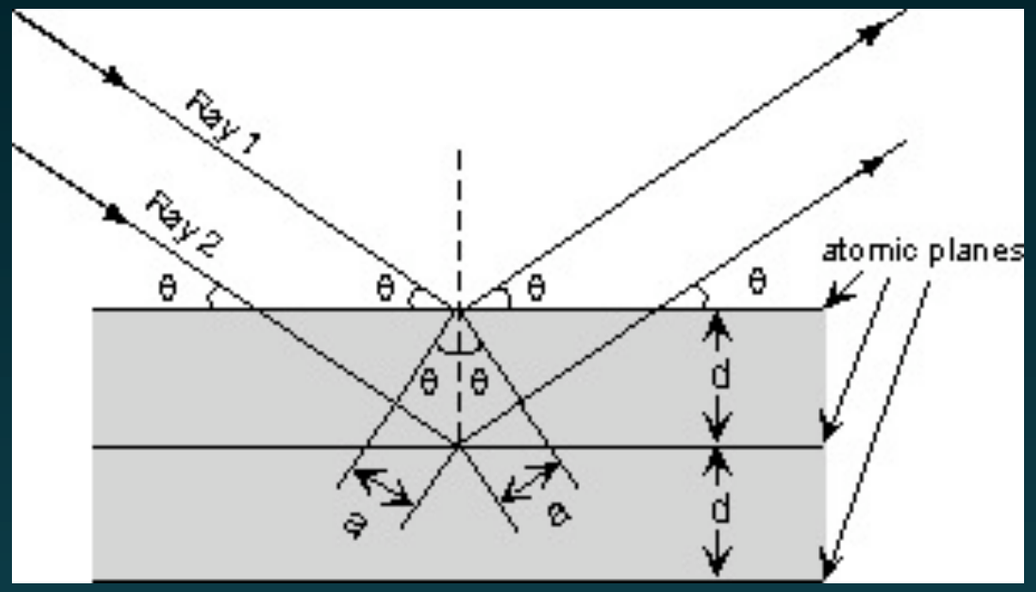
Extra distance travelled by Ray 2 is:

$$2a = 2d\sin\theta$$

- Geometric Structure
  - Atomic Positions
  - Strain

$$n\lambda = 2d\sin\theta$$

X-ray Scattering

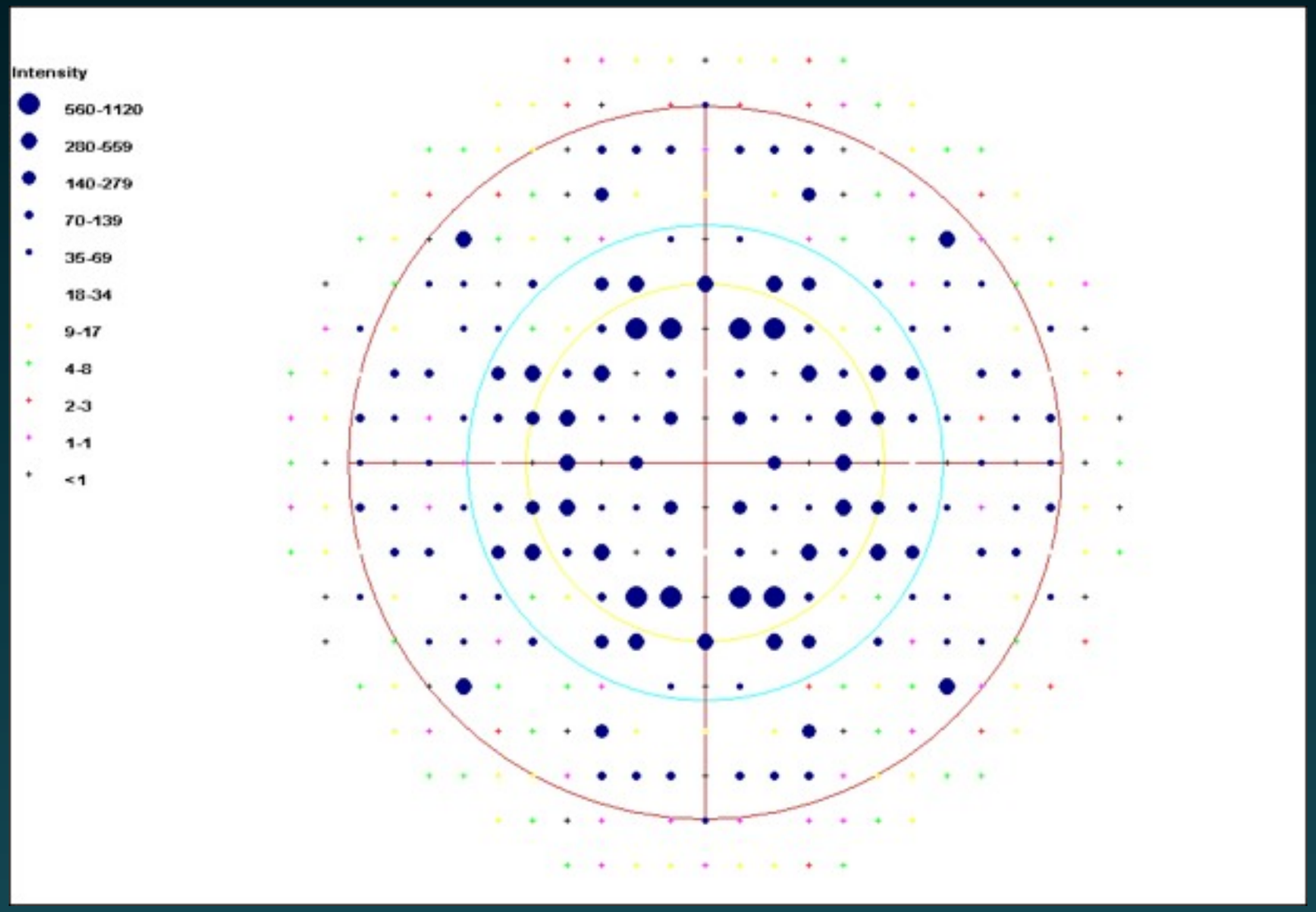


Constructive Interference Occurs When:

$$n\lambda = 2d\sin\theta$$

- Geometric Structure
  - Atomic Positions
  - Strain

X-ray Scattering

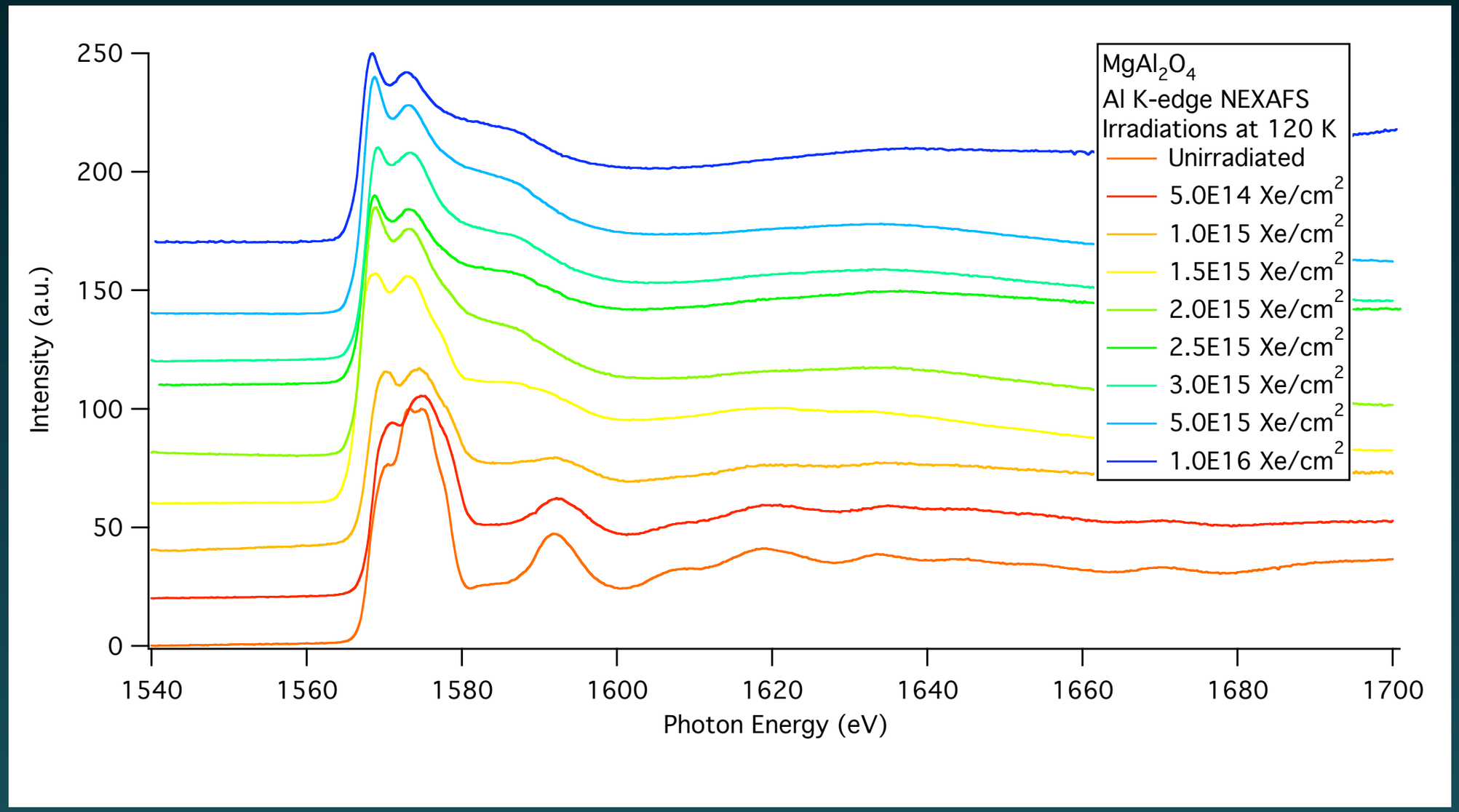


- Geometric Structure
  - Atomic Positions
  - Strain

Radiation Damage

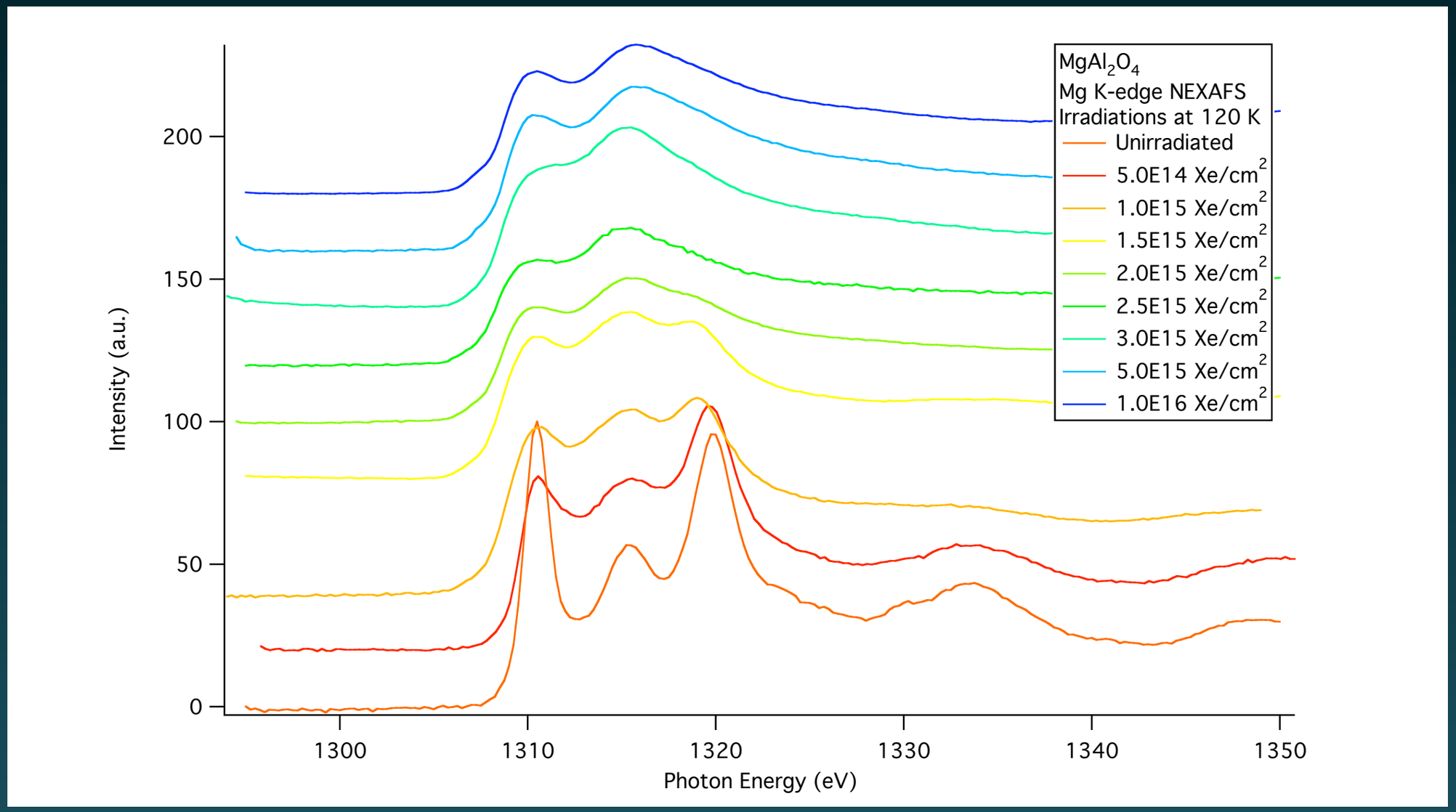
- Spinel - $MgAl_2O_4$
- MOX Fuel
- Cladding
- Structural Coating
- Xe Irradiation
- X-ray Absorption Spectroscopy

Spinel



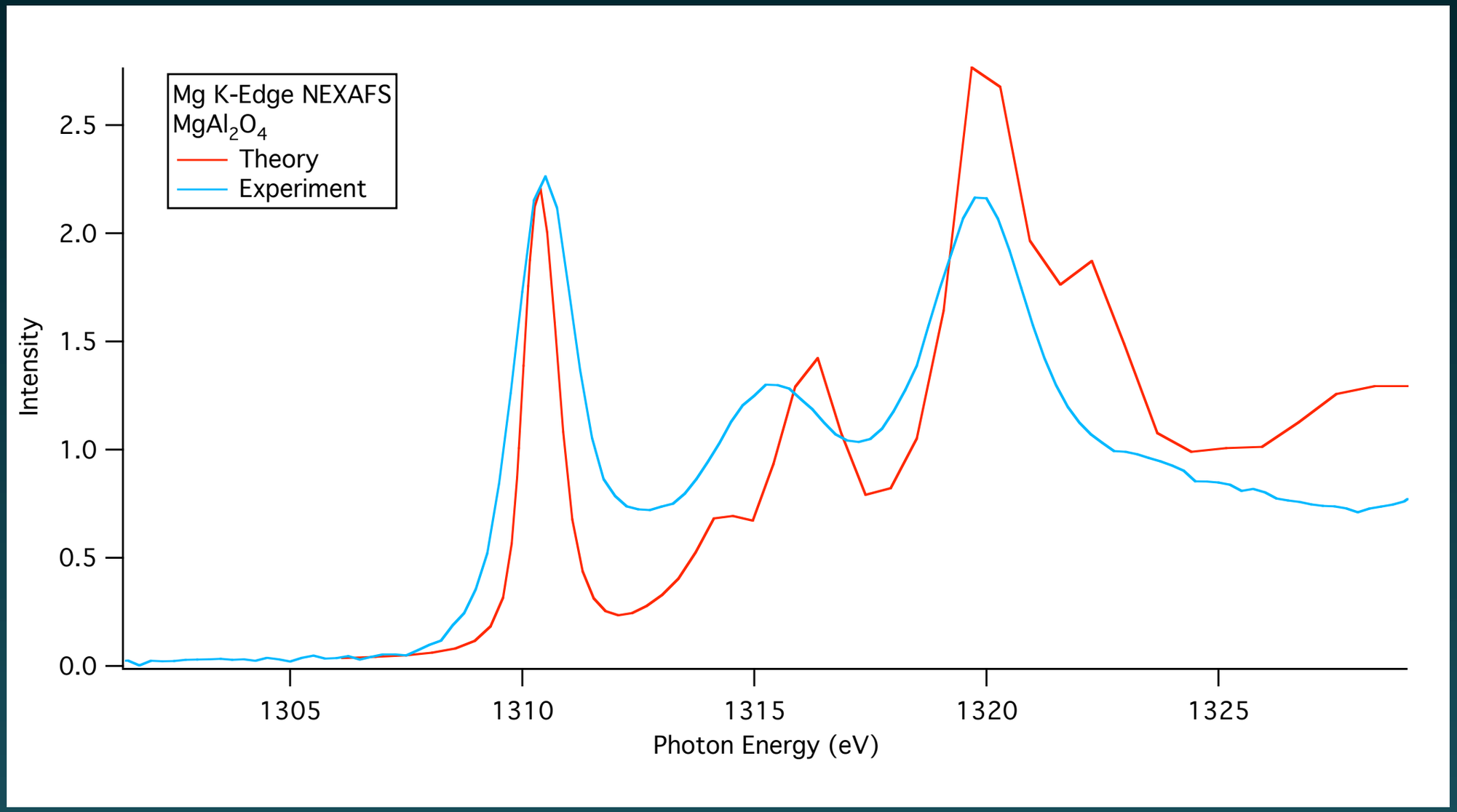
- Al K-Edge

Spinel



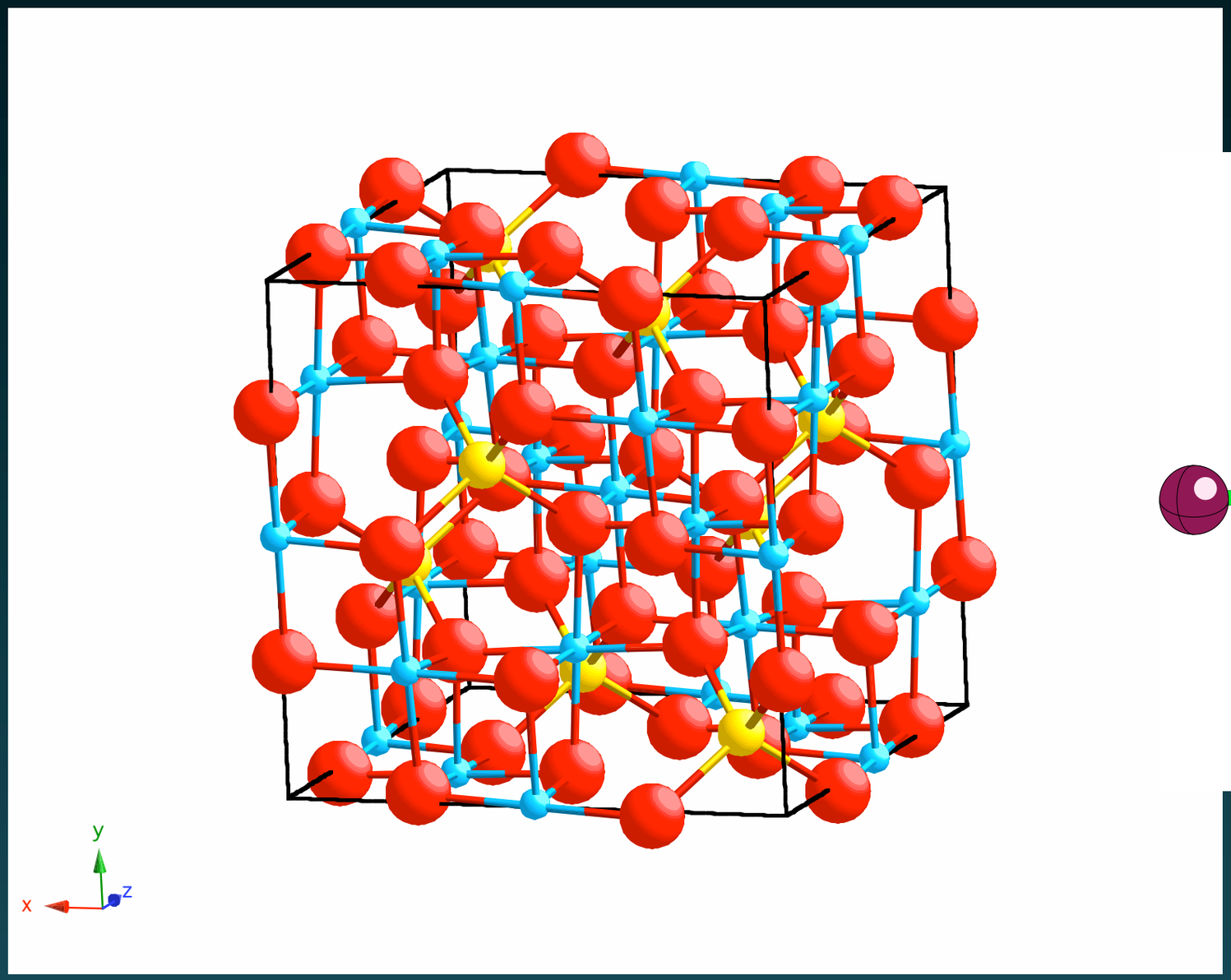
- Mg K-Edge

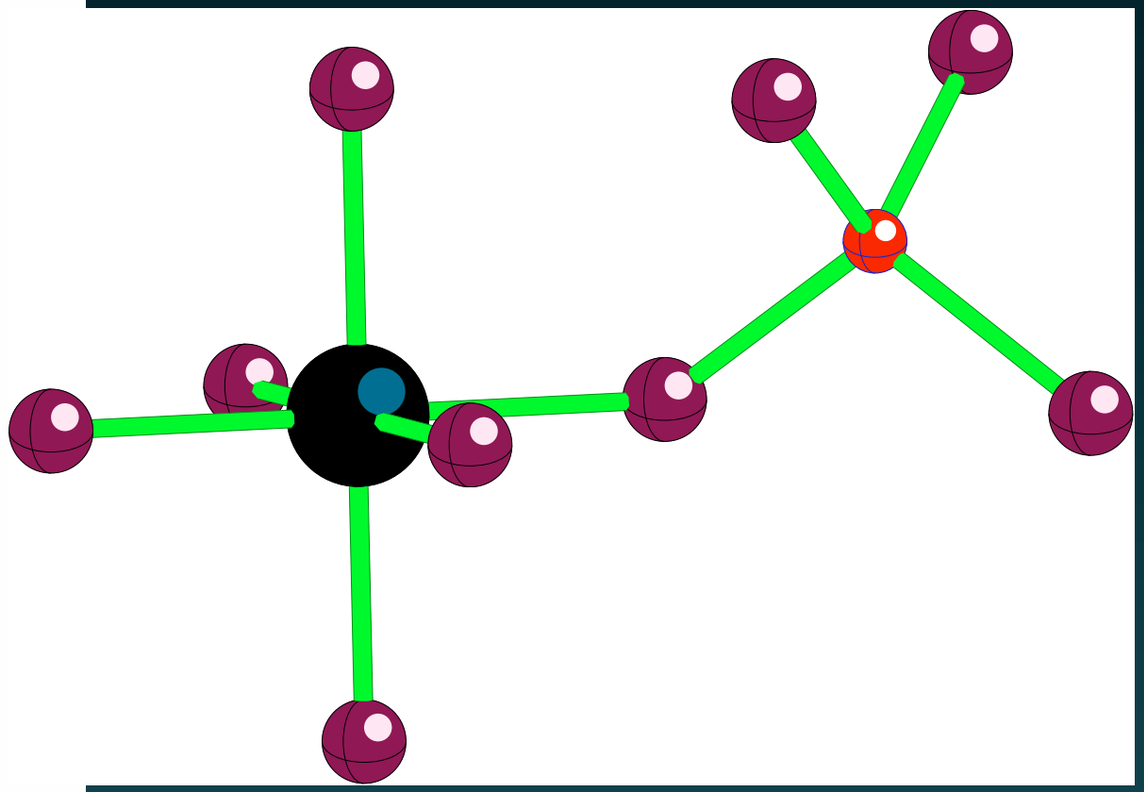
Spinel



- Theoretical Modeling

Spinel





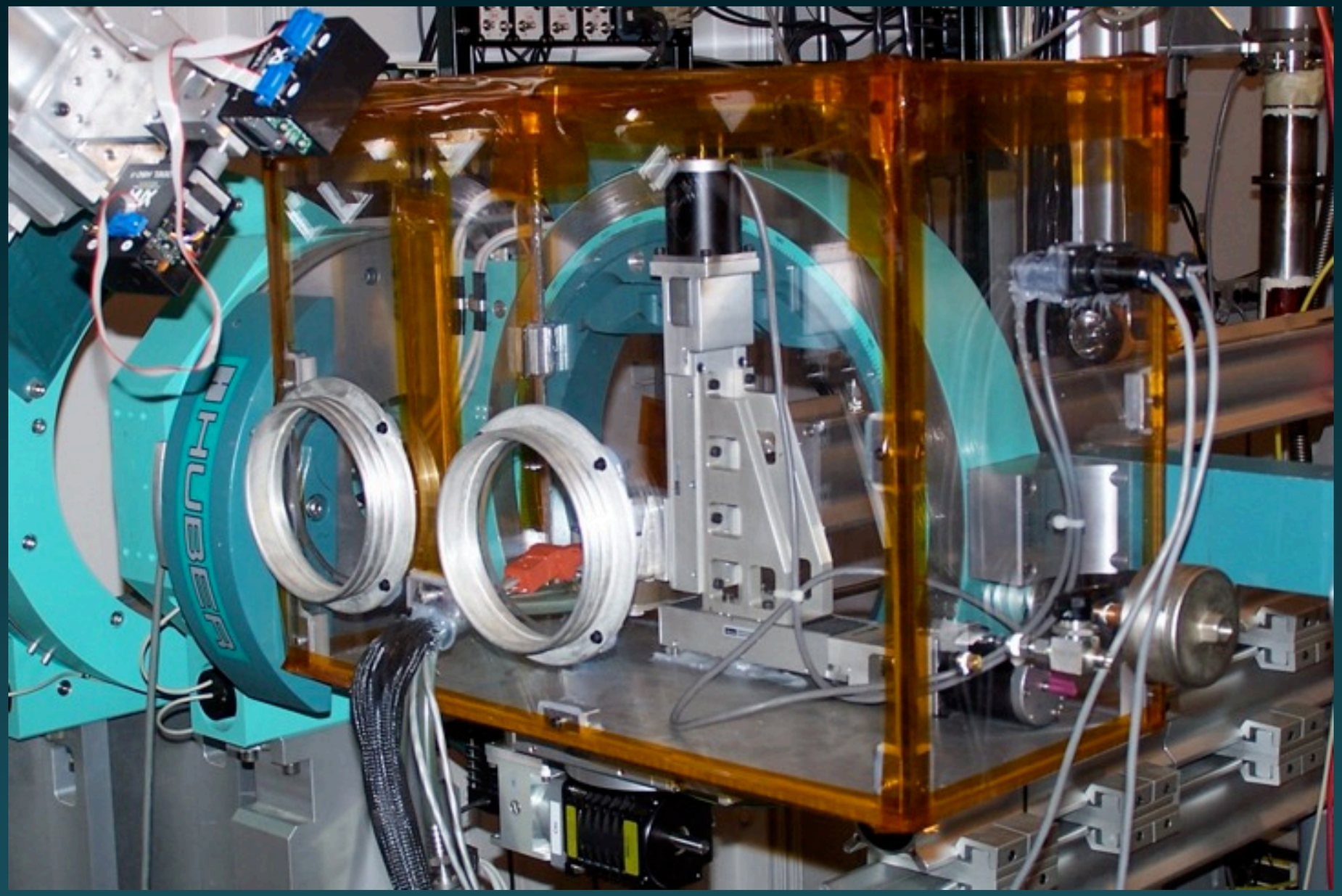
- Octahedral Al
- Tetrahedral Mg
- Site Swapping

- Al - O 1.92 Å
- Mg - O 1.93 Å

Xe Irradiation

- Small Penetration Depth
- Really Hard To Study
- Why Not Use Neutrons?
- Radioactive Samples Are Hard To Handle At Synchrotrons

Graphite Irradiation



- X-ray Glove Box

Graphite Irradiation

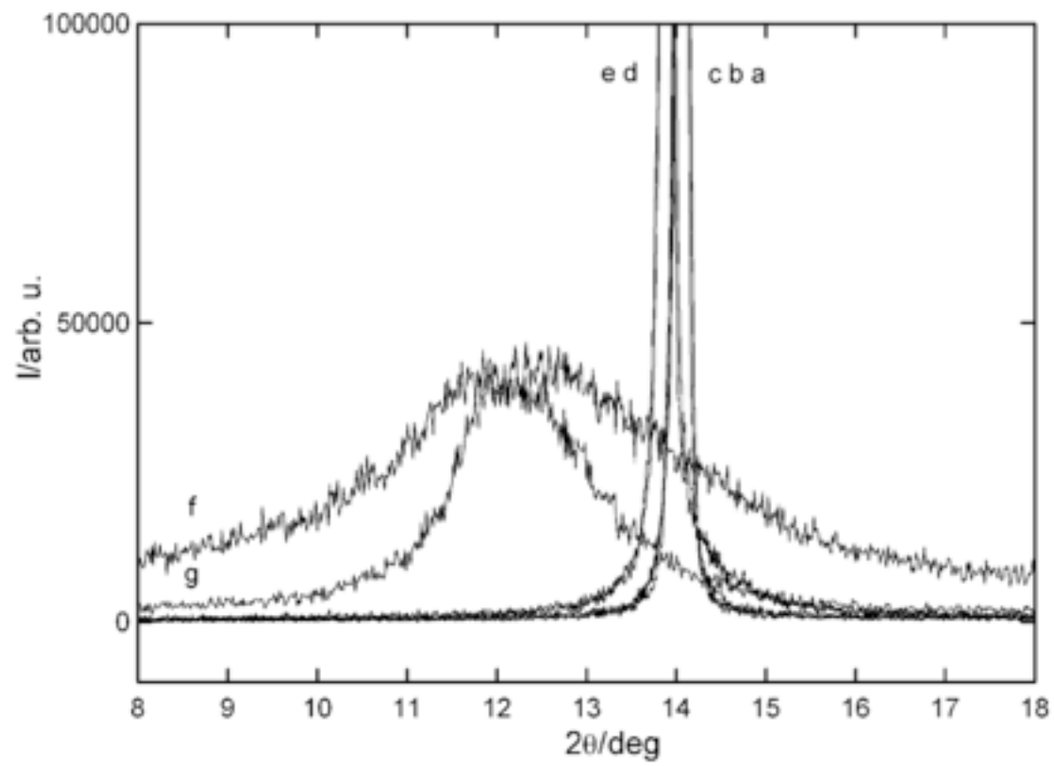


Fig. 4. Section of X-ray diffraction patterns at 25 °C in the vicinity of the 002 graphite diffraction peak.

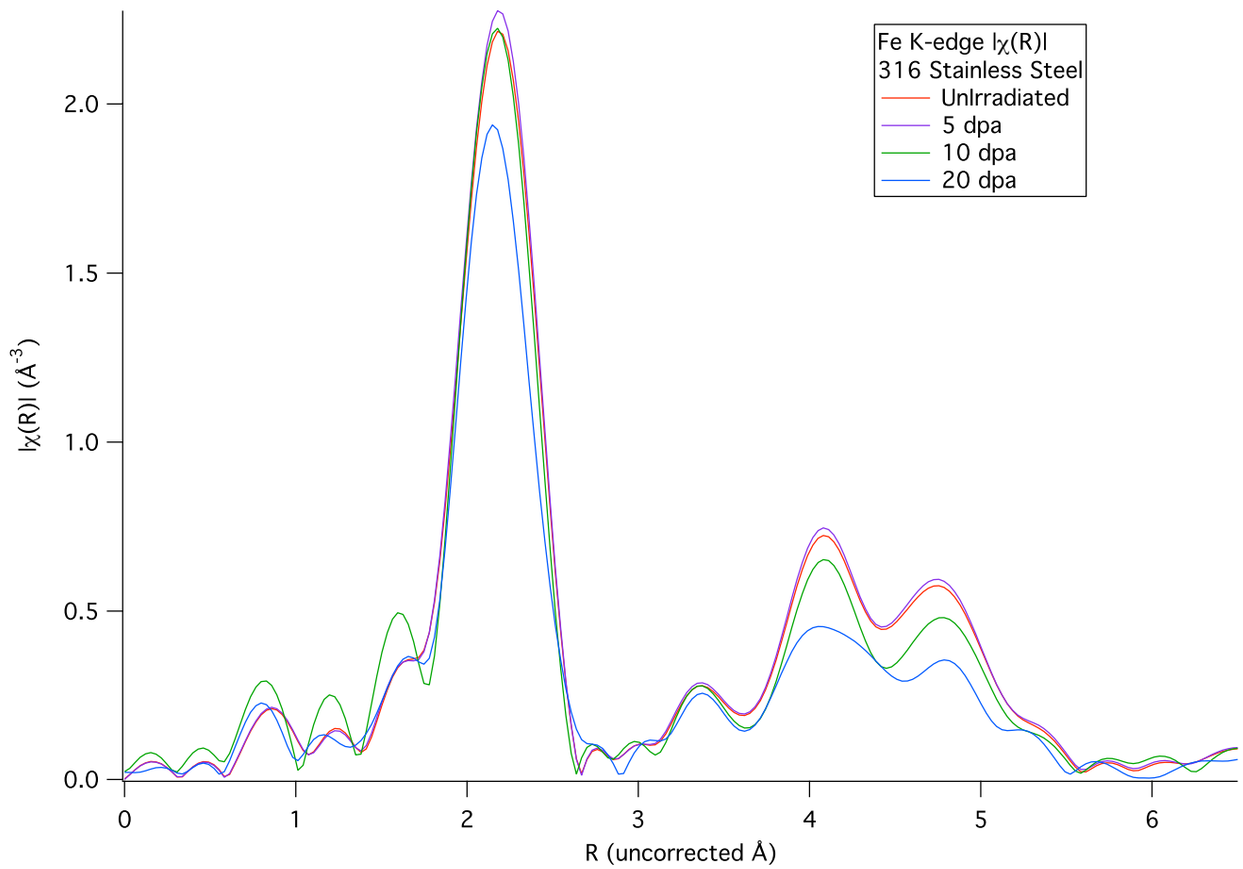
Lexa Journal of Nuclear Materials 348 (2006) 122–132

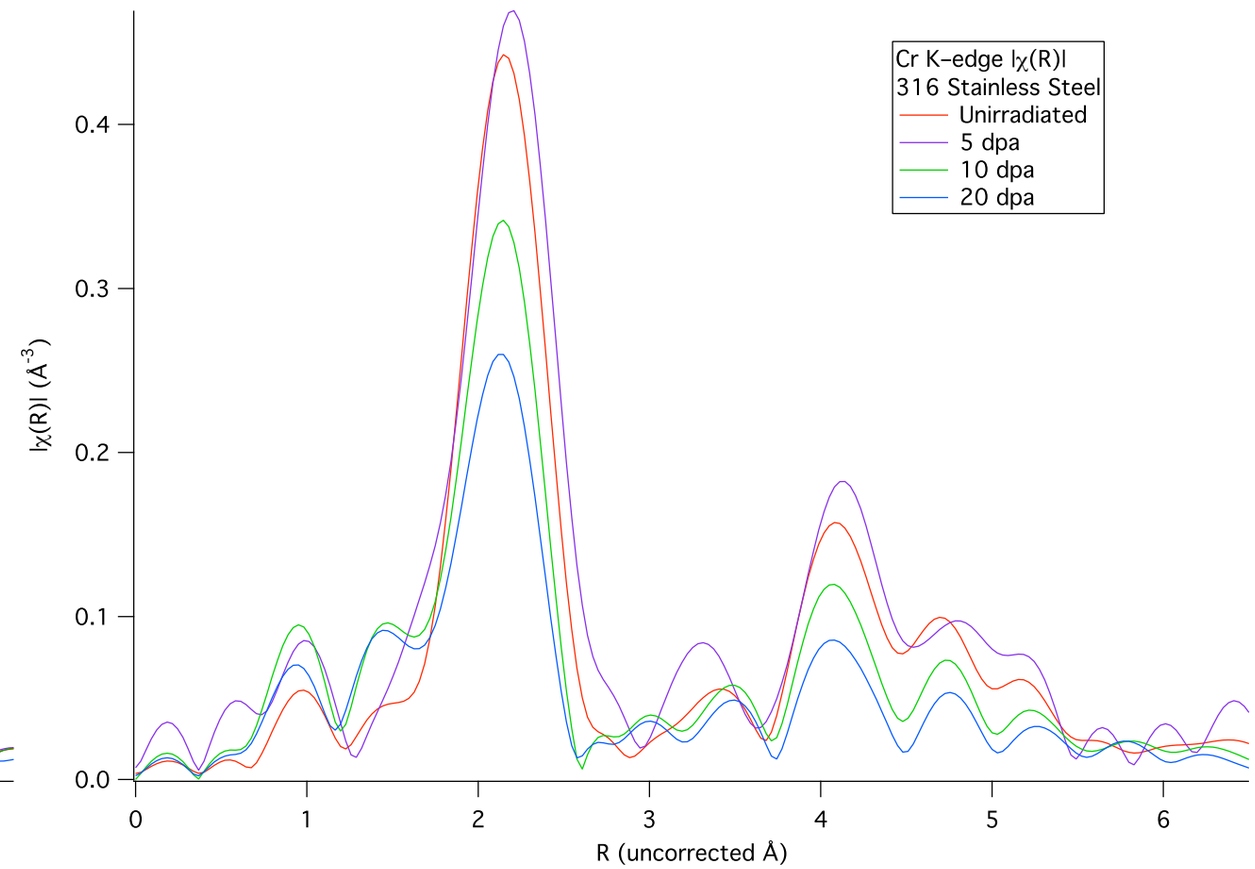
- Graphite From ASTRA Core
- Amorphous At ~5x10^{19} n/cm^{2}

NRR CAT Test

- X-ray Absorption
- X-Ray Diffraction
- Sample Handling
- Detector Performance
- Safety Protocols

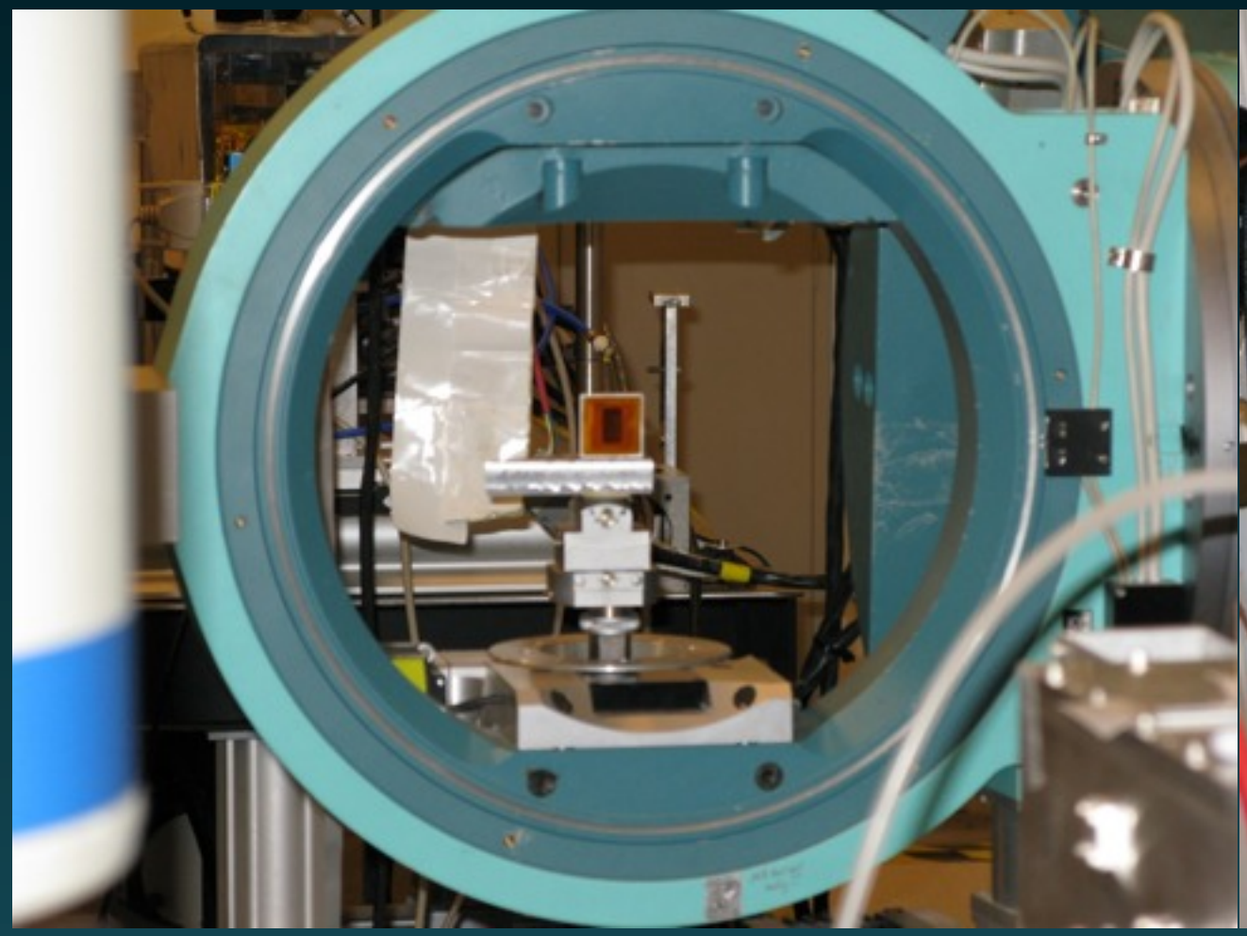
NRR CAT Irradiated Samples

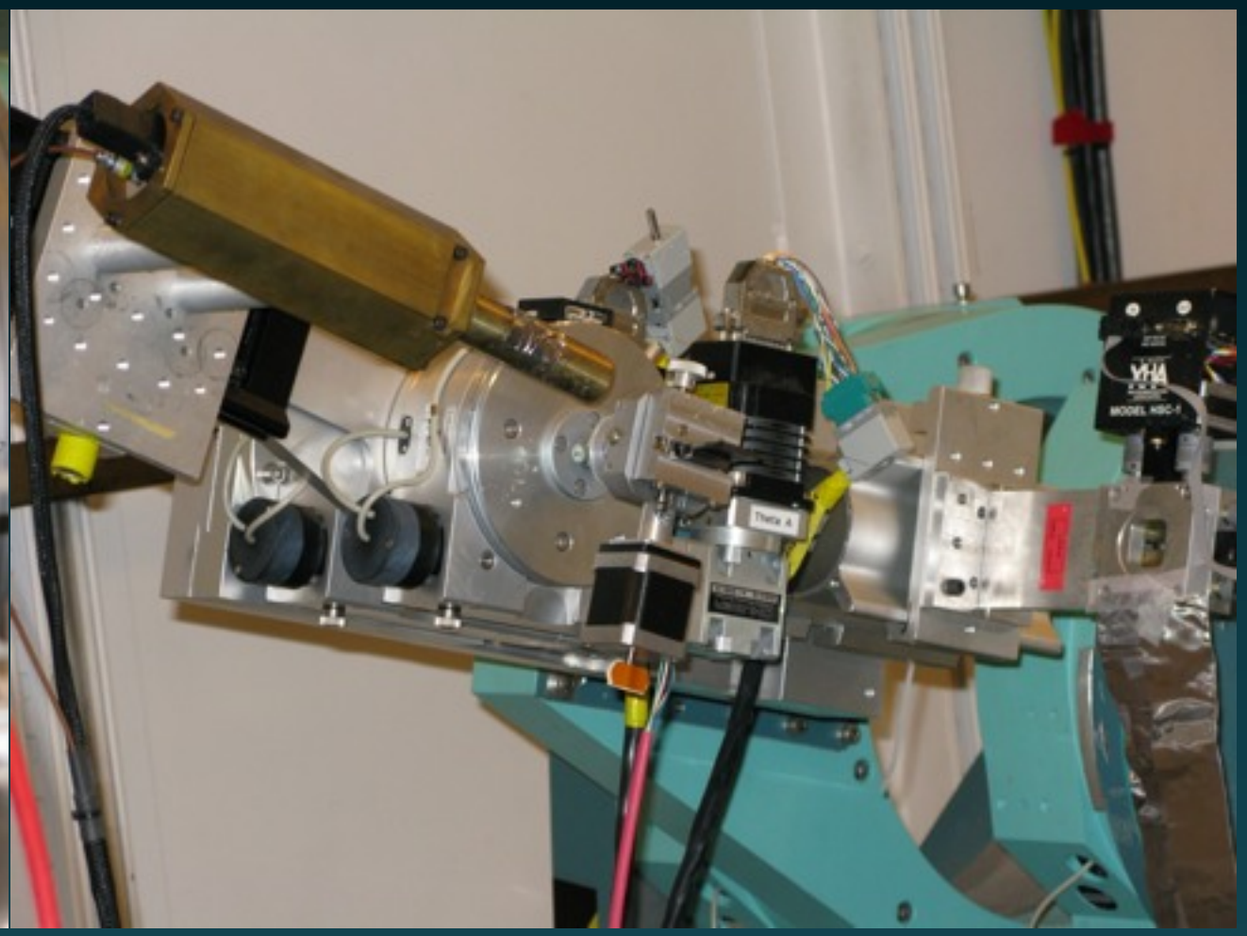




- SS 316 Irradiated In Reactor
- Fe data: near-neighbors are not Lost at doses to 10 dpa.
- Cr data: near neighbors Lost at lower doses

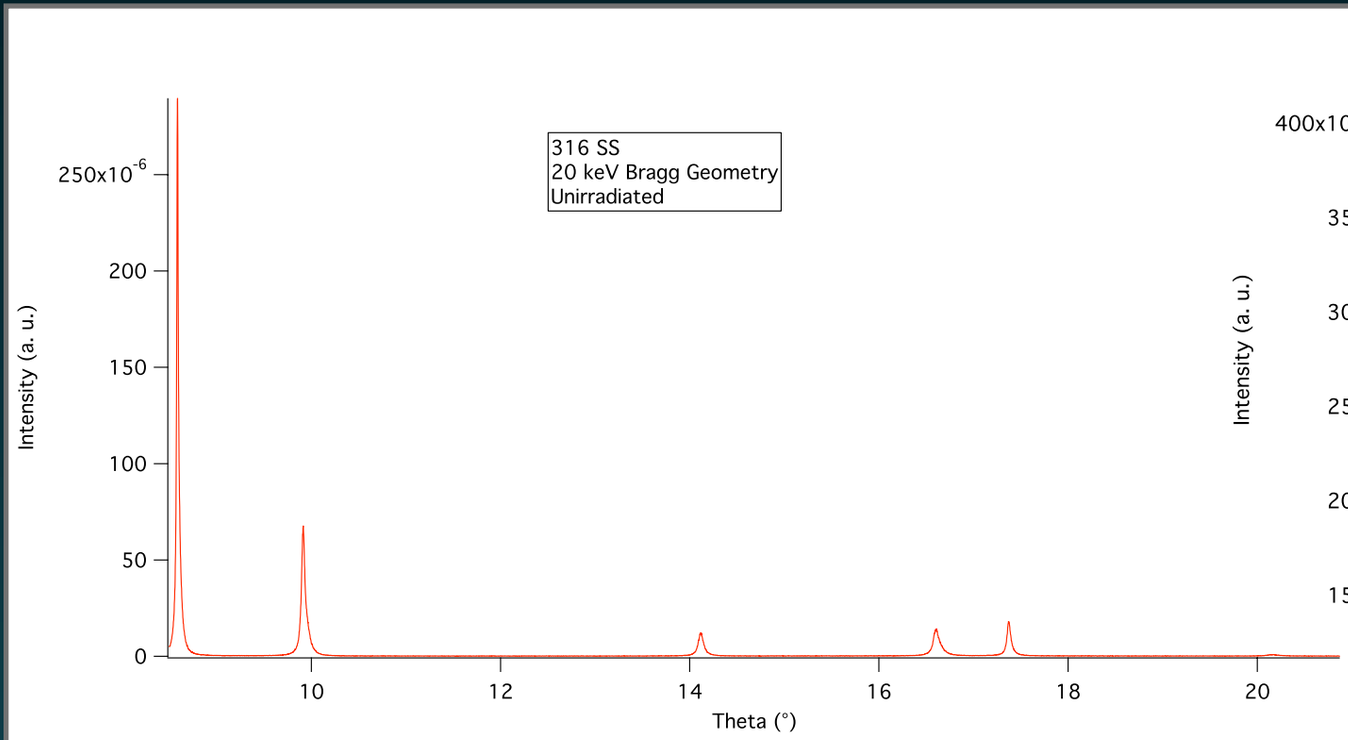
NRR CAT Diffraction

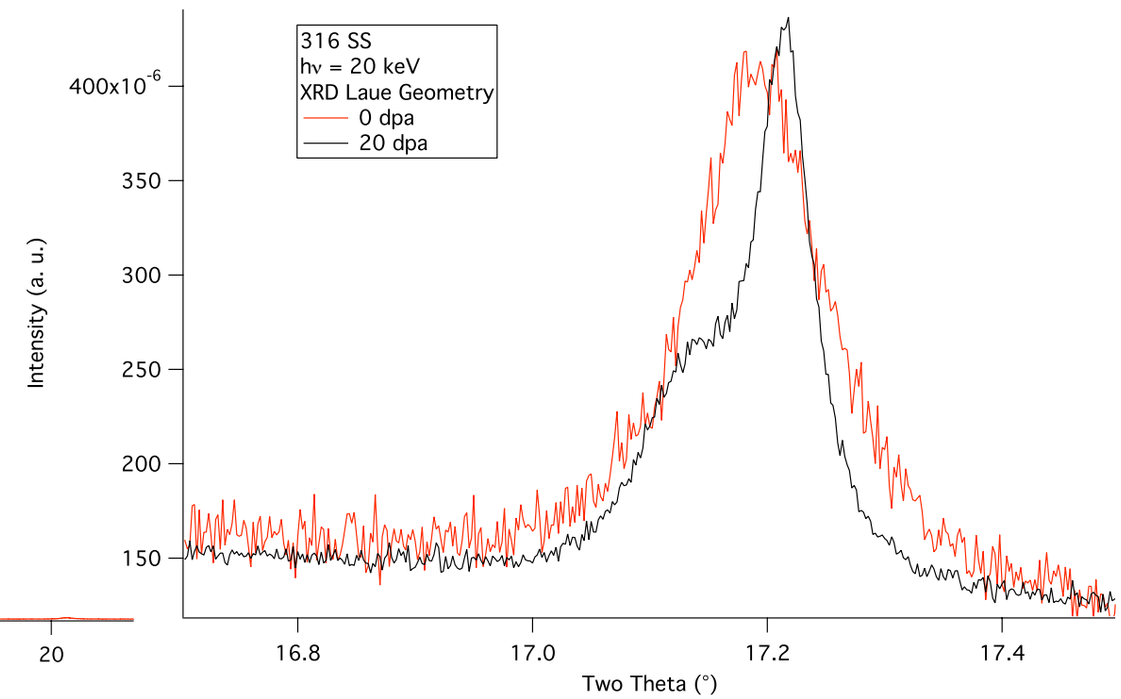




- Triple Containment Cell
- 0.2 mCi
- Analyzer Crystal

NRR CAT Diffraction

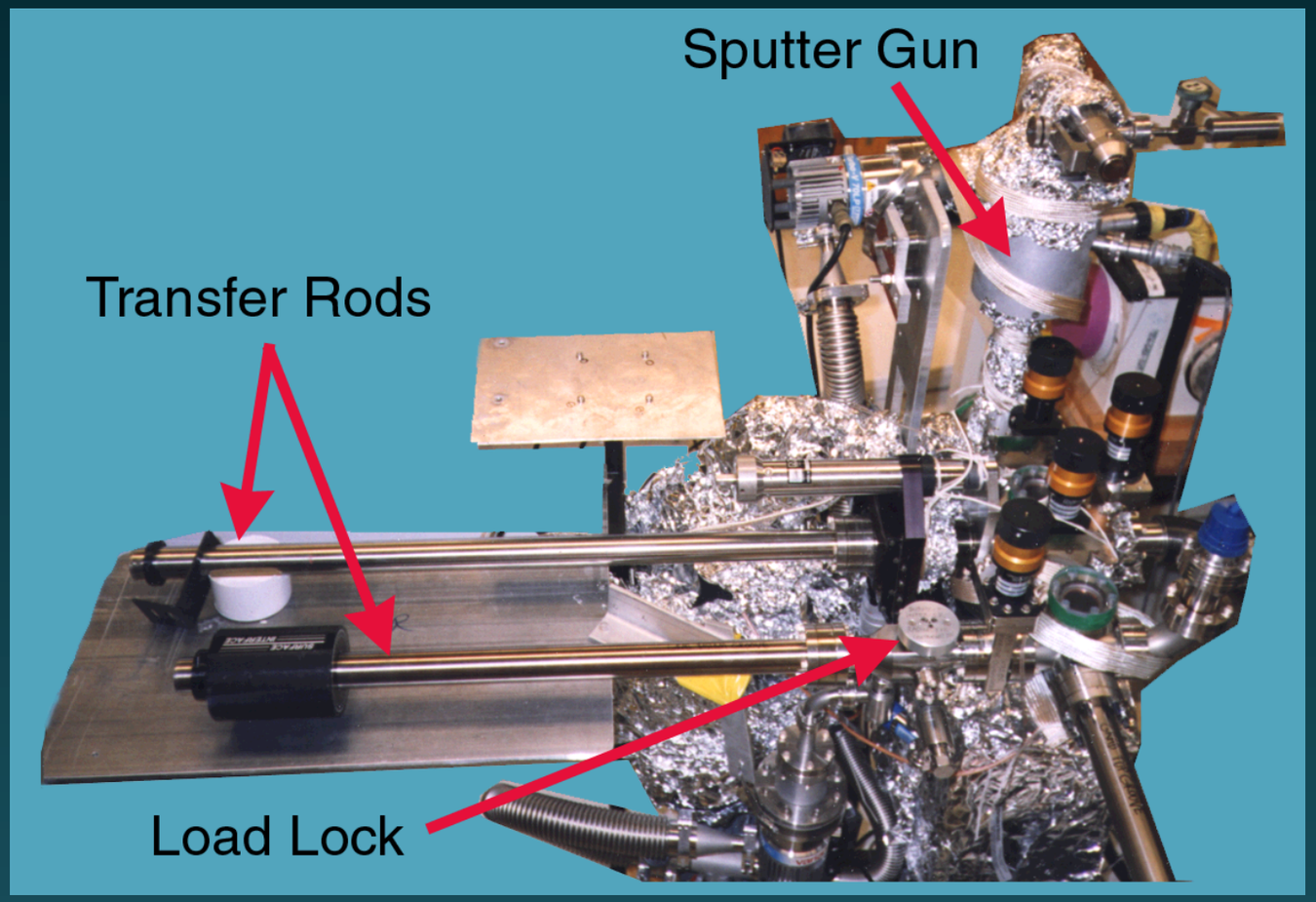




- Triple Containment
- Laue Or Bragg
- 20 keV
- New Results No Idea What It Means

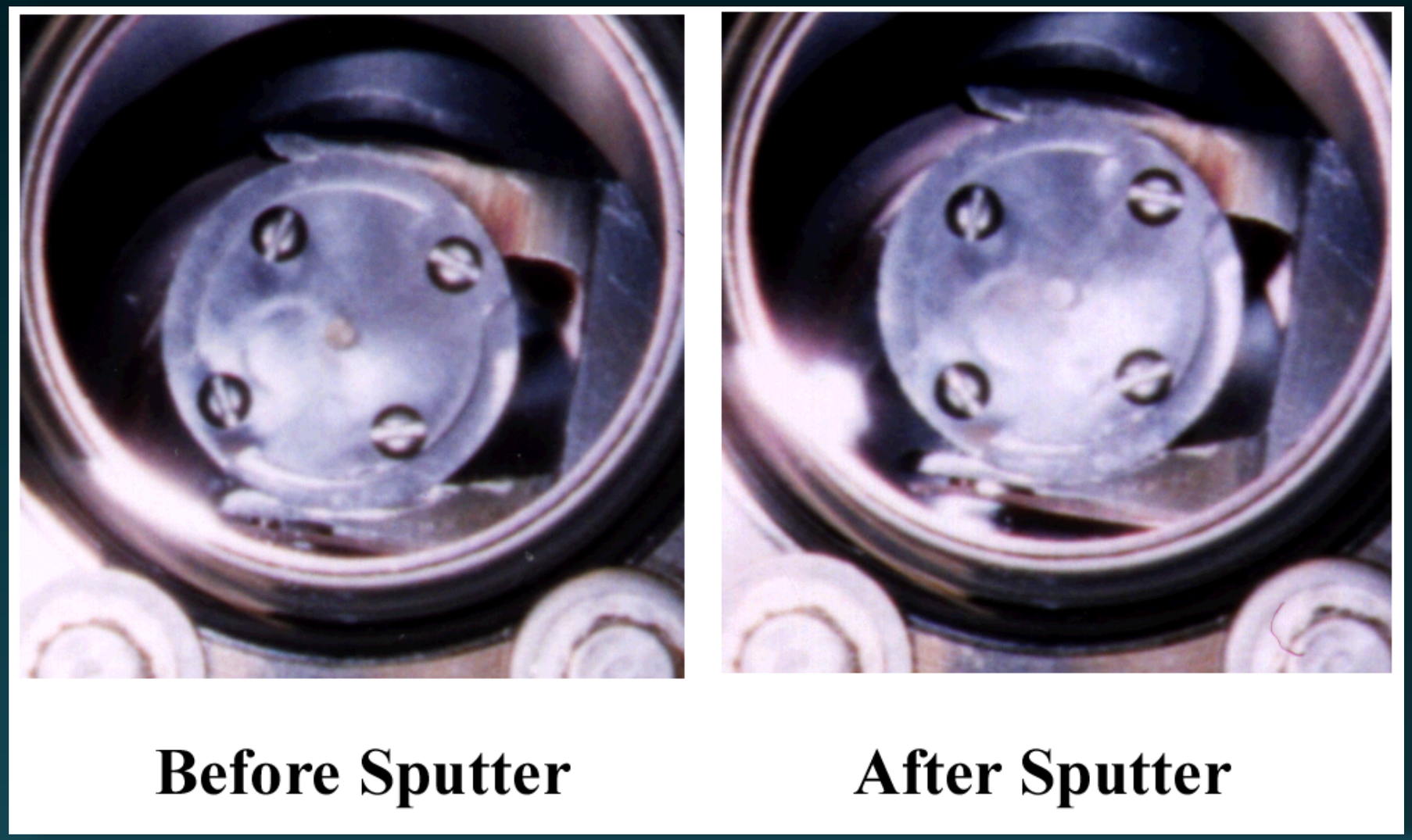
Specialized Handling

- Repeated Sputter-Anneal Cycles to Remove Dissolved O_2
- Samples Transferred to Vacuum Transfer Vessel
- Samples Shipped in Vacuum Transfer Vessel at 10^{-8} Torr
- Sputter 5 kV Ar ions
- Anneal 75 °C



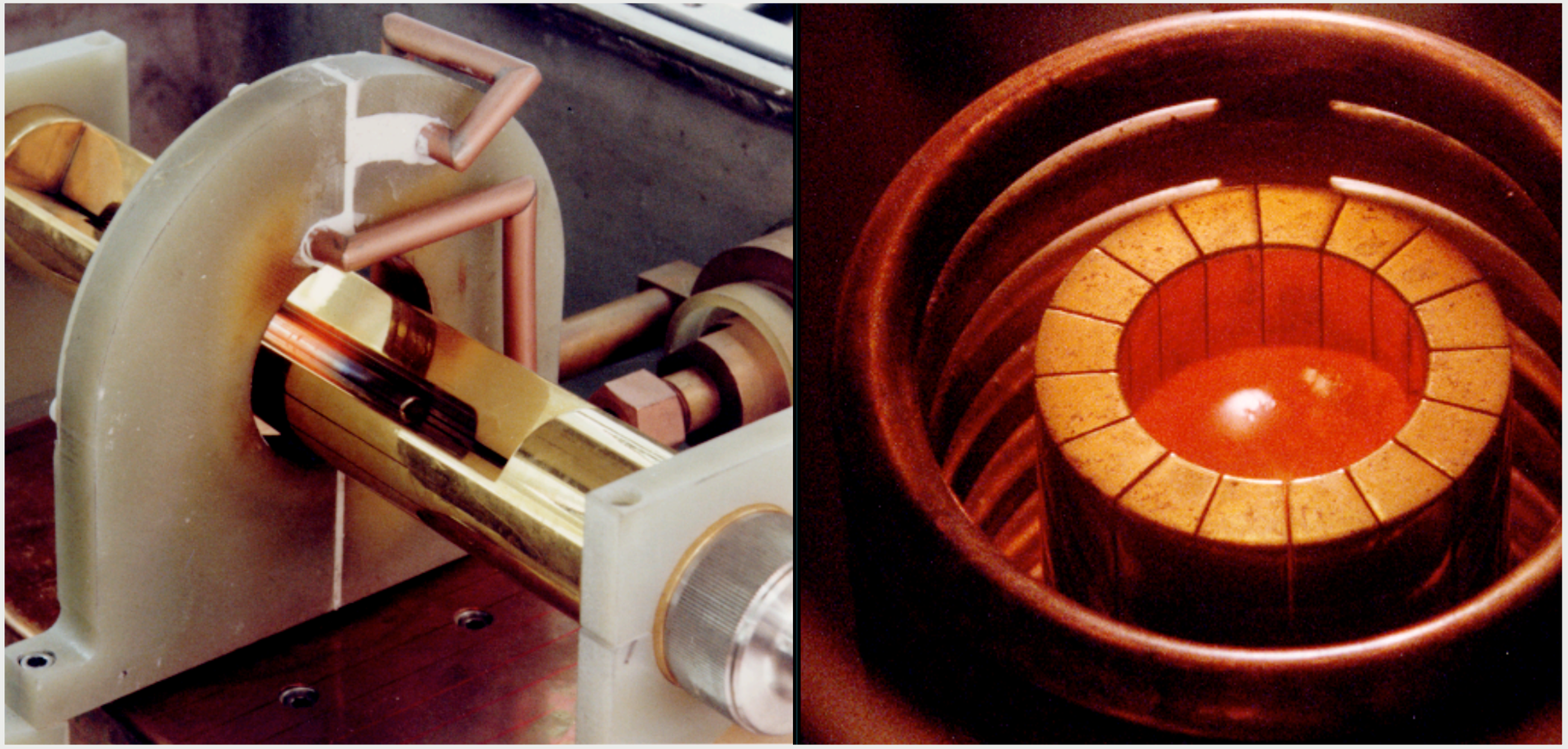
Sample Handling Integral Transfer Plutonium Intense Light Experiments

Pu



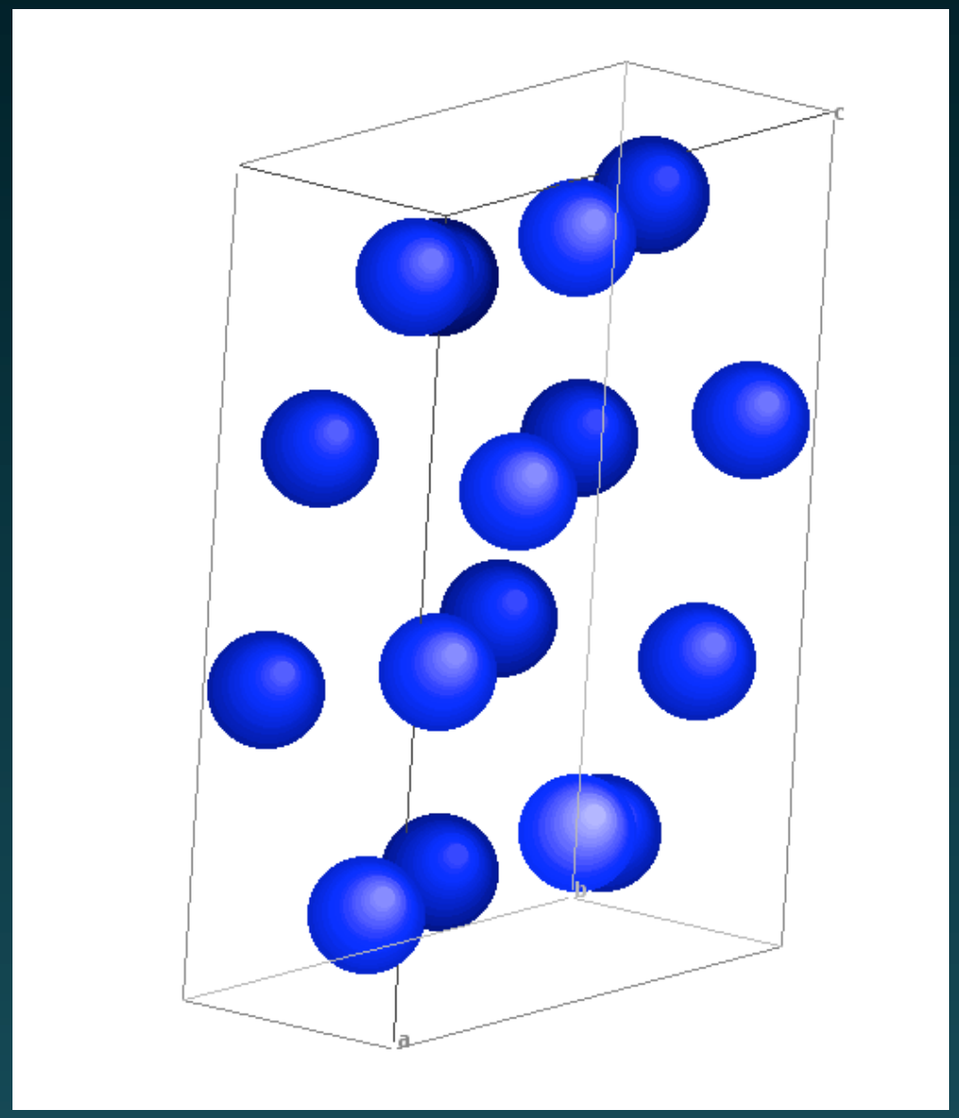
- Cleaning

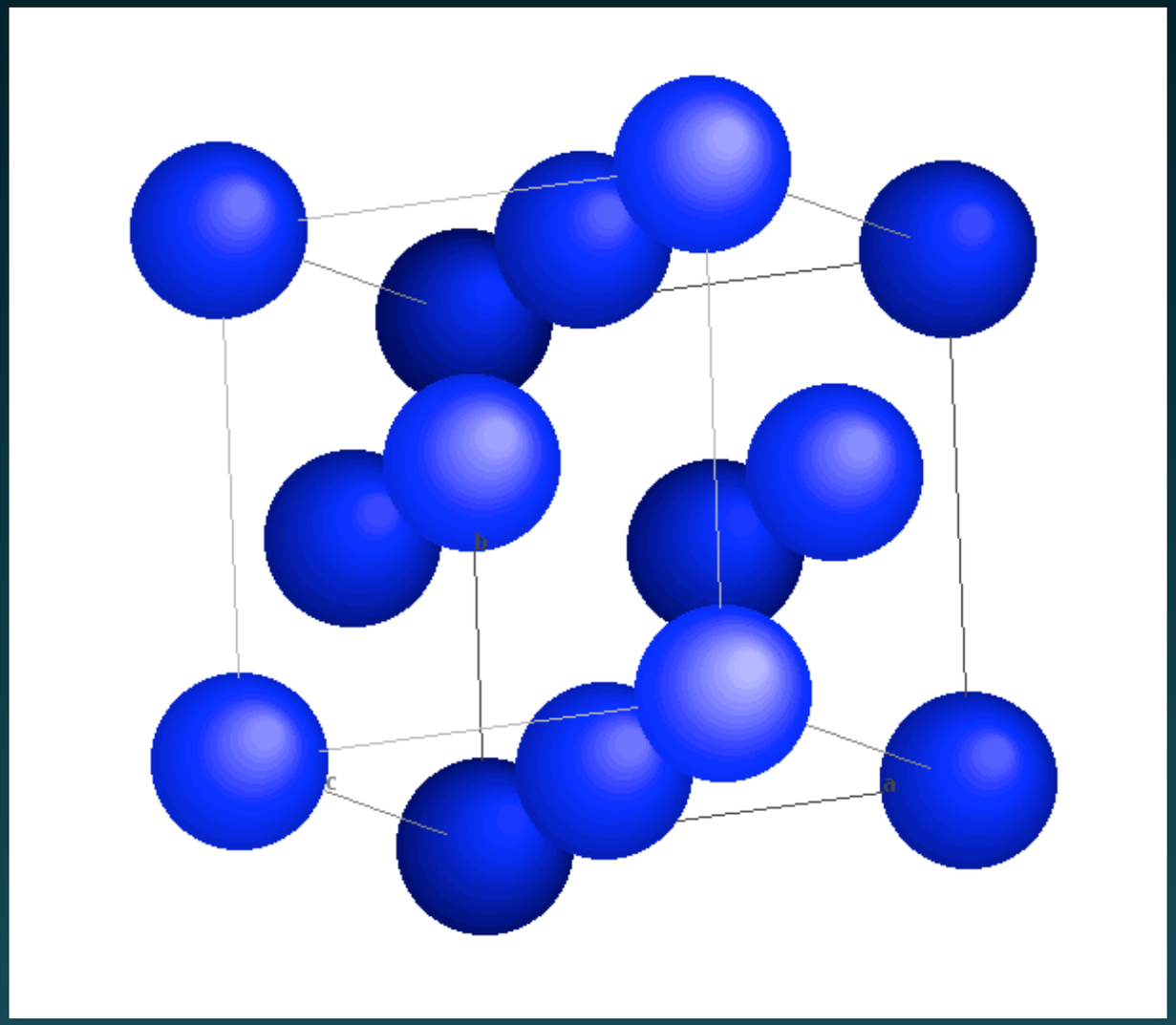
Pu



- Leviation Zone Refinement
  - 1.5 cm/min Zone Travel Rate
  - 800 °C Molten Zone (red glow)
- Levitated Molten Plutonium
  - 180 ppm Impurities

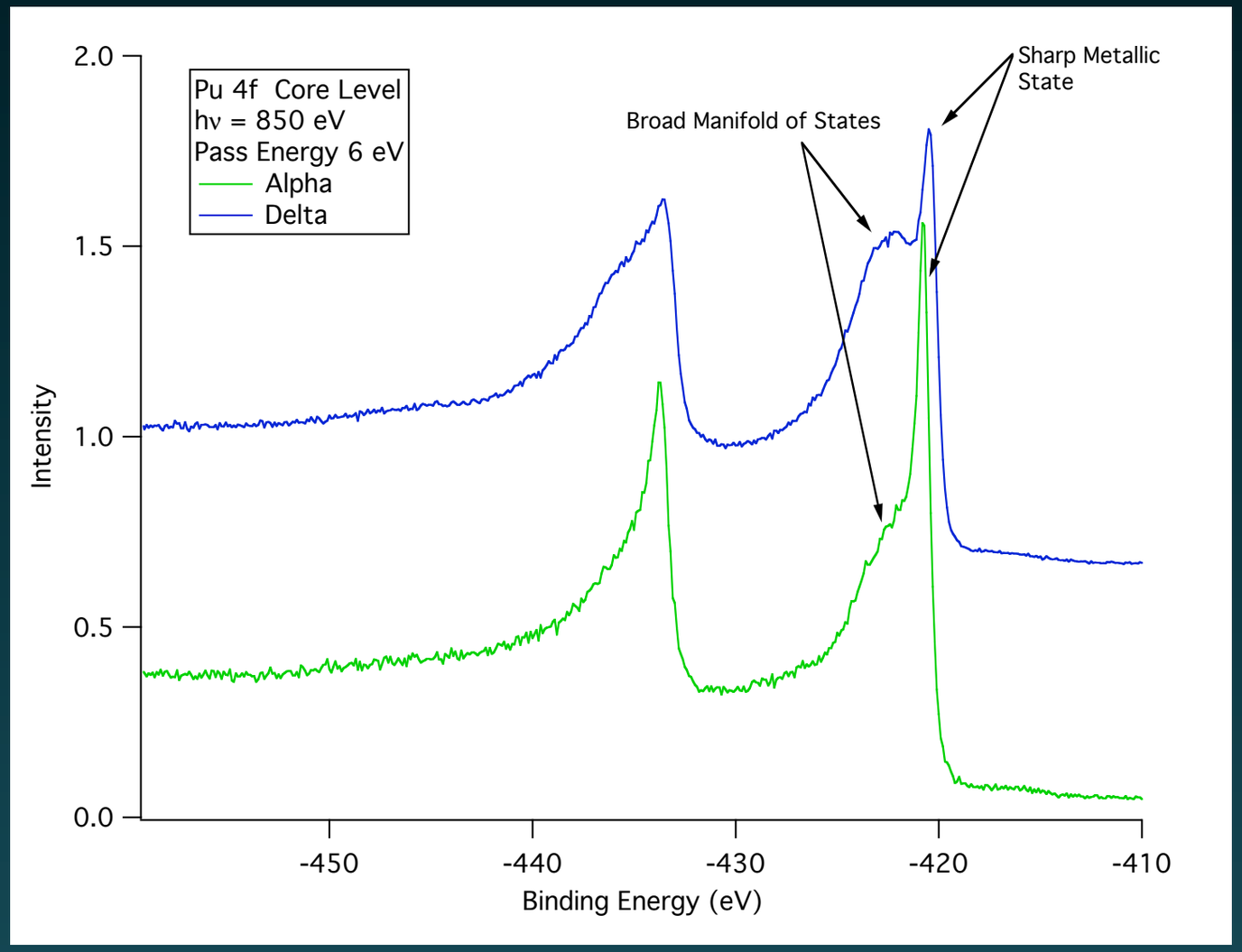
Pu





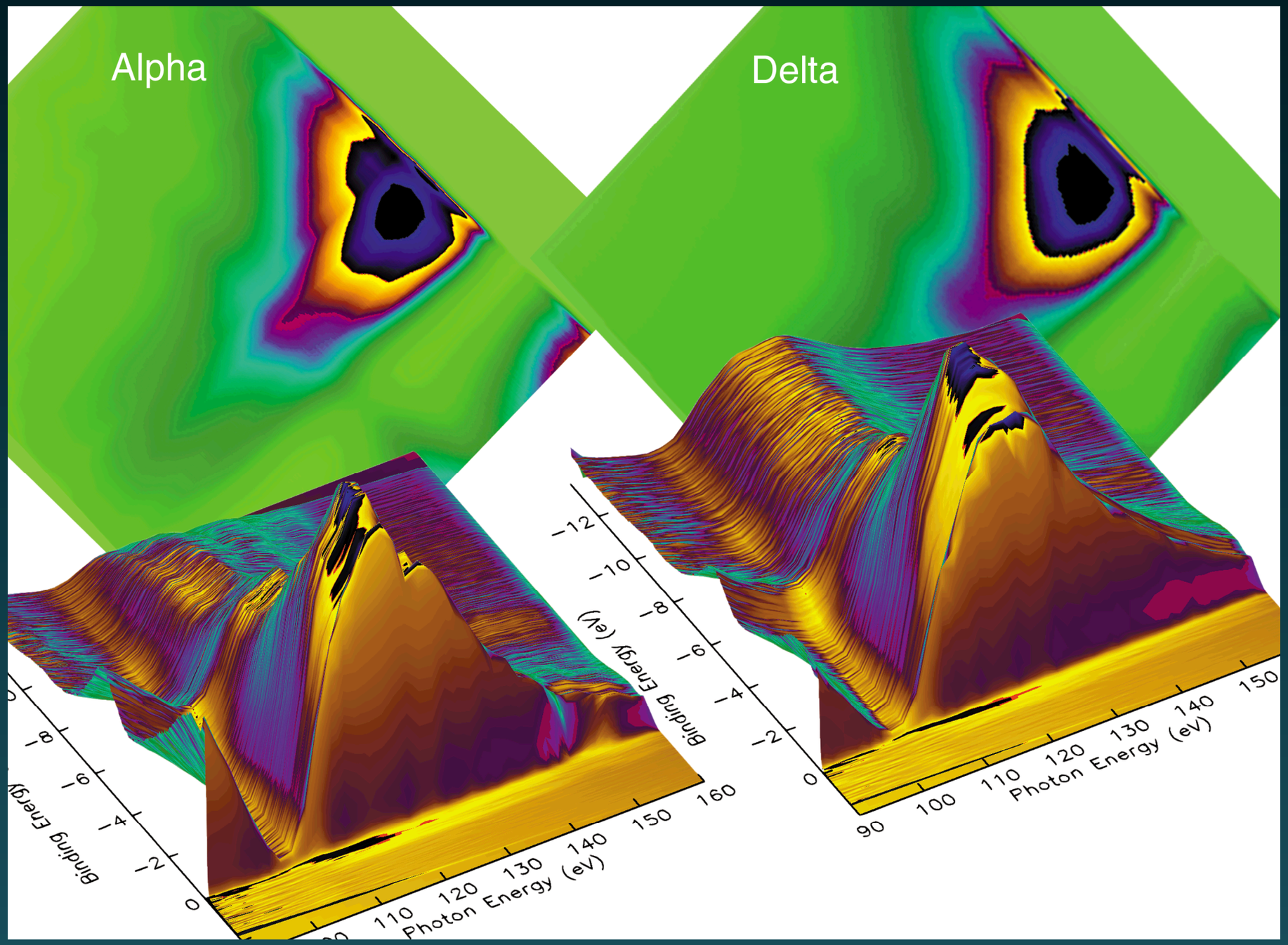
- α - Pu Monoclinic
- δ - Pu FCC

Pu



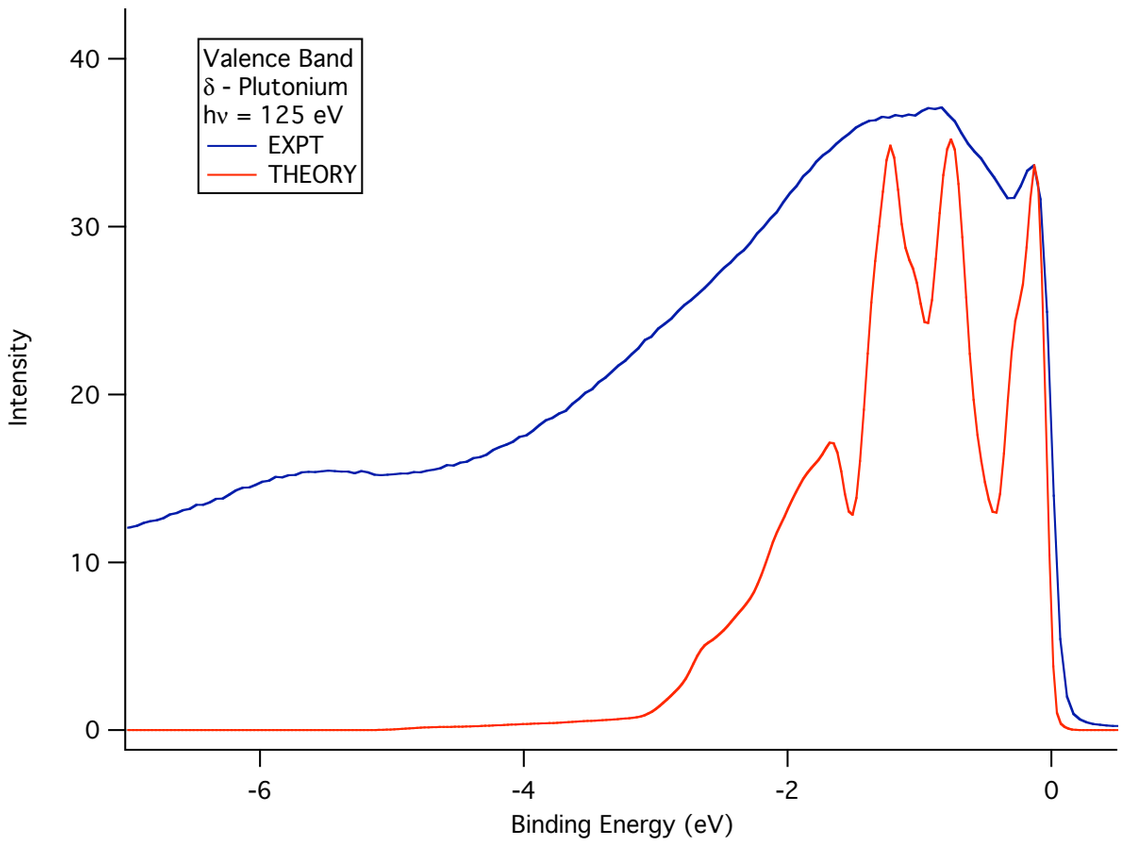
- Plutonium 4f Core Level Spectra
  - Itinerant vs Localized Valence Electrons

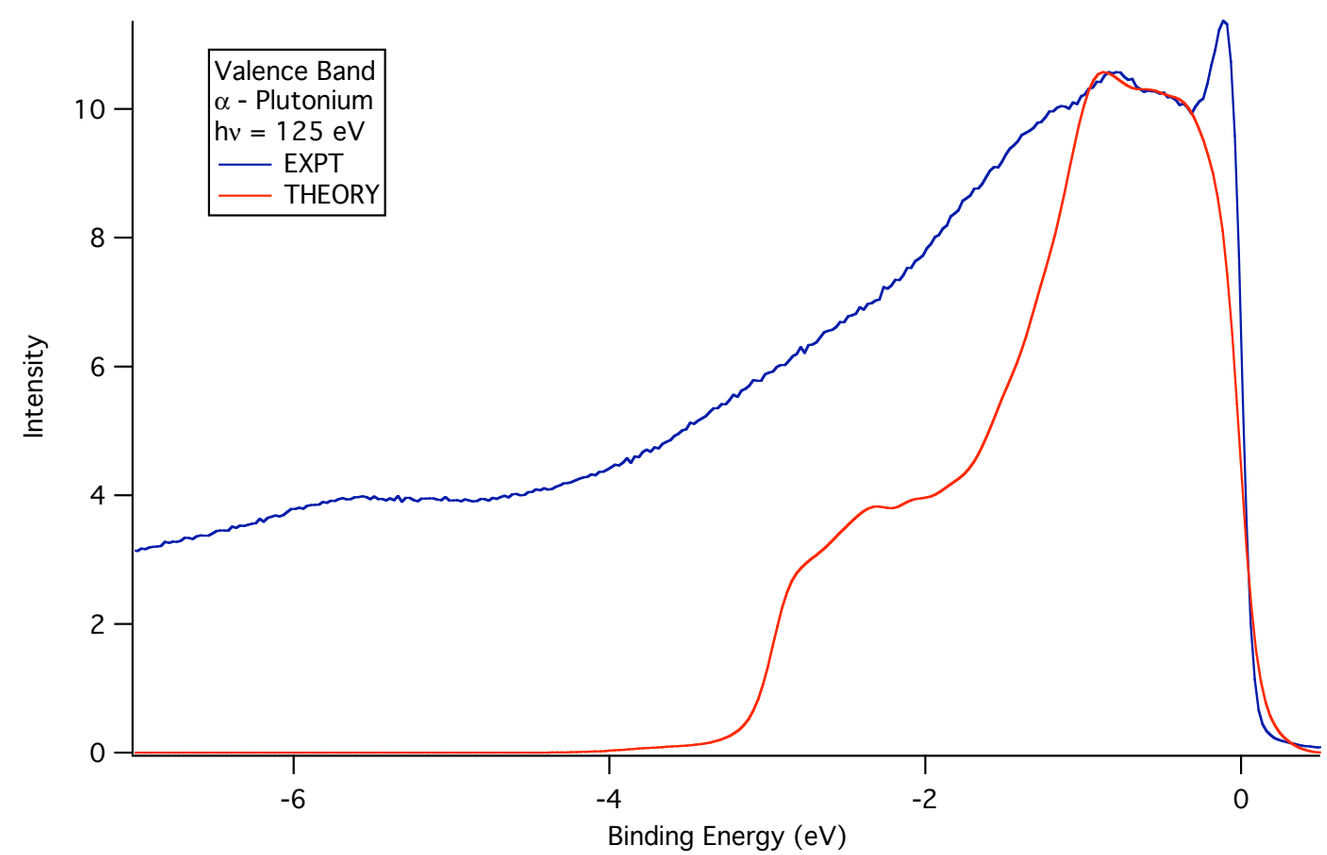
Pu



- Plutonium 5f Valence Band Spectra

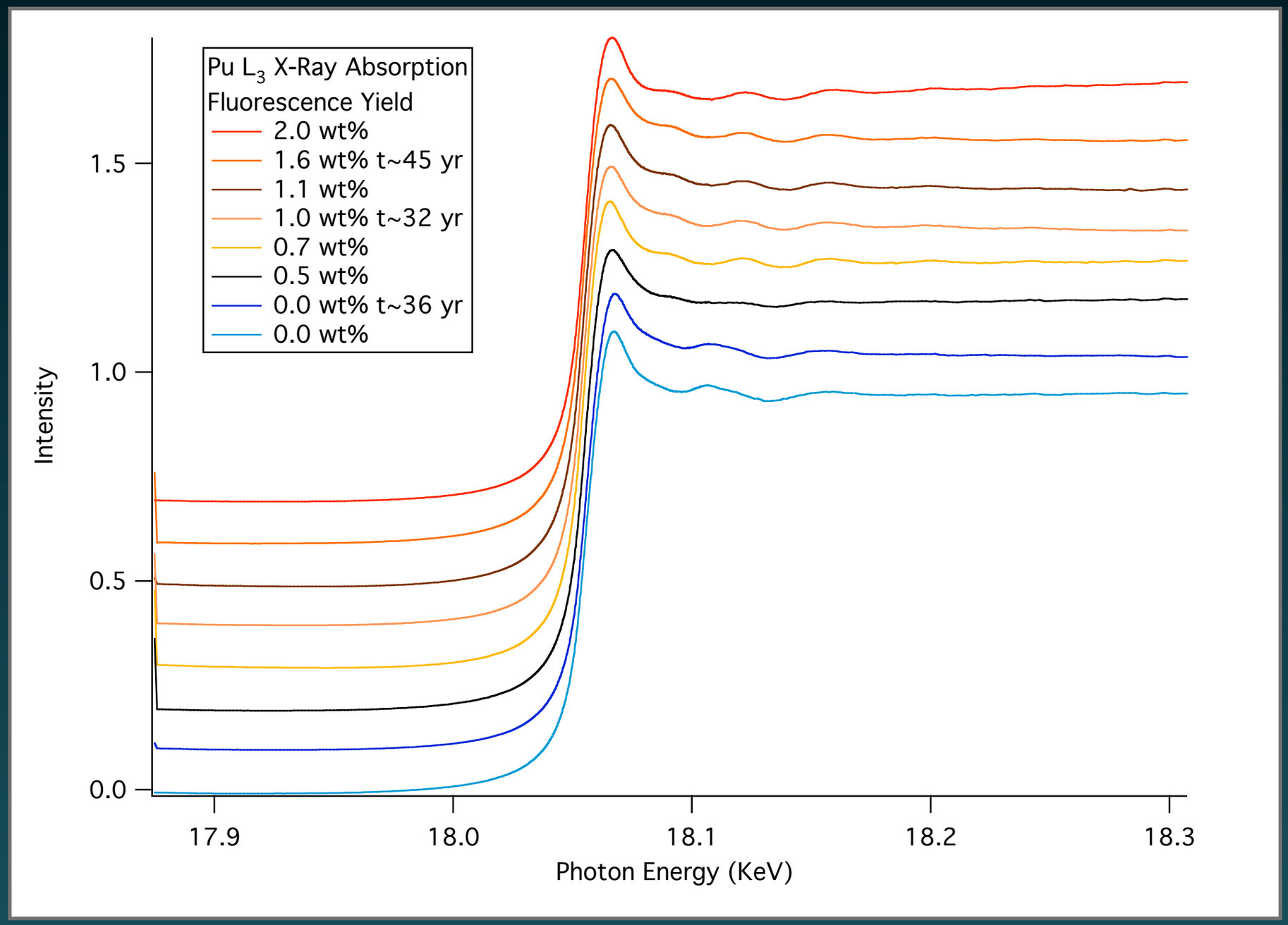
Pu





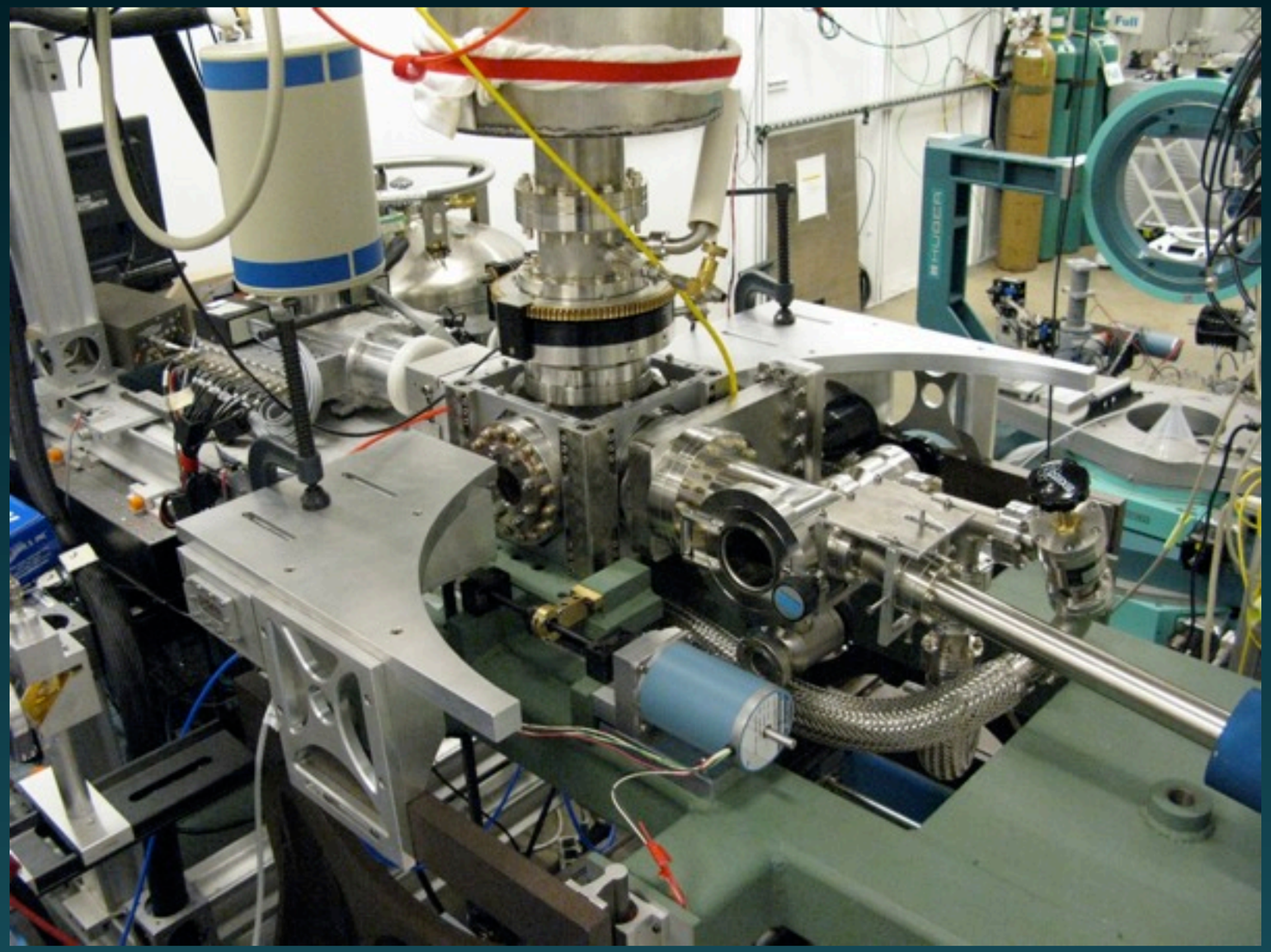
- δ - Pu 4 localized 5f electrons 2 Itinerant
- α - Pu 5f electrons itinerant
- Photoemission Matrix Elements NOT Accounted for in Calculation

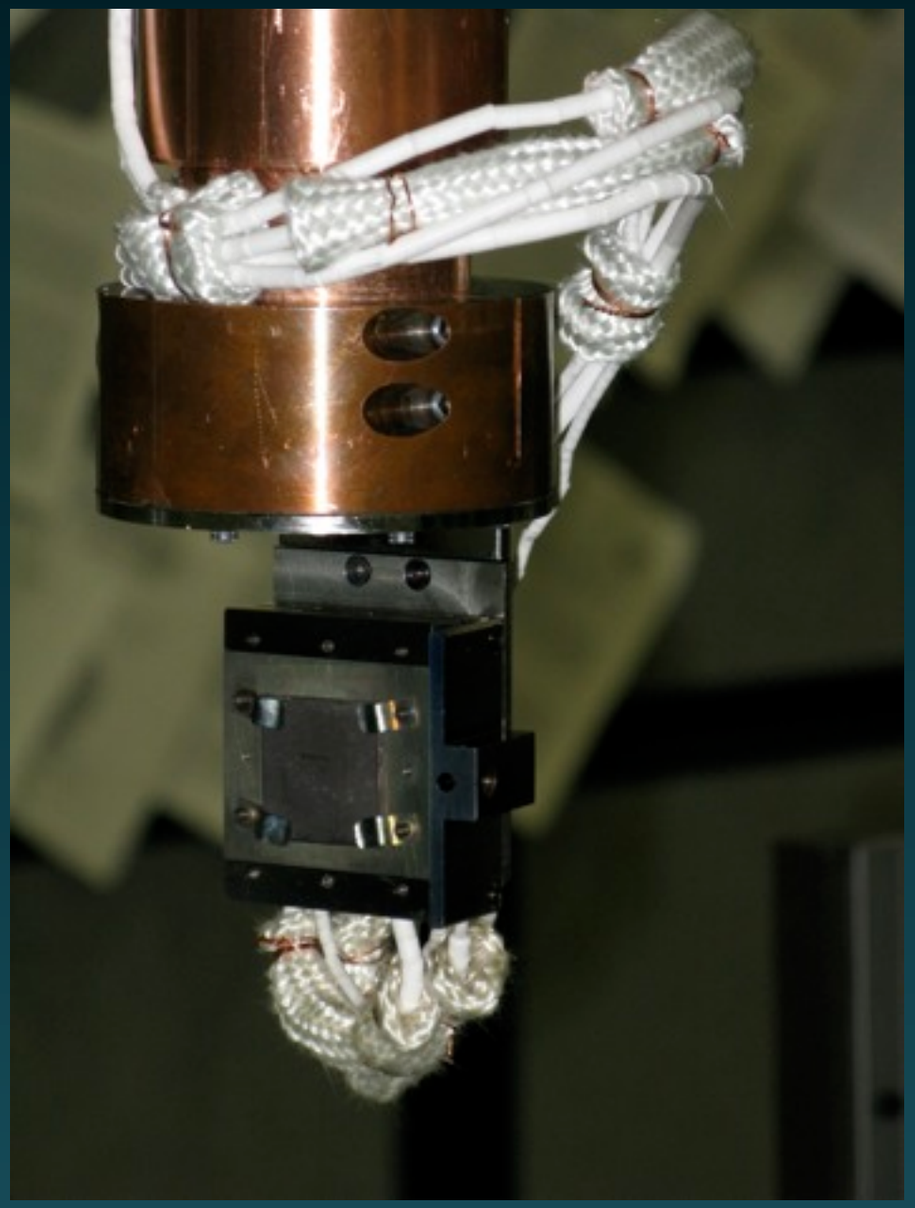
Pu Aging



- X-ray Absorption

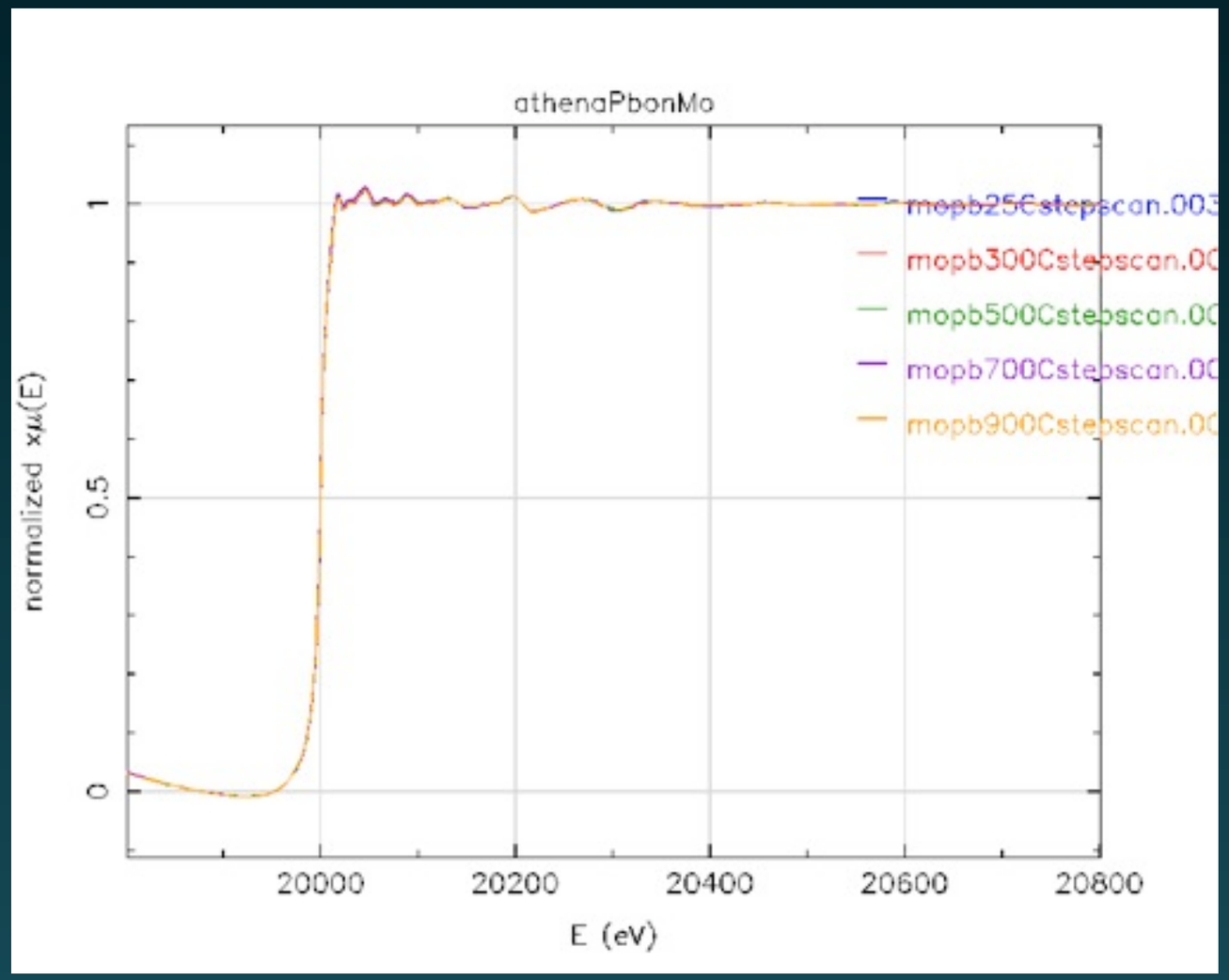
Corrosion Chemistry





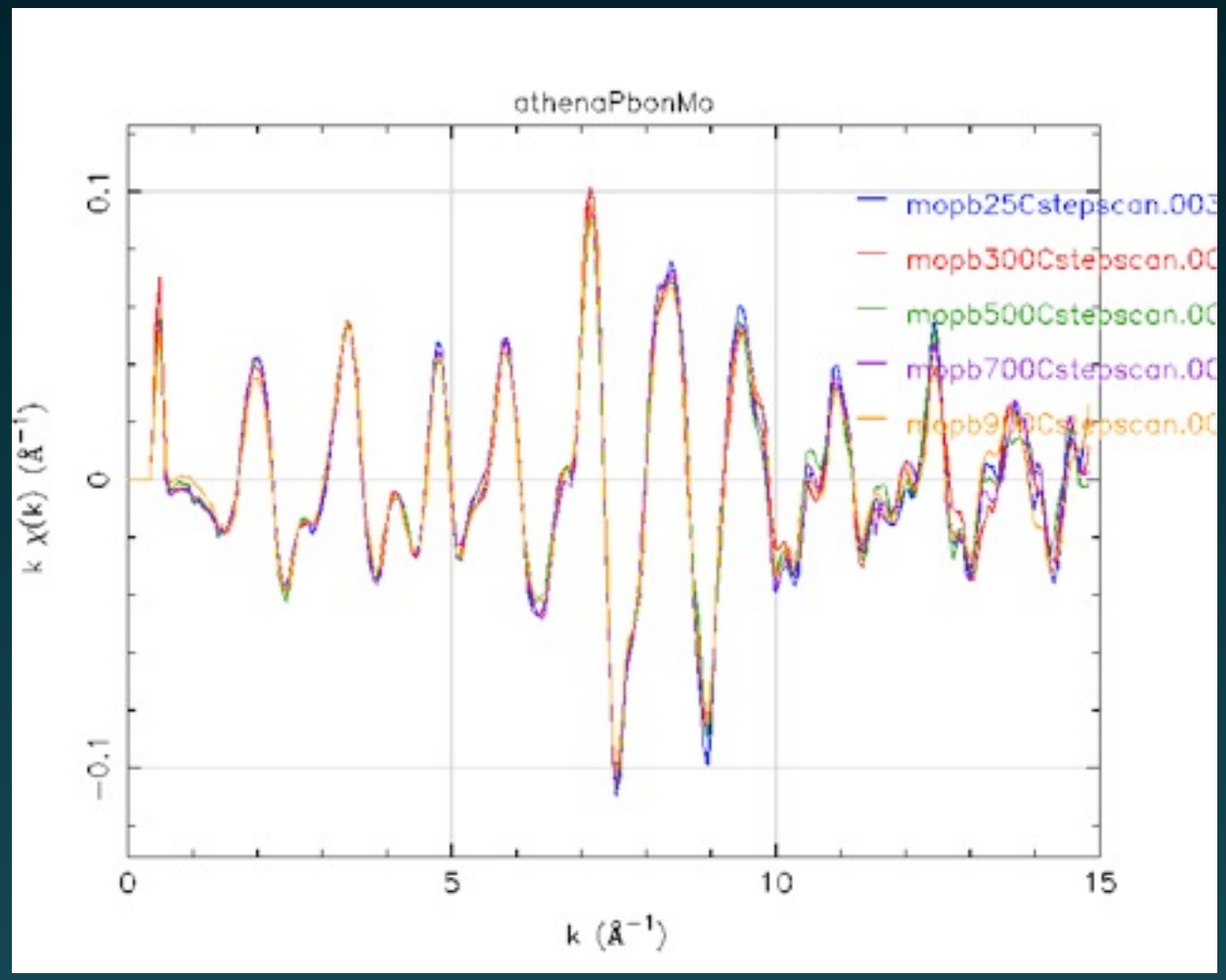
- Develop Systems For Studying Corrosion Chemistry Under Controlled Conditions
- Surface Sensitive Angles

Pb on Mo



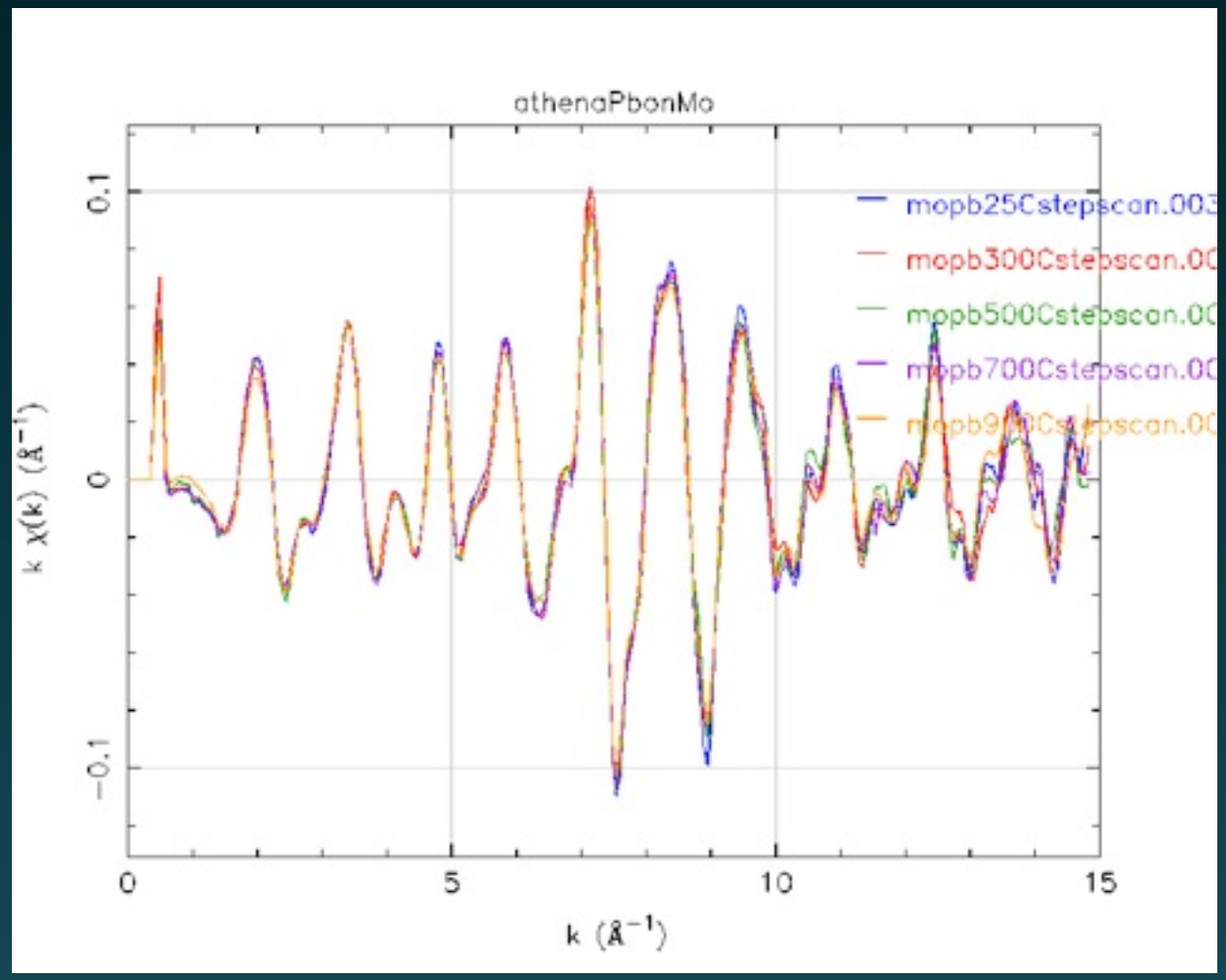
- No Shift In Edge Position

Pb on Mo



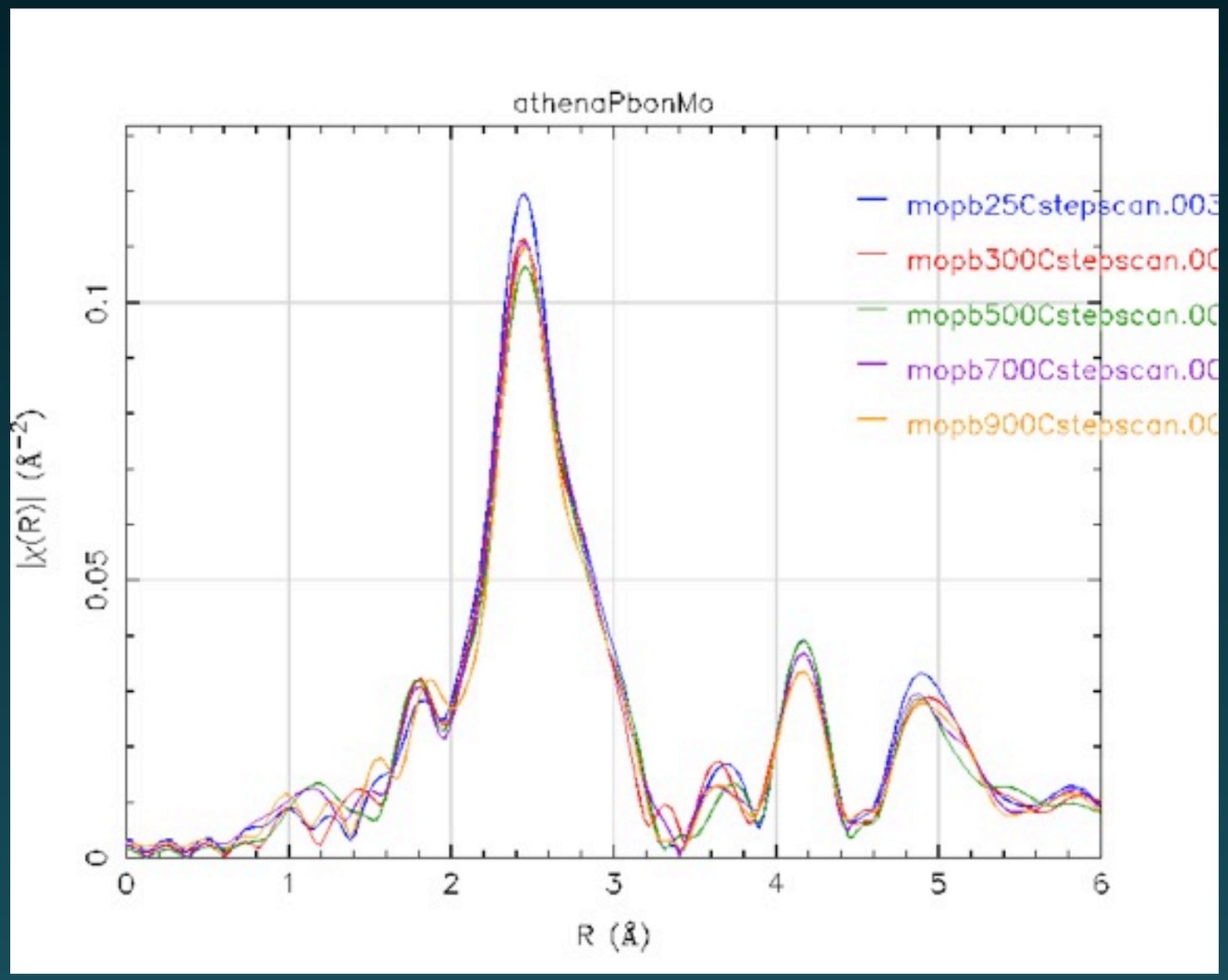
- Not Much Change

Pb on Mo



- Not Much Change

Pb on Mo



- No Significant Oxidation

Pb on Mo

Molybdenum

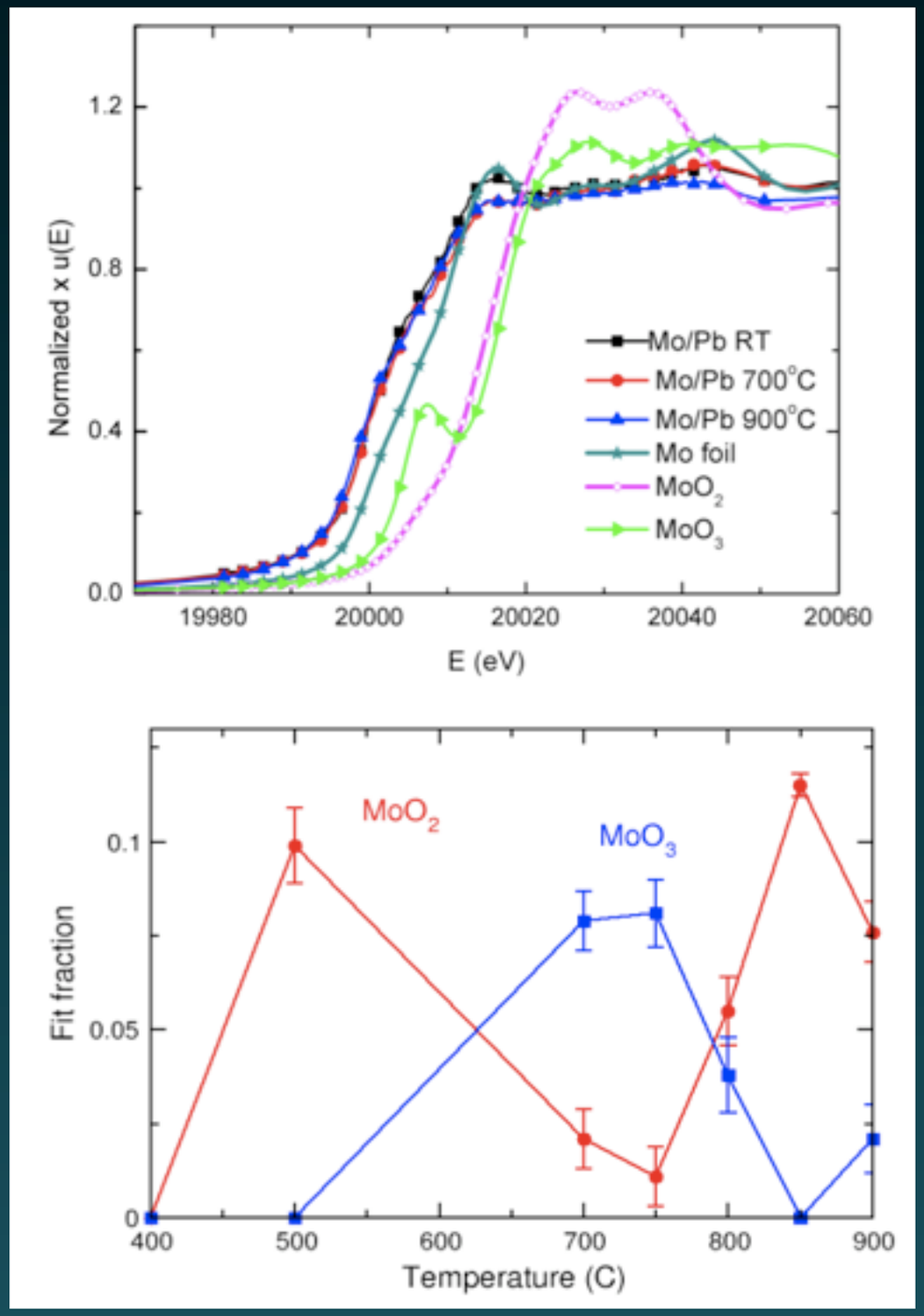
316L SS

Spinel

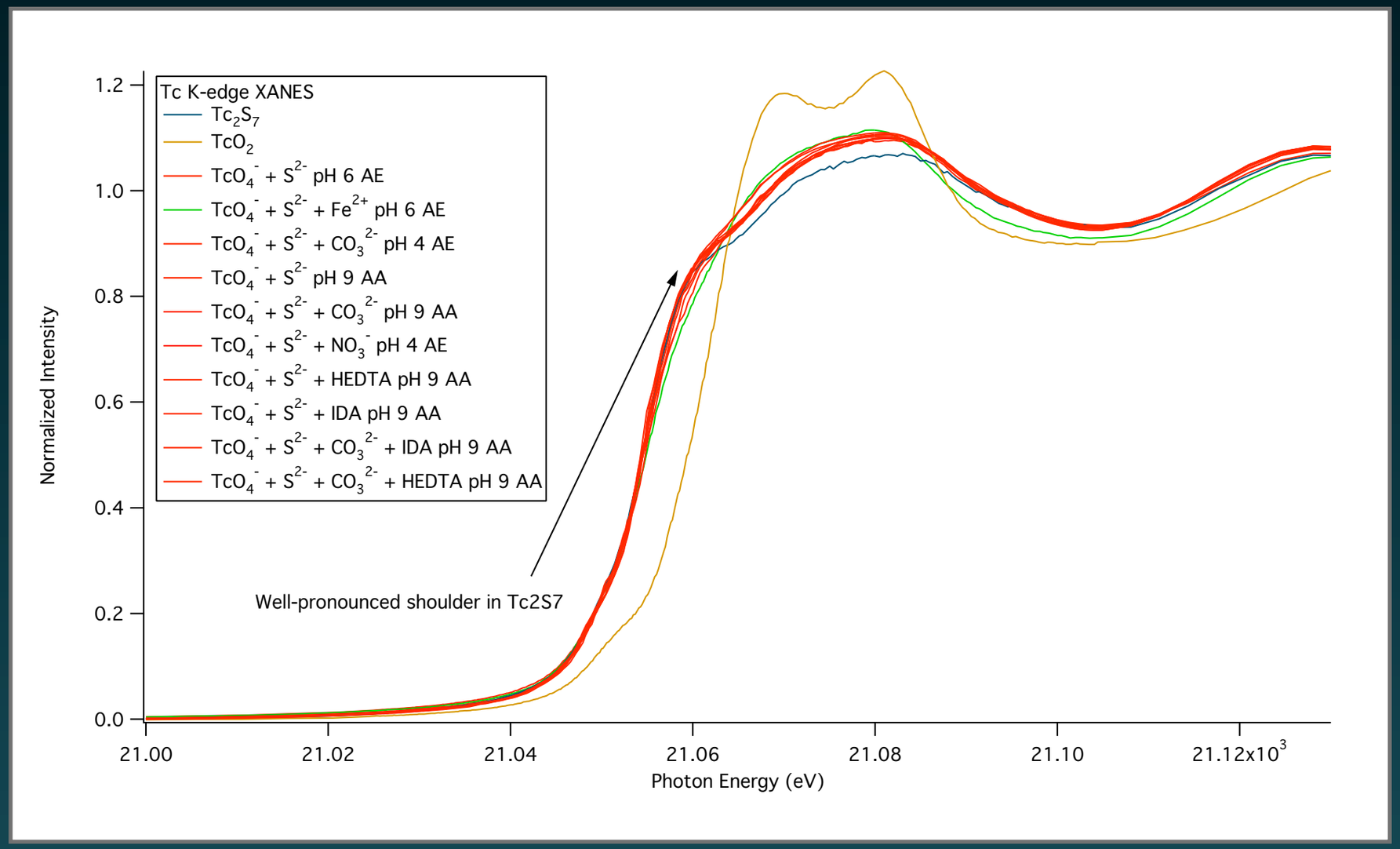
- Old System
  - Tube Furnace With Kapton Window
  - Impossible To Control Oxygen Concentration
  - Could Reach 900 °C

Pb On Mo

- Oxidation Of Mo
- 900 °C
- Large Edge Shift
- Surface Sensitive Angle of 18°

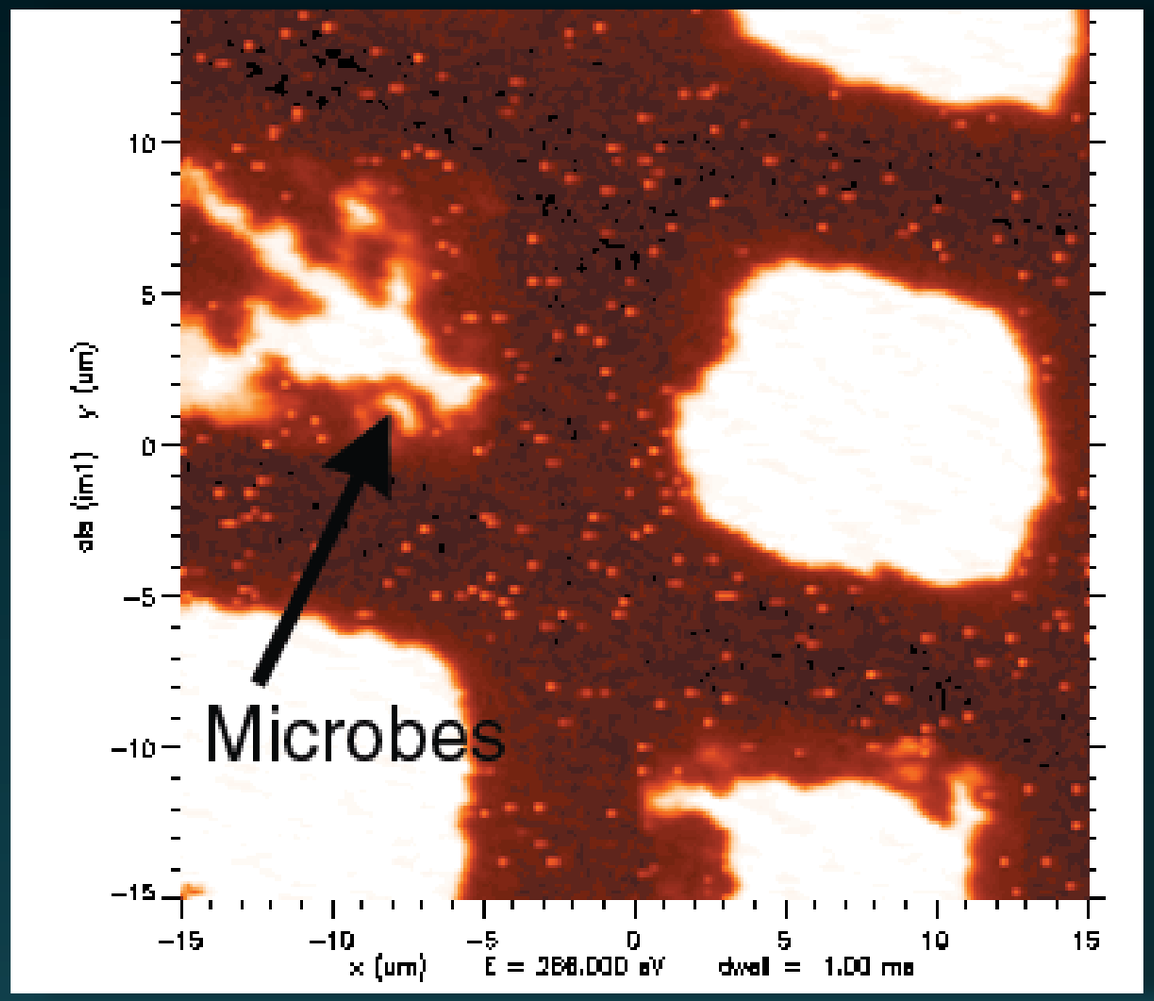


Nuclear Waste



- Fix Tc Plumes At Hanford By Pumping H_2S Into The Ground
- RXTN Product Is >90% Tc_2S_7 under Most Conditions

Detectors



Bioadsorption of Pu(VI) by WIPP 1A

C K-Edge Image

10^{-6} M Pu solution

100 nm Spacial Resolution

Bright Dots -Alpha Particles Striking Detector

- Radioactive Materials Cause Problems For Detectors
  - Can Saturate
  - Can Interfere
- Bent Laue Detector - Extremely High Resolution ~ 100 eV

Bent Laue

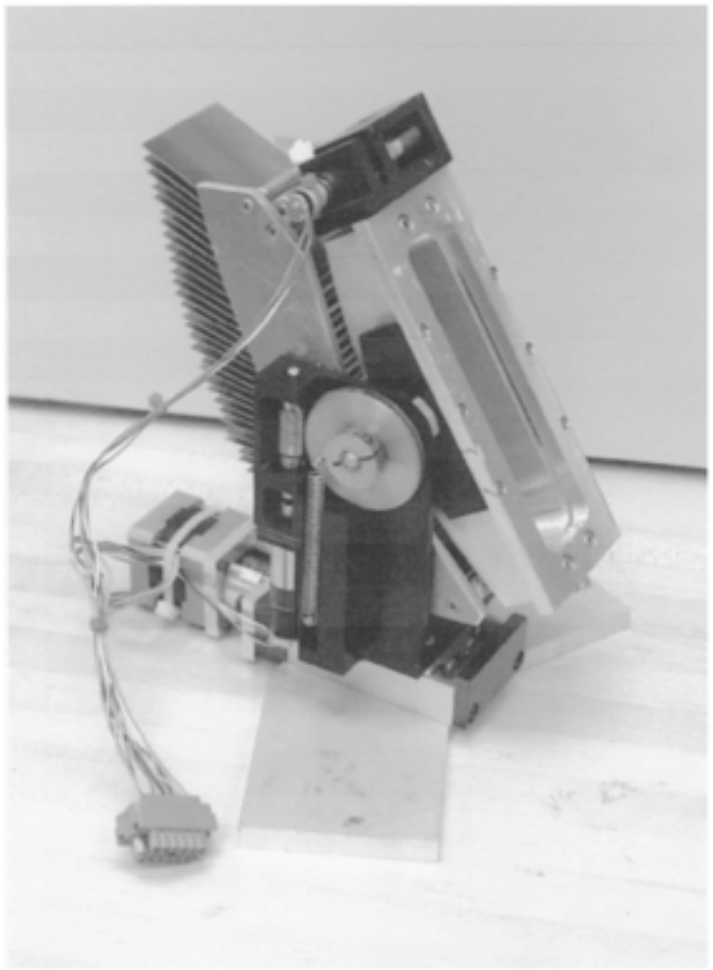


FIG. 1. Photograph of the bent Laue analyzer.

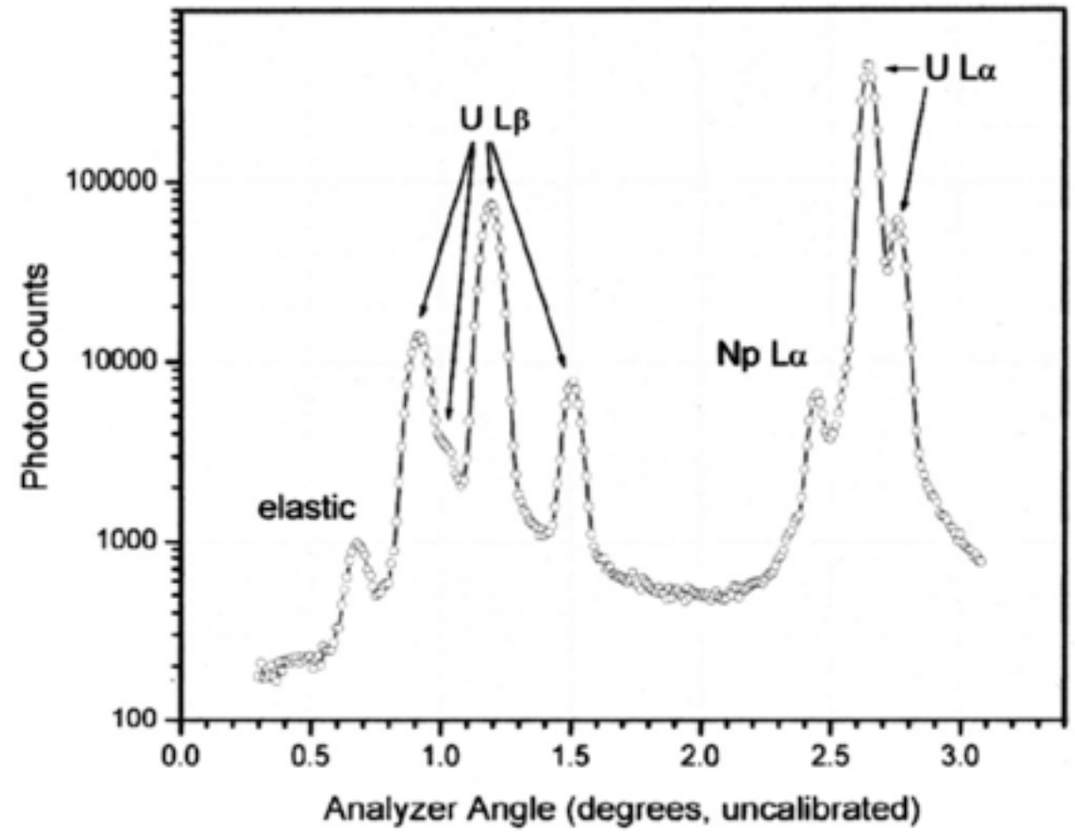


FIG. 2. Bent Laue analyzer spectrum for 1 part Np, 160 parts U (configuration C); incident beam energy: 17 700 eV.

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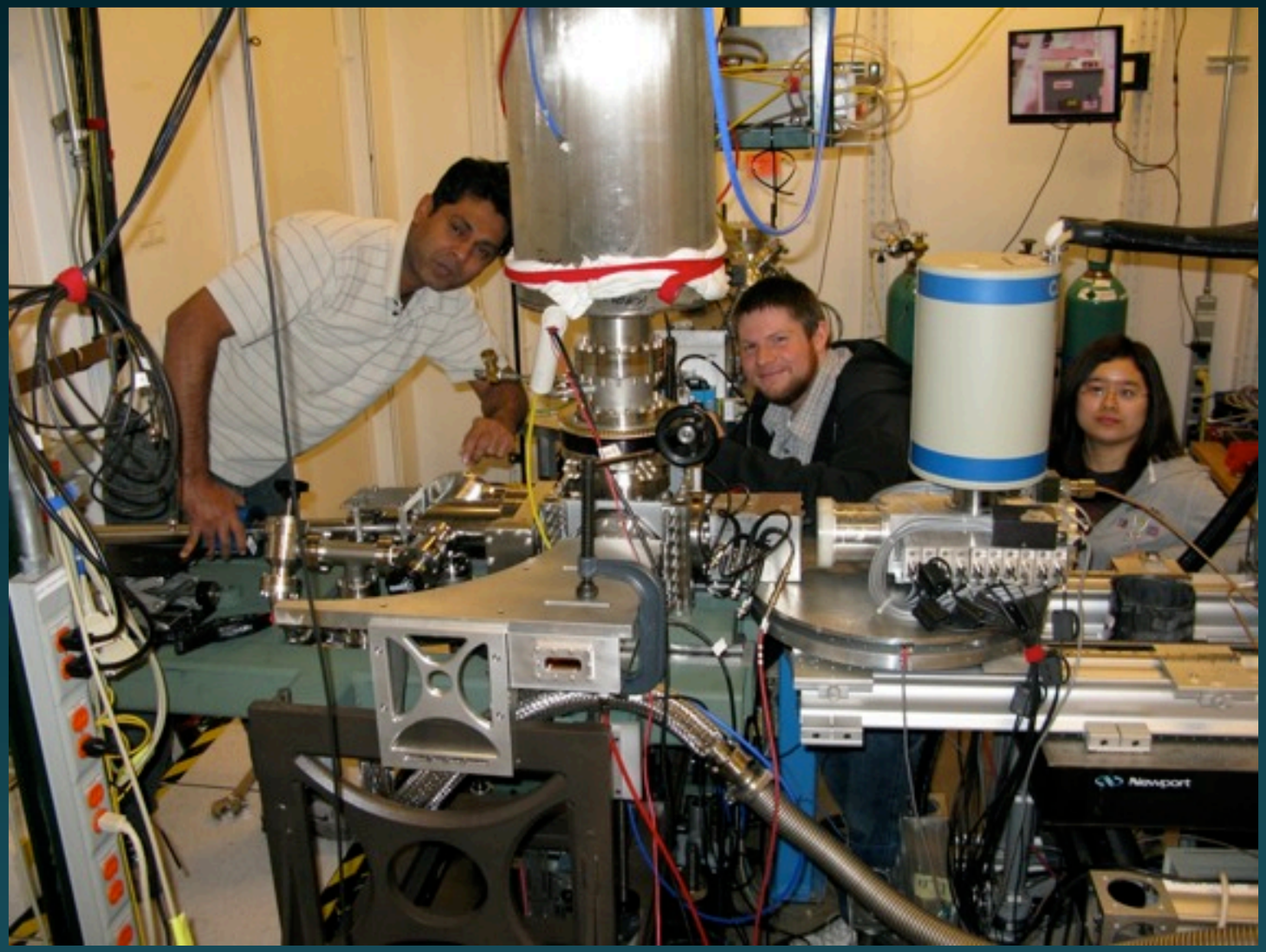
- High Resolution Analyzer 25-70 eV
- Energy Rejection With W Slits

Students



- Lack Of Qualified Nuclear Personnel

Nuclear And Radiological Research CAT

- Prior Work On Radioactive Materials
  - Provided Important Data
  - Limitations
    - Unirradiated Materials
    - Low Activity Samples
    - Minimal Processing
- Designed To Facilitate Experiments With:
  - Moderate Activity
  - Handling of Radioactive Material
  - Neutron Irradiated Samples

NRRCAT

- Expandable
- Canted Undulator Design (U3.1 - 4.3 keV)
- Single Undulator Initial Implementation
- Kohzu Double Crystal Monochromator
- XAS
- XRD (8 Circle Diffractometer)
- XPS
- Microfocussing (Xradia KB mirror, Zone Plates)
- Bent Laue Detectors

NRRCAT

- Fundamental Scientific Understanding
  - Surface Corrosion
  - Fuel-Cladding Interactions
  - Chemistry Under Highly-Ionizing Conditions
  - Material Strength Under Highly-Ionizing Conditions
- Develop high burn-up fuel.
- Develop cladding alloys that will hold integrity at high temperatures.
- Develop structural materials that will function at high-temperature in highly corrosive and highly ionizing environments.

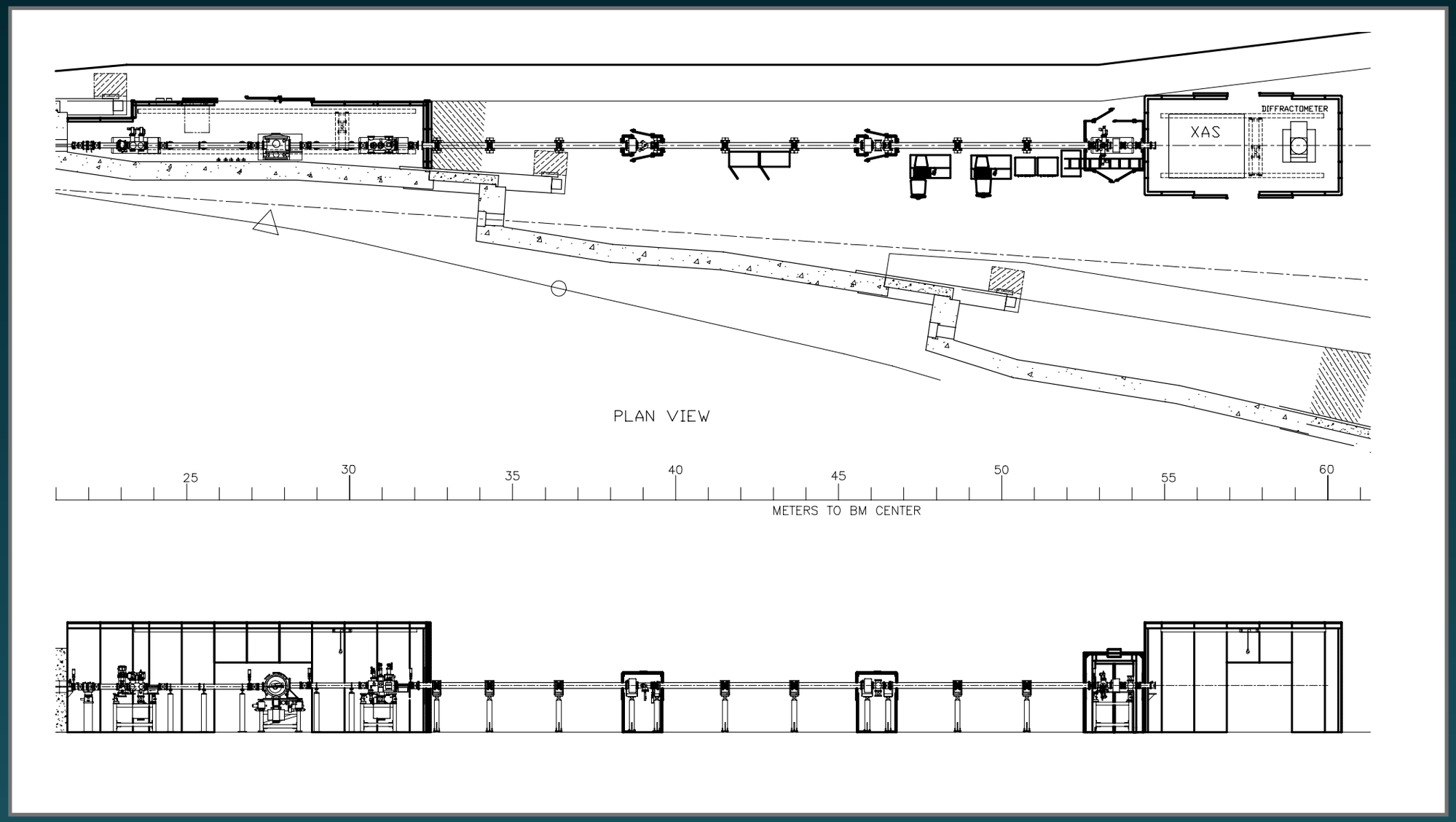
NRRCAT

- The proposed NRR CAT will provide a unique facility for the study of radioactive materials.
- It will greatly expand the post-irradiation evaluation capabilities available to experimenters in the U. S.

NRRCAT

- Current Membership Of NRRCAT
  - ATR NSUF
  - Argonne NE Division
  - Illinois Institute of Technology
  - New Brunswick Laboratory
  - Penn State University
  - University of California, Santa Barbara
  - University of Illinois, Urbana-Champaign
  - University of Wisconsin, Madison

Questions??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METERS TO BM CENTER