Characterization of Irradiated Nuclear Fuels with Pulsed Neutrons – including LDNS

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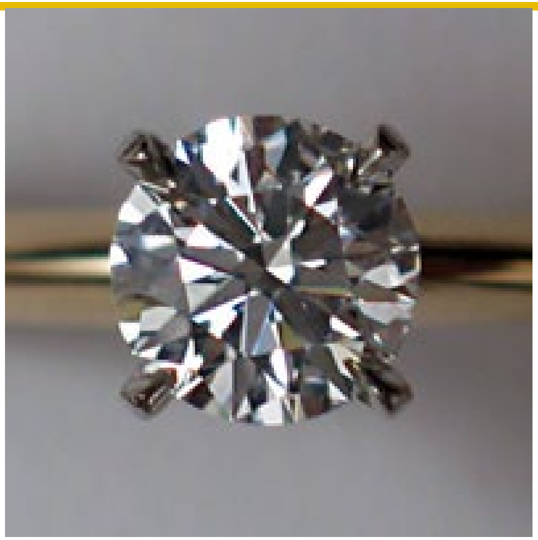
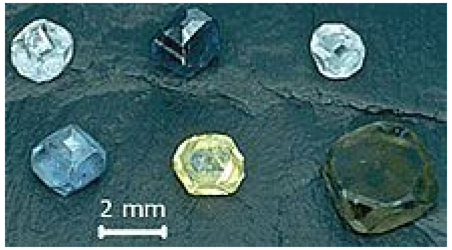
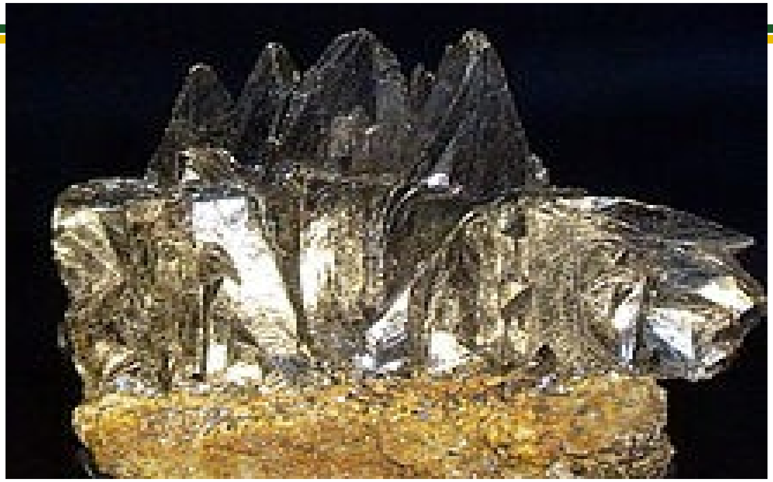
LA-UR-22-29701

Audience participation: What is this?

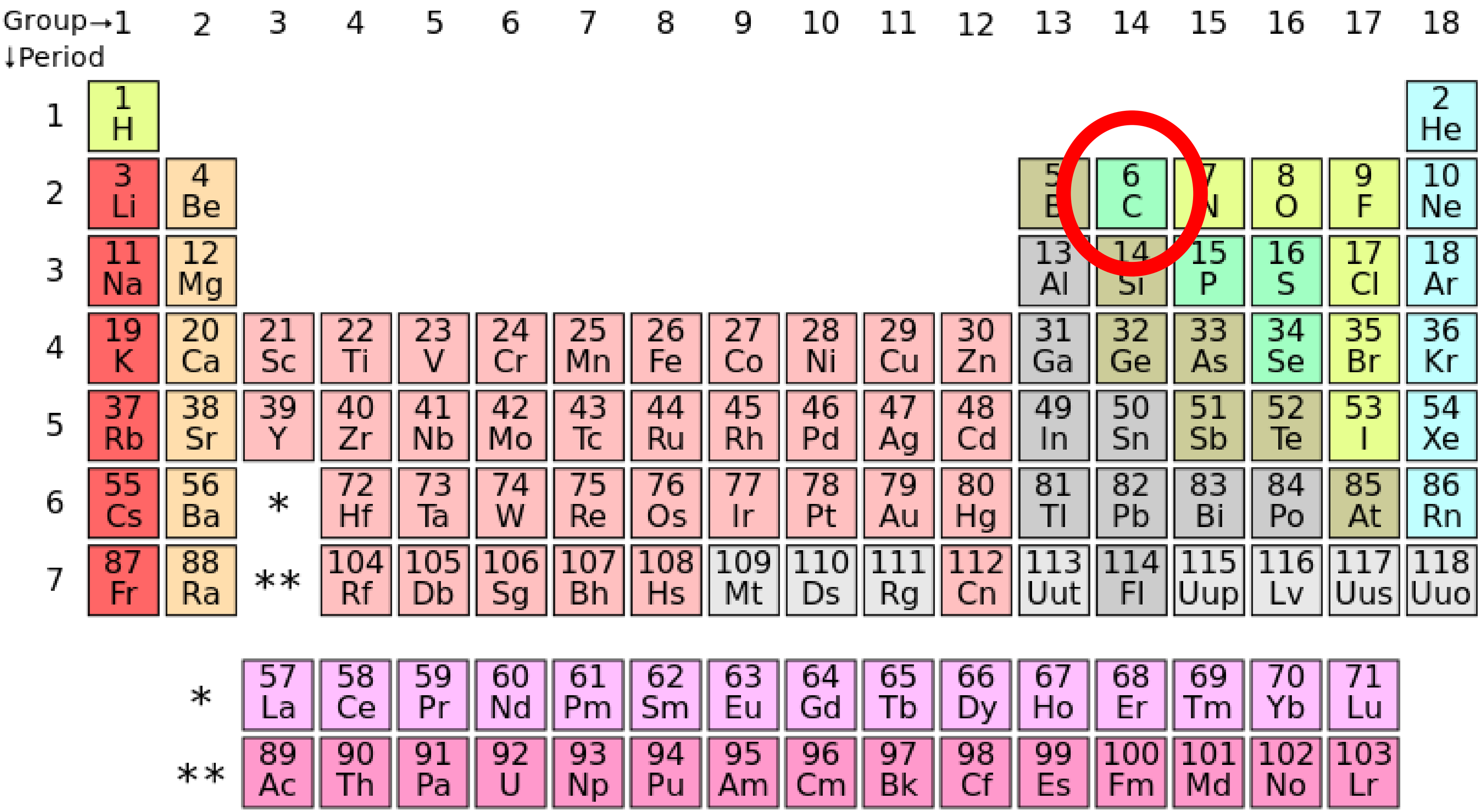




And what is this?

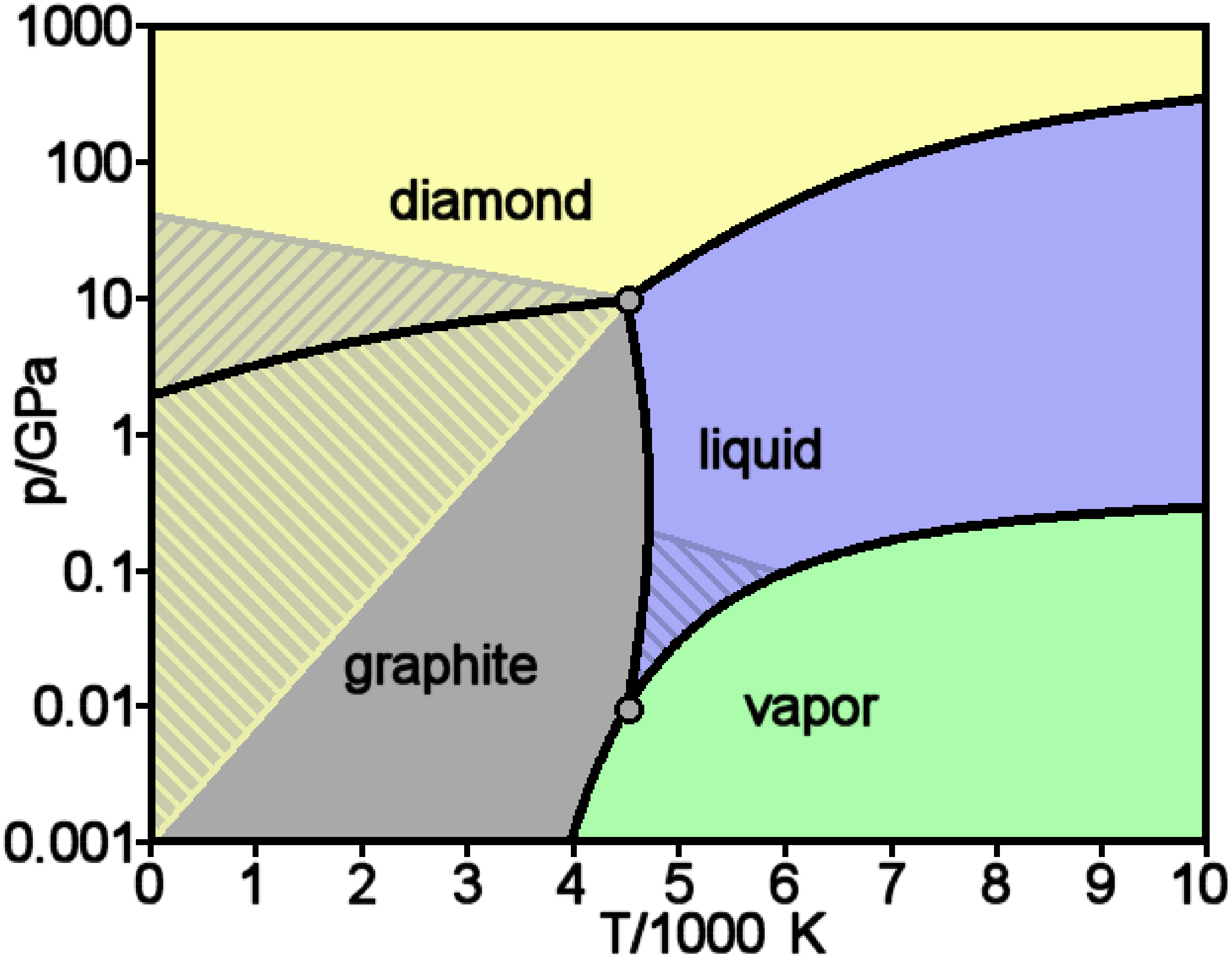


What do diamonds and graphite have in common?



Group→1 ↓Period	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	*	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	**	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Fl	115 Uup	116 Lv	117 Uus	118 Uuo
		*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu	
		**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr	

When is carbon graphite and when is it a diamond? Phase diagram!

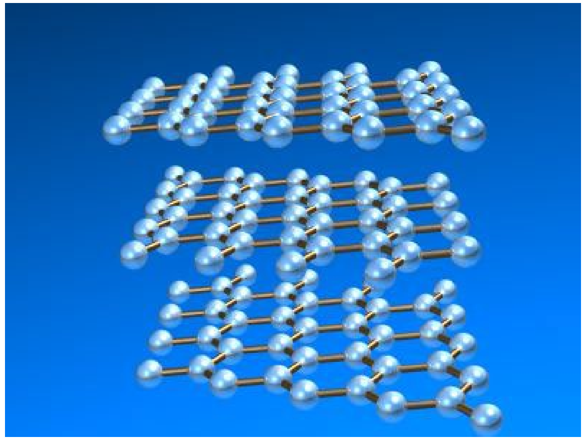


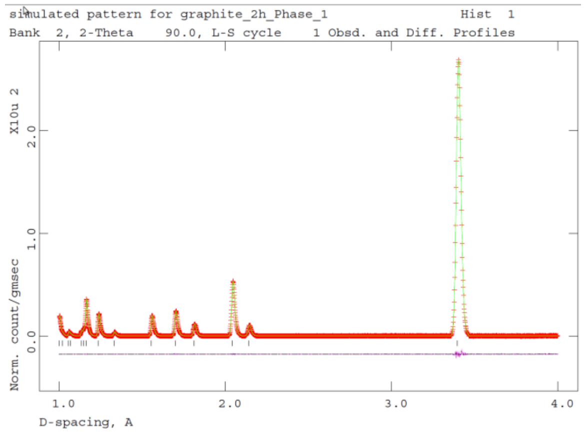
Viewing Atoms: Carbon
Crystal structure is like a finger-print!

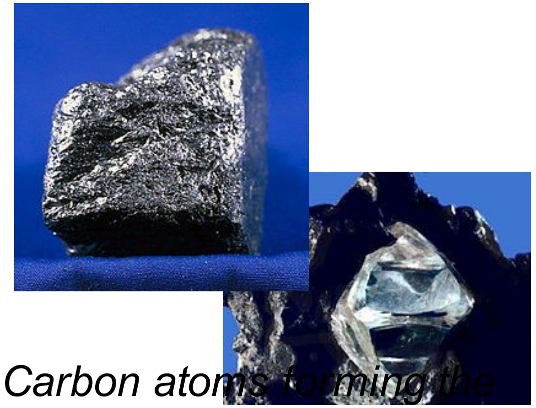
Arrangement of the atoms (the crystal structure)

Viewed with neutrons in a diffraction experiment

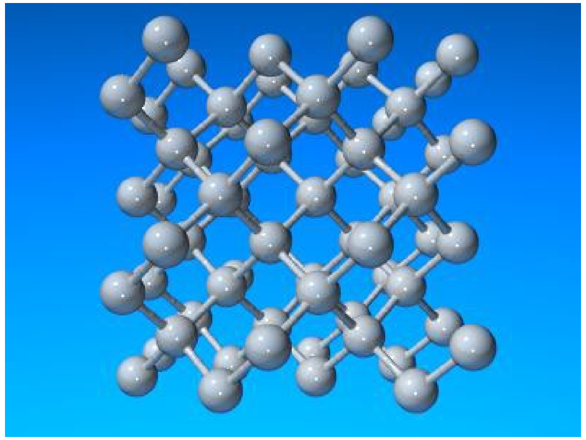
Carbon atoms forming the graphite structure (graphite is soft because of its crystal structure)



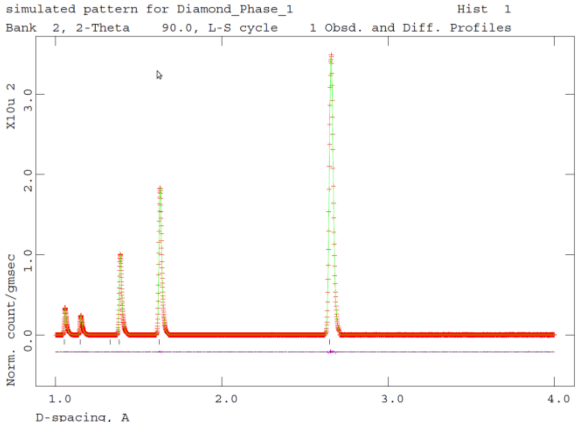




Carbon atoms forming the diamond crystal structure (diamond is hard because of its crystal structure)

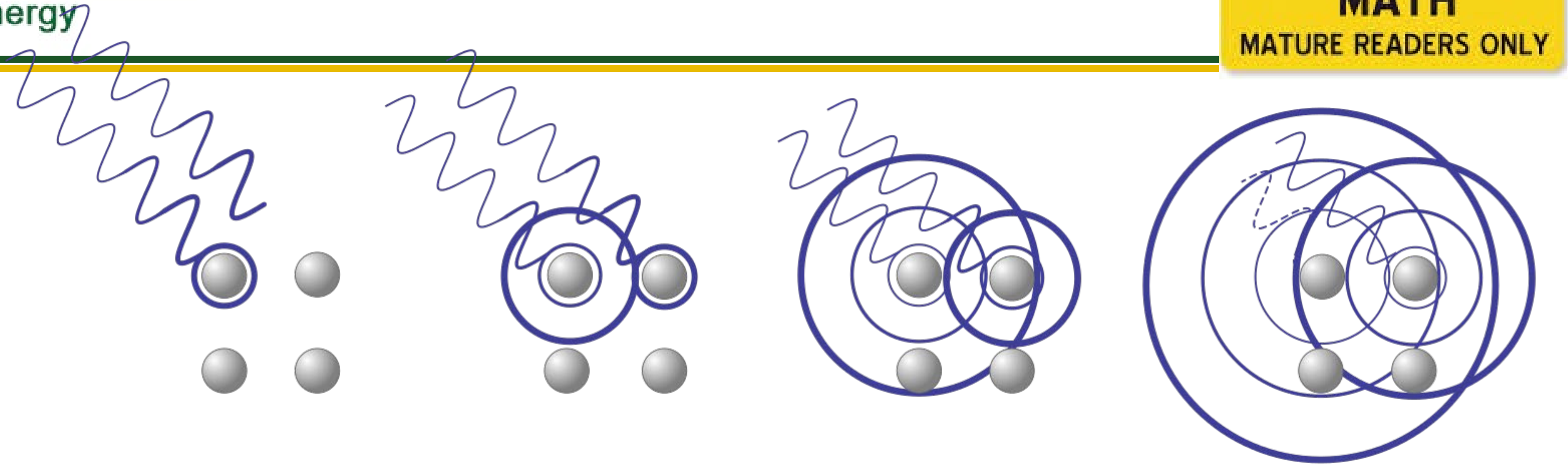


Crystal structures from www.webelements.com



How does diffraction work?

CAUTION
CONTAINS MATH
MATURE READERS ONLY



We need a wavelength! Neutrons are small enough particles to have both wave and particle properties, therefore there are two ways to write the momentum of a neutron:

$$p_{quantum\,mechanics} = \hbar k = \frac{h}{2\pi}\frac{2\pi}{\lambda} = \frac{h}{\lambda} \text{ and } p_{classical\,mechanics} = mv = m\frac{L}{t}$$

$$p_{quantum\,mechanics} = p_{classical\,mechanics} \text{ and therefore } \frac{h}{\lambda} = m\frac{L}{t} \Leftrightarrow \boxed{\lambda = \frac{ht}{mL}}$$

Lattice spacing of a diffracting crystal lattice plane

Bragg's law must be fulfilled:

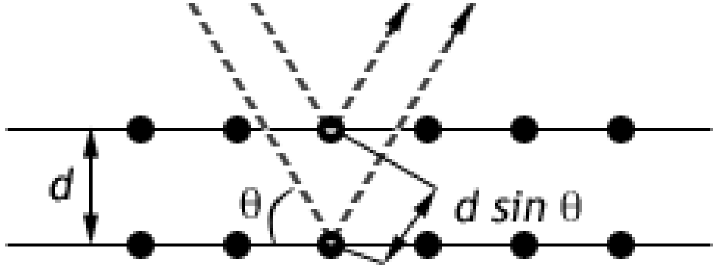
$$\lambda = 2d \sin(\vartheta) = ht/mL$$

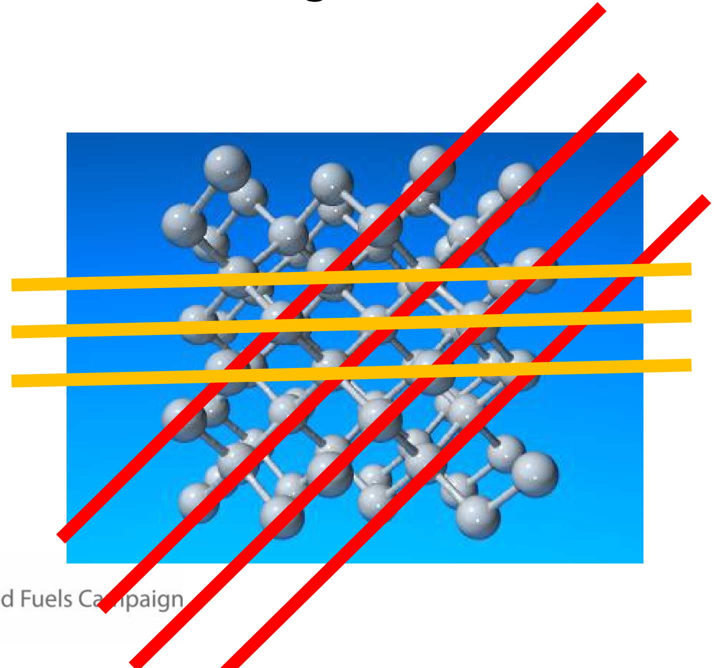
$$\Leftrightarrow$$

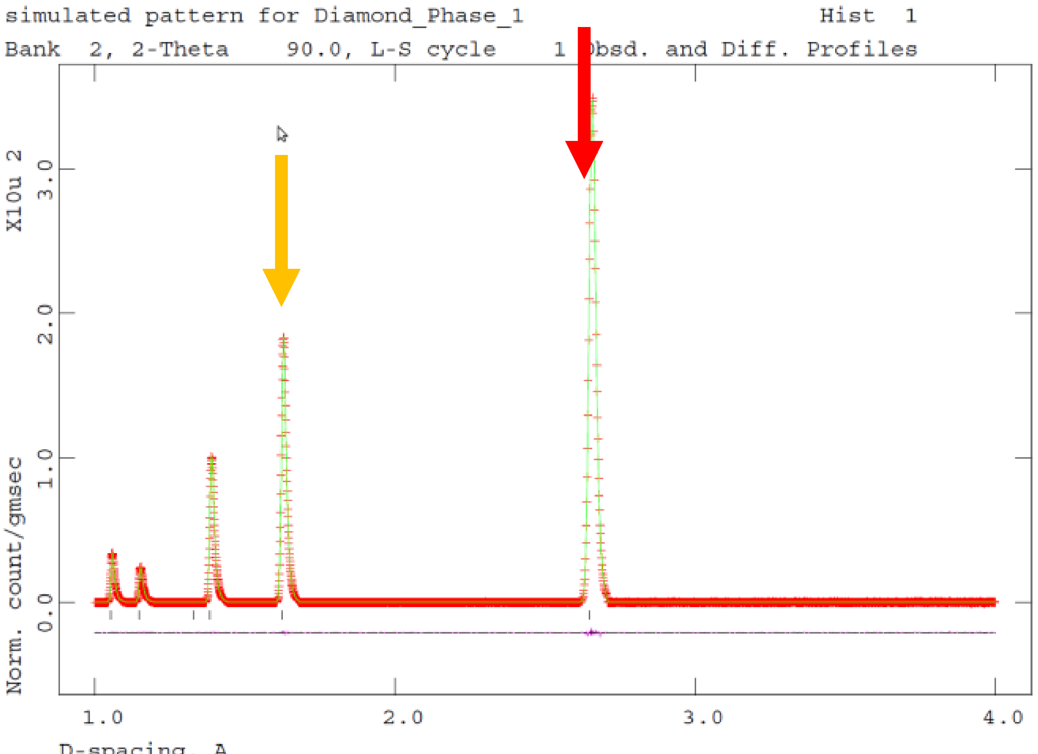
$$d = t \times h \,/\, 2mL \sin(\vartheta)$$

$$d = t \times const$$

Time-of-flight diffraction

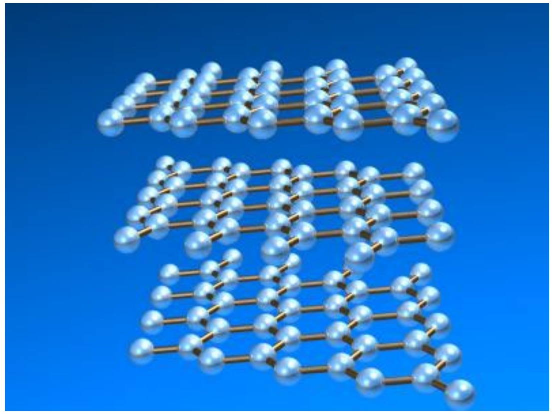


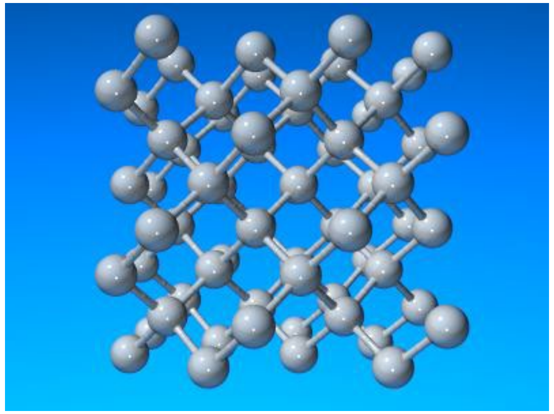




Neutron Diffraction for Material Characterization

- *Diffraction ⇒ Accessible quantities are*
  - *Crystal structure (space group, atomic positions, thermal motion)*
  - *Volume fractions of phases, establish phase transition P/T*
  - *Lattice strains*
  - *Texture*
  - *Dislocation types & densities*
- *Deep penetration ⇒ Sample environments possible*
  - *furnaces*
  - *load frames*
  - *pressure cells etc.*
- *~cm^2 beam-spot ⇒ Bulk probe*
  - *information averaged over ~1 cm^3*
  - *large grained materials*
  - *good grain statistics*
- *Scattering power depends on isotope ⇒ Different contrast than X-rays*
  - *Crystallography of systems consisting of atoms practically indistinguishable with X-rays*
  - *Crystallography of systems consisting of high and low Z-number elements*





Vogel, "A Review of Neutron Scattering Applications to Nuclear Materials" (2013)

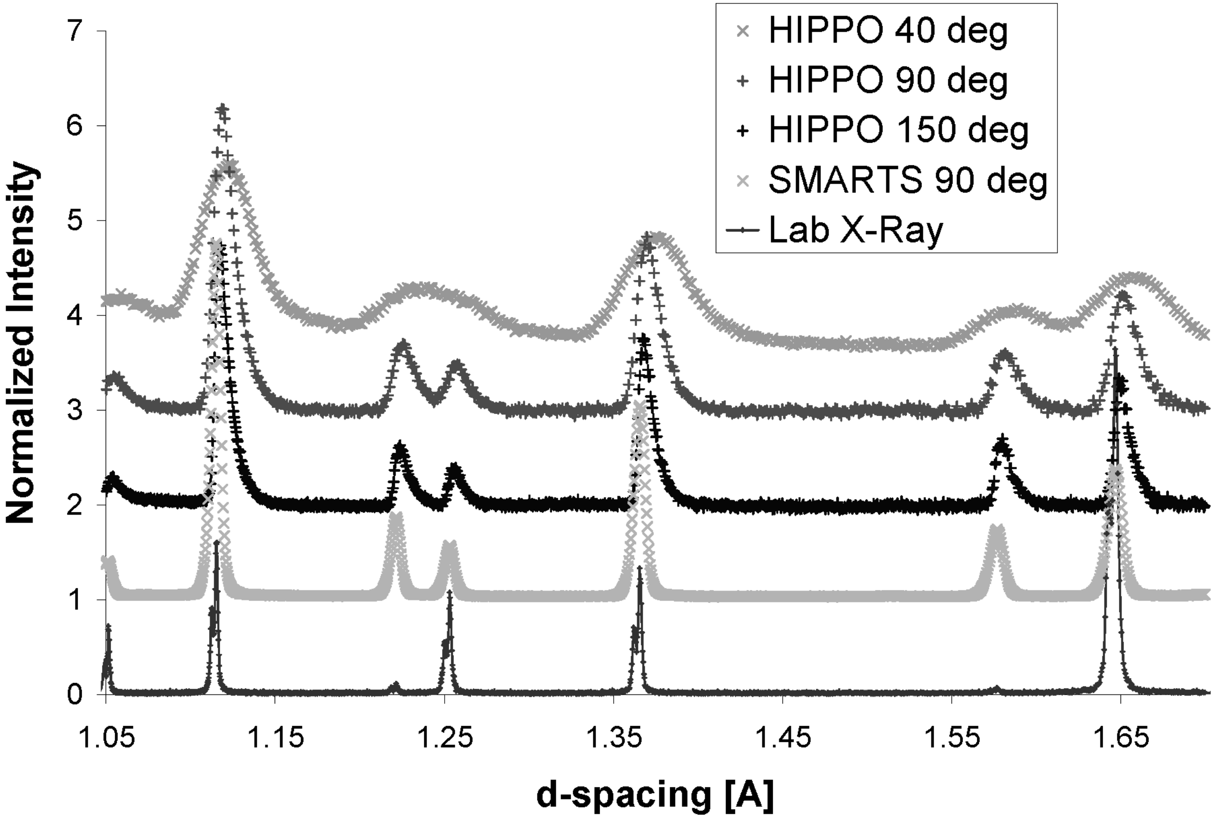
Any laboratory X-ray source is better than neutrons!

...in some aspects only, of course:

- *XRD flux >10^9 is higher than 10^7 particles per cm^2 and second on HIPPO*
- *Resolution (peak width) of standard X-ray machine is better than that of even high-resolution neutron machines*

⇒ Make sure that neutrons are a good choice!

⇒ Combined refinement of XRD & ND can be beneficial!



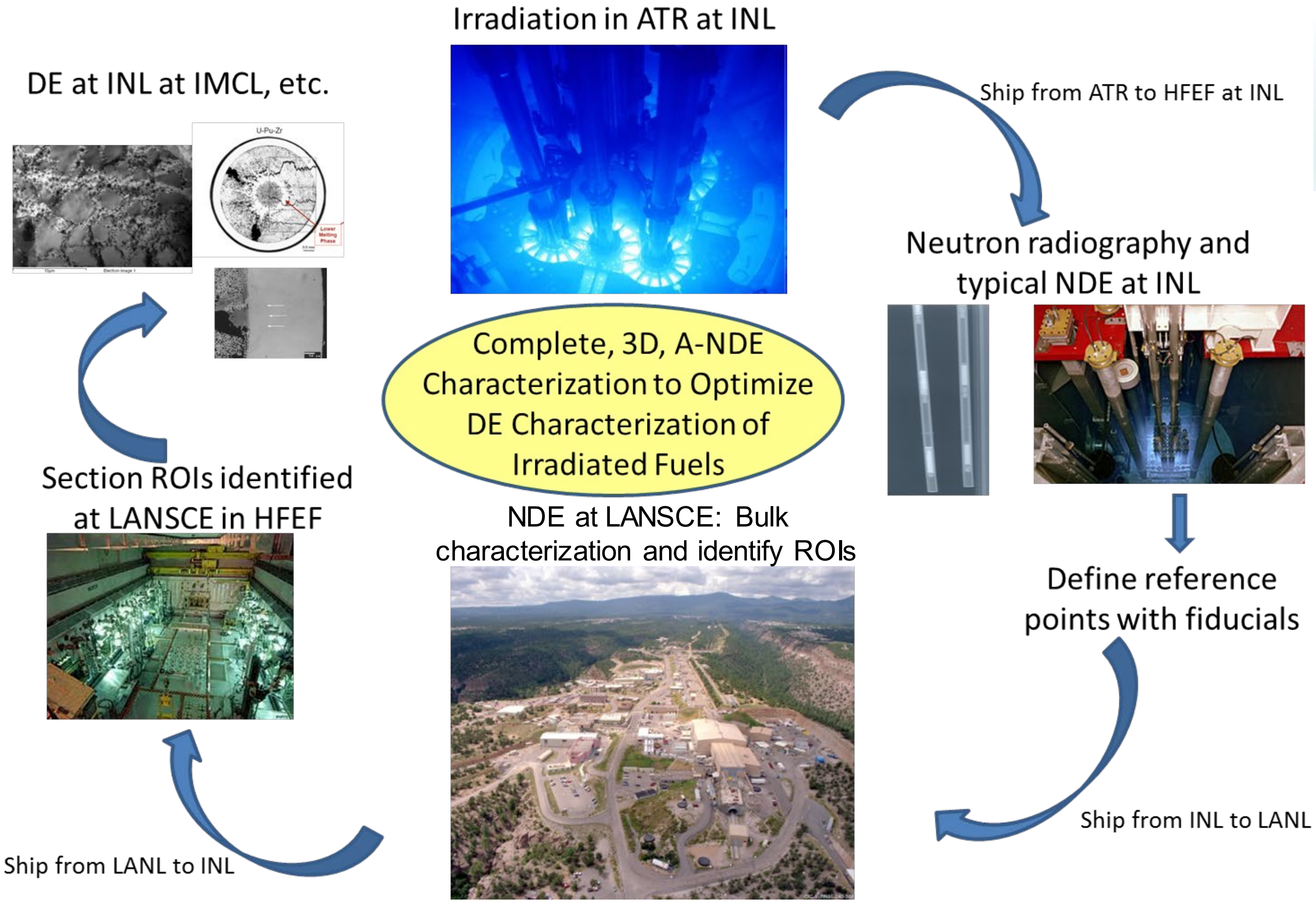
Summary: Why use neutrons for material characterization?

Development of Advanced Fuels

- Understanding irradiation behavior of nuclear fuel is of paramount importance for safe reactor operation
  ⇒ key for licensing of new fuel forms and reactors
- Irradiation tests are done on a few cm^3 of fuel contained in steel irradiation capsules in reactors such as the Advanced Test Reactor (ATR) at Idaho National Laboratory
- Irradiation tests can last up to multiple years
  ⇒ samples are expensive to produce
- Depending on initial isotopes and irradiation duration, a few cm^3 of fuel can emit 900 R/hr
- Destructive post-irradiation examination (PIE) in hot cells provides mm^3 samples with “manageable” dose rates ⇒ characterization is expensive
- Key questions:
  - **Which regions of the sample are “normal”, which regions are “unusual”?**
  - **Which regions provide the best return of investment when prepared in destructive testing?**
- Pulsed neutron techniques add unique data to the data from only few tools available to characterize the entire irradiated sample volume of a few cm^3
- Besides post-irradiation examination, pulsed neutrons offer also unique opportunities for phase diagram studies, microstructure evolution during processing etc.

Los Alamos, Oak Ridge, and Idaho National Laboratories: Fuel Development

ATR: Advanced Test Reactor
DE: Destructive Evaulatuon
HFEF: Hot Fuel Examination Facility
IMCL: Irradiated Material Characterization Lab
NDE: Non-destructive Evaluation
ROI: Region of Interest



Irradiation in ATR at INL

Ship from ATR to HFEF at INL

Neutron radiography and typical NDE at INL

Define reference points with fiducials

Ship from INL to LANL

NDE at LANSCE: Bulk characterization and identify ROIs

Ship from LANL to INL

Section ROIs identified at LANSCE in HFEF

DE at INL at IMCL, etc.

Complete, 3D, A-NDE Characterization to Optimize DE Characterization of Irradiated Fuels

LANSCE
Los Alamos Neutron Science Center

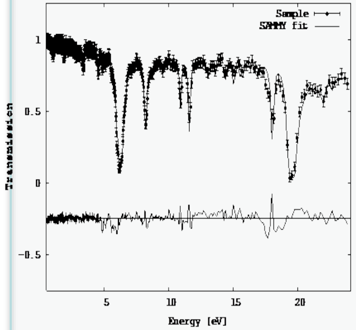
AFC Advanced Fuels Campaign

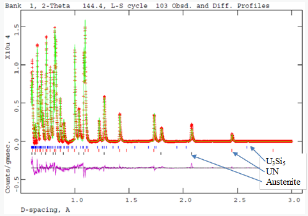
Vogel et al., "Pathway to Characterization of Irradiated Specimen with Pulsed Neutrons", DOE/NE milestone report M3FT-19LA020201012

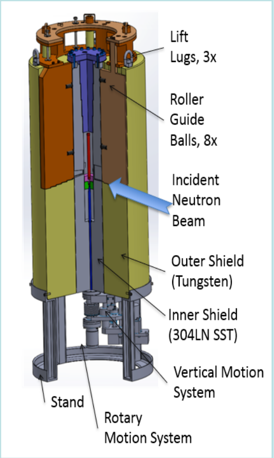
INL Idaho National Laboratory

Pillars of Advanced Post-Irradiation Examination at LANL

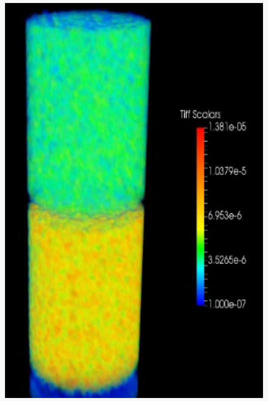
Method Development

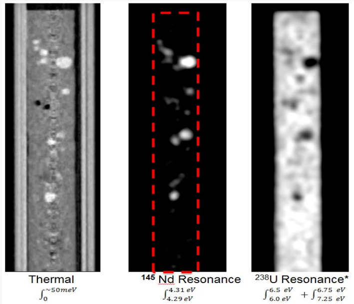


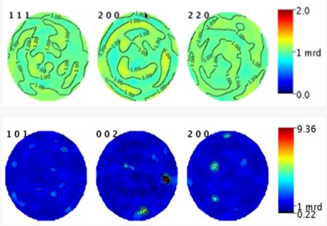




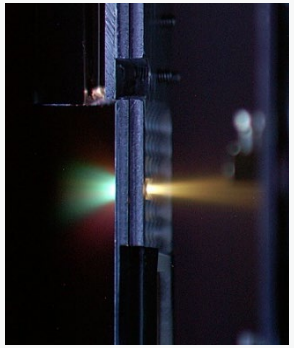
Demonstration & Application

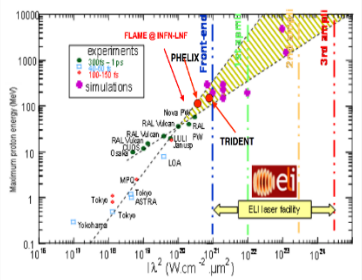






Develop Pool-side Capability





Fast Reactor Fuel Performance Challenges & New Concepts

Historical Fuel Performance Issues

- Metallic/fast reactor fuels have fundamentally different irradiation behavior than e.g. ceramic fuels such as UO_2
- Swelling - limited burnup to 3 at. %, Solved early in EBR-II testing with lowering Smeared Density to 75% to allow for interconnected porosity releasing fission gas, solid fission product build-up limits fuel to 15-20 at.% burnup
- Alloying elements to raise the fuel melting temperature and tailor the phase of U or U+Pu in the fuel (Zr, "Fs", Mo, Ti)
- Fuel Cladding Chemical Interaction (FCCI)
  - *FCCI occurs at nominal operating conditions in U and U-Mo fuels and limits burnup to 10at. % (U-Fe, U-Ni interaction typically)*
  - *FCCI occurs at nominal operation conditions in U-Zr and U-Pu-Zr fuels beyond 10at.% burnup (Lanthanide – Fe interaction typically)*
- Fuel Constituent Redistribution – an effect of phase transitions
  - *U, U-5Fs, and U-10Mo do not redistribute*
  - *U-10Zr does redistributes where Zr migrates to the center of the fuel*
  - *U-Pu-10Zr redistributes with Zr migrating to the central region and the periphery*

"Fs"" – 49.8Mo-38Ru-6Rh-4Pd-2Zr-0.2Nb

New concepts

Annular / low smear density

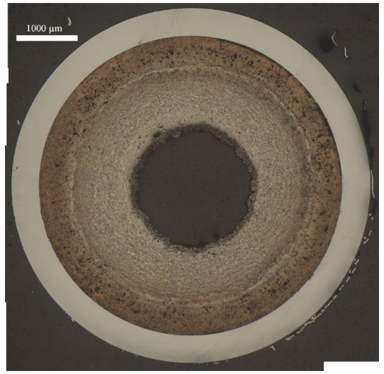
New alloys

Additives

New alloys

More irradiation tests!

U-4Pd-10/13Zr, annular, 55% sd



AFC-3 A/B/C/D & AFC-4A series

- **AFC-3A/B/C/D & AFC-4A are alloys exploration tests**
  - Alternate alloys and forms to U-10Zr: U-10Mo
  - Pd additive to mitigate fuel-cladding chemical interaction (FCCI)
  - Annular Forms to eliminate Na treatment issues (He bonded)
  - Lower smear density
- **Irradiation Issues**
  - Capsule Fabrication 3A/B
  - Reactor Uncertainty
- **Characterization at LANSCE:**
  - 6 mm ∅, ~1.5 mm thick disk prepared from AFC-3A-R5A
  - Irradiated for Nuclear Technology Research and Development (NTRD) Advanced Fuel Campaign (AFC) program at ATR
  - Burnup of 2.5 % fissions per initial metal atom (FIMA)
  - Received at LANSCE November 2019 with NSUF grant

Rodlet ID	Alloy	Fuel Form	Bond Material	Nominal Smear Density
3A-R1	U-10Mo	Solid	Sodium	75%
3A-R2	U-10Mo	Annular	Helium	55%
3A-R4	U-10Zr	Annular	Helium	55%
3A-R5A	U-1Pd-10Zr	Solid	Sodium	75%
3A-R5B	U-2Pd-10Zr	Solid	Sodium	75%
3B-R1	U-4Pd-10Zr	Solid	Sodium	55%
3B-R2	U-4Pd-10Zr	Annular	Helium	55%
3B-R4	U-10Mo	Solid	Sodium	55%
3B-R5	U-10Mo	Solid	Sodium	55%
3C-R1	U-10Mo	Solid	Sodium	75%
3C-R2	U-10Mo	Annular	Helium	55%
3C-R3	U-10Zr	Sodium	Solid	65%
3C-R4	U-10Zr	Annular	Helium	55%
3C-R5A	U-1Pd-13Zr	Solid	Sodium	75%
3C-R5B	U-2Pd-13Zr	Solid	Sodium	75%
3D-R1	U-10Zr	Annular	Helium	55%
3D-R2	U-4Pd-13Zr	Solid	Sodium	55%
3D-R3	U-10Mo	Solid	Sodium	55%
3D-R4	U-10Mo	Annular	Helium	55%
3D-R5	U-4Pd-13Zr	Annular	Helium	55%
4A-R1	U-10Mo	Annular	Helium	65%
4A-R3	U-5Mo-4.3Ti-0.7Zr	Solid	Sodium	75%
4A-R4	U-5Mo-4.3Ti-0.7Zr-2Pd	Solid	Sodium	75%
4A-R5	U-10Zr	Solid	Sodium	75%

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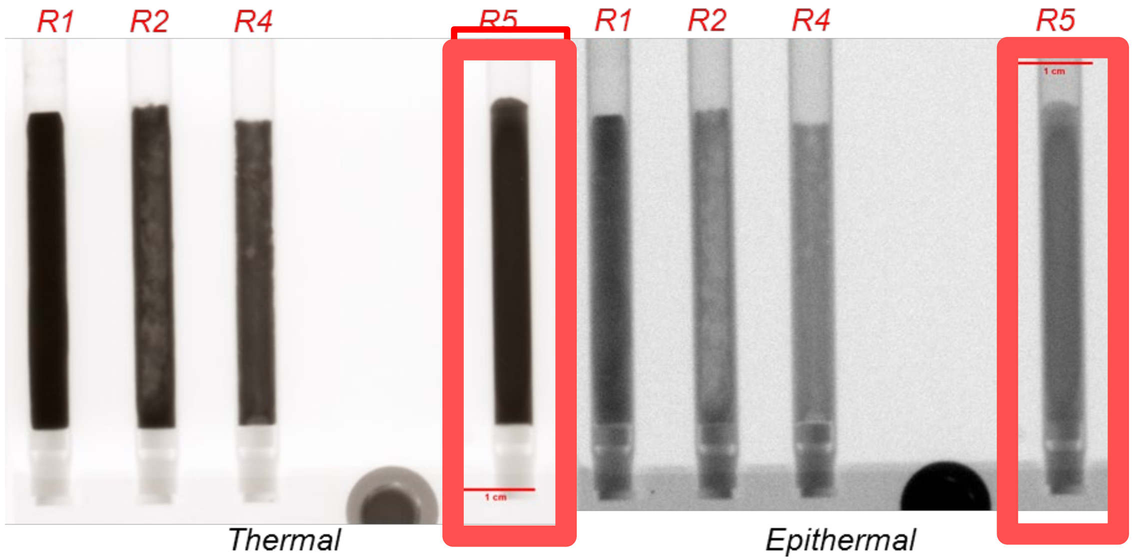
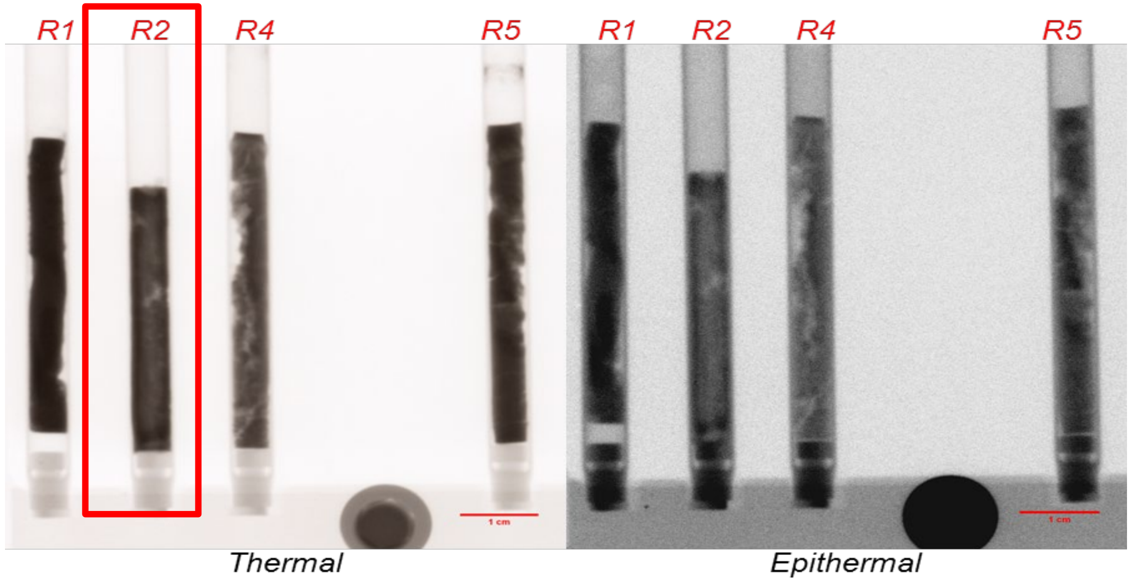
AFC-3A/B PIE Highlights – Neutron Radiography at nRAD@INL



Neutron radiography of AFC-3A

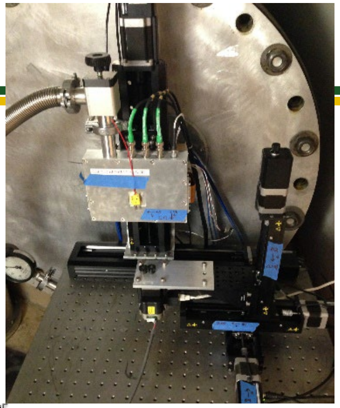


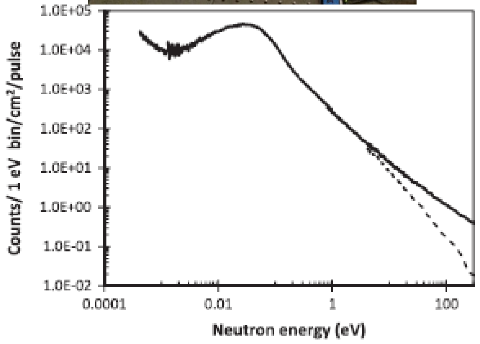
Neutron radiography of AFC-3B

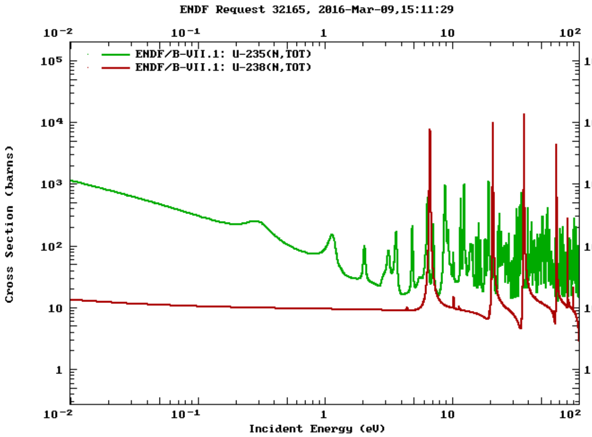
J.M. Harp et al. / Journal of Nuclear Materials 509 (2018) 377e391

At LANSCE: Energy-resolved Neutron Imaging

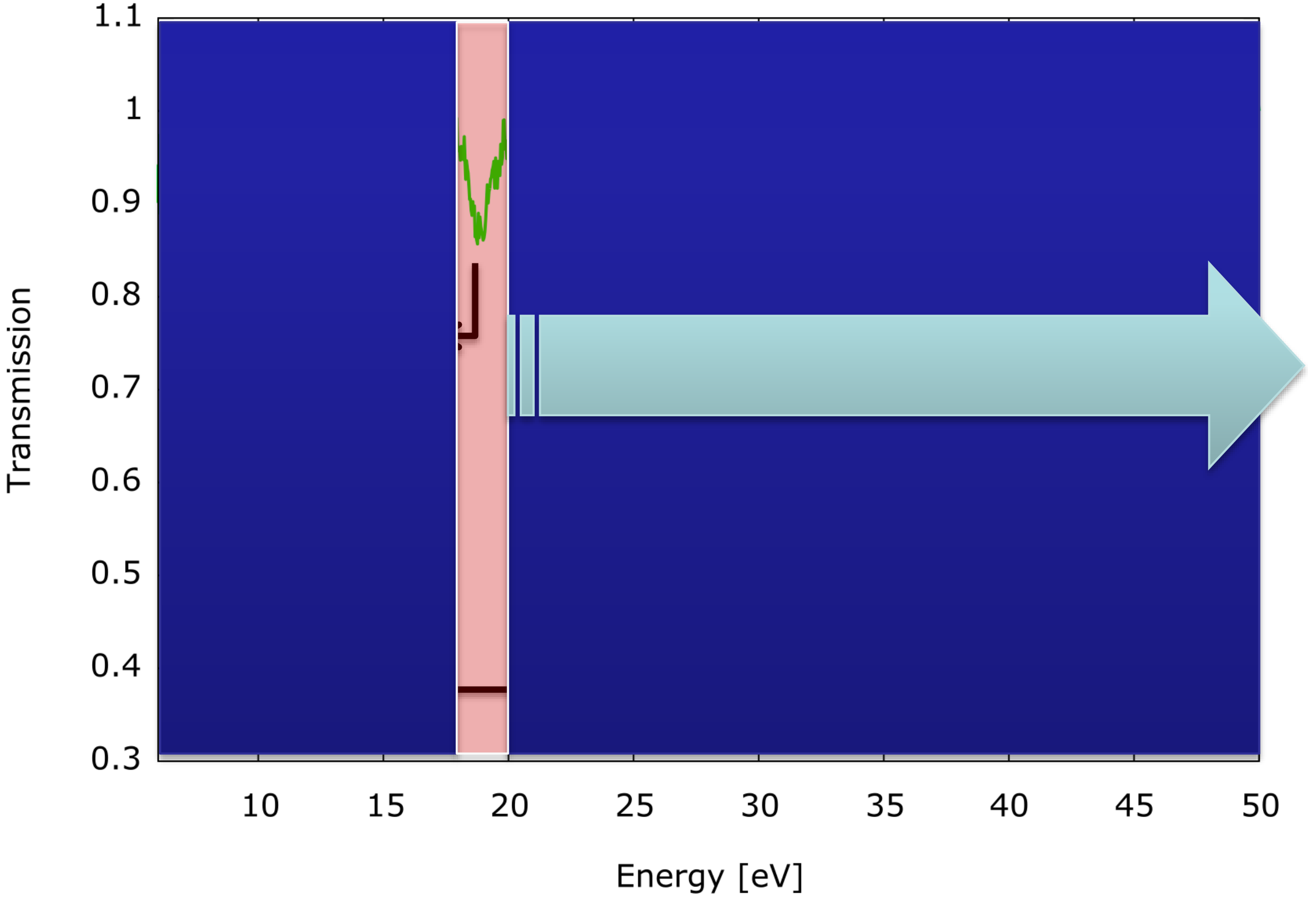
- Pulsed LANSCE neutron source allows to select neutron energy by time-of-flight
- Pixilated time-of-flight detector allows to record ~3000 neutron radiographs/pulse ⇒ 512×512 transmission spectra (28×28 mm^2)
- Selecting higher neutron energies allows imaging of isotopes opaque for thermal neutrons
- Isotopes can be identified by their absorption resonance “finger-print”
- Isotope densities can be quantified by the well-known cross-sections

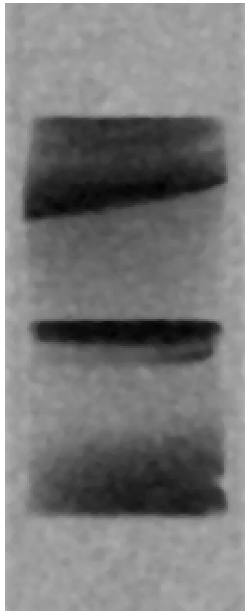




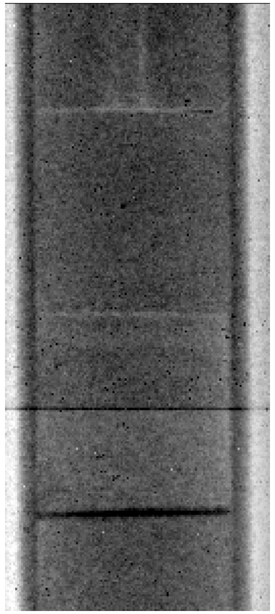


Mapping Isotopes by Neutron Energy Selection





Transmission Image (Tungsten only by energy selection)

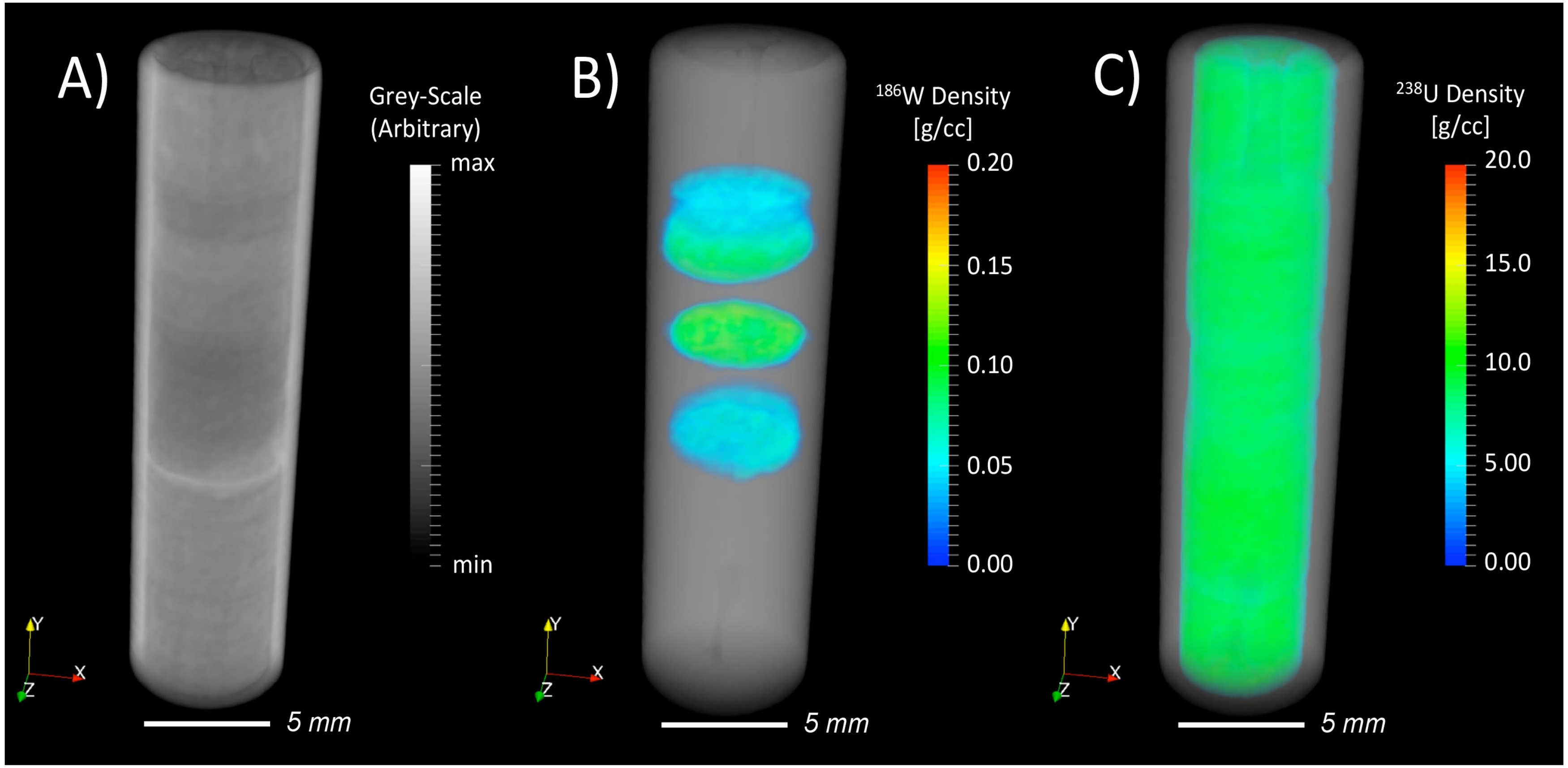


Transmission Image (Thermal spectrum)

Selecting an energy range of a given isotope allows for mapping of the specific isotope in the presence of other isotopes.

Principle of Non-destructive Bulk Isotope Density Measurements with Energy-Resolved Neutron Imaging

- Known cross-section & fit areal density ⇒ number of absorbing or resonating nuclei in beam path
- Nuclei per voxel divided by voxel volume from CT reconstruction ⇒ absolute density (“partial density”)
- Applicable to Xe or Kr fission gases

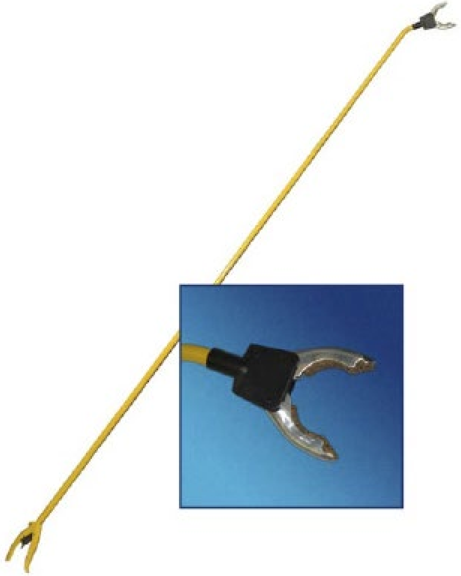


Neutron Characterization of Irradiated U-10Zr-1Pd at LANSCE

- 6 mm ∅, ~1.5 mm thick disk prepared from AFC-3A-R5A
- Burnup of 2.5 % fissions per initial metal atom (FIMA)

- Designed Al sample holder with cavity to hold sample
- Sample loaded in hot cell at INL
- Received at LANSCE November 2019 with NSUF grant
- Data collected in December 2019, fall of 2020, summer 2021

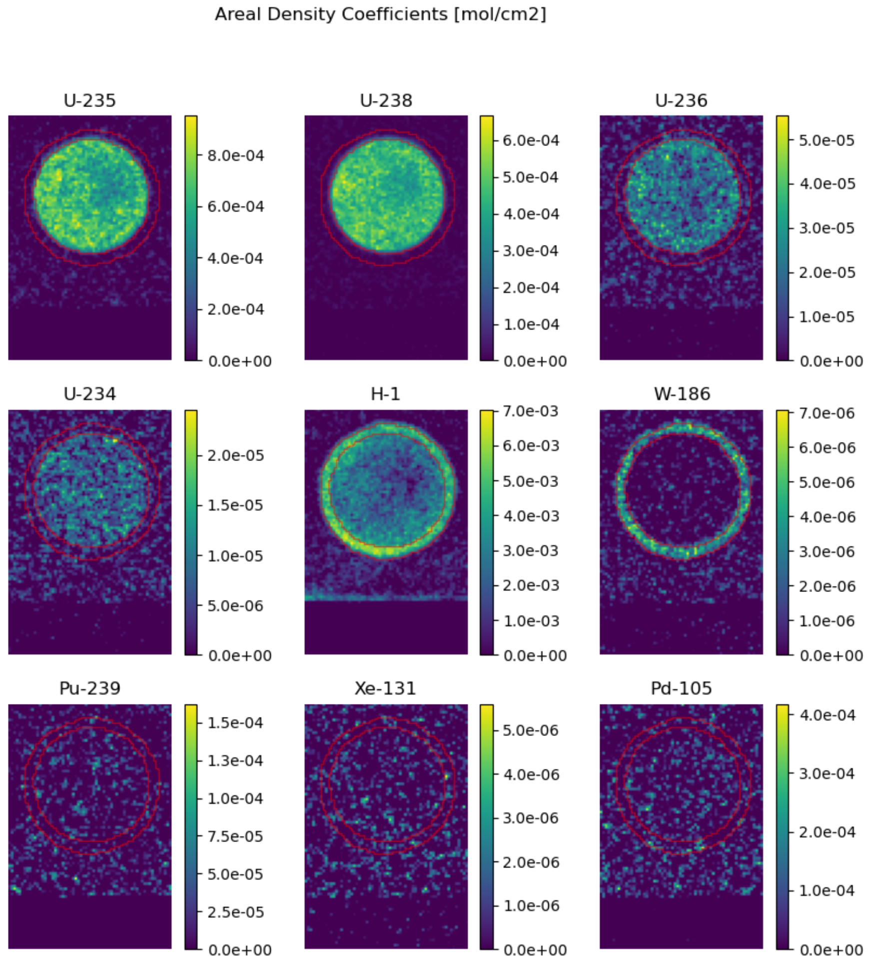
- Dose rate on contact: ~3R/hr (DOE allowable dose for public is 0.1R)
- Dose rate at 2m: ~10 mR/hr ⇒ Remote handling possible
- Pre-irradiation enrichment level: 56.5 at.% U-235 (of U atoms)
  ⇒ strong thermal neutron absorber
  ⇒ 1/e penetration depth for thermal neutrons: ~1.1 mm
  ⇒ still probing entire 1.5mm thickness (XRD: ~1μm for Cu K_α)
  ⇒ diffraction difficult…



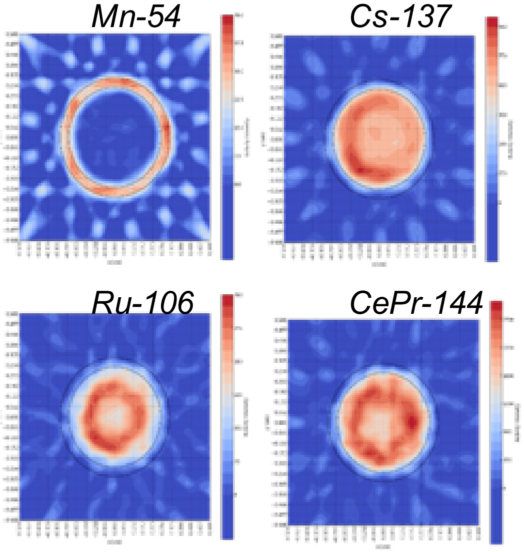
Energy-resolved Neutron Imaging (ERNI)

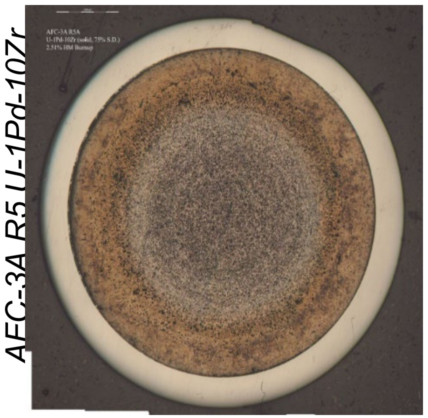
- **Pulsed neutrons with proper time-of-flight detector allow mapping of isotope distributions**
- **For the disk-shaped sample done in 2D**
- **Ultimately doable in 3D using tomographic reconstruction**
- **Complements gamma-emission tomography which cannot detect non-radioactive isotopes**

Areal density of select isotopes from ERNI



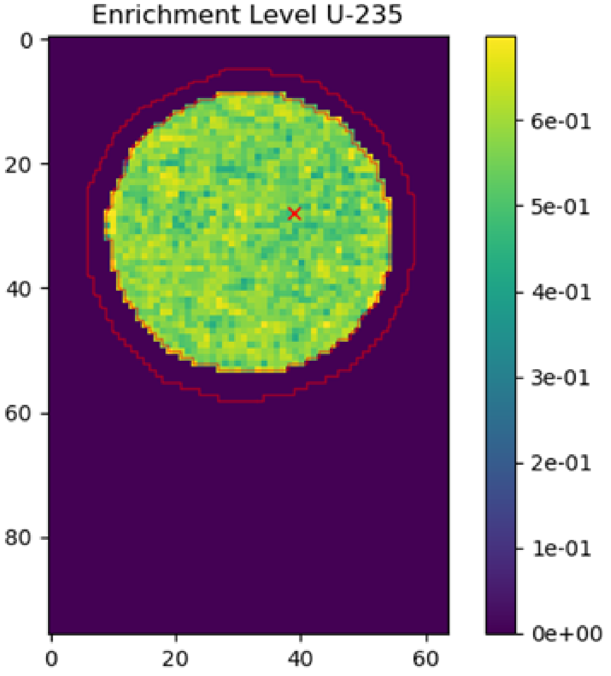
Distribution of radioactive fission products from gamma-emission tomography

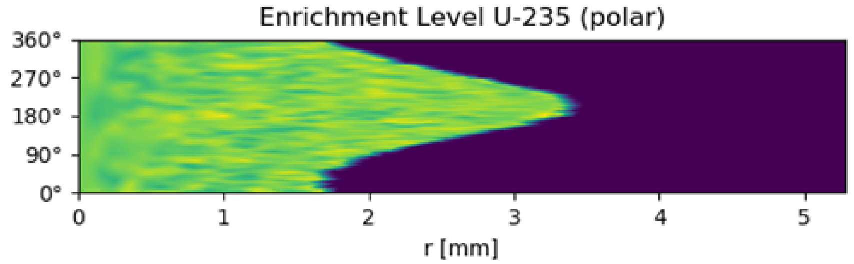


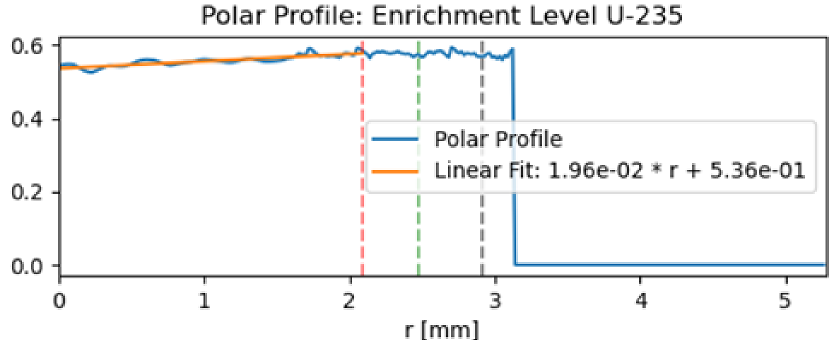


U-235 areal density map

- U-235 areal density shown as fraction relative to all U isotopes ⇒ enrichment level
- Convert 2D maps into polar coordinates (density as a function of azimuth angle and radius)
- Average densities as a function of radius to observe trends
- Mean U235 enrichment level: 56.8+/-5.1% (but strong trend, not statistical fluctuation!
  53.6% in the center, increases at ~2%/mm to the outer radius
- Nominal initial U235 enrichment level was 56.5% of all U atoms pre-irradiation
  ⇒ Do not see the 2.5% FIMA that this was irradiated to - but we are close

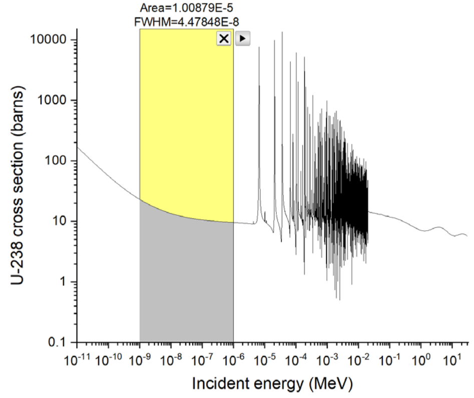


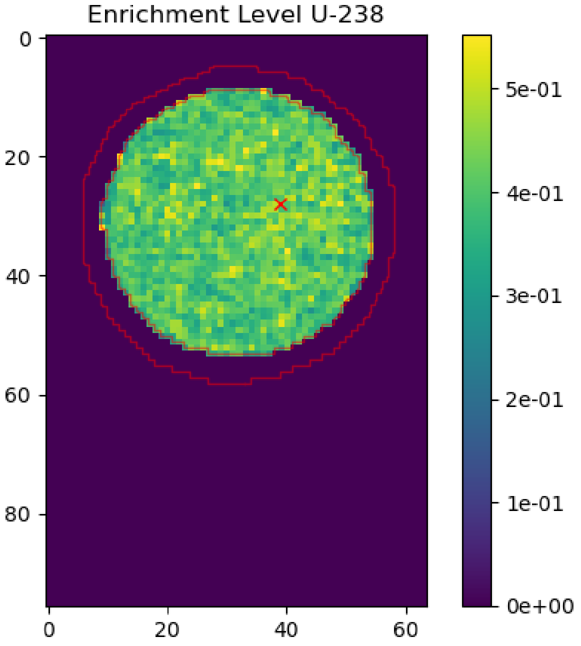


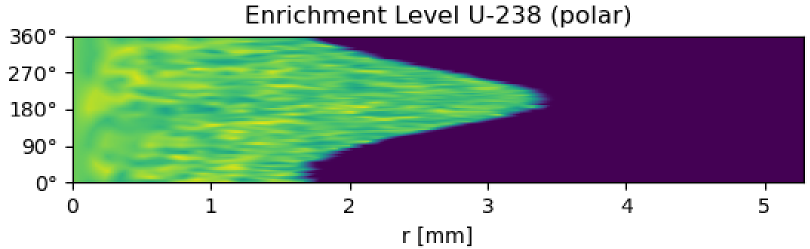


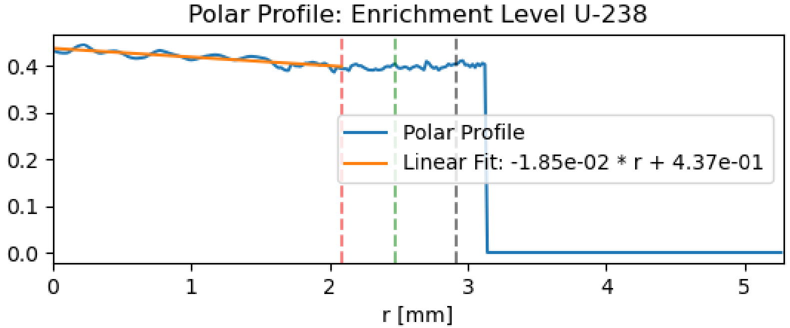
U-238 areal density map

- **U-238 fraction (relative to areal density of all uranium isotopes) *decreases* as a function of radius ⇒ counter-intuitive?**
- **Answer: Cd shroud during irradiation shields material from thermal neutrons!**
- **Integrated cross-section of U-238 above Cd cut-off > U-235!**
- **MCNP calculation will provide more accurate test**









	Cross section [barns*MeV]	
Energy [eV]	U-235	U-238
0.4-10	0.0007727	0.00101
0.4-100	0.00714	0.00929
0.4-1000	0.03543	0.03807

Energy-resolved Neutron Imaging: Increased Uranium density at outer radius

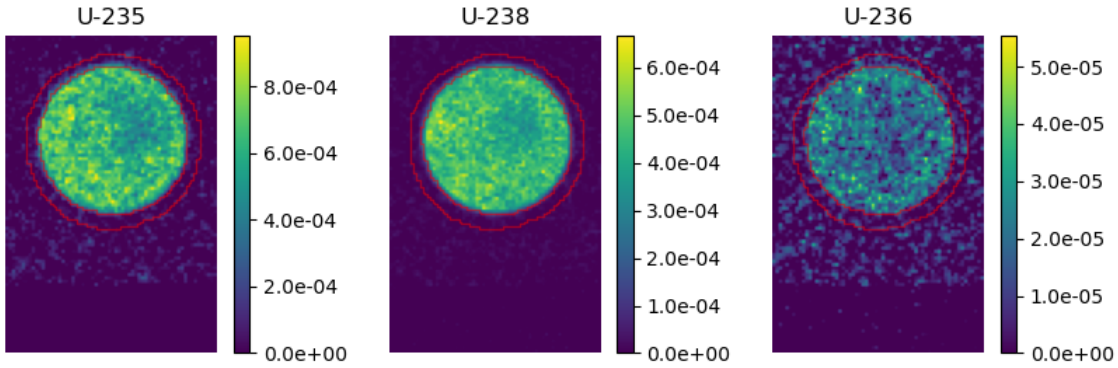
- Uranium density increases towards the outer radial region
  ⇒ consistent with findings from electron microscopy
  (e.g. Yao et al. JNucMat, 2020)

- Nominal composition: 77.5U-22.5Zr (atom %)

- Observed compositions by electron microscopy on outer radial region by Yao et al.:
- 97U-3Zr: 25% more U than nominal
- 93U-7Zr: 20% more U than nominal

⇒ Energy-resolved neutron imaging results agree

⇒ Results averaged over entire cross-section



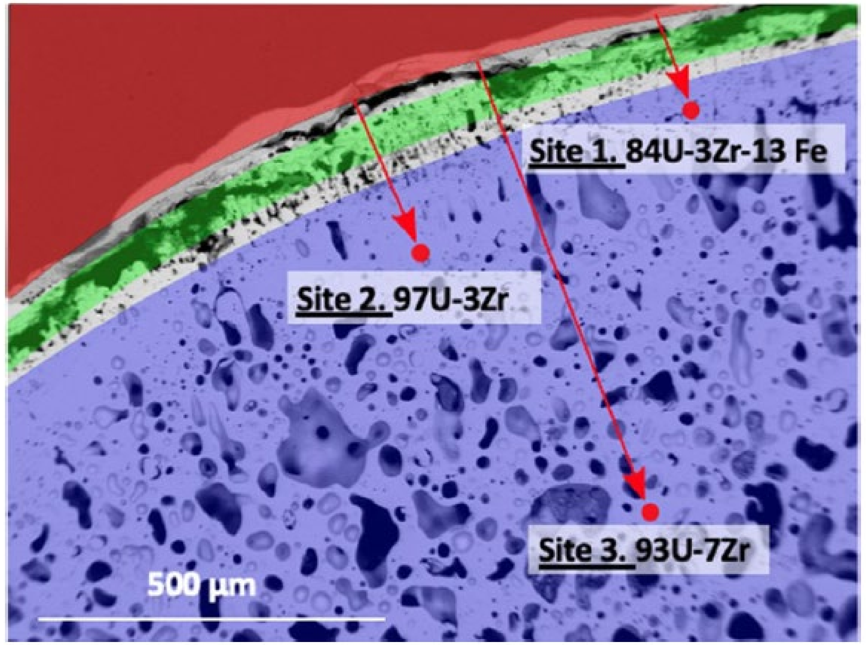
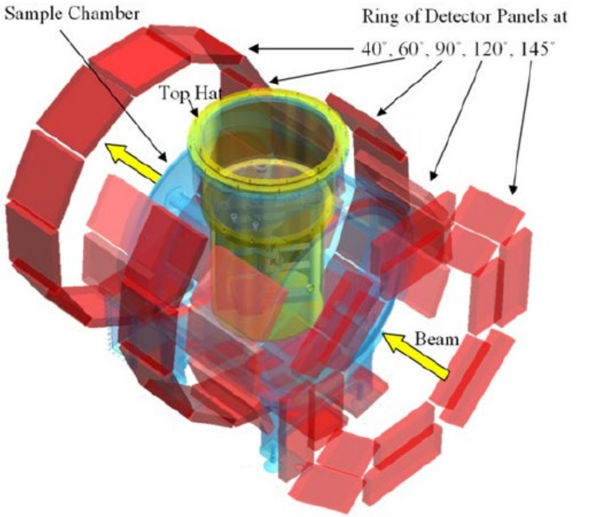
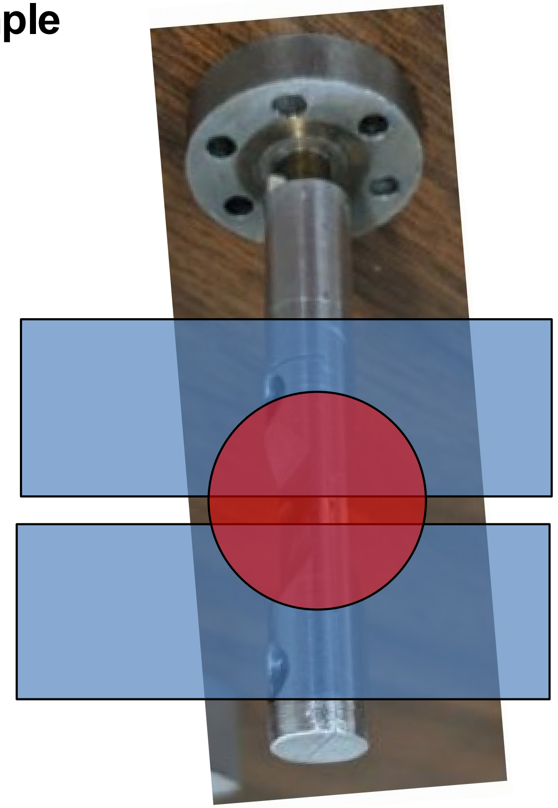


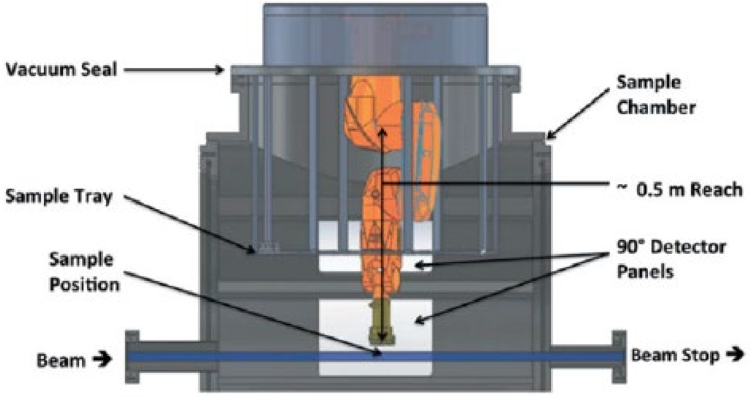
Fig. 3. The variation of major chemical compositions inside fuel near the fuel-cladding interface.

Results for irradiated U-10Zr-1Pd Sample from Neutron Diffraction

- High Pressure/Preferred Orienation (HIPPO) neutron time of flight diffractometer at Los Alamos Neutron Science Center
- Sample loaded on robotic sample changer
- Scanned with 2mm wide horizontal slit along sample axis to improve signal to noise ration
- Rietveld analysis of diffraction data
- Diffraction signal dominated by aluminum from sample holder



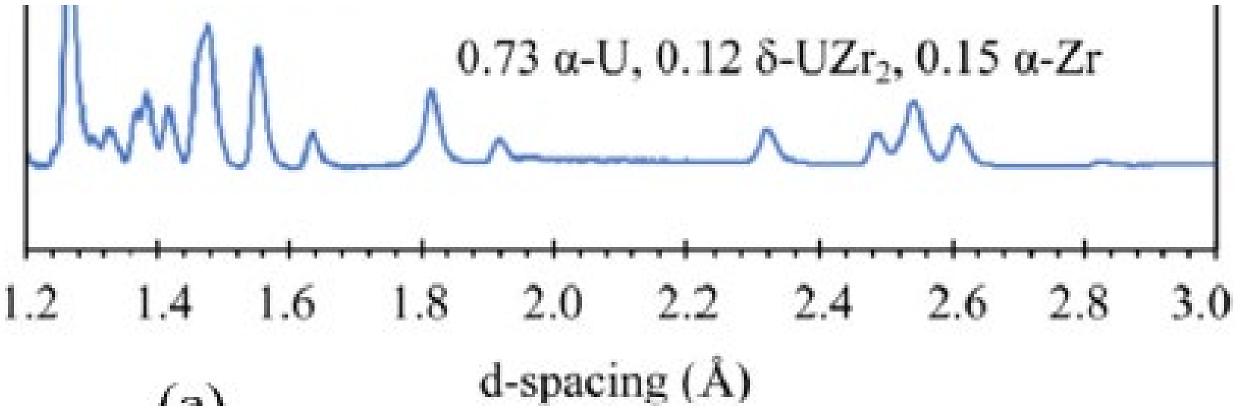


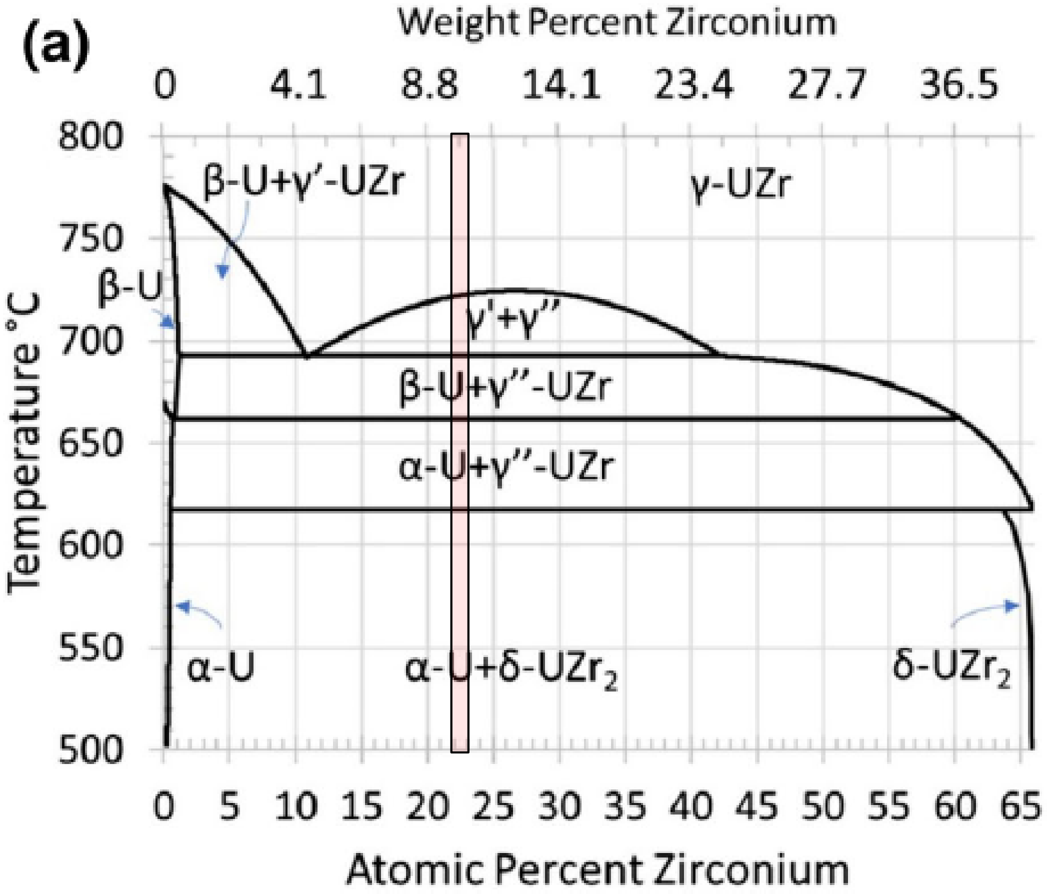


*Wenk et al. NIMA **515** (2003) 575, Vogel et al. Powder Diffraction **19** (2004) 65, Losko et al. J. Appl. Cryst. **47** (2016) 2109, Takajo & Vogel, J. Appl. Cryst. **51** (2018) 895*

Expectation: α-U + δ-UZr_2

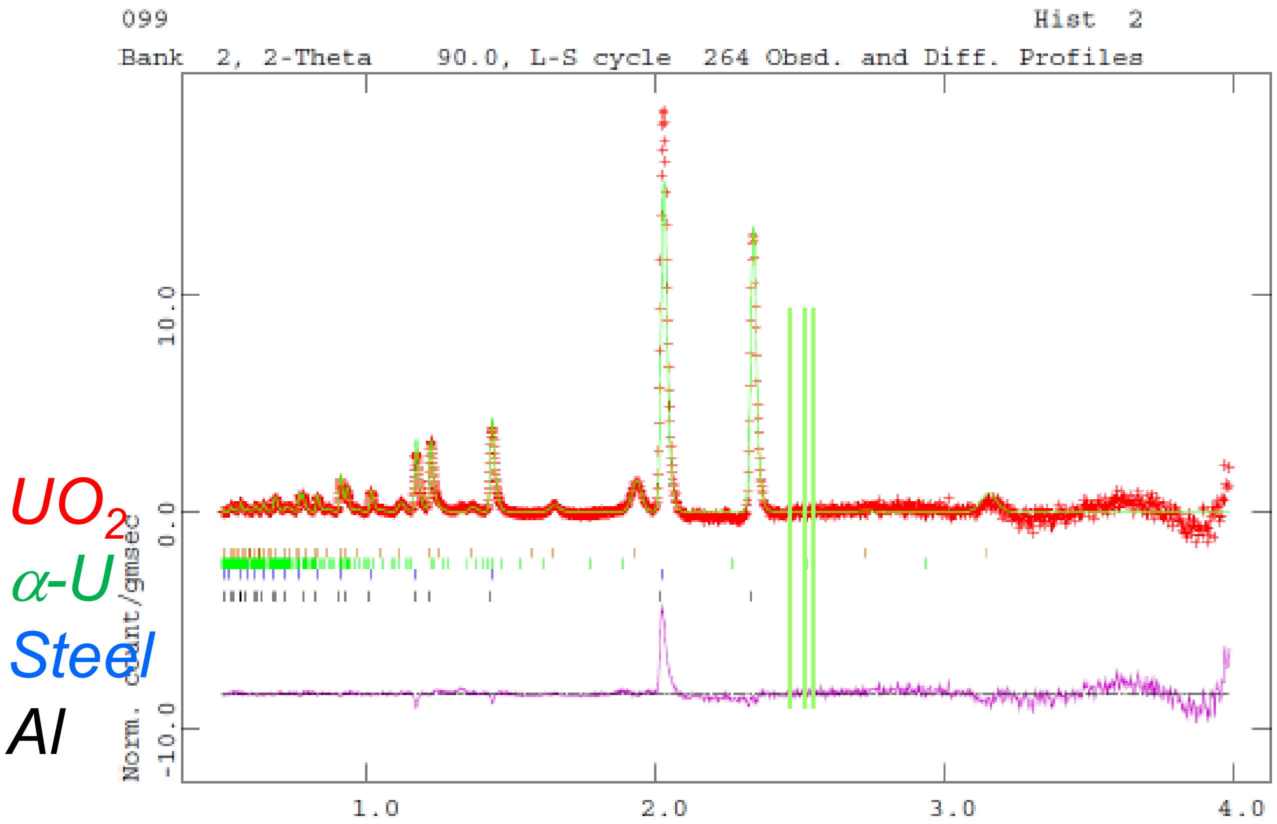
- **Per phase diagram, U-10wt%Zr (U-22.5 at% Zr) should consist of α-U and δ-UZr_2**
- **Crystal structures well known**
- **In fresh material, Rietveld analysis results in ~85 wt% α-U in a U-10wt%Zr sample**
- **More studies on U-10Zr with HIPPO:**
  - Williams et al. JOM 72 (2020) 2042;
  - Xie et al. J. Nuc. Mat. 544 (2021): 152665.



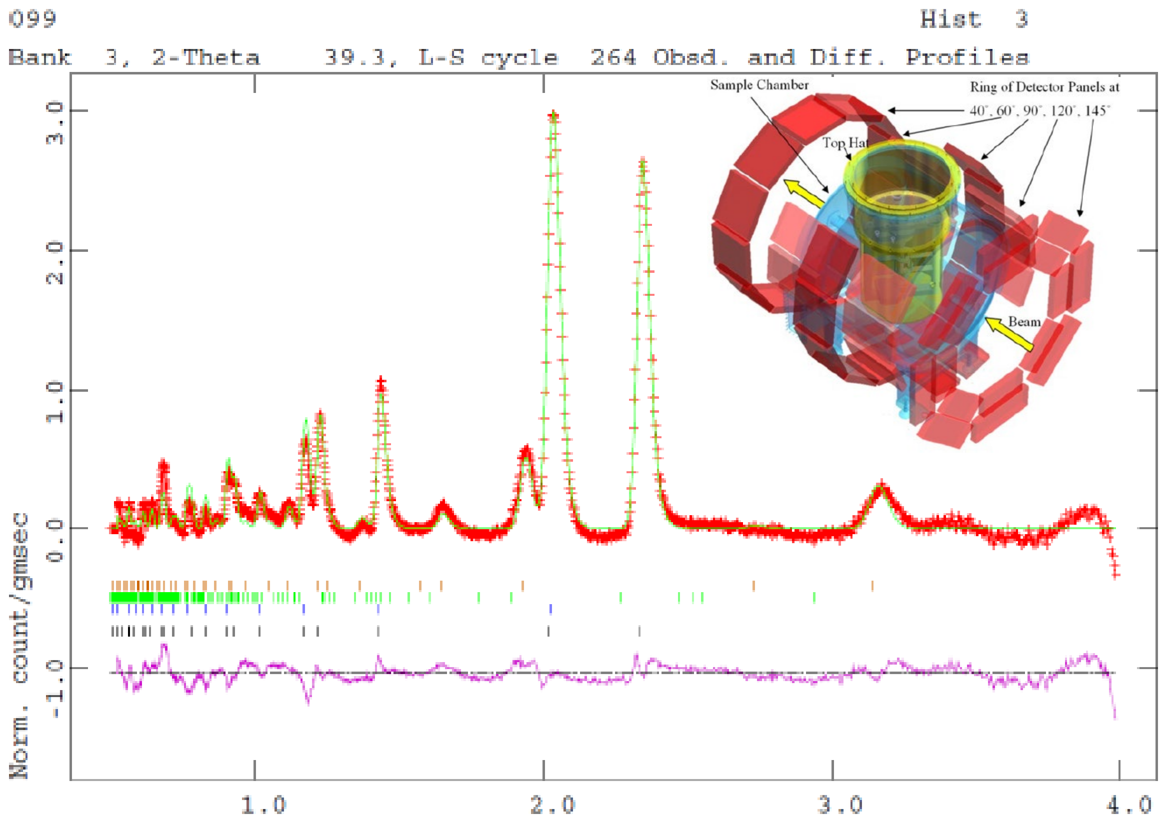


Diffraction shows sample is fully oxidized

90° bank

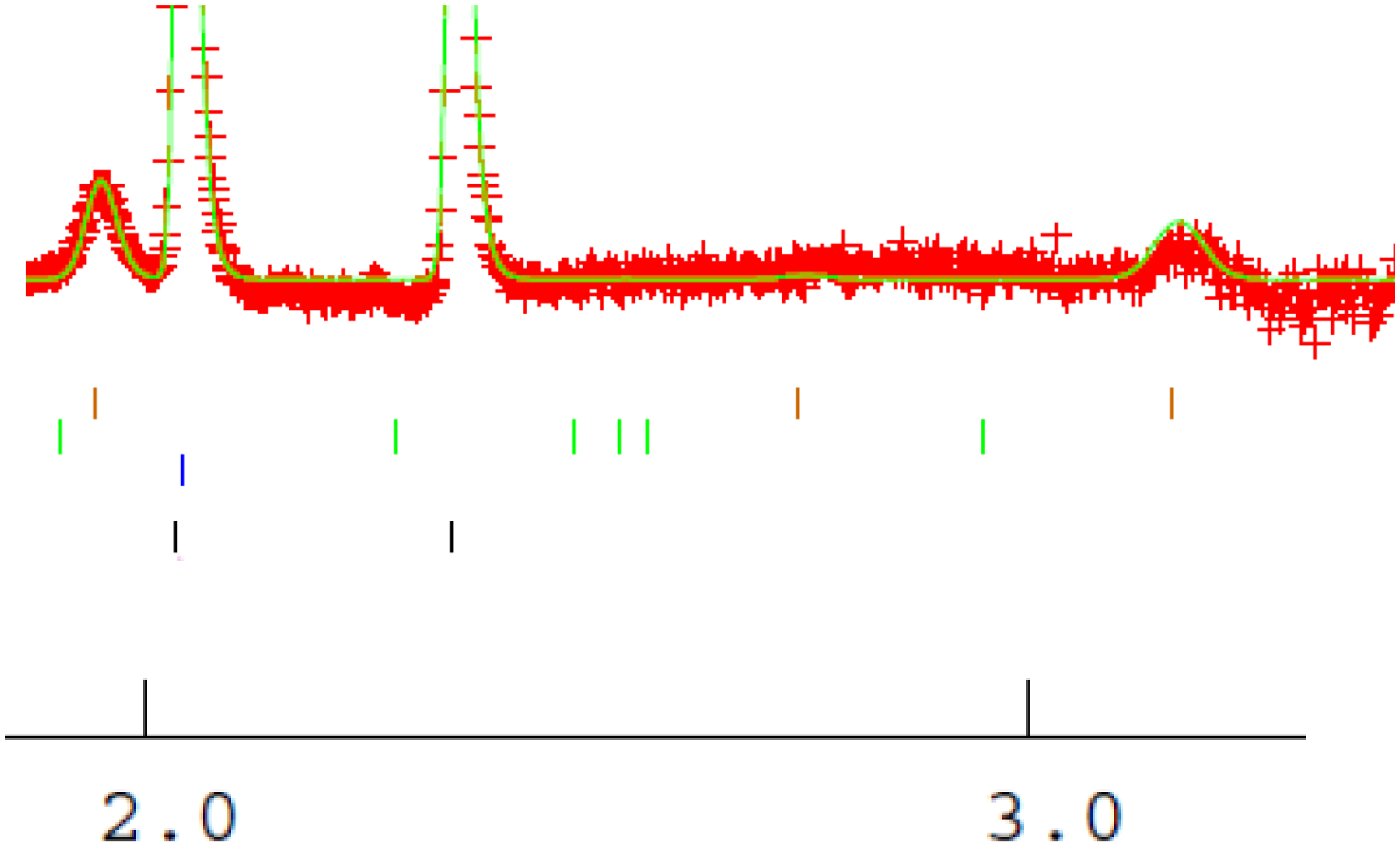


40° bank (transmitted!)

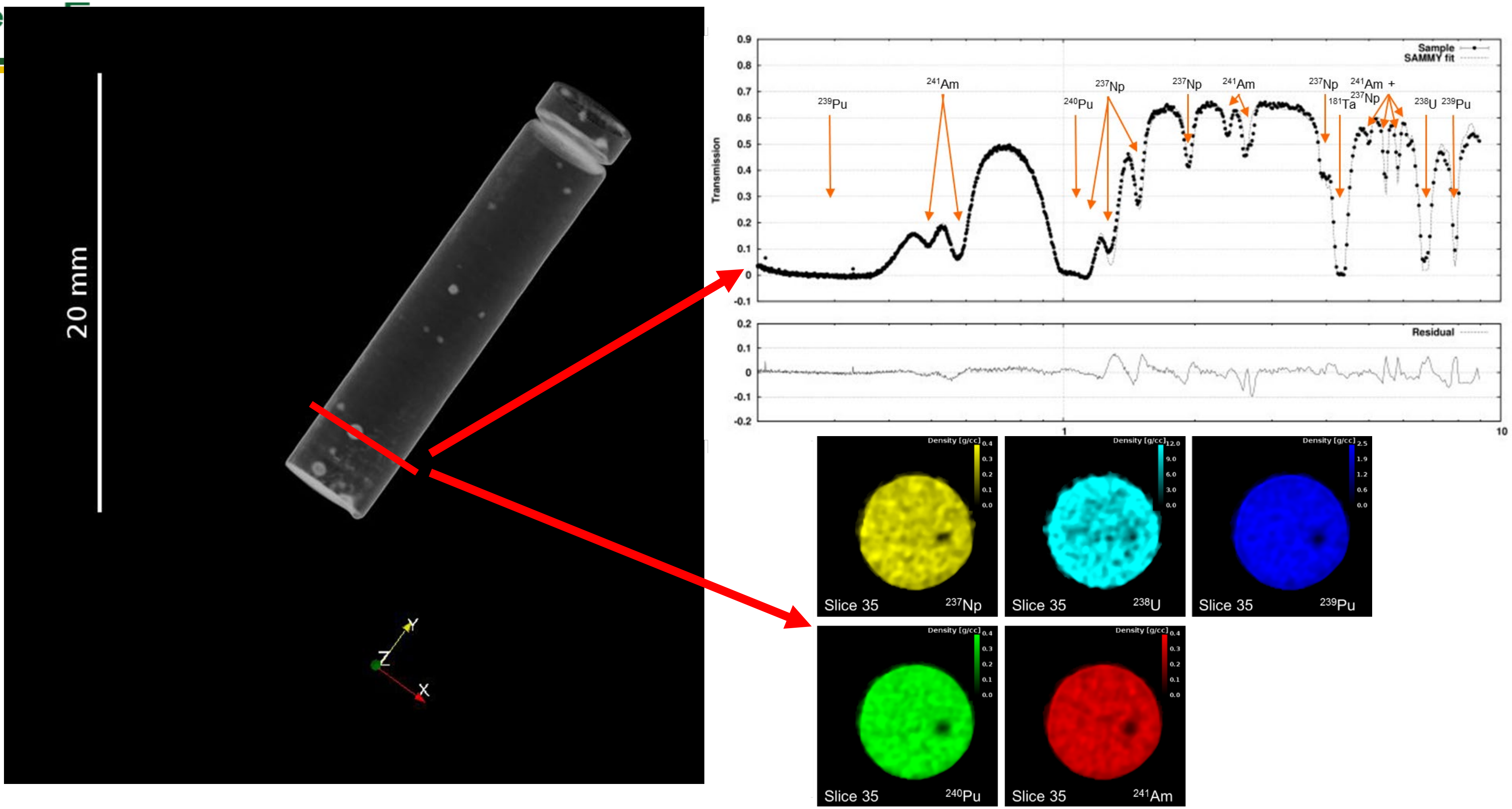


No intensity where α-U should be

- **Diffraction data shows no peaks at d-spacings where α-U should have strongest peaks around 2.5Å**
- **In the probed volume, no measurable amount of α-U is present (otherwise diffraction peaks would occur)**
- **40° detector requires neutrons to travel through sample**
  ⇒ entire thickness is probed, not a surface effect
- **No Zr-oxide detected**
- **Next time air-tight sample container is needed**

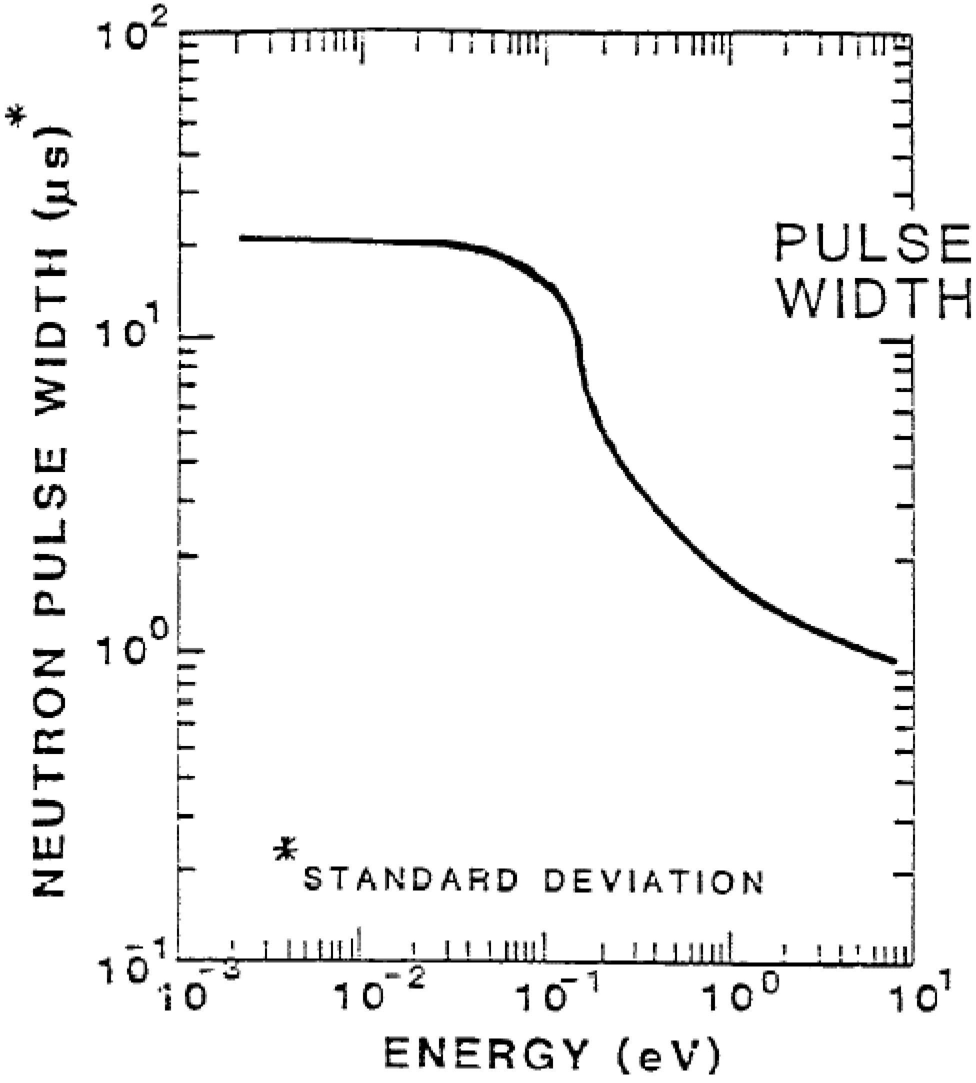


3D Reconstruction of isotope densities in dU-20Pu-10Zr-3Np-2Am (Transmutation fuel) using energy-resolved neutron imaging (ERNI)



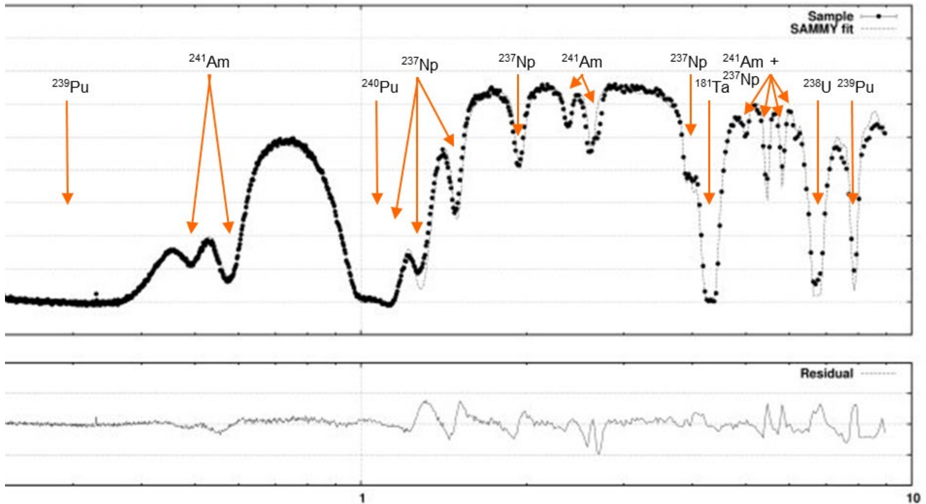
- Pixel-wise reconstruction of areal densities followed by tomographic reconstruction creates 3D isotope density maps
- Requires short-pulsed neutrons

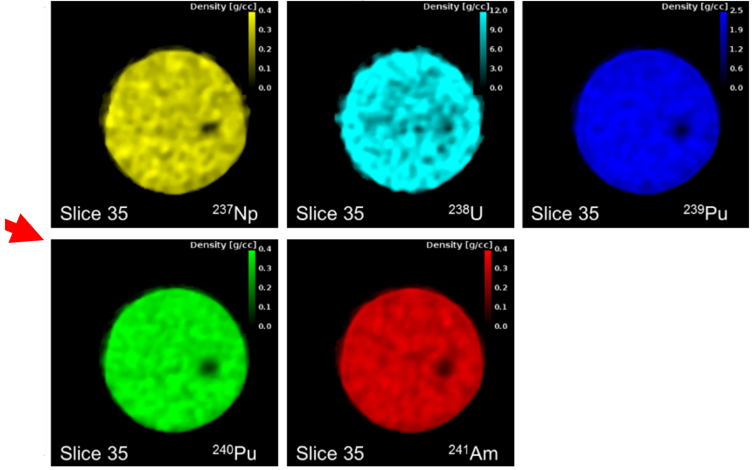
3D Reconstruction of isotope densities in dU-20Pu-10Zr-3Np-2Am (Transmutation fuel) using energy-resolved neutron imaging (ERNI)



Allowable initial p/d pulse width should be less than required neutron pulse width for desired energy (~1μs for 10 eV epithermal neutrons)

From: Gary Russell et al. ICANS-VIII Proceedings (1985)





ies followed by tomographic
ty maps

Application Example: Characterization of UCl_3 and NaCl-35.2mol%UCl_3 Salts using Neutron Scattering

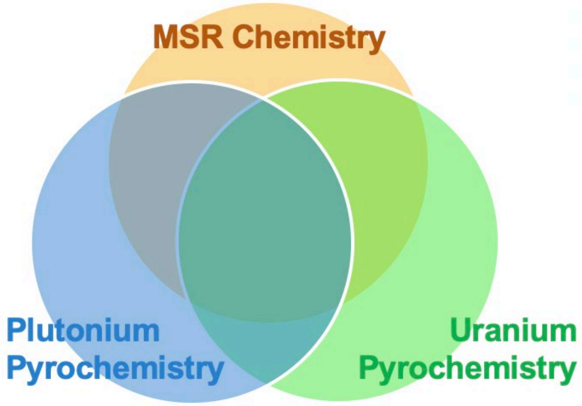
Sven C. Vogel[1], A. David R. Andersson[1], Marisa J. Monreal[1], J. Matthew Jackson[1], S. Scott Parker[1], Gaoxue Wang[1], Ping Yang[1], Boris Khaykovich[2], Sean Fayfar[2], Jianzhong Zhang[1]

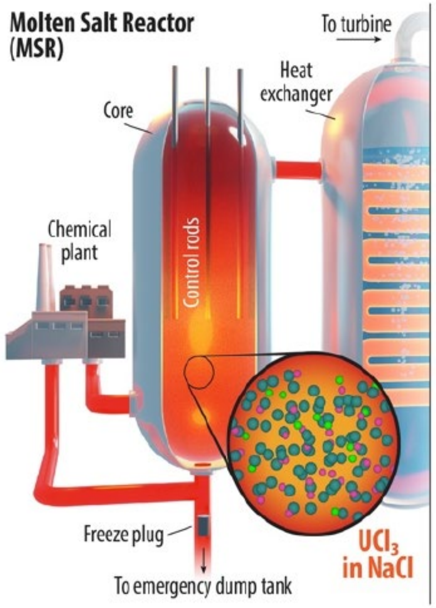
[1]Los Alamos National Laboratory
[2]MIT

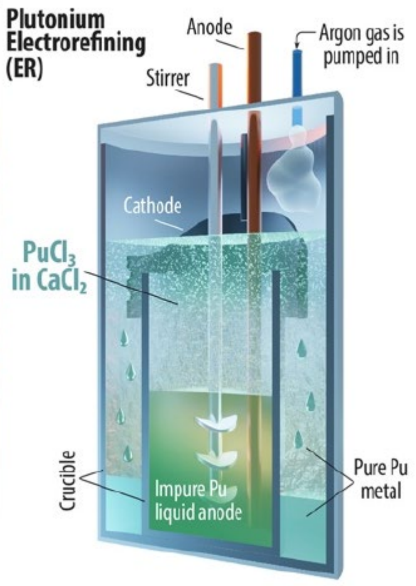
LA-UR-23-22572

Why research molten salts?

- Actinide-molten salts are used in
  - Next-generation nuclear power plants (molten salt reactors/MSRs)
⇒ liquid material is inherently resistant to radiation damage
  - Spent fuel re-processing
  - Weapons metal purification
- Experimental data on physical properties is sparse, inaccurate, and rarely includes actinides, esp. plutonium
  ⇒ Data is needed for licensing!
  ⇒ Novel techniques needed
- LANL offers
  - Infrastructure & expertise to make and handle samples, incl. Pu
  - Modeling expertise for actinide salts
  - Neutrons@LANSCE enable characterization (radiography & scattering)

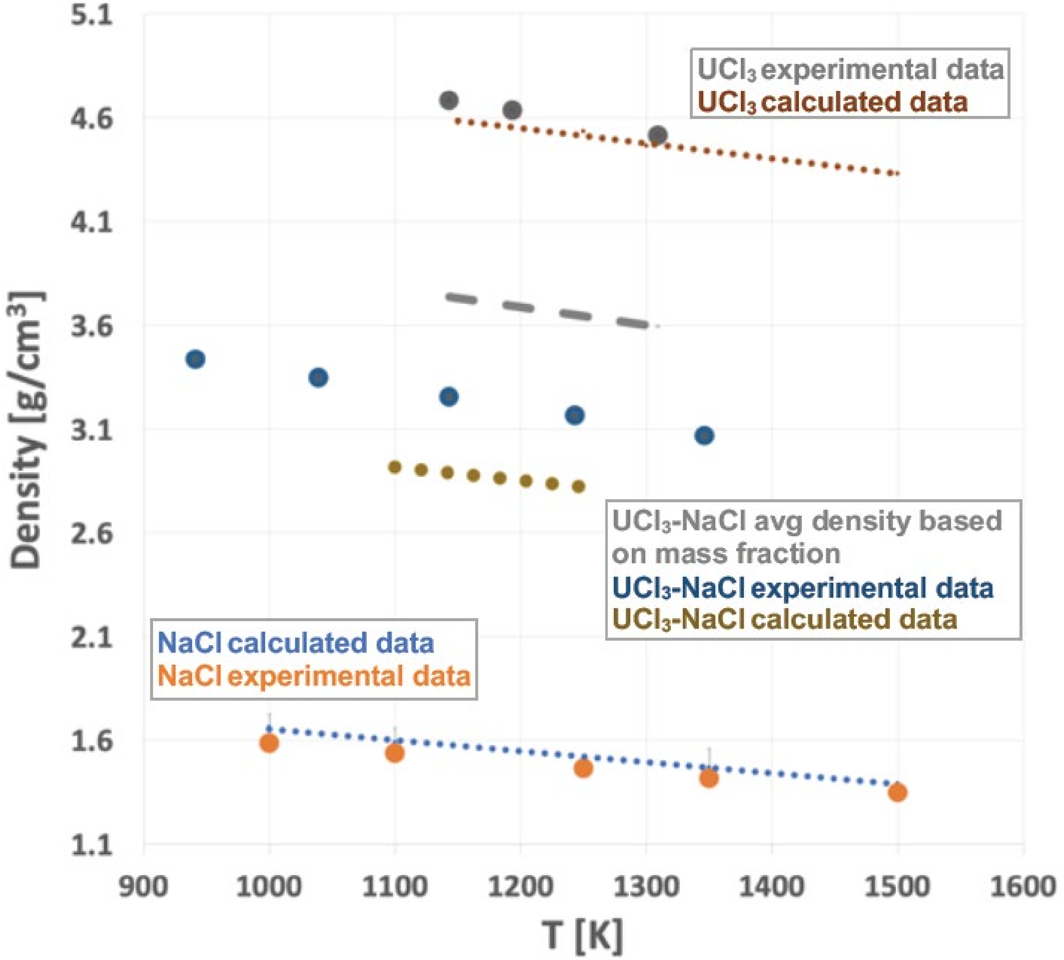






Example problem: Density

- Density normally straight forward to measure
- Chloride salts melt at T>800C, require special containers
  ⇒ not trivial
  ⇒ U, Pu salts complicate further
- Measured density of NaCl, UCl_3 and NaCl+UCl_3 mixture as a function of temperature with neutron radiography
- Prediction of densities works well for pure salts, but not for mixture
  ⇒ Model is missing "something"
  ⇒ Do experiments to find out



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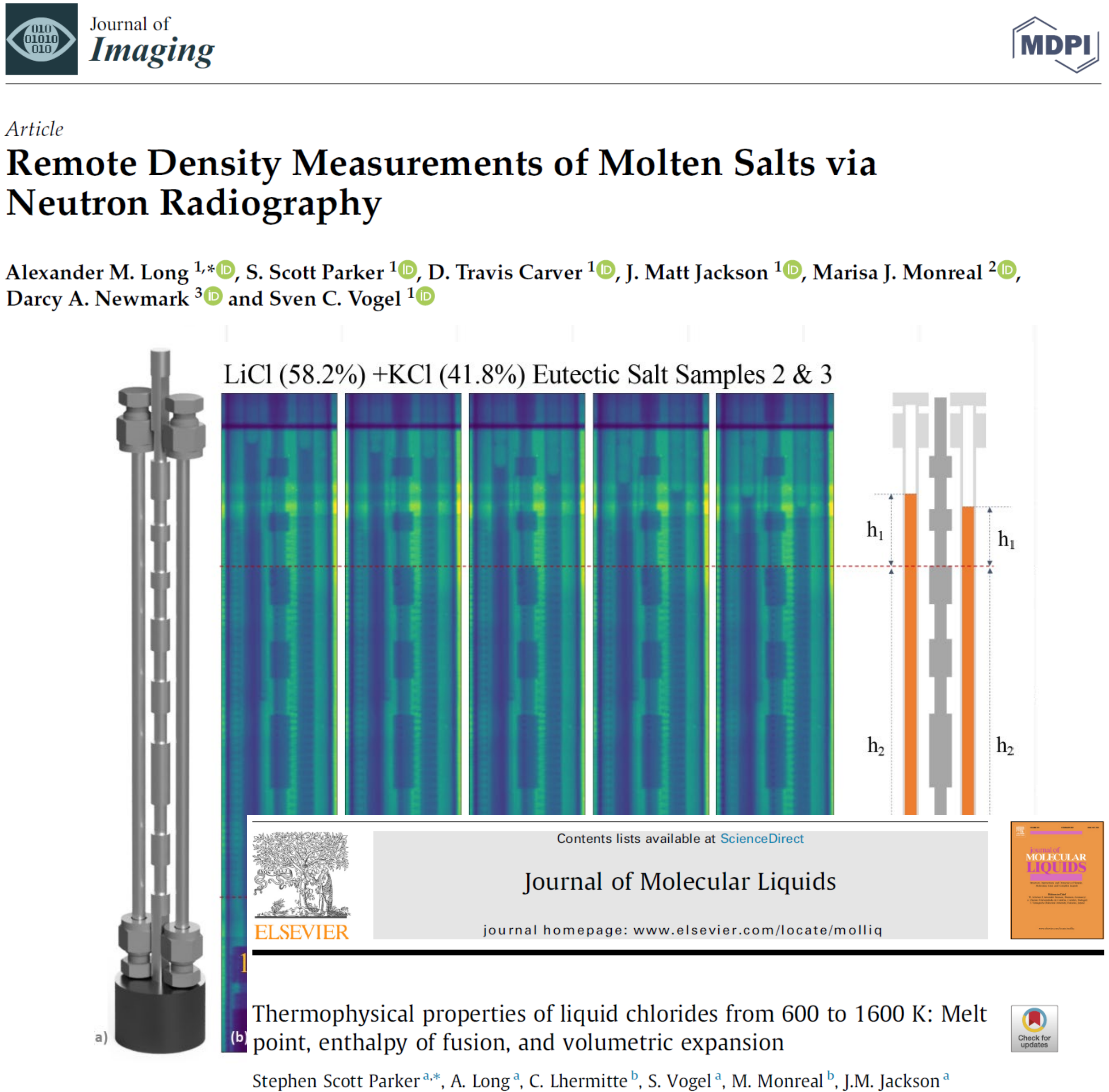


Journal of *Imaging*

MDPI

Article

Remote Density Measurements of Molten Salts via Neutron Radiography

Alexander M. Long [1,*], S. Scott Parker [1], D. Travis Carver [1], J. Matt Jackson [1], Marisa J. Monreal [2], Darcy A. Newmark [3] and Sven C. Vogel [1]

Contents lists available at ScienceDirect

Journal of Molecular Liquids

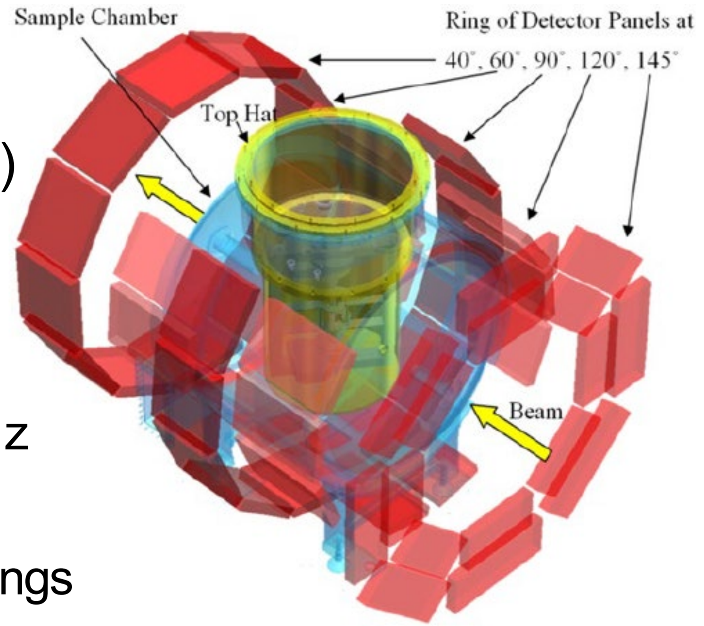
journal homepage: www.elsevier.com/locate/molliq

ELSEVIER

Thermophysical properties of liquid chlorides from 600 to 1600 K: Melt point, enthalpy of fusion, and volumetric expansion

Stephen Scott Parker [a,*], A. Long [a], C. Lhermitte [b], S. Vogel [a], M. Monreal [b], J.M. Jackson [a]

Neutron Diffraction with HIPPO



- High Pressure/Preferred Orientation (HIPPO) neutron time of flight diffractometer at Los Alamos Neutron Science Center (LANSCE)
  - Short pulse (270 ns) spallation neutron source
  - 800 MeV protons produce neutron pulses at 20Hz
- HIPPO:
  - 1,200 ^{3}He tubes arranged on 45 panels on five rings
  - Moderator to sample distance ~8.9 m
  - Detectors cover ~22.4 % of 4π around the sample
- So-called ILL furnace used for heating:
  - Vacuum ~10^{-6} Torr
  - Vanadium heating elements and heat shields
  - Maximum temperature 1150°C

Characterization of UCl_3

- Conducted two experiments:
  - Pure UCl_3 in 2020
    - *Fused inside in 3mm diameter SiO_2 glass capillary*
    - *…inside 5mm diameter glass capillary*
    - *…inside 6mm diameter vanadium can*
⇒ If inner capillary breaks, open outer capillary contains material
    - *Heated with constant heating rate of 1°C/min to 850°C, 5 minute data collection time*
    - *Heated to molten state (T_m=835°C) with 16 hrs hold for pair-distribution function data collection*
  - Eutectic UCl_3/NaCl mixture in 2022
    - *More relevant to application*
    - *Did not melt, stayed ~400°C (T_m=520°C for x_{UCl3}=0.329)*
    - *Powder sample contained in 6 mm diameter vanadium can*
⇒ Much larger sample volume, less background from glass
⇒ better neutron diffraction signal
    - *Counted for ~45 minutes at each temperature point*

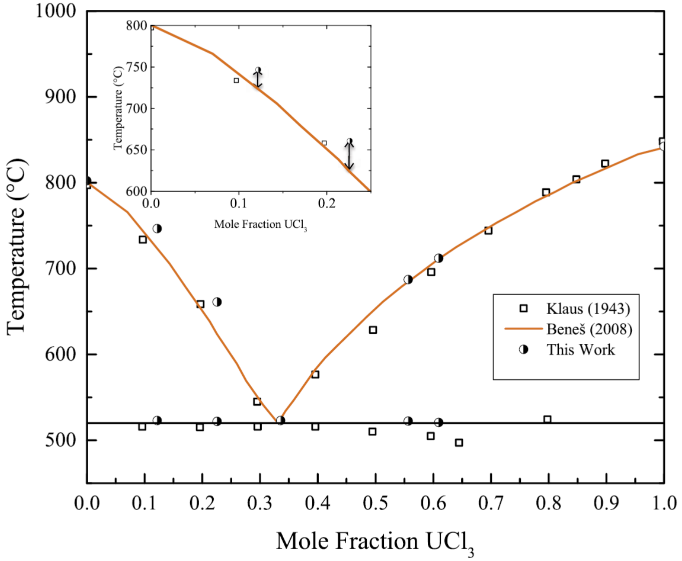


Fig. 4. The NaCl–UCl_3 melt point results plotted along side the experimental results from Kraus [13,3,11] and the modeled results of Beneš [10]. The inset highlights the discrepancy between the previously measured liquidus cure and the data obtained here.

From Sooby et al., J. Nuc. Matls. ***466*** *(2015) 280-285.*

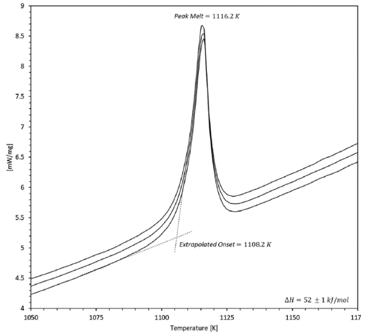
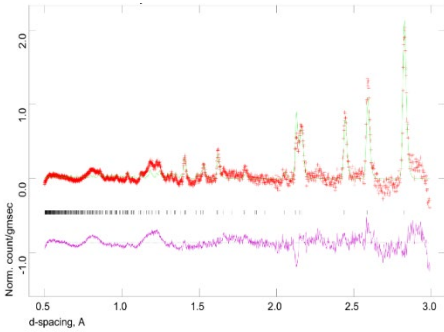


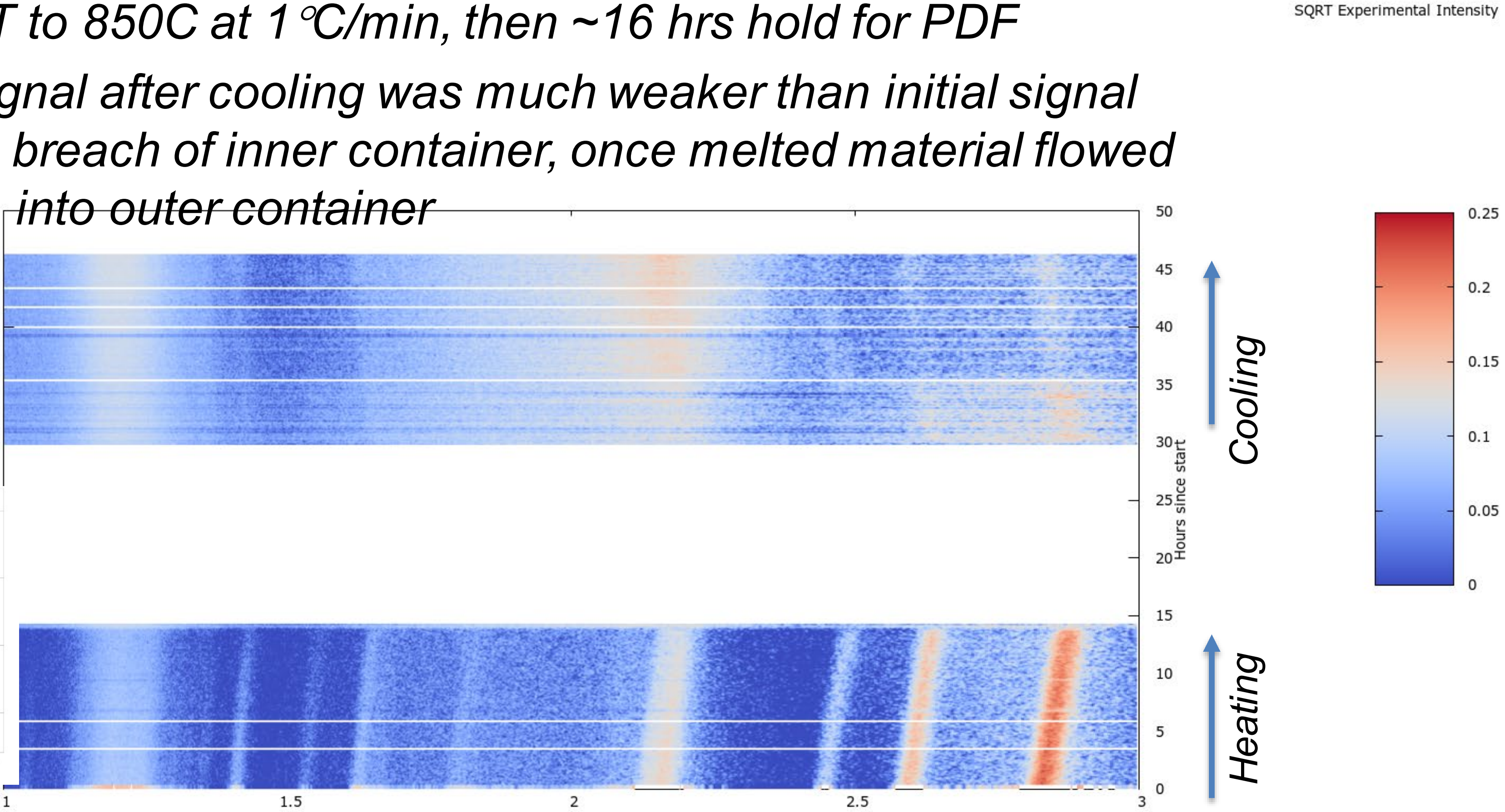
Fig. 2. DSC traces of three measurements establishing the melting point of UCl_3 to be 1108.2 ± 0.2 K.

From Vogel et al. JOM ***73*** *(2021): 3555-3563.*

Heating pure UCl_3 to melt

- *Heated UCl_3 in SiO_2 glass capillary inside vanadium can*
- *Salt previously annealed*
- *RT to 850C at 1°C/min, then ~16 hrs hold for PDF*
- *Signal after cooling was much weaker than initial signal*
  $\Rightarrow$ breach of inner container, once melted material flowed into outer container

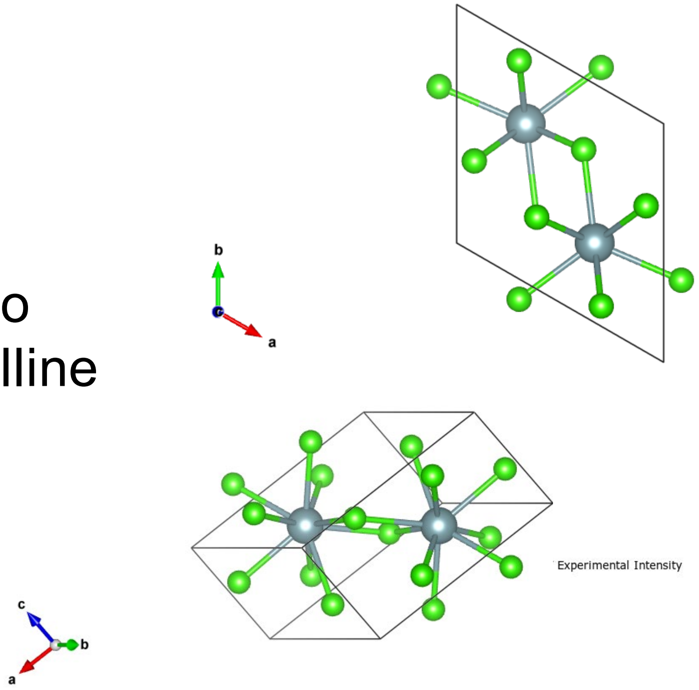


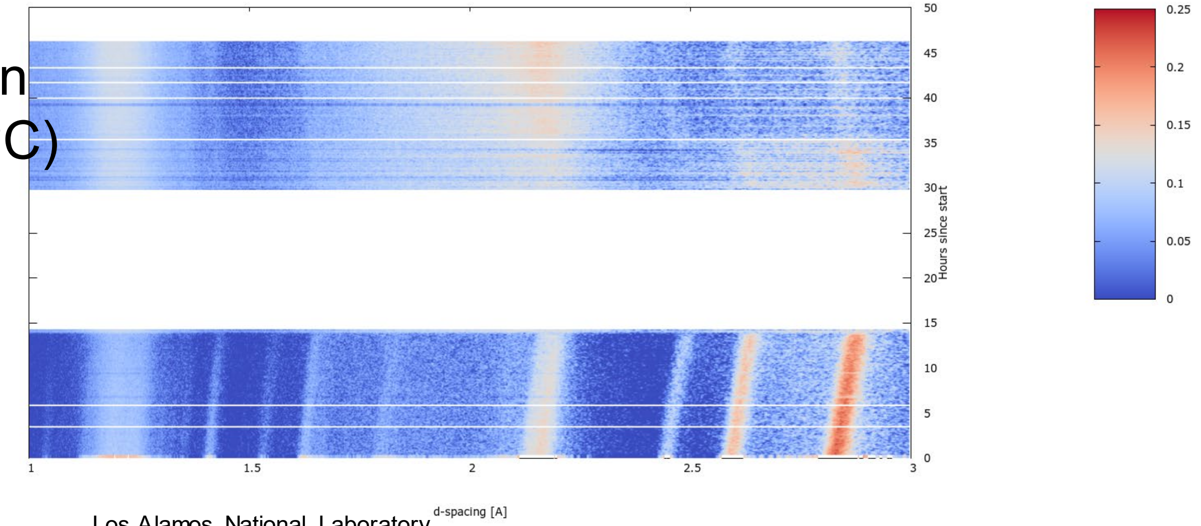


Vogel et al. JOM 2021

Diffraction data analysis

- Diffraction patterns collected with 5 minutes count time (ΔT=5°C)
- Diffraction from UCl_3 during heating allows to derive crystallographic parameters of crystalline phase (Rietveld analysis)
- Pair distribution function analysis needs development (container signal)
- ~160 patterns analyzed until melting
- Melting occurs within one 5 minute run at ~839°C (DSC.:835°C)
- UCl_3 is hexagonal, space group P 6_3/m
- U on 1/3, 2/3, ¼
- Cl on ~0.39, ~0.30, ¼

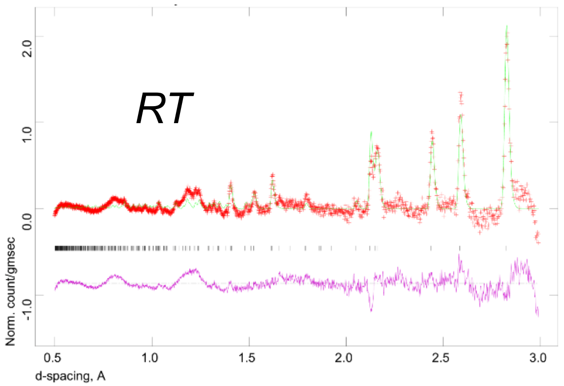


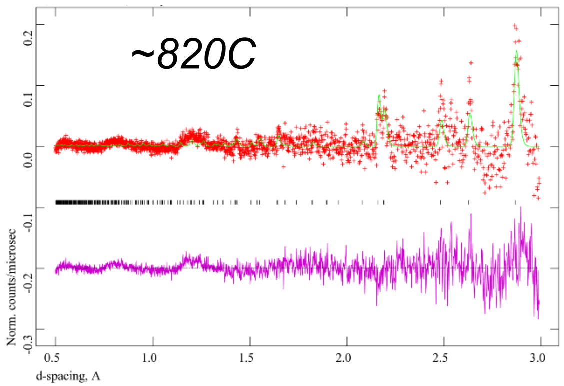


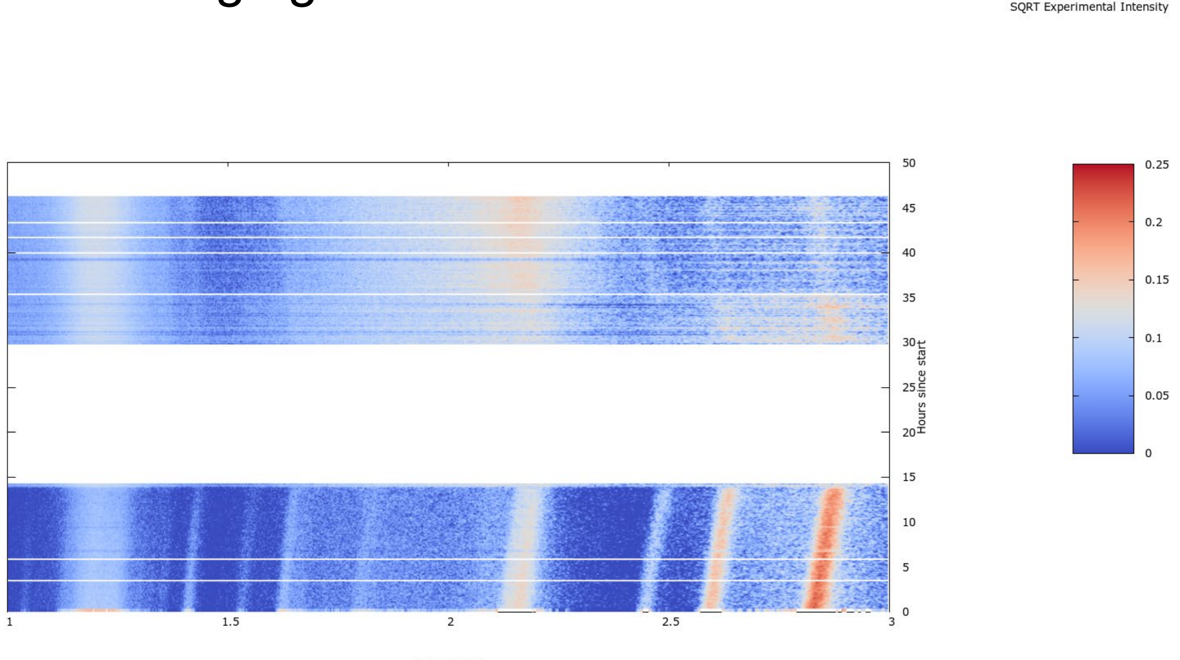
Vogel et al. JOM 2021

Diffraction data analysis

- Rietveld analysis of data in the crystalline state
- Short integration time (5 minutes) of strong thermal neutron absorber (Cl) in small diameter (3mm) double-walled SiO2 glass container ⇒ challenging…



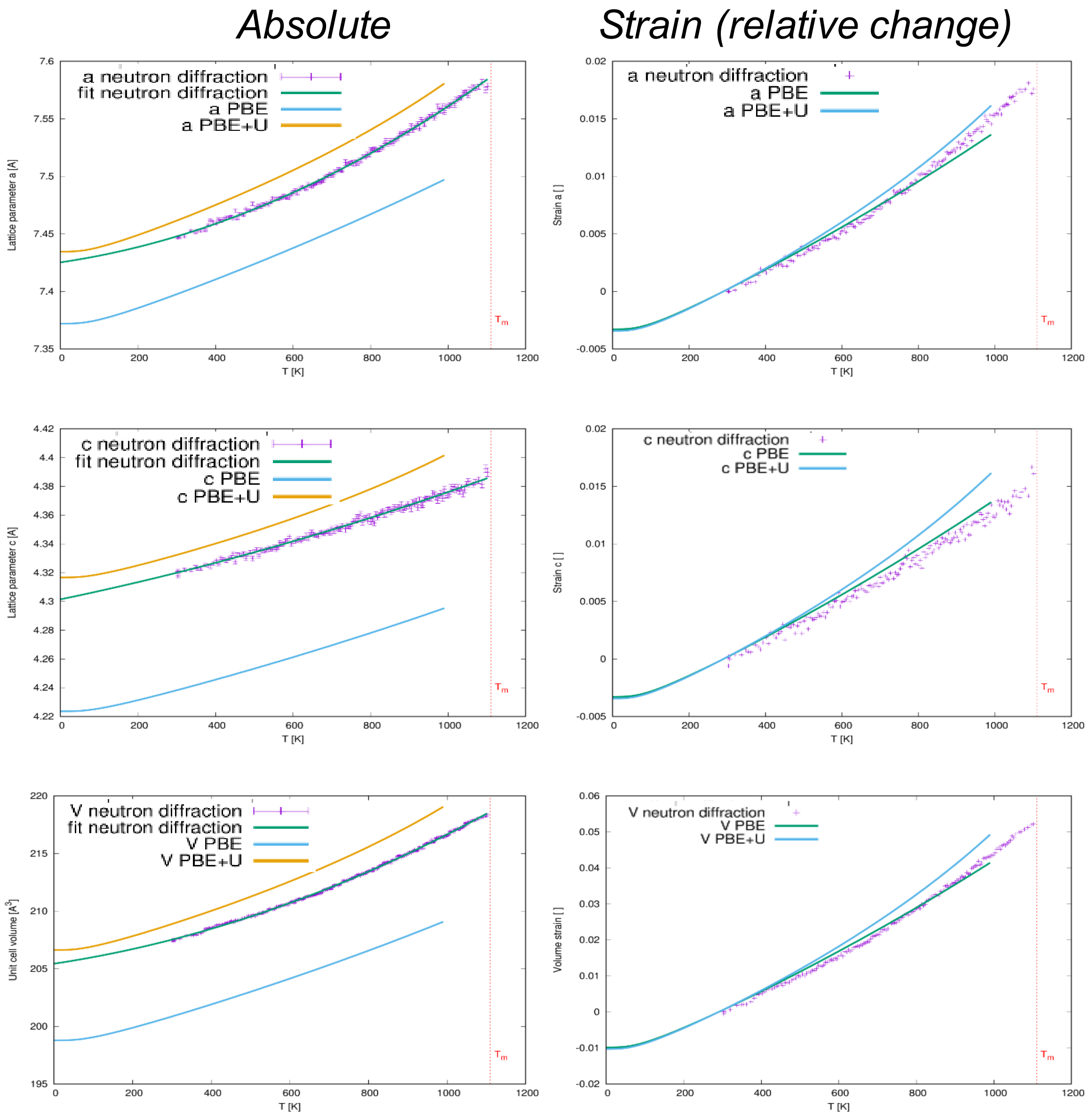




Vogel et al. JOM 2021

Comparison of lattice parameters with DFT

- Absolute *a, c,* and unit cell volume measured with neutron diffraction
- Same parameters predicted with DFT with and without Hubbard on-site correction on 5f orbitals of uranium atoms
- Correction improves agreement of absolute lattice parameters
- *a* lattice parameter has stronger contribution from quadratic term than *c* lattice parameter
- Both a and c expand ~1.8% before melting
  ⇒ Overall anisotropy small
  ⇒ Cracking due to anisotropy unlikely
- First determination of thermal expansion behavior of UCl_3 to the best of our knowledge



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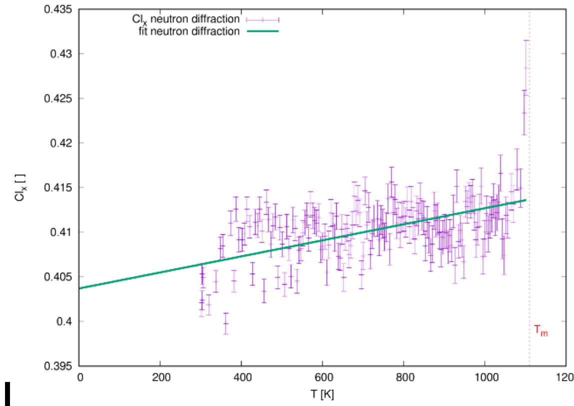
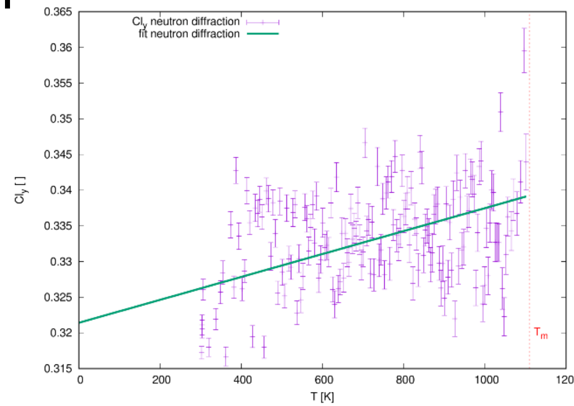
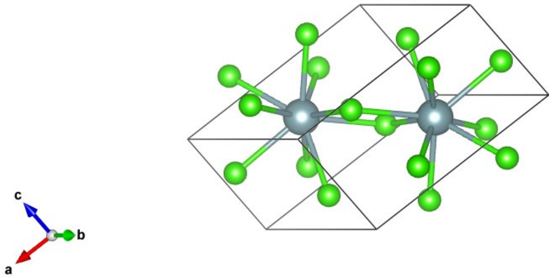
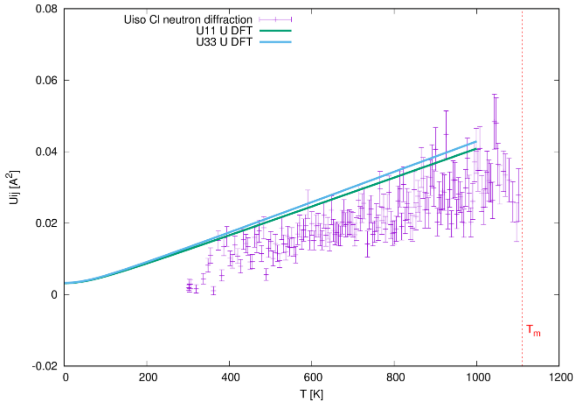
Results atomic positions and atomic displacement parameters

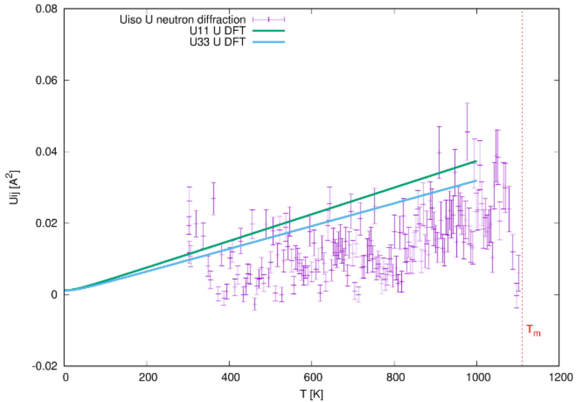
- Fractional coordinates of Cl atoms change slightly with temperature (linear fit shown)
  ⇒ Can be compared or included into future predictions
- Atomic displacement parameters are “integral of all phonons
  ⇒ Can be compared with DFT calculations (same as lattice parameters, predictions shown)
- DFT predict very small anisotropy in atomic displacement (not observable by current neutron data)







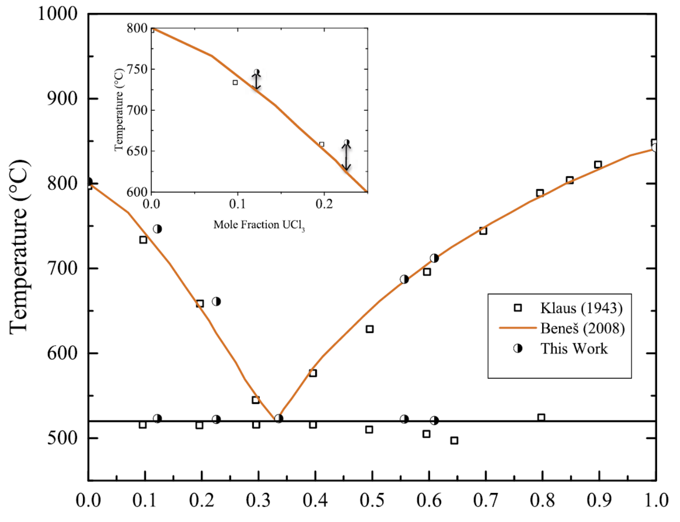




Vogel et al. JOM 2021

NaCl/UCl$_3$ Eutectic

- Excellent Rietveld fit quality for NaCl and UCl$_3$ phase mixture
- Current analysis shows very little interaction in the solid state
  ⇒ NaCl can be used as internal temperature standard
- Data allowed to refine weight fractions, lattice parameters, atom positions, anisotropic atomic displacement parameters
- Weight fractions ~constant (as they should) except for highest T data points
  ⇒ pre-melting?



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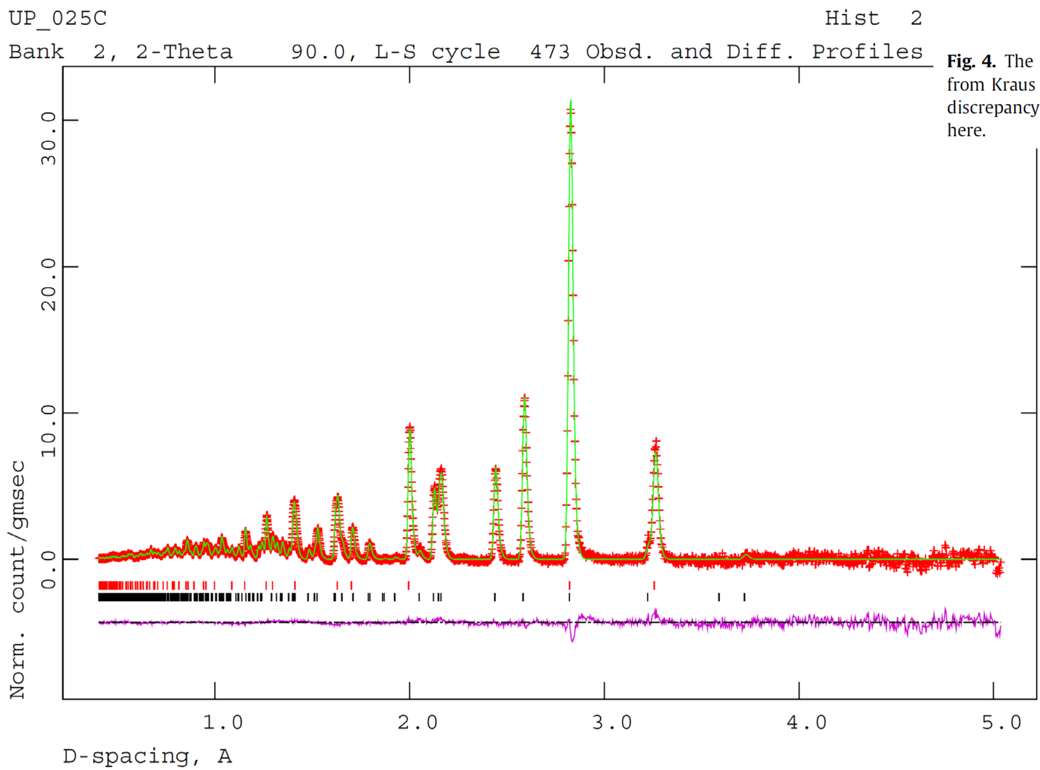
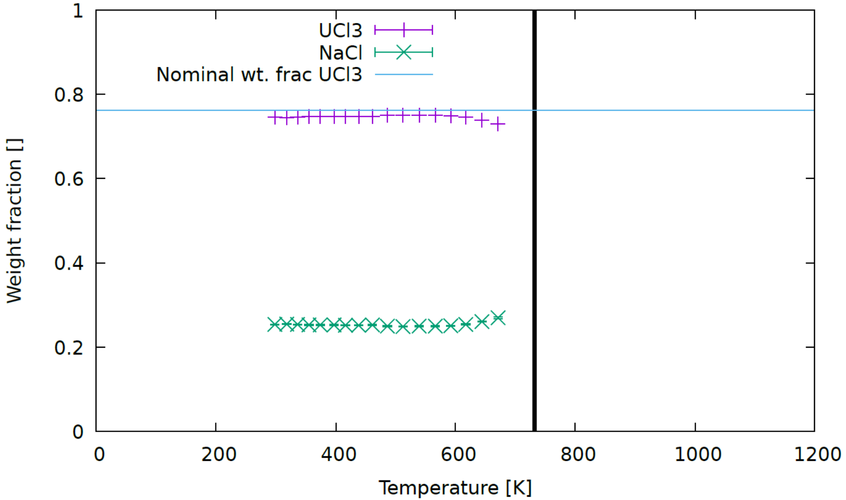
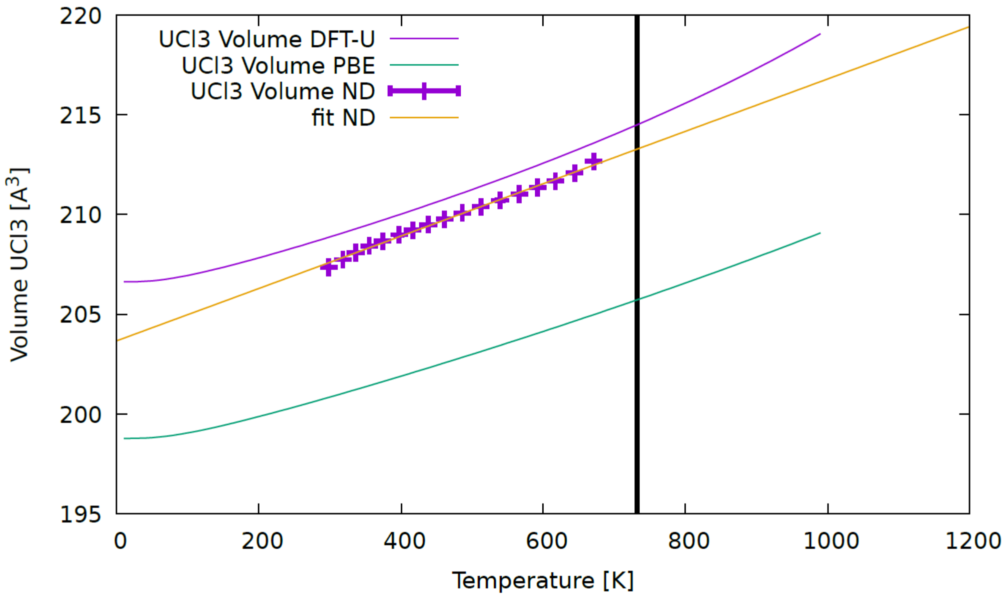
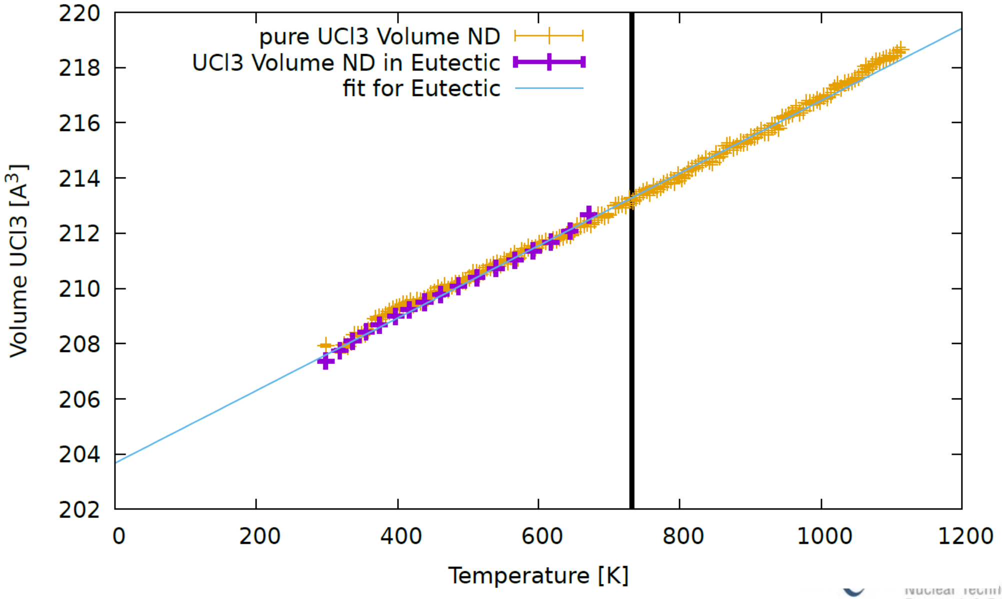




UCl_3 Unit cell volume as a function of temperature

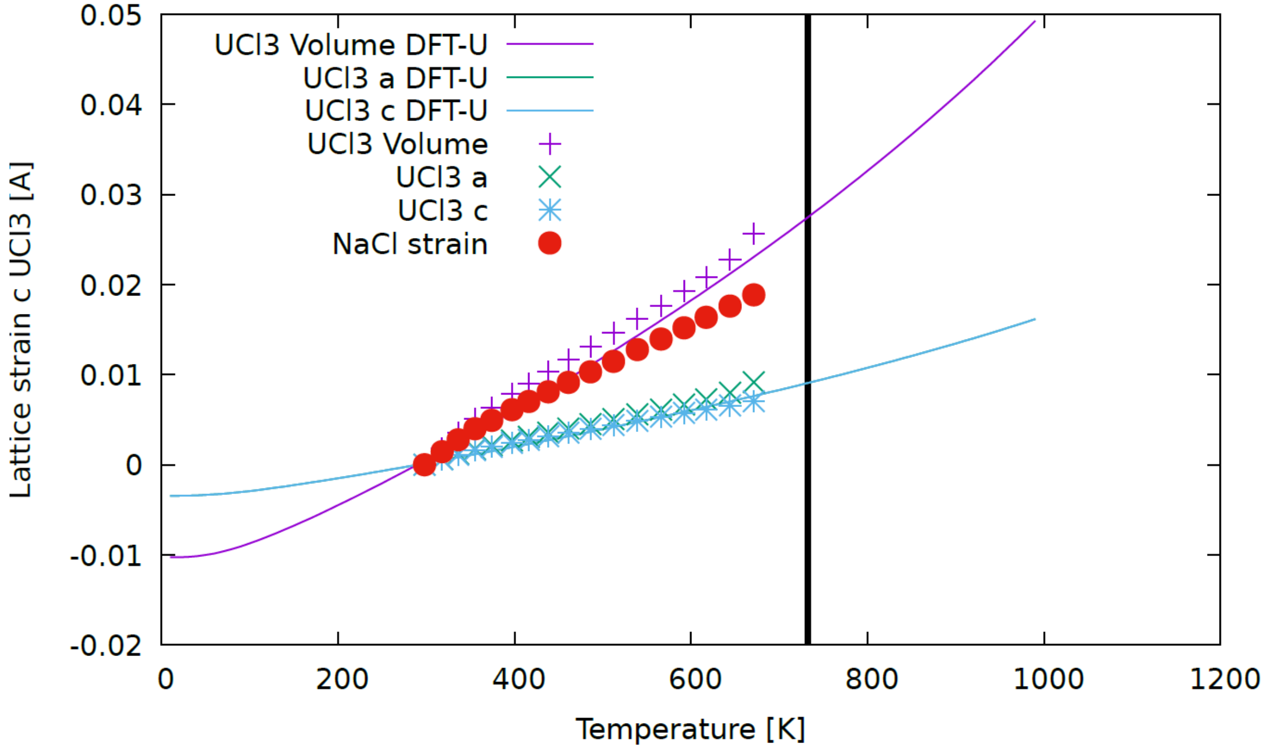
- DFT-U model predict absolute unit cell volume well, similar slope/thermal expansion
- Last data point slightly above thermal expansion fit
  $\Rightarrow$ pre-melting?
- Excellent agreement with continuous heating rate study
  $\Rightarrow$ pure UCl_3 and UCl_3 in eutectic mixture behave the same





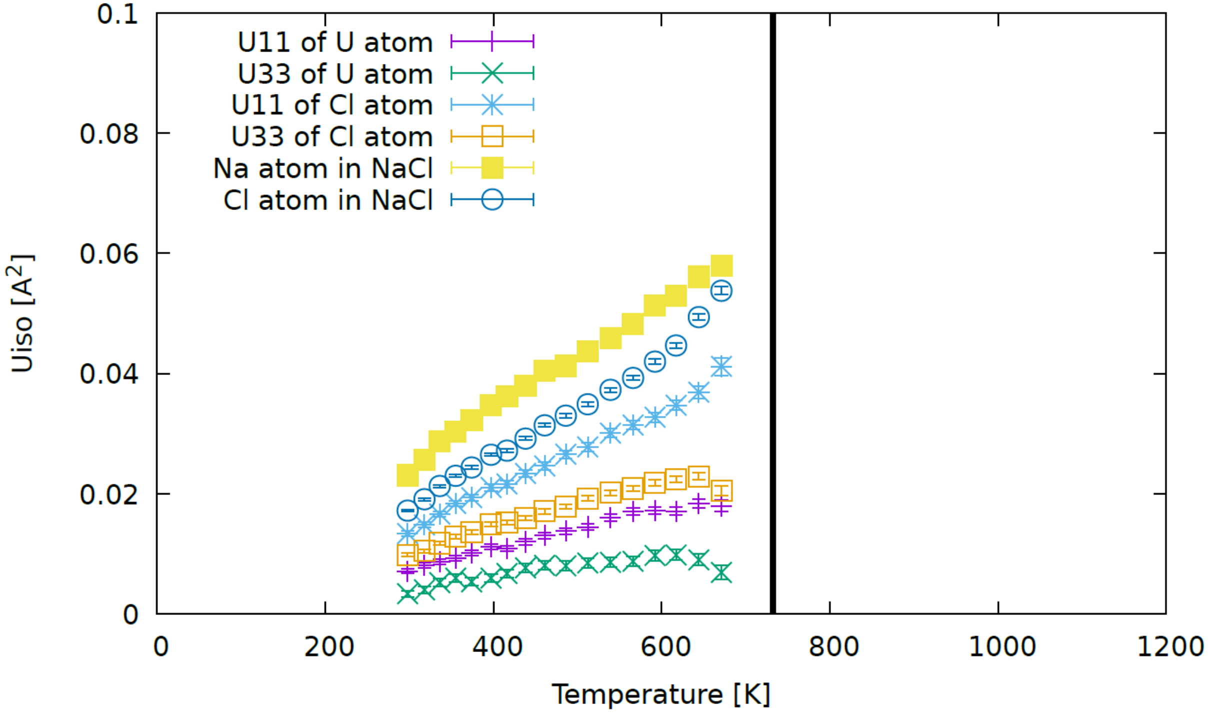
Lattice strains

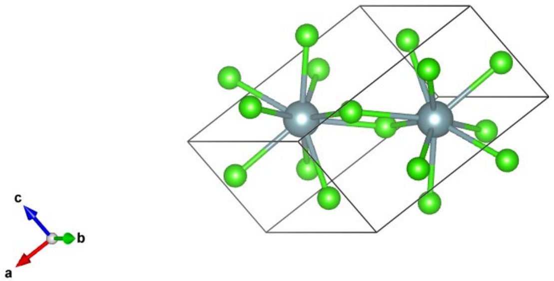
- Very small difference between expansion of lattice parameters a and c
  ⇒ small anisotropy of UCl_3
- Relative expansion very well predicted by DFT model
- NaCl expansion less than UCl_3
  ⇒ Thermal stresses between phases will build up during heating/cooling
  ⇒ Cracking in the solid state possible/likely



Anisotropic atomic displacement parameters

- Atomic displacement/thermal motion is clearly anisotropic for both U and Cl atoms
- Both Cl and U are displaced more in the a/b plane than along the c-axes
- Cl atom displacement amplitude is larger than U atoms
- Last few data points show deviation from ~linear behavior ⇒ could be pre-melting phenomenon
- Cl in NaCl shows larger amplitude than Cl atoms in UCl_3, amplitude for Na atom in NaCl even larger





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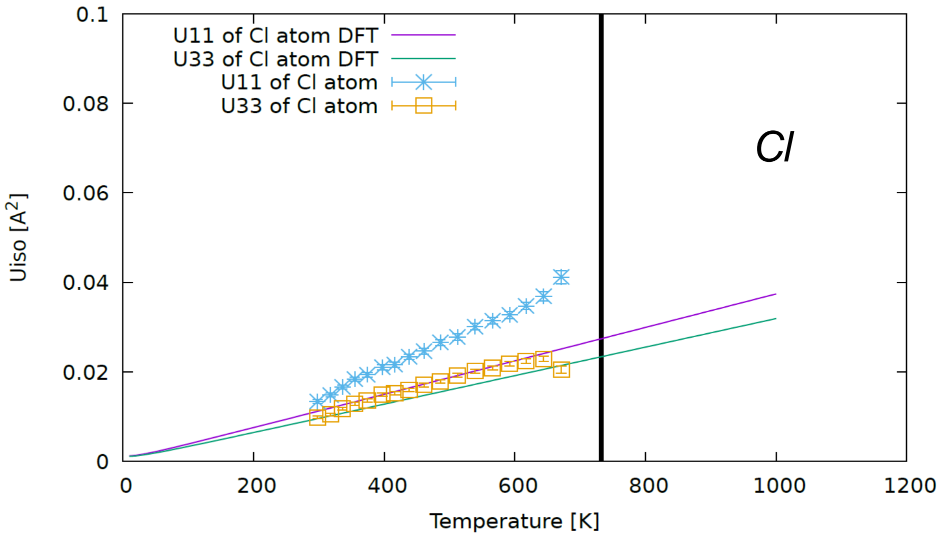
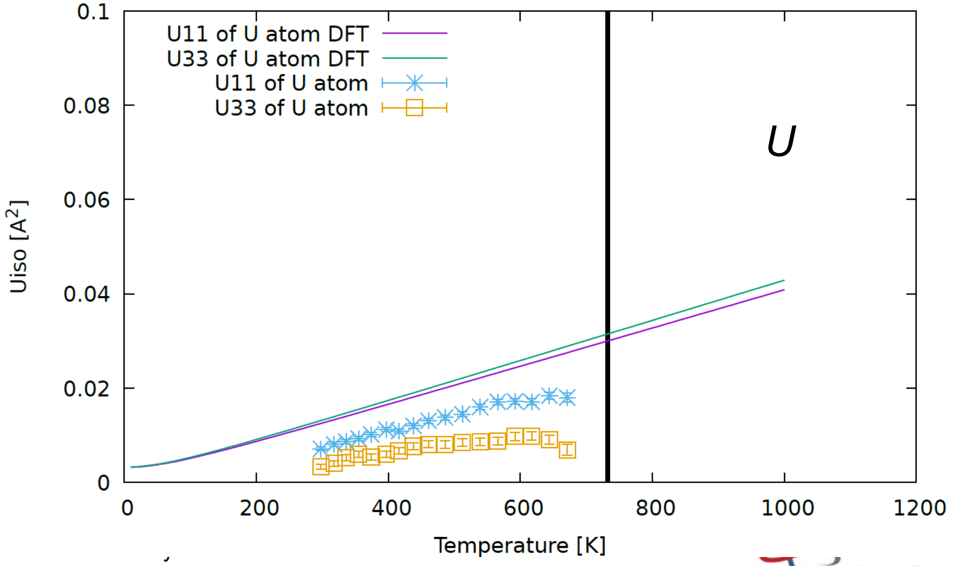
Comparison with DFT predictions

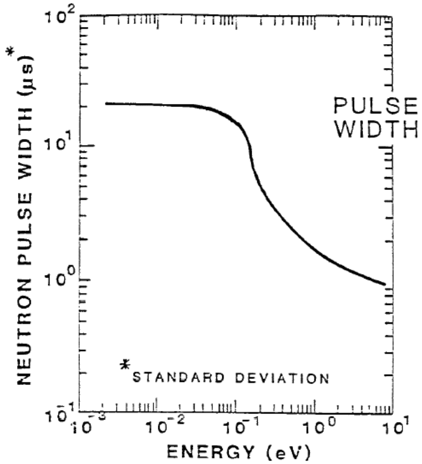
- Experimental data shows larger atomic displacement in a/b-plane than along c-axis by almost a factor of 2
- DFT predicts this for Cl atoms but with smaller difference
- DFT predicts small anisotropy of atomic displacement for U atoms with higher amplitude along c-axis
- Atomic displacement amplitudes result from phonons
  ⇒ DFT predicts phonon densities of state
  ⇒ Other thermodynamic parameters are derived from phonon predictions
  ⇒ benchmarking with experiment is important!





A few words on neutron sources (great minds think alike but not the same…)

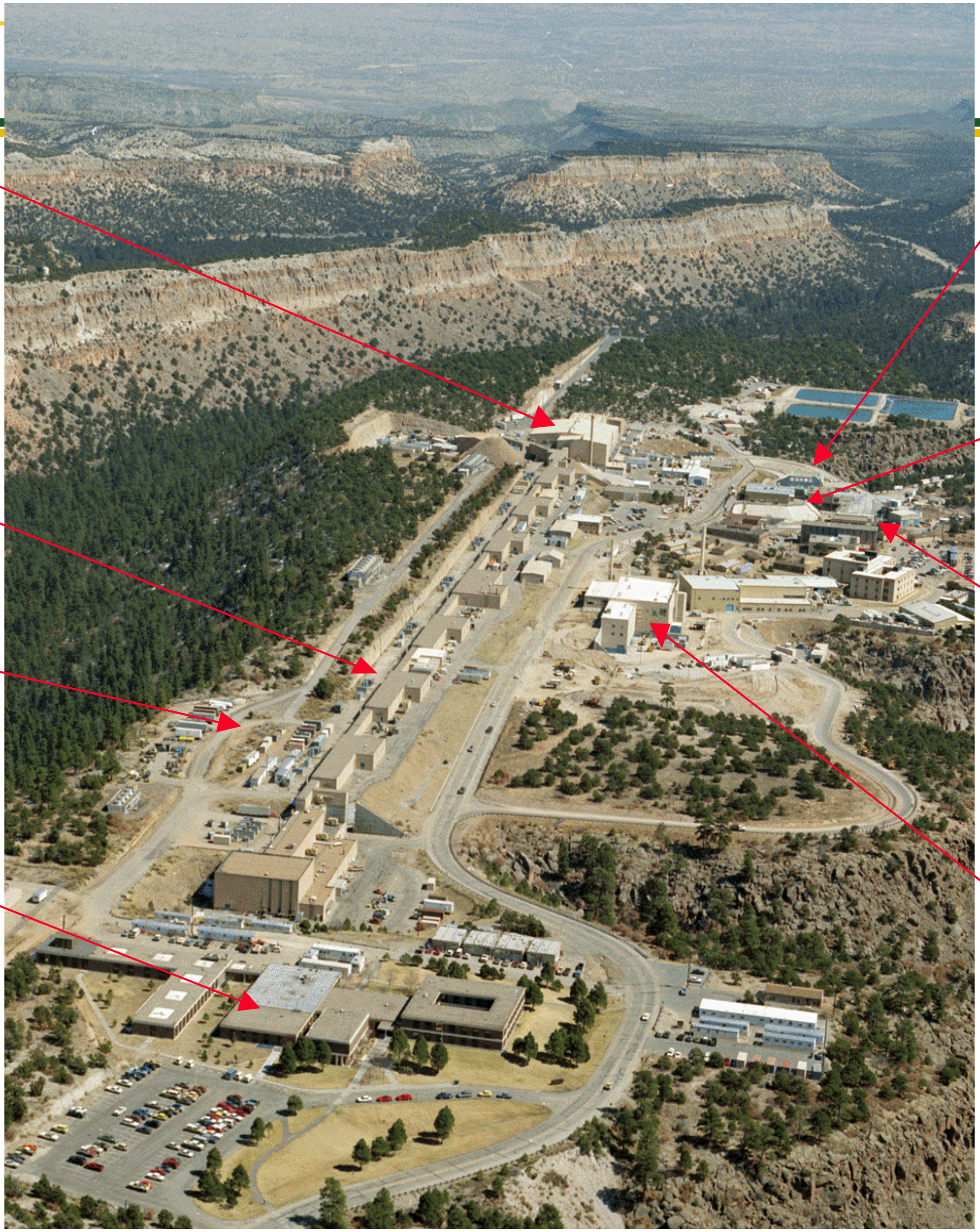
- Reactors are continuous sources
  ⇒ hard to do pulsed neutron techniques like energy-resolved neutron imaging or time-of-flight diffraction
  ⇒ fuel supply and disposal, licensing, operation are substantial headaches
  ⇒ unlikely that a lot more reactors are built
- Operational advantage: accelerator source is governed by policies more similar to an X-ray machine in a dentist office ⇒ much less headaches, less restriction on what samples can run
- **Pulsed sources are the future!**
- Spallation neutron sources exist (LANSCE/US, ISIS/UK, PSI/Switzerland, SNS/US, J-PARC/Japan, C-SNS/China) as user facilities ⇒ very large scale, hard to get beam time
- Big user facilities are built (ESS/Sweden, SNS STS/US, ISIS-2/UK…) ⇒ again very large (~2$B), hard to get beam time
- Medium-size sources exist, e.g. SARAF@SOREQ, Israel
- Small pulsed neutron sources exist, e.g. Riken Advanced Neutron Source RANS-1 to RANS-3
  - Minimum pulse length ~10 µs (good enough for TOF diffraction, not good for resonances)
  - Fits in a room
- More on compact sources: Union of Compact Accelerator-driven Neutron Sources, ucans.org



RIKEN Advanced Neutron Source

- Yoshie Otake/RIKEN is project leader (find her papers for more, e.g. Otake, Yoshie, et al. "RIKEN Compact Neutron Source Systems RANS Project." Nuclear Physics News 33.2 (2023): 17-21.)
- RANS-2 source fits in 10x5 m^2 area, large room sufficient (plus beam lines), RANS-1 about twice as large, RANS-3 designed for truck operation
- **Moderator can be changed on the fly, flexible setup**
  ⇒ switch from thermal for diffraction to cold for phase contrast imaging, high flux/low resolution to medium flux/high resolution etc.
- Demonstrated among others
  - **Radiography, including phase contrast imaging** (Takano, Hidekazu, et al. "Demonstration of Neutron Phase Imaging Based on Talbot–Lau Interferometer at Compact Neutron Source RANS." Quantum Beam Science 6.2 (2022): 22.)
  - **Time-of-flight diffraction including texture measurements** (Xu, P. G., et al. "In-house texture measurement using a compact neutron source." Journal of Applied Crystallography 53.2 (2020): 444-454.
- Promising source!

LANSCE – Los Alamos Neutron Science Center



Proton Radiography, formerly Los Alamos Meson Physics Facility (LAMPF) Site of proposed MaRIE facility

800- MeV linear proton accelerator (125μA)

Isotope Production Facility (IPF)

LANSCE Visitor Center

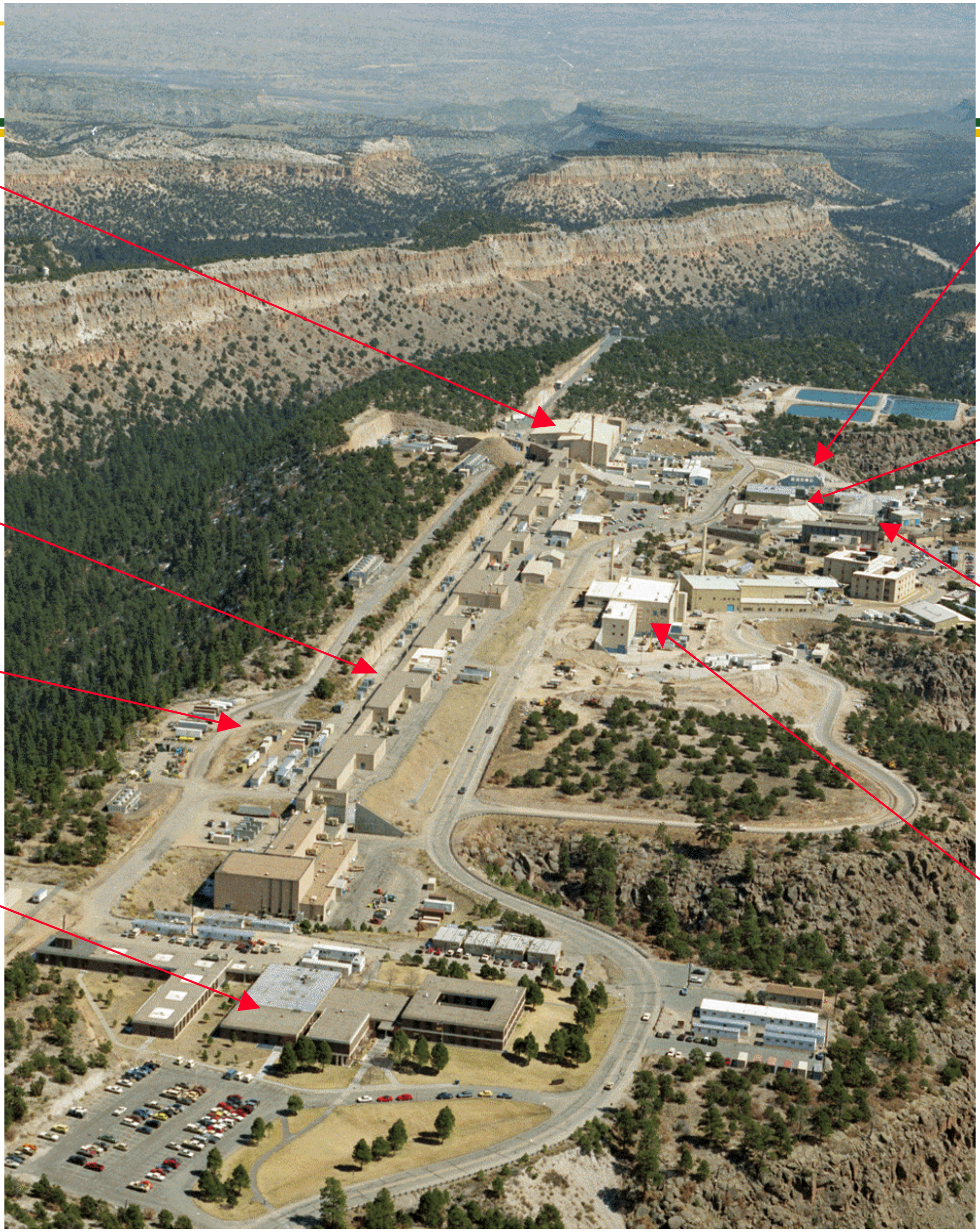
Manuel Lujan Jr. Neutron Scattering Center

Proton Storage Ring (PSR)
⇒ Pulse generation for MLNSC

Weapons Neutron Research (WNR)

Accelerator-based Production of Tritium (APT) and Low-Energy Demonstration Accelerator (LEDA), both decomissioned

LANSCE – Los Alamos Neutron Science Center



Proton Radiography, formerly Los Alamos Meson Physics Facility (LAMPF) Site of proposed MaRIE facility

800- MeV linear proton accelerator ($125\mu A$)

Isotope Production Facility (IPF)

LANSCE Visitor Center

Proton storage ring needed to make short pulses, cost >$100M – LDNS does not need that!

Proton Storage Ring (PSR)
⇒ Pulse generation for MLNSC

Weapons Neutron Research (WNR)

Isotope production, proton radiography, use of neutrons of all energy ranges could be done with one source, especially if similar detector technology can be used for all (Losko camera)

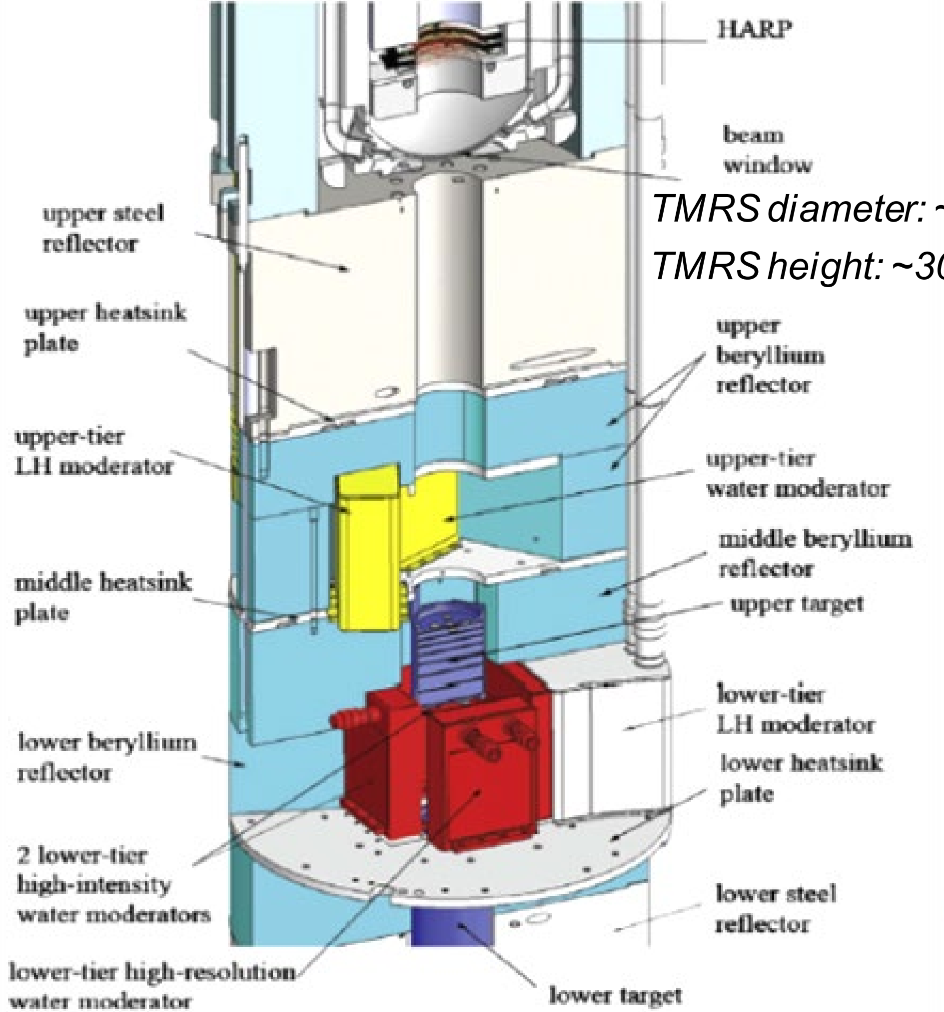
Conventional Pulsed Neutron Source: LANSCE

LANSCE – 800 MeV linear proton accelerator, ½ mile long, 100 μA on target, 20 Hz, spallation

⇒ >$1B investment, ~$10M for new target (plus installation)

⇒ ~100 people to operate just the source

⇒ ~$1M/month electricity bill

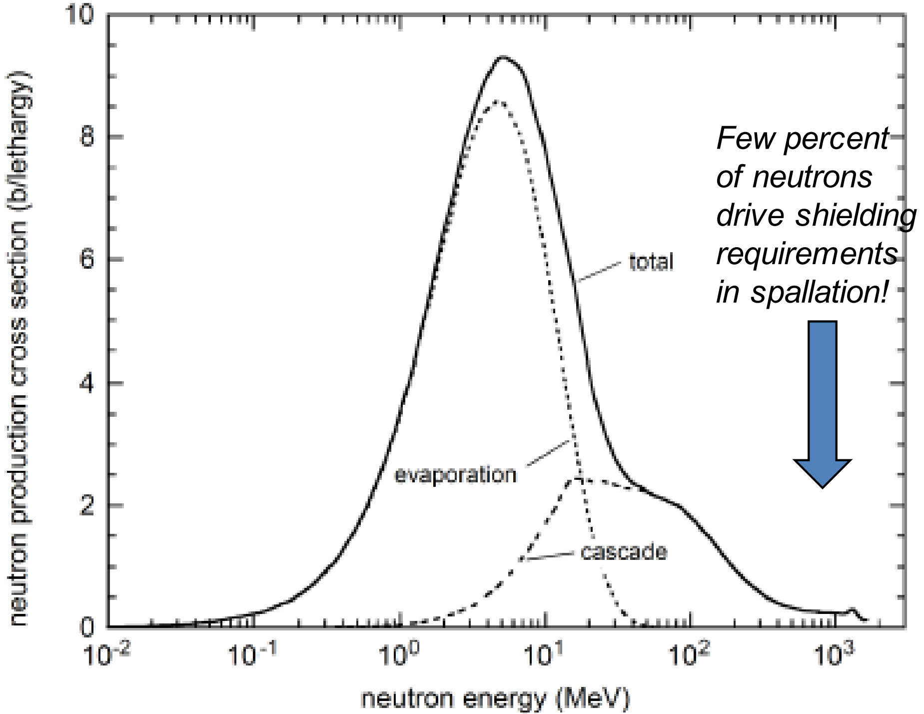


TMRS diameter: ~60 cm

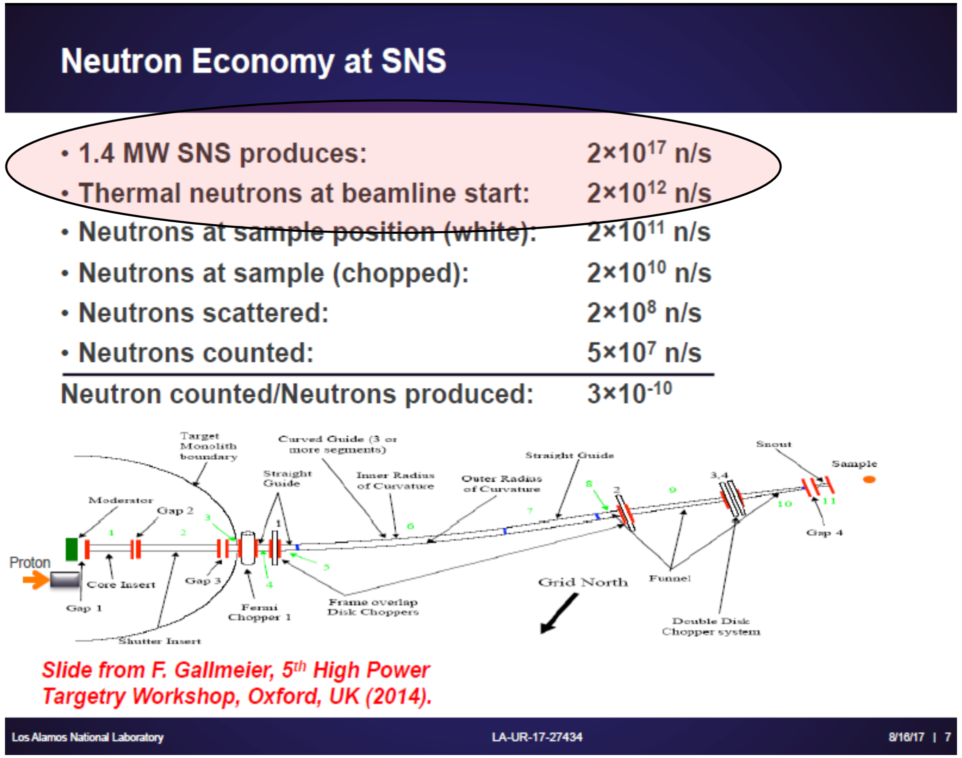
TMRS height: ~300 cm

Neutron Production LDNS vs. SNS

Neutron production cross section for 1.7-GeV protons on tungsten



Few percent of neutrons drive shielding requirements in spallation!



Neutron Economy at SNS

- 1.4 MW SNS produces: 2×10^{17} n/s
- Thermal neutrons at beamline start: 2×10^{12} n/s
- Neutrons at sample position (white): 2×10^{11} n/s
- Neutrons at sample (chopped): 2×10^{10} n/s
- Neutrons scattered: 2×10^{8} n/s
- Neutrons counted: 5×10^{7} n/s

Neutron counted/Neutrons produced: 3×10^{-10}

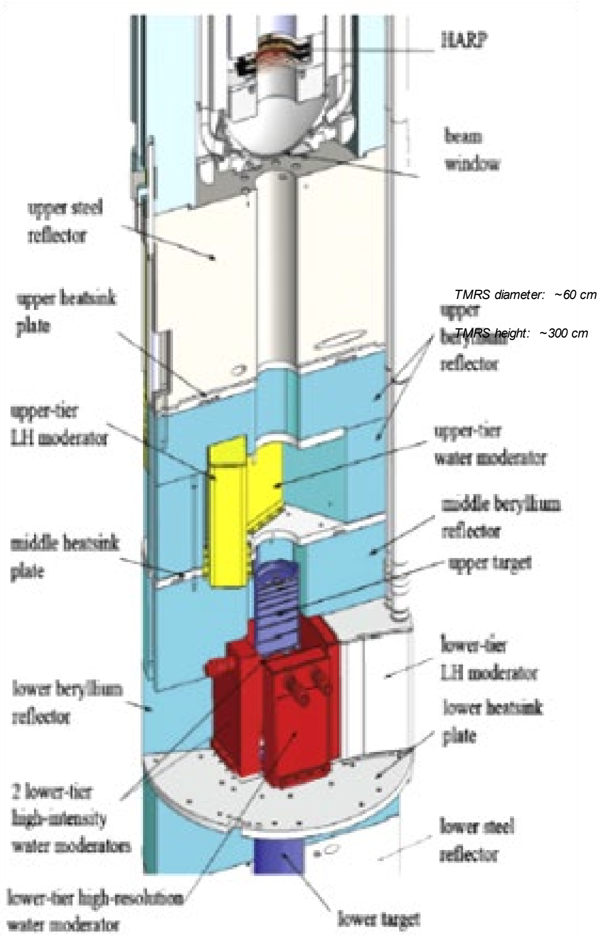
Slide from F. Gallmeier, 5th High Power Targetry Workshop, Oxford, UK (2014).

Los Alamos National Laboratory LA-UR-17-27434 8/16/17 | 7

- *LDNS utilizing deuteron breakup (or photoneutrons) requires much less shielding than spallation neutron source ⇒ sample can be closer to the source, $1/L^2$!!!*
- *Neutrons produced with directionality provide ~orders of magnitude better source-to-moderator coupling*

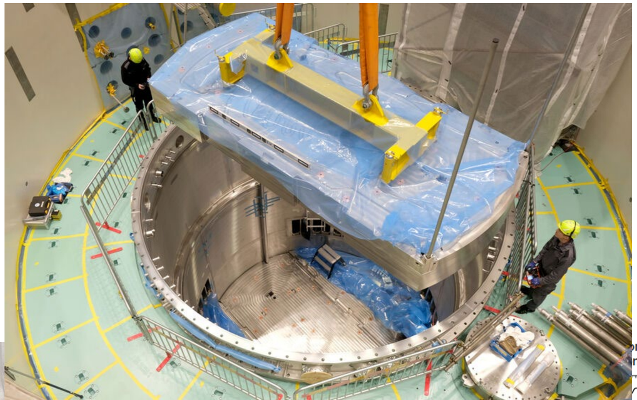
How far away are we from Laser-LANSCE?

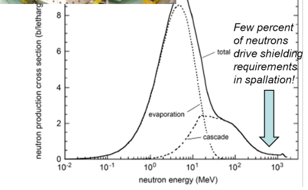
- $\sim 10^{10}$ n/pulse achieved @ TRIDENT
  (March & July 2016, 70J output energy $\Rightarrow$ 20 MeV deuterons
  70J/600 fs=0.1 PW)
- Neutrons pre-dominantly forward
  $\Rightarrow$ majority reaches moderator
  $\Rightarrow$ $\sim 10^{10}$ moderated n/pulse (~1 ns pre-moderation pulse width)
- LANSCE:
  $\Rightarrow$ 100 μA proton current @ 20 Hz, 800 MeV
  $\Rightarrow$ $\sim 3\times10^{13}$ p/pulse
  $\Rightarrow$ ~20 n/spallation process
  $\Rightarrow$ $\sim 6\times10^{14}$ n/pulse, but isotropic, out of a 10cm $\varnothing$, 20cm target)
  $\Rightarrow$ $\sim 1\times10^{13}$ moderated n/pulse (~270 ns pre-moderation pulse width,
  ~2% of neutrons cross moderator surface)
- TRIDENT – LANSCE: 10^{10} : 1×10^{13}
- Laser system, deuteron & neutron target optimizations: Factor 10 $\Rightarrow$ 10^{11}
- kJ laser: Breakup cross-section predicts factor ~20 $\Rightarrow$ 2×10^{12}
- 0.2×10^{13} moderated n/pulse feasible (have 5 lasers?)
- Smallest source-to-sample distance at LANSCE: ~6 m
- Source-to-sample distance for laser-driven source: <2 m ($1/L^2$ ~factor 10)
  $\Rightarrow$ setup for e.g. resonance imaging/NRTA/mini-HIPPO possible!

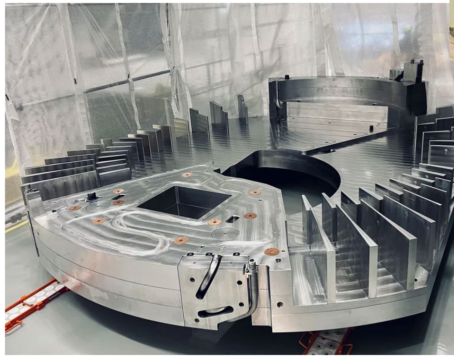


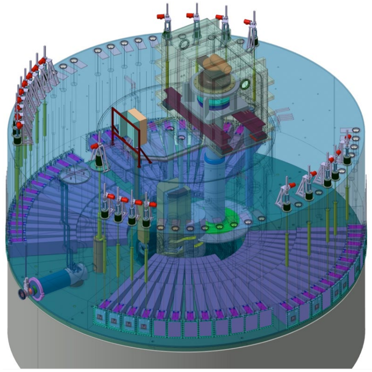
Cost Drivers for Spallation Sources

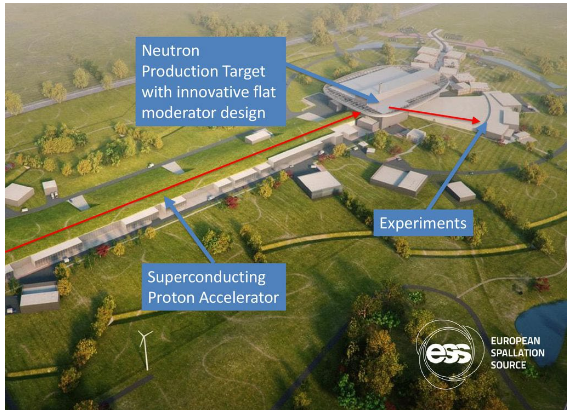
- **Linear accelerator**
  - Lots of energy used to keep protons together over ~km distance
  - Proton storage ring needed to compress
- **Target system**
  - Isotope inventory needs to be monitored
  - Complex, heavy & expensive system in itself
- **Target building**
  - Shielding must be designed for neutrons of energy close to proton energy
  - Expensive, heavy, drives closest sample position to >15m from source ($1/L^2$ bites…)
  - Significant amounts of funding to manage sagging of floor in the building to keep beamlines aligned
  - Huge chunk of cost of source
- **LDNS would not need any of that**







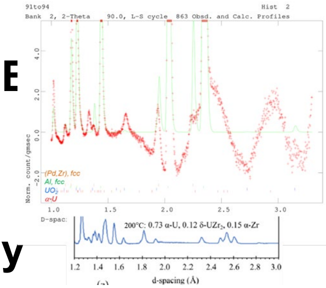


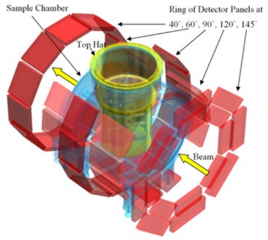


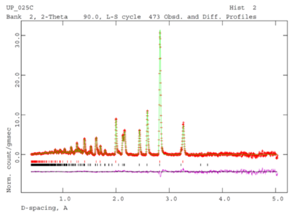
Picture credit: J. Womersley ESS slides, August 2018 & ESS website "How it works

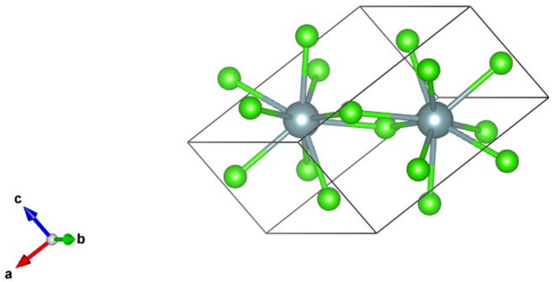
Summary & Conclusions

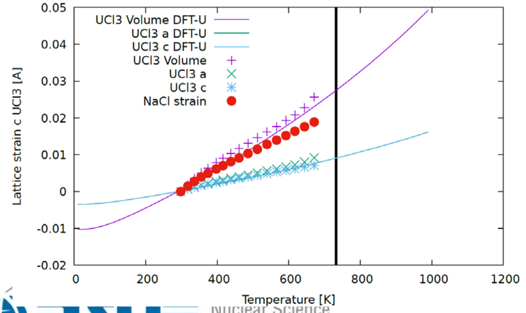
- Pulsed neutron offer bulk (cm^3 volumes) characterization capabilities for irradiated (and fresh) nuclear fuels
- Irradiation happens at locations different from location of pulsed neutron sources
- Some materials (uranium, plutonium, chlorides, highly radioactive etc.) are not allowed at neutron user facilities (HFIR reactor at Oak Ridge irradiates fuels, but cannot bring those samples to SNS)
- Cannot spend $2B to build another SNS at locations where irradiation happens (Idaho National Lab)
  ⇒ need for small compact sources such as laser-driven neutron sources (or RANS)
- Bulk characterization increases parameter space utilized for selection of volumes for destructive PIE
  ⇒ these techniques increase value of expensive irradiation campaigns
- Resonance techniques require short pulses that e.g. RANS cannot provide
- Pulsed neutron methods (energy-resolved neutron imaging, diffraction) were demonstrated on many fresh fuels and some irradiated fuels
  ⇒ Now we need those laser-driven neutron sources!

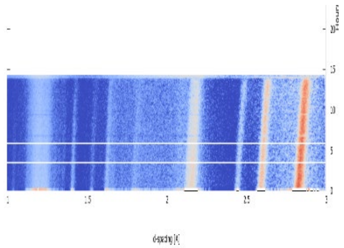












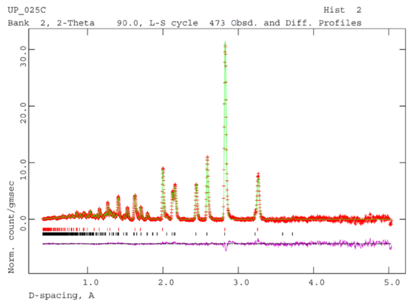
Advanced Fuels Campaign

LANSCE
Los Alamos Neutron Science Center

NSUF Nuclear Science User Facilities

NTRD Nuclear Technology Research & Development

Summary & Outlook



- Characterization of molten salts (including irradiated materials) is crucial to make molten salt reactors are reality
- Infra-structure to handle them is beyond what most neutron sources allow even without irradiated
- Solid phase data can be used to benchmark models, which are important for designing but also licensing or reactors
- Short pulse neutron sources can provide unique insight