Uranium Carbide and Uranium-Metal Thermal Scattering Law Evaluations and Cross Sections

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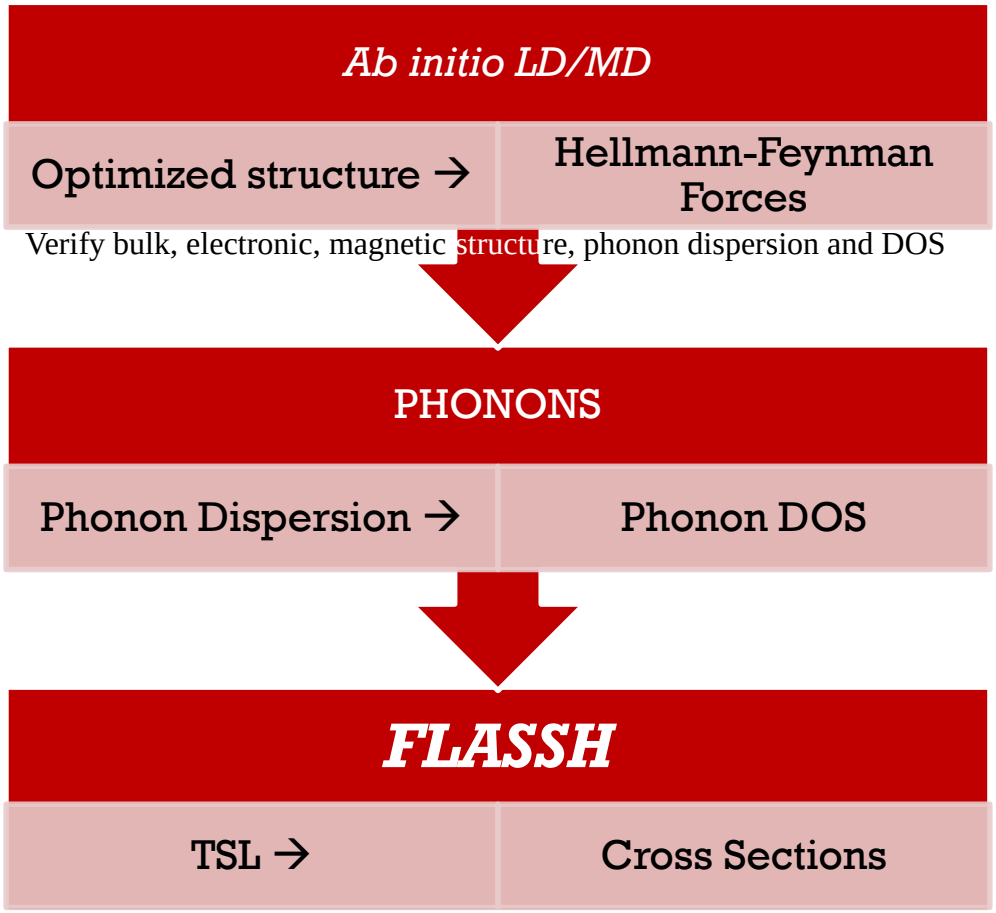
Brookhaven National Lab, NY, USA

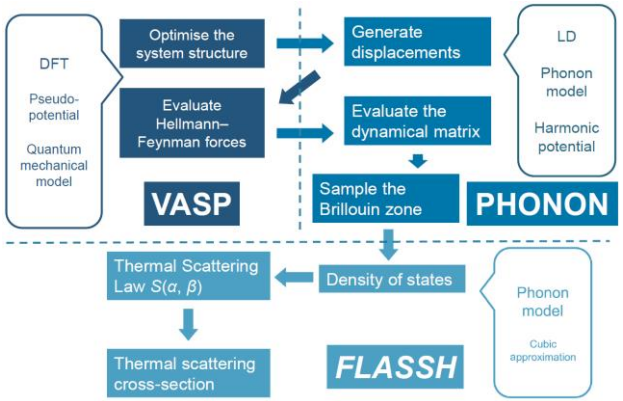
Motivation

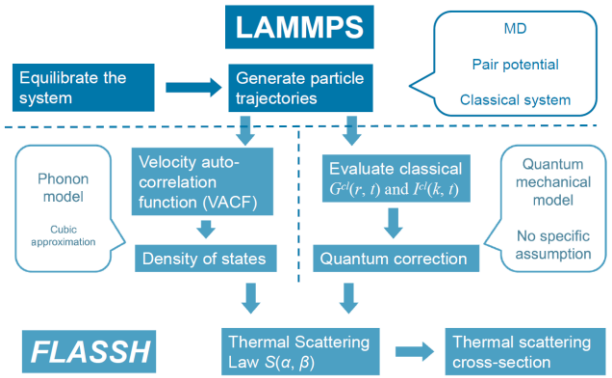
- **TSL describes scattering system: impacts neutron thermalization and may inform resonance broadening**
  - **TSL impacts thermal cross sections, thermal neutron spectrum, criticality safety, Doppler broadening**
- **Generate *ab initio* thermal scattering cross sections for advanced reactor fuel materials: *Uranium Carbide and Uranium Metal***
  - **UC and U-Metal have high thermal conductivities and fissile isotope densities**
  - **UC considered for thermal nuclear propulsion, HTGR TRISO fuel particles, fast breeder fuel**

Method

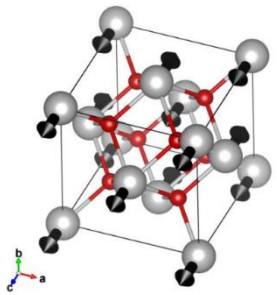


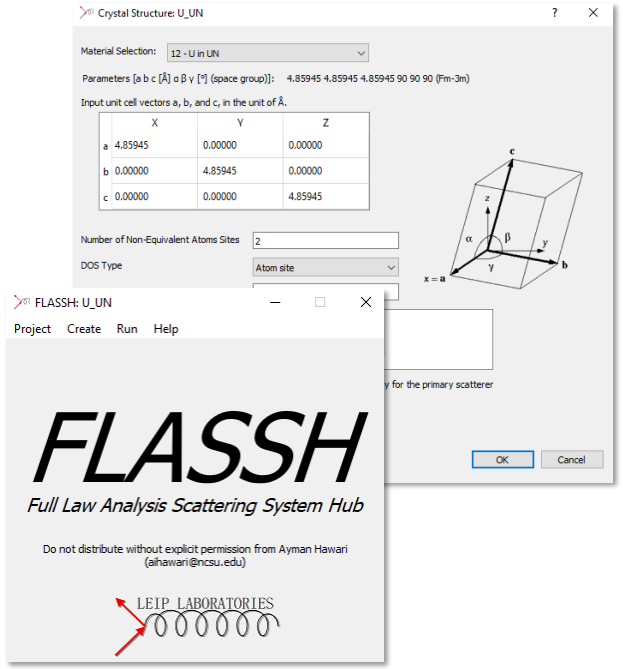
TSL Methodology



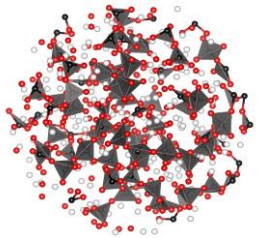


DFT/LD approach





MD approach

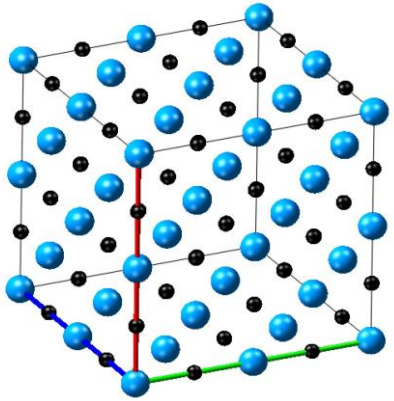


Uranium Carbide AILD

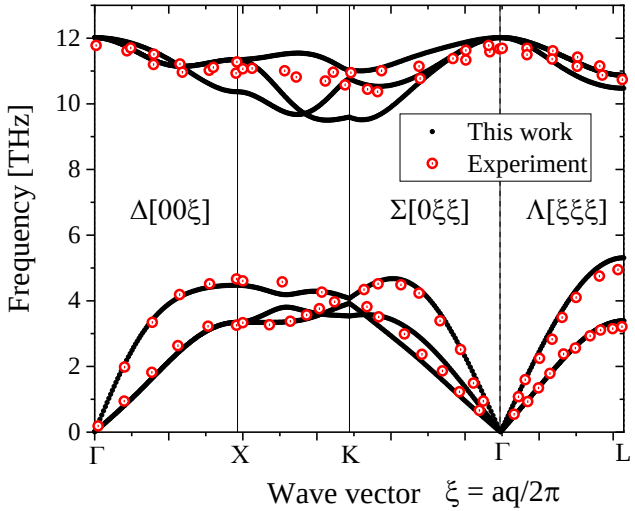
- **VASP DFT Framework**
  - **GGA-PBEsol Pseudopotential**
  - **1.9 eV Effective Hubbard Term**
  - **Magnetic structure captured with Spin-Orbit-Coupling**
    - **AFM-I polarizations in x,y,z → PM**
  - **2x2x2 Supercell, 64 Atoms**
  - **.02 Å Displacements in ± x-direction for non-equivalent sites**

Lattice Constant	DFT	Experiment	Deviation (%)
a (Å)	4.9275	4.960	-0.654

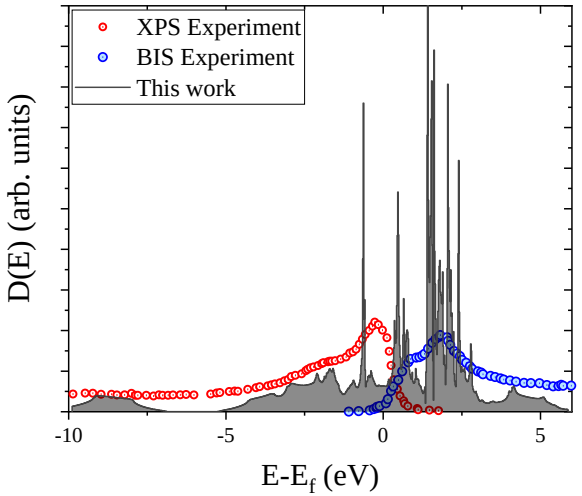
UC Supercell



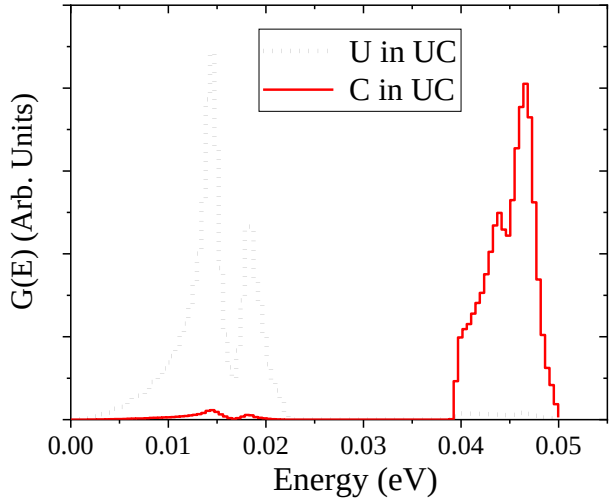
Phonon Dispersion



Electronic DOS

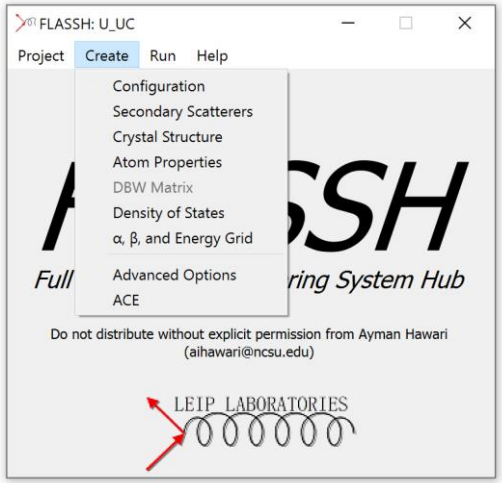


Phonon DOS



FLASSH


FLASSH: U_UC
Project Create Run Help
FLASSH
Full Law Analysis Scattering System Hub
Do not distribute without explicit permission from Ayman Hawari
(aihawari@ncsu.edu)
LEIP LABORATORIES


FLASSH: U_UC
Project Create Run Help
Configuration
Secondary Scatterers
Crystal Structure
Atom Properties
DBW Matrix
Density of States
α, β, and Energy Grid
Advanced Options
ACE
Do not distribute without explicit permission from Ayman Hawari
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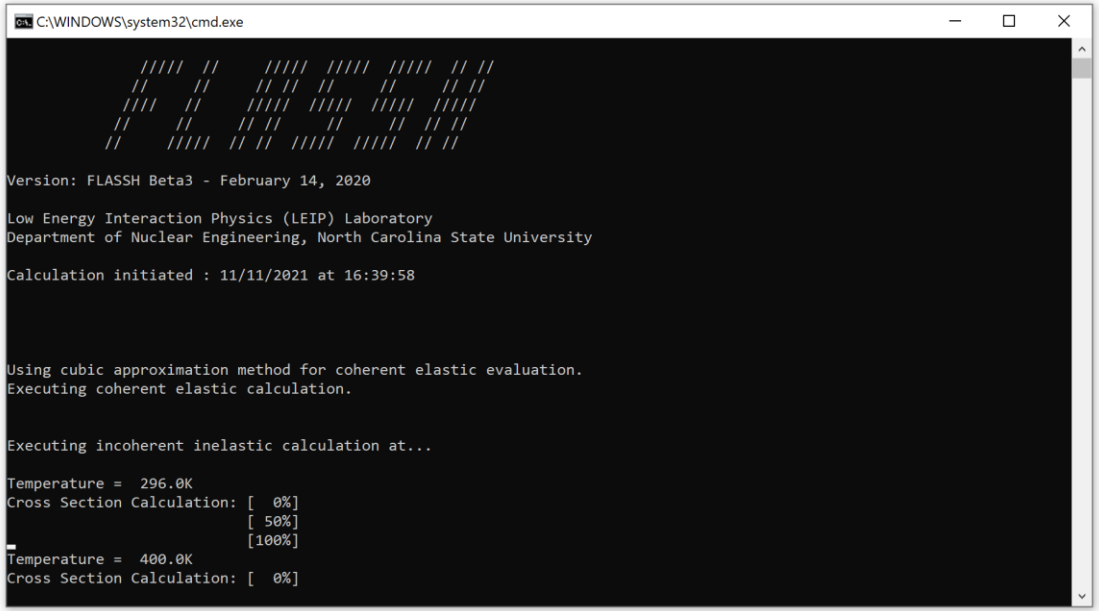
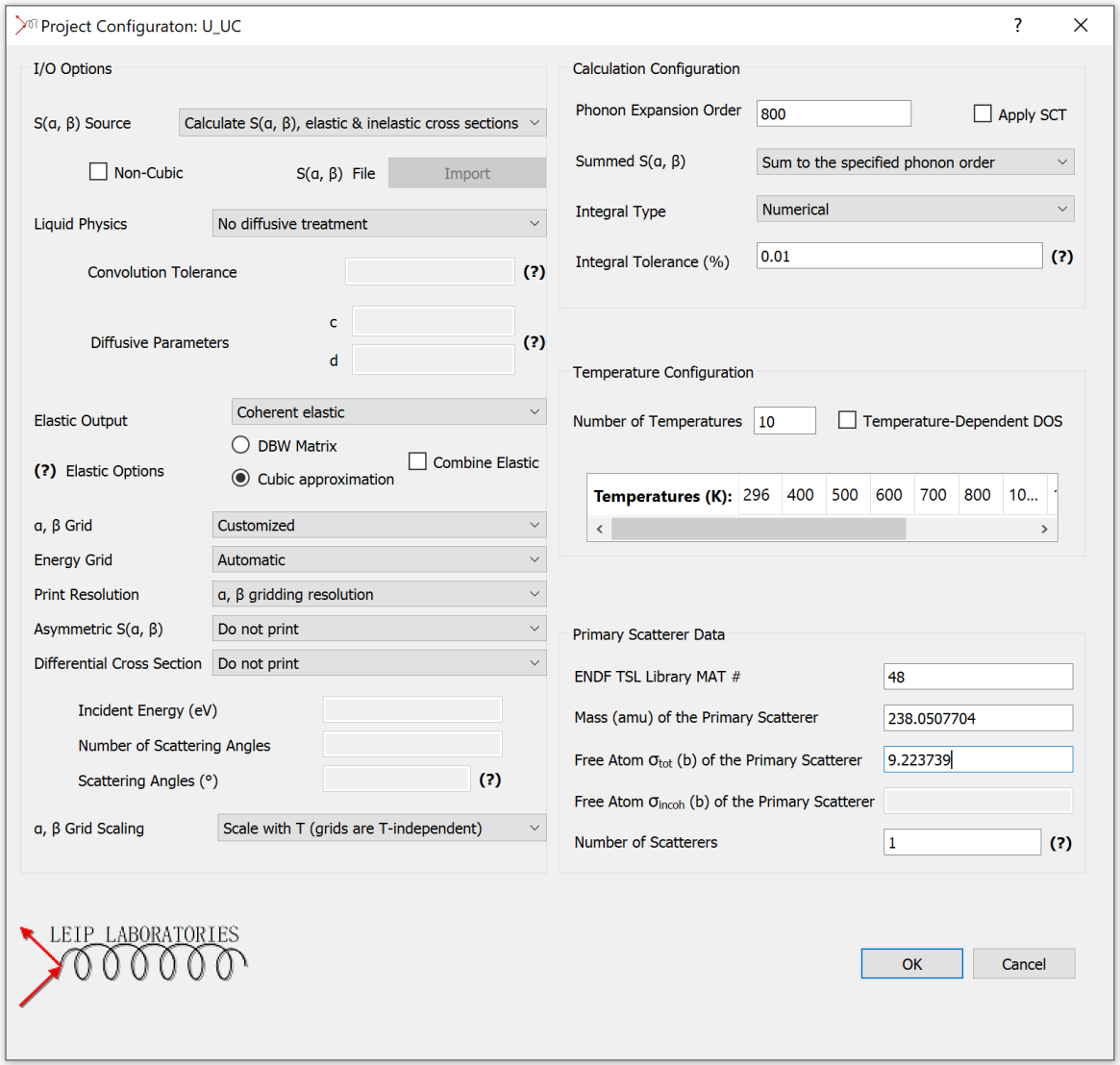
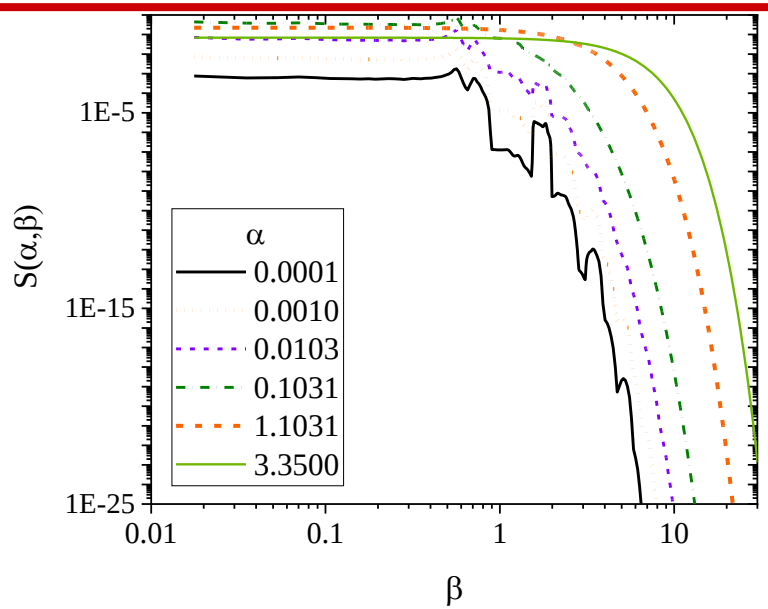
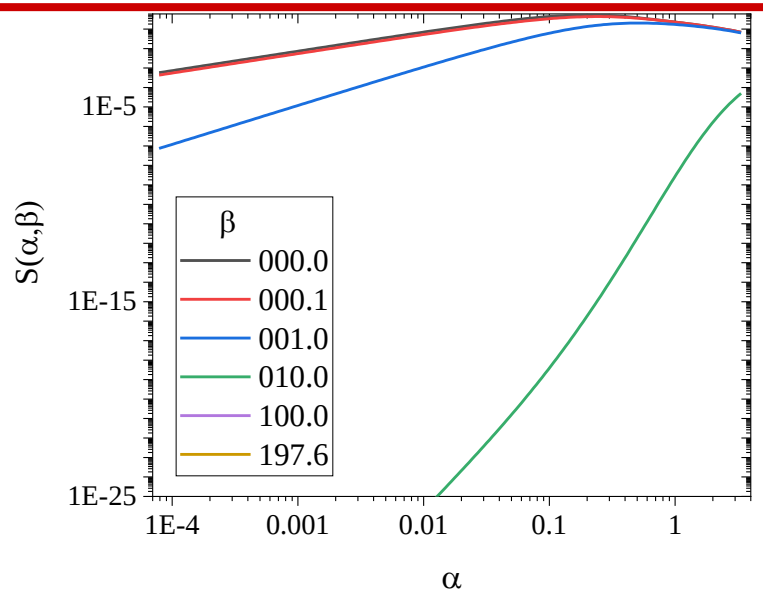

C:\WINDOWS\system32\cmd.exe
Version: FLASSH Beta3 - February 14, 2020
Low Energy Interaction Physics (LEIP) Laboratory
Department of Nuclear Engineering, North Carolina State University
Calculation initiated : 11/11/2021 at 16:39:58
Using cubic approximation method for coherent elastic evaluation.
Executing coherent elastic calculation.
Executing incoherent inelastic calculation at...
Temperature = 296.0K
Cross Section Calculation: [0%]
[50%]
[100%]
Temperature = 400.0K
Cross Section Calculation: [0%]


Project Configuraton: U_UC
I/O Options
S(α, β) Source
Calculate S(α, β), elastic & inelastic cross sections
Non-Cubic
S(α, β) File
Import
Liquid Physics
No diffusive treatment
Convolution Tolerance
Diffusive Parameters
c
d
Elastic Output
Coherent elastic
(?) Elastic Options
DBW Matrix
Cubic approximation
Combine Elastic
α, β Grid
Customized
Energy Grid
Automatic
Print Resolution
α, β gridding resolution
Asymmetric S(α, β)
Do not print
Differential Cross Section
Do not print
Incident Energy (eV)
Number of Scattering Angles
Scattering Angles (°)
α, β Grid Scaling
Scale with T (grids are T-independent)
Calculation Configuration
Phonon Expansion Order
800
Apply SCT
Summed S(α, β)
Sum to the specified phonon order
Integral Type
Numerical
Integral Tolerance (%)
0.01
Temperature Configuration
Number of Temperatures
10
Temperature-Dependent DOS
Temperatures (K): 296 400 500 600 700 800 10...
Primary Scatterer Data
ENDF TSL Library MAT #
48
Mass (amu) of the Primary Scatterer
238.0507704
Free Atom σtot (b) of the Primary Scatterer
9.223739
Free Atom σincoh (b) of the Primary Scatterer
Number of Scatterers
1
LEIP LABORATORIES
OK
Cancel

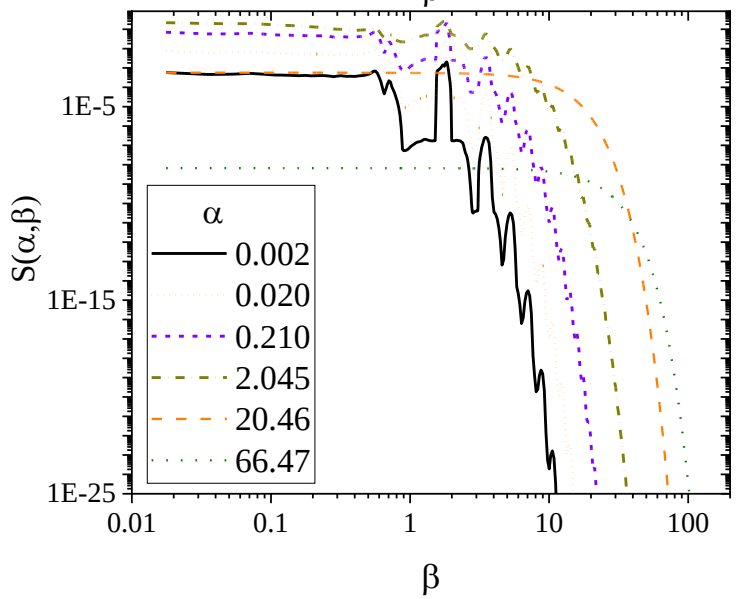
Uranium Carbide TSL at 296 K

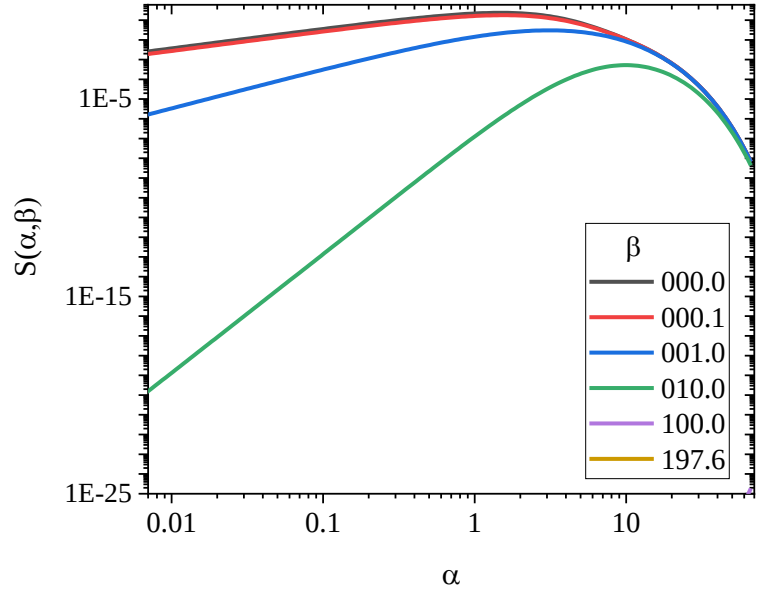
Uranium in UC





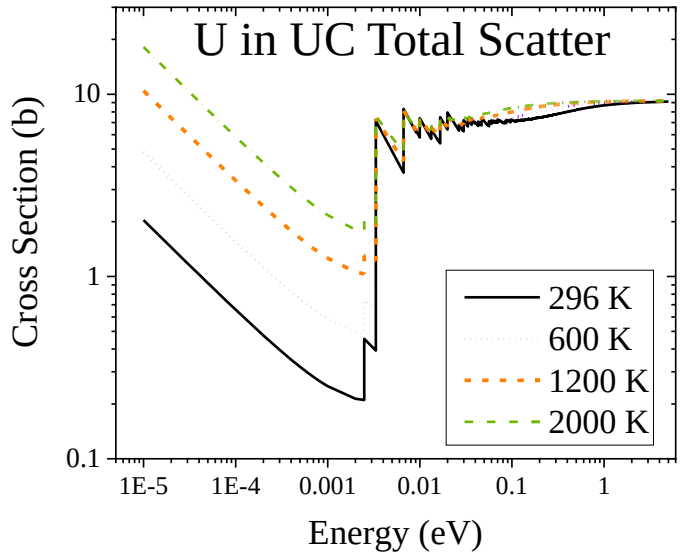
Carbon in UC

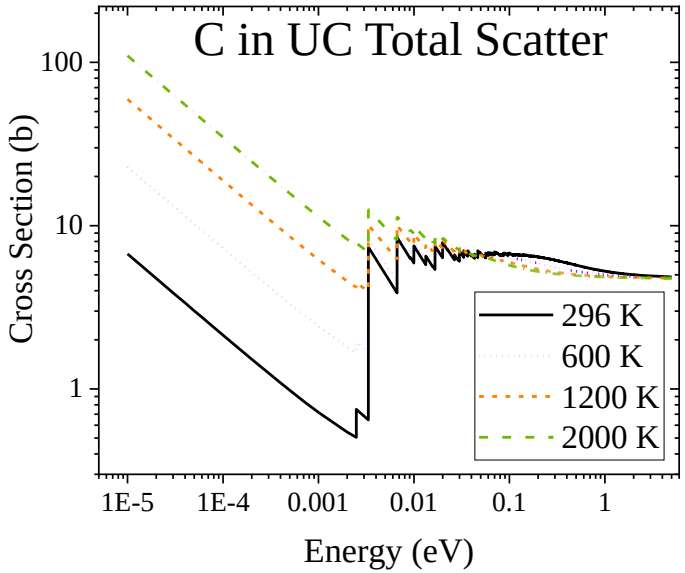


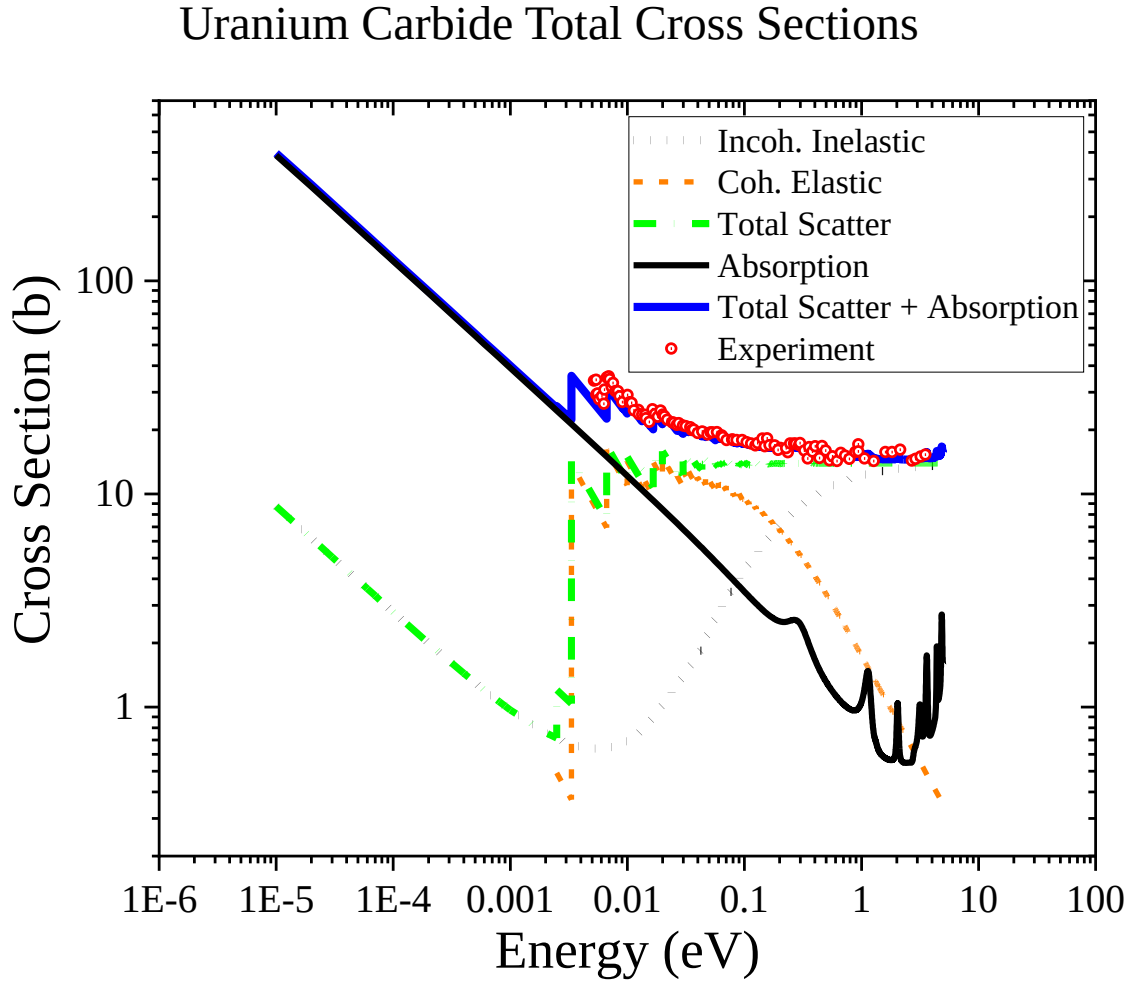


- **Evaluated for 296, 400, 500, 600, 700, 800, 1000, 1200, 1600, 2000 K**

Uranium Carbide Cross Sections







Uranium Carbide TSL

```
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 0.000000+0 0.000000+0          0          0          0          6  48 1451
 1.000000+0 5.000000+0          0          0         12          8  48 1451
 0.000000+0 0.000000+0          0          0         33          3  48 1451
 U in UC    LEIP LAB    EVAL-Jun21 J.P.W. Crozier, A.I. Hawari       48 1451
                        DIST-                                        48 1451
----ENDF/B-VIII         MATERIAL  48                                 48 1451
-----THERMAL NEUTRON SCATTERING                                      48 1451
------ENDF-6 FORMAT                                                 48 1451
                                                                     48 1451
  Temperatures = 296 400 500 600 700 800 1000 1200 1600 2000 K       48 1451
                                                                     48 1451
 HISTORY                                                             48 1451
 -------                                                             48 1451
 This library was produced by the Low Energy Interaction Physics     48 1451
 (LEIP) group at North Carolina State University, USA. The           48 1451
 thermal scattering law data for Uranium in Uranium Carbide was      48 1451
 developed using ab initio lattice dynamics (AILD)[1,2]. Ten         48 1451
 temperatures are available in this library. The Full Law            48 1451
 Analysis Scattering System Hub (FLASSH) system was used to          48 1451
 produce File 7 MT = 2, 4 data for Uranium in UC [3]. MAT=48 and     48 1451
 ZA=148 are used for U in UC.                                        48 1451
                                                                     48 1451
 REFERENCES                                                          48 1451
 ----------                                                          48 1451
 1. A.I. Hawari, "Modern Techniques in Inelastic Thermal Neutron     48 1451
    Scattering Analysis," Nucl. Data Sheets 118, 172 (2014).         48 1451
                                                                     48 1451
 2. J.P.W. Crozier, A.I. Hawari, "Generation of the Thermal          48 1451
    Scattering Law of Uranium Carbide with Ab Initio Lattice         48 1451
    Dynamics," Trans. American Nuclear Society, Vol. 124, (2021).    48 1451
                                                                     48 1451
 3. Y. Zhu, A.I. Hawari, "Full Law Analysis Scattering System Hub    48 1451
    (FLASSH)," PHYSOR 2018: Reactor Physics Paving the Way           48 1451
    Towards More Efficient Systems, Cancun, Mexico, 2018.            48 1451
                                                                     48 1451
 ------------------------------------------------------------------  48 1451
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                           7          2       1672          0        48 1451
                           7          4     134411          0        48 1451
```

```
                                                                     48 0  0
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 2.960000+2 0.000000+0          9          0          1        903  48 7  2
        903          1                                              48 7  2
 2.493869-3 6.121195-4 3.325158-3 2.306956-2 6.650316-3 5.374672-2  48 7  2
 9.144185-3 5.484663-2 9.975474-3 7.097774-2 1.330063-2 8.110025-2  48 7  2
 1.579450-2 8.204143-2 1.662579-2 1.170336-1 1.995095-2 1.479011-1  48 7  2
 2.244482-2 1.490659-1 2.660126-2 1.615509-1 2.909513-2 1.632259-1  48 7  2
 2.992642-2 1.916711-1 3.325158-2 2.125411-1 3.574545-2 2.133564-1  48 7  2
 3.657674-2 2.325951-1 3.990190-2 2.385320-1 4.239577-2 2.401312-1  48 7  2
 4.322705-2 2.566788-1 4.655221-2 2.875218-1 4.904608-2 2.898836-1  48 7  2
 5.320253-2 2.932578-1 5.569640-2 2.940342-1 5.652769-2 3.193687-1  48 7  2
 5.985284-2 3.372345-1 6.234671-2 3.390219-1 6.317800-2 3.502394-1  48 7  2
 6.650316-2 3.608198-1 6.899703-2 3.630867-1 6.982832-2 3.830729-1  48 7  2
 7.315348-2 3.925230-1 7.564735-2 3.940126-1 7.980379-2 4.024914-1  48 7  2
 8.229766-2 4.046918-1 8.312895-2 4.147460-1 8.645411-2 4.376572-1  48 7  2
 8.894798-2 4.398216-1 8.977927-2 4.494981-1 9.559829-2 4.509159-1  48 7  2
 9.642958-2 4.706223-1 9.975474-2 4.831343-1 1.022486-1 4.845257-1  48 7  2
 1.064051-1 4.873684-1 1.088989-1 4.907779-1 1.097302-1 5.016272-1  48 7  2
```

```
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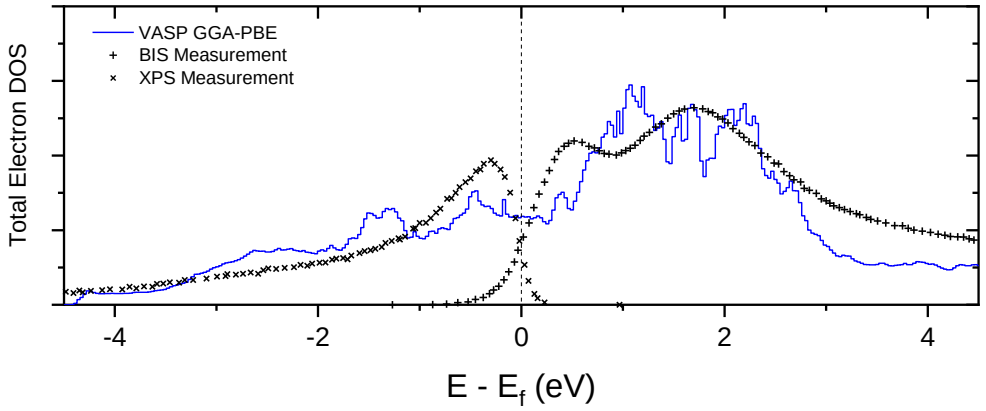
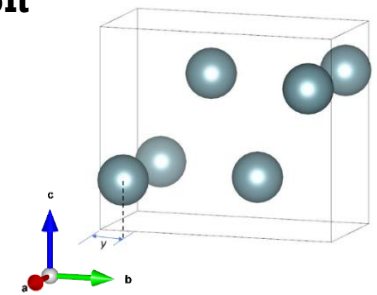
 9.223739+0 1.976285+2 2.360058+2 5.000000+0 0.000000+0 1.000000+0  48 7  4
 0.000000+0 0.000000+0          0          0          1        350  48 7  4
        350          4                                              48 7  4
 2.960000+2 0.000000+0          9          0          1        200  48 7  4
        200          4                                              48 7  4
 7.880988-5 5.781342-4 8.314555-5 6.099267-4 8.771974-5 6.434669-4  48 7  4
 9.254557-5 6.788505-4 9.763689-5 7.161790-4 1.030083-4 7.555589-4  48 7  4
 1.086752-4 7.971031-4 1.146539-4 8.409303-4 1.209615-4 8.871658-4  48 7  4
 1.276161-4 9.359418-4 1.346368-4 9.873977-4 1.420438-4 1.041681-3  48 7  4
 1.498582-4 1.098945-3 1.581025-4 1.159356-3 1.668004-4 1.223084-3  48 7  4
 1.759768-4 1.290313-3 1.856581-4 1.361233-3 1.958719-4 1.436048-3  48 7  4
 2.066477-4 1.514971-3 2.180162-4 1.598226-3 2.300102-4 1.686051-3  48 7  4
 2.426640-4 1.778697-3 2.560140-4 1.876427-3 2.700984-4 1.979519-3  48 7  4
 2.849577-4 2.088268-3 3.006344-4 2.202982-3 3.171736-4 2.323987-3  48 7  4
```

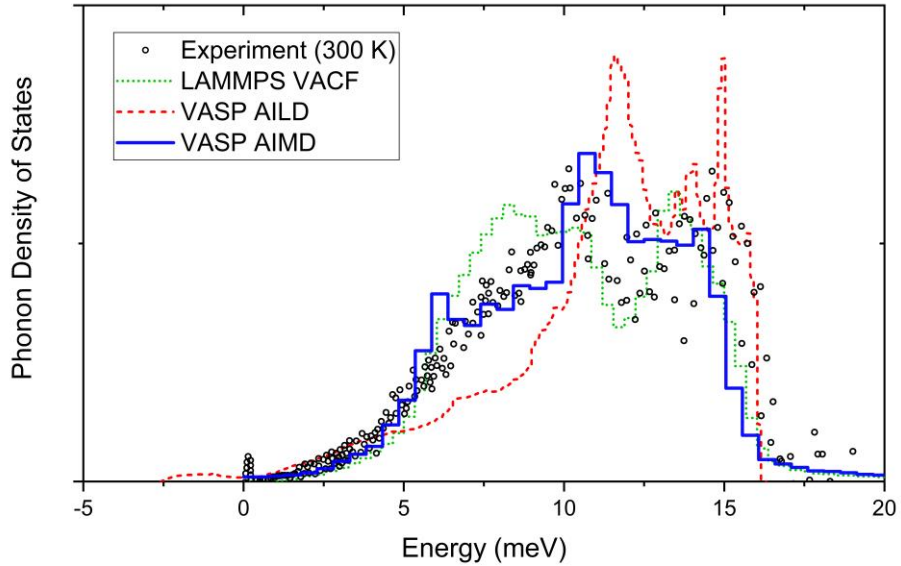
Uranium Metal AIMD

- ***Ab initio* Molecular Dynamics**
  - ***Ab initio* required to capture accurate electronic structure**
  - **MD required to capture temperature dependence**

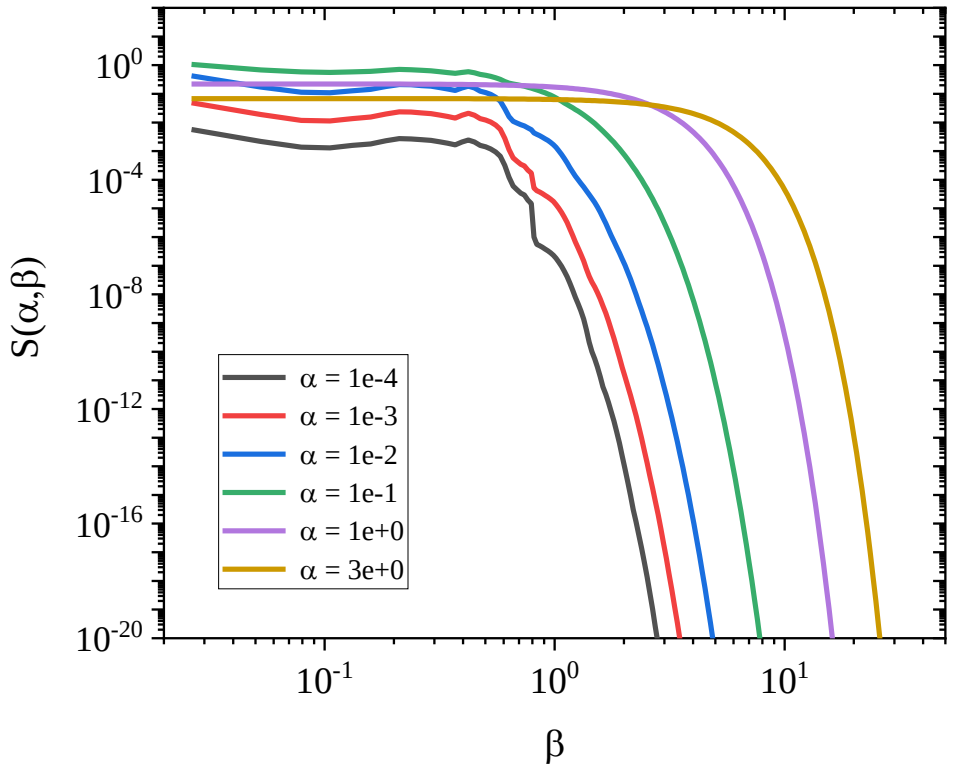


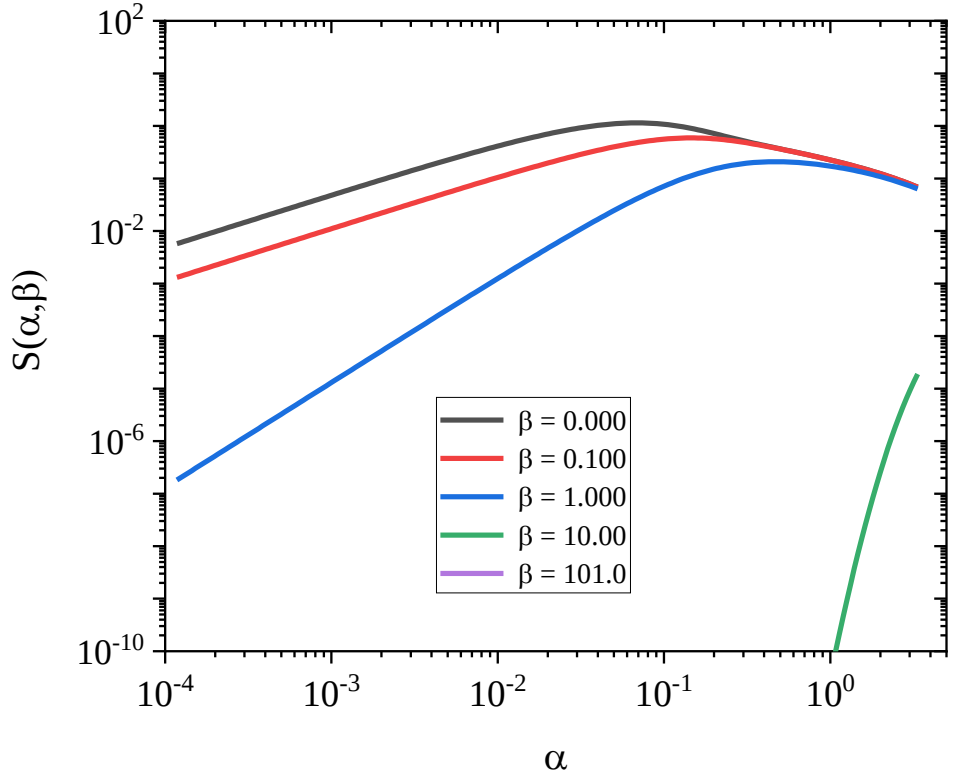
- **VASP Molecular Dynamics Inputs**
  - **GGA-PBE pseudopotential**
  - **No Hubbard corrections or spin-orbit coupling**
  - **5x2x3 Supercell, 120 Atoms**
  - **0.05 fs timestep**
  - **296 K**
  - **3 ps – equilibration**
  - **10 ps – VACF calculation**





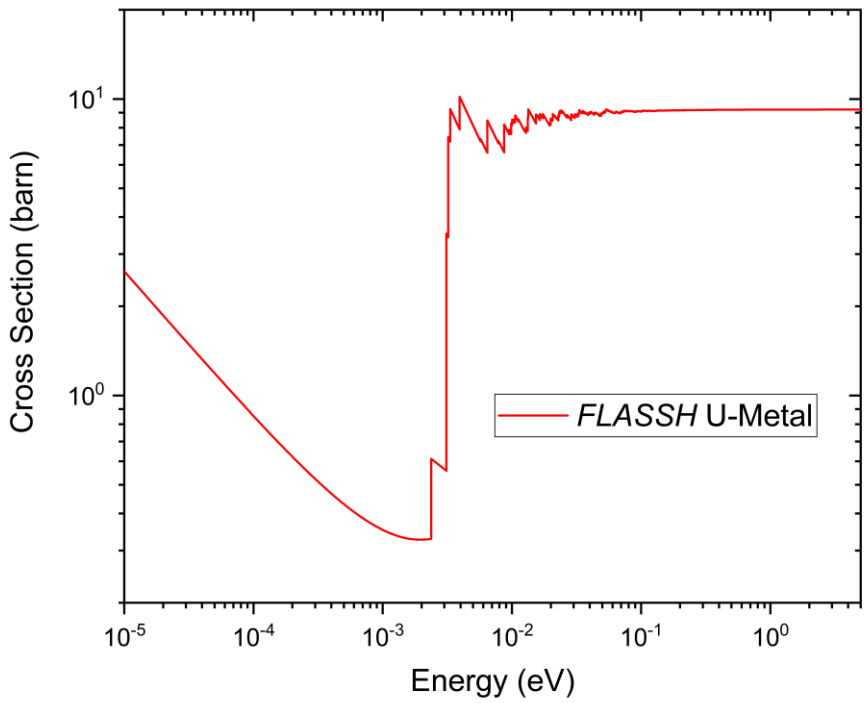
Uranium Metal TSL at 296 K



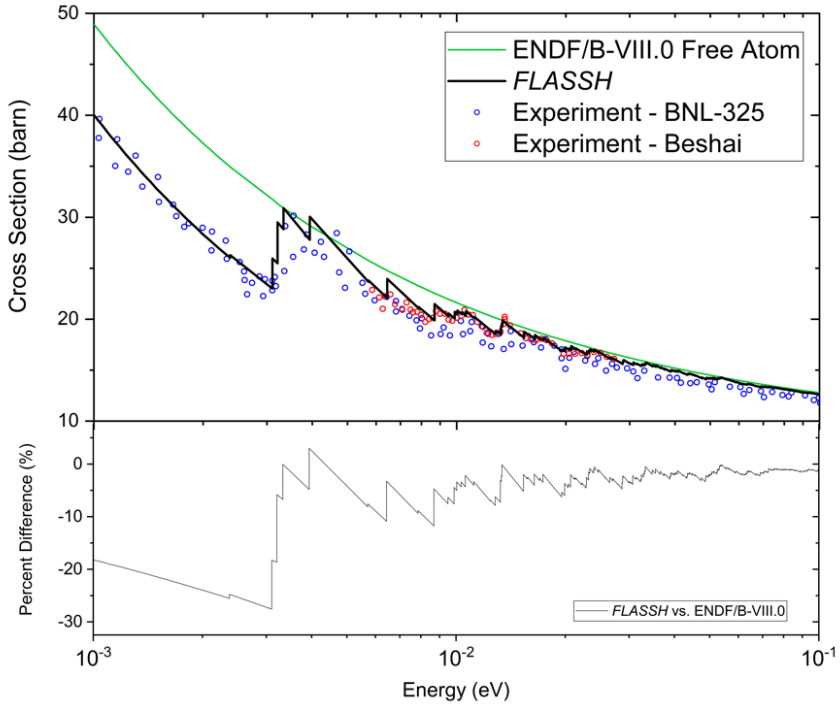


- **Evaluated for 296, 400, 500, 600, 700, 800, and 900 K**
- **α-phase stable up to ~668°C**
- **Room temperature traditionally used in cross section measurements**

Uranium Metal Cross Sections



Total Scattering (300 K)



Total Cross Section (300 K)

Uranium-Metal TSL

```
                                                                   1 0  0
 1.730000+2 2.360058+2          -1          0          0          0  73 1451
 0.000000+0 0.000000+0           0          0          0          6  73 1451
 1.000000+0 5.000000+0           0          0         12          8  73 1451
 0.000000+0 0.000000+0           0          0         28          3  73 1451
 U-metal    LEIP LAB    EVAL-JULY21 N.C. Fleming, A.I. Hawari        73 1451
                        DIST-                                        73 1451
----ENDF/B-VIII         MATERIAL  73                                 73 1451
-----THERMAL NEUTRON SCATTERING                                      73 1451
------ENDF-6 FORMAT                                                 73 1451
                                                                     73 1451
  Temperatures = 296  400  500  600  700  800  900 K                 73 1451
                                                                     73 1451
 HISTORY                                                             73 1451
 -------                                                             73 1451
 This library was produced by the Low Energy Interaction Physics     73 1451
 (LEIP) group at North Carolina State University, USA. The           73 1451
 thermal scattering law data for uranium metal was developed         73 1451
 using ab initio molecular dynamics (AIMD). The coherent elastic     73 1451
 cross sections were calculated based on the lattice constants       73 1451
 obtained from the MD structure. Seven temperatures are available    73 1451
 in this library. The Full Law Analysis Scattering System Hub        73 1451
 (FLASSH) system was used to produce File 7 MT = 2, 4 data for       73 1451
 U-metal [1]. The coherent elastic data were prepared using the      73 1451
 cubic approximation. MAT=73 and ZA=173 are used for U-metal.        73 1451
                                                                     73 1451
                                                                     73 1451
 REFERENCES                                                          73 1451
 ----------                                                          73 1451
 1. N. C. Fleming et al., "FLASSH 1.0: Full Law Analysis             73 1451
    Scattering System Hub," Trans. Am. Nucl. Soc., 125 (2021).       73 1451
                                                                     73 1451
 -------------------------------------------------------------------- 73 1451
                               1        451         35          0  73 1451
                               7          2       9576          0  73 1451
                               7          4     103240          0  73 1451
                                                                  73 1  0
```

```
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 2.960000+2 0.000000+0          6          0          1       7172  73 7  2
       7172          1                                              73 7  2
 2.410606-3 5.433518-4 3.152133-3 9.722178-3 3.256306-3 2.318624-2  73 7  2
 3.382800-3 3.023454-2 3.997833-3 3.984172-2 5.793406-3 4.051481-2  73 7  2
 6.534933-3 5.275715-2 7.973345-3 5.407972-2 8.819045-3 6.794808-2  73 7  2
 9.642424-3 7.088838-2 1.002191-2 7.796448-2 1.019793-2 8.170507-2  73 7  2
 1.048812-2 8.347478-2 1.076343-2 8.887317-2 1.135614-2 9.100141-2  73 7  2
 1.260853-2 9.142183-2 1.302522-2 9.628016-2 1.345423-2 1.080015-1  73 7  2
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 1.594181-2 1.273243-1 1.599133-2 1.280412-1 1.668333-2 1.348245-1  73 7  2
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 0.000000+0 0.000000+0          0          0          6          0  73 7  4
 9.223739+0 1.976285+2 2.360058+2 5.000000+0 0.000000+0 1.000000+0  73 7  4
 0.000000+0 0.000000+0          0          0          1        370  73 7  4
       370          4                                              73 7  4
 2.960000+2 0.000000+0          6          0          1        200  73 7  4
       200          4                                              73 7  4
 1.840107-5 4.729665-5 1.955581-5 5.026687-5 2.078303-5 5.342375-5  73 7  4
 2.208725-5 5.677905-5 2.347332-5 6.034527-5 2.494638-5 6.413568-5  73 7  4
 2.651187-5 6.816441-5 2.817560-5 7.244647-5 2.994374-5 7.699782-5  73 7  4
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Summary

- New Uranium Carbide and Uranium Metal evaluations using the *FLASSH* platform
  - ***Ab initio* LD/MD models–** converged electronic, magnetic and dynamical properties demonstrate good agreement with experimental data
  - **Thermal scattering cross sections–** generate inelastic and elastic thermal neutron scattering libraries in *FLASSH*
  - **New TSL evaluations–** production of ENDF File 7 data for Uranium Carbide and Uranium Metal submitted to NNDC for ENDF/B-VIII.1 release

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Contributed TSL Evaluations 2020-2021

	Material	Motivation	Theory	Validation
1	**FLiBe (beyllium)**	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	MD *FLASSH*	Ongoing against IRPhEP benchmark
2	**FLiBe (flourine)**			
3	**FLiBe (lithium)**			
4	**Liquid hydrogen fluoride (hydrogen)**	NCSP applications No TSL data in ENDF/B-VIII.0	MD *FLASSH*	Ongoing total cross section
5	**Heavy oil (hydrogen)**	NCSP/NR applications No TSL data in ENDF/B-VIII.0	MD *FLASSH*	Ongoing total cross section
6	**Sapphire (Al in Al2O3)**	Neutron science / Research Reactors No TSL data in ENDF/B-VIII.0 (cryogenic temperatures)	DFT/LD *FLASSH*	Total cross section
7	**Sapphire (O in Al2O3)**			
8	**Polyethylene (hydrogen, cryogenic temperatures)**	NCSP applications – update Extended ENDF/B-VIII.0 to cryogenic temperature	MD *FLASSH*	Total cross section & benchmarks
9	**Nuclear graphite (20% porosity)**	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	MD *FLASSH*	Various measurements & Benchmarks
10	**Graphite (crystalline with Sd)**	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	TSL measurements
11	**Beryllium (crystalline with Sd)**	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	TSL measurements
12	**Beryllium (with cryogenic temperatures)**	Neutron science / Research Reactors No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
13	**Uranium carbide (uranium)**	NCSP applications No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
14	**Uranium carbide (carbon)**	NCSP applications No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
15	**Silicon carbide (silicon)**	NCSP applications – update No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
16	**Silicon carbide (carbon)**	NCSP applications – update No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
17	**Uranium metal**	NCSP applications No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Total cross section
18	**Calcium Hydride (calcium)**	NCSP applications – update No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Ongoing
19	**Calcium Hydride (Hydrogen 1)**	NCSP applications – update No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Ongoing
20	**Calcium Hydride (Hydrogen 2)**	NCSP applications – update No TSL data in ENDF/B-VIII.0	DFT/LD *FLASSH*	Ongoing
21	**Amorphous carbon (carbon)**	DOE NE Advanced nuclear reactors No TSL data in ENDF/B-VIII.0	MD *FLASSH*	Ongoing

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Thank You