A First Look at the Thermal Neutron Scattering Law for H-UH_3

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2018 ANS Annual Meeting
Philadelphia, PA June 17-21 , 2018

NAVAL NUCLEAR LABORATORY

Outline

- Background
  - Benchmarks
  - Phase Diagram
  - Crystal Structure
- Atomistic Simulation
  - AILD Evaluation Process
  - DFT/LD Calculations
  - Phonon Density of States
- TSL Evaluation
- Conclusions
- Next Steps

Background

- Uranium Hydride (UH_3) has been used in several historical critical experiments
  - G. A. Linenberger, et al., *Nucl. Sci. Eng.*, **7**, 44-57 (1960).
  - HEU-COMP-INTER-003, "Reflected Uranium-Hydride Critical Assemblies"
    - 3 doubly reflected cases
    - 4 singly reflected cases
- Goal of this work is to evaluate TSLs for UH_3
  - Using *ab initio* lattice dynamics (AILD) approach
  - Determine bias associated with free gas approximation

HEU-COMP-INTER-003

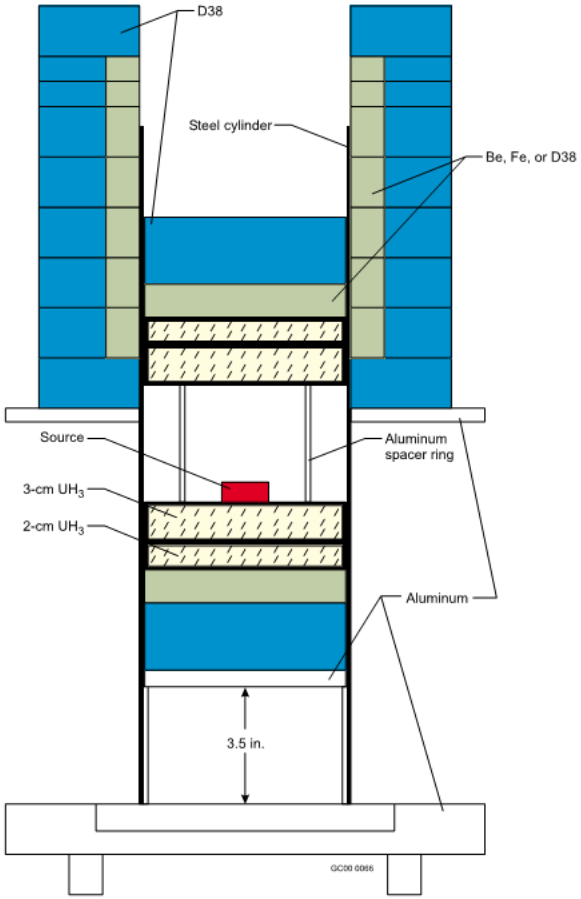


Figure 2. Schematic of the Doubly Reflected Assembly.

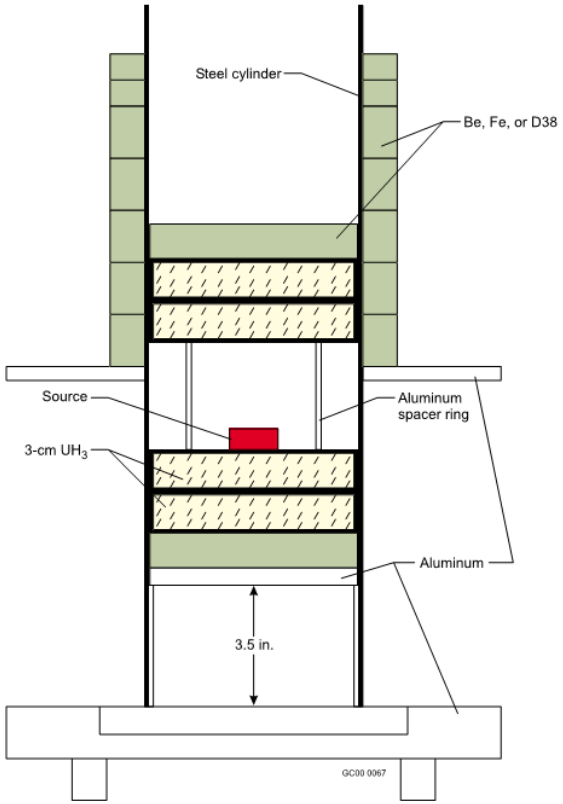
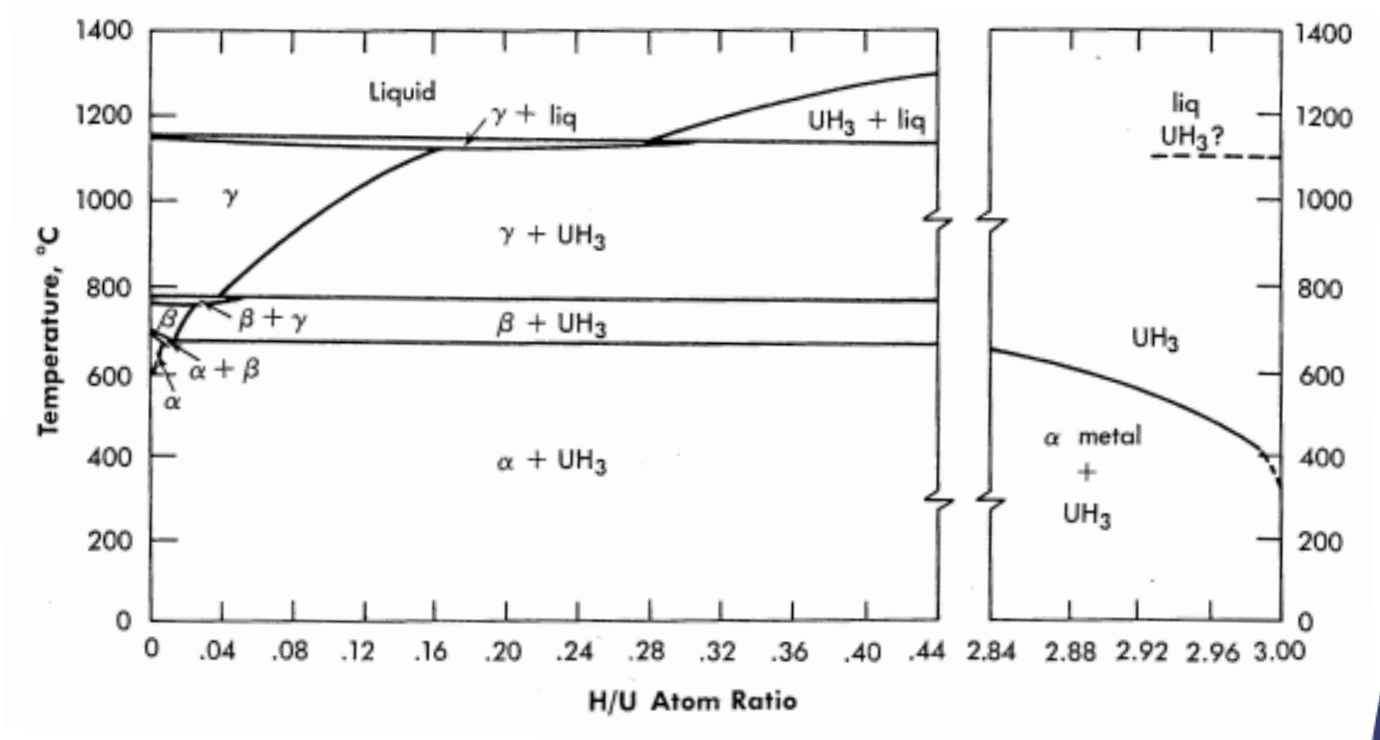


Figure 3. Schematic of a Singly Reflected Assembly.

Uranium-Hydrogen Phase Diagram

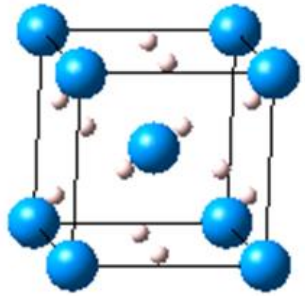
- H_2 (gas) reacts with U (metal) to form uranium hydride (UH_3)
- At room temperature forms two phase solid solution of α-U (metal) + UH_3
- UH_3 fraction increases with H/U atom ratio



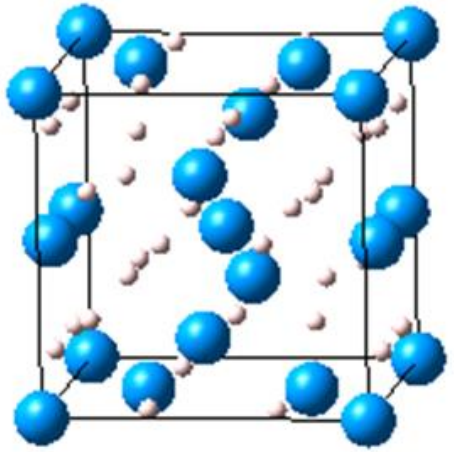
Phase diagram for the Uranium-Hydrogen system. From W. M. Mueller, *et. al*., *Metal Hydrides*, Academic Press, New York, 1968.

UH_3 Crystal Structure

- UH_3 has two allotropes, both cubic
  - Pm3n symmetry group
- α-UH_3 only stable at low (cryogenic) temperatures
  - 2 molecules (8 atoms) per unit cell
  - 4.16 Å lattice constant
- β-UH_3 stable at room temperature and above
  - 8 molecules (32 atoms) per unit cell
  - 6.643 Å lattice constant
- H-UH_3 TSL developed using first-principles or *ab initio* lattice dynamics (AILD) approach
  - Density Functional Theory (DFT) to calculate interatomic Hellman-Feynman forces for crystal structure
  - Lattice Dynamics (LD) to determine dispersion relations and phonon density of states (PDOS)
  - H-UH_3 TSL evaluated in incoherent approximation using NJOY/LEAPR

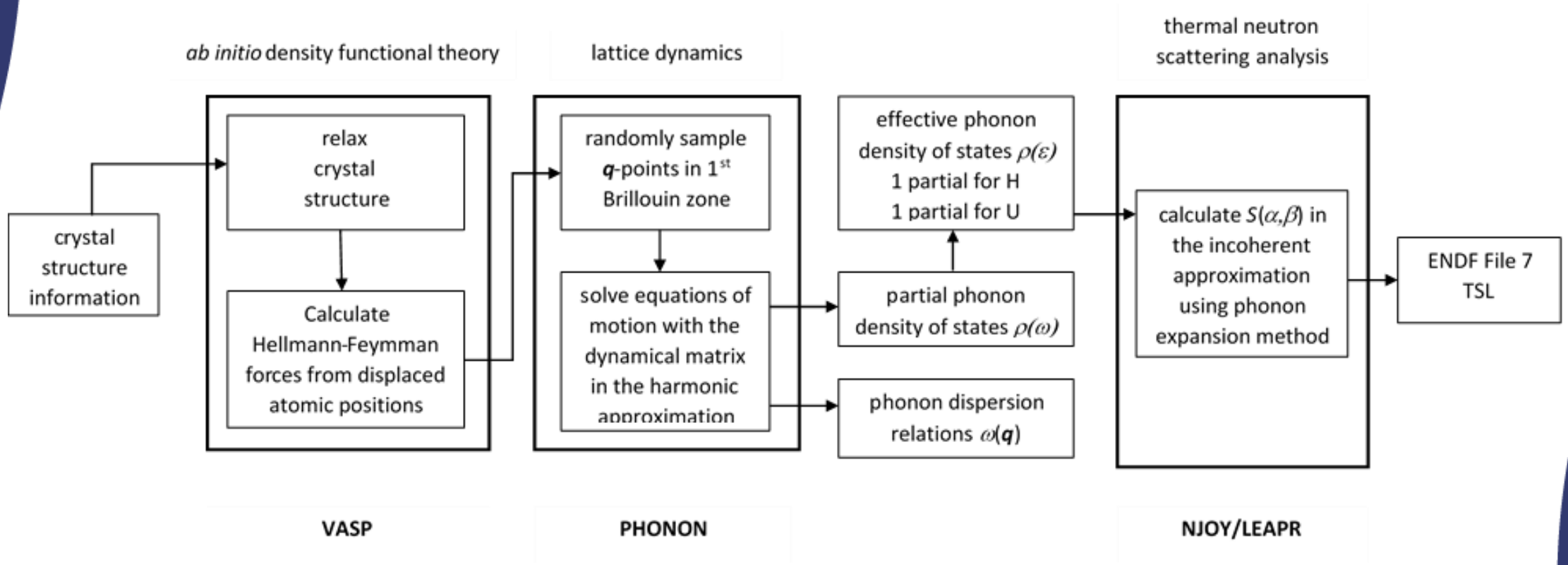


α-UH_3 Unit Cell



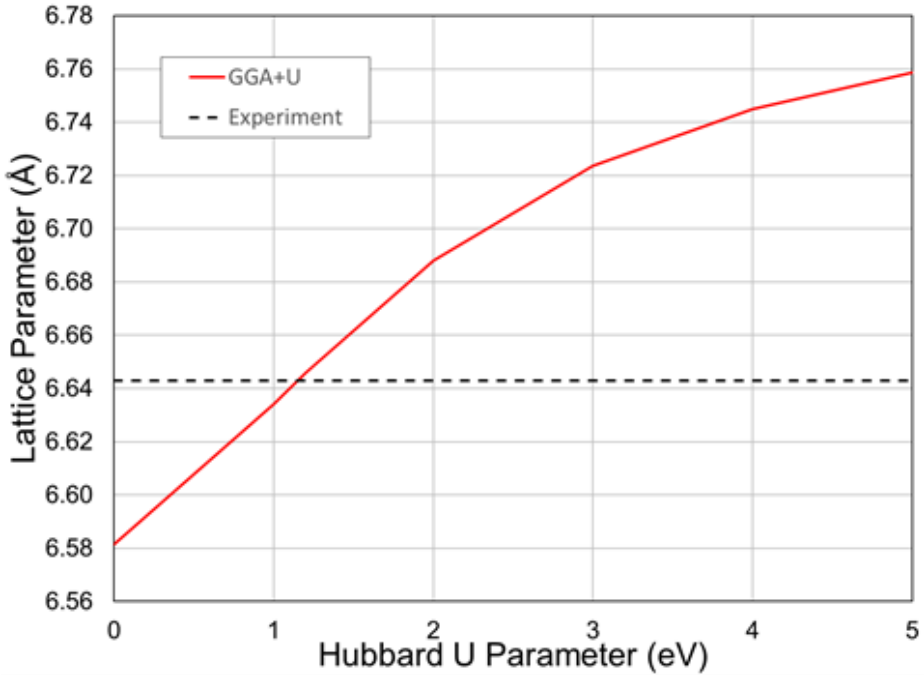
β-UH_3 Unit Cell

AILD-based TSL Evaluation Process



UH_3 Structure Optimization

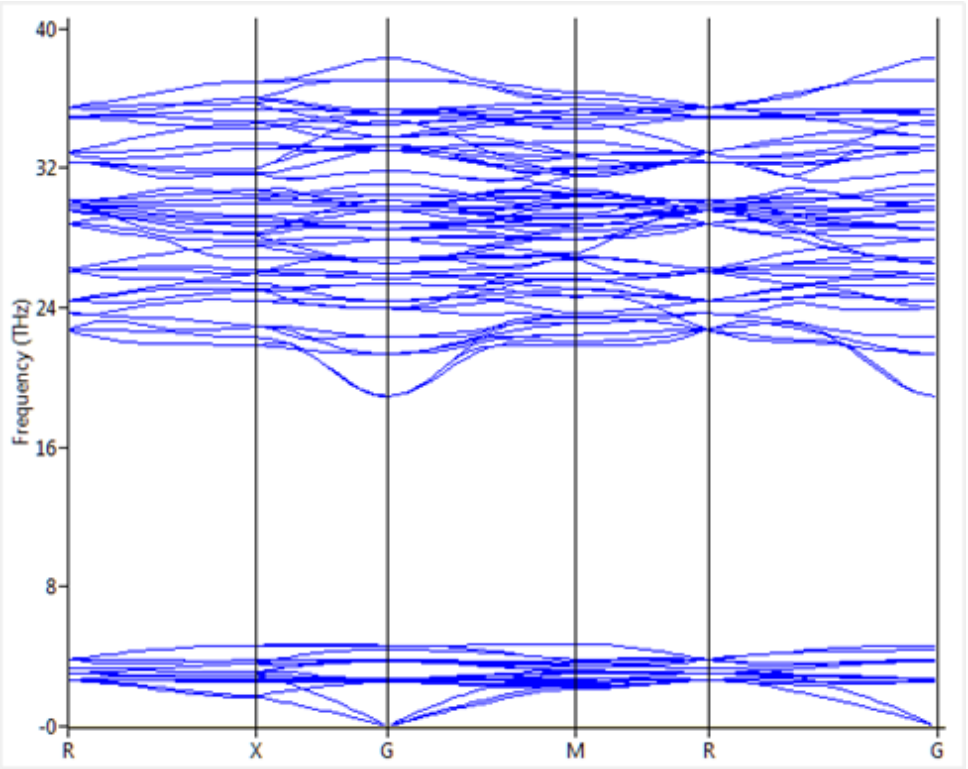
- DFT structure optimization using VASP (*Vienna Ab Initio Simulation Package*)
  - GGA exchange and correlation functional
  - Hubbard *U* parameter correction applied to U 5*f* electrons
    - Account for effect of strong correlation of 5*f* electrons on chemical binding of U molecules
  - Spin-polarized magnetism
  - 500 eV planewave cutoff
  - *k*-point spacing of 0.2 $Å^{-1}$ (5×5×5 k-mesh)
  - 10^{-6} eV total electronic energy threshold
- **Hubbard U = 1.2 eV yields lattice parameter of *a* = 6.6458 Å**
  - 0.04% higher than measured lattice parameter of *a* = 6.643 Å



VASP structure optimization of UH_3 using GGA+U.

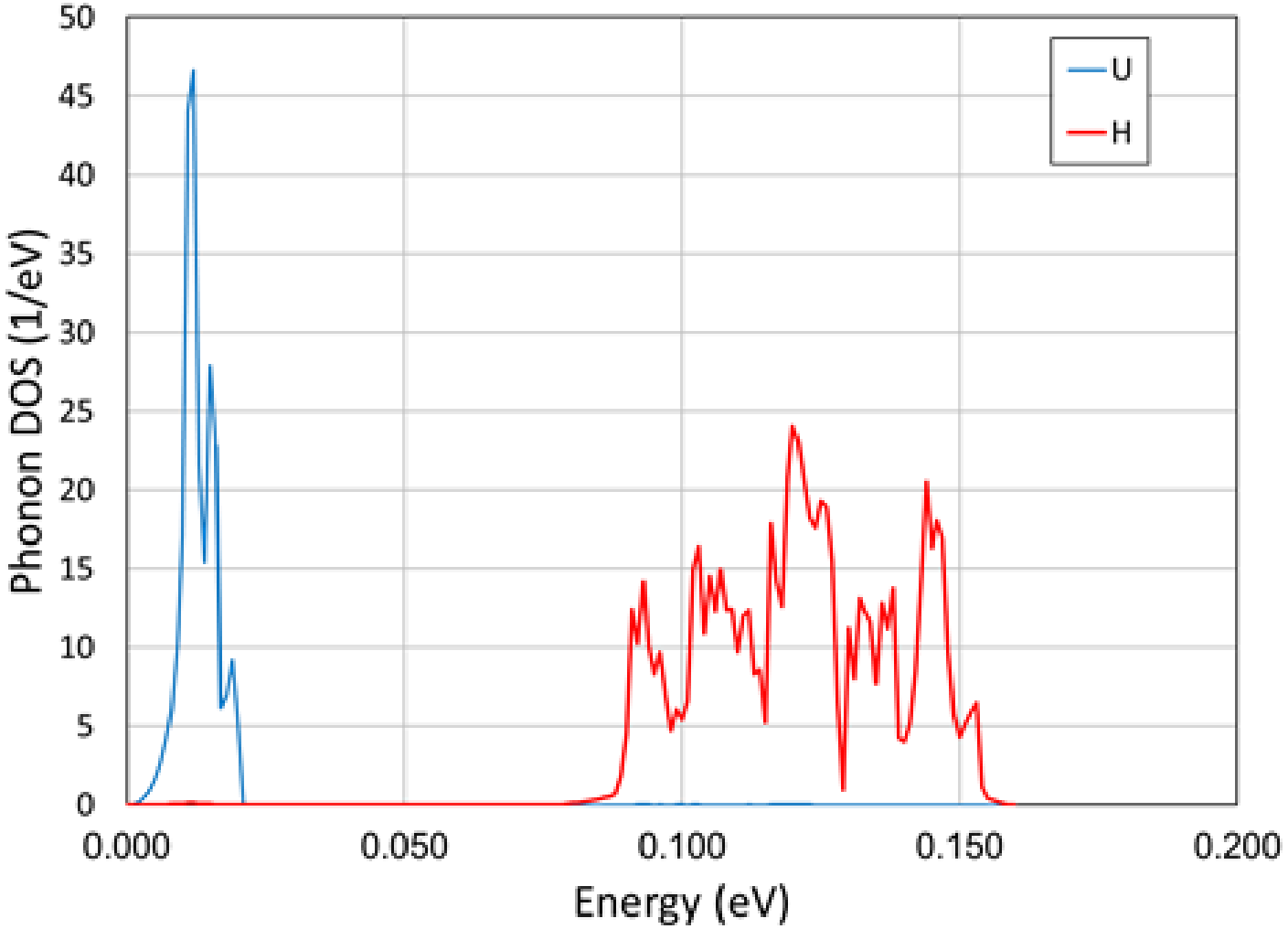
UH_3 Lattice Dynamics

- LD calculations performed using PHONON
  - Interatomic forces calculated by VASP
  - 2×2×2 supercell (256 atoms)
  - ±0.02 Å atom displacements
  - 0.02 $Å^{-1}$ *k*-point spacing (3×3×3 *k*-point mesh)
- Dispersion relations (at right)
  - Well separated acoustic and optical modes
  - Lower branches are acoustic modes mainly due to heavy U atom vibrations
  - Higher branches are optical modes mainly due to light H atom vibrations
    - Relatively wide and dense optical mode



Calculated dispersion relation for UH_3 along the highest-symmetry points of the Brillouin zone.

Calculated Phonon DOS for UH_3



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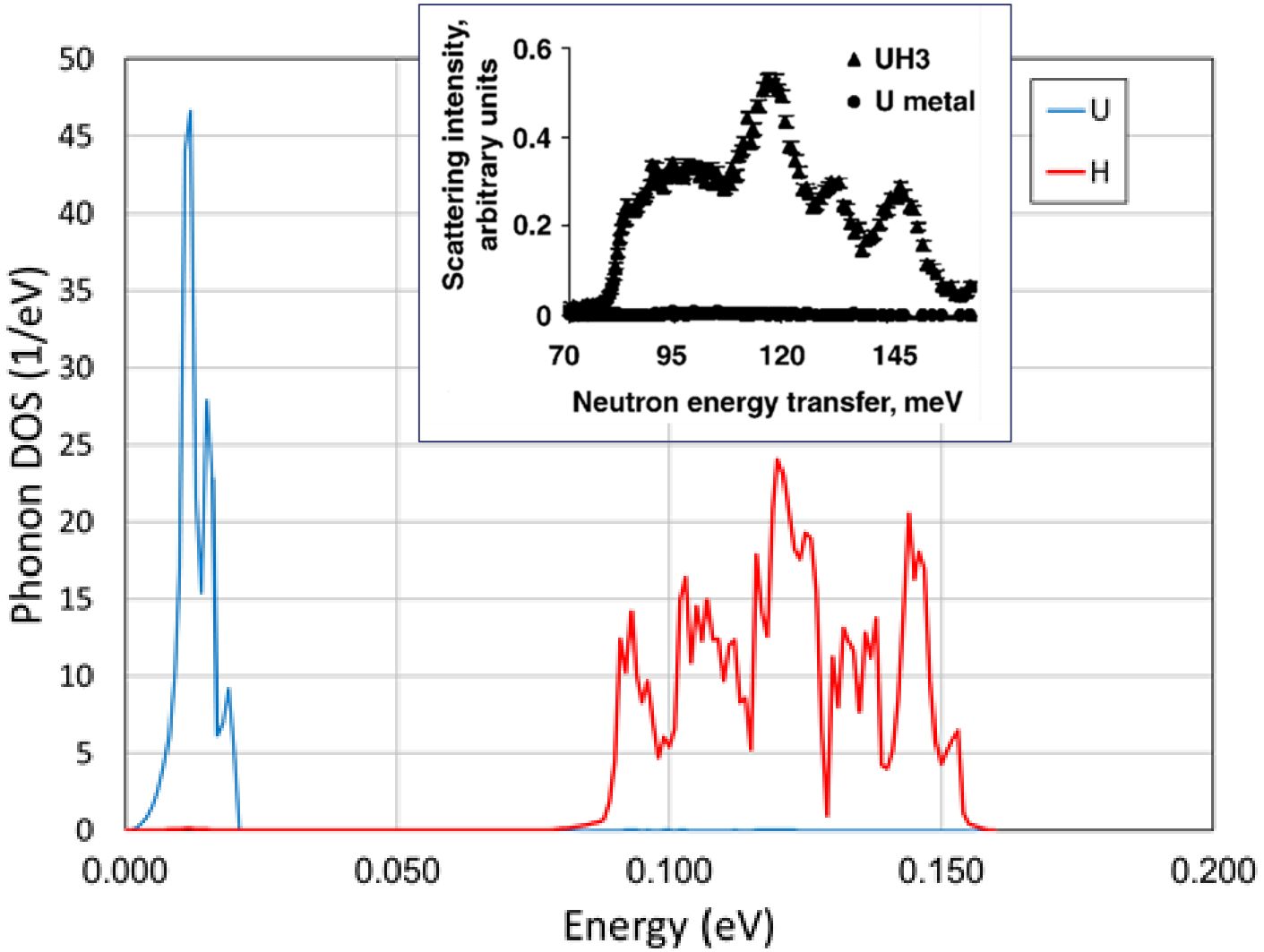
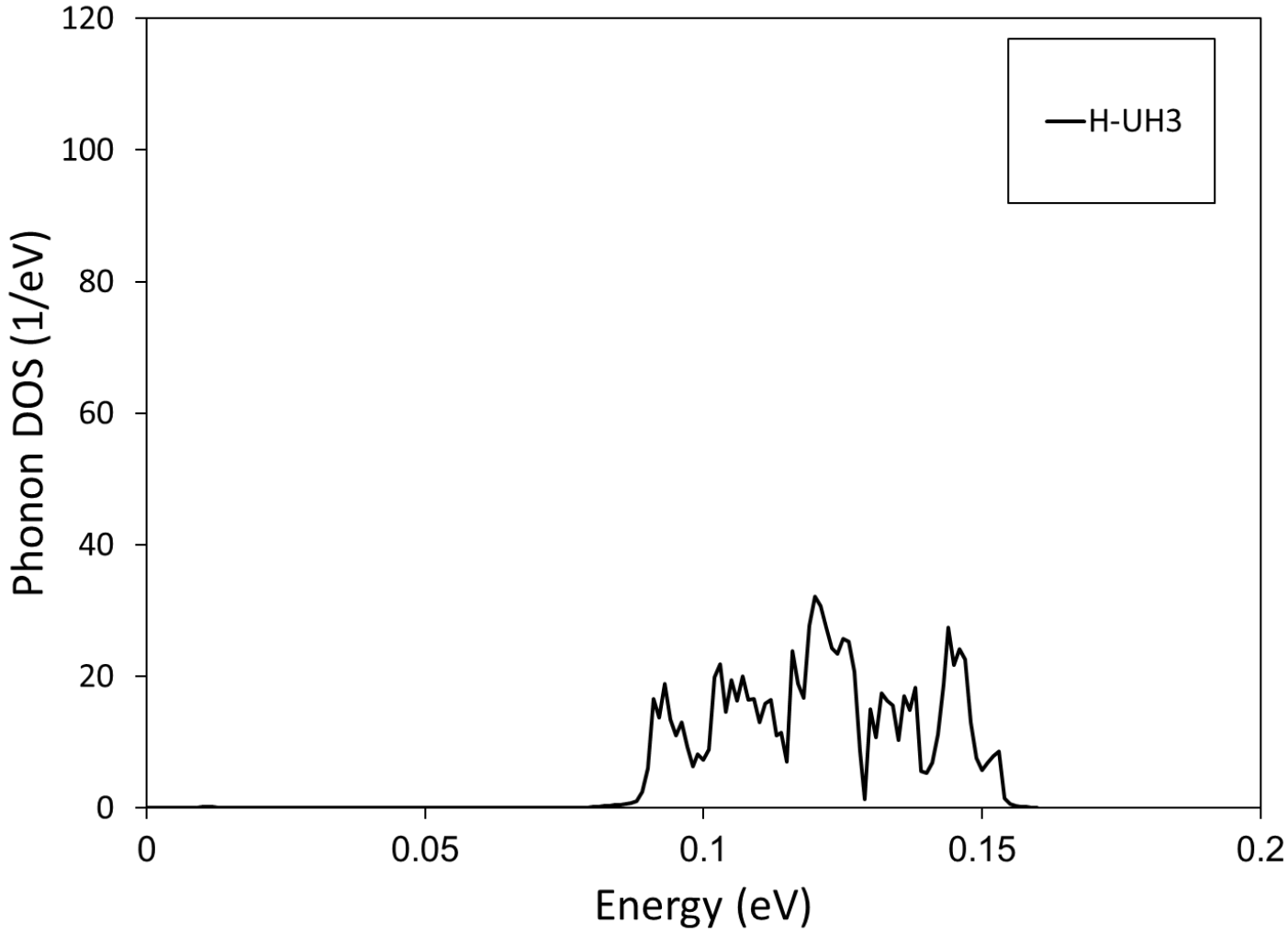


Comparision of H Phonon DOS for Several Metal Hydrides



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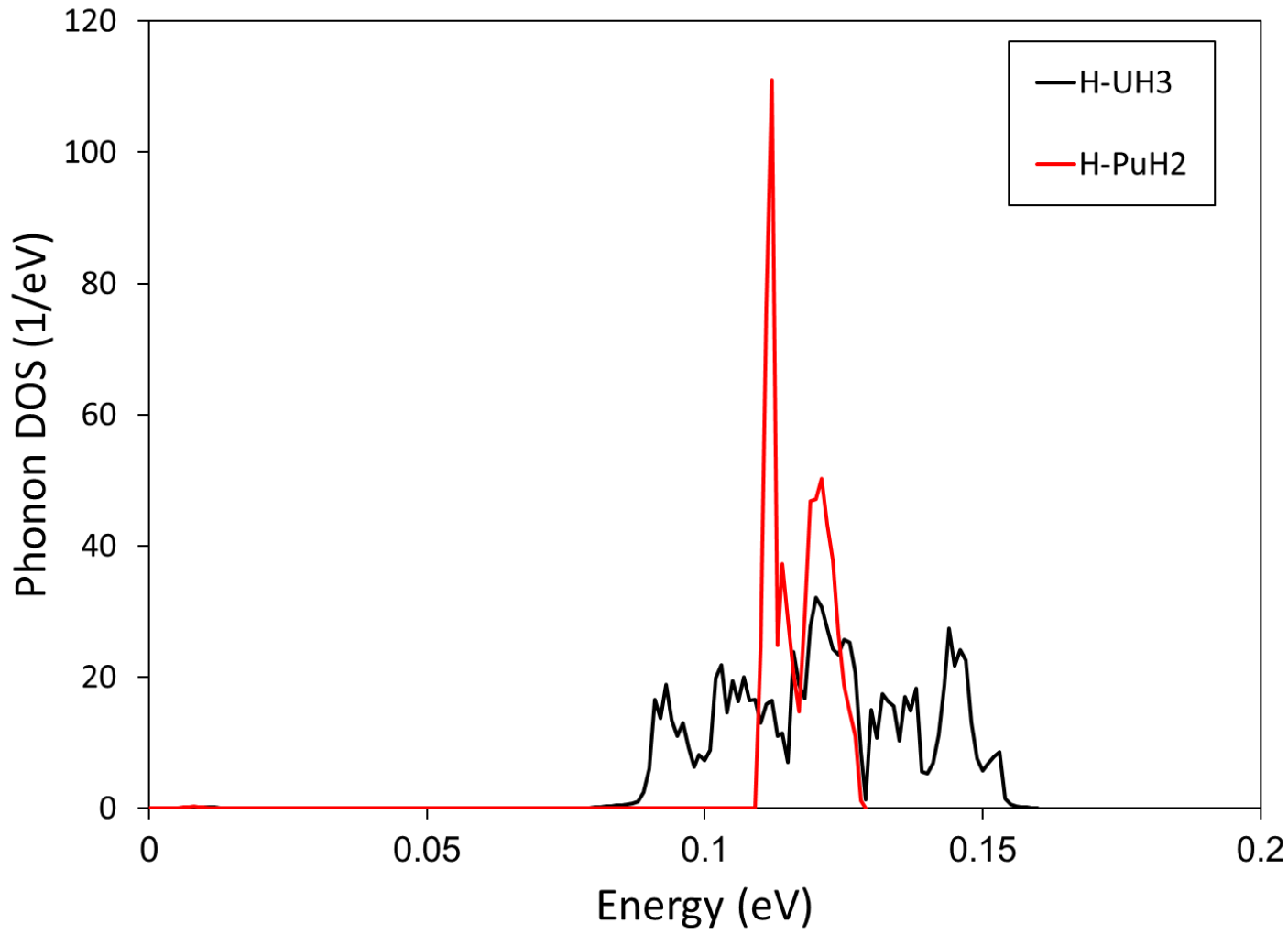

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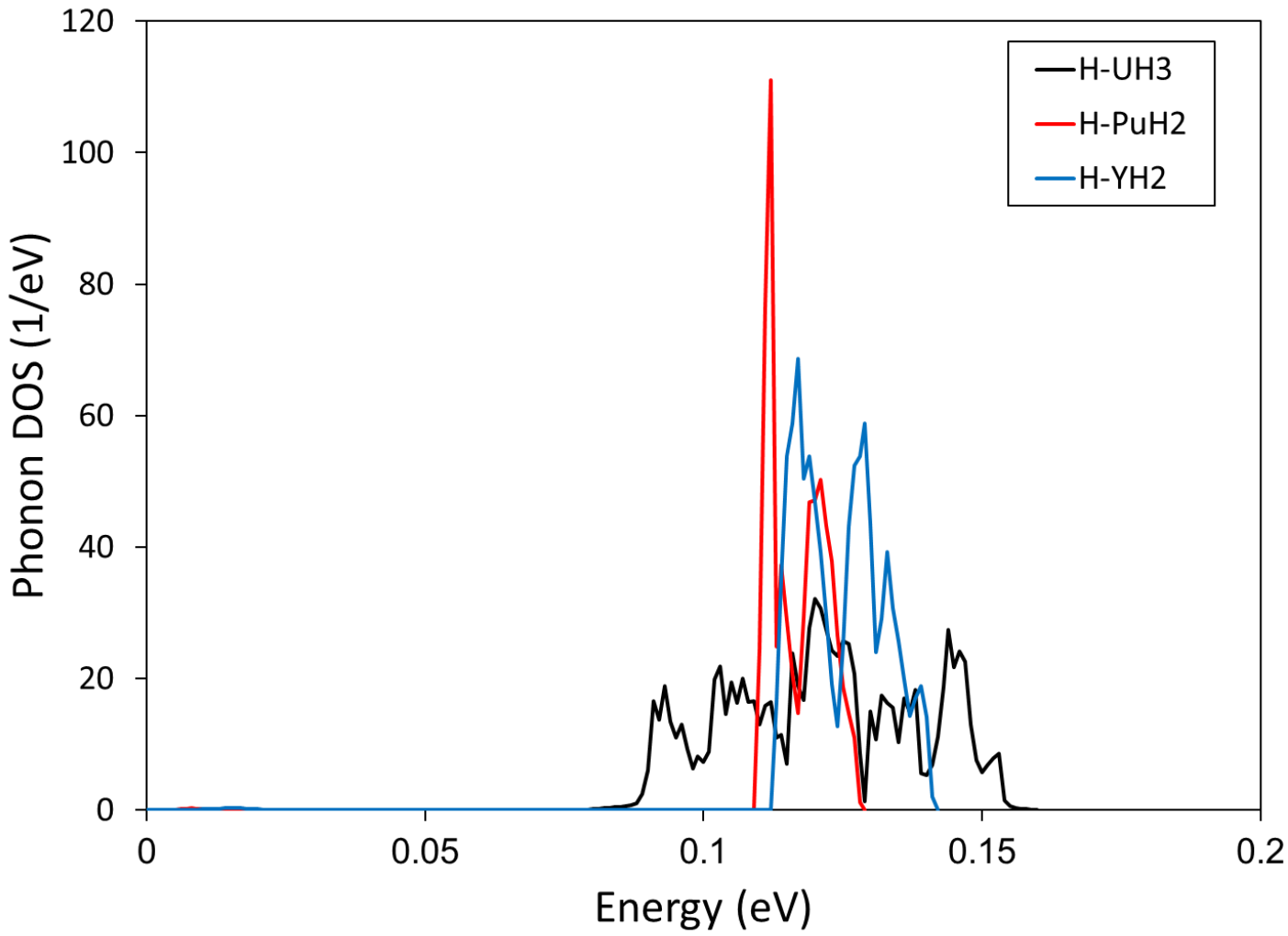

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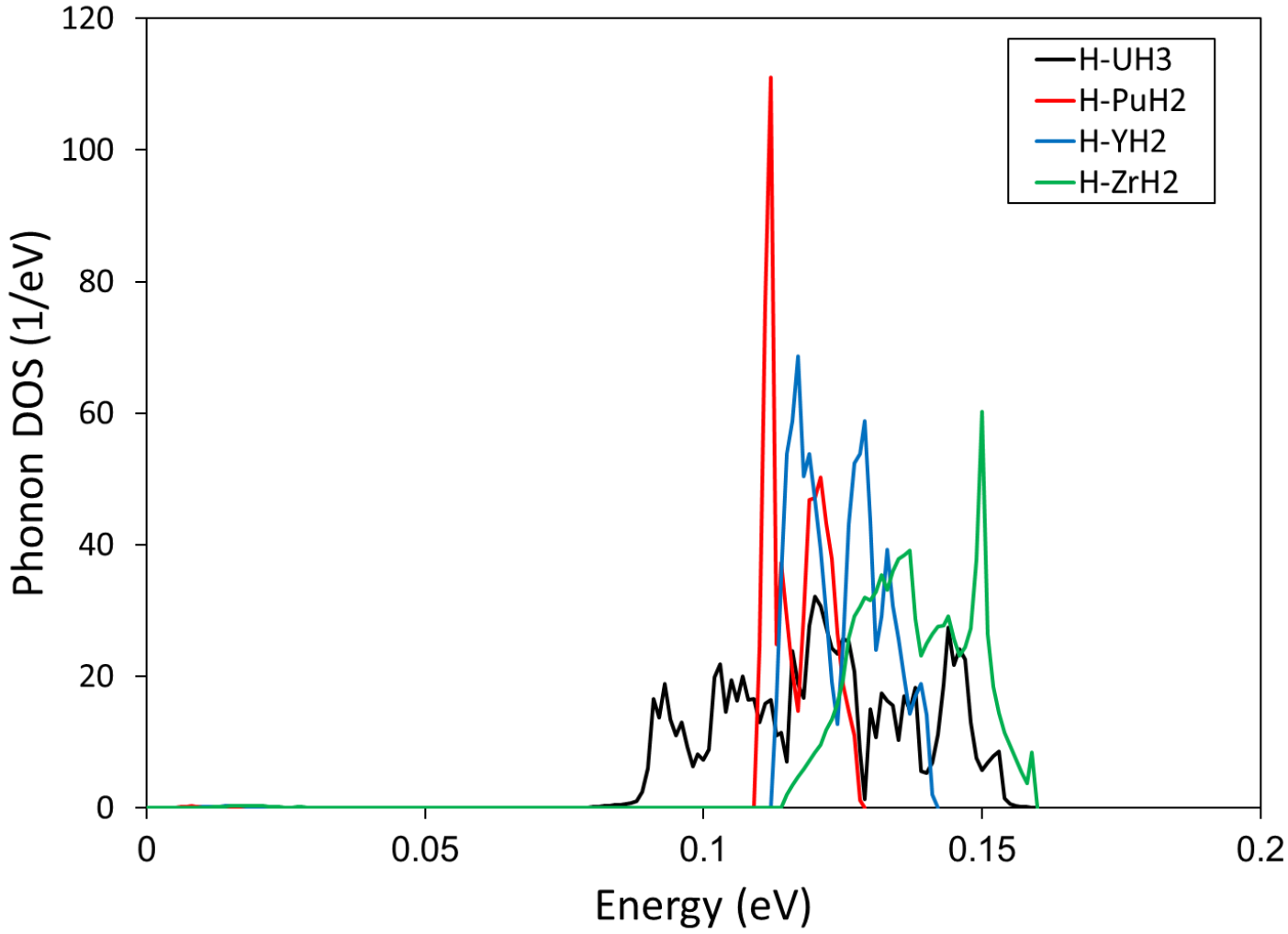
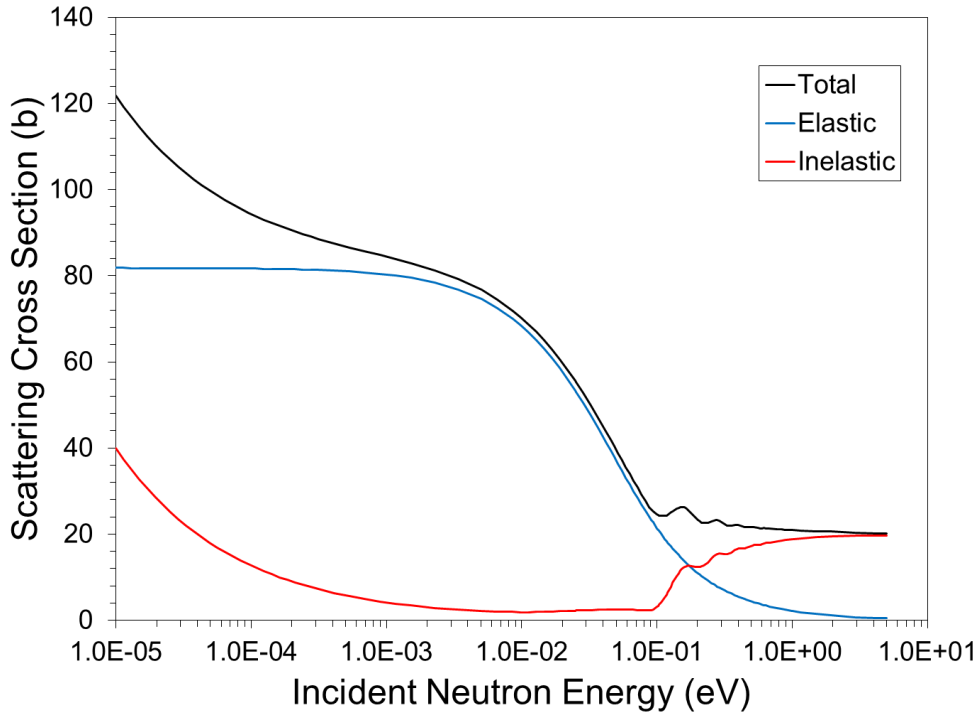


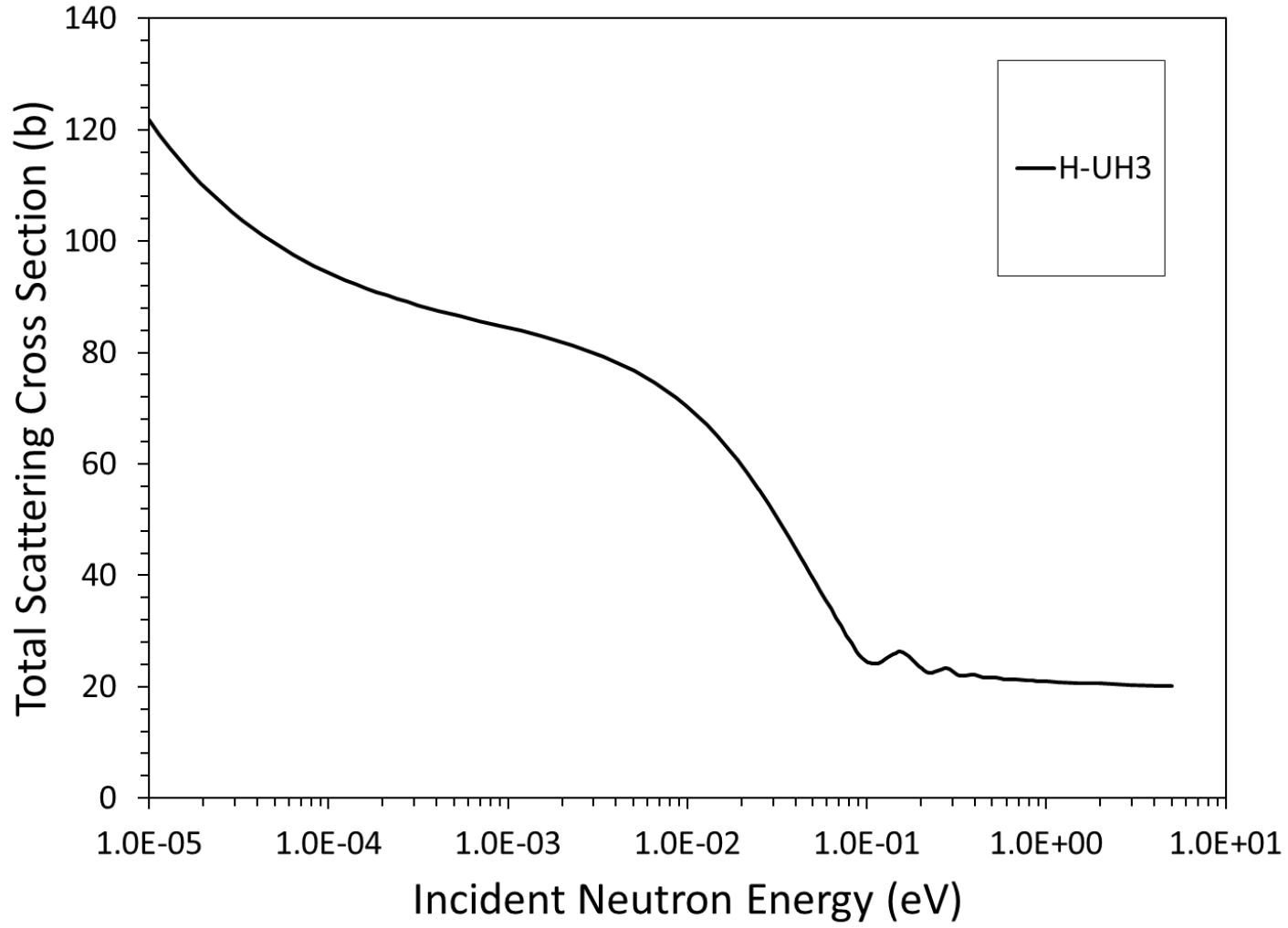
H-UH_3 TSL Evaluation

- H-UH_3 TSL generated using NJOY/LEAPR
  - H-UH_3 phonon DOS from PHONON calculation
  - Incoherent approximation
  - Atomic mass ratio and free atom scattering cross section for ^{1}H from ENDF/B-VII.1
  - α and β grids optimized to treat scattering up to 5 eV without SCT approximation
  - Temperature evaluated at 293.6 K (room temp.)
- U-UH_3 TSL not evaluated at this time
  - LEAPR can't properly treat U coherent elastic scattering in UH_3 without extensive modifications
  - Plan to evaluate U-UH_3 using FLASSH
  - Proper treatment of coherent elastic scattering and relax incoherent approximation



Total, elastic, and inelastic scattering cross section for H-UH_3 at 293.6 K generated by NDEX

Comparision of Total Scattering Cross Sections for Several Metal Hydrides



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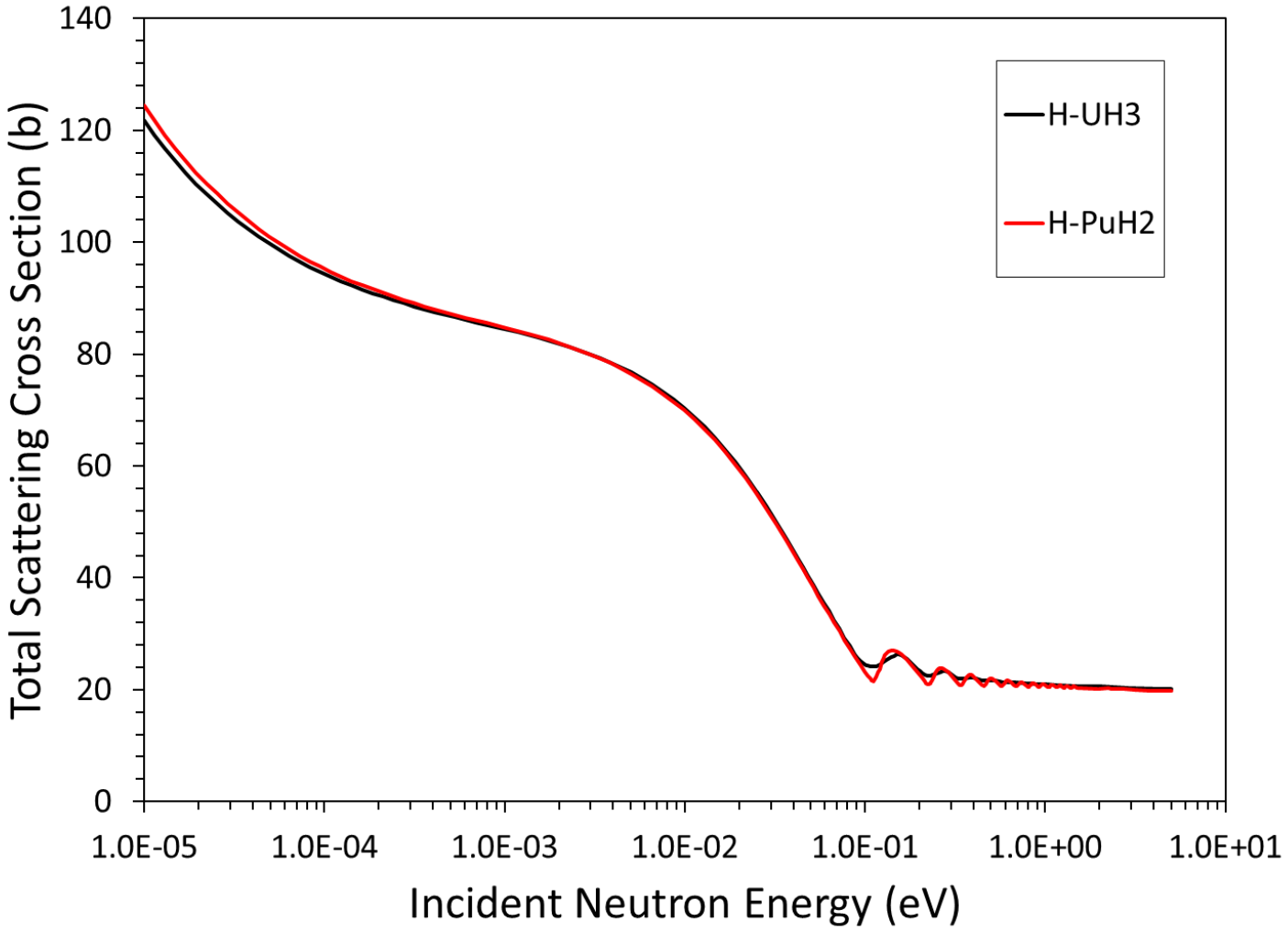

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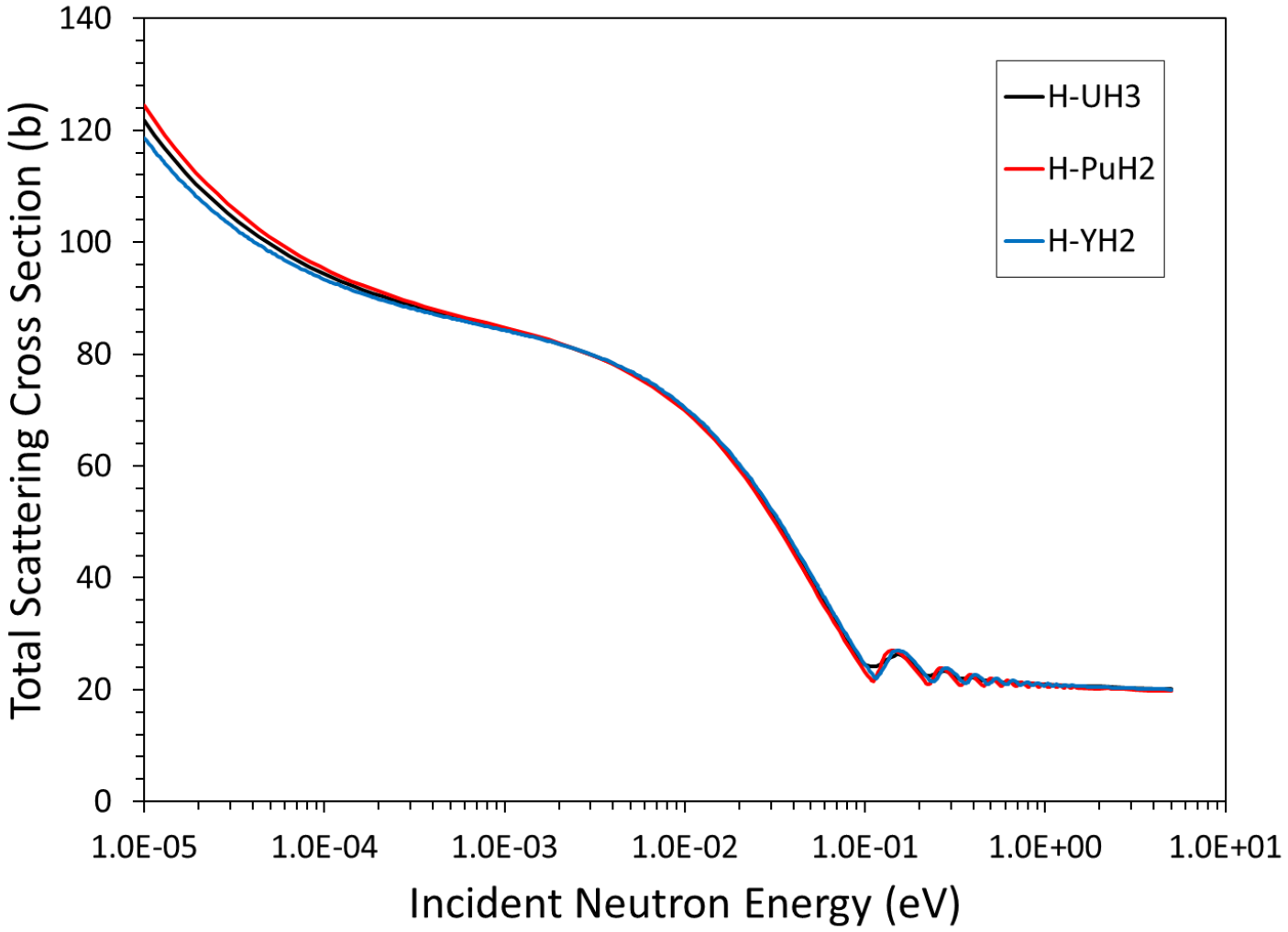

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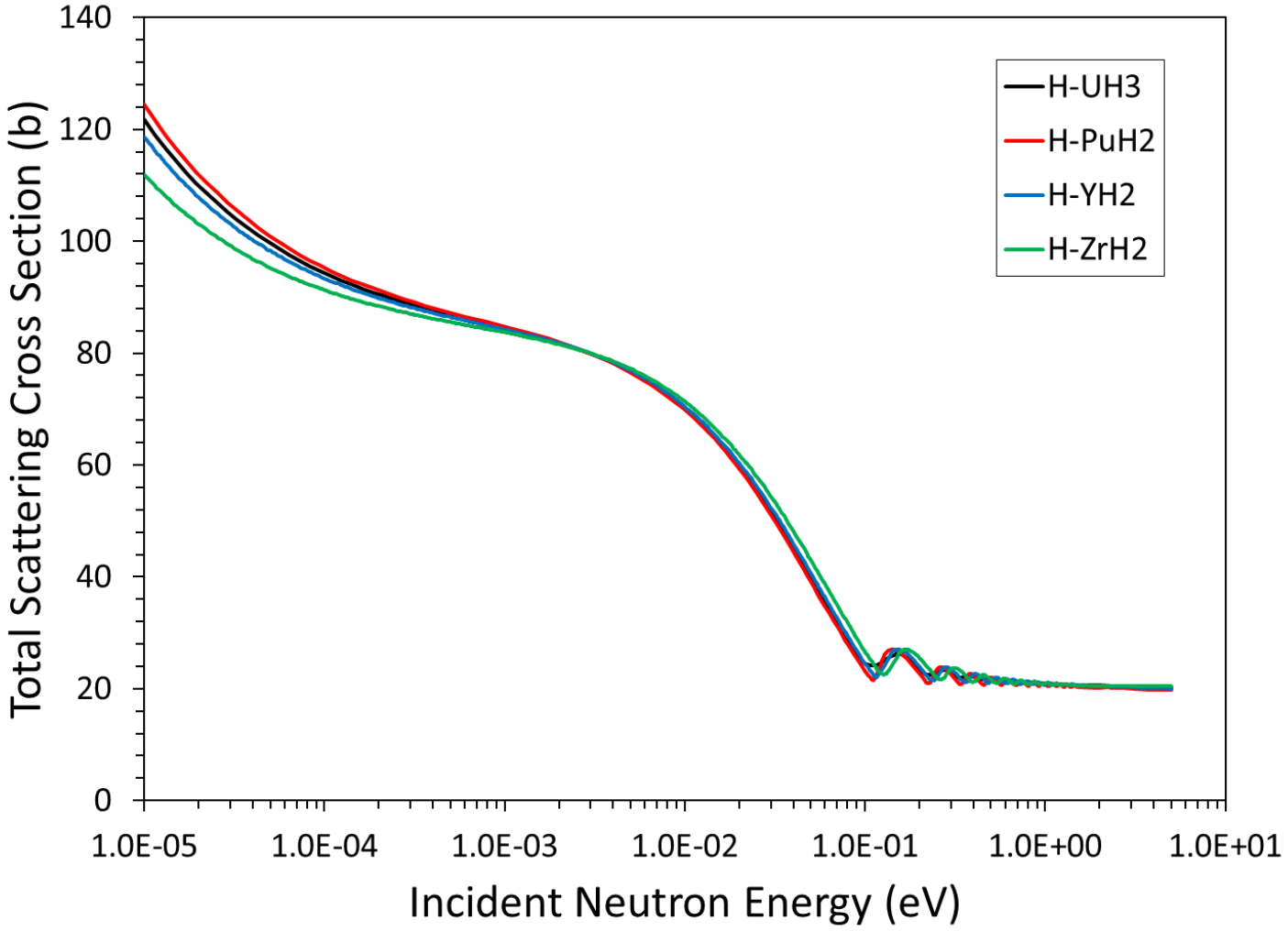

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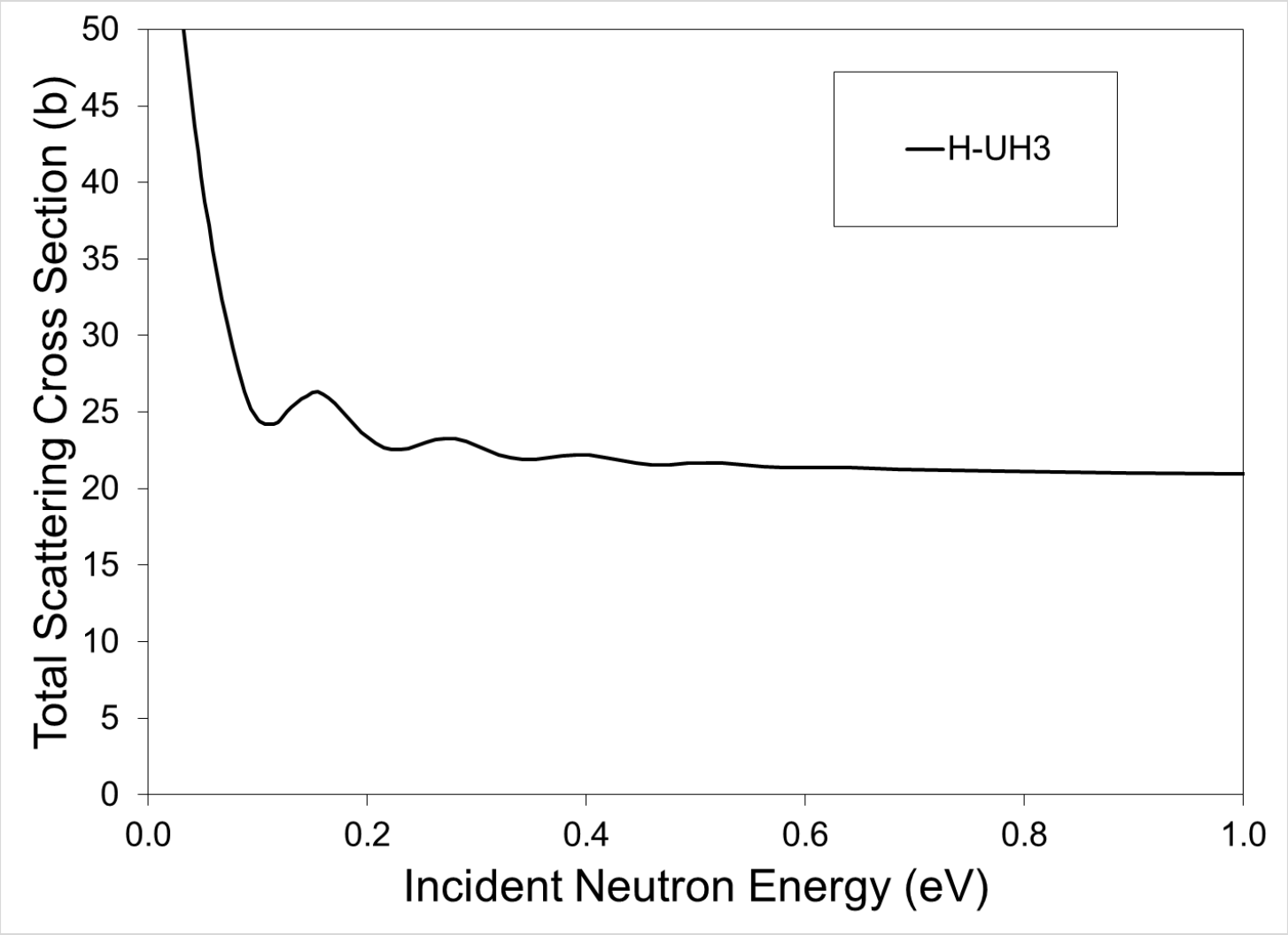

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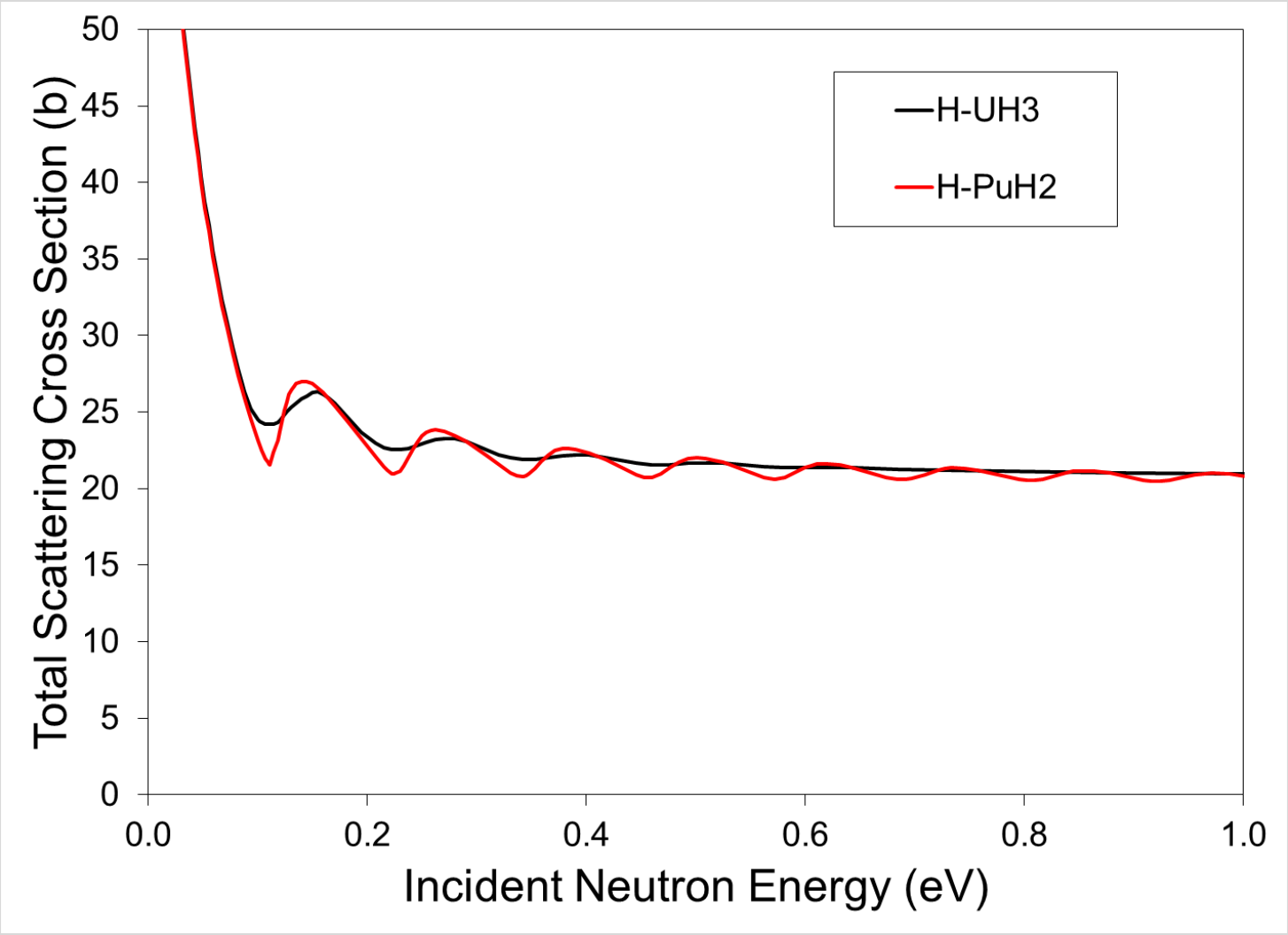

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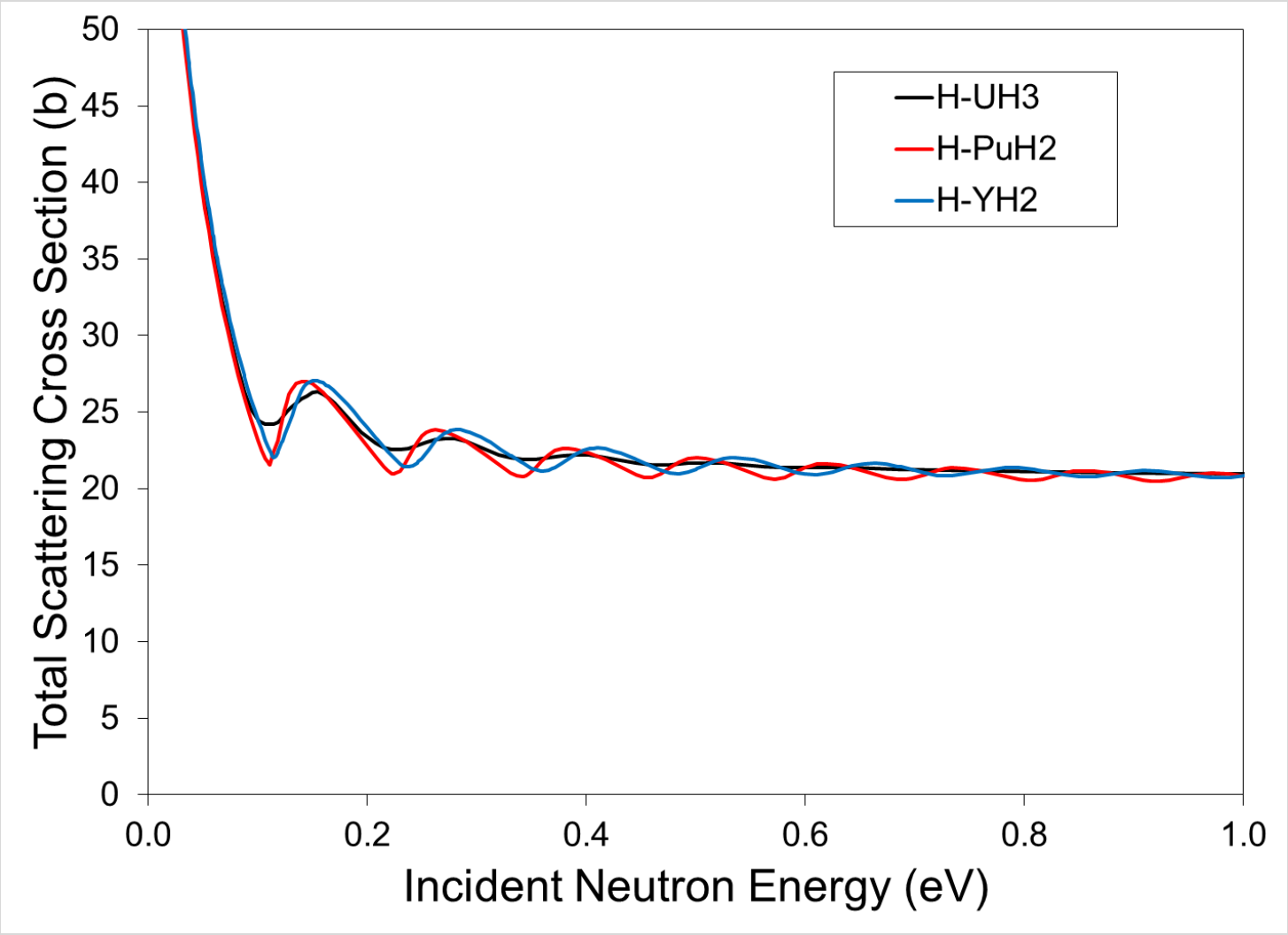

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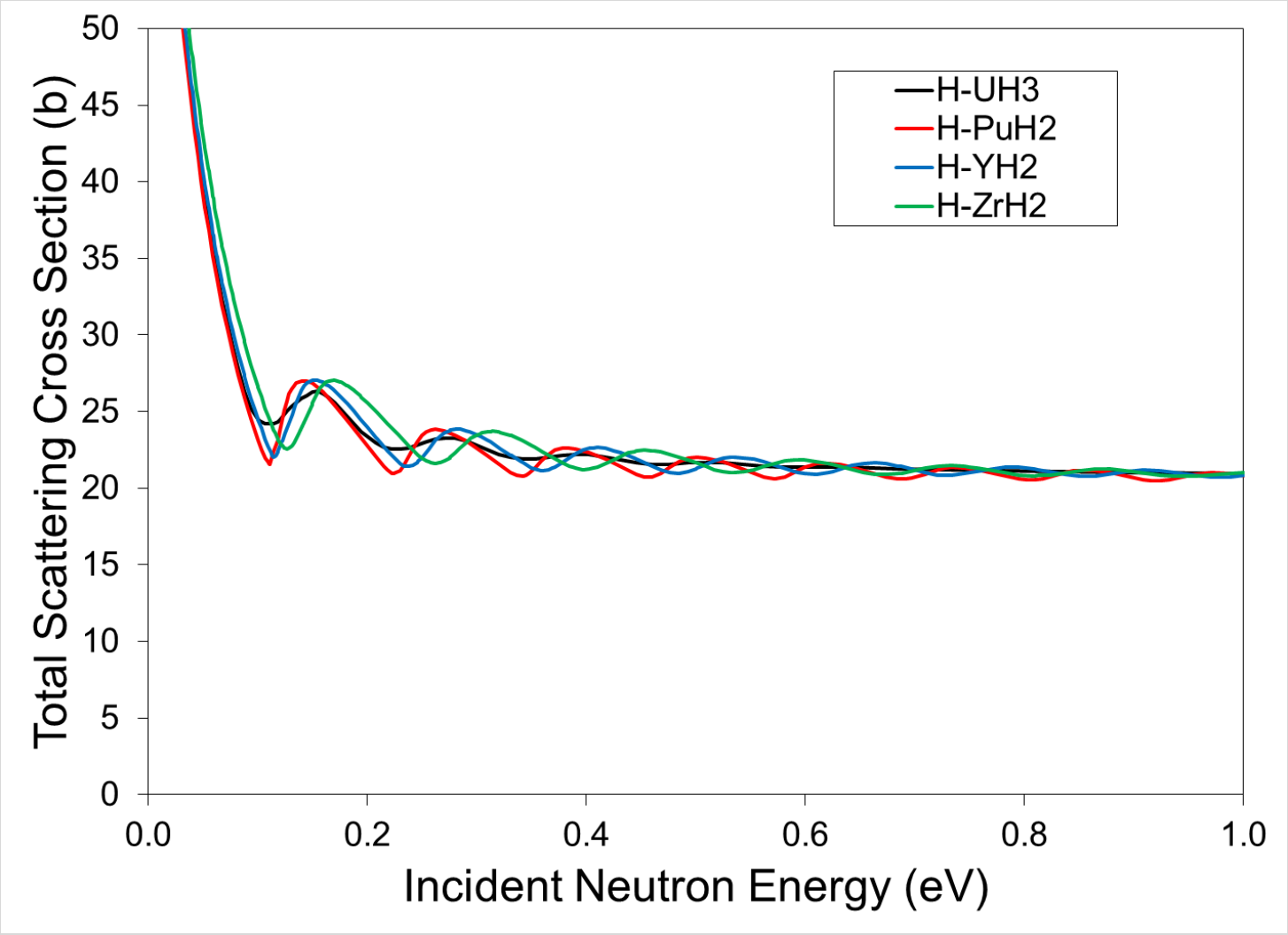


Conclusions

- Calculated phonon DOS for UH_3 using AILD methods
- Optical mode consistent with inelastic neutron scattering measurements performed at NIST
- H-UH_3 TSL evaluated using calculated phonon DOS
- Broader optical mode in UH_3 results in shallower multiphonon scattering peaks in the H-UH_3 inelastic and total scattering cross section relative to other metal hydrides that have been evaluated

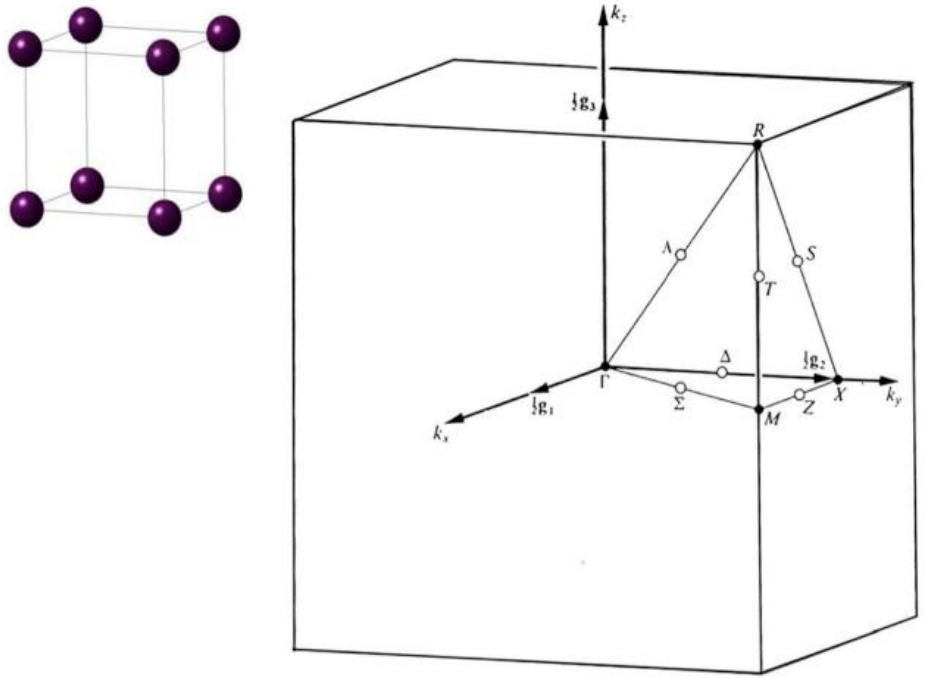
Next Steps

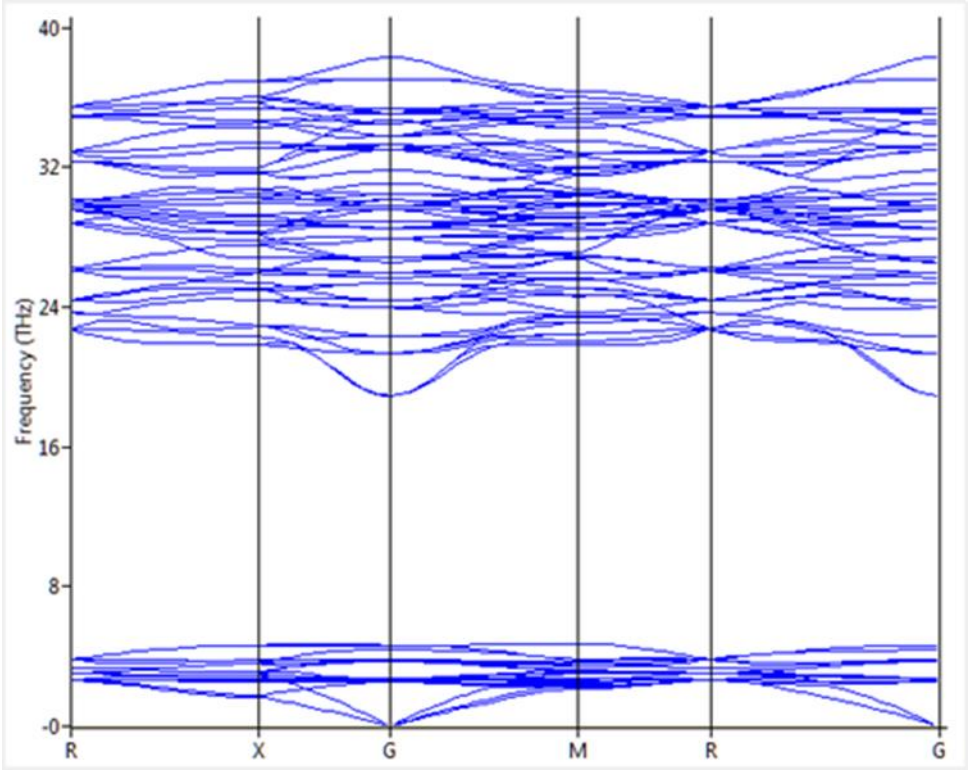
- Evaluate U-UH_3 in FLASSH
  - Generic coherent elastic scattering capability
  - Relax incoherent approximation
- Re-evaluate H-UH_3 in FLASSH
  - Best to have H-UH_3 and U-UH_3 evaluated in same code using consistent approximations
- MC21 benchmark testing using HEU-COMP-INTER-003
  - Impact of H-UH_3 and U-UH_3 TSLs
  - Bias associated with free gas approximation

Backup Slides

UH_3 Lattice Dynamics

Simple Cubic Brillion Zone





Calculated dispersion relation for UH_3 along the highest-symmetry points of the Brillouin zone.