Accomplishments of Thermal Neutron Scattering Research at North Carolina State University

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NUCLEAR CRITICALITY SAFETY PROGRAM (NCSP)TECHNICAL PROGRAM REVIEW
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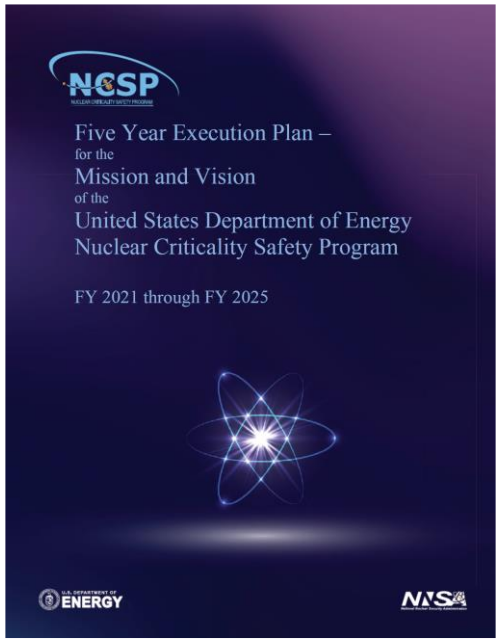
Acknowledgment

- NNSA Nuclear Criticality Safety Program (NCSP)
  - collaboration with LLNL
- Naval Nuclear Propulsion Program (NNPP)
- The LEIP Team



FY 2020 Tasks

Task	Task Title	Budget
ND2	Generation and Benchmarking of Thermal Neutron Scattering Cross Sections in Support of Advanced Nuclear Reactor Concepts	$72 K
ND3	Development and Implementation of an Advanced and Rigorous Computational Platform for Thermal Neutron Scattering Analysis	$100 K
ND5	Development and Implementation of a Modern Doppler Broadening Approach Including Atomic Binding Effects	$95 K

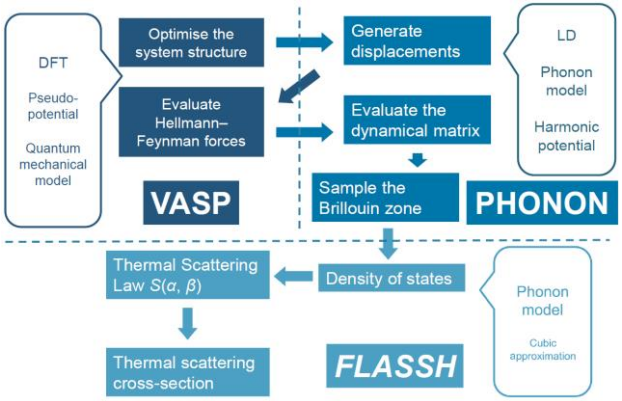
APPENDIX B: Nuclear Data Priorities, Basis Statements, and Milestones

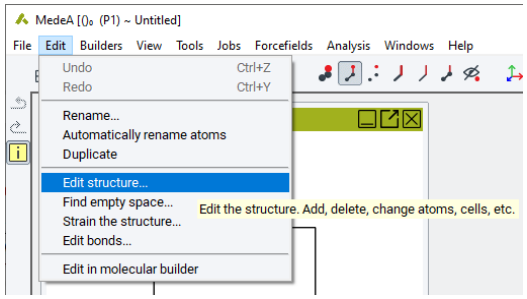
Nuclear Data Evaluations							
Materials	**Pre-FY2021**	**FY2021**	**FY2022**	**FY2023**	**FY2024**	**FY2025**	**Post-FY2025**
Zirconium ($^{90,91,92,94,96}Zr$)					ORNL	ORNL	ORNL
Basis	Resonance evaluations. Zirconium is a key structural element that is primarily used in cladding for fuel rods and is currently in consideration for use with advanced nuclear fuel matrices in the form of zirconium hydride. The latest ENDF/B-VII.1 resonance evaluation relies on JENDL-4 data and resonance parameters from the Atlas of Neutron Resonances. As a result, the evaluated resonance parameters are not based on detailed R-matrix analyses. In addition, newer RPI total cross-section measurements on natural zirconium indicate that the older ENDF/B-VI.8 data match the recent RPI measurements better than the newer isotopic evaluations. Furthermore, improved differential measurements of the zirconium isotopes have been identified on the OECD/NEA nuclear data High Priority Request List (HPRL). Differential measurements are needed in the resonance region to accurately predict the neutron resonances for the zirconium isotopes, and corresponding resonance evaluations are needed to provide detailed resonance parameters and covariance data. In addition, the SAMMY evaluation software has the capability to generate angular scattering distributions from the resonance parameters thereby providing detailed resonance scattering structure that will improve the elastic scattering modeling for the zirconium isotope evaluations. NR continues to be unsatisfied with Zr evaluations in ENDF.						
Water (H_2O)	LLNL/NCSU	LLNL/NCSU					
Basis	TSL evaluation. Water is this most important moderator and moderating reflector material for criticality safety and light water reactor physics. Problems with evaluations submitted by CAB at elevated temperatures (that were noticed during the ENDF/B-VIII.0 evaluation process) warrant re-evaluating this essential material using the latest methods developed under LLNL ND2, ND3.						
Hydrofluoric Acid (HF)	LLNL/NCSU	LLNL/NCSU					
Basis	TSL evaluation. HEU-SOL-THERM-039, "Mixture of Uranium (93%) Hexafluoride and Hydrofluoric Acid (Low H/U Ratio) in a Hot-Water-Reflected Spherical Tank," critical experiments overpredict k-eff from 2-6% regardless of cross-section library or code utilized. An appropriate thermal scattering law for the liquid Hydrofluoric acid (HF) moderator will likely resolve this calculational discrepancy.						
Uranium Hexafluoride (UF_6)	LLNL/NCSU	LLNL/NCSU					
Basis	TSL evaluation. As the H/U ratio is "low" in HEU-SOL-THERM-039, correcting for F in UF_6 may be necessary as a moderator. A thermal scattering law for this fissile compound will be useful for the advanced Doppler broadening methods currently under development as LLNL ND5.						
Uranium Metal (U)	LLNL/NCSU	LLNL/NCSU	LLNL/NCSU				
Basis	TSL evaluation. Requested by the RPI for use in U-235 resonance parameter analysis.						
Uranium Carbide (UC)		LLNL/NCSU	LLNL/NCSU	LLNL/NCSU			
Basis	TSL evaluation. A common fissile compound under consideration for high-temperature advanced nuclear reactor fuel. A thermal scattering law for UC will improve Doppler broadening using advanced methods currently under development as LLNL ND5.						

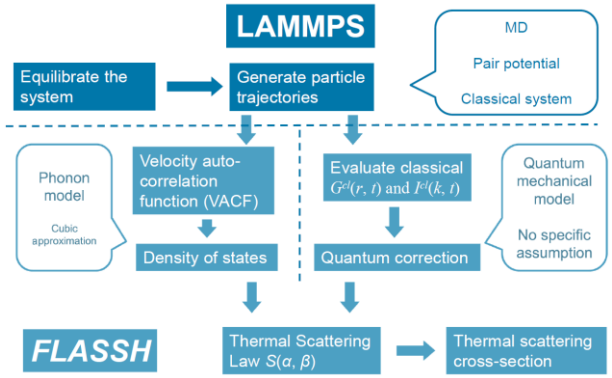
Contributed TSL Evaluations

Material	Motivation	NCSP 5 Year Plan	
		Expected	Actual
Heavy oil (hydrogen)	NCSP/NR applications No TSL data in ENDF/B-VIII.0	FY 2020	FY 2019
Liquid hydrogen fluoride (hydrogen)	NCSP applications No TSL data in ENDF/B-VIII.0	FY 2021	FY 2020
Light water (hydrogen)	NCSP applications Extended ENDF/B-VIII.0	FY 2021	FY 2020
Polyethylene (hydrogen)	NCSP applications Extended ENDF/B-VIII.0	----	----
Uranium metal	NCSP applications No TSL data in ENDF/B-VIII.0	FY 2022	Near completion
Uranium carbide	NCSP applications No TSL data in ENDF/B-VIII.0	FY 2023	ongoing
Sapphire (Al in Al2O3)	Neutron science / Research Reactors No TSL data in ENDF/B-VIII.0 (cryogenic temperatures)	----	----
Sapphire (O in Al2O3)			
20% Porous Reactor Graphite	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	----	----
FLiBe (beyllium)	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	----	----
FLiBe (flourine)			
FLiBe (lithium)			

TSL Methodology

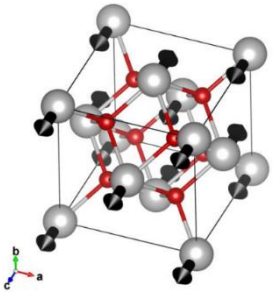


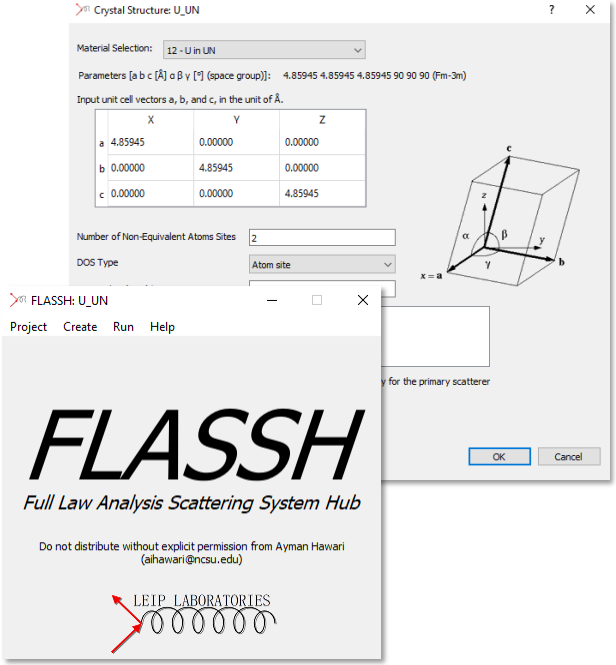


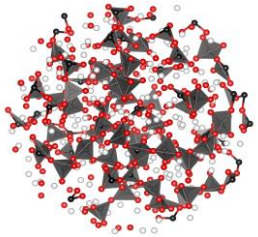


DFT/LD approach

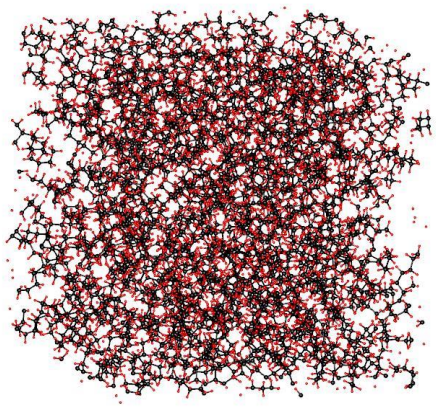
MD approach

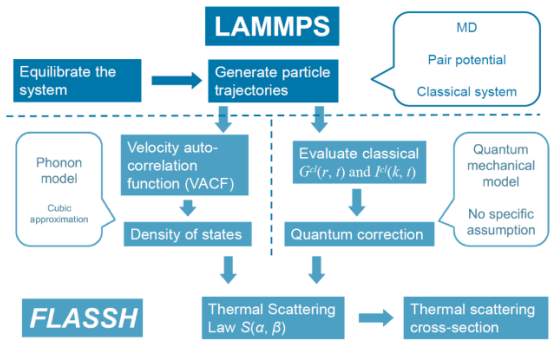


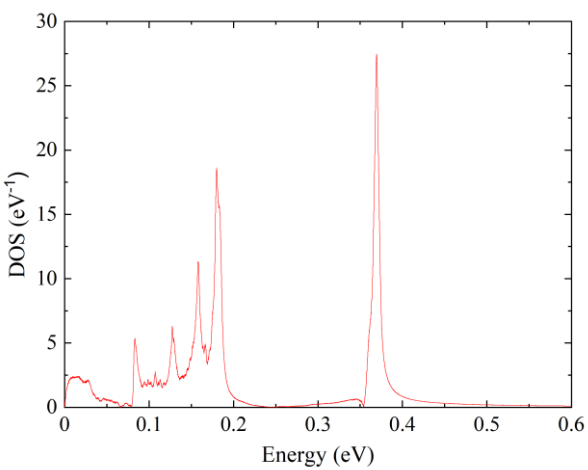


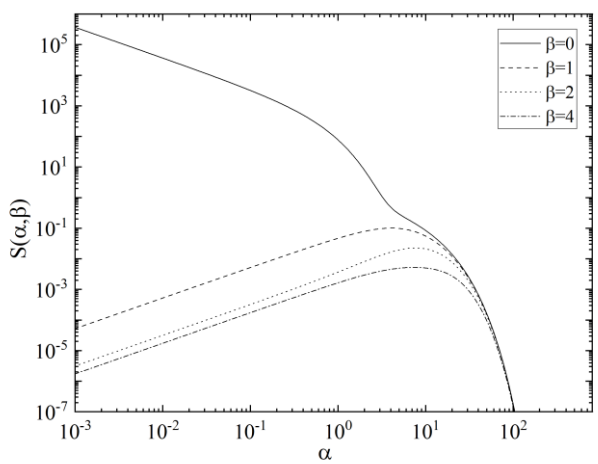


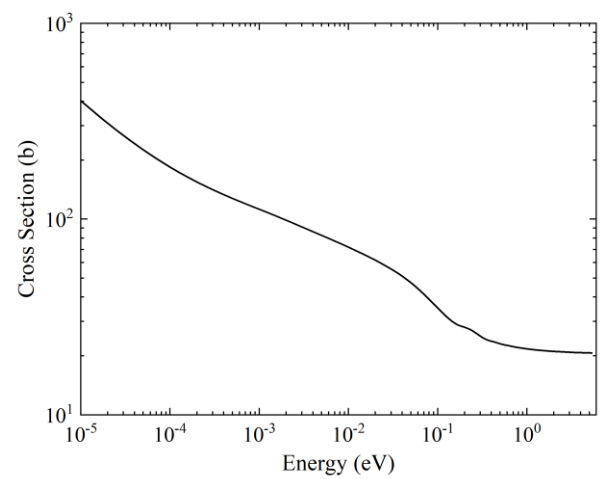
ND2 – New to NNDC
Heavy Paraffinic Oil Data – NCSP/NR



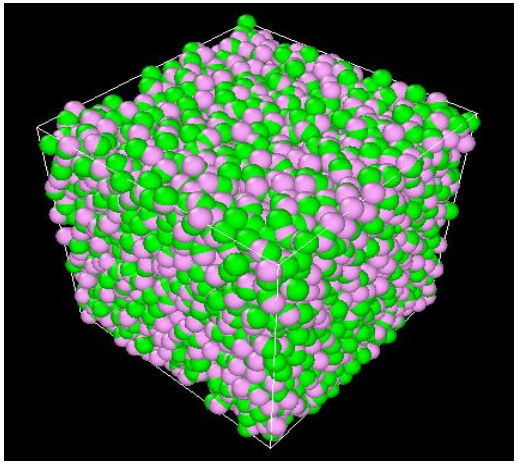


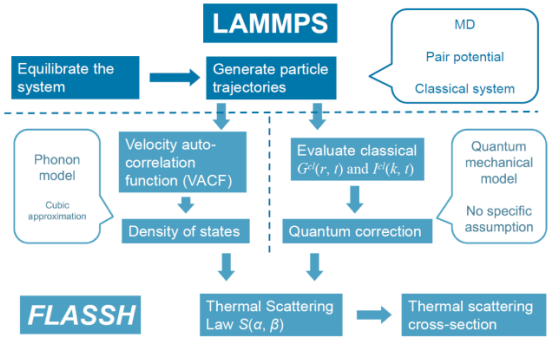


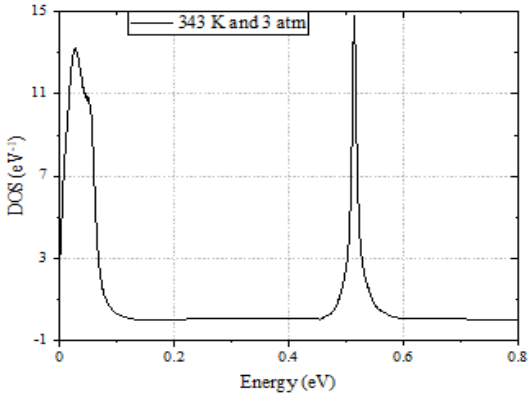


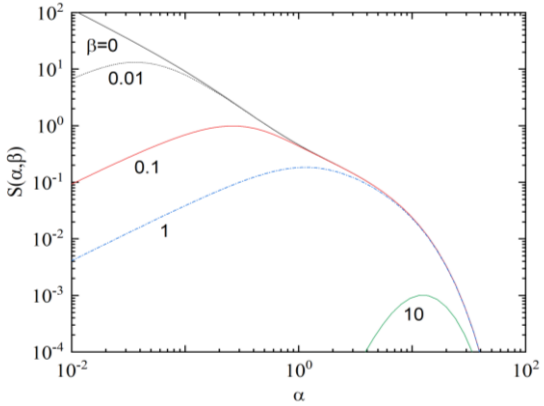


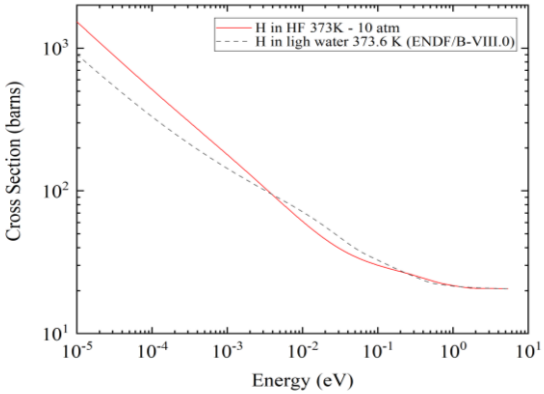
ND2 – New to NNDC Hydrofluoric Acid Data - NCSP



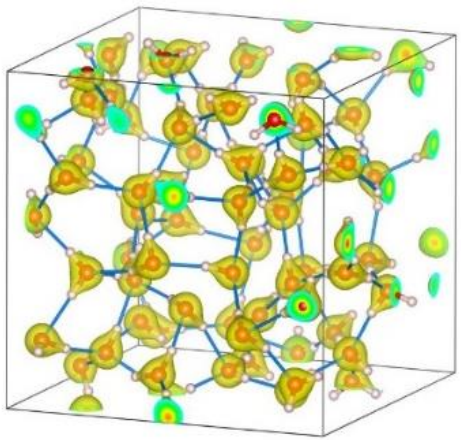


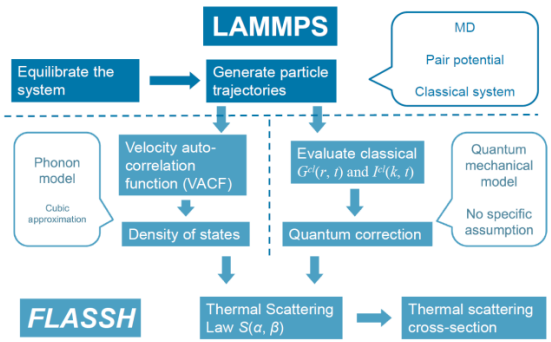


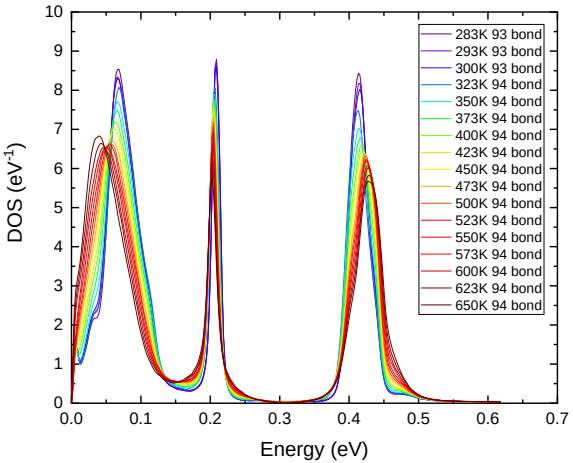


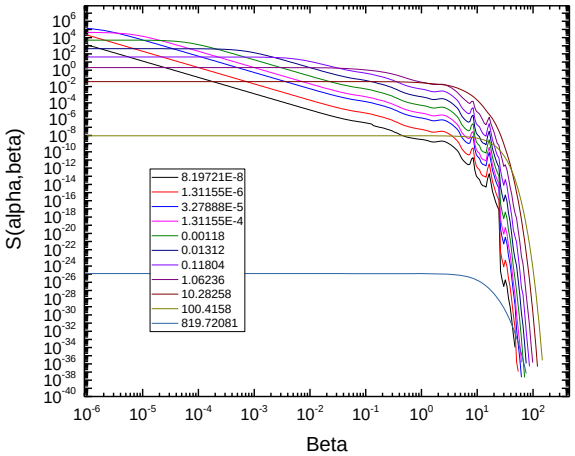


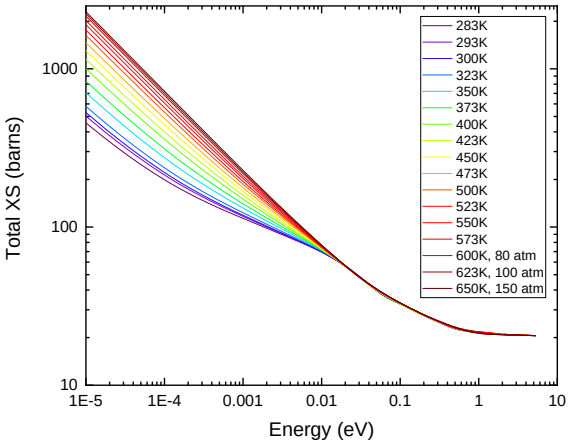
ND2 – Testing at NR Light Water - NCSP



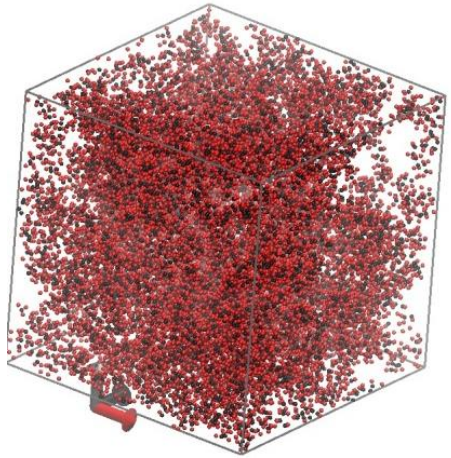


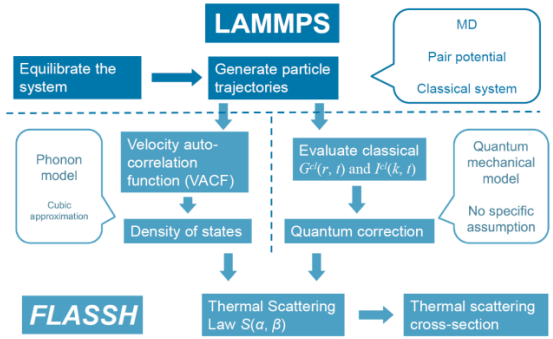


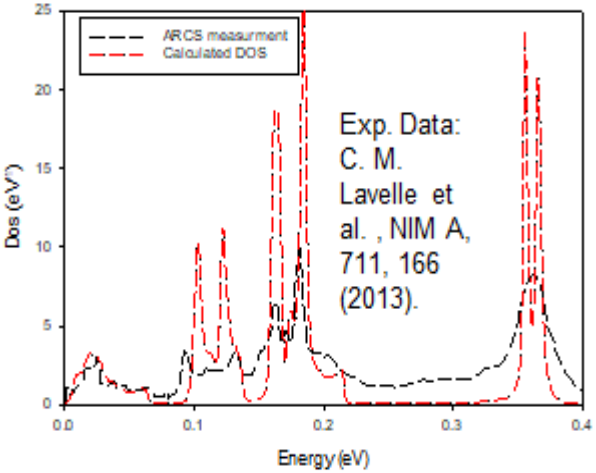


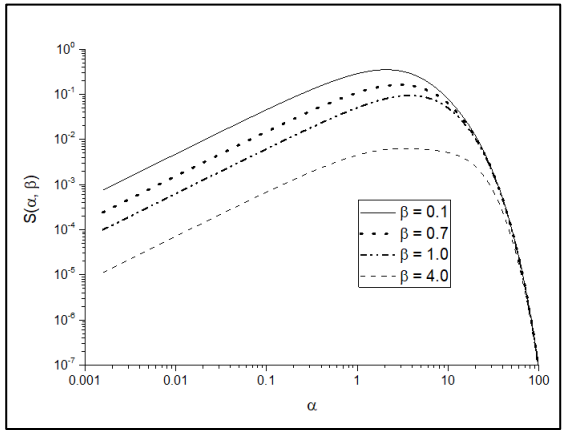


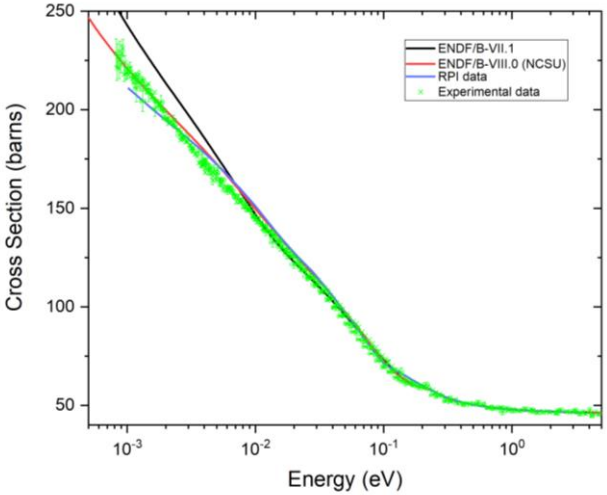
ND2 – Update to NNDC Polyethylene Data - NCSP





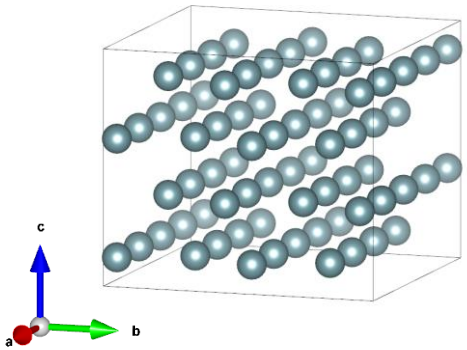


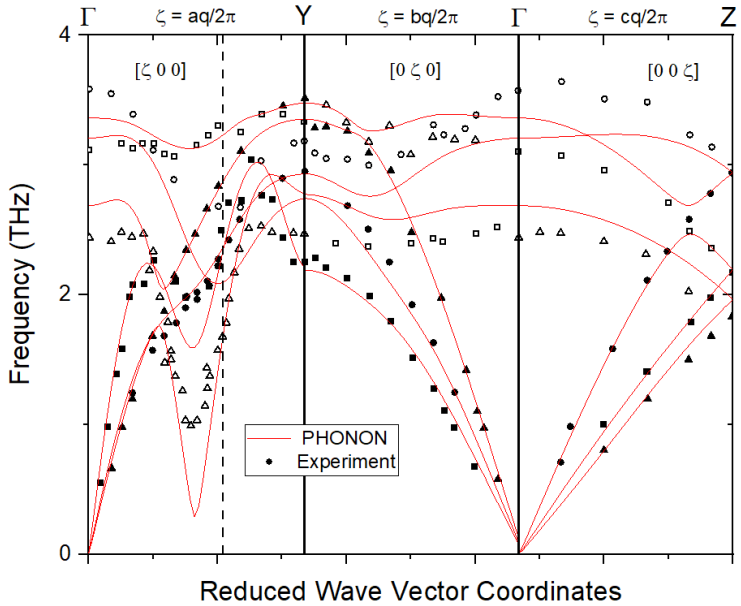




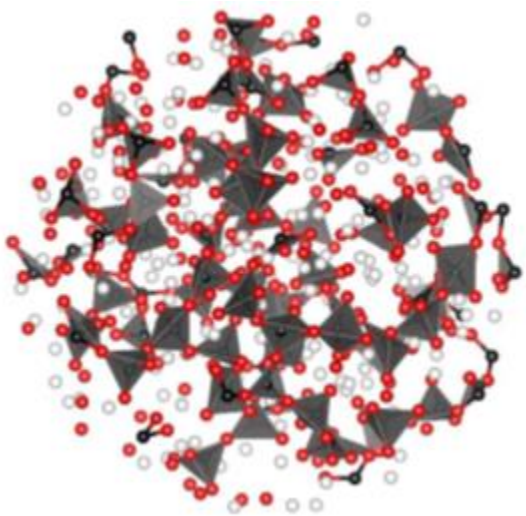
ND2
Uranium Metal – NCSP

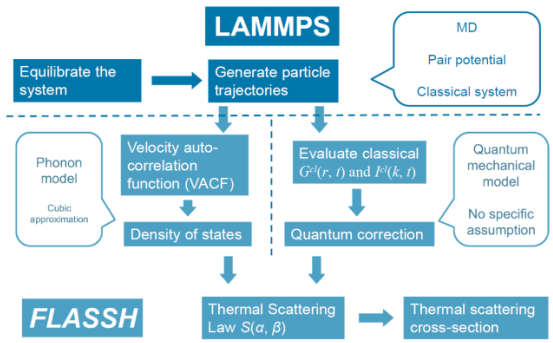
- **DFT Model**
  - GGA-PBE, Spin Polarized Calculation
  - Hubbard Term, +U = 1.5 eV
  - No SOC (insignificant impact)
  - 4x2x2 to 4x4x4 Supercells (64-256 atoms)
- **Cross Sections**
  - Uranium total absorption 7.57 b at 2.53 eV

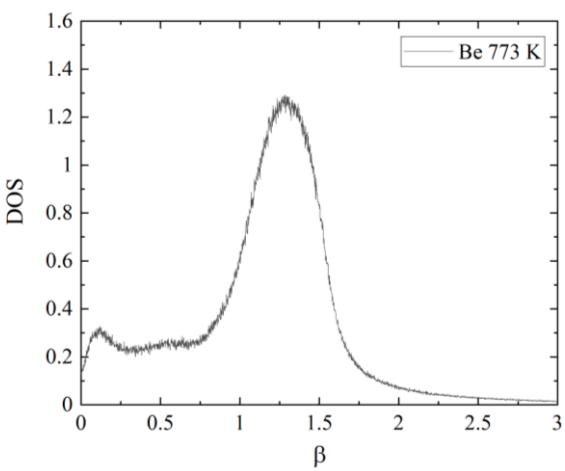


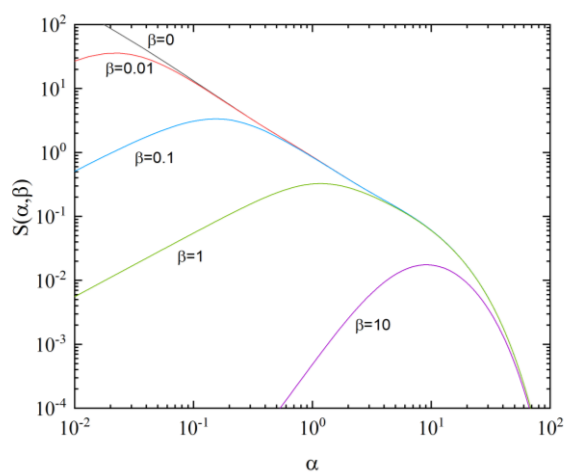


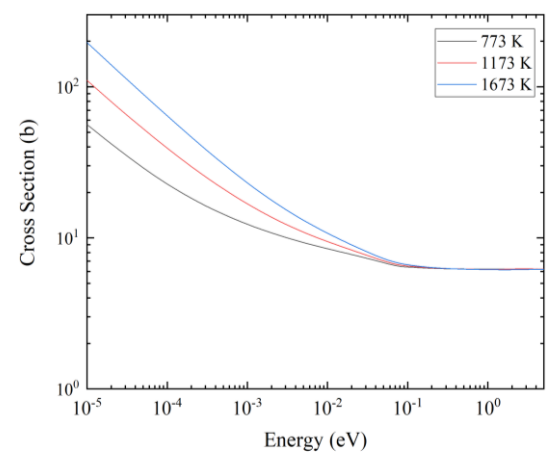
New to NNDC
Molten Salt FLiBe Data – DOE NE



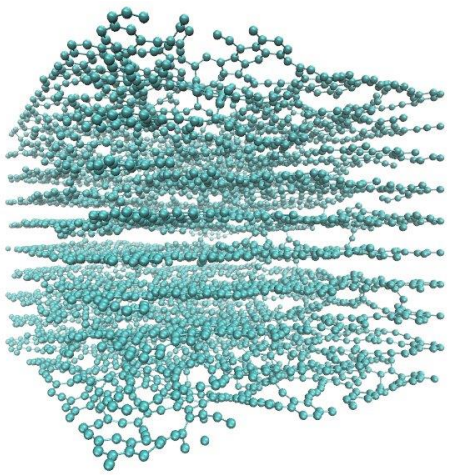


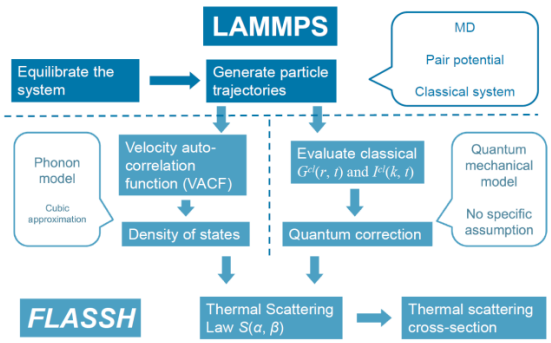


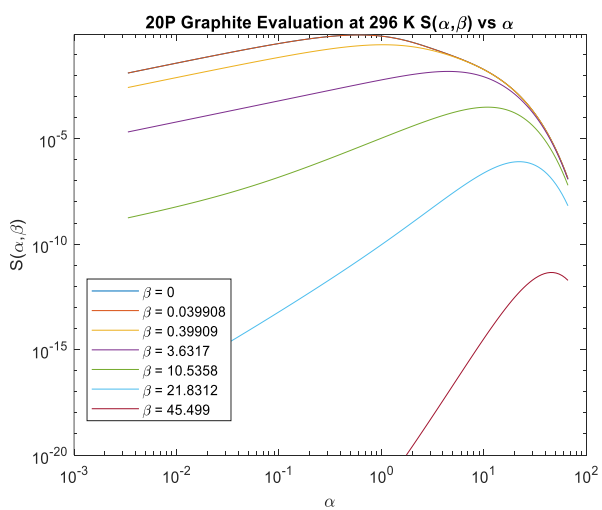


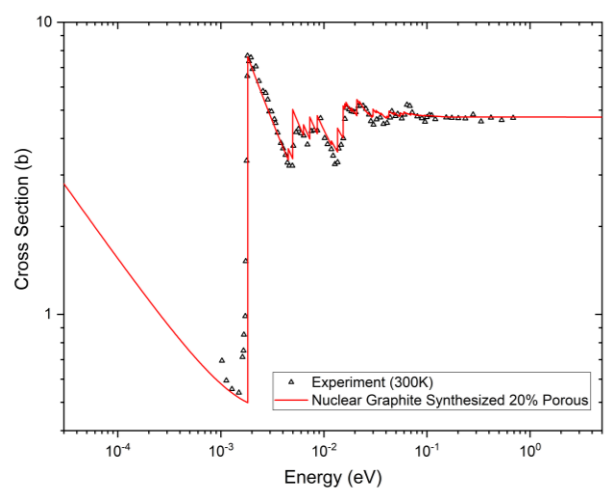


20% Porous Nuclear Graphite – DOE NE

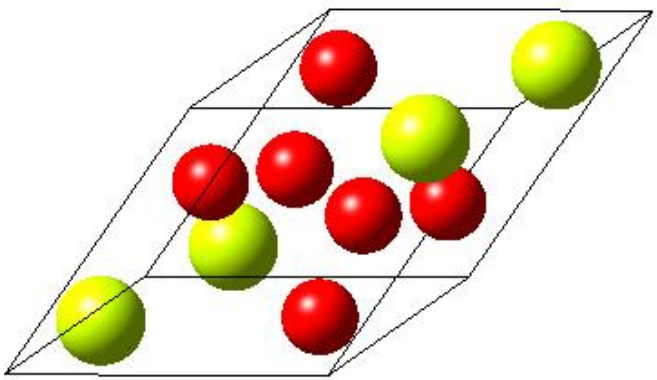


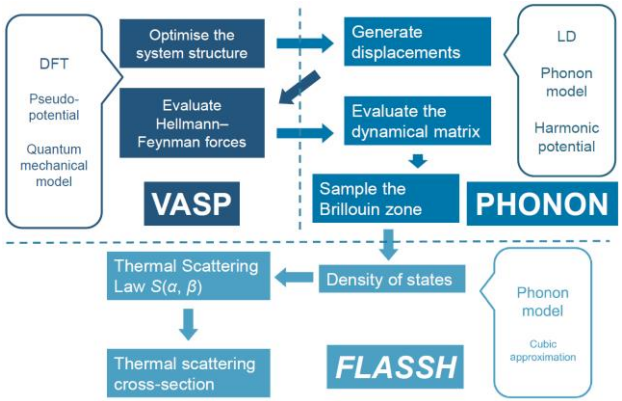


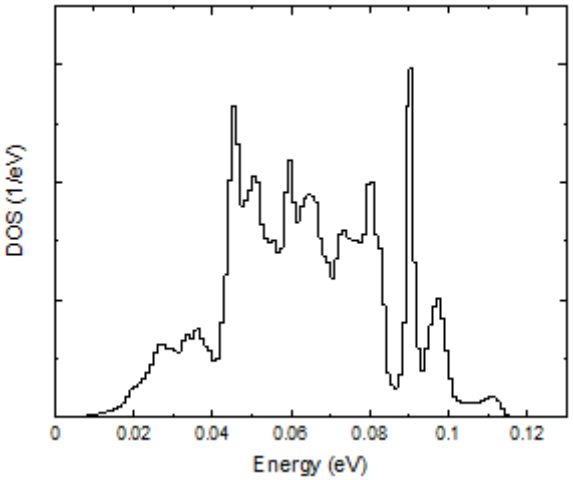


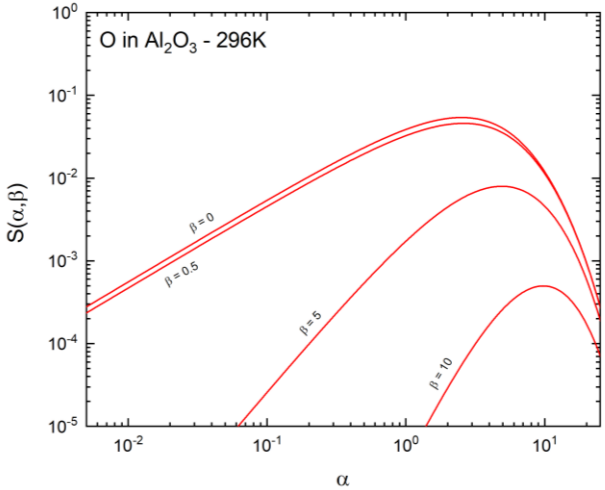


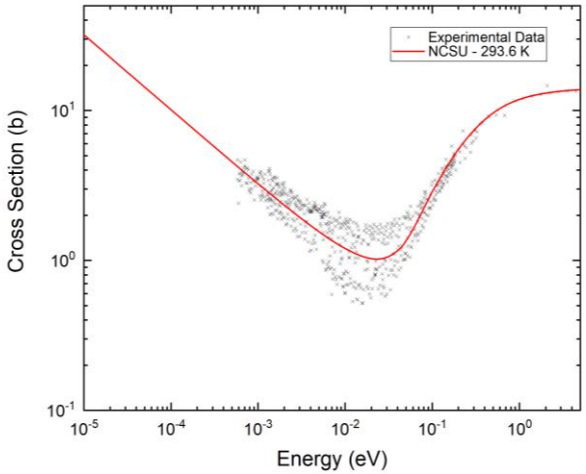
New to NNDC
Single Crystal Sapphire Data - RR



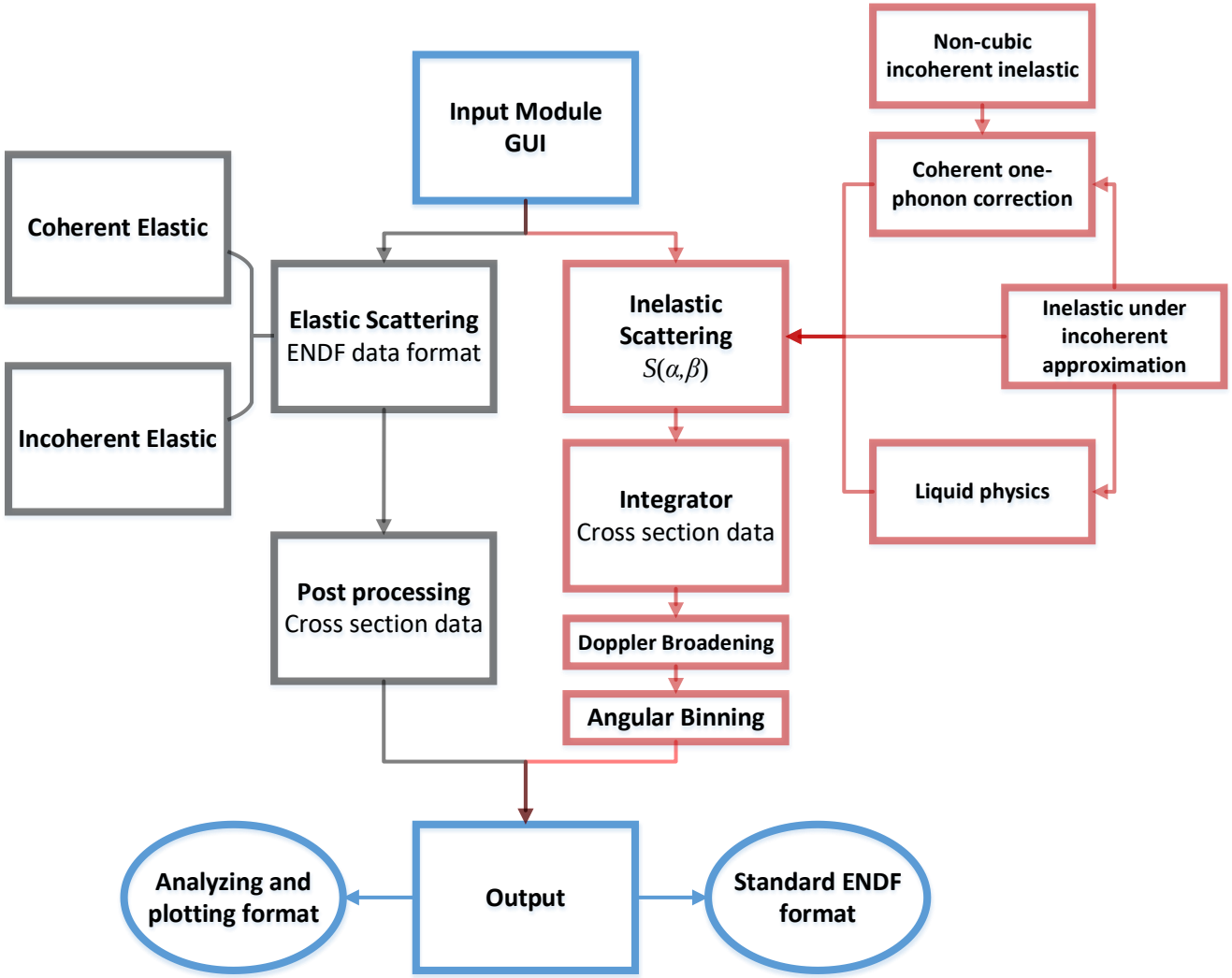






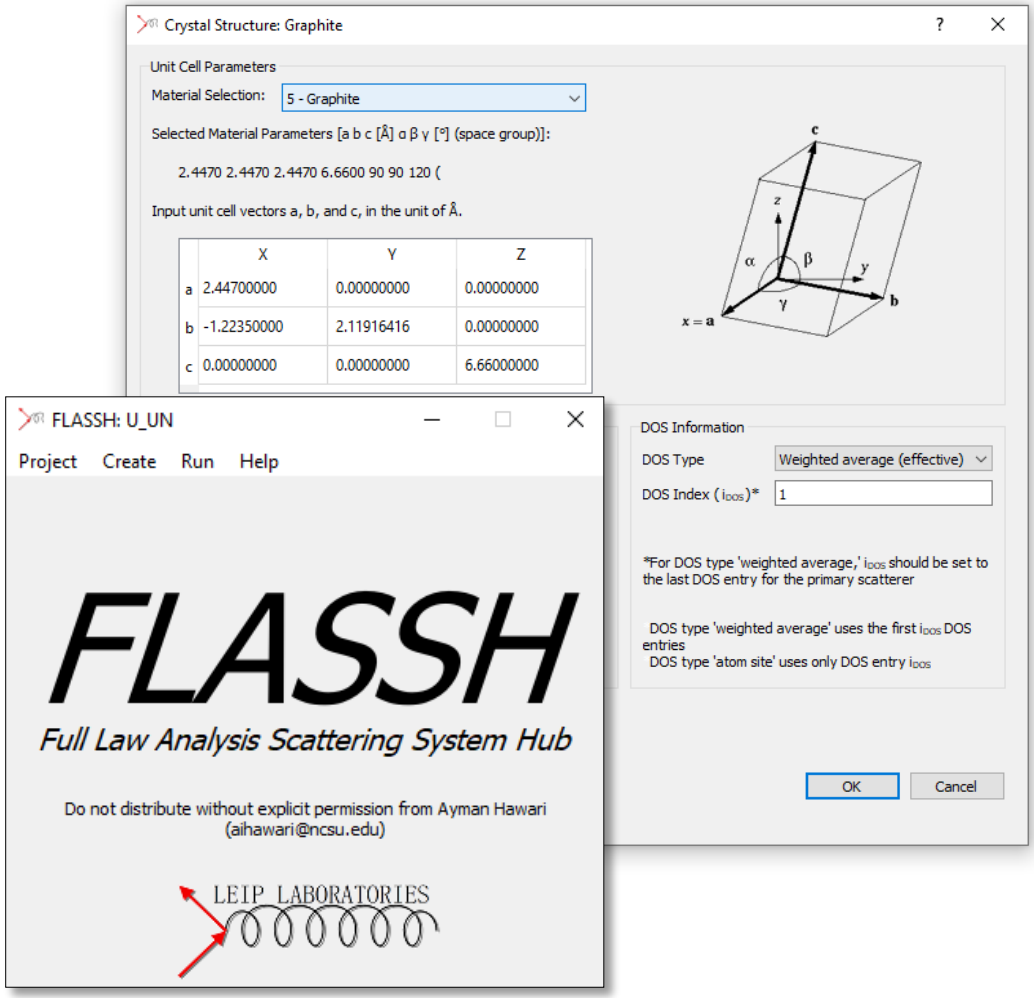


ND3 – *FLASSH*


Input Module
GUI
Coherent Elastic
Incoherent Elastic
Elastic Scattering
ENDF data format
Post processing
Cross section data
Inelastic
Scattering
$S(\alpha,\beta)$
Integrator
Cross section data
Doppler Broadening
Angular Binning
Non-cubic
incoherent inelastic
Coherent one-
phonon correction
Inelastic under
incoherent
approximation
Liquid physics
Analyzing and
plotting format
Output
Standard ENDF
format

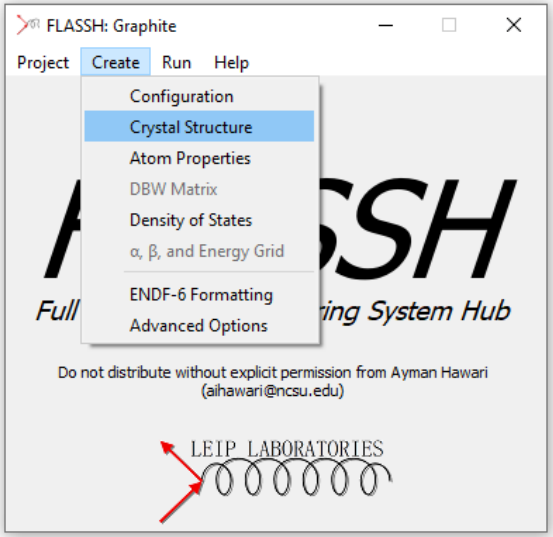
FLASSH

- ***FLASSH***
  - Calculations and ENDF TSL library formatting modules implemented in FORTRAN 95 using a modular design
  - Parallel computing realized by OpenMP 4.0 bindings
  - GUI implemented by cross platform QT® C++ API
  - Error checks
  - Input Generator (for both *FLASSH* and NJOY)
  - ENDF / ACE Formatting
  - Warning Messages Based on Material Physics
  - Crystal Structure Dependent Calculation

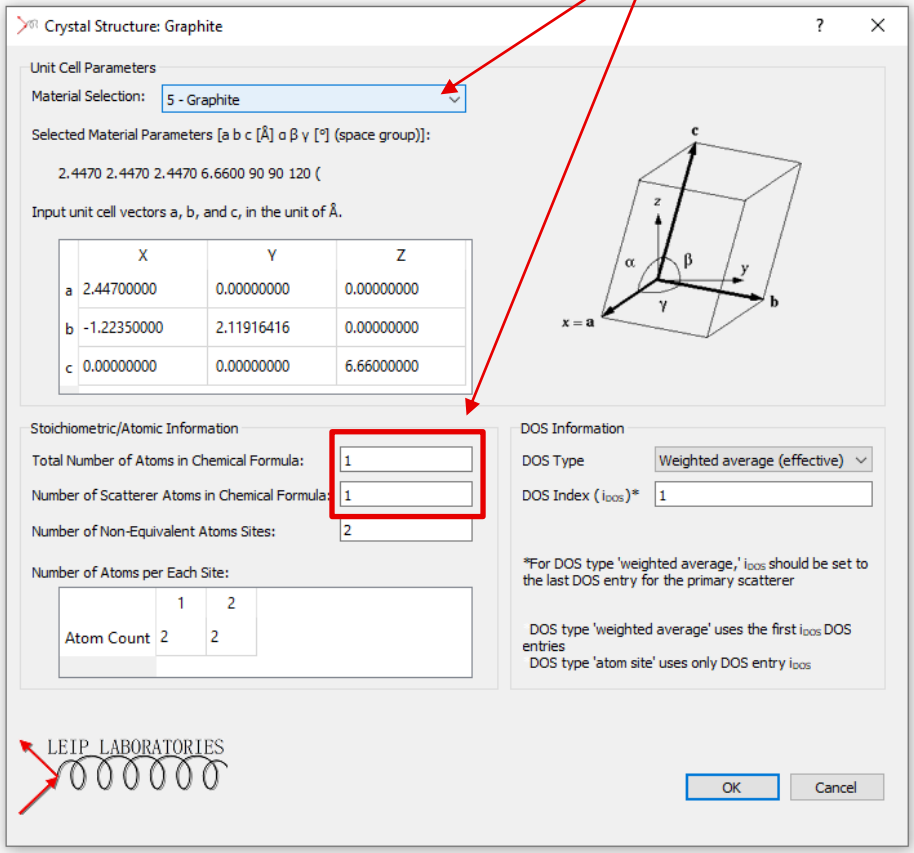


FLASSH

- ***FLASSH* GUI**
  - Crystal Structure (Defaults Shown)

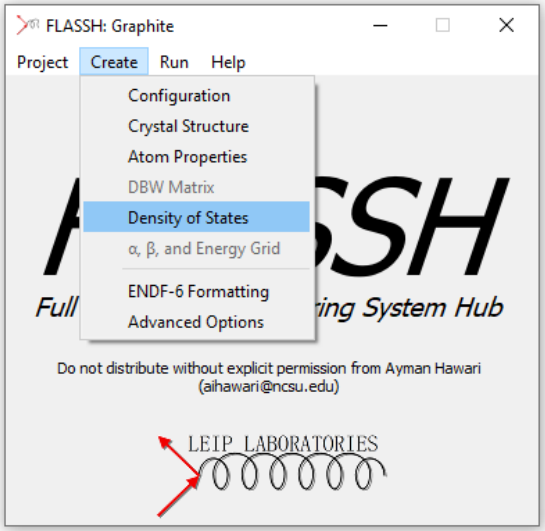


Required
User Input

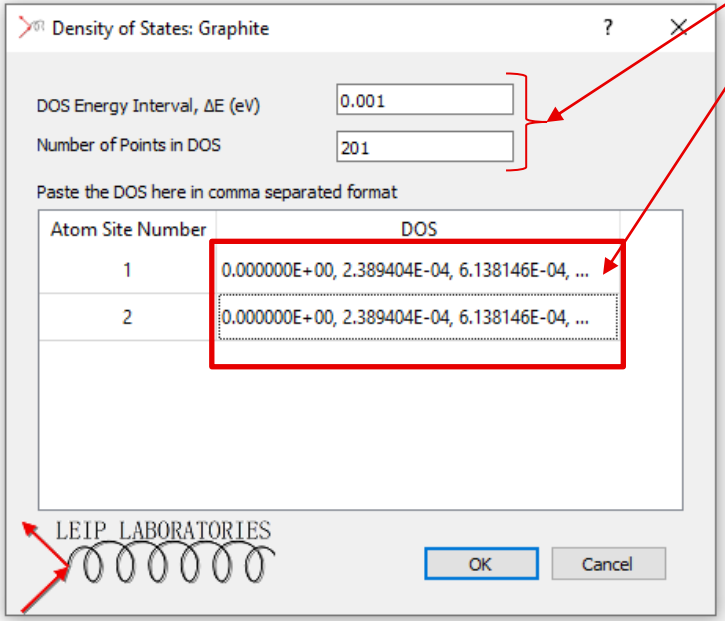


FLASSH

- ***FLASSH* GUI**
  - Density of States



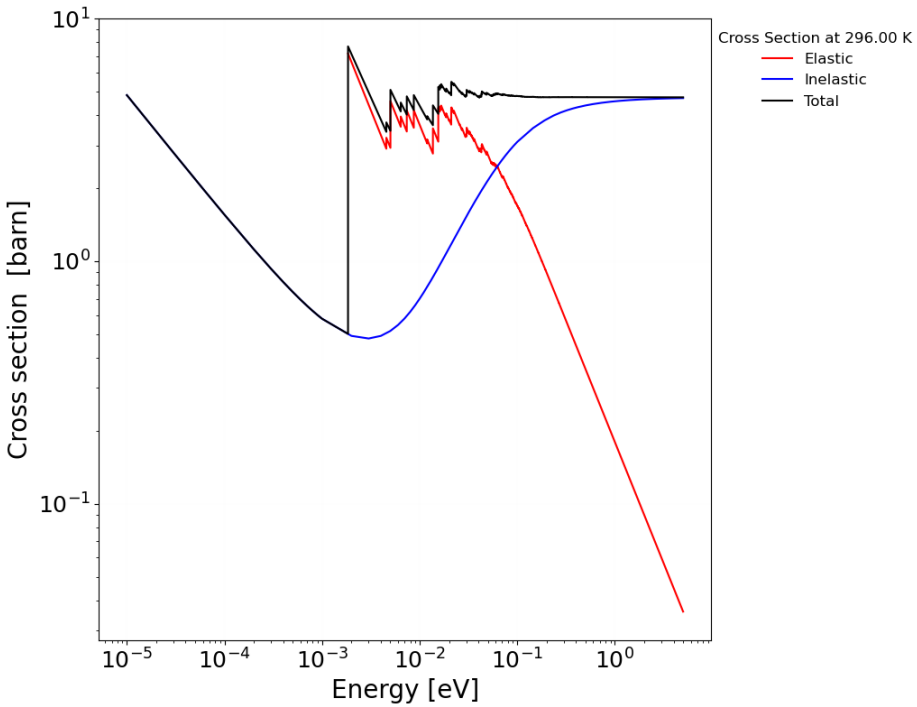
Required User Input



FLASSH

- **GUI: Plotting**
  - *FLASSH* built-in plotter produces cross section and TSL plots
  - Plots can be used by evaluators to check accuracy of results

- **ACE Output Capability**
  - *FLASSH* generated ACE files are directly compatible with codes such as MCNP
  - High resolution cross sections without requiring transfer of files

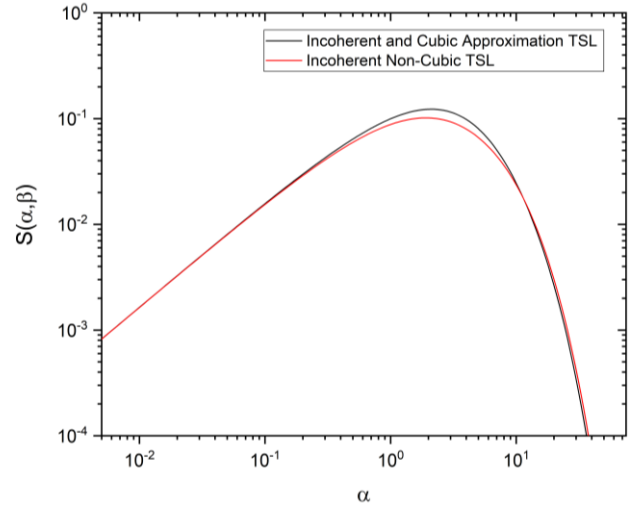


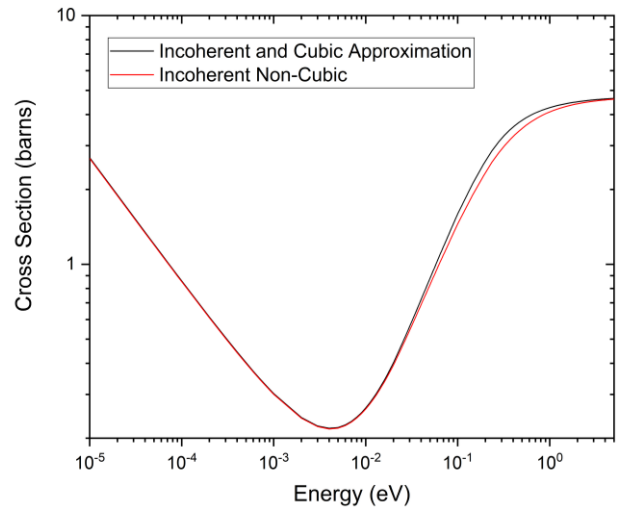
ND3/ND5 – Doppler Analysis *FLASSH* Generalized TSL

- Generalized Non-Cubic Evaluation
  - Exact material structure
  - Direction-dependent calculations averaged into α/β space
  - Effects at highest α-values

2.33E-03	-1.65E-06	2.15E-09
-1.65E-06	2.33E-03	-5.65E-09
2.15E-09	-5.65E-09	1.35E-02

Exact Debye-Waller Matrix
(Å²)





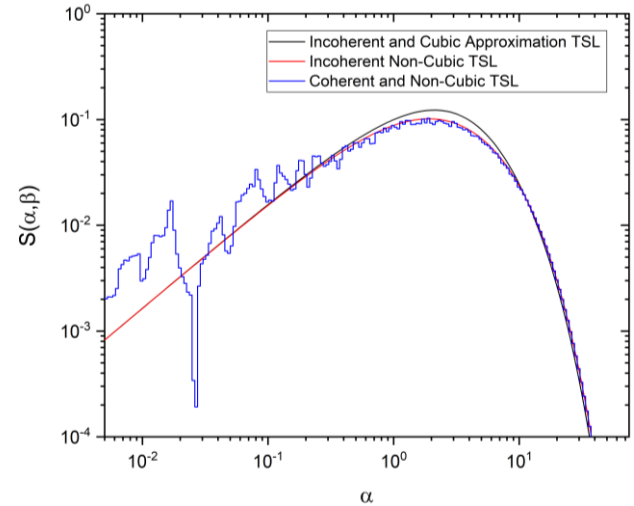
Graphite TSL data and inelastic cross sections comparing cubic and non-cubic impacts at room temperature.

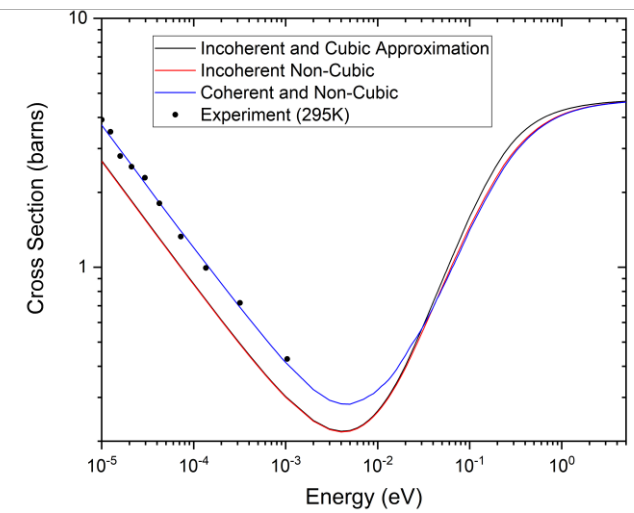
ND3/ND5 – Doppler Analysis *FLASSH* Generalized TSL

- Generalized Self and Distinct (Coherent) Evaluation
  - Exact material structure
  - Direction-dependent calculations averaged into α/β space
  - Effects at lowest α-values

2.33E-03	-1.65E-06	2.15E-09
-1.65E-06	2.33E-03	-5.65E-09
2.15E-09	-5.65E-09	1.35E-02

Exact Debye-Waller Matrix
($Å^2$)

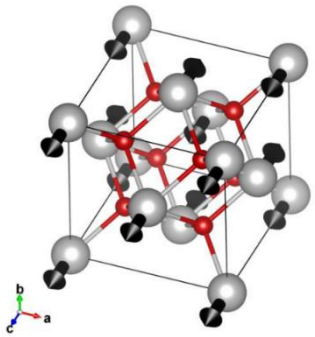




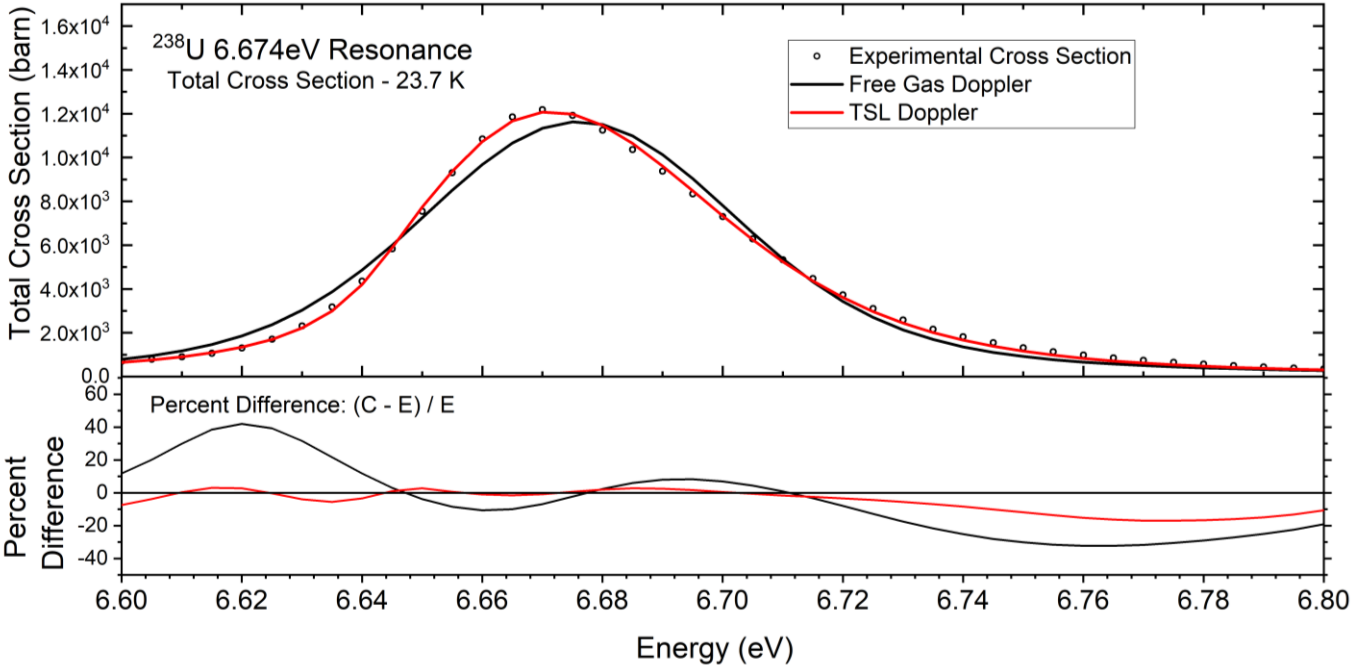
Graphite TSL data and inelastic cross sections comparing incoherent cubic and generalized full TSL impacts at room temperature.

ND5 – Doppler Broadening

- Structure Impacts
  - Initial testing with cubic UO_2 material
    - Significant impacts between free gas and thermal scattering broadening
  - Improvements in TSL theorized to improve Doppler broadening



- Fluorite Structure
- 2x2x2 supercell
- GGA-PBE+U

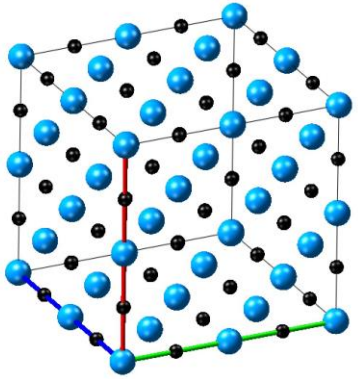


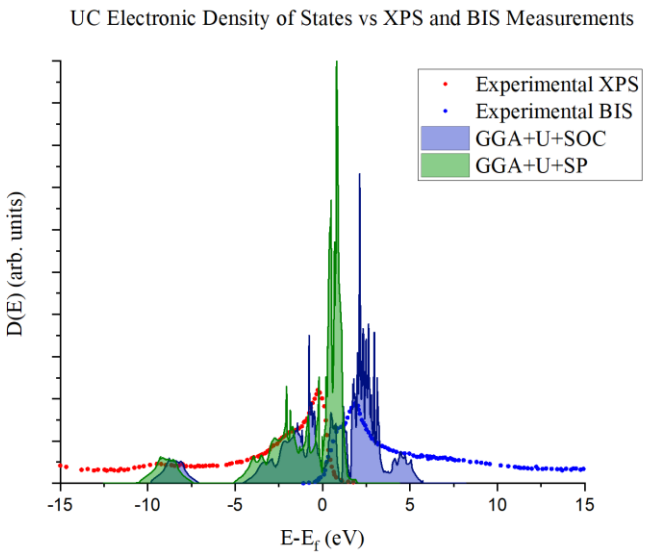
FY 21

ND2 – Uranium Carbide

DFT Model

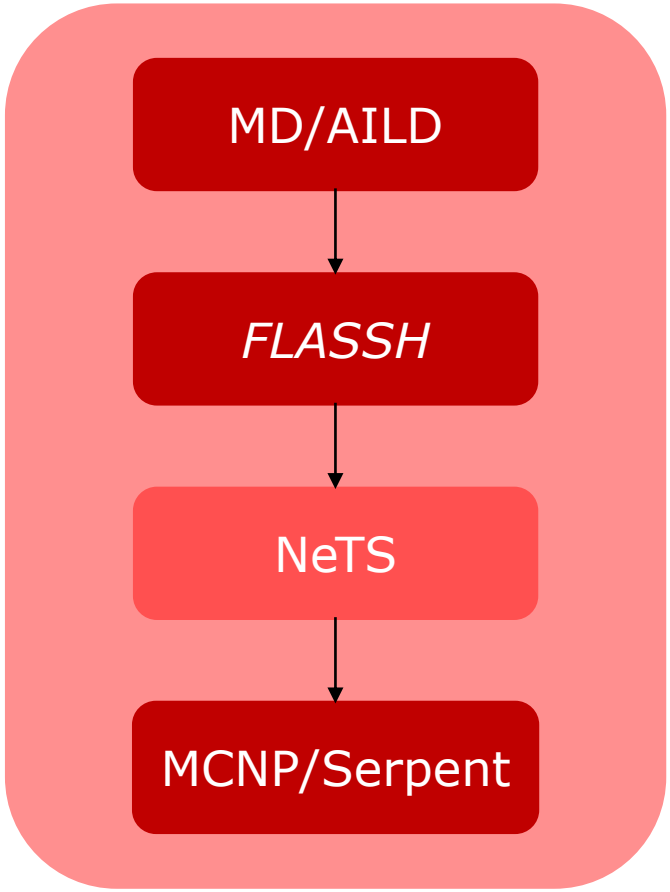
- GGA-PBE +U
- Spin Polarized: AFM-1 between (100) planes
- 2x2x2 Supercell → 64 Atoms (32 U, 32 C)
- 4 Displacements : .03 Å in ± x-direction





FY 21
ND10 – TSL NeTS

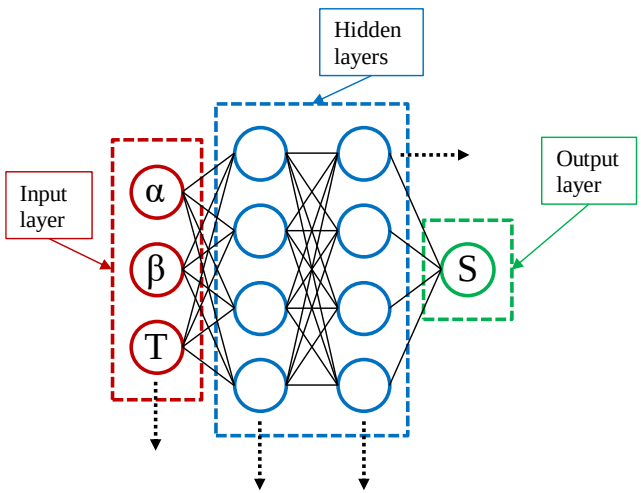
- New TSL paradigm
  - DL Neural Thermal Scattering (NeTS) modules
- See papers and presentations
  - ANS 2019 Winter Meeting, Washington, DC, USA
  - PHYSOR 2020 Meeting, Cambridge, UK



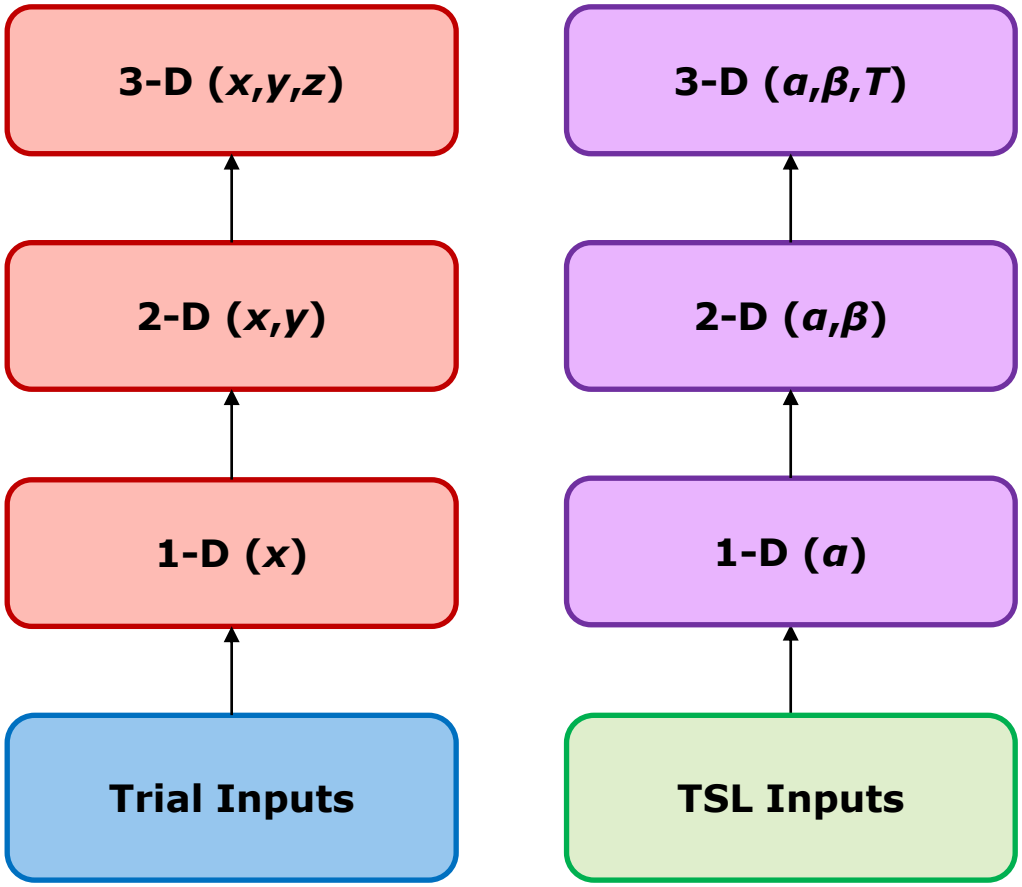
FY 21

ND10 – Progressive Development

- **Start simple**
- **Adaptive iteration**



PyTorch



Measurements
Total Cross Section & Structure

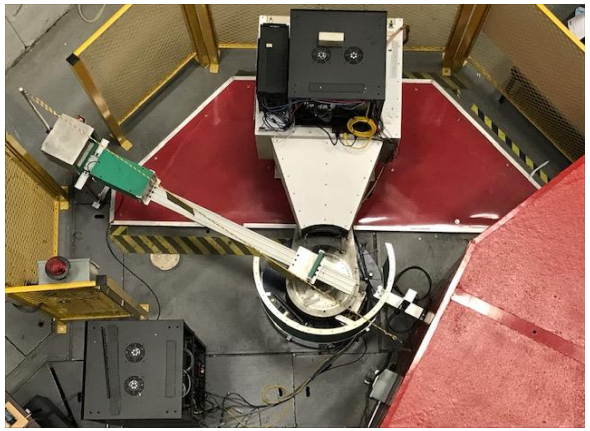
NPDF Facility Upgrades – Dual Purpose:

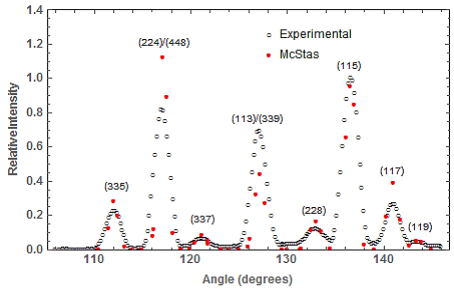
- Diffraction Measurements:

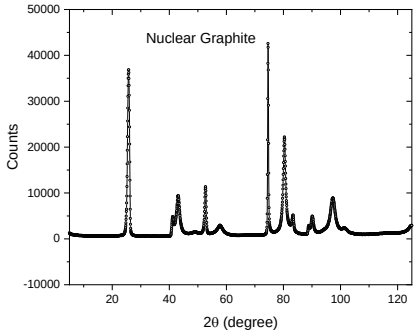
  15 New Position Encoding Modules (PEM) – improved diffraction measurement resolution $\Delta d/d$ of 2.9×10^{-3} for 3mm holder

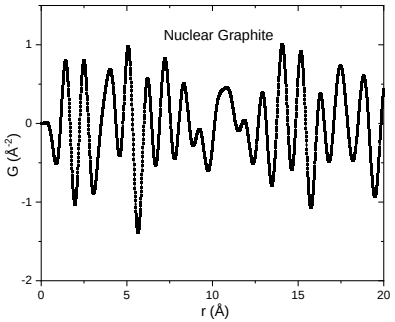
- **Transmission Measurement Capabilities:**

  - 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.









Summary

- Thermal neutron scattering law (TSL) data evaluations have been completed and contributed to ENDF
  - New evaluations are underway
- Advanced TSL techniques in combination with Doppler analysis have been developed
- *FLASSH* is developing as a modern platform for thermal neutron data analysis
- ML/DL NeTS method development initiated
- Experimental capabilities for validation have been developed at the PULSTAR reactor