NNL Developed TSL Evaluations

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CSEWG – Evaluation Committee
December 1, 2020

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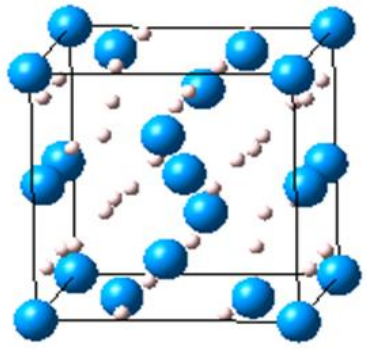
12 New and Revised TSL Evaluations

Material	Evaluation	MAT	Type	Temperatures	Eval. Code	Notes
UH_3	$H(UH_3)$	9	New	293.6	LEAPR	AILD, H incoherent approx.
YH_2	$Y(YH_2)$	55	Rev	293.6,400,500,600,700,800,1000,1200,1400,1600	FLASSH	AILD, Y coherent elastic
δ-ZrH_x	$H(ZrH_x)$	5	Rev	293.6,400,500,600,700,800,1000,1200	FLASSH	AIMD, H incoherent
	$Zr(ZrH_x)$	58	Rev	293.6,400,500,600,700,800,1000,1200	FLASSH	AILD. Zr coherent elastic
ε-ZrH_2	$H(ZrH_2)$	5	New	293.6,400,500,600,700,800,1000,1200	FLASSH	AIMD, H incoherent
	$Zr(ZrH_2)$	58	New	293.6,400,500,600,700,800,1000,1200	FLASSH	AILD, Zr coherent elastic,
Be_2C	$Be(Be_2C)$	28	New	293.6,400,500,600,700,800,1000,1200,1600,2000	FLASSH	AILD, Be coherent elastic
	$C(Be_2C)$	36	New	293.6,400,500,600,700,800,1000,1200,1600,2000	FLASSH	AILD, C coherent elastic
^7LiH	$H(^7LiH)$	4	New	293.6,400,500,600,700,800	FLASSH	AILD, mixed elastic scattering
	$^7Li(^7LiH)$	21	New	293.6,400,500,600,700,800	FLASSH	AILD, mixed elastic scattering
^7LiD	$D(^7LiD)$	15	New	293.6,400,500,600,700,800	FLASSH	AILD, mixed elastic scattering
	$^7Li(^7LiD)$	22	New	293.6,400,500,600,700,800	FLASSH	AILD, mixed elastic scattering

Also have ^7LiH and ^7LiD evaluations based on incoherent approximation

$H(UH_3)$



β-UH_3 Unit Cell

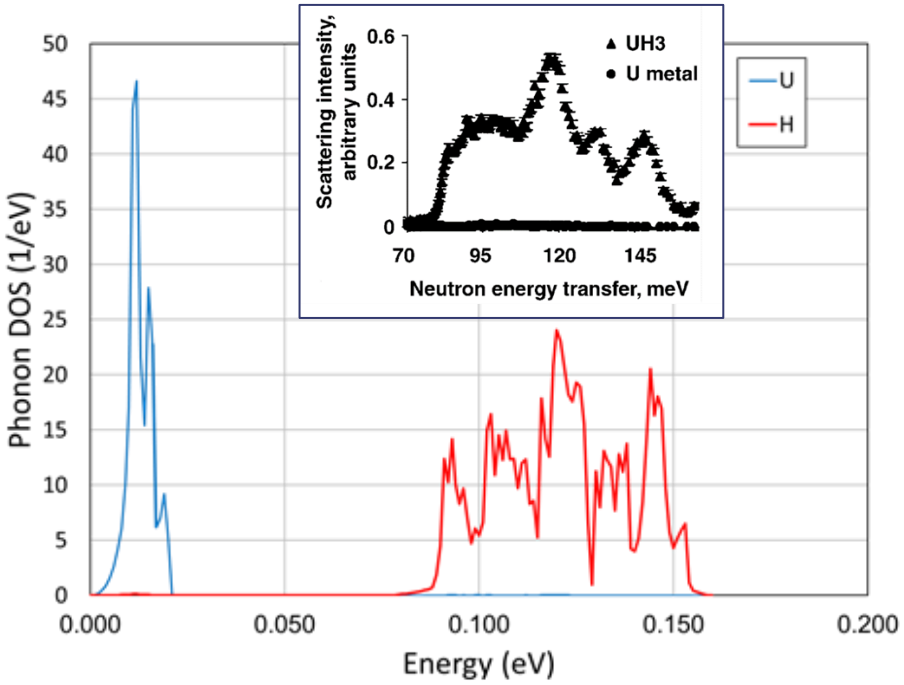
β-UH_3 stable at room temperature and above

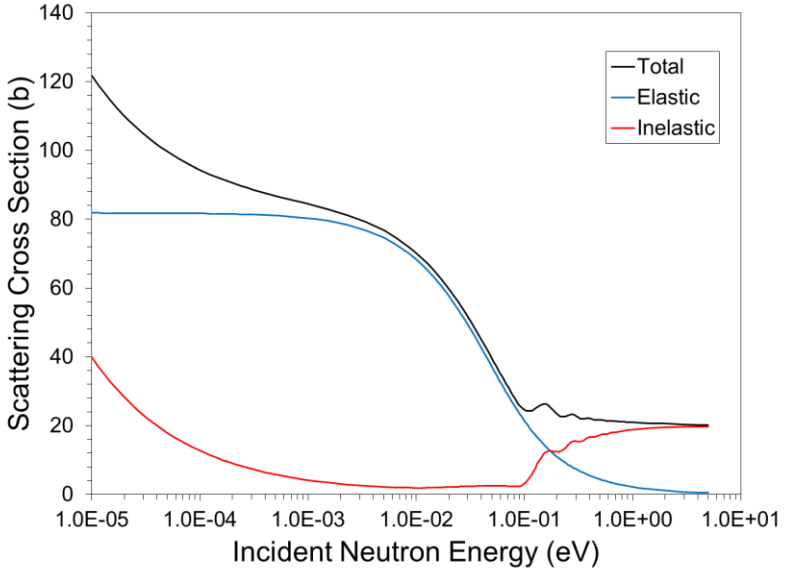
- 8 molecules per unit cell
- 6.643 Å lattice constant

PDOS calculated using VASP/PHONON

- Spin-polarized magnetism
- GGA+U to account for 5f electrons

Benchmark testing in progress





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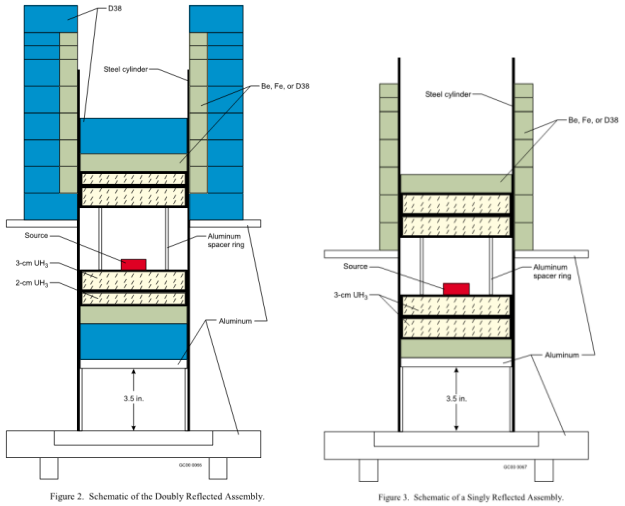
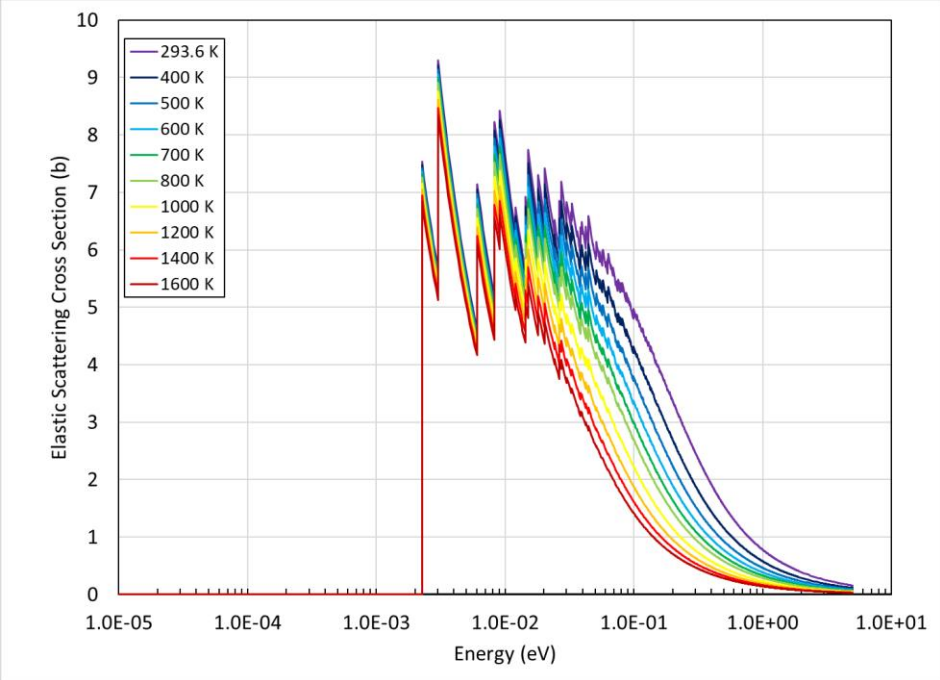


Figure 2. Schematic of the Doubly Reflected Assembly.

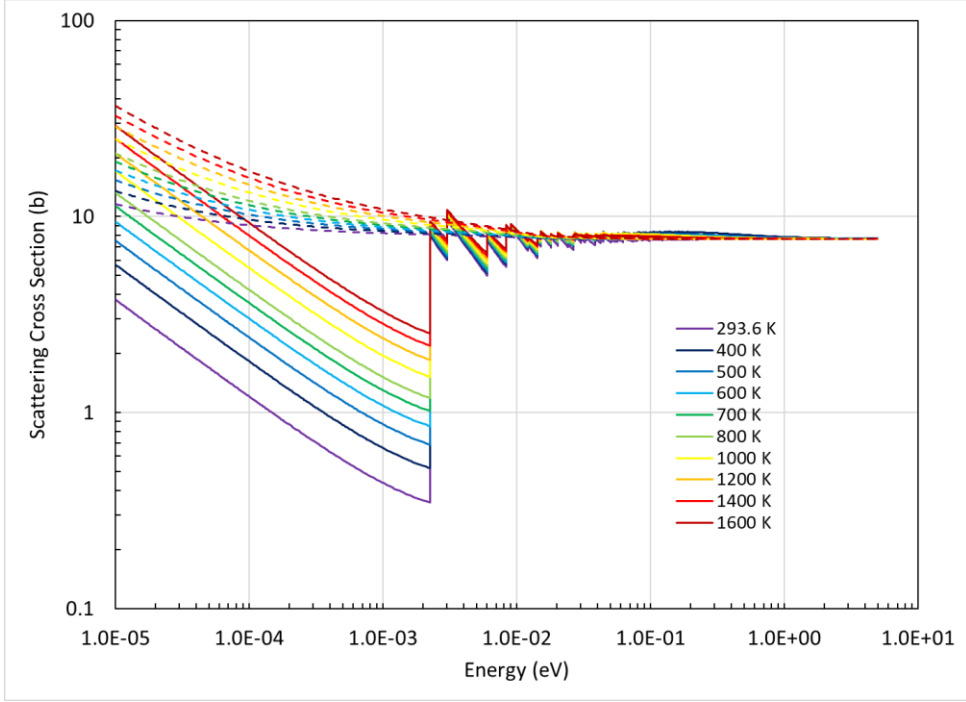
Figure 3. Schematic of a Singly Reflected Assembly.

$Y(YH_2)$ Revision

- Revision of NNL $Y(YH_2)$ TSL evaluation
- Same physical model for inelastic scattering
- Elastic scattering improved (relaxed incoherent approx.)
  - Bragg edges calculated using FLASSH general elastic scattering treatment based on YH_2 unit call structure
- Validation expected to be difficult since $H(YH_2)$ dominates
  - Might be able to see effect in modern neutron transmission measurements



Coherent elastic scattering cross sections for $Y(YH_2)$

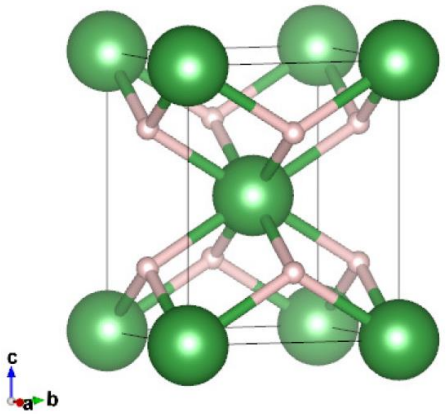


Comparison of revised (solid) and original (dotted) NNL $Y(YH_2)$ evalution total scattering cross sections showing effect of correctly treating coherent scattering

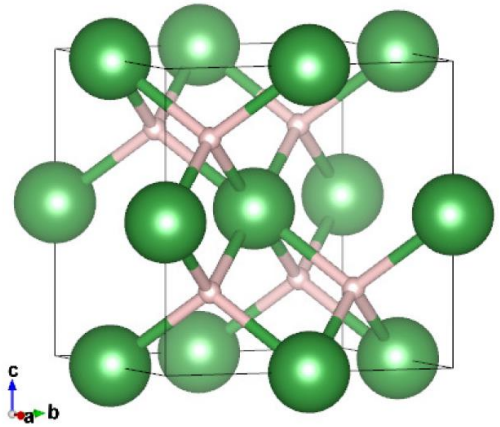
Zirconium Hydride Phonon Model

$Zr(ZrH_x)$

- Ab initio Lattice Dynamics (AILD)
  - Large mass and small vibrational displacement
  - Harmonic approximation reasonable
  - 2x2x2 δ-$ZrH_{1.5}$ supercell (80 atoms)
  - 3x3x2 ε-ZrH_2 supercell (108 atoms)
- VASP-PHONON
  - GGA-PBE
  - 450 eV plane wave cut-off
  - < 1.8 nm^{-1} k-point mesh



ε-phase



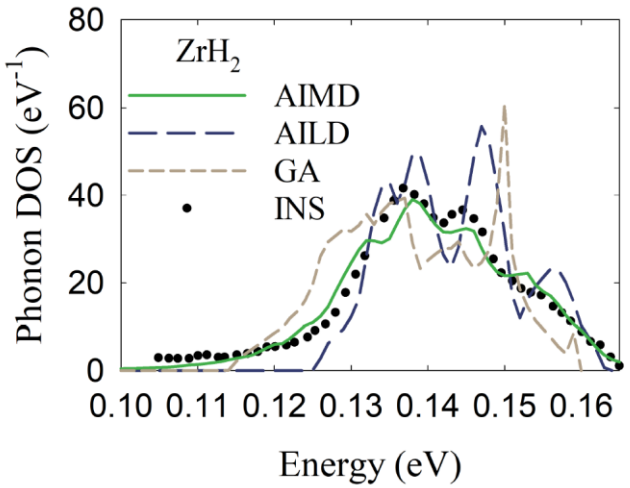
δ-phase

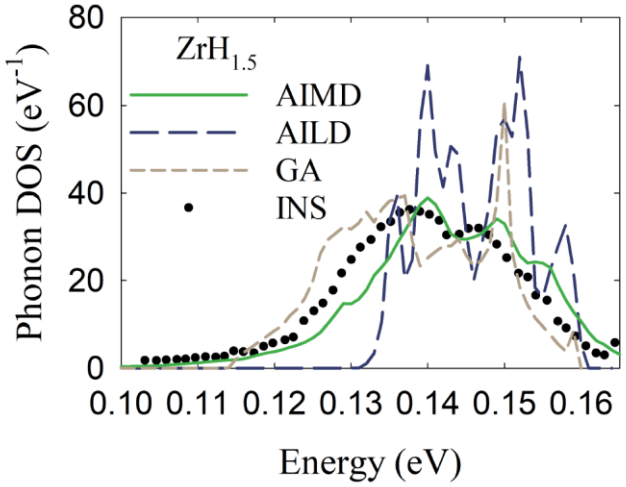
$H(ZrH_x)$

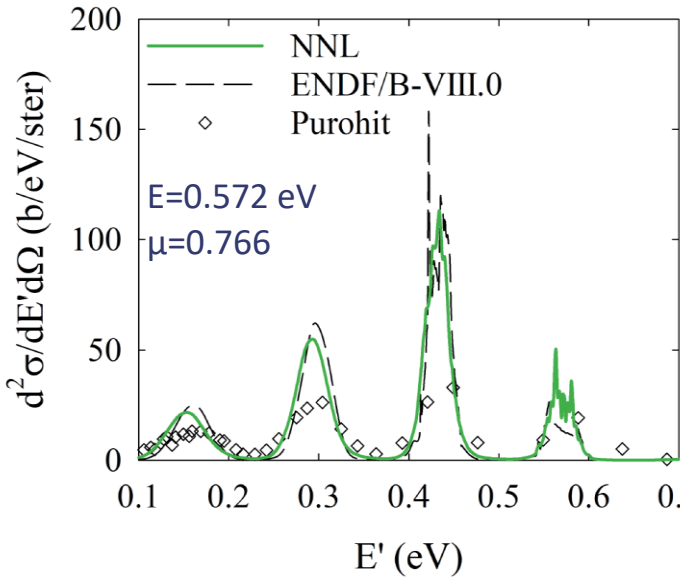
- Ab Initio Molecular Dynamics (AIMD)
  - Low H mass results in large vibrational displacements
  - Anharmonicity even at 0 K
  - 3x3x3 δ-$ZrH_{1.5}$ supercell (270 atoms)
  - 4x4x3 ε-ε-ZrH_2 supercell (288 atoms)
- VASP MD
  - GGA-PBE
  - 350 eV plane wave cut-off
  - < 0.6 nm^{-1} k-point mesh
  - 300 K with NVT ensemble
  - 0.00025 fs time steps
  - 1 ps equilibration + 1.5 ps simulation

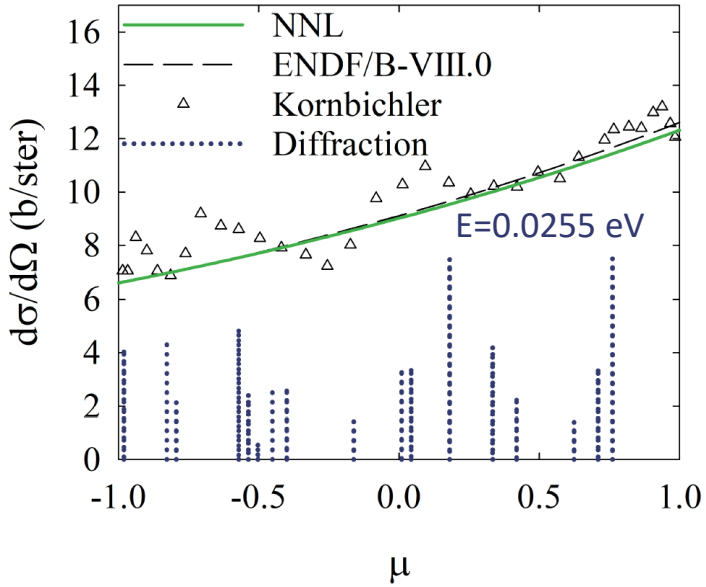
Zirconium Hydride Model Validation

- AIMD hydrogen phonons improved agreement compared to both
  - General Atomics (GA) model for current ENDF
  - AILD calculations
- Initial TSL validation performed through direct comparison with experiment for ε-phase
  - Current evaluations and ENDF/B-VIII.0
- Differential and double differential cross sections computed direct from FLASSH
- Current evaluations consistent with ENDF/B-VIII.0 and experiment in inelastic behavior
- New elastic treatment consistent with experiment
  - Bragg peaks for ZrH_2 model trend with behavior of measured differential cross section



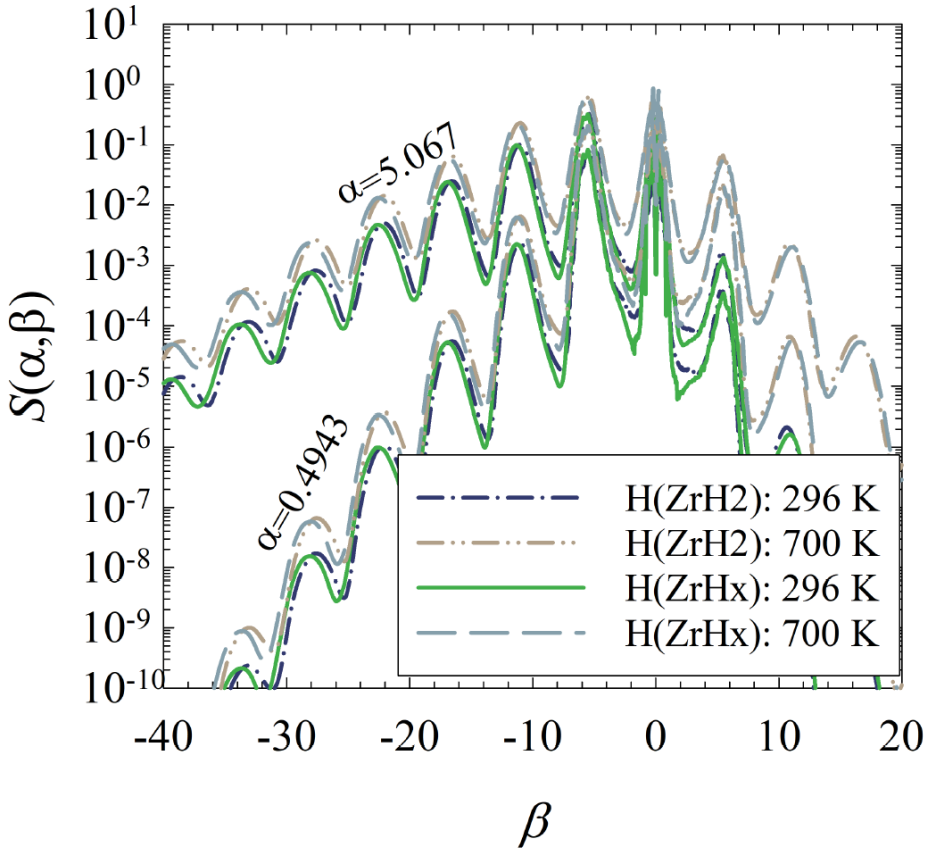


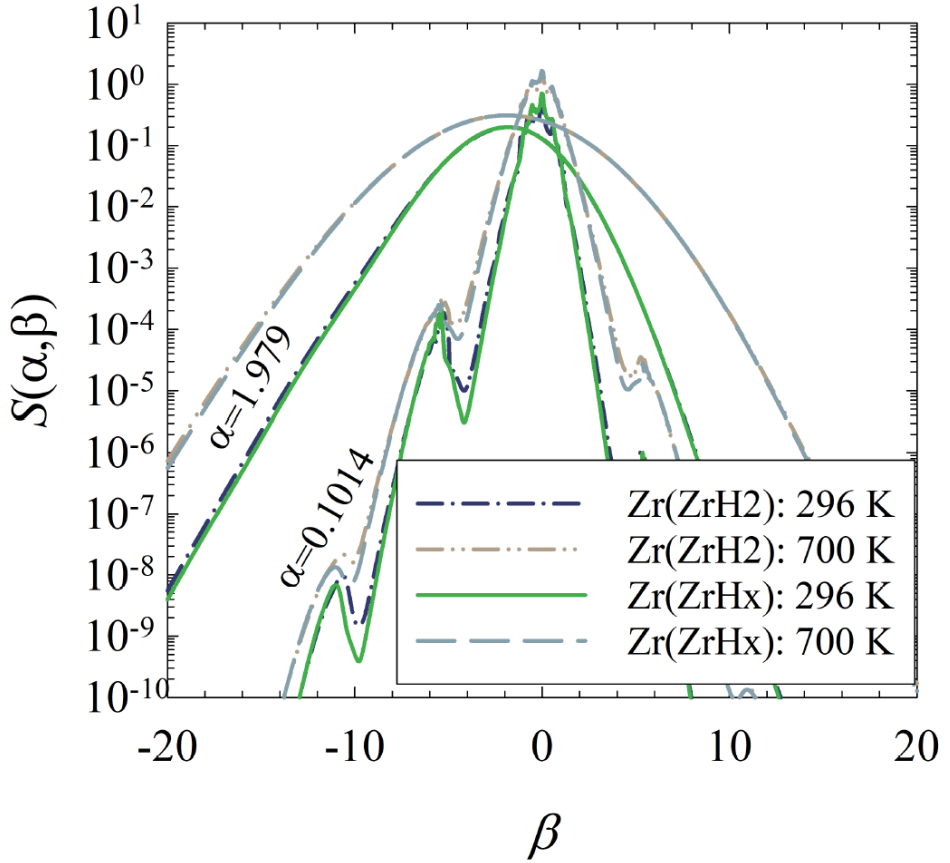




TSL for Different Phases

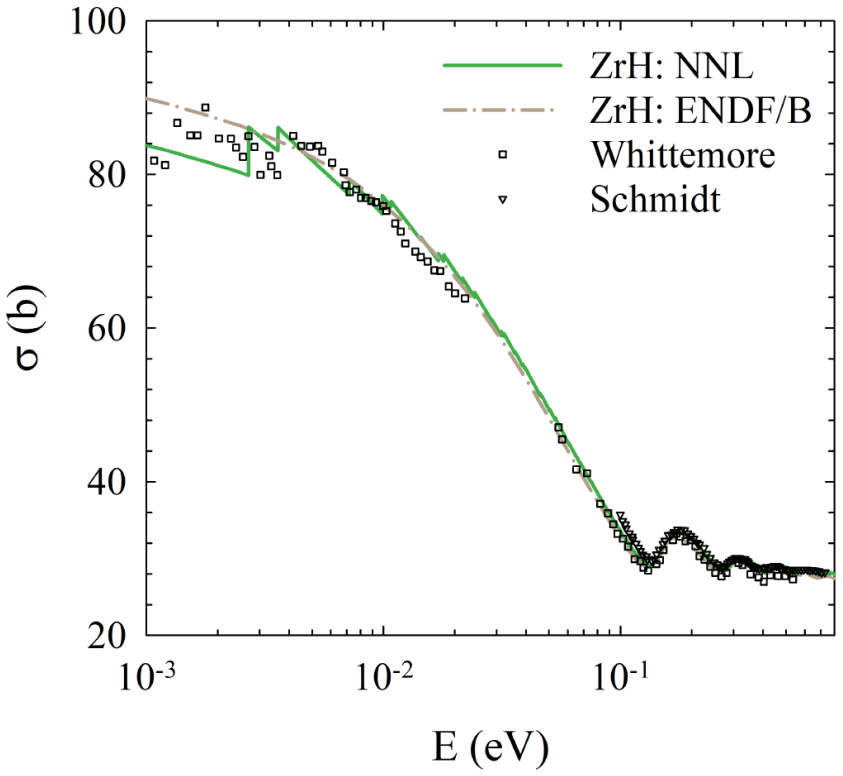
- TSLs for δ-phase and ε-phase are compared
- δ-H(ZrH_x) and ε-H(ZrH_2) both have strong quantum oscillator effect
- δ-phase diverges from ε-phase with increasing phonon order
  - Higher energy phonon spectra in δ-phase relative to ε-phase
- δ-Zr(ZrH_x) and ε-Zr(ZrH_2) are similar
  - Quantum oscillator effect is observed though small contributor

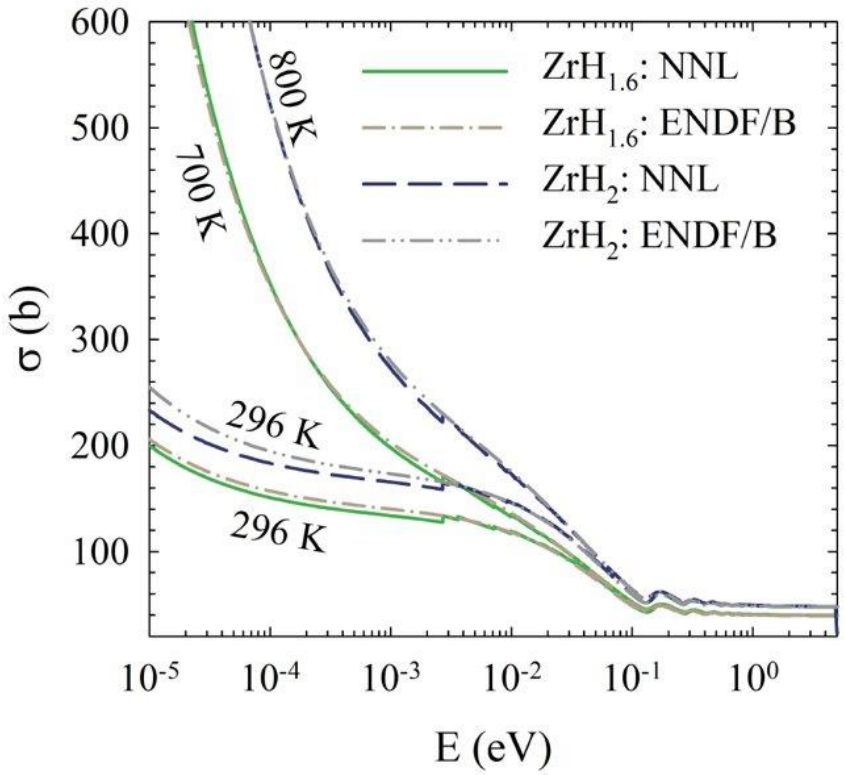




Scattering Cross Section Validation

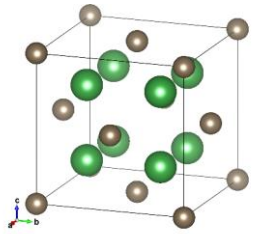
- Total Scattering Cross Section generated from TSL using NDEX
  - Adaptive energy mesh based on tabulated β-mesh captures oscillations to all phonon orders (ANE 149, 15 (2020) 107773)
- Coherent elastic cross section improve agreement with experiment compared to ENDF/B-VIII.0
- Consistent agreement in oscillations between current evaluation and ENDF/B-VIII.0
- NNL and ENDF/B: δ-ZrH_x parallel in 1/*v* region but ε-ZrH_2 diverge in 1/*v* region
  - Less pronounced acoustic phonons in ε-ZrH_2
- At high temperatures coherent elastic scattering persists but is reduced, optical phonon up-scattering dominant
  - Cross section of current evaluation and ENDF/B-VIII.0 nearly the same for both phases



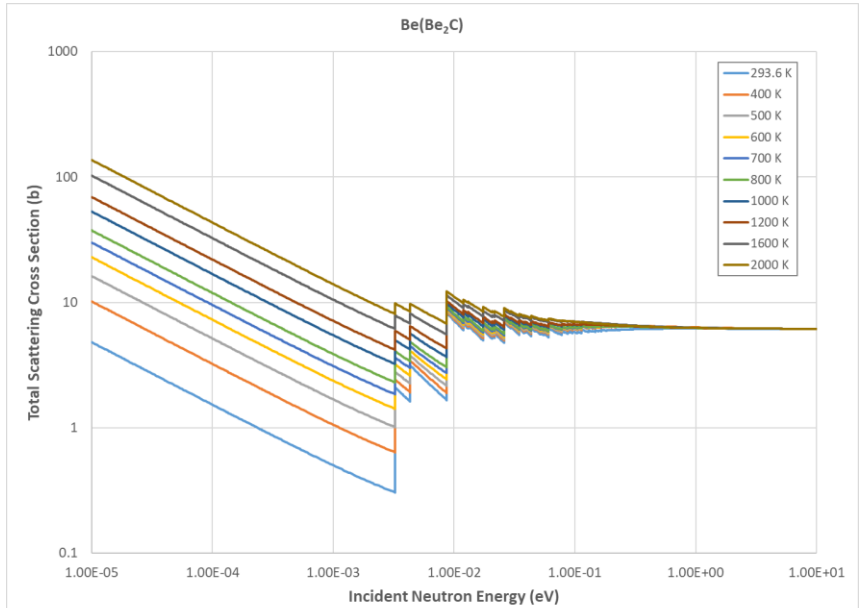


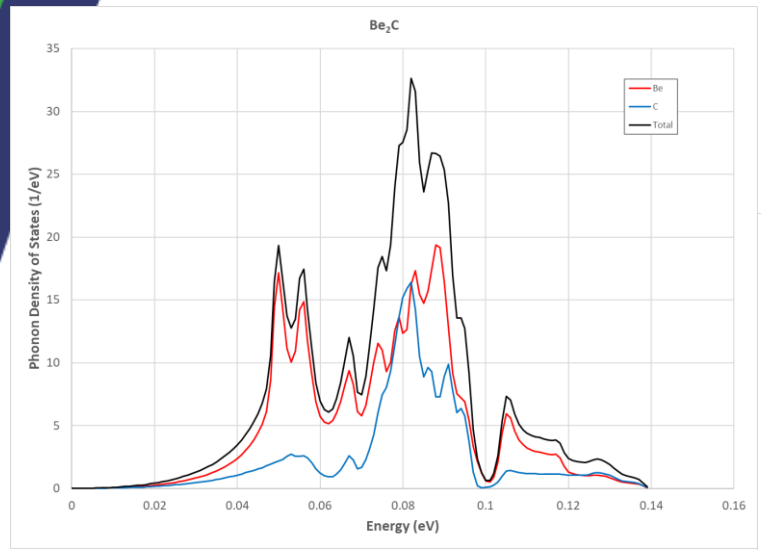
Be_2C

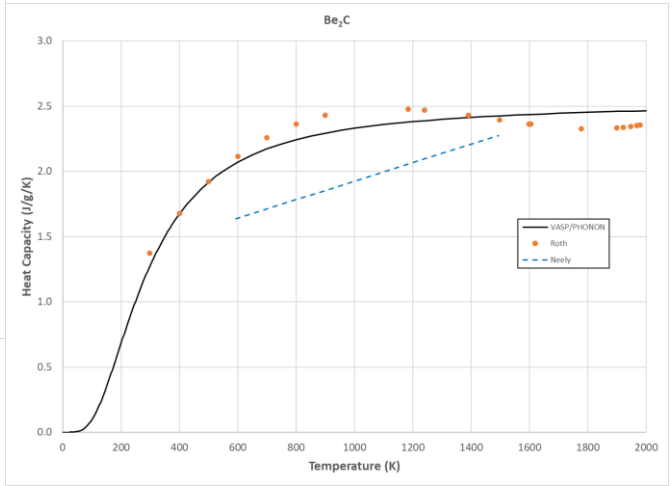
- PDOS calculated using AILD (VASP/PHONON)
  - 3x3x3 supercell, LO/TO splitting
- Heat capacity in reasonable agreement with SNL measurements, 1950s NEPA data suspect
- FLASSH used in order to capture coherent elastic scattering effects
- Be(Be_2C) and C(Be_2C) evaluated at 10 temperatures between 293.6 - 2000 K
- Transmission and INS measurements needed

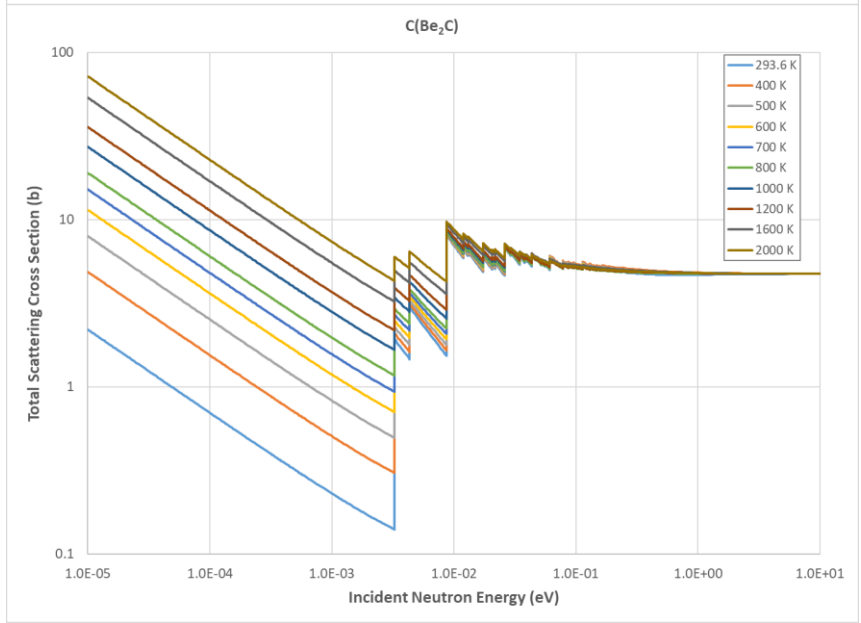


Be_2C unit cell (4.324 Å)



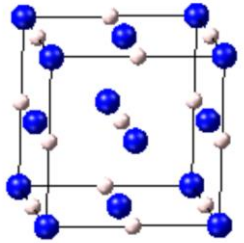




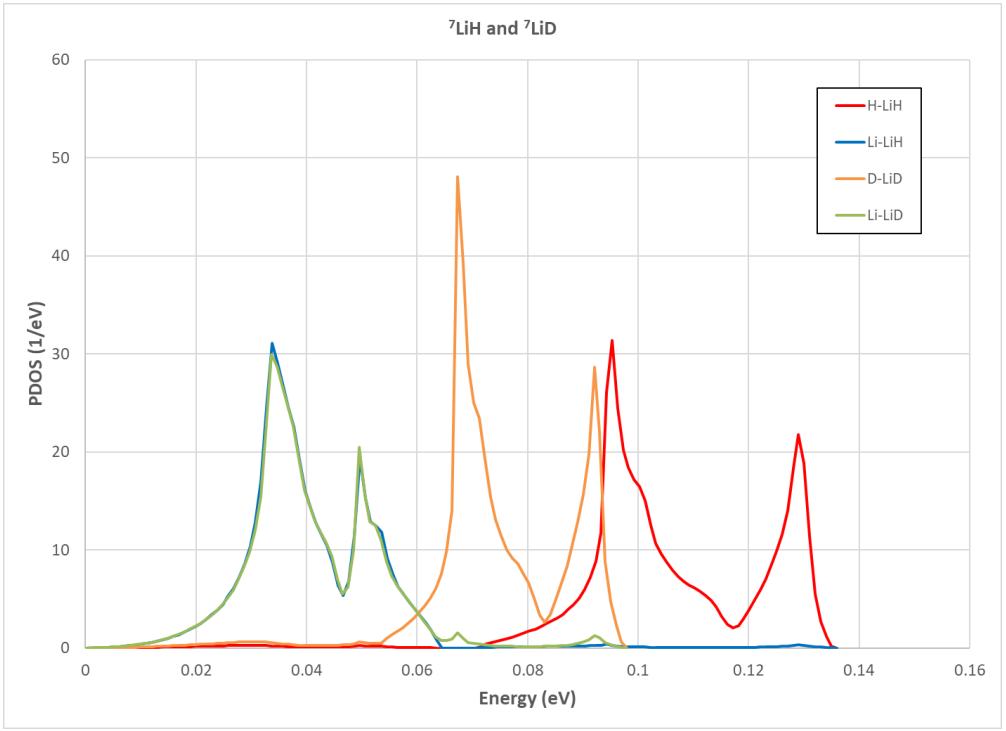


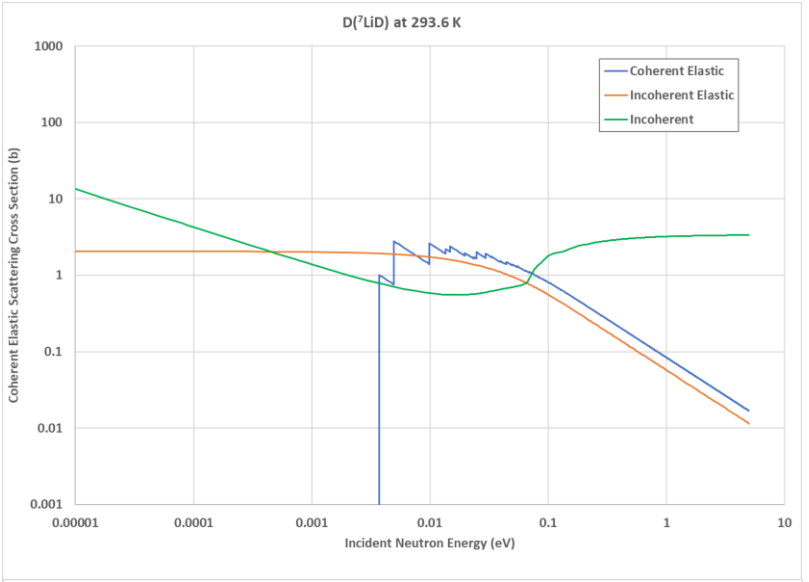
^{7}LiH and ^{7}LiD

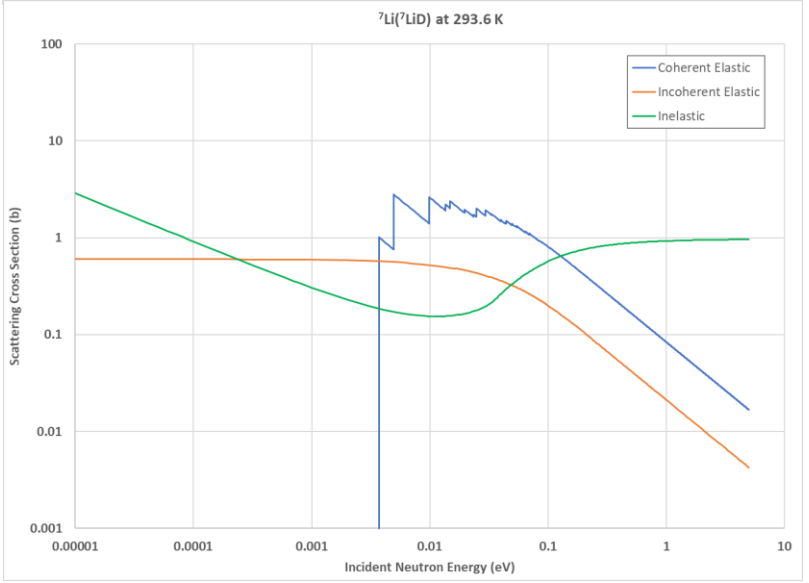
- Mixed elastic scattering treatment used to capture coherent and incoherent scattering effects in ^{7}Li and D
- AILD used to calculate PDOS
- TSL calculated at 6 temperature between 293.6 – 800 K using FLASSH
- Coherent elastic based on crystal structure and measured RT lattice constant by Zimmerman (*Phys. Rev. B*, **5**, 4704 (1972))



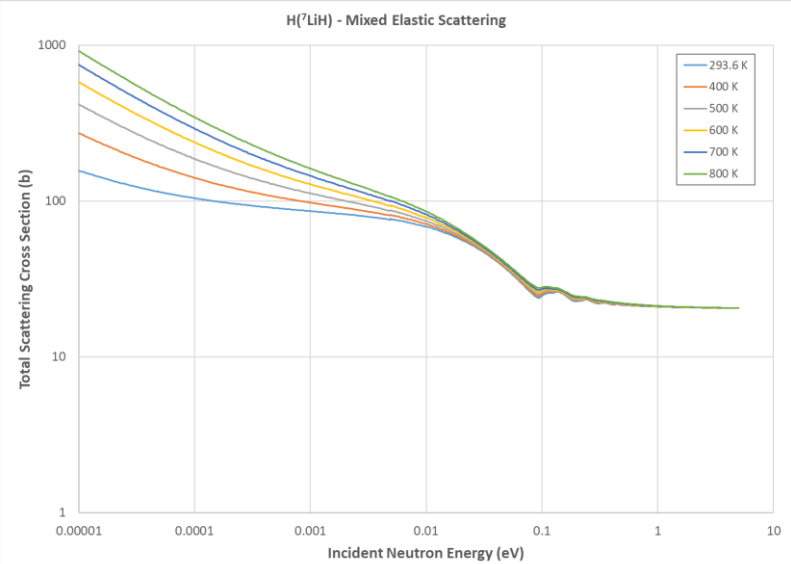
LiH unit cell (4.0831 Å for LiH, 4.0684 Å for LiD)

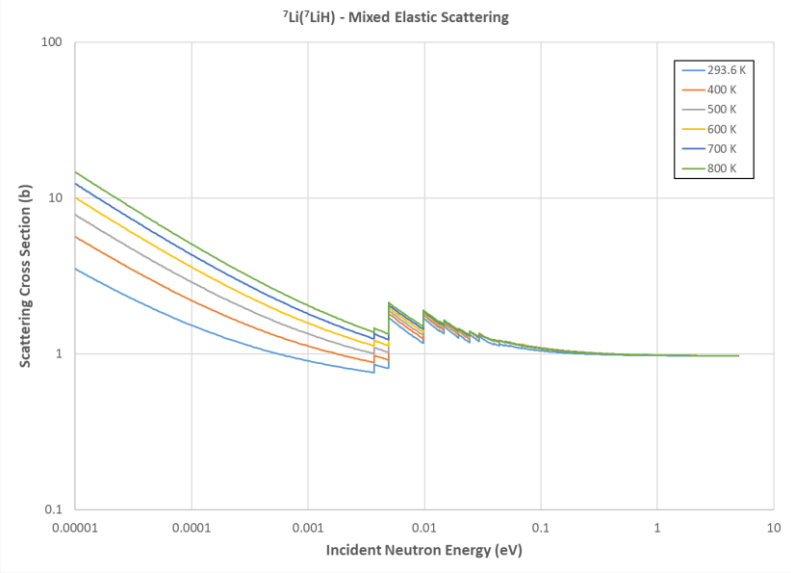




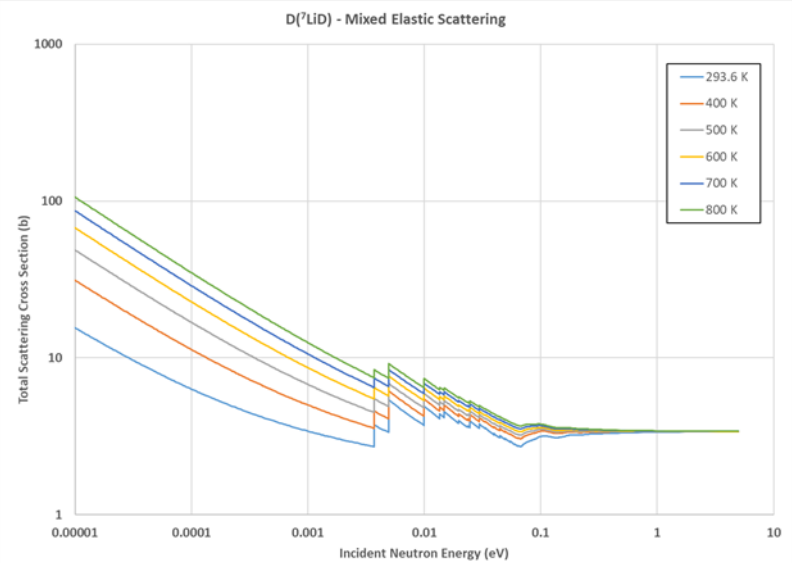


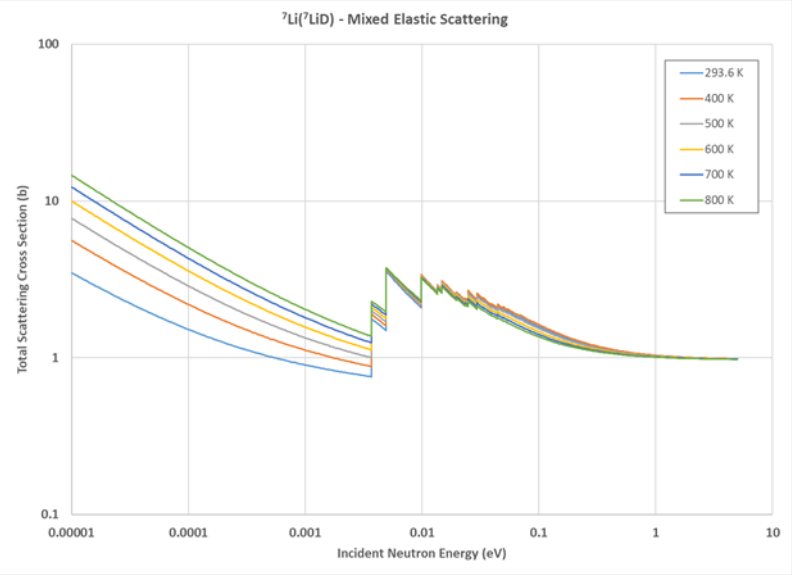
^{7}LiH


H(7LiH) - Mixed Elastic Scattering
Total Scattering Cross Section (b)
1000
100
10
1
0.00001
0.0001
0.001
0.01
0.1
1
10
Incident Neutron Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800 K


7Li(7LiH) - Mixed Elastic Scattering
Scattering Cross Section (b)
100
10
1
0.1
0.00001
0.0001
0.001
0.01
0.1
1
10
Incident Neutron Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800 K

^{7}LiD


D(7LiD) - Mixed Elastic Scattering
Total Scattering Cross Section (b)
1000
100
10
1
0.00001
0.0001
0.001
0.01
0.1
1
10
Incident Neutron Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800 K


7Li(7LiD) - Mixed Elastic Scattering
Total Scattering Cross Section (b)
100
10
1
0.1
0.00001
0.0001
0.001
0.01
0.1
1
10
Incident Neutron Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800 K