Thermal Neutron Scattering Law Research at North Carolina State University

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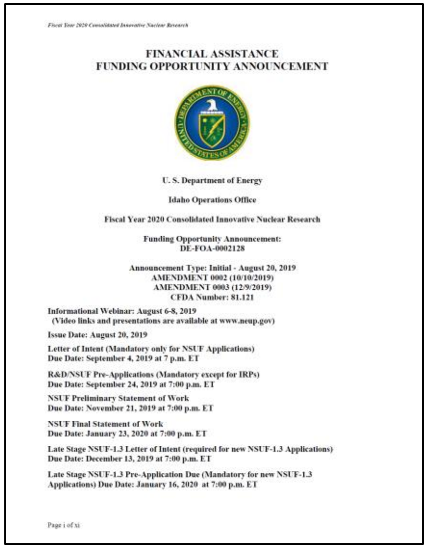
NEA Working Party on International Nuclear Data Evaluation Co-operation
May 11 – 15, 2019 • WebEx Meeting

Acknowledgment

- DOE NE through the Nuclear Energy University Program (NEUP)
- NNSA Nuclear Criticality Safety Program (NCSP)
  - in collaboration with LLNL
- Naval Nuclear Propulsion Program (NNPP)



Fiscal Year 2020 Consolidated Innovative Nuclear Research

FINANCIAL ASSISTANCE
FUNDING OPPORTUNITY ANNOUNCEMENT

U. S. Department of Energy

Idaho Operations Office

Fiscal Year 2020 Consolidated Innovative Nuclear Research

Funding Opportunity Announcement:
DE-FOA-0002128

Announcement Type: Initial - August 20, 2019
AMENDMENT 0002 (10/10/2019)
AMENDMENT 0003 (12/9/2019)
CFDA Number: 81.121

Informational Webinar: August 6-8, 2019
(Video links and presentations are available at www.neup.gov)

Issue Date: August 20, 2019

Letter of Intent (Mandatory only for NSUF Applications)
Due Date: September 4, 2019 at 7 p.m. ET

R&D/NSUF Pre-Applications (Mandatory except for IRPs)
Due Date: September 24, 2019 at 7:00 p.m. ET

NSUF Preliminary Statement of Work
Due Date: November 21, 2019 at 7:00 p.m. ET

NSUF Final Statement of Work
Due Date: January 23, 2020 at 7:00 p.m. ET

Late Stage NSUF-1.3 Letter of Intent (required for new NSUF-1.3 Applications)
Due Date: December 13, 2019 at 7:00 p.m. ET

Late Stage NSUF-1.3 Pre-Application Due (Mandatory for new NSUF-1.3 Applications) Due Date: January 16, 2020 at 7:00 p.m. ET

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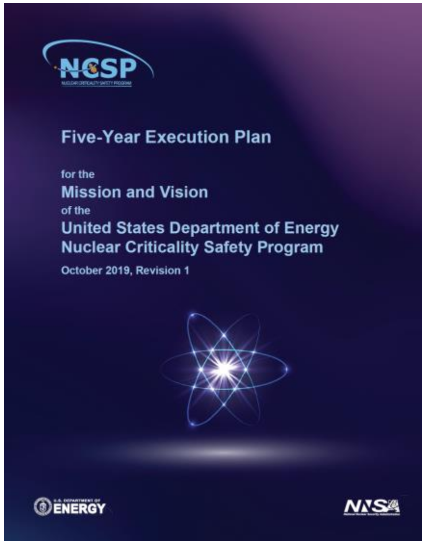
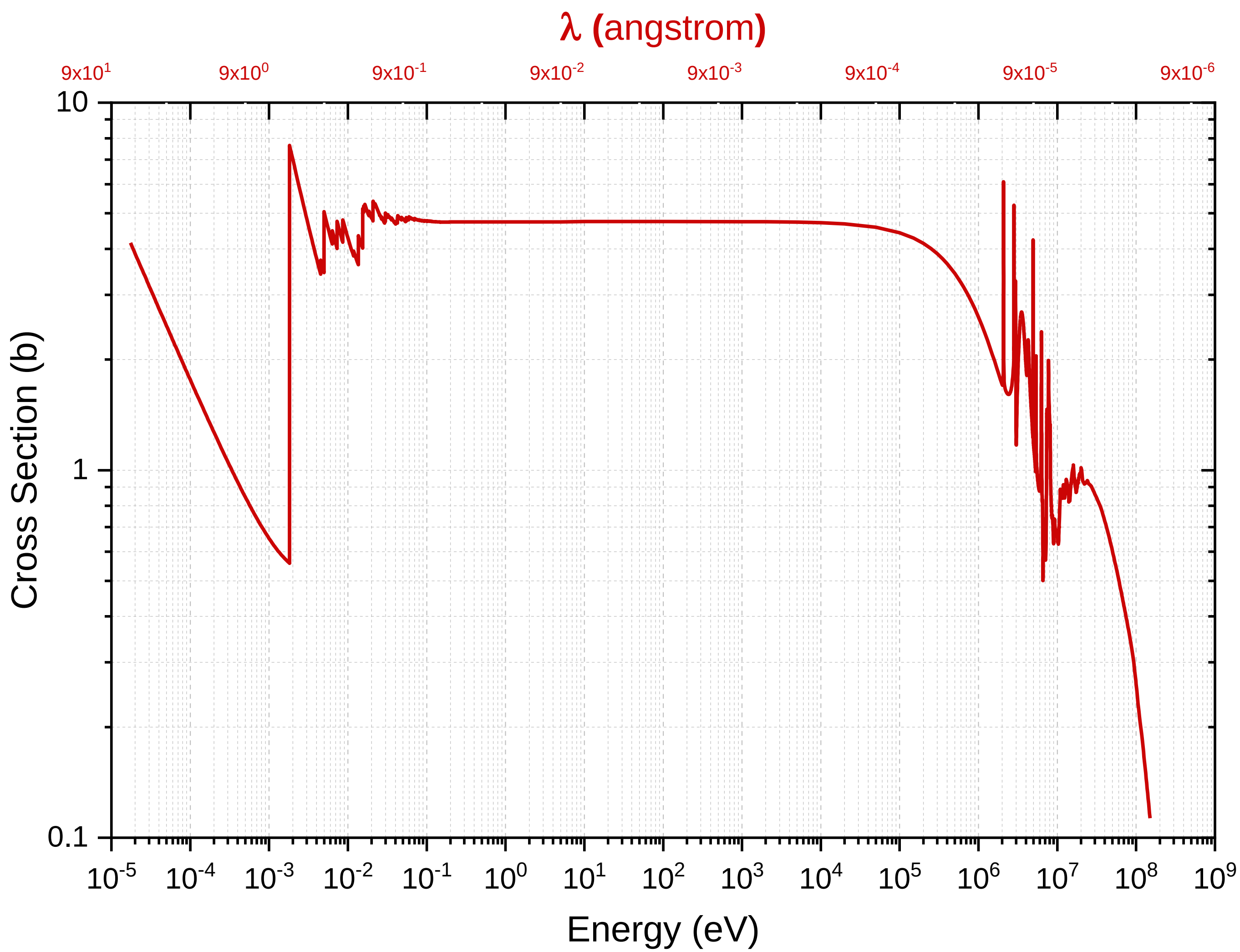


LEIP LABORATORIES

Neutron Interactions – Carbon



Neutron Thermalization

Using first Born approximation combined with Fermi pseudopotential, it can be shown that the double differential scattering cross section has the form

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left\{\sigma_{coh} S(\vec{\kappa},\omega) + \sigma_{incoh} S_s(\vec{\kappa},\omega)\right\}$$

The dynamic structure factor $S(\vec{\kappa},\omega)$ is composed of two parts

$$S(\vec{\kappa},\omega) = S_s(\vec{\kappa},\omega) + S_d(\vec{\kappa},\omega)$$

Van Hove's space-time formulation

$$I(\vec{\kappa},t) = \int G(\vec{r},t)\exp(i\vec{\kappa}\cdot\vec{r})d\vec{r}$$

$$S(\vec{\kappa},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} G(\vec{r},t)e^{i(\vec{\kappa}\cdot\vec{r}-\omega t)}d\vec{r}dt$$

where $G(\vec{r},t)$ is the *dynamic pair correlation function* and can be expressed in terms of time dependent atomic positions.

Since 1960s
GASKET
NJOY/LEAPR
INCOHERENT APPROXIMATION

$$S_s(\alpha,\beta) = k_B T \cdot S_s(\vec{\kappa},\omega)$$

$$\left.\frac{d^2\sigma}{d\Omega dE'}\right|_{inelastic} = \frac{\sigma}{2k_B T}\sqrt{\frac{E'}{E}}S_s(\alpha,\beta)$$

$\beta = \frac{E - E'}{k_B T}$ Energy transfer

$\alpha = \frac{(E + E' - 2\sqrt{EE'}\cos\theta)}{k_B T}$ Momentum transfer

The scattering law (TSL) is the Fourier transform of a Gaussian correlation function

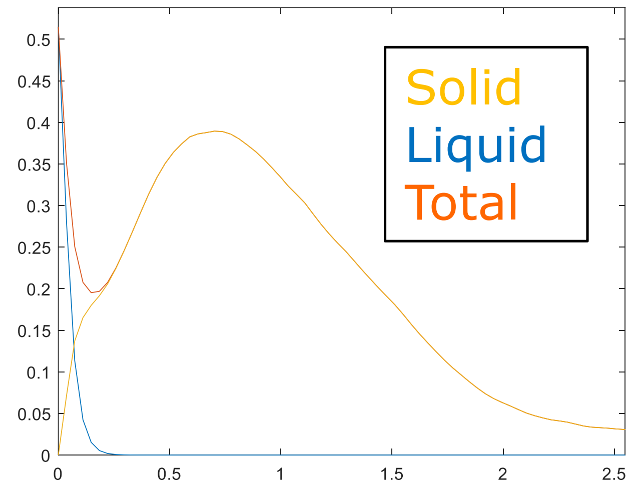
$$S_s(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\beta t} e^{-\gamma(t)} dt$$

$$\gamma(t) = \frac{\alpha}{2}\int_{-\infty}^{\infty} \frac{\rho(\beta)}{\beta \sinh(\beta/2)}\left[1 - e^{-i\beta t}\right] e^{\beta/2} d\beta$$

$\rho(\beta)$ – density of states (e.g., phonon frequency distribution)

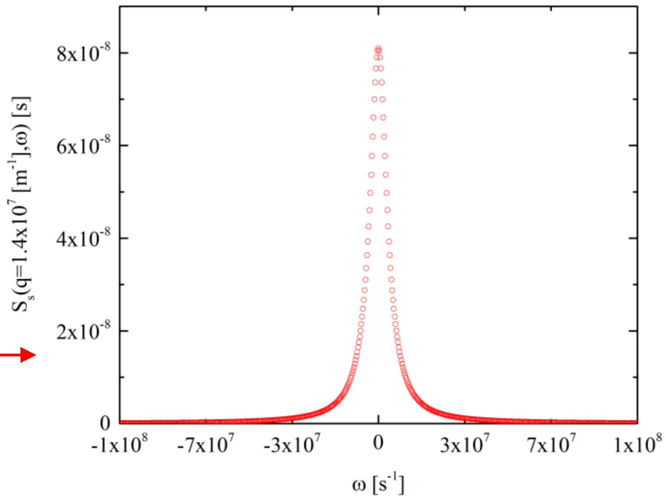
Thermalization in Liquids

- **Separation** of the diffusive DOS from the continuous (solid) DOS



- **Convolution** of the solid and liquid TSL components

$$S_{total}(\alpha,\beta) = \left(S_{diff.}(\alpha,\beta) * S_{cont.}(\alpha,\beta)\right)$$



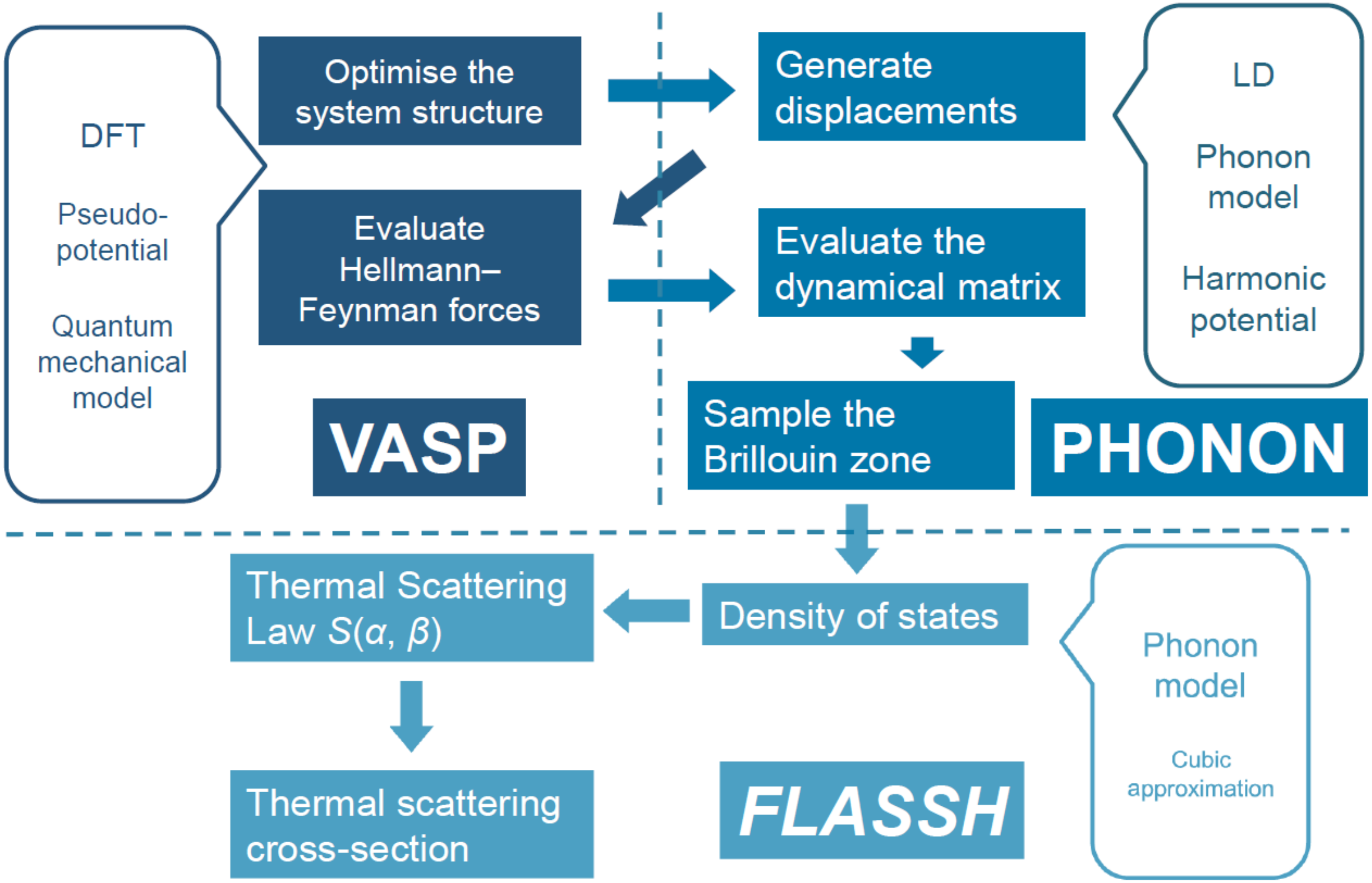
Thermal Scattering Law Analysis

- Key development in the last 20 years is the use of atomistic simulations methods to support the evaluation process
  - Produce data necessary to calculate the TSL including
    - DOS for evaluation of TSL
    - Direct access to TSL using correlation analysis
    - Support computational and experimental analysis

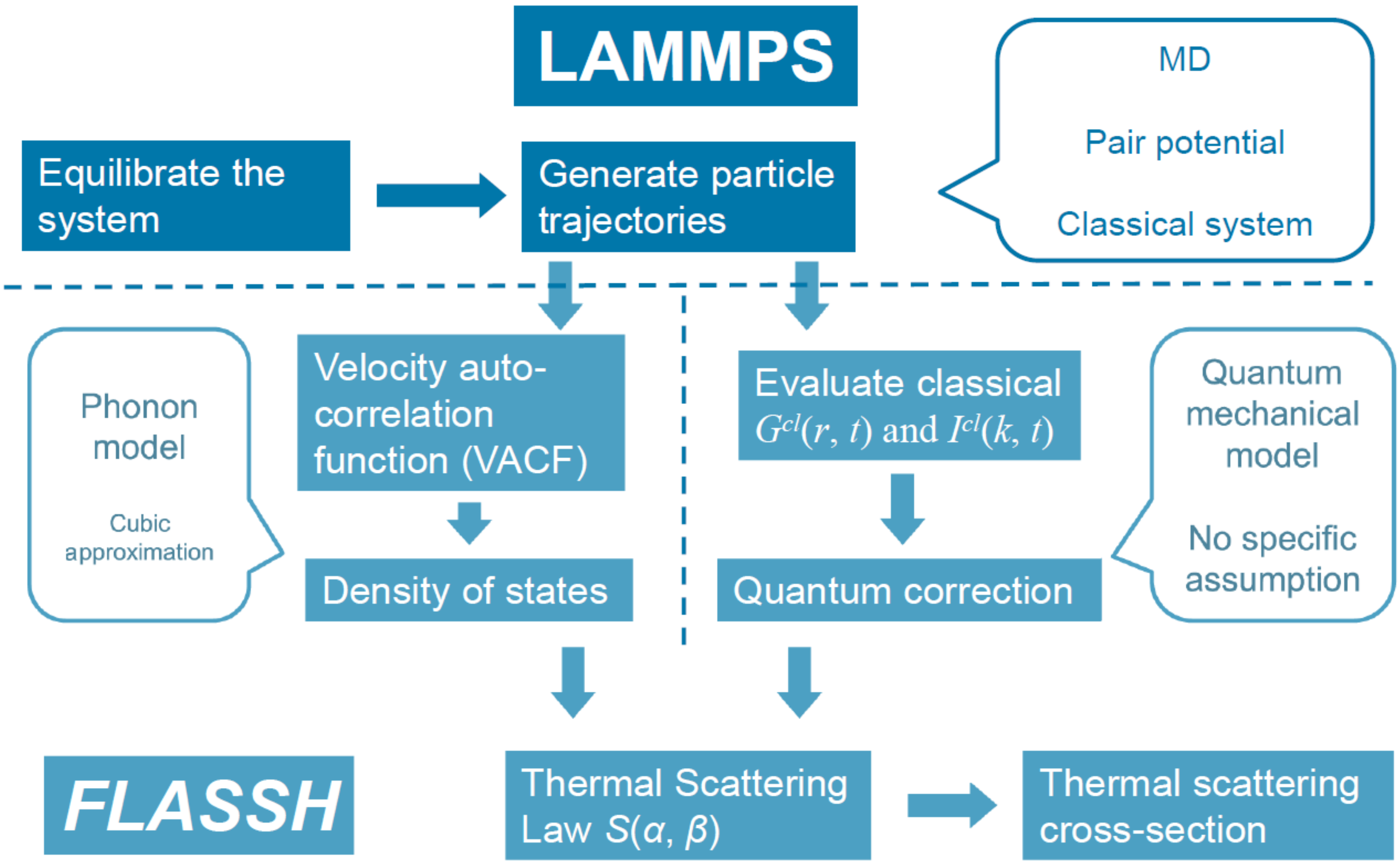
Analysis Approach

- Construct atomistic model of a material
- Verify ability of model to reproduce physical properties of the material (equilibrium conditions)
  - Density, thermal expansion, thermal conductivity,...
  - Ergodic behavior, correlations,...
- Generate input (DOS, ...) for TSL calculations
- Calculate TSL and produce thermal scattering cross sections
  - Check consistency of results with computational assumptions/models
  - Compare to experimental data

Thermal Scattering Law
DFT/LD

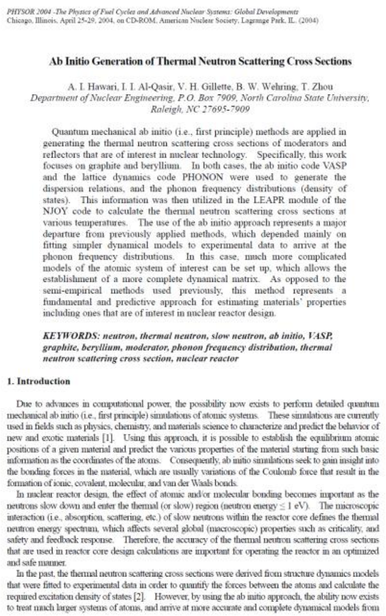


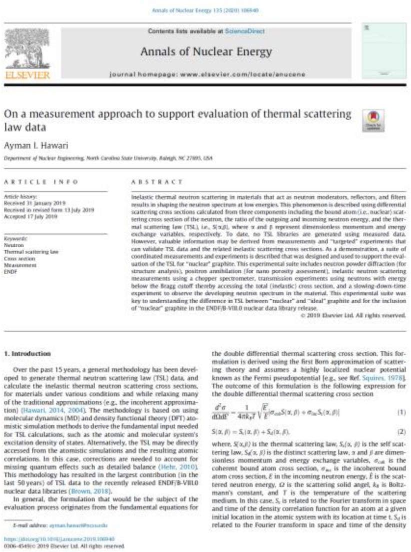
Thermal Scattering Law
MD/QM



Thermal Scattering Law Analysis

2000 - Present



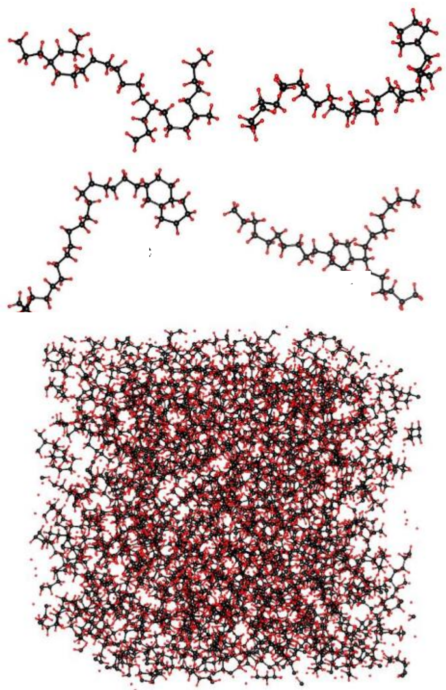


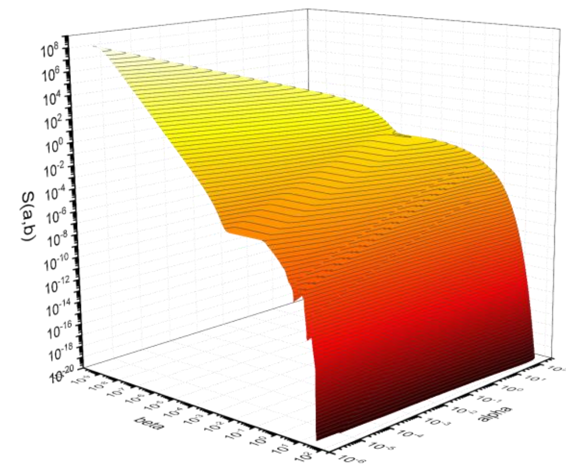
ENDF/B-VIII TSL Evaluations

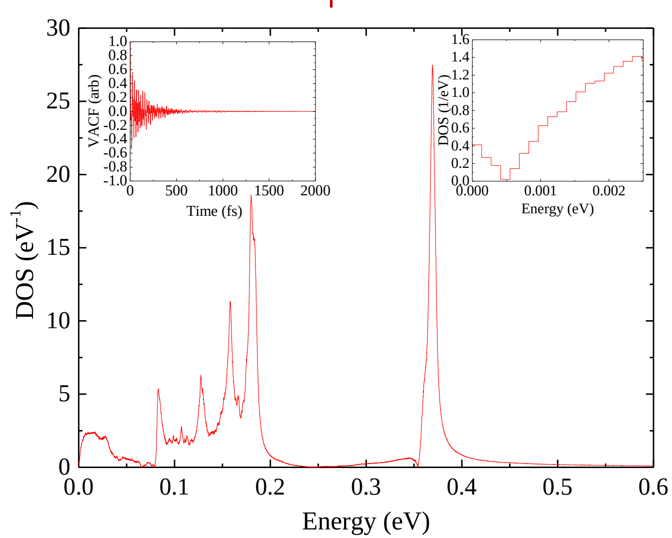
Material	Status	Evaluation Basis	Institution
Beryllium metal	tsl-Be-metal.endf	DFT/LD	NCSU
Beryllium oxide (beryllium)	tsl-BeinBeO.endf	DFT/LD	NCSU
Beryllium oxide (oxygen)	tsl-OinBeO.endf	DFT/LD	NCSU
Light water (hydrogen)	tsl-HinH2O.endf	MD	CAB
Light water ice (hydrogen)	tsl-HinIceIh.endf	DFT/LD	BAPL
Light water ice (oxygen)	tsl-OinIceIh.endf	DFT/LD	BAPL
Heavy water (deuterium)	tsl-DinD2O.endf	MD	CAB
Heavy water (oxygen)	tsl-OinD2O.endf	MD	CAB
Polymethyl Methacrylate (Lucite)	tsl-HinC5O2H8.endf	MD	NCSU
Polyethylene	tsl-HinCH2.endf	MD	NCSU
Crystalline graphite	tsl-graphite.endf	MD	NCSU
Reactor graphite (10% porosity)	tsl-reactor-graphite-10P.endf	MD	NCSU
Reactor graphite (30% porosity)	tsl-reactor-graphite-30P.endf	MD	NCSU
Silicon carbide (silicon)	tsl-CinSiC.endf	DFT/LD	NCSU
Silicon carbide (carbon)	tsl-SiinSiC.endf	DFT/LD	NCSU
Silicon dioxide (alpha phase)	tsl-SiO2-alpha.endf	DFT/LD	NCSU
Silicon dioxide (beta phase)	tsl-SiO2-beta.endf	DFT/LD	NCSU
Yttrium hydride (hydrogen)	tsl-HinYH2.endf	DFT/LD	BAPL
Yttrium hydride (yttrium)	tsl-YinYH2.endf	DFT/LD	BAPL
Uranium dioxide (oxygen)	tsl-OinUO2.endf	DFT/LD	NCSU
Uranium dioxide (uranium)	tsl-UinUO2.endf	DFT/LD	NCSU
Uranium nitride (nitrogen)	tsl-NinUN.endf	DFT/LD	NCSU
Uranium nitride (uranium)	tsl-UinUN.endf	DFT/LD	NCSU
Paraffinic Oil	To NNDC	MD	NCSU
Sapphire (Al2O3)	To NNDC	DFT/LD	NCSU
Molten Salt (FLiBe)	Completed	MD	NCSU

New to NNDC

Heavy Paraffinic Oil



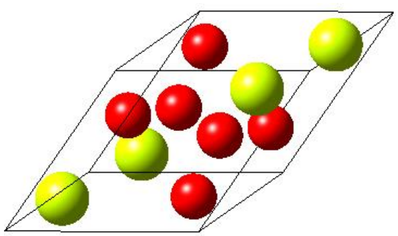


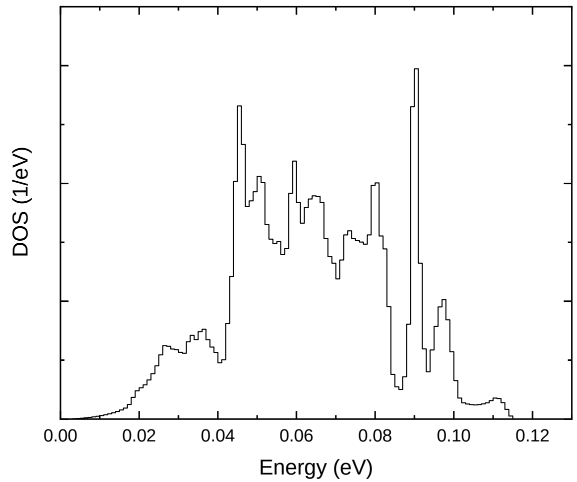


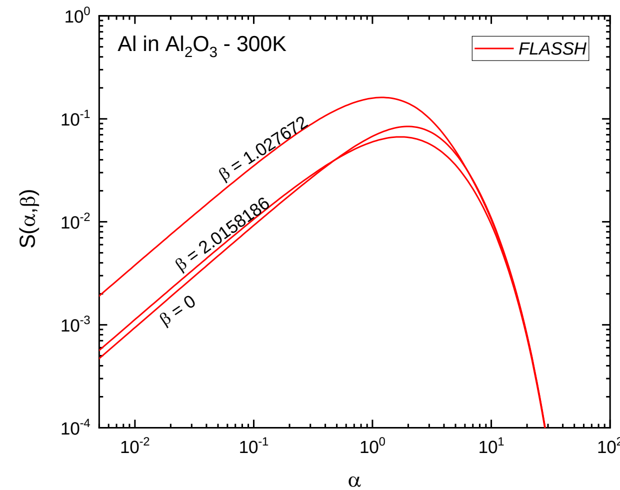
New to NNDC

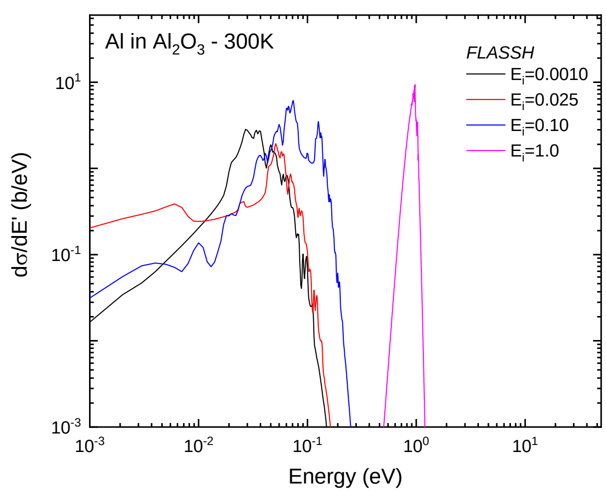
Single Crystal Sapphire TSL Data

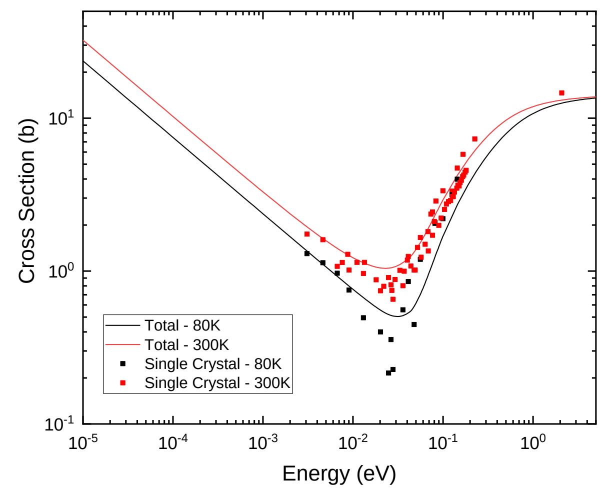
- *Ab initio* lattice dynamics
  - Predictive density of states (DOS)
  - Rhombohdral structure
  - 2x2x2 supercell
  - GGA-PBE











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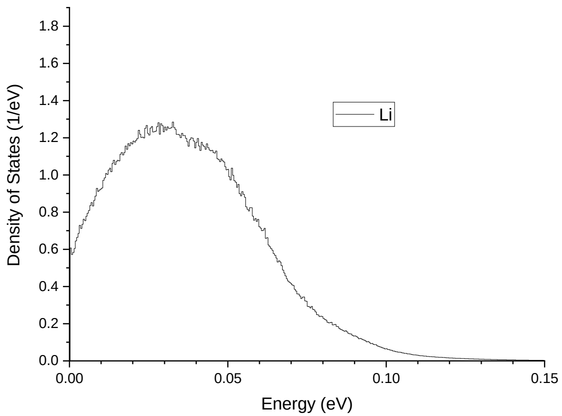
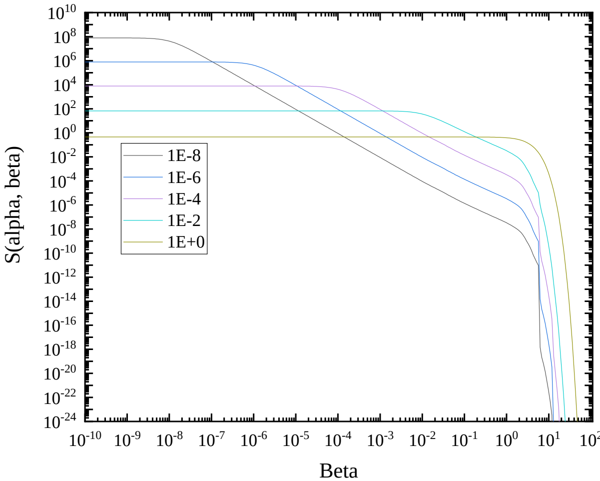
Completed FLiBe TSL Data

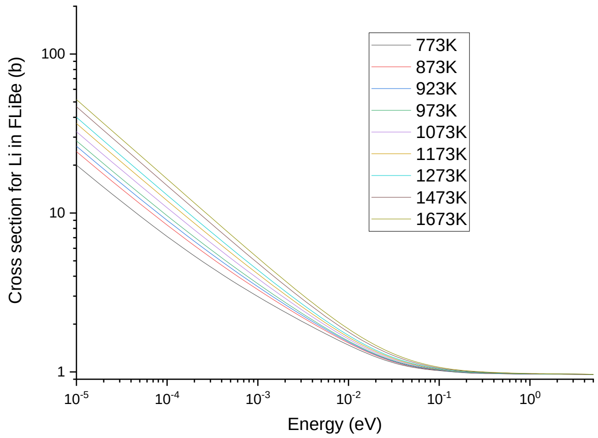
DOS using MD 773 K



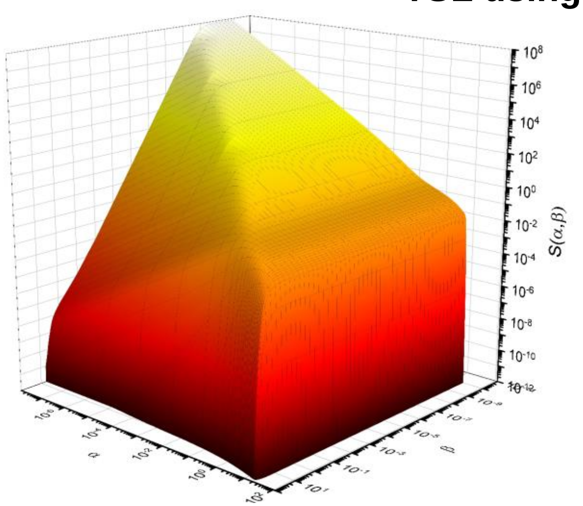
TSL using *FLASSH 773 K*



Cross section using *FLASSH*

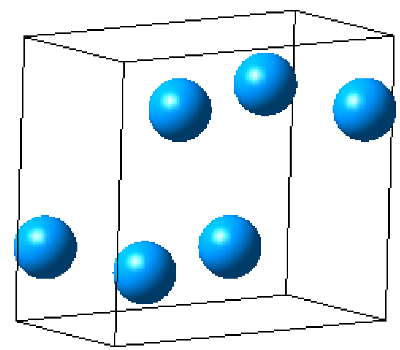


TSL using *FLASSH 773 K*

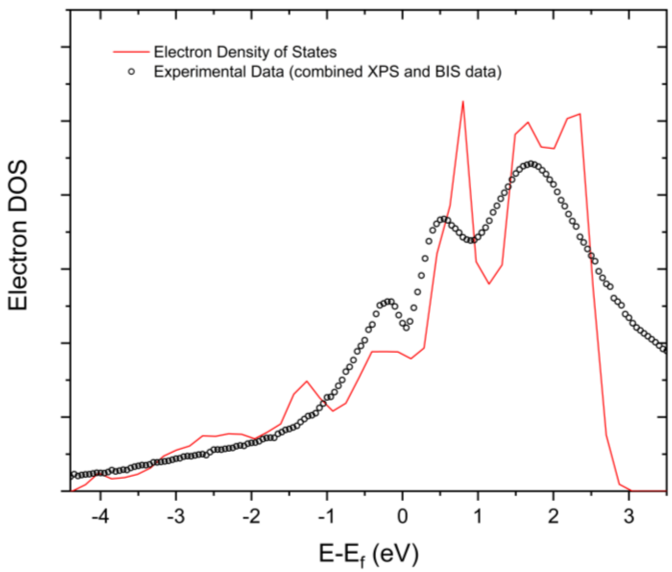


Uranium Metal

- α-Uranium Metal
  - Stable up to 668ºC
- *Ab initio* lattice dynamics
  - DFT – GGA-PBE plus an effective Coulomb term (+U) of 1eV for the *5f* electrons plus spin-orbit coupling



- Orthorhombic structure
- 550 eV plane wave cutoff
- 12x12x7 k-mesh



	Experiment (4.2 K)	Calculated	Diff. (%)
a (Å)	2.8444	2.8565	0.42
b (Å)	5.8689	5.8706	0.03
c (Å)	4.9316	4.9834	1.05

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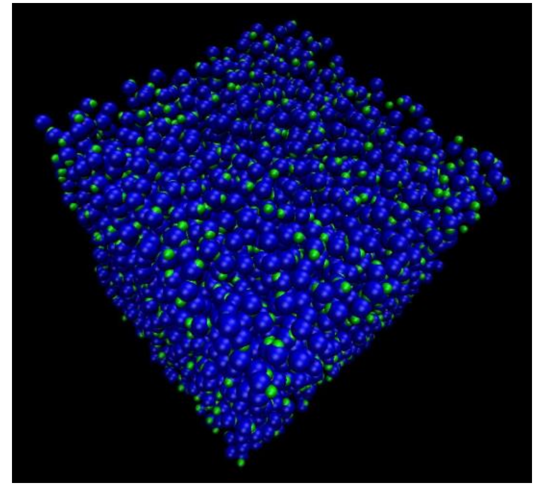
Hydrofluoric Acid MD

- Constructed potential function
- Molecular dynamics model
  - LAMMPS code
  - 4000 molecules



$$U^{total} = \sum_i \frac{1}{2} k_D \left| \boldsymbol{r}_{F,i} - \boldsymbol{d}_i \right|^2 + \frac{1}{2} k_{HF} (r_{HF} - d_{FH})^2 + \sum_{i<j} \sum_{s,\acute{s}} \frac{q_s q_{\acute{s}}}{\left| \boldsymbol{r}_{s,i} - \boldsymbol{r}_{\acute{s},i} \right|} + \sum_{i<j} 4\,\varepsilon_{FF} \left[\left(\frac{\sigma_{FF}}{\left| \boldsymbol{r}_{F,i} - \boldsymbol{r}_{F,j} \right|} \right)^{12} - \left(\frac{\sigma_{FF}}{\left| \boldsymbol{r}_{F,i} - \boldsymbol{r}_{F,j} \right|} \right)^{6} \right]$$

- Testing is underway using experimental data derived from "CRITICALITY OF LIQUID MIXTURES OF HIGHLY 235U-ENRICHED URANIUM HEXAFLUORIDE AND HYDROFLUORIDE ACID"

Y-CDC-9

UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE Y-12 PLANT
operated for the
U.S. ATOMIC ENERGY COMMISSION

CRITICALITY OF LIQUID MIXTURES OF HIGHLY ^{235}U-ENRICHED URANIUM HEXAFLUORIDE AND HYDROFLUORIC ACID

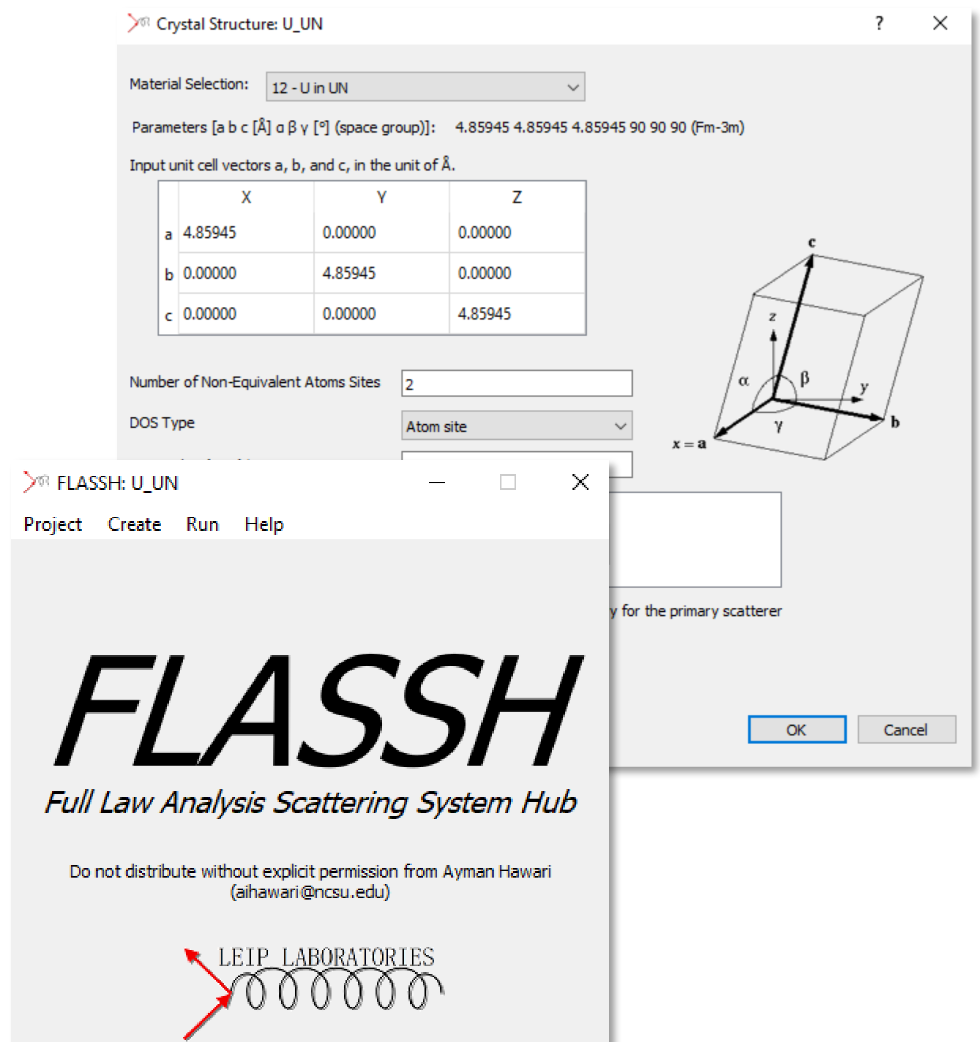
Robert Caizergues, Edouard Deilgat, Pierre Lécorché, Louis Maubert, and Henri Revol

Commissariat A L'Énergie Atomique
Direction de la Protection et de la Sûreté Radiologiques
Service D'Études de Criticité

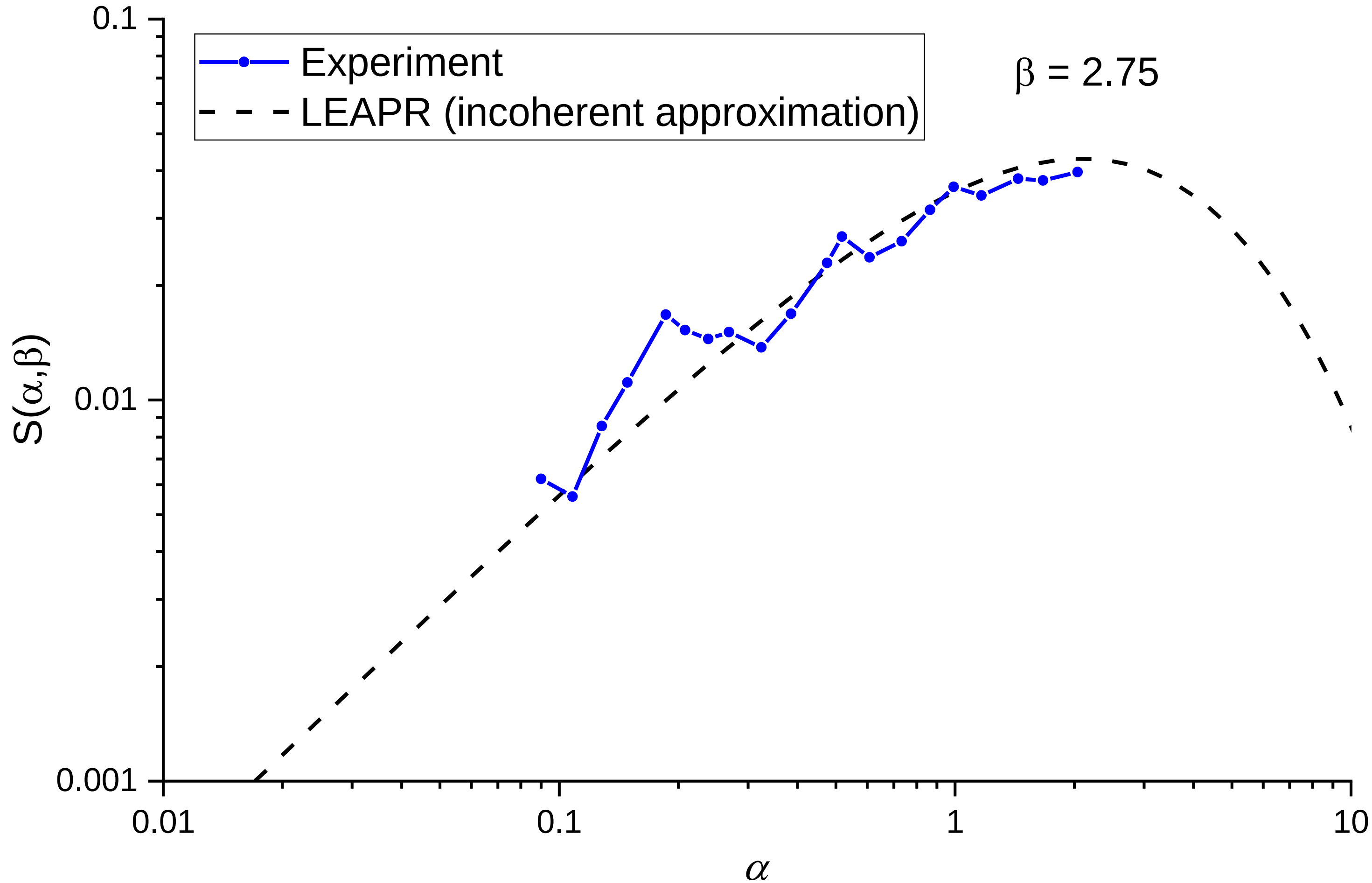
CRITICALITY DATA CENTER

FLASSH Code

- *FLASSH* Code
  - Relaxed major approximations (incoherent, cubic, ect.)
  - Improved liquid physics
  - Improved output formatting
  - Warning Messages for the User
- *FLASSH* GUI
  - Error Checks
  - Crystal Structure Window/ Inputs
  - ENDF Header Formatting

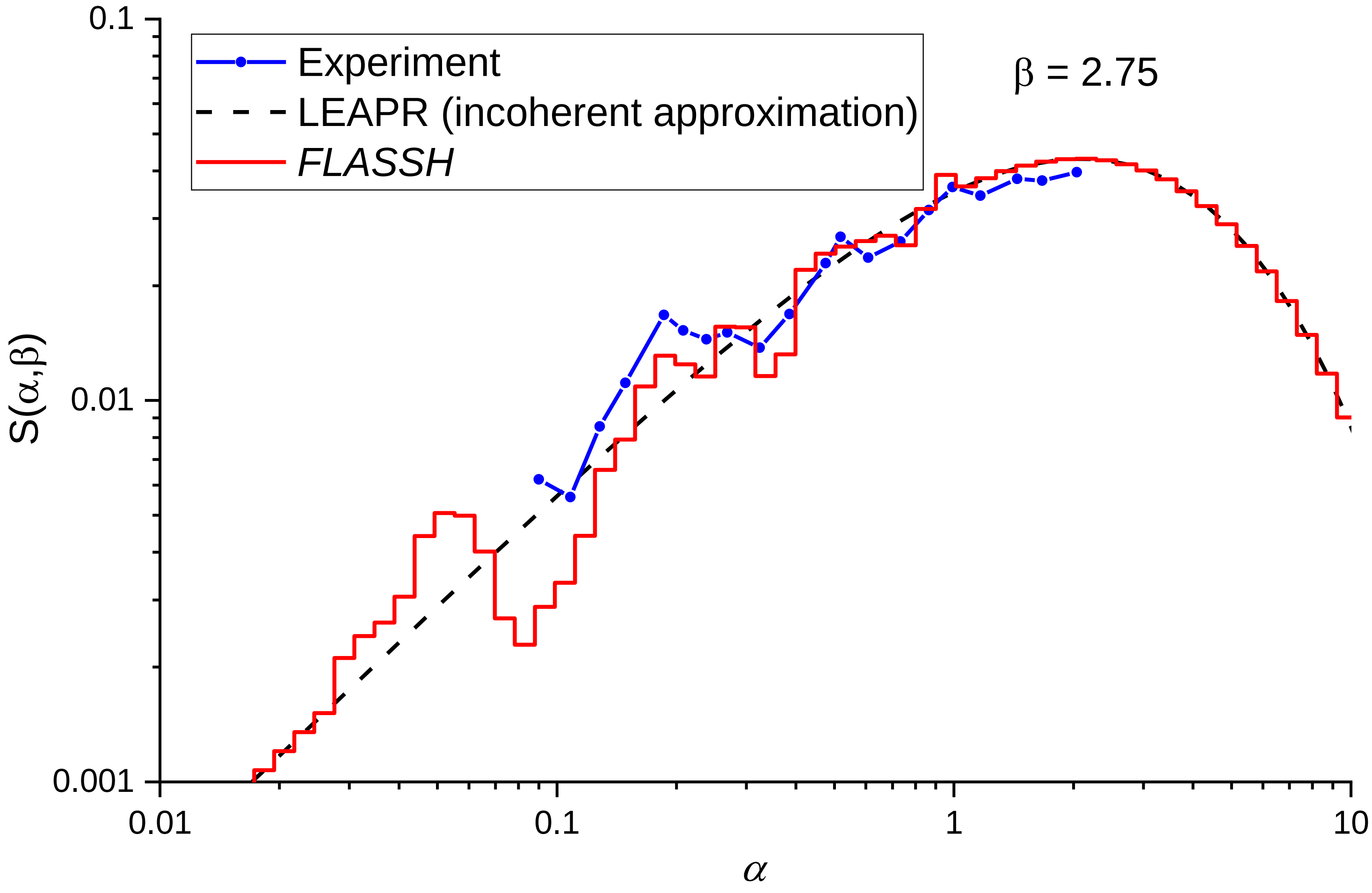


NJOY
Be Scattering Law $S(\alpha, \beta)$



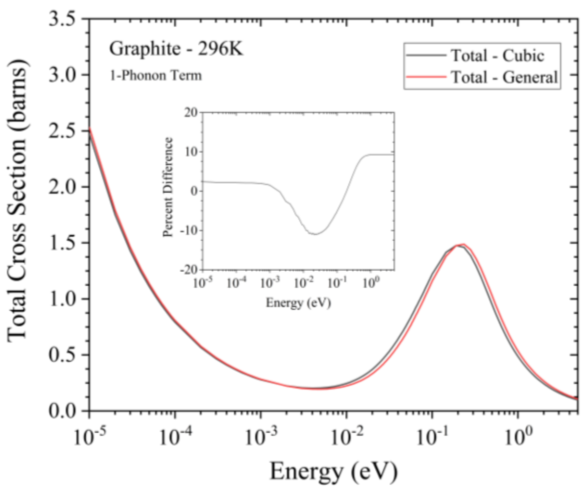
FLASSH

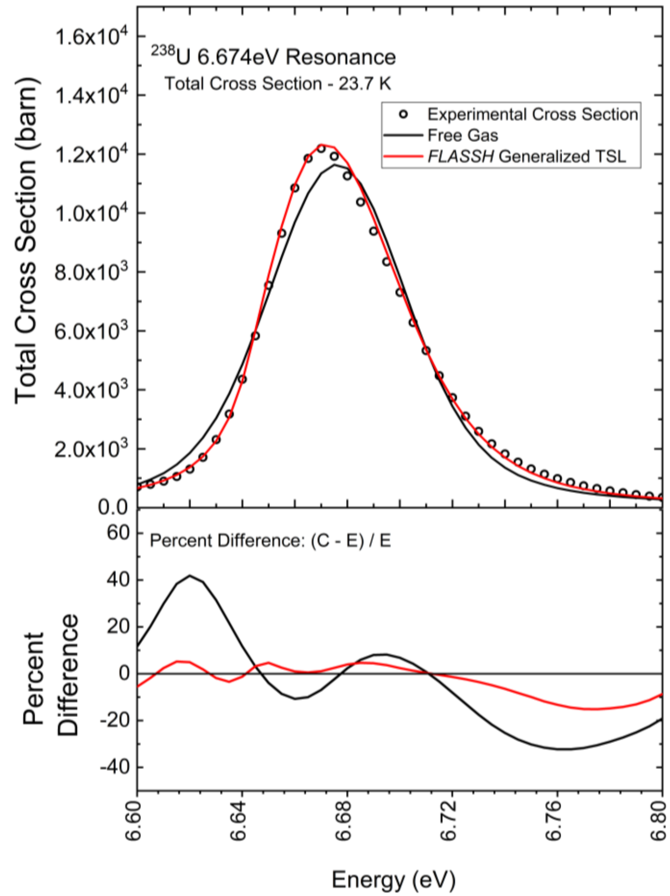
Be Scattering Law $S(\alpha, \beta)$



FLASSH – Generalized TSL & Doppler Treatment

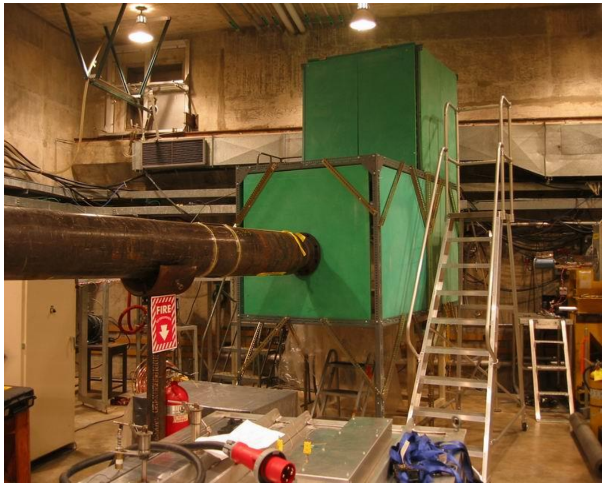
- **Generalized TSL**
  - Exact Structure
  - Polarization vectors and frequencies directly input
  - Removes cubic and atom site approximation
- **Doppler Broadening**
  - *FLASSH* TSL to determine velocity distribution for broadening
  - Implements exact material structure
  - Consistent evaluation for both thermal scattering and Doppler broadening

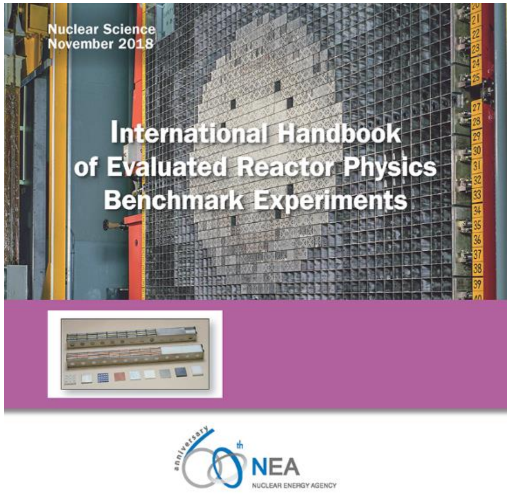


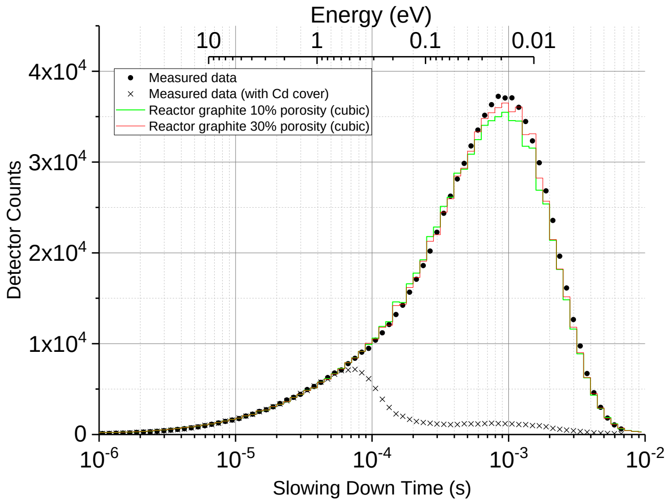


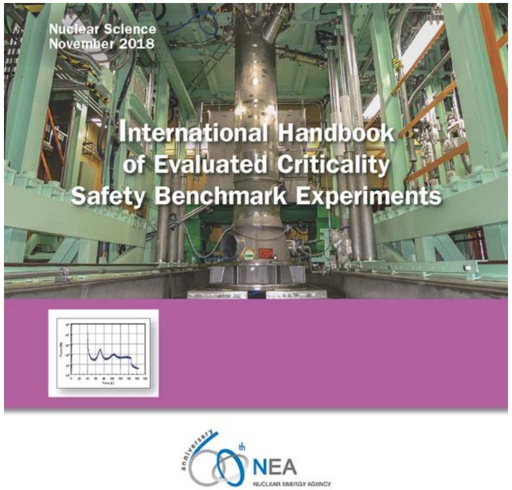
$$S_s^m(k,\omega) = \int e^{-i\omega t} I_s^m(k,t)dt$$
$$= \int e^{-i\omega t} e^{\langle U^2\rangle_m + \langle UV_0\rangle_m} dt$$

TSL Specific Benchmarks





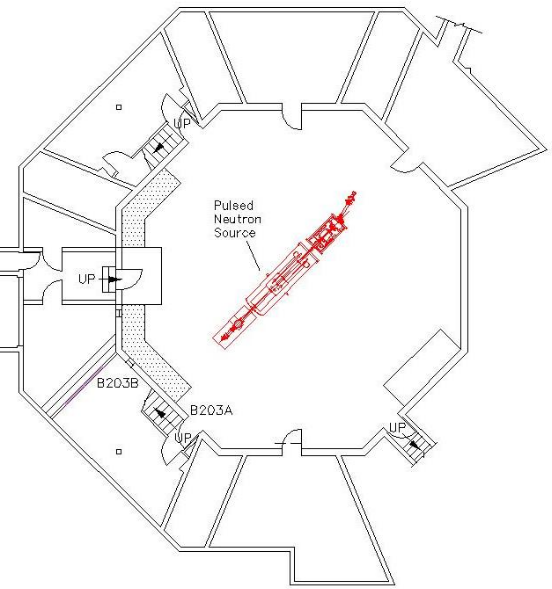


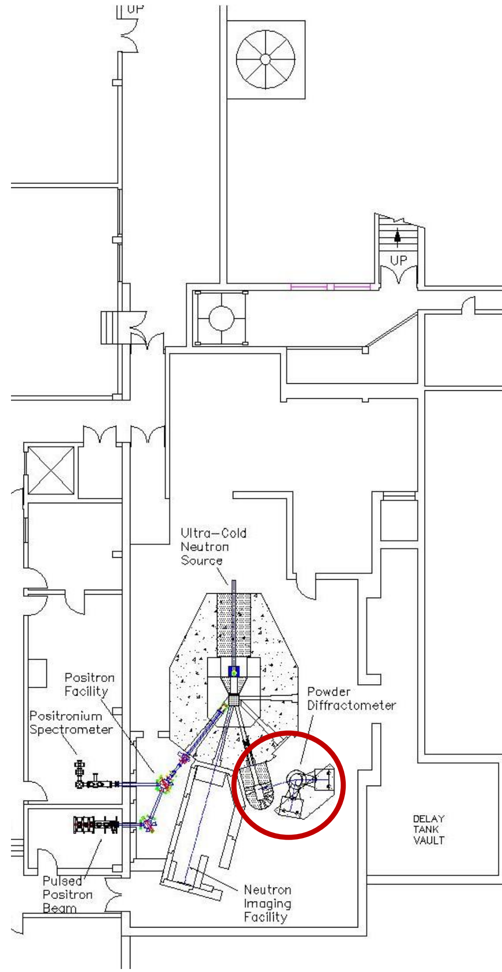


PULSTAR reactor

- **Current major facilities/capabilities**
  - **Neutron powder diffraction**
  - Neutron imaging
  - Intense positron beam
  - Ultracold neutron source (under testing)
  - Neutron activation analysis
  - In-pool irradiation testing facilities

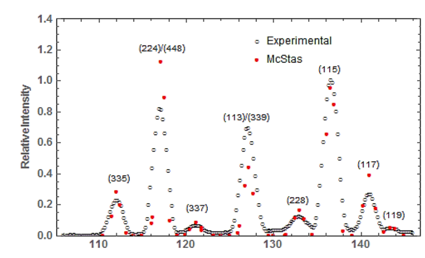
- **Current projects**
  - PULSTAR power upgrade 1-2 MW (licensing stage)
  - Various instrument and facility upgrades
  - Pulsed accelerator neutron source (under testing)
  - Fuel loop for fission gas release studies





Old reactor bay

reactor bay



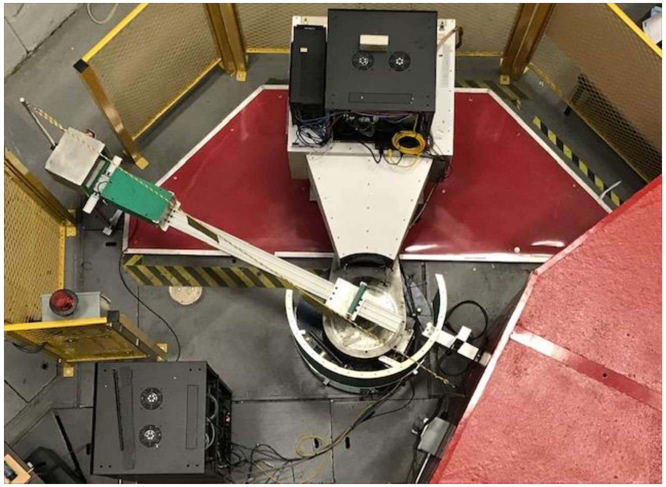
Cross Section Measurement Project

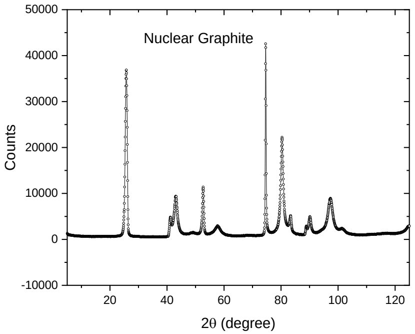
NPDF Facility Upgrades – Dual Purpose:

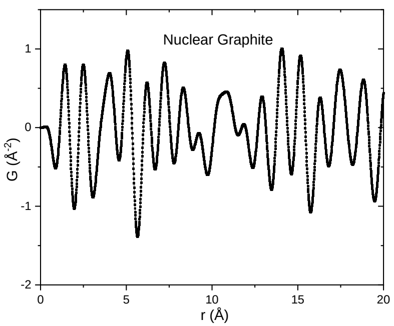
- Diffraction Measurements:

  15 New Position Encoding Modules (PEM) – improved diffraction measurement resolution $\Delta d/d$ of 2.9×10^{-3} for 3mm holder

- **Transmission Measurement Capabilities:**
  - Monochromator capable of providing beam wavelengths of 1.085 Å, 1.180 Å, 1.479 Å, and 1.762 Å
  - Transmission Detection Apparatus with collimator.

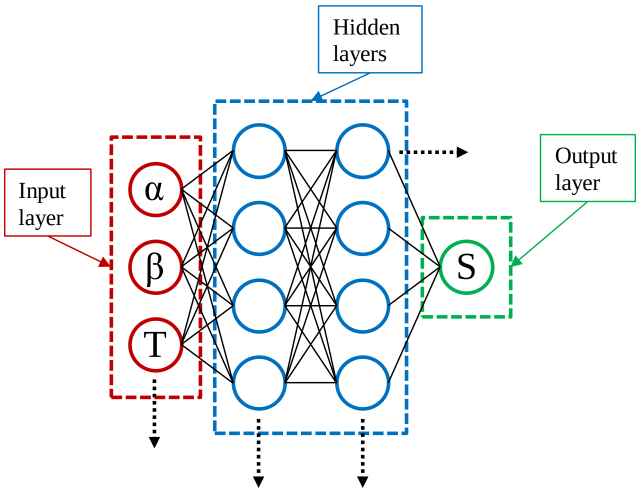


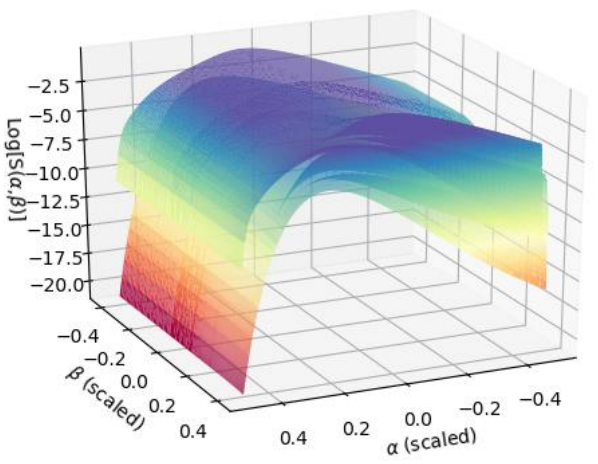




TSL NeTS

- New TSL paradigm
  - DL Neural Thermal Scattering (NeTS) modules
- See papers and presentations
  - ANS 2019 Winter Meeting, Washington, DC, USA
  - PHYSOR 2020 Meeting, Cambridge, UK





Summary

- TSL Activities continue including evaluations and methods development
- Evaluations are contributed to NNDC
  - Paraffinic oil and single crystal sapphire
  - Molten salt FLiBe will be submitted soon
  - Hydrofluoric acid is initiated
  - Uranium metal is initiated
- *FLASSH* testing and several evaluations are underway
- Activities in data measurements and benchmark development are underway
- DL NeTS approach is under testing