HIPPO meets ERNI & BERT - combining diffraction and imaging for characterization of nuclear materials

Alexander M. Long[1] (PI of NSUF IS proposal),
Sven C. Vogel[1], Tsviki Y. Hirsh[2], Adrian S. Losko[3]

HIPPO: High-Pressure/Preferred Orientation
ERNI: Energy-resolved neutron imaging
BERT: Bragg-edge radiography & tomography

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[2]Soreq Nuclear Research Center, Yavne, Israel
[3]Forschungs-Neutronenquelle Heinz Maier-Leibnitz, Garching, Germany

LA-UR-25-22742

NSUF Nuclear Science User Facilities

LANSCE Los Alamos Neutron Science Center

Los Alamos NATIONAL LABORATORY

NSUF Instrument Scientist Proposal by Alex Long

- PI Alex Long (FP5/ERNI instrument scientist)
- 110K awarded FY24
- 0.04 FTE (~2 weeks) for Sven Vogel (HIPPO instrument scientist)
- Goals:
  - Integrate imaging with TOF diffraction $\Rightarrow$ less beam time needed
  - Simplify data analysis $\Rightarrow$ easier access

NSUF Visions @ LANSCE

Elevating Neutron Resonance Imaging Techniques at LANSCE for Next Generation Nuclear Materials Research

Mission Statement:

We aim to advance neutron resonance imaging (NRI) techniques at the Los Alamos Neutron Science Center (LANSCE) by integrating NRI capabilities with other pulsed-neutron characterization techniques, expanding its sensitivity to lighter isotopes, and enhancing its accessibility to the broader NSUF scientific community. Through these initiatives, we will provide more comprehensive measurements of nuclear materials, expedited executions of NSUF RTE awards, and foster a collaborative and engaged community of researchers, committed to advancing NRI-based characterization techniques within the NSUF program.

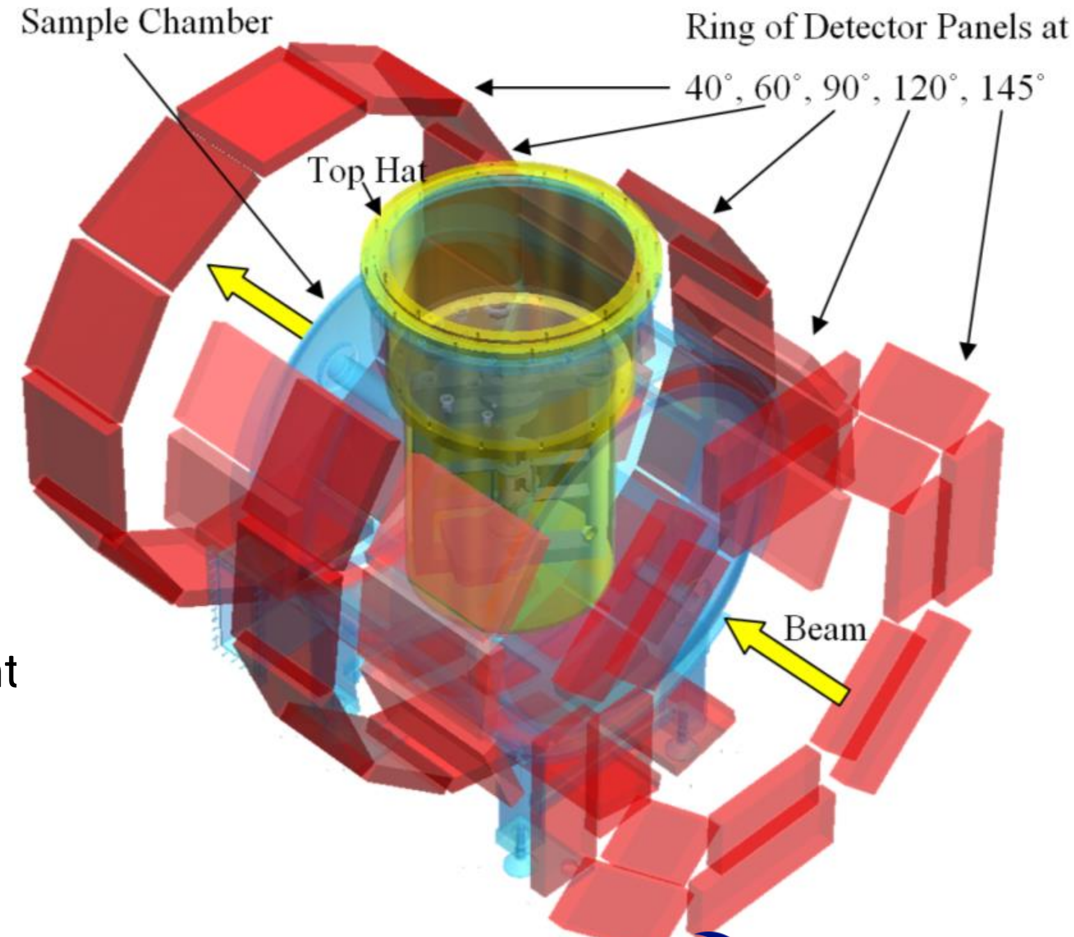
This presentation:

- What was accomplished
- Why is this useful for NSUF

High-Pressure/Preferred Orientation diffractometer HIPPO at LANSCE

General purpose TOF diffractometer
- Crystal structure analysis
- Texture measurements
- Sample environments for temperature, stress, pressure, magnetic field etc.

- $35° < 2\vartheta < 150°$, $0.1Å < \lambda < 18Å$, $0.37\% < \Delta d/d < 1.8\%$
- Moderator-sample: 8.87 m
- Measurement time for texture: ~10 minutes
- Measurement time for powder: <1 minute
- Beam spot: 10 mm Ø, 14mm penumbra
- Thermal flux: ~10^7 n/s/cm^2 at 90 microA proton current
- Detector coverage: 4.9 m^2, ~20% of 4π (1200 ^{3}He tubes on 53 panels)
- Sample chamber: ~1m^3, 73.4cm $\varnothing$ opening



Wenk et al., NIM, ***515*** (2003) 575. Vogel et al., Powder Diffraction, **19** (2004) 65.

Neutron Diffraction: Micro- & Crystal Structure

- Diffraction provides information on microstructure (phase fractions, texture, strains etc.) and crystal structure (lattice parameters, atom position, atomic displacement parameters etc.)
- Applications include changes of micro- and crystal structure as a function of temperature, irradiation etc.
- Data relevant to inform and benchmark models
- Example: NSUF funded investigation of the U-Zr phase diagram (“In-Situ Phase Analysis of Phase Transitions in U-(6, 10, 20, 30) wt%Zr Fresh Fuels”, 18-1437, Walter Williams)

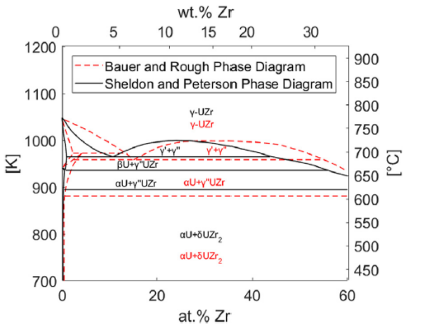
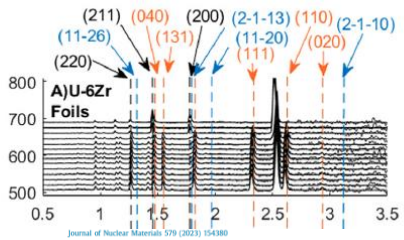
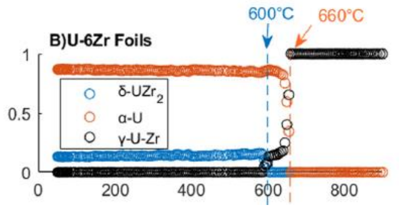


Fig. 1. A view of the Bauer and Rough phase diagram [13,14] (red) plotted with Sheldon and Peterson phase diagram [12] (black) from 0 to 60 at.% Zr (0–36.5 wt.% Zr) illustrating the different number of isotherms in each description of the U-Zr system.





Contents lists available at ScienceDirect

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journal homepage: www.elsevier.com/locate/jnucmat

ELSEVIER

Phase transformations and thermal expansion coefficients of unirradiated U-X wt.% Zr (X = 6, 10, 20, 30) measured via neutron diffraction

W.J. Williams[a,*], S.C. Vogel[b], M.A. Okuniewski[c]

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https://doi.org/10.1007/s11837-020-04086-8
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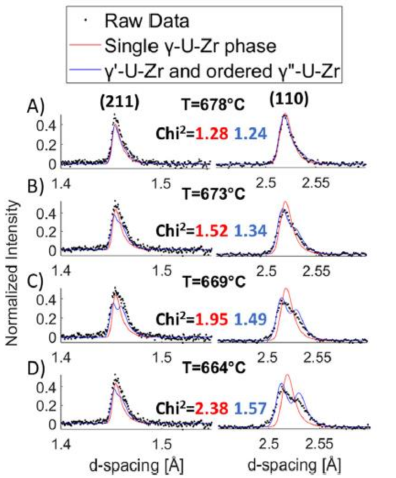
IN-SITU CHARACTERIZATION TECHNIQUES FOR INVESTIGATING NUCLEAR MATERIALS

In Situ Neutron Diffraction Study of Crystallographic Evolution and Thermal Expansion Coefficients in U-22.5 at.%Zr During Annealing

W.J. WILLIAMS,[1,2,4] M.A. OKUNIEWSKI,[1] S.C. VOGEL,[3] and J. ZHANG[3]



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HIPPO

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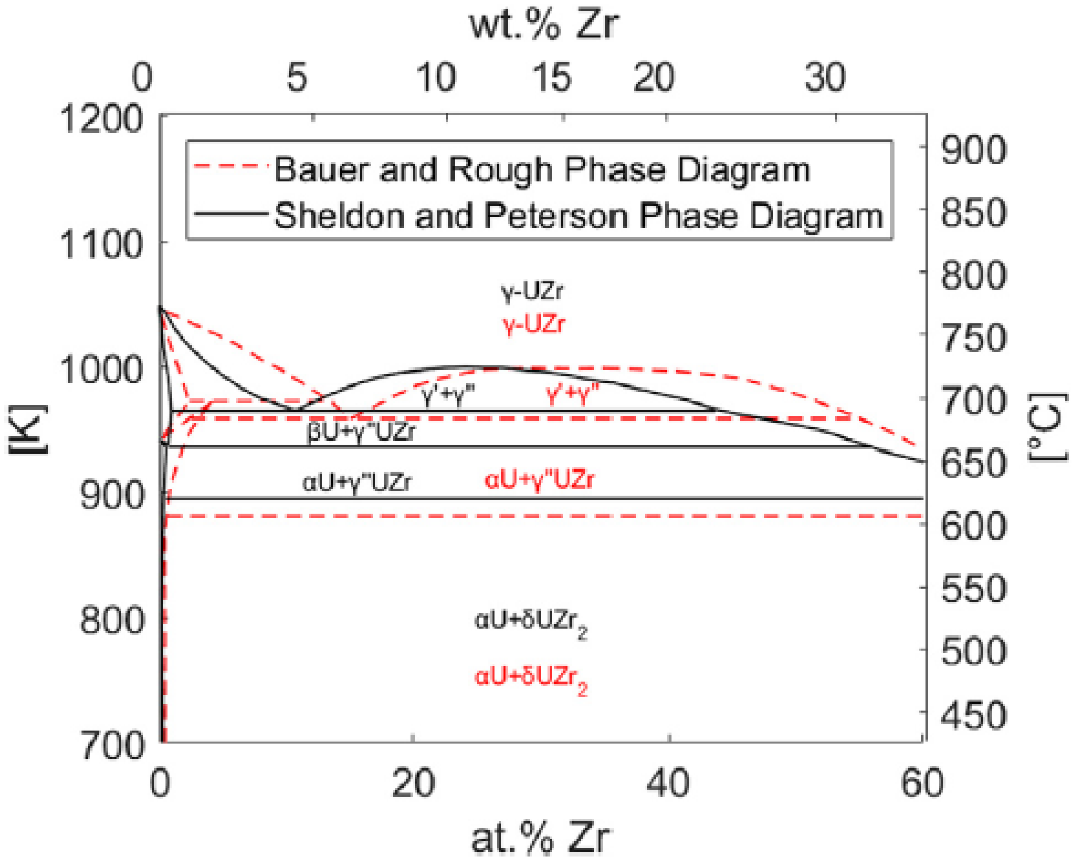
- Example: NSUF funded investigation of phase diagram (“In-Situ Phase Analysis Phase Transitions in U-(6, 10, 20, 30) wt Fresh Fuels”, 18-1437, Walter Williams)



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W.J. WILLIAMS,[1,2,4] M.A. OKUNIEWSKI,[1] S.C. VOGEL,[3] and J. ZHANG[3]

1.4 1.5 d-spacing [Å] 2.5 2.55 d-spacing [Å]

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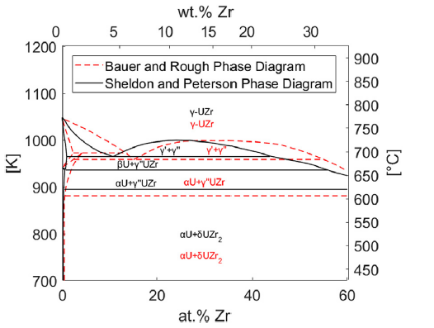
Neutron Diffraction: Micro- & Crystal Structure

- Diffraction provides information on microstructure (phase fractions, texture, strains etc.) and crystal structure (lattice parameters, atom position, atomic displacement parameters etc.)



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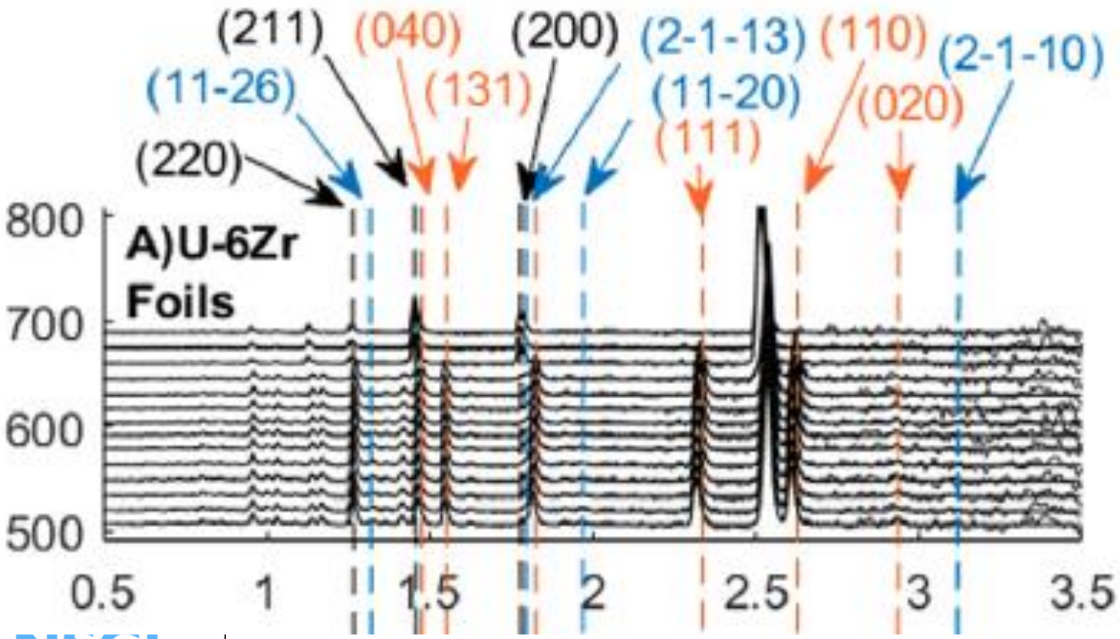
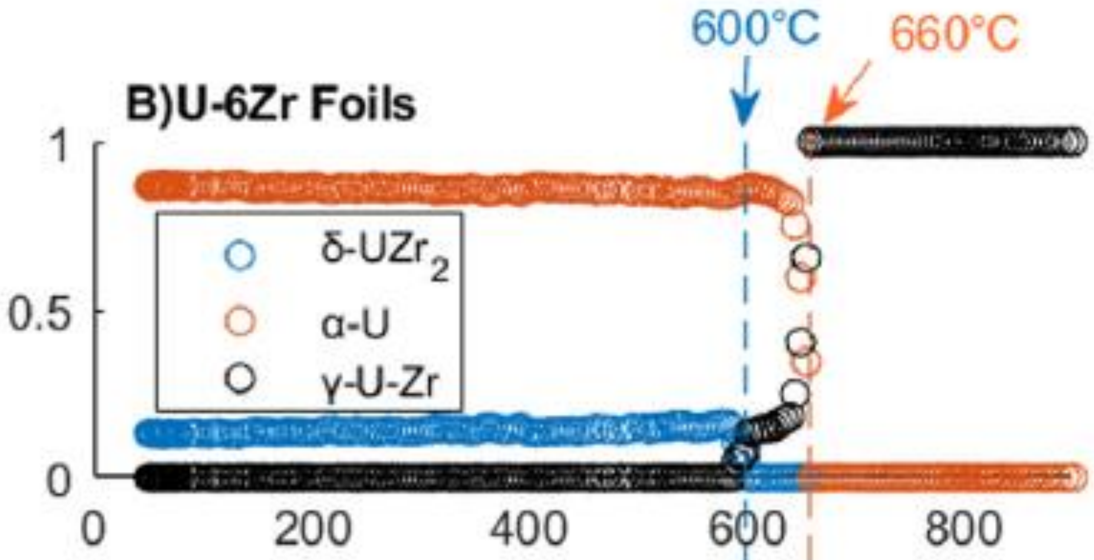
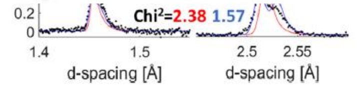




and Thermal Expansion Coefficients in U-22.5 at.%Zr During Annealing

W.J. WILLIAMS,[1,2,4] M.A. OKUNIEWSKI,[1] S.C. VOGEL,[3] and J. ZHANG[3]



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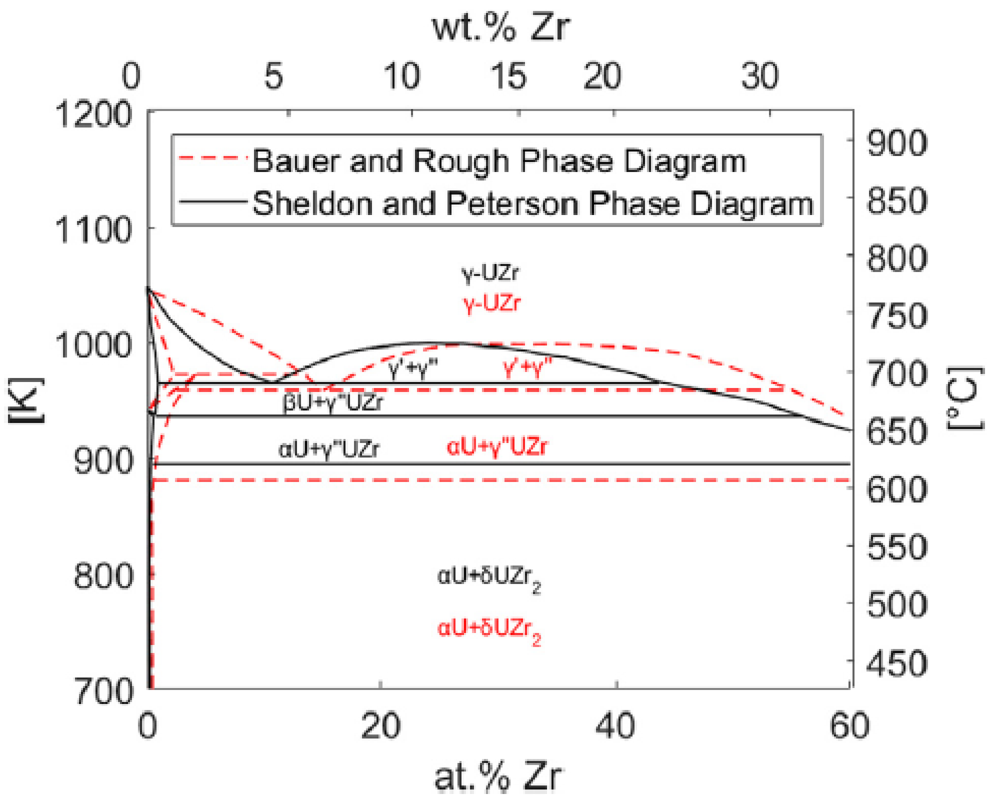
Neutron Diffraction: Micro-



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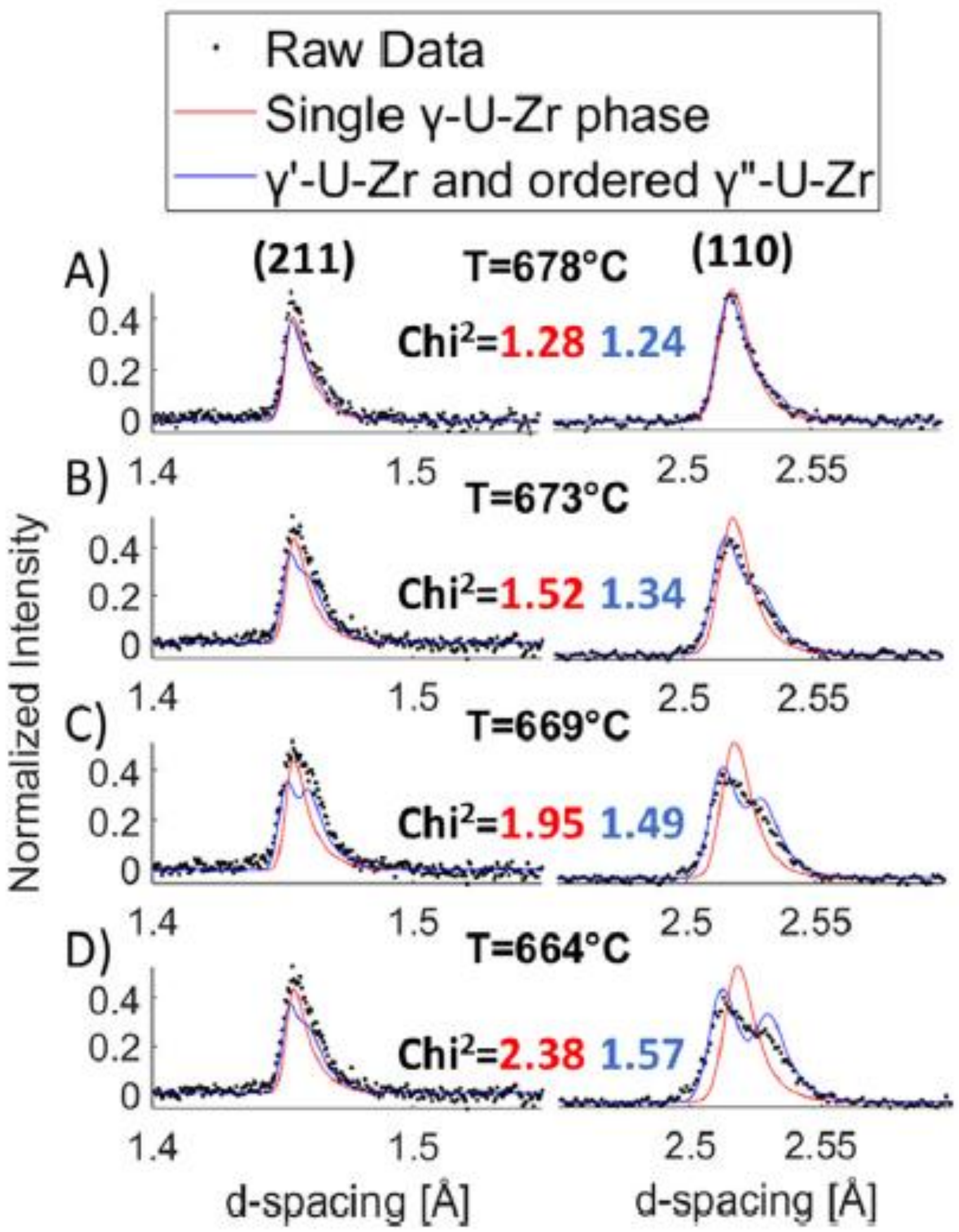

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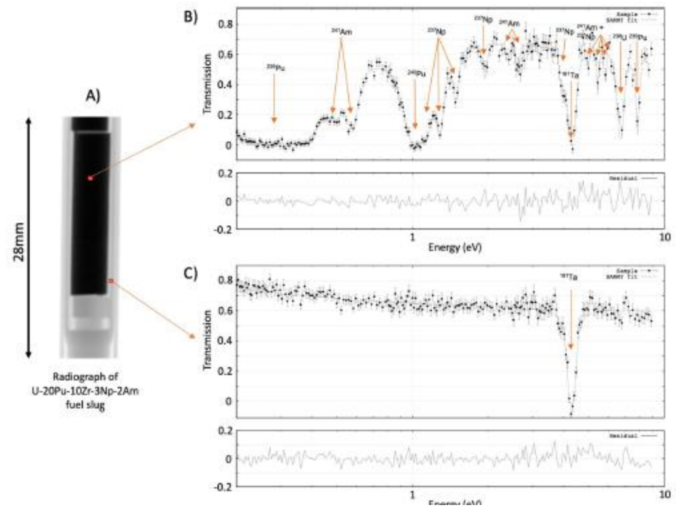
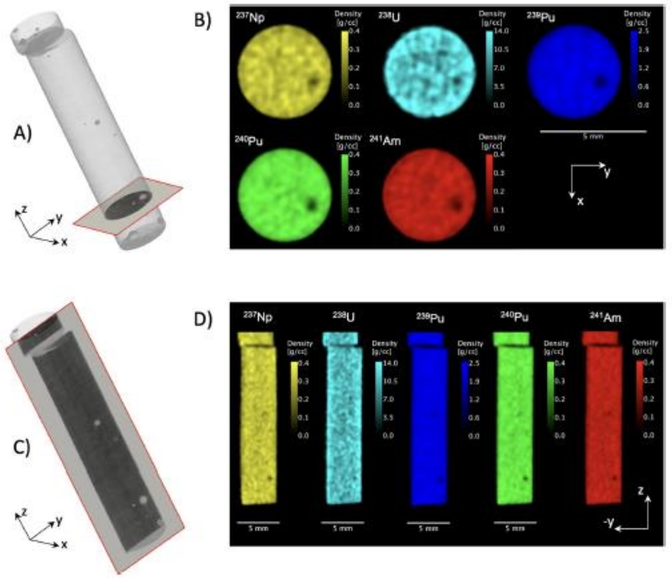
Energy-resolved Neutron Imaging: Spatial distribution of isotopes, micro-, & crystal structure

- Combination of short-pulsed neutron source at LANSCE and suitable detectors enables analysis of neutron transmission data for each pixel in a radiography
- Map isotopes densities (U235, U238, Pu239, H etc.) from resonances
- Map phases and lattice parameters from Bragg-edges
- Example: 3D isotope densities in U-20Pu-10Zr-3Np-2Am metallic fuel sample





scientific reports

Check for updates

OPEN

3D isotope density measurements by energy-resolved neutron imaging

A. S. Losko[1,2] & S. C. Vogel[1]

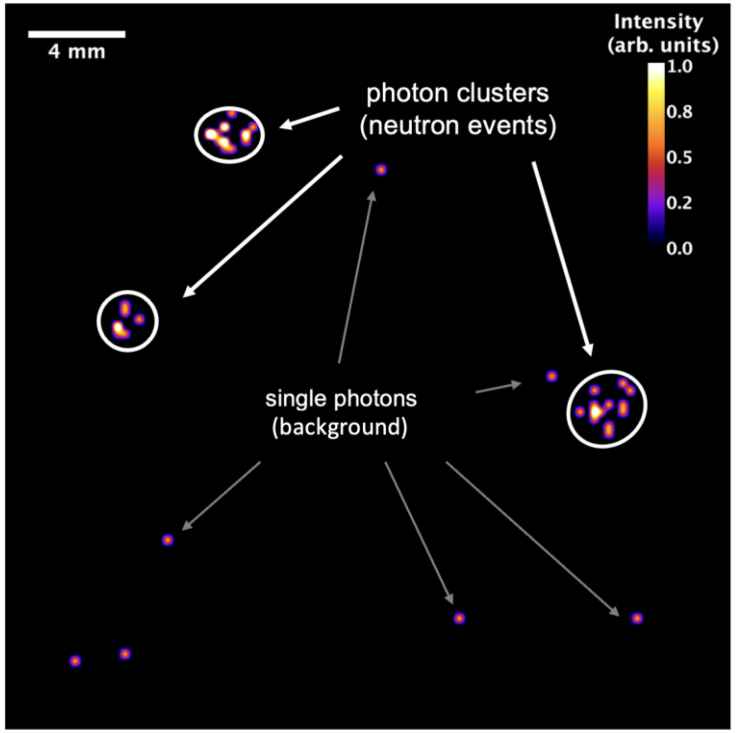
Los Alamos Neutron Science Center

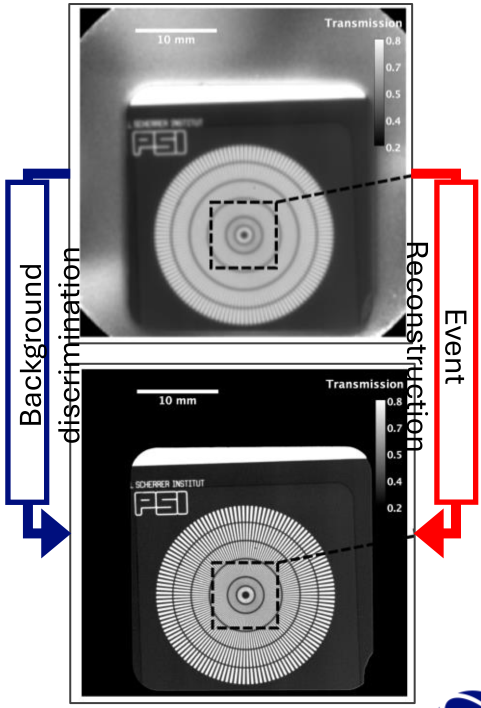
Enabling technology: Compact event-mode imaging cameras

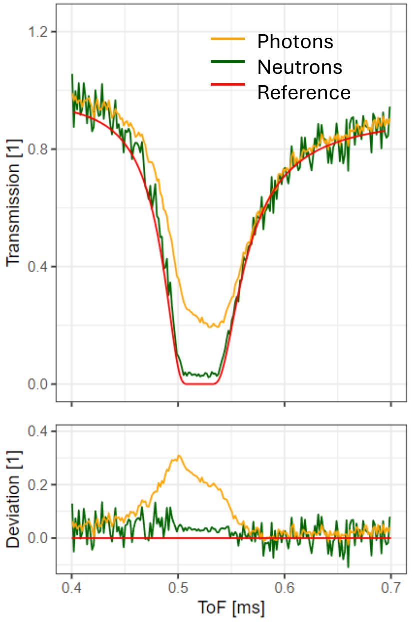
- Event-mode camera analyzes photon output of scintillator, enables neutron/gamma discrimination

Ideal setup for NRI-PIE!

- Proposed ~2020, novel technique
- Improves spatial and temporal resolution
- Gamma rejection makes it ideal tool to characterize radioactive samples such as irradiated fuels
- LANSCE team involved in developing this technique
- Some technique development supported by NSUF IS grant







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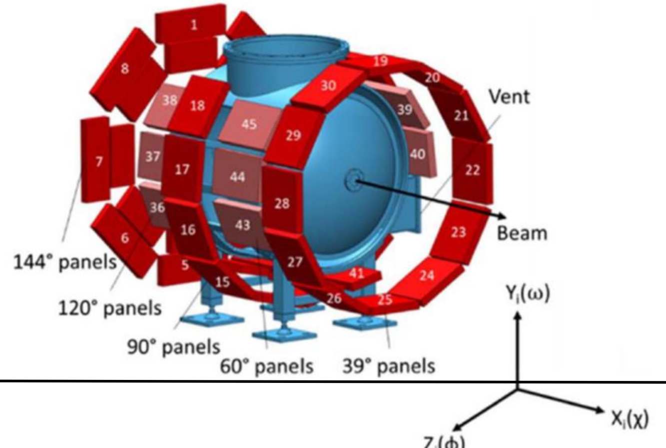
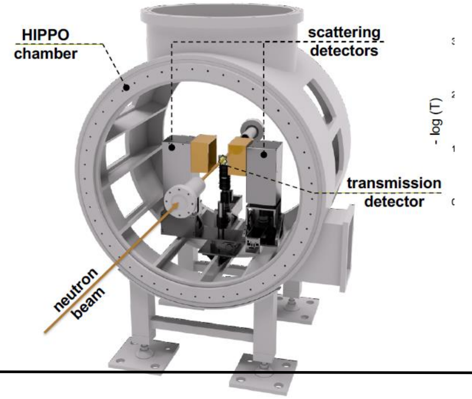
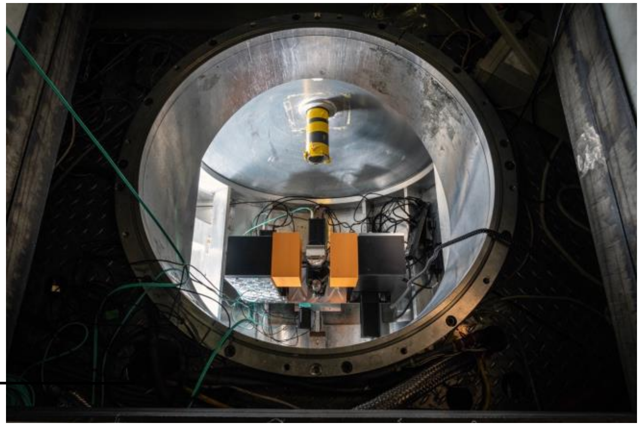
"New perspectives for neutron imaging through advanced event-mode data acquisition" Losko et. al. Scientific Reports (2021)
"LumaCam: A Novel Class of Position-Sensitive Event Mode Particle Detectors using Scintillator Screens" Wolfertz et. al. Scientific Reports (submitted)

Event-mode camera in HIPPO: Combining neutron diffraction with energy-resolved neutron imaging

- TPX3Cam/Event-mode camera first installed in HIPPO December 2022
- Improved version in FY24 with this grant, but only ~10 days of beam time for commissioning and demonstration
- 10mm ∅ HIPPO beam spot large enough for some imaging
  ⇒ Scans for e.g. irradiated fuel also provides Bragg-edge data for spatially resolved lattice parameters (e.g. O density in high burnup UO_2)
- 1,200 He-3 tubes provide simultaneous diffraction data
- Count times for imaging ~50 times longer (one to several hours) than for diffraction (one to several minutes)
- Immediate access to HIPPO sample environments (furnaces, cryostats, sample changer robot etc.)
- While diffraction analysis is mature, energy-resolved neutron imaging requires analysis tools







Master Camera Installers Practicing their Craft (also head-shots of my co-authors)

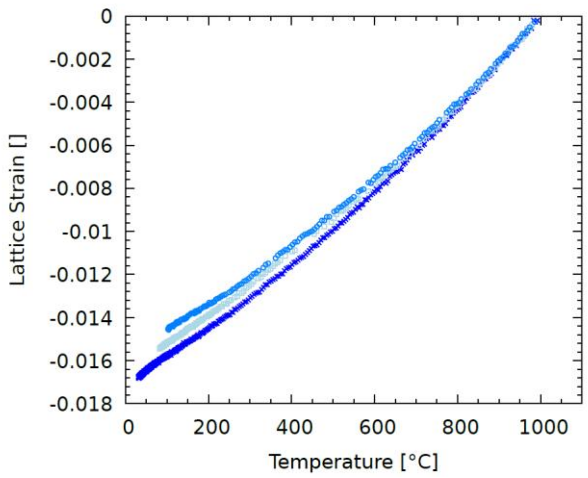
Tsviki Hirsh

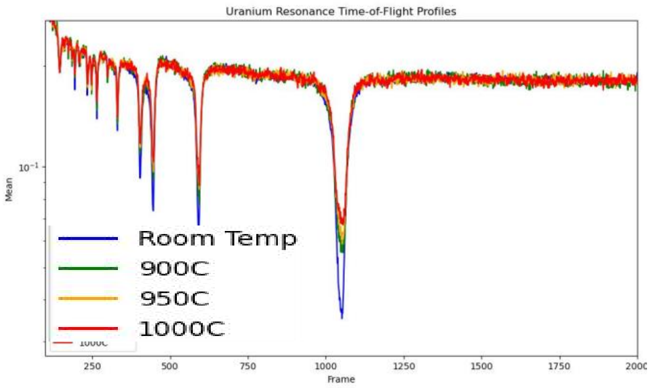
Alex Long

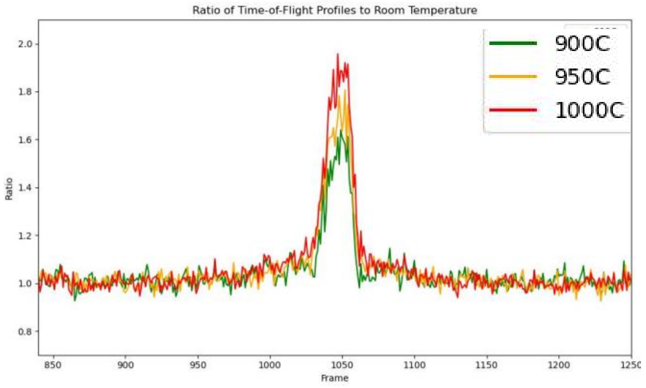
Adrian Losko

Thermal Expansion of U-Mo alloy

- U-Mo, U-Zr, and U-Nb researched at LANL for var. LANL missions
- Diffraction allows to study
  - phase diagrams/phase transformations,
  - ordering processes
  - lattice parameters
- Lattice parameters: density & thermal expansion as a function of temperature
- Three compositions of U-Mo characterized between RT and 1000°C
- High neutron flux & ~20% of 4pi detector coverage
  ⇒ one minute time resolution, 5C/min temperature ramp rate
- Lattice strains relative to lattice parameter measured at 1000°C
  ⇒ three alloys shows differences in thermal expansion behavior,
- Event-mode neutron imaging camera
  ⇒ transmission data simultaneously with diffraction data
  ⇒ Doppler broadening of U-238 neutron absorption resonances could enable sensor-less temperature measurements
  ⇒ Depth of resonances of U-238 and U-235 would allow to map enrichment levels (present samples were depleted uranium)





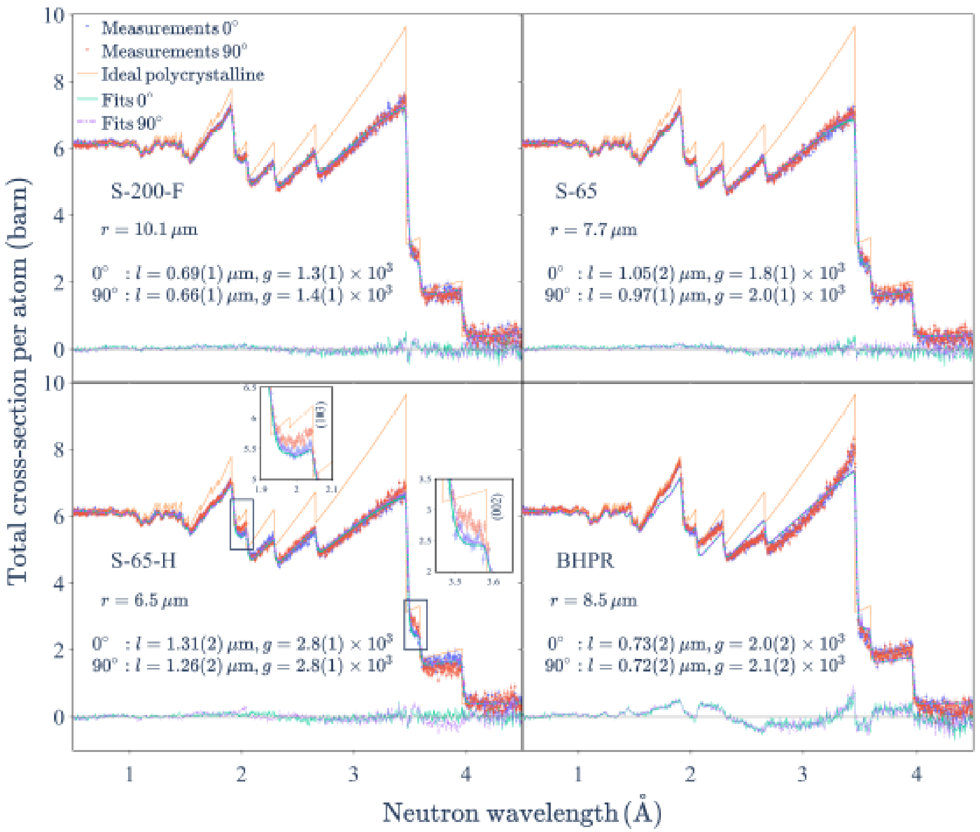


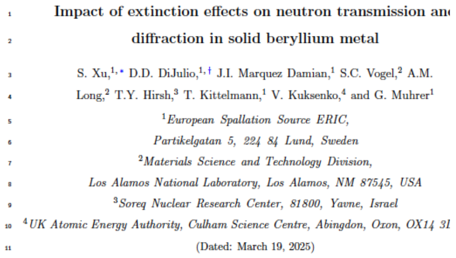
Nuclear Science User Facilities

HIPPO

Micro-structure aware cross-section of Be

- Microstructure affects cross-section of materials in the thermal neutron energy range
- Ability to measure BOTH microstructure AND cross-section allows to rule out e.g. texture as cause for discrepancies
- Added extinction to NCRYSTAL code that can be plugged into MCNP calculations for thermal cross-sections
- For neutron reflector applications, but can be applied to cladding, structural materials etc.
- Collaboration with European Spallation Source, Lund, Sweden
- Paper under review with J. Appl. Cryst.




Impact of extinction effects on neutron transmission an

diffraction in solid beryllium metal

S. Xu,[1,*] D.D. DiJulio,[1,†] J.I. Marquez Damian,[1] S.C. Vogel,[2] A.M. Long,[2] T.Y. Hirsh,[3] T. Kittelmann,[1] V. Kuksenko,[4] and G. Muhrer[1]

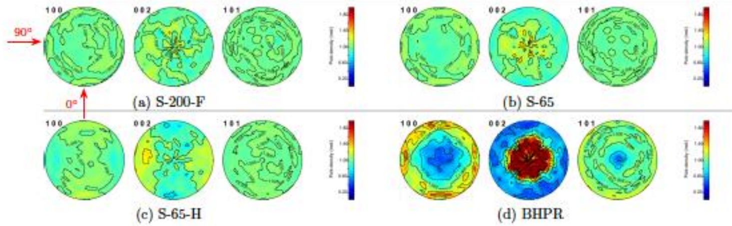
[1]European Spallation Source ERIC, Partikelgatan 5, 224 84 Lund, Sweden

[2]Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

[3]Soreq Nuclear Research Center, 81800, Yavne, Israel

[4]UK Atomic Energy Authority, Culham Science Centre, Abingdon, Oxon, OX14 3I

(Dated: March 19, 2025)



Other relevant application examples

- Grain orientation mapping in large-grained reactor pressure vessel or primary cooling loop pipe steel to inform acoustic inspection methods
- Cross-section of hydrazine above and below freezing point for satellite-based neutron detectors (diffraction provides evidence of freezing and integrity of sample)
- Cross-section and texture of HEU uranium fuel for NATRIUM
- Interest expressed to measure thermal cross-sections above and below ambient conditions (e.g. solid-state moderator materials, fuels for space reactors)
- NSUF proposals that will benefit from this capability:
  - RTE "Isotope density mapping using Energy Resolved Neutron Resonance Imaging of a High Burnup UO2 Fuel Fragment " (fuel from H.B. Robinson reactor, 23-4584, funded, waiting for beam time)
  - CINR on Fast Flux Test Facility irradiated steel (CINR 24-31447, funded)
  - CINR "Post-Irradiation Examination (PIE) of BR2-Irradiated SiC-Based Accident-Tolerant Fuel (ATF) Cladding Materials & Constituents Thereof (POSEIDON)" (25-34175, submitted)
  - CINR "Title: Effect of neutron irradiation on NF616 (Grade 92) at LWR and fast reactor relevant temperatures " (25-34154, submitted)

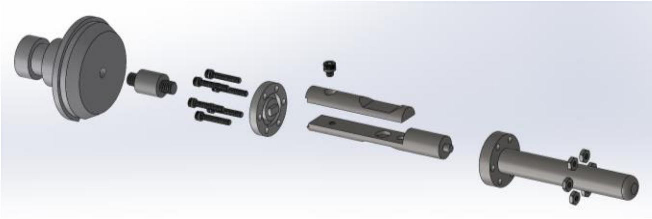
Update on high burnup fuel characterization

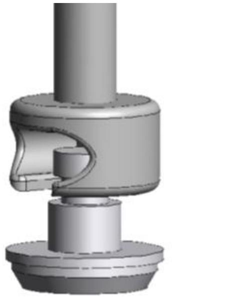
- 23-4584, PI Will Cureton, collaboration between ORNL, INL, LANL
- Funded, but delayed because sample holder needed re-design and 2024 LANSCE run cycle was cancelled
- Issues with previous sample holder for irradiated U-10Zr-1Pd (20-2958):
  - Sample turned out to be fully oxidized (no alpha-U detected)
  - Pathway for oxygen to reach sample could also provide pathway for contamination to leave containment
  - Sample holder made from aluminum contributed greatly to diffraction signal (especially for 55% enriched sample)
  - Heating not possible because spacers and adapter for robotic sample changer were also made from aluminum
- Improved sample holder:
  - Sample sealed in vanadium can and in holder (ORNL design)
  - Replacement of aluminum with vanadium will reduce diffraction signal
  - One less screw for hot cell operators
  - No more aluminum parts, sample can be heated, devised furnace adaptor allowing remote handling

  $\Rightarrow$ After room temperature characterization of HBU sample under existing RTE a follow-up RTE will be submitted for high temperature structure changes in irradiated fuel (up to 800C)

Old

New





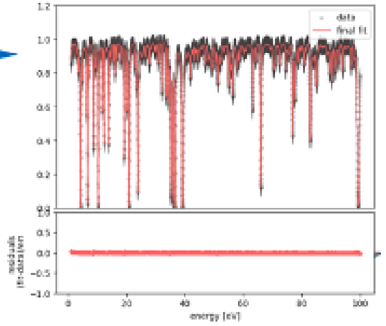
Software Development & Publications

- Sofware:
  - PLEIADES: Resonance analysis
  - Lead by LANL in collaboration with other neutron sources, funded by NSUF
  - Contributed to several others such as nBragg (Bragg-edge analysis, Soreq/Israel) and NCrystal (thermal cross-sections, Danish Technical University), MAUD (diffraction analysis, Unversity of Trento, Italy)
- Publications:
  - 2 Publications almost in print
  - 1 Publication close to submission

PLEIADES

Python Libraries Extensions for Isotopic Analysis via Detailed Examinations of SAMMY results

docs passing DOI 10.5281/zenodo.1414282

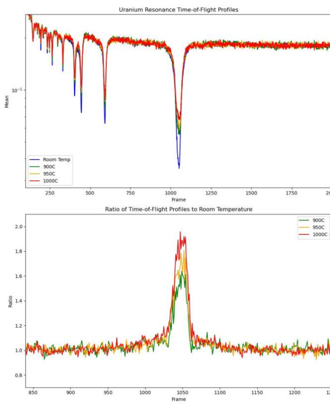


nature SCIENTIFIC REPORTS — **Energy-Resolved Neutron Imaging and Diffraction including Grain Orientation Mapping Using Event Camera Technology** — Tsviki Y. Hirsh[1], Andrew F.T. Leong[2], Alexander M. Long[2], Adrian S. Losko[3], Alexander Wolfertz[3], Daniel J. Savage[2], Tim T. Jäger[4], John Rakovan[5], and Sven C. Vogel[2] — Accepted

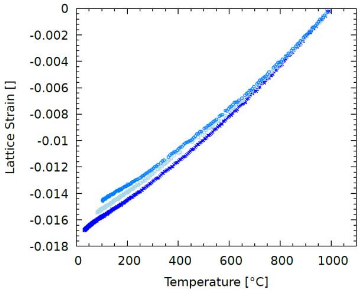
nature SCIENTIFIC REPORTS — **Large Field-of-View Event-Mode Camera for High-Precision Epithermal Neutron Resonance Imaging** — Tsviki Y. Hirsh[1], Alexander M. Long[2], Adrian S. Losko[3], Tim T. Jaeger[4], Alexander Wolfertz[3], and Sven C. Vogel[2] — Accepted

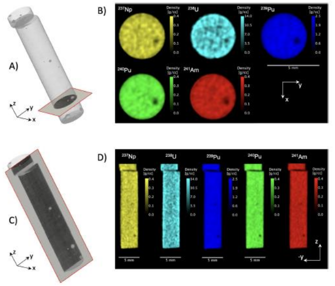
JOSS The Journal of Open Source Software — **PLEIADES: Python Libraries Extensions for Isotopic Analysis via Detailed Examination of SAMMY results** — Alexander M. Long[1], Tsviki Hirsh[2], Jean Bilheux[3], Adrian Losko[4], Chen Zhang[3], and Yuxuan Zhang[3] — In Prep.

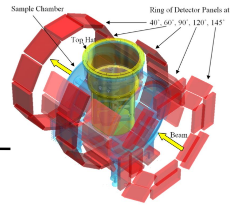
Conclusions

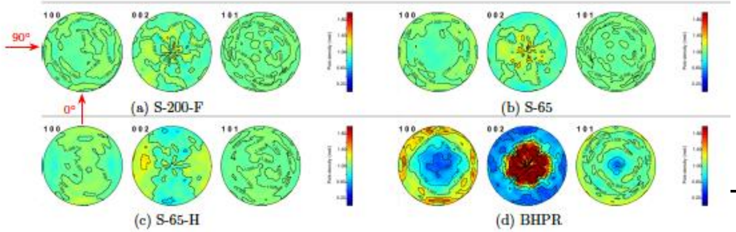


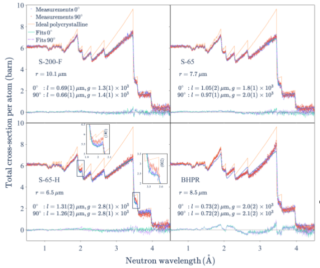
- Simultaneous characterization with diffraction and energy-resolved neutron imaging (resonances, Bragg-edges, inelastic) is a powerful characterization tool
- Count times will be dominated by imaging if mapping desired, similar for global cross-section
- For cross-section measurements, knowledge about microstructure (texture, phase composition etc.) is often very useful
- For non-ambient condition measurements, energy-resolved neutron imaging adds information to e.g. spatial distribution of decomposition reaction in hydrides or as a diagnostic for sensor-less bulk sample temperature measurements
- Both atomic displacement parameters in diffraction analysis and Doppler-broadening of resonances depend on thermal motion of atoms predictable by e.g. DFT or MD calculations => Combination could be a powerful technique to benchmark DFT at non-ambient conditions
- Python-based data analysis pipelines for both Bragg-edge and resonance fits using NCrystal (nBragg) and SAMMY (PLEIADES), respectively, are very powerful
- Funding from NSUF for this effort is gratefully acknowledged!



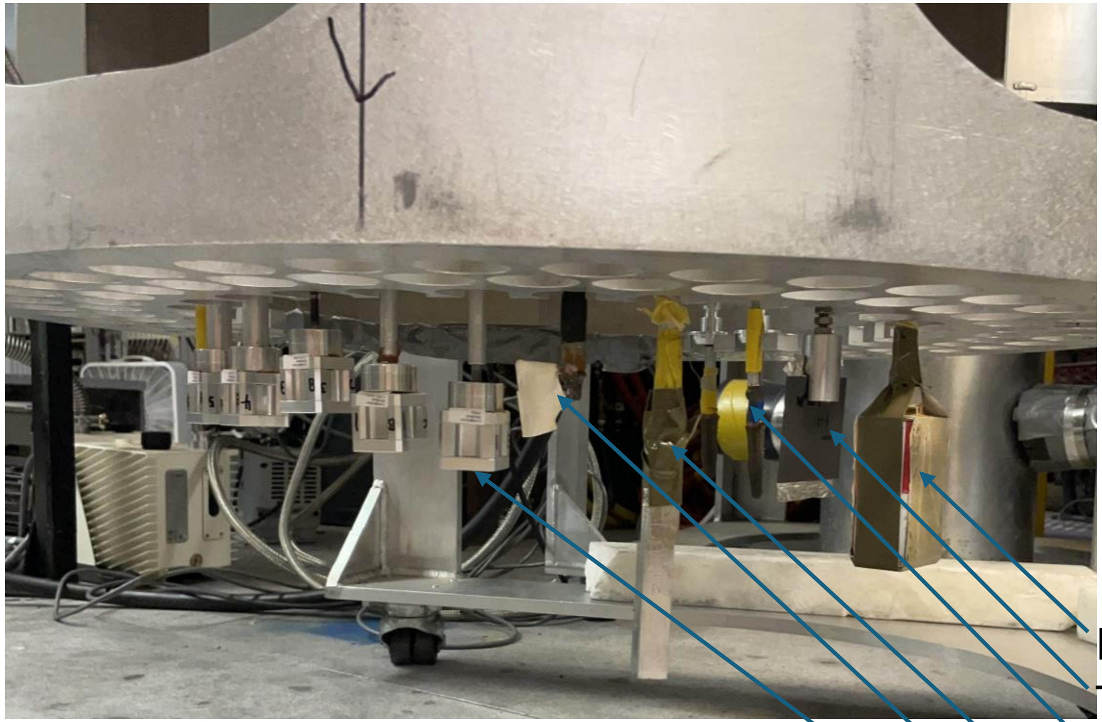








HIPPO workhorse sample environments: Robotic sample change & ILL furnace

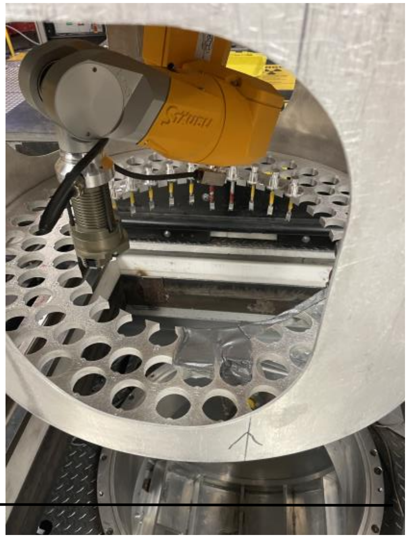


Fe powder
Ta sheet
dU cylinder
Large grained steel
Natural silver
ESS beryllium

A lot of the data presented here collected in one shift! (time was limited due to end of run cycle looming…) ⇐

Imaging on HIPPO benefits from existing sample environments

Beam pipes of furnace and cryostat require larger distance of camera from sample than for robotic sample changer (exit pipes could be modified)



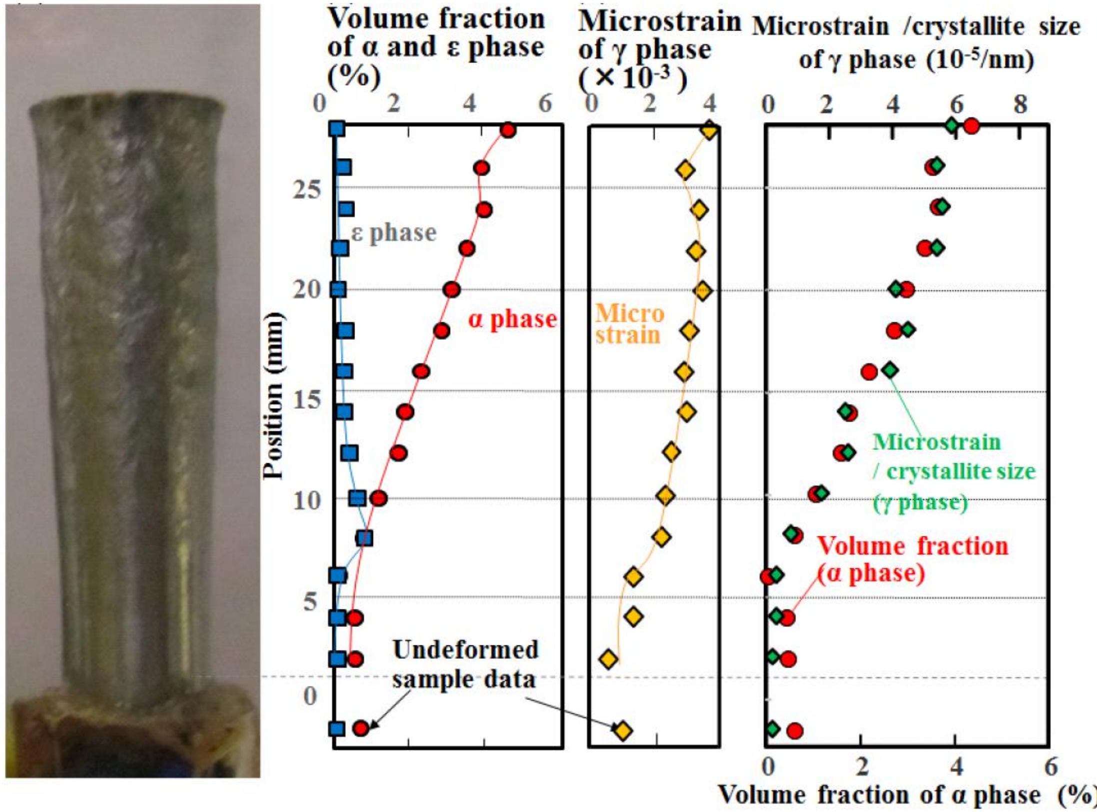
LANSCE
Los Alamos Neutron Science Center

Los Alamos
NATIONAL LABORATORY

Cd slit allows “spatially-resolved” diffraction => Characterization in ~2mm slices

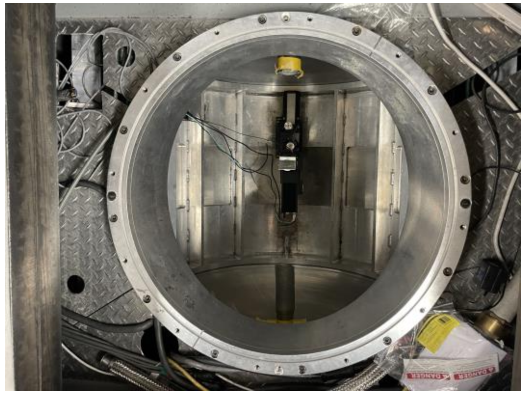
Caliber 7.62 mm additively manufactured steel cylinder was driven into the wall at 235 m/s into an anvil in the Taylor Anvil Gas Gun Facility at LANL. Microstructure as a function of distance from impact studied with HIPPO

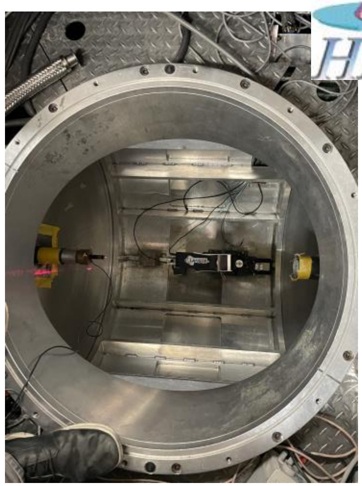
Cd slit in front of sample position



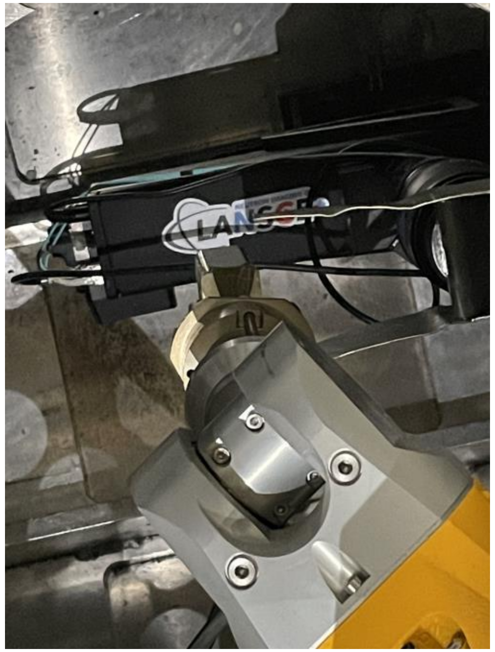
S. Takajo et al., J. Powder Diffraction, 2018

Camera in HIPPO – A few observations





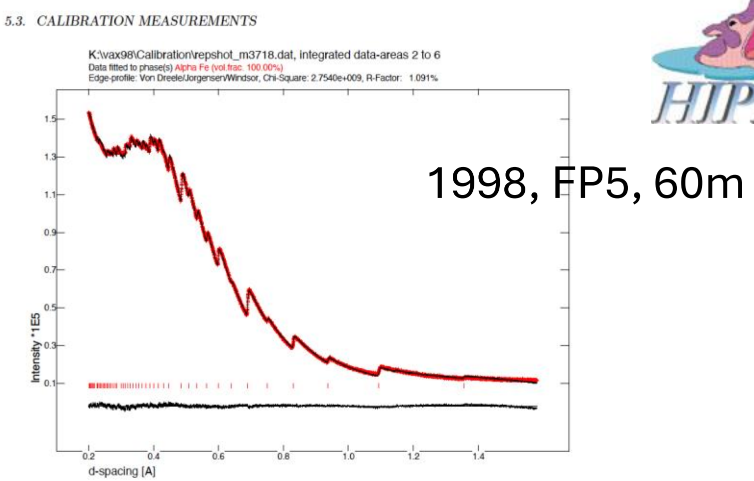
- Any polycrystalline camera components in the beam cause background for the diffraction signal
  => need to get rid of aluminum in mirror and scintillator backplate
- Any upset requiring reboot of the camera requires removal of sample environments by crane, requiring in turn possible new empty instrument runs etc.
  => need to be able to reboot camera remotely
- Need motion control to align camera with beam spot, move along beam for robot and furnace/cryostat beam positions
- Camera operation is fully integrated with HIPPO experiment automation, diffraction and ERNI data receive same run number, runs are started and ended together, camera is treated like a sample environment WRT experiment automation

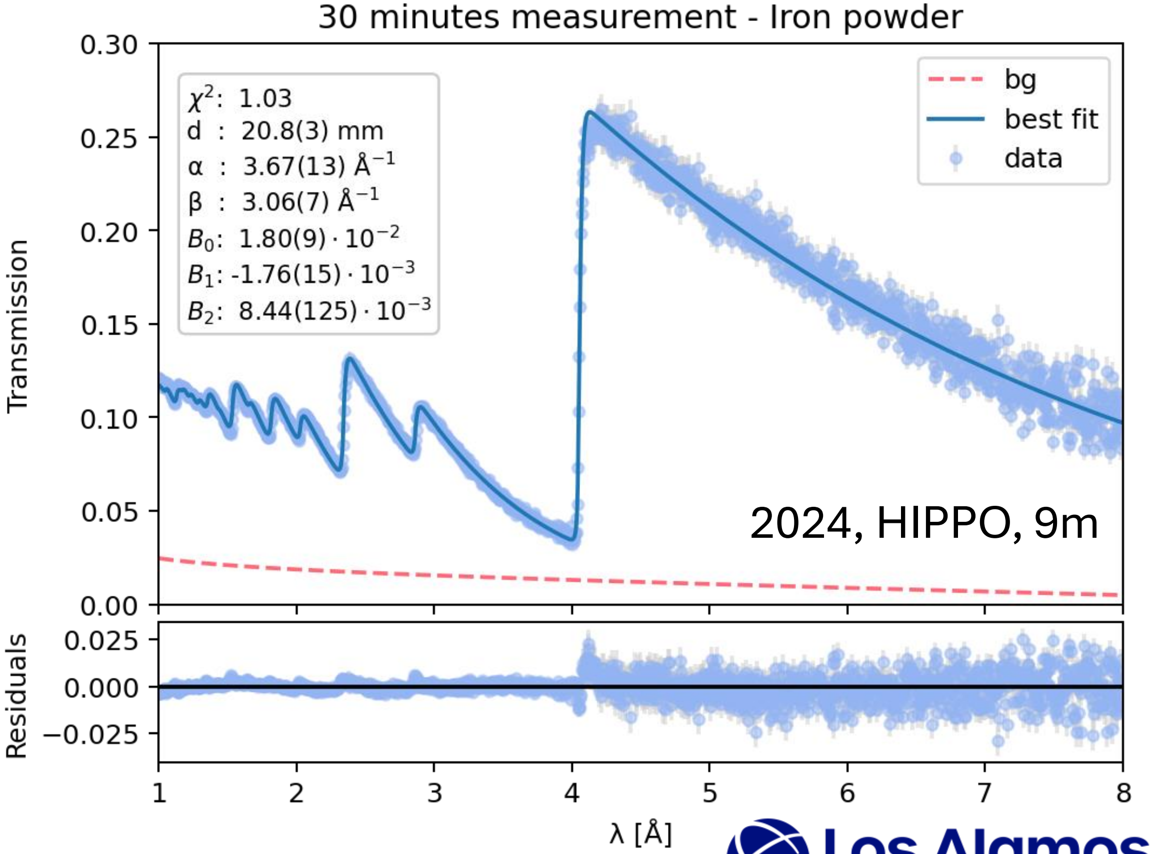


T. Hirsh et al., submitted to Nature Scientific Reports (2024)

Iron powder

- Same iron powder used in 1998 for first Rietveld fit of a Bragg-edge pattern with BETMAn
- All transmission data analysis by Tsviki Hirsh using NCrystal, jupyter-notebooks, and python
- Low background
- Resolution comparable with adjacent FP5
- Resulting sample thickness of 20.8(3)mm compares well with the ~25mm thick container given that powder is loosely packed
  => Physics model for Bragg-edge analysis is reasonable

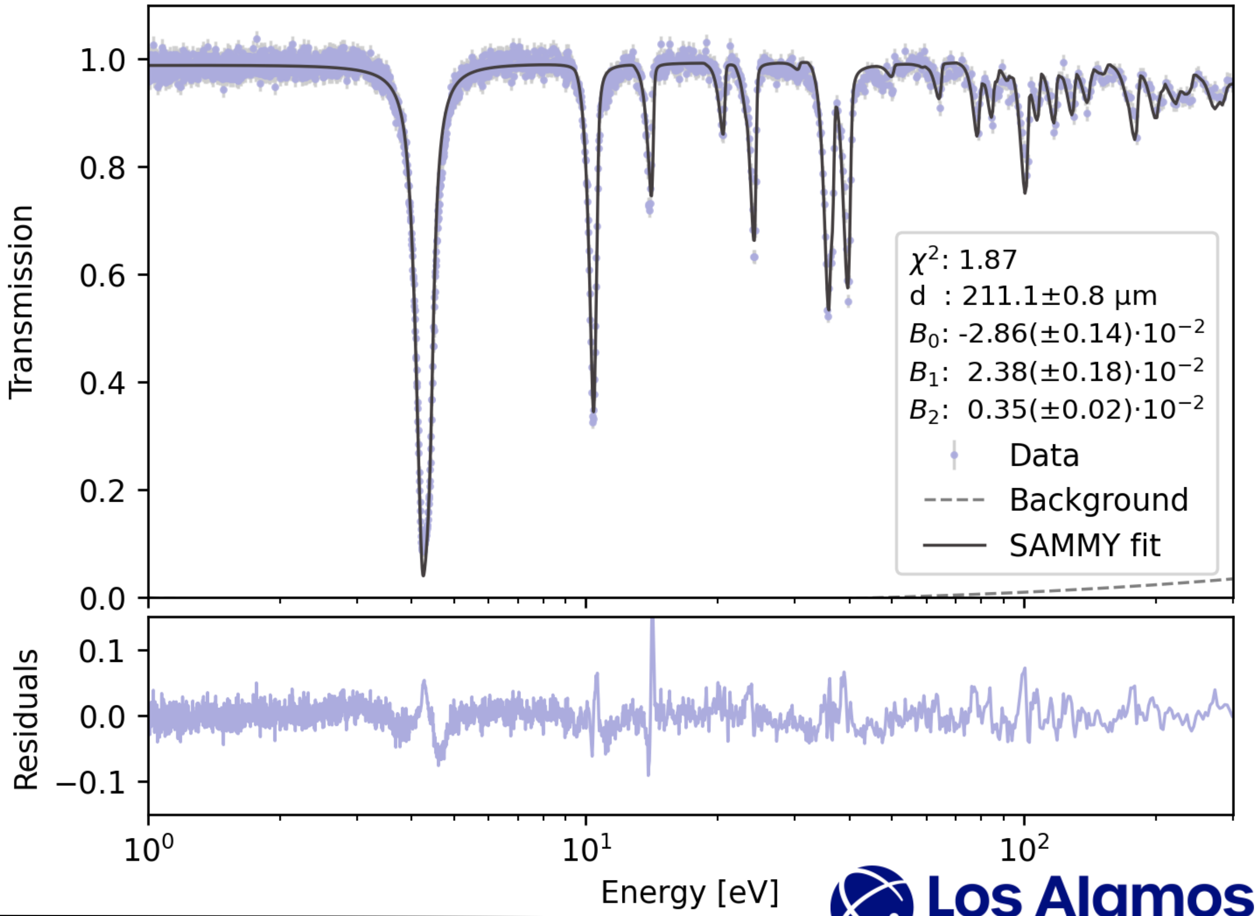




T. Hirsh et al., submitted to Nature Scientific Reports (2024)

Ta foil for resonance

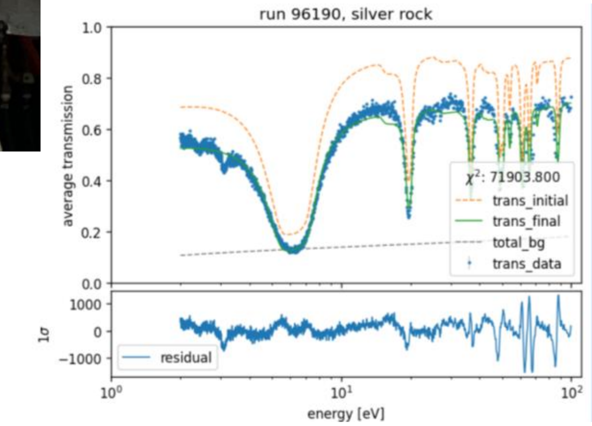
- Ta resonances match up to ~300 eV with a 30 minute count time!!!
- Background extremely low => benefit over FP5 due to incident collimation of HIPPO at the cost of a <14 mm diameter beam spot
- Resulting sample thickness from resonance analysis is within 5% of micrometer-measured sample thickness

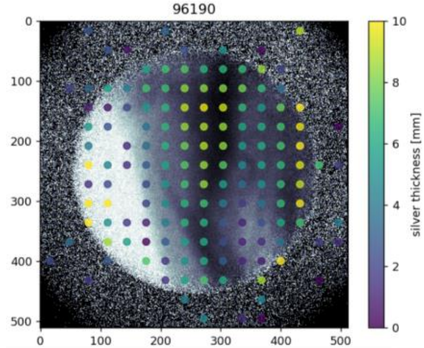


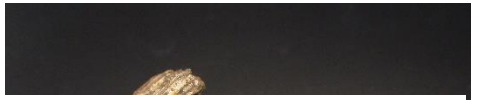
T. Hirsh et al., submitted to Nature Scientific Reports (2024)

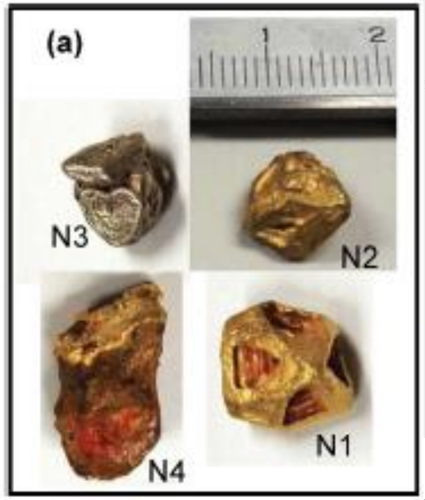
Natural Silver Sample

- Mineral specimens are often too precious for destructive characterization by other techniques and benefit from neutron characterization
- Neutron diffraction provides e.g. phase composition and texture
- Natural silver specimen from the collection of the New Mexico Bureau of Geology & Mineral Resources' Mineral Museum at the New Mexico Institute of Mining and Technology was provided by New Mexico S John Rakovan
- Goal was to characterize the texture of a wir to compare with previously characterized sil
- Size of the sample is ~4cm, non-wire parts m masking tape painted with Gd_2O_3 paint, 10 m
- Diffraction data allows to quantify texture (n orientation detected) and identify additional
- Neutron absorption resonance data allows to thickness (ultimately tomography of sample) presence of impurities of elements with neutron absorption resonances



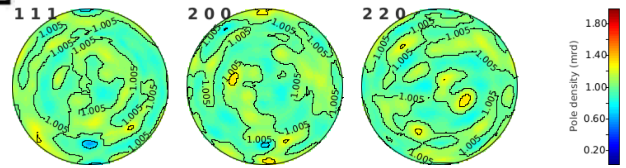






Non-Destructive Study of Bulk Crystallinity and Elemental Composition of Natural Gold Single Crystal Samples by Energy-Resolved Neutron Imaging

Anton S. Tremsin[1], John Rakovan[2], Takenao Shinohara[3], Winfried Kockelmann[4], Adrian S. Losko[5] & Sven C. Vogel[5]



T. Hirsh et al., submitted to Nature Scientific Reports (2024)