Technical Program Review
Nuclear Criticality Safety Program
March 27 – 28, 2018 • Oak Ridge, TN, USA

Thermal Scattering Law Research and Development At North Carolina State University

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Nuclear Reactor Program
Department of Nuclear Engineering
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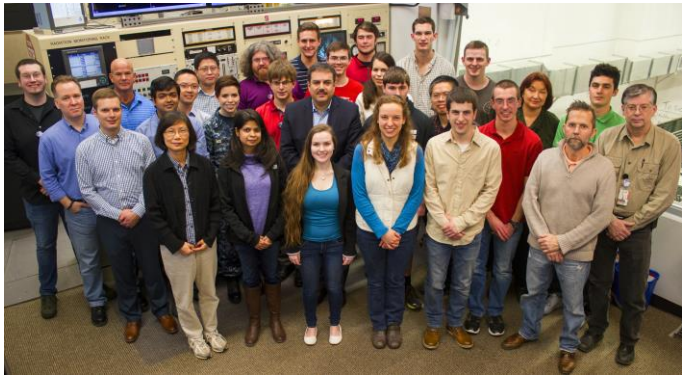
Acknowledgement

- The many graduate students, postdocs, and research staff at North Carolina State University

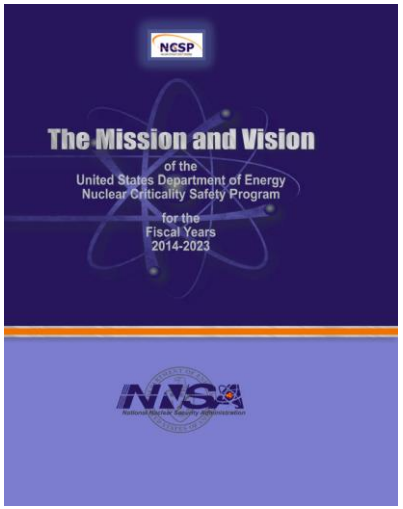


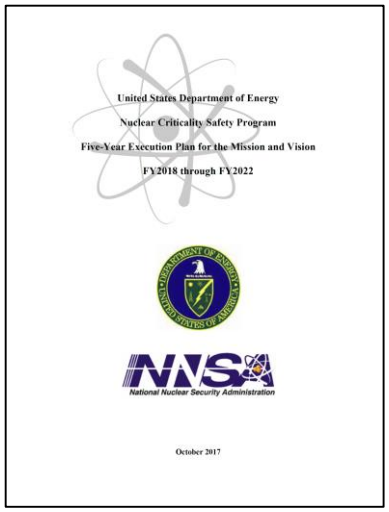
- Collaboration with LLNL and Bettis labs
  - David Heinrichs, Michael Zerkle, Jesse Holmes
- Funding
  - US NNSA Nuclear Criticality Safety program
  - US Naval Nuclear Propulsion Program

FY 2017

- 10 new TSL evaluations contributed to ENDF/VIII
  - 5 first-of-a-kind evaluations
- Modern predictive methods for thermal neutron cross section calculations based on the use of atomistic simulations
  - Ab initio lattice dynamics
  - Molecular dynamics (ab initio and classical)
    - New materials
    - All states of matter (solid, liquid, gas)
    - Imperfect structure
- Implemented a new thermal scattering analysis platform that uses a generalized theoretical approach for TSL calculations.
  - Removed approximations such as incoherent approximation, cubic approximation, atom site approximation and SCT approximation.

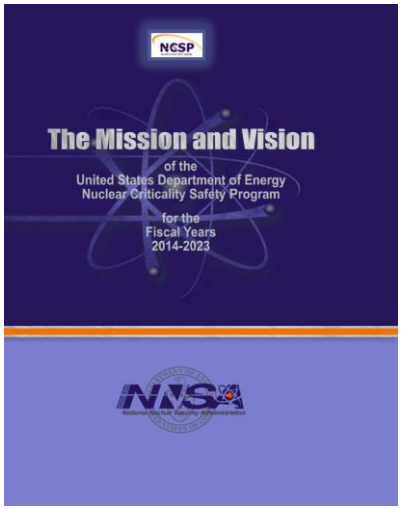
Objectives

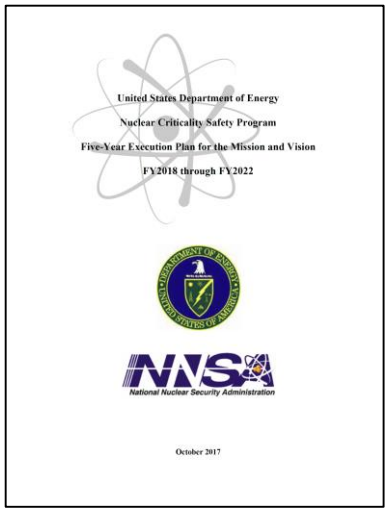




Priority Needs */ **Additional Needs**	Thermal scattering (Paraffinic Oil, HF, Silicone Oil, UO_2F_2, PuH_2, UH_3, Paraffin, U_3O_8, U_3Si_2, UC, PuO_2, etc.), ^{239}Pu, Fe, Cr, ^{237}Np, Pb, ^{55}Mn, Ti, ^{240}Pu/ ^{233}U, Th, Be, ^{51}V, Zr, F, K, Ca, Mo, Na, La
Completed Evaluations (FY)	Minor Actinides (13), SiC(17), SiO_2 (17), $C_5O_2H_8$ (16), CH_2 (17), Be (17), BeO (17), Graphite (17), UO_2 (17), UN (17), ^{55}Mn (12), ^{58,60}Ni (14), ^{180,128,183,184,186}W (14), Ca (16), ^{59}Co (17), ^{63,65}Cu(17)

Objectives





Priority Needs */ Additional Needs	Thermal scattering (Paraffin, Oil, HF, Silicone Oil, UO_2F_2, PuH_2, UH_3, Paraffin, U_3O_8, U_3Si_2, UC, PuO_2, etc.), ^{239}Pu, Fe, Cr, ^{237}Np, Pb, ^{55}Mn, Ti, ^{240}Pu/^{233}U, Th, Be, ^{51}V, Zr, F, K, Ca, Mo, Na, La
Completed Evaluations (FY)	Minor Actinides (13), Si (17), Si (17), $C_5O_2H_8$ (16), C (17), Be (17), B (17), Graphite (17), U (17), U (17), ^{55}Mn (12), ^{58,60}Ni (14), ^{180,128,183,184,186}W (14), Ca (16), ^{59}Co (17), ^{63,65}Cu(17)

Working on light water (H_2O)

Evaluations

Material	Method	Status
Lucite $(C_5H_8O_2)_n$	MD	ENDF/B-VIII.0
Polyethylene $(CH_2)_n$	MD	ENDF/B-VIII.0
Beryllium (Be metal)	DFT/LD	ENDF/B-VIII.0
Beryllium oxide (BeO)	DFT/LD	ENDF/B-VIII.0
Silicon carbide (SiC)	DFT/LD	ENDF/B-VIII.0
Silicon dioxide (SiO_2)	DFT/LD	ENDF/B-VIII.0
Graphite (crystalline)	DFT/LD	ENDF/B-VIII.0
Graphite (nuclear)	MD	ENDF/B-VIII.0
Uranium mononitride (UN)	DFT/LD	ENDF/B-VIII.0
Uranium dioxide (UO_2)	DFT/LD	ENDF/B-VIII.0
H_2O (light water)	MD	On going
Paraffinic Oil	MD	ENDF File 7 ready
FLiBe liquid	MD	ENDF File 7 ready

Evaluations

Material	Method	Status
Lucite $(C_5H_8O_2)_n$	MD	ENDF/B-VIII.0
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Silicon dioxide (SiO_2)	DFT/LD	ENDF/B-VIII.0
Graphite (crystalline)	DFT/LD	ENDF/B-VIII.0
Graphite (nuclear)	MD	ENDF/B-VIII.0
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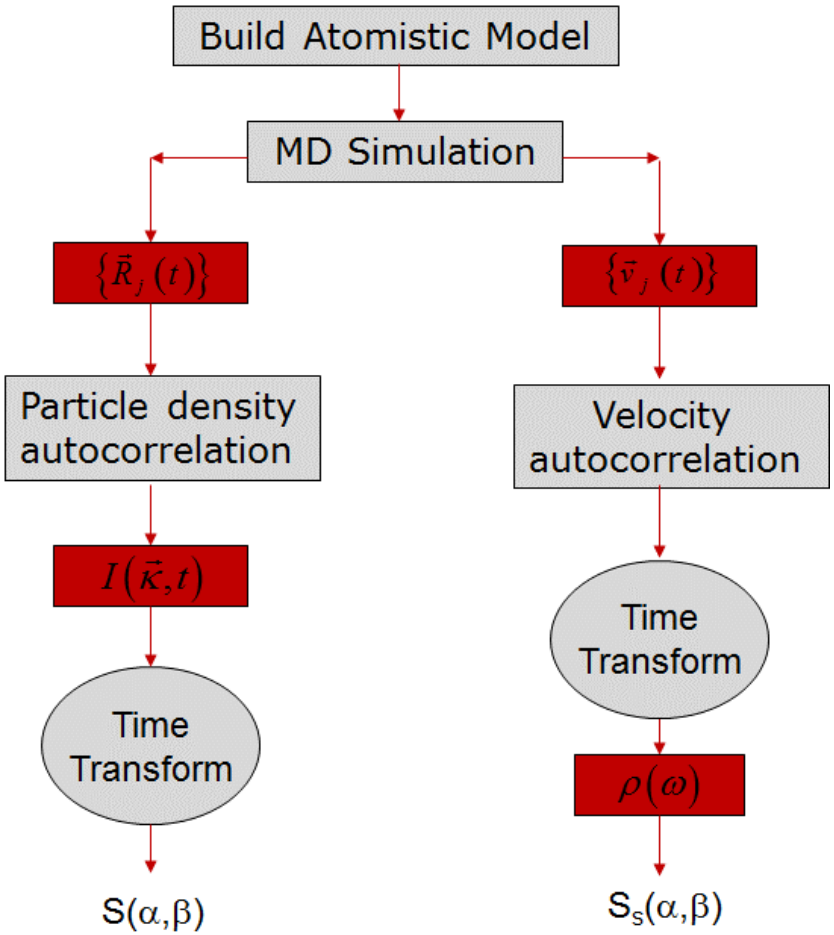
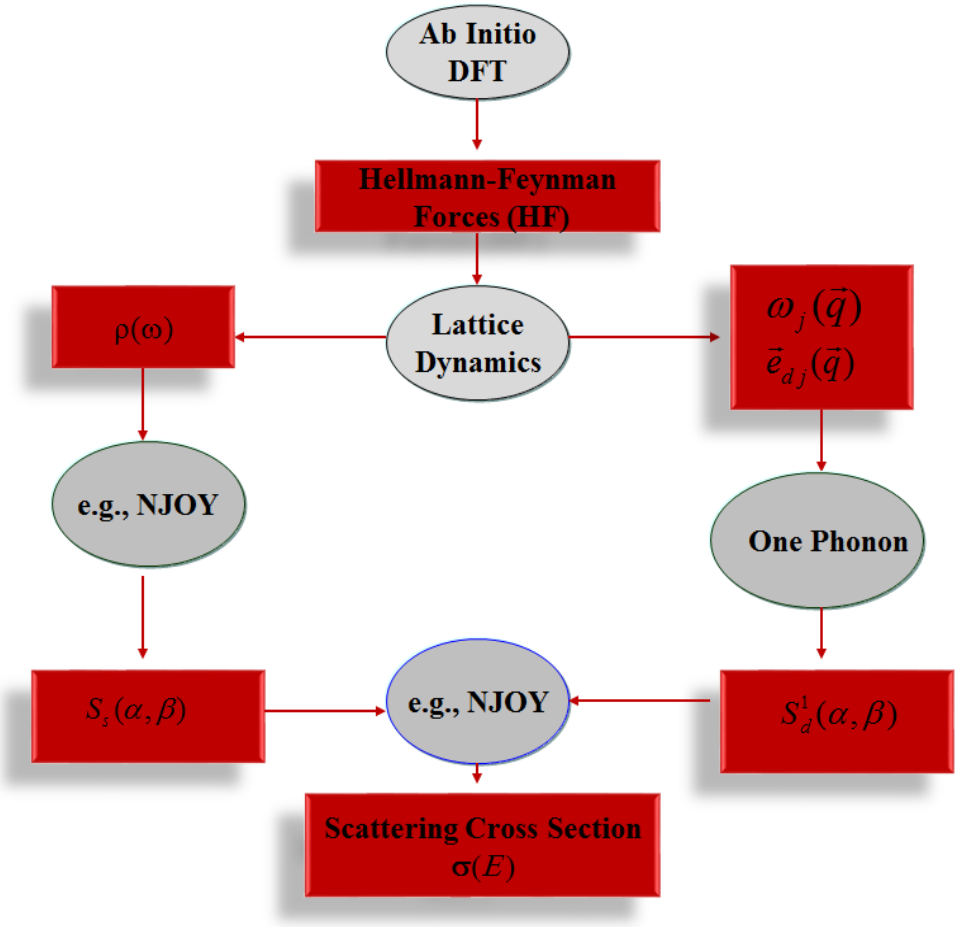
H_2O (light water)	MD	On going
Paraffinic Oil	MD	ENDF File 7 ready
FLiBe liquid	MD	ENDF File 7 ready

Methods





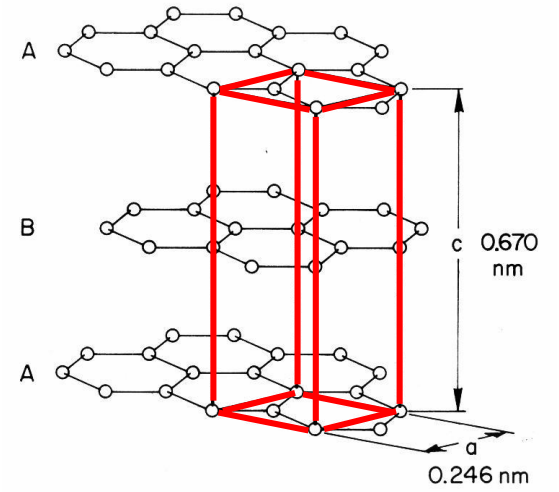
A. I. Hawari, "Modern Techniques for Inelastic Thermal Neutron Scattering Analysis," Nuclear Data Sheets, Vol. 118, 172, 2014.

Computational Capabilities

- Hybrid mini cluster - 16 nodes
  - 312 CPU cores
  - 22 computational GPU
  - Expanding…..
- Parallel computations
  - Atomistic simulations
  - TSL analysis
  - Neutronic simulations
  - System design
- VASP, PHONON, LAMMPS NJOY, MCNP, Serpent, GEANT4, McStas, PARET, RELAP, COMSOL

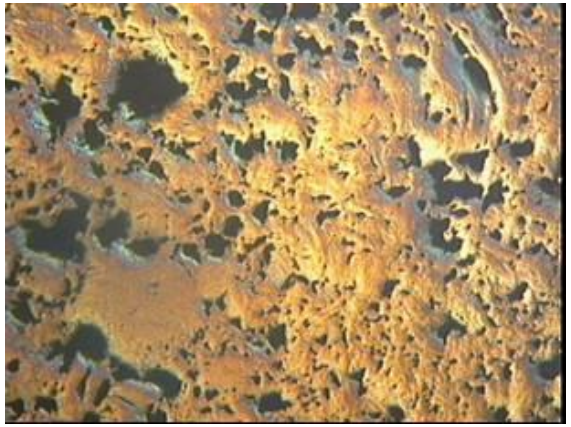
Graphite

Ideal "crystalline" graphite consists of planes (sheets) of carbon atoms arranged in a hexagonal lattice. Covalent bonding exits between intraplaner atoms, while the interplaner bonding is of the weak Van der Waals type. The planes are stacked in an "abab" sequence.



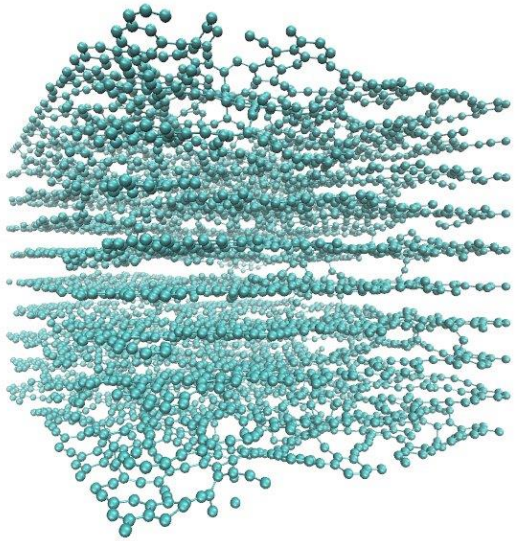
- Hexagonal Structure
- 4 atoms per unit cell
- a = b = 2.46 Å
- c = 6.7 Å
- Density = 2.25 g/cm^3

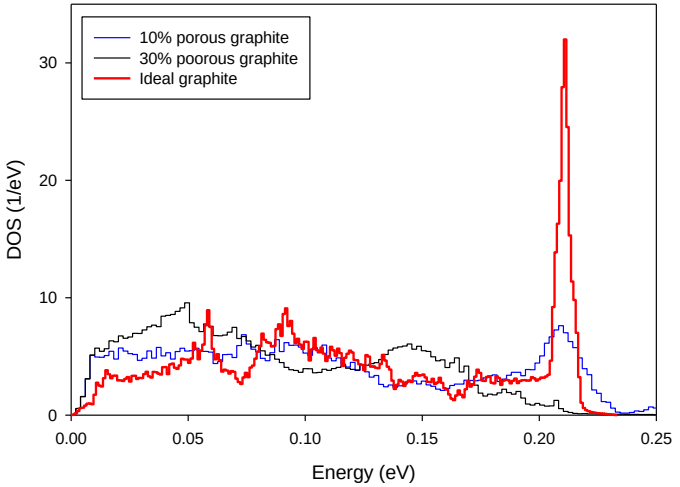
Reactor graphite consists of ideal graphite crystallites (randomly oriented) in a carbon binder. It is highly porous structure with porosity level ranging between 10% and 30%.

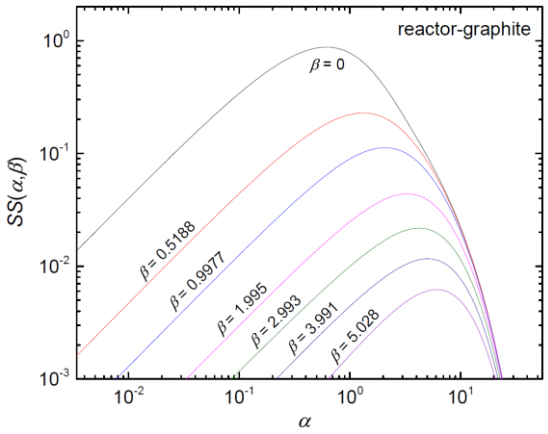


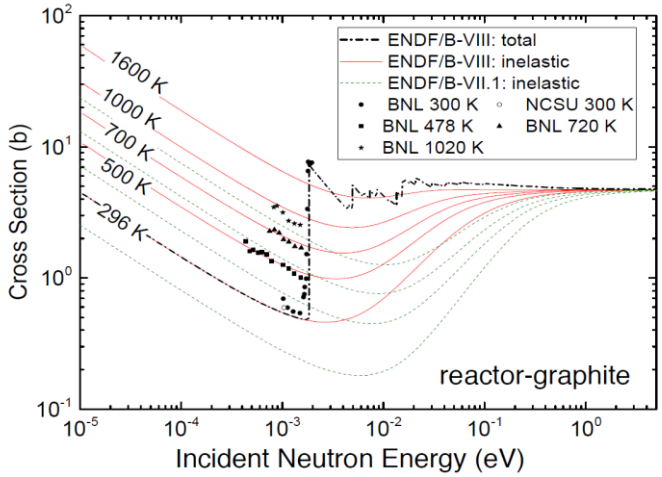
Nuclear Graphite (SEM at NCSU)
Density = 1.5 – 1.8 g/cm^3

Nuclear/Reactor Graphite

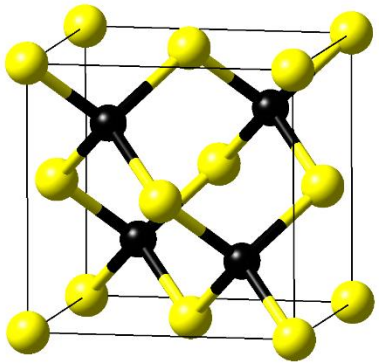


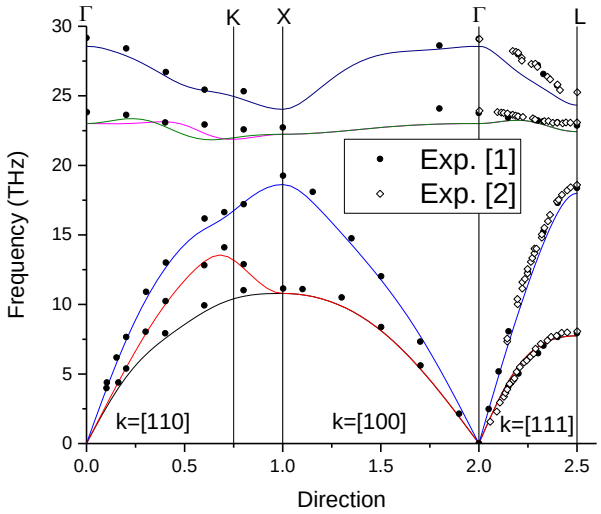


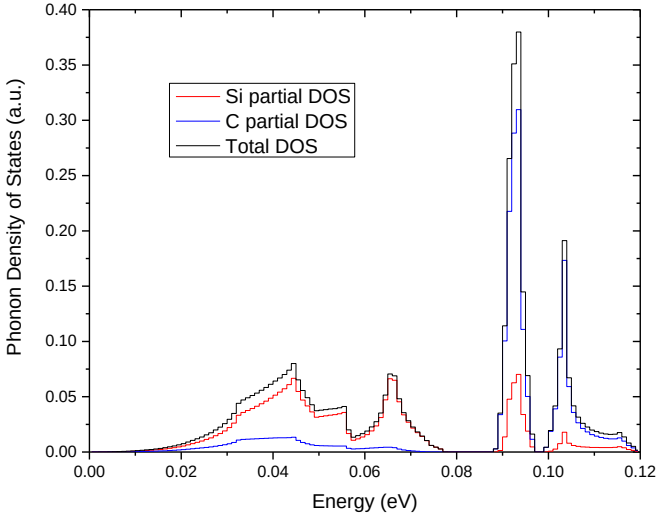




Silicon Carbide(SiC)

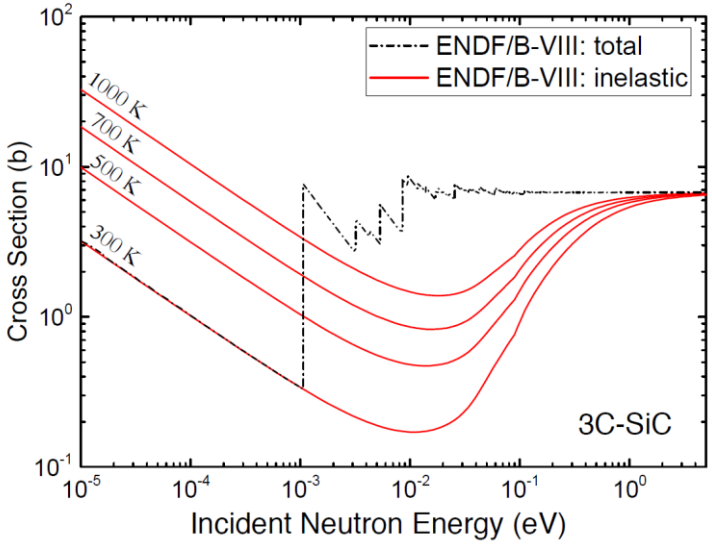






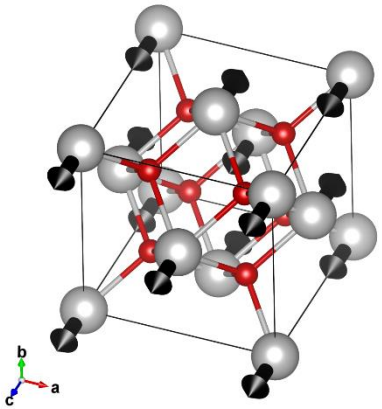
- FCC Structure
- 3x3x3 super-cell
- GGA

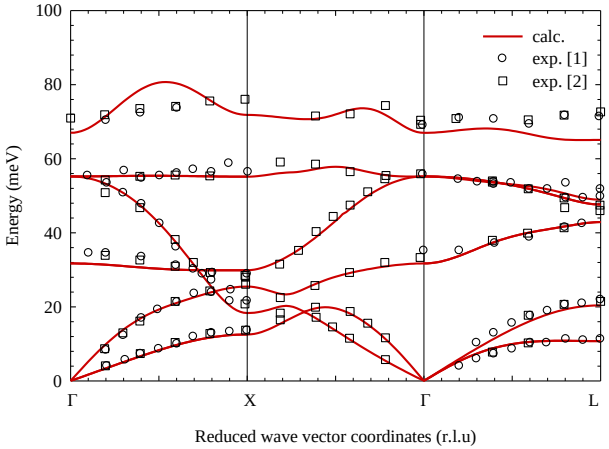
	a (Å)
DFT	4.379
Exp.	4.3596

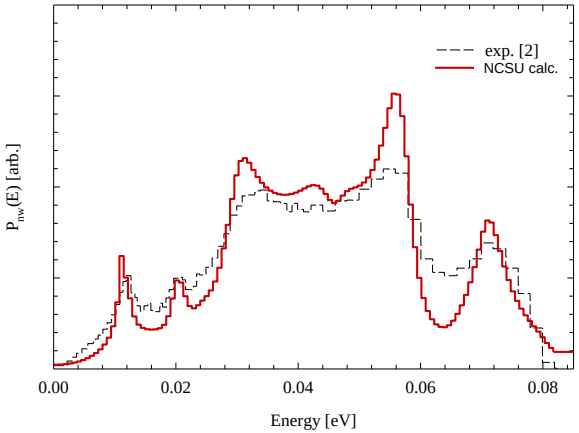


[1] S. Bagci, S. Duman, H.M. Tutuncu, G.P. Srivastava, "Theoretical studies of SiC, AlN and their (110) surfaces", DIAM. RELAT. MAT. 18, 1057 (2009).
[2] K. Karch, P. Pavone, W. Windl, D. Strauch, F. Bechstedt, "Ab initio calculation of structural, lattice dynamical, and thermal properties of cubic silicon carbide", INTERNATIONAL JOURNAL OF QUANTUM CHEMISTRY. 56, 801 (1995).

Uranium Dioxide (UO_2)

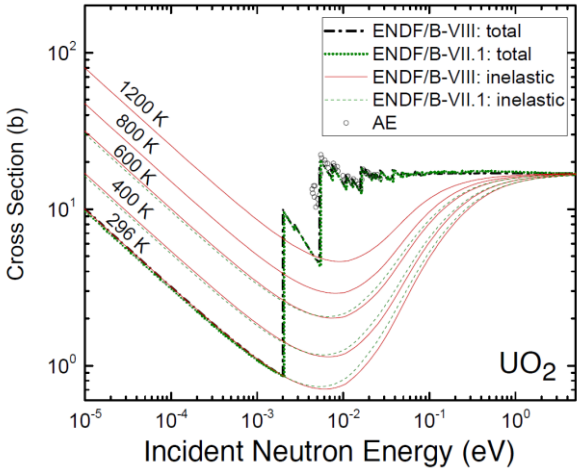






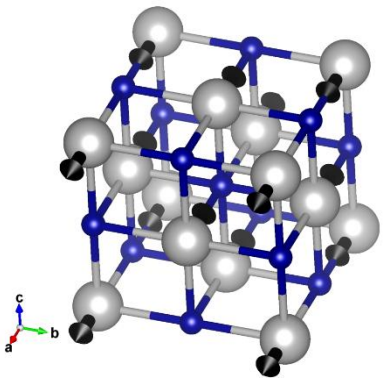
- Fluorite structure
- 2x2x2 super-cell
- GGA-PBE+U

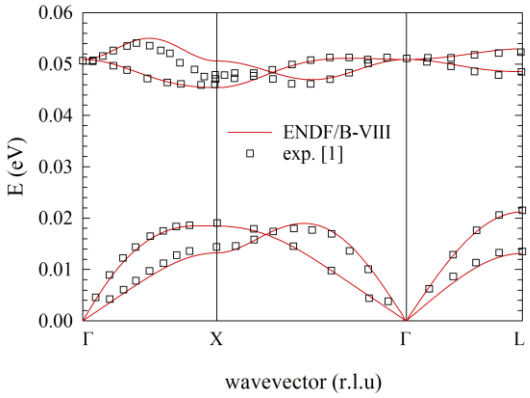
	DFT	Exp.
a [Å]	5.547	5.471
E_g [eV	1.96	2.0
μ [μ_B]	1.96	1.74

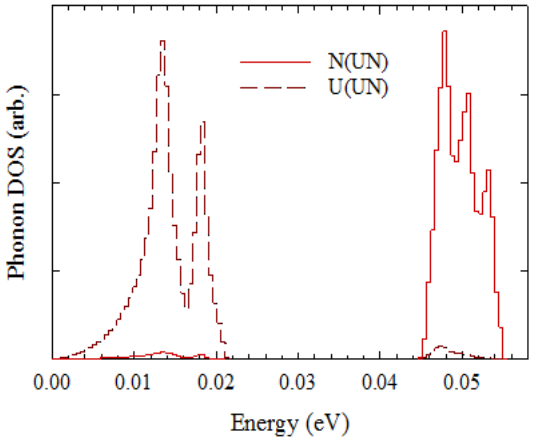


1) G. Dolling, R. A. Cowley, and A. D. B. Woods, "The crystal dynamics of Uranium Dioxide," Can. J. Phys., 43, 8 (1965) 1397
2) J. W. L. Pang, A. Chernatynskiy, B. C. Larson, W. J. L. Buyers, D. L. Abernathy, K. J. McClellan, and S. R. Phillpot, "Phonon density of states and anharmonicity in UO2," Phys. Rev. B, 89, (2014) 115132

Uranium Nitride (UN)

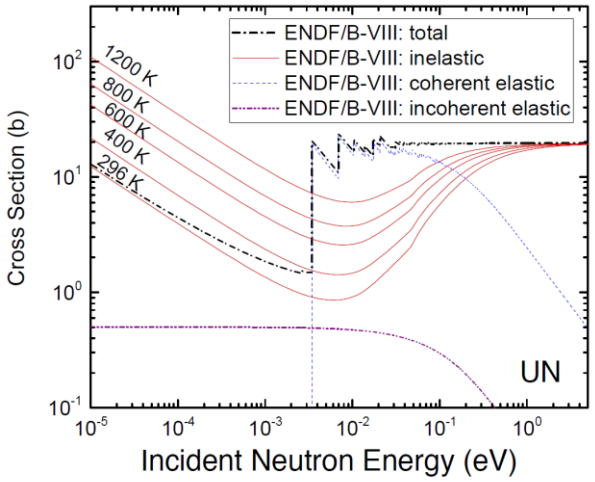


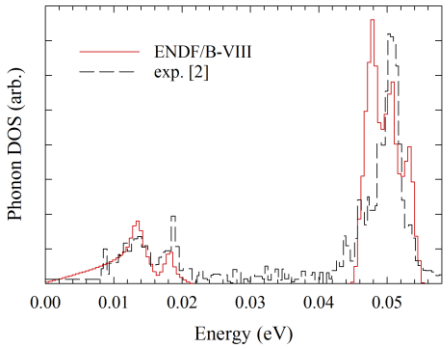




- Rock-salt structure
- 2x2x2 super-cell
- GGA-PBE

	a (Å)	μ (μB)	Bulk Modulus (GPa)
DFT	4.859	0.98	221
Exp.	4.880-4.890	0.75	203

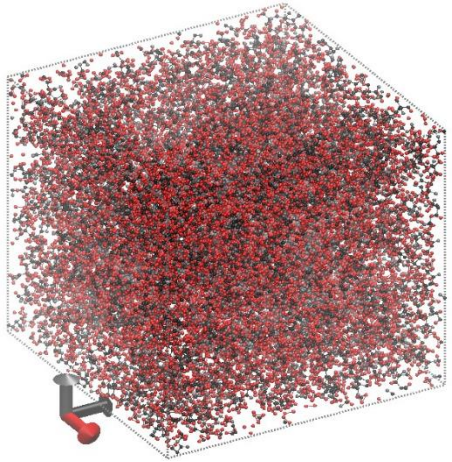


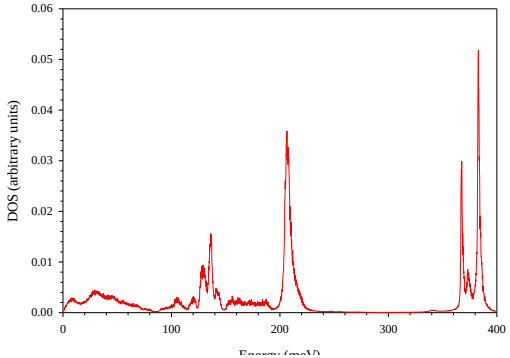


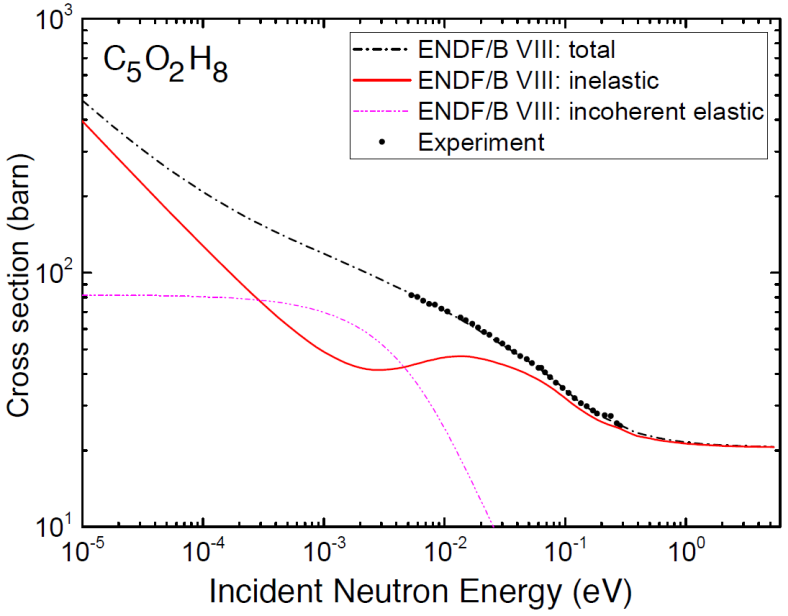
1) G. Dolling, T. M. Holden, E. C. Evenson, W. J. L. Buyers, and G. H. Lander,"Phonon dispersion relation of uranium nitride above and below the Neel temperature," International Conference on Lattice Dynamics, Paris, Frances, September 5-10 (1977).

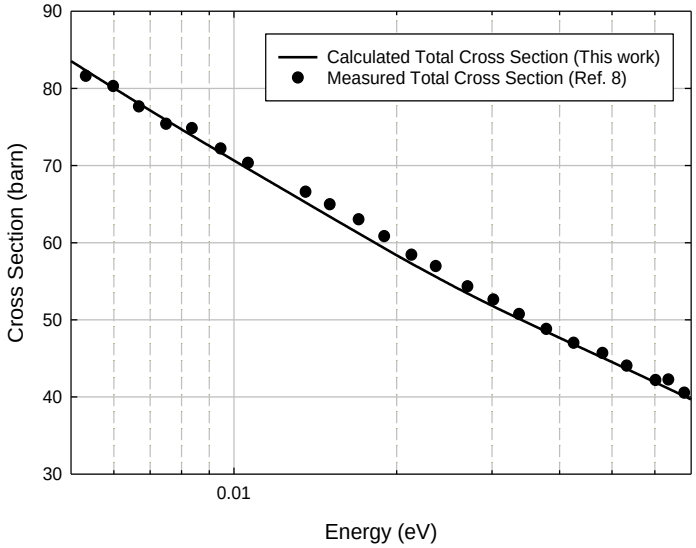
2) A. A. Aczel, G. E. Granroth, G. J. MacDougall, W. J. L. Buyers, D. L. Abernathy, G. D. Samolyuk, G. M. Stocks, and S. E. Nagler,"Quantum oscillations of nitrogen atoms in uranium nitride," Nature Communications, 3 (2012)

Lucite $(C_5O_2H_8)_n$



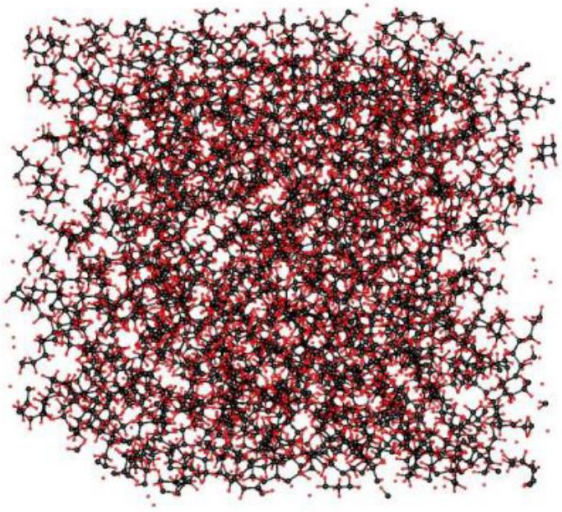


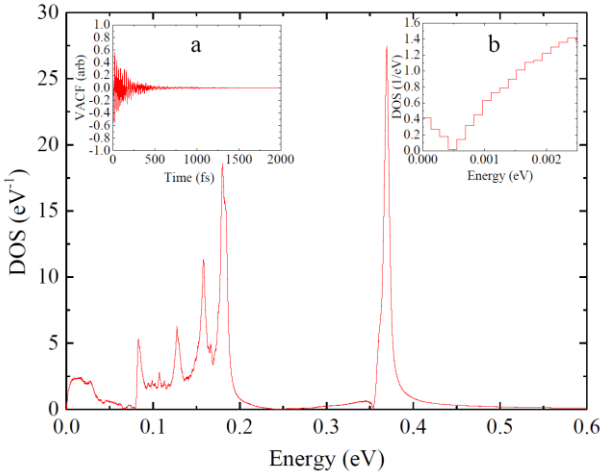


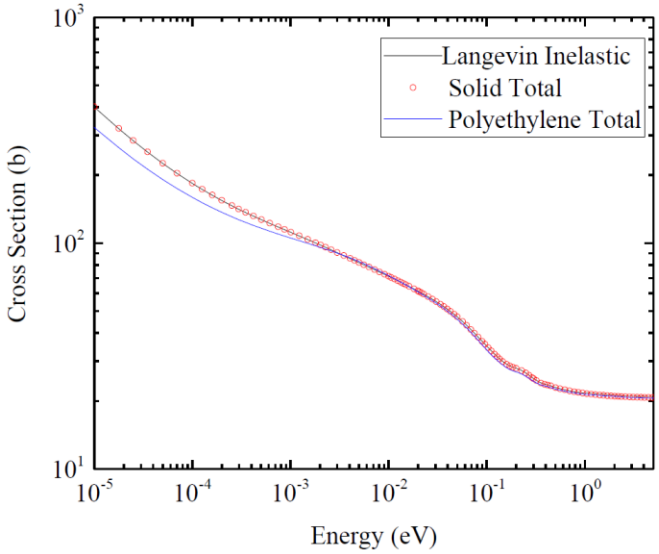


1) A. I. Hawari et al "Analysis of Thermal Neutron Scattering in Polymethyl Methacrylate (Lucite)," Transactions of the American Nuclear Society, 113, 2015.
2) Exp. Data: G. SIBONA et al., Anna. Nucl. Energy, 18, 689 (1991).

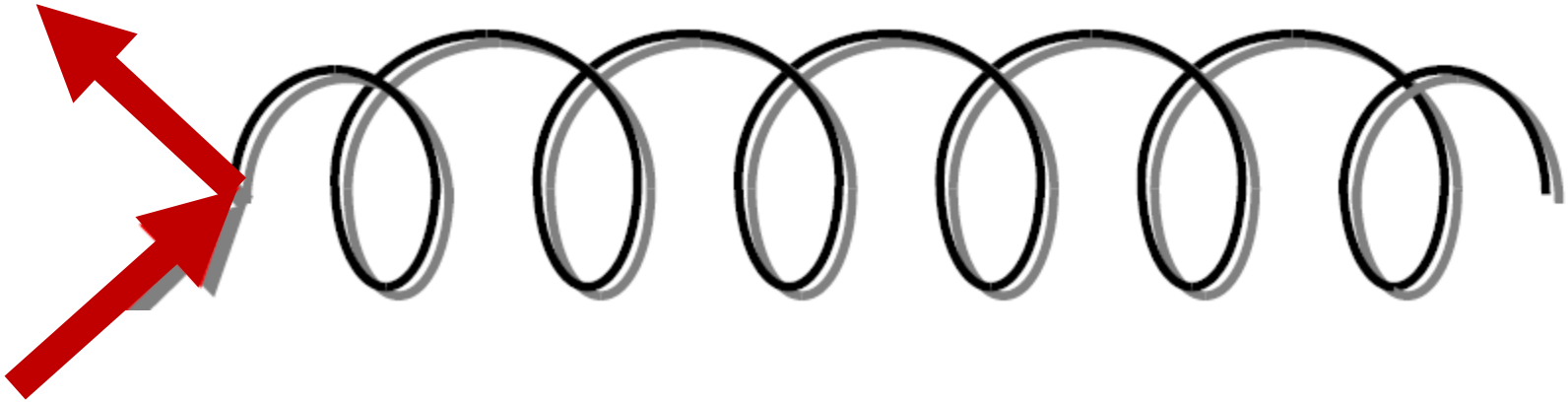
Heavy Paraffinic Molecular Materials





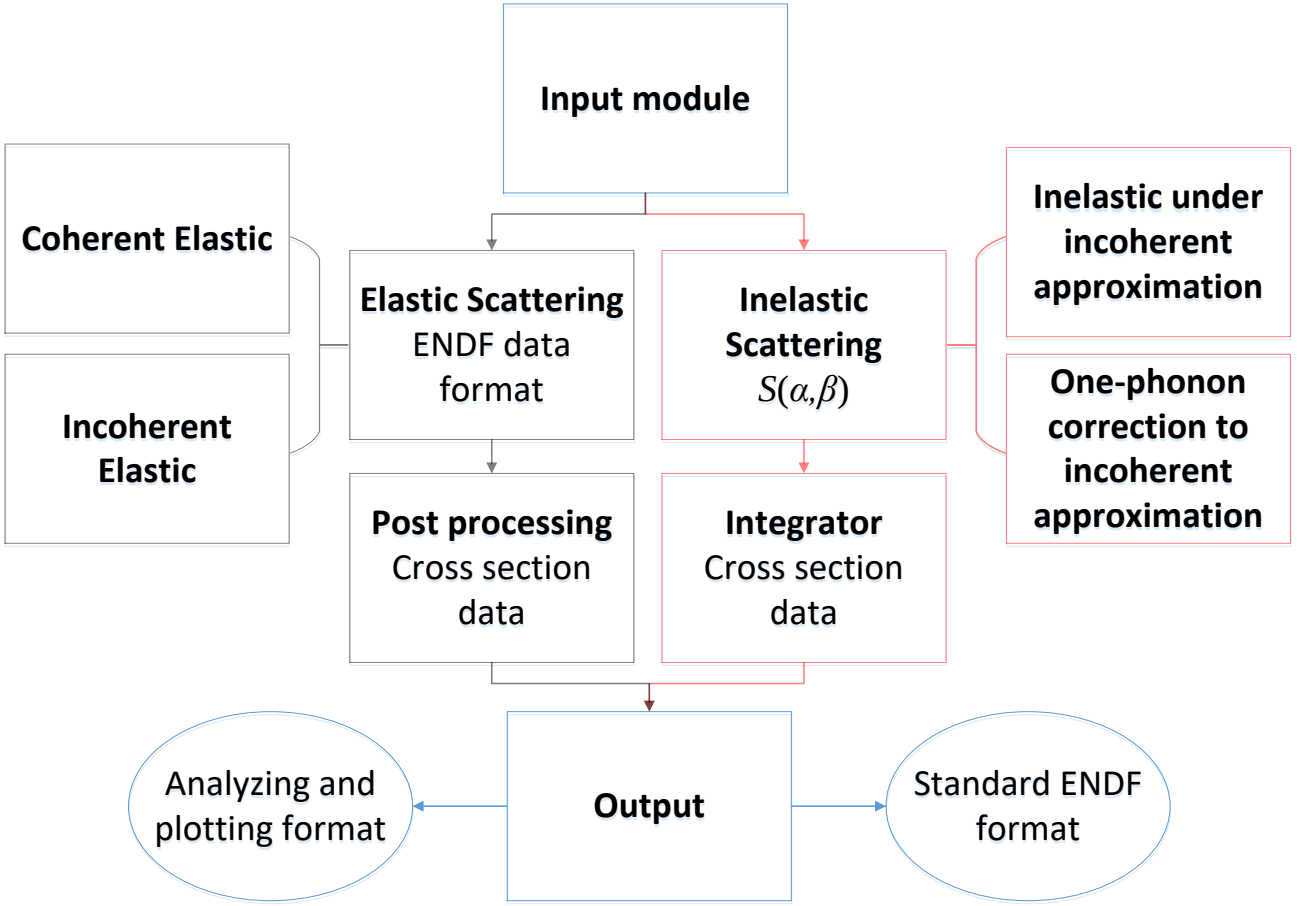


FLASSH



Full Law Analysis Scattering System Hub

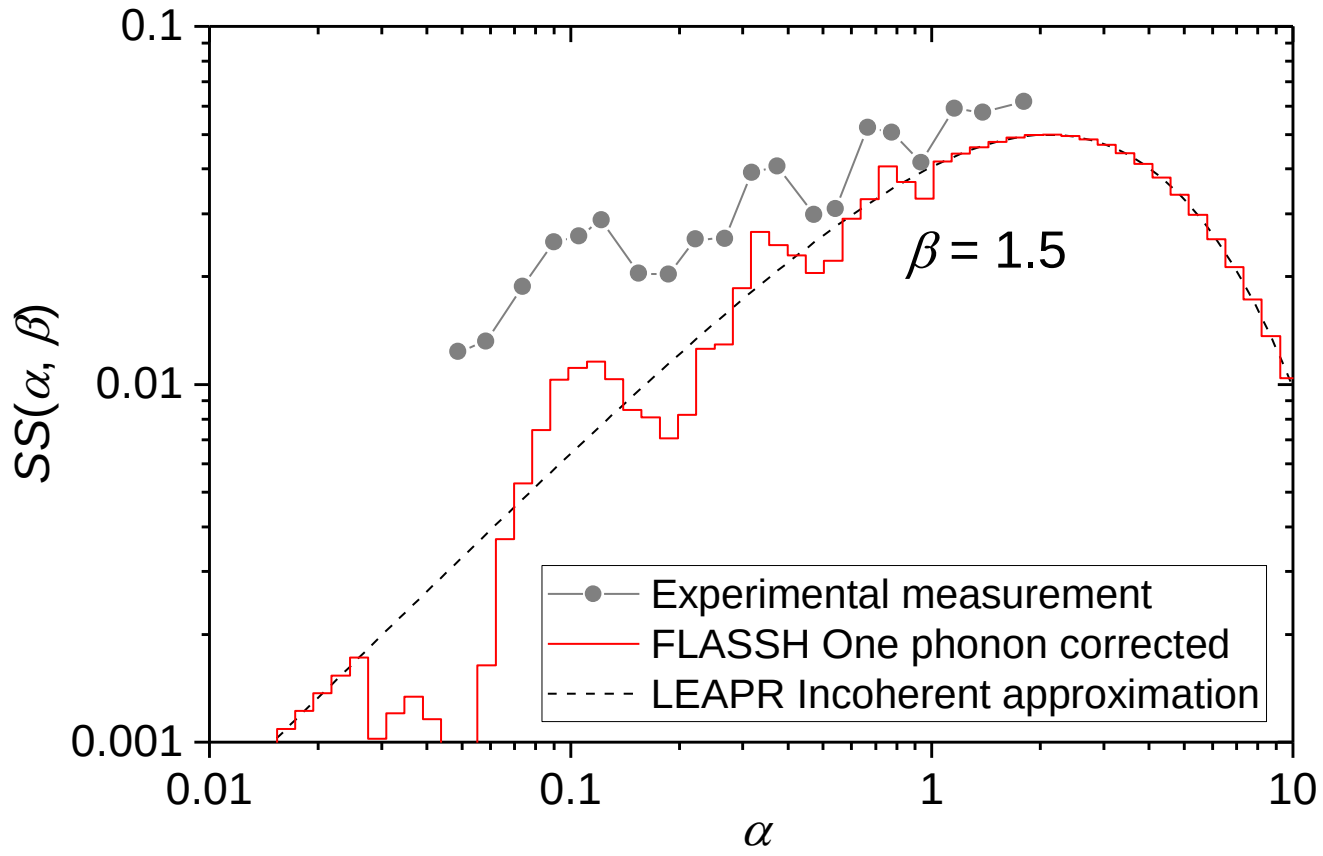
FLASSH


Input module
Coherent Elastic
Incoherent Elastic
Elastic Scattering
ENDF data format
Inelastic Scattering
$S(\alpha,\beta)$
Inelastic under incoherent approximation
One-phonon correction to incoherent approximation
Post processing
Cross section data
Integrator
Cross section data
Analyzing and plotting format
Output
Standard ENDF format

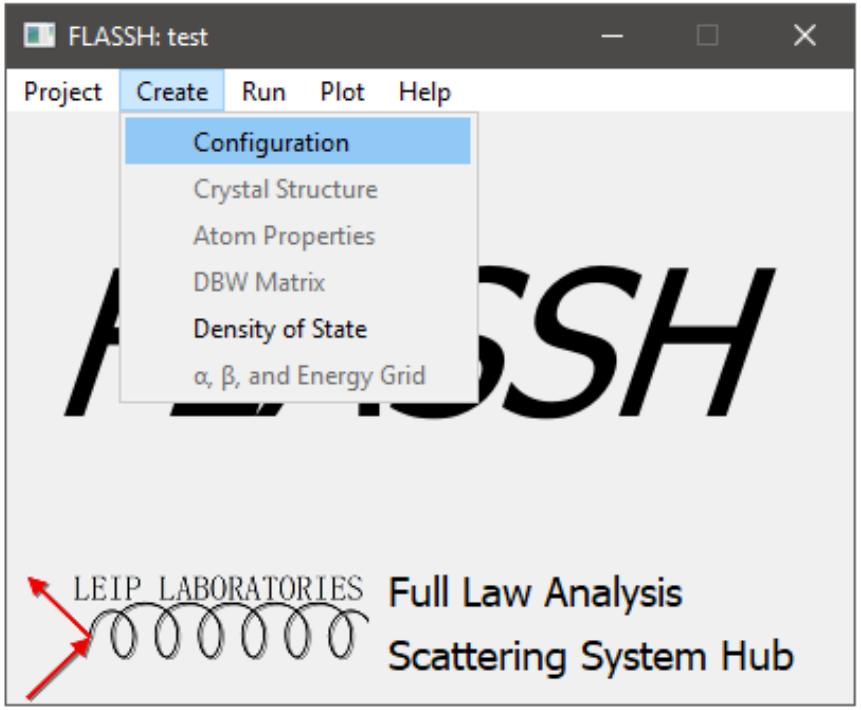
FLASSH Features

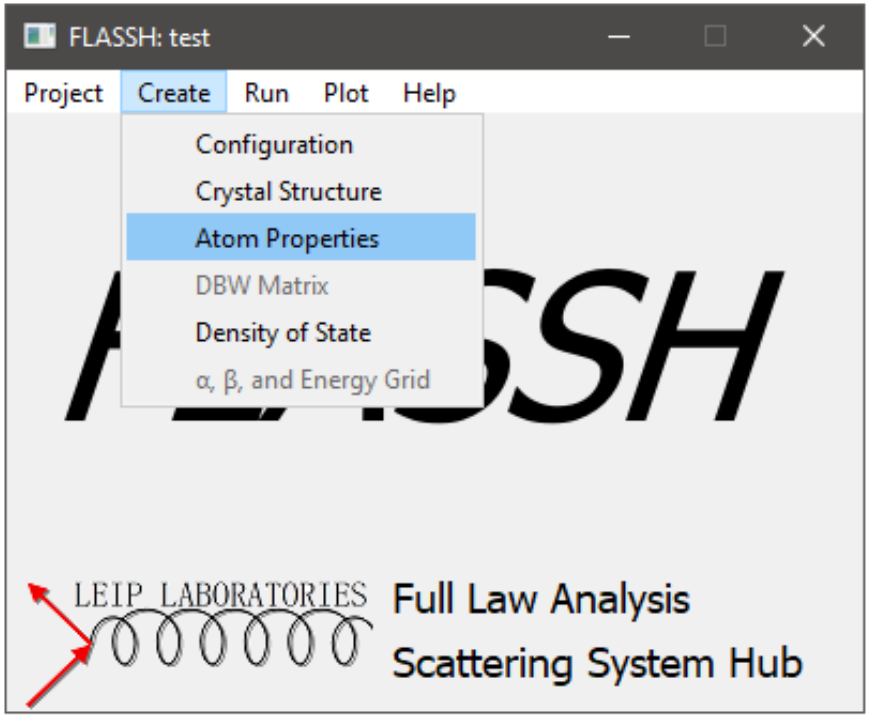
Feature	*FLASSH*	NJOY
Incoherent approximation	No	Yes
Cubic approximation	No	Yes
Atom site approximation	No	Yes
Short collision time approximation	No	Yes
Gaussian approximation	Yes	Yes
Harmonic approximation	Yes	Yes
Parallel processing	Yes	No
GUI	Yes	No

One-phonon Corrected $S(\alpha, \beta)$ Be Metal



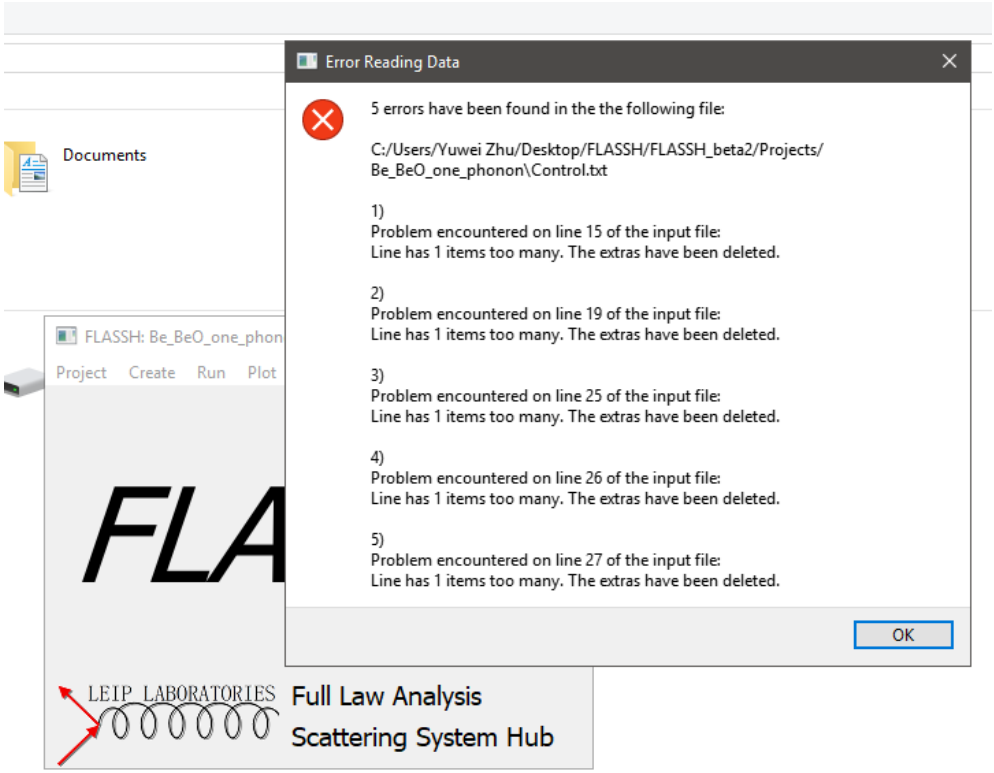
FLASSH GUI

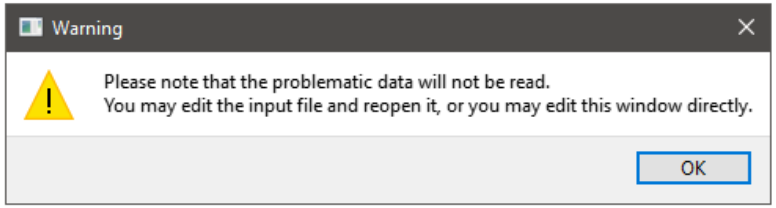




- Guide the user through input items by input file logic

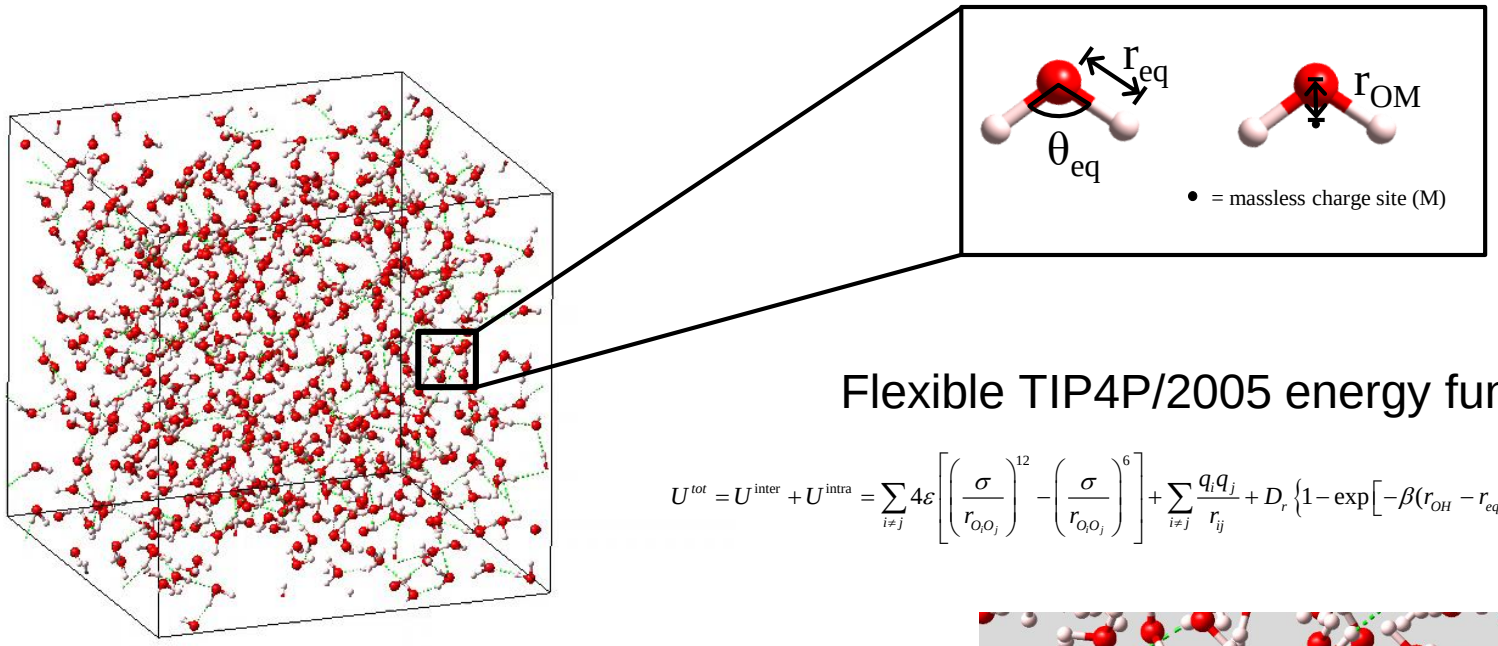
FLASSH GUI





- Input file syntax checking
- Warning about input file error

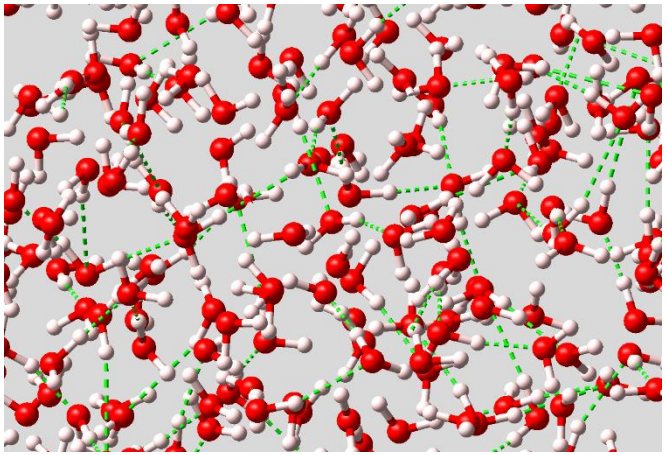
Light Water (H_2O) – FY 2018



Flexible TIP4P/2005 energy function:

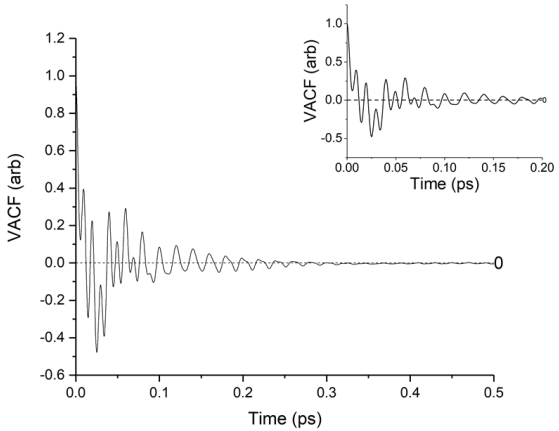
$$U^{tot} = U^{inter} + U^{intra} = \sum_{i \neq j} 4\varepsilon \left[\left(\frac{\sigma}{r_{O_i O_j}} \right)^{12} - \left(\frac{\sigma}{r_{O_i O_j}} \right)^{6} \right] + \sum_{i \neq j} \frac{q_i q_j}{r_{ij}} + D_r \left\{ 1 - \exp\left[-\beta (r_{OH} - r_{eq}) \right] \right\}^2 + \frac{1}{2} K_\theta (\theta - \theta_{eq})^2$$

Static view of hydrogen-bond network in liquid H_2O:
Red atoms are oxygen, white atoms are hydrogen and hydrogen-bonds are shown as dashed green lines (<3.0 Å)



Light Water (H_2O) – FY 2018

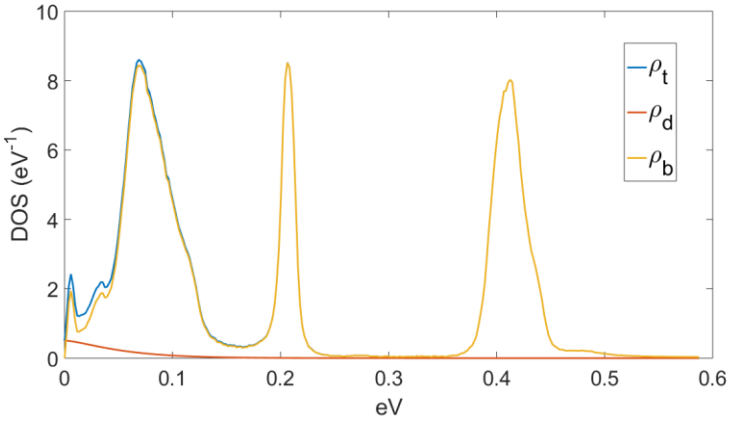
Velocity autocorrelation function (VACF) of liquid H_2O @ 300 K



$$\rho_t(\omega) = \int_0^\infty \exp(-i\omega t) C(t) dt$$

Fourier transform of VACF

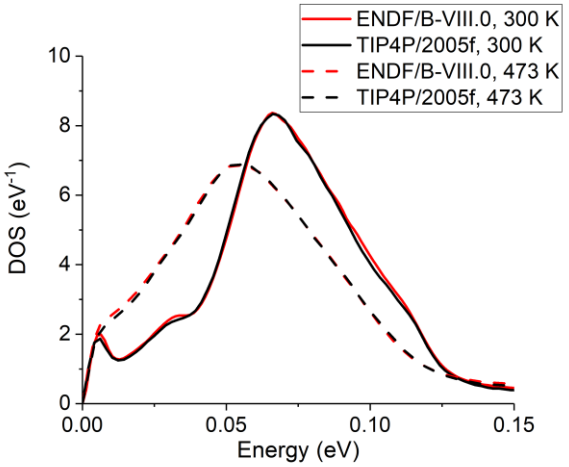
Vibrational DOS of liquid H_2O @ 300 K



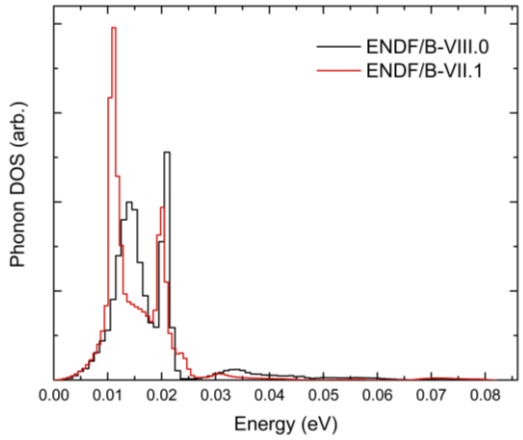
	Density (g/cm³)		D_{self} (10^{-5} cm²/s)	
	300 K	473 K	300 K	473 K
TIP4P/2005f	0.987	0.837	2.33	18.2
Experiment	0.997	0.904 *	2.30	23.8

*Extrapolated from experimental data up to 373 K

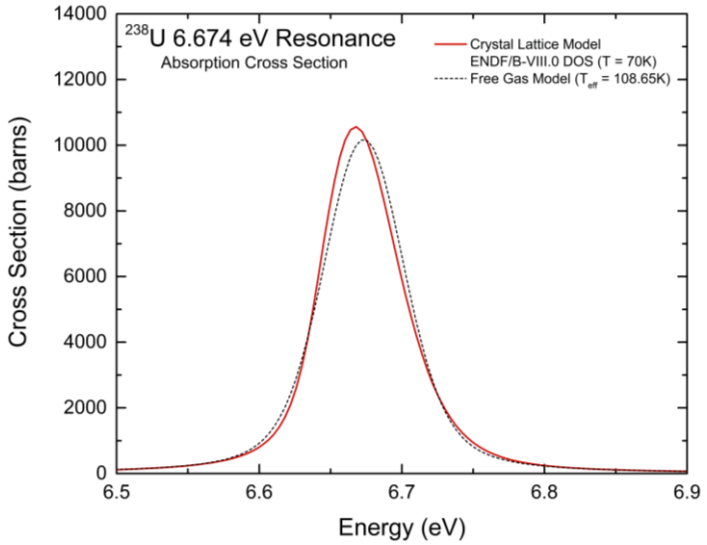
TIP4P/2005f parameter	Value (units)
ε	0.7749 (kJ/mol)
σ	0.31644 (nm)
q_H	0.5564 (e)
q_M	-1.1128 (e)
r_{OM}	0.13194 (Å)
r_{eq}	0.93 (Å)
D_r	432.581 (kJ/mol)
β	22.87 (nm^{-1})
K_θ	367.81 (kJ/mol*rad²)
θ_{eq}	107.4 (°)

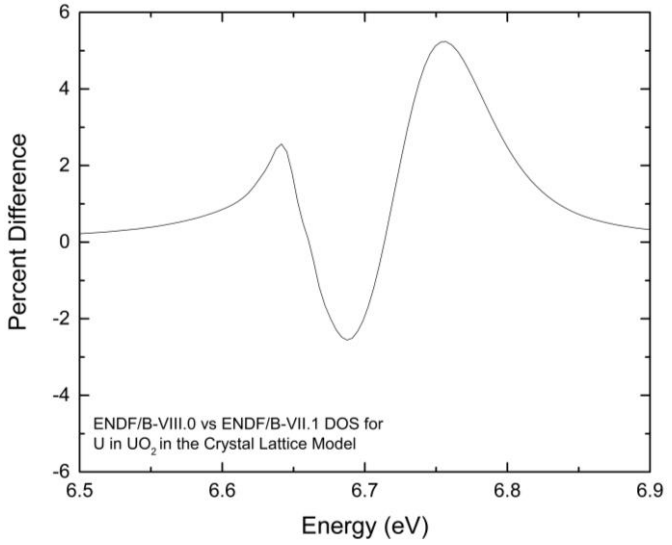


Doppler Broadening – FY 2018



- Density of States for Uranium in UO_2
  - Comparison of the DOS for the current ENDF/B-VIII.0 with the historical ENDF/B-VII.1
- Doppler Broadening of Absorption Resonances
  - Using the ENDF/B-VIII.0 DOS and the effective temperature calculated using the same DOS, the resulting Doppler broadened absorption resonances at 70K are displayed.
  - Differences in the resulting Doppler broadened cross sections using the crystal lattice model with the current and historical ENDF/B DOS are on the order of 5% (~200 barns).





Summary

- 10 new TSL evaluations contributed to ENDF/VIII
  - 5 first-of-a-kind evaluations
- Modern predictive methods for thermal neutron cross section calculations based on the use of atomistic simulations
  - Ab initio lattice dynamics
  - Molecular dynamics (ab initio and classical)
    - New materials
    - All states of matter (solid, liquid, gas)
    - Imperfect structure
- *FLASSH* is a new thermal scattering analysis platform that uses a generalized theoretical approach for TSL calculations.
  - Removed approximations such as incoherent approximation, cubic approximation, atom site approximation and SCT approximation.
- Progress on FY 2018 tasks