NNL TSL Evaluation Work for ENDF/B-VIII.1

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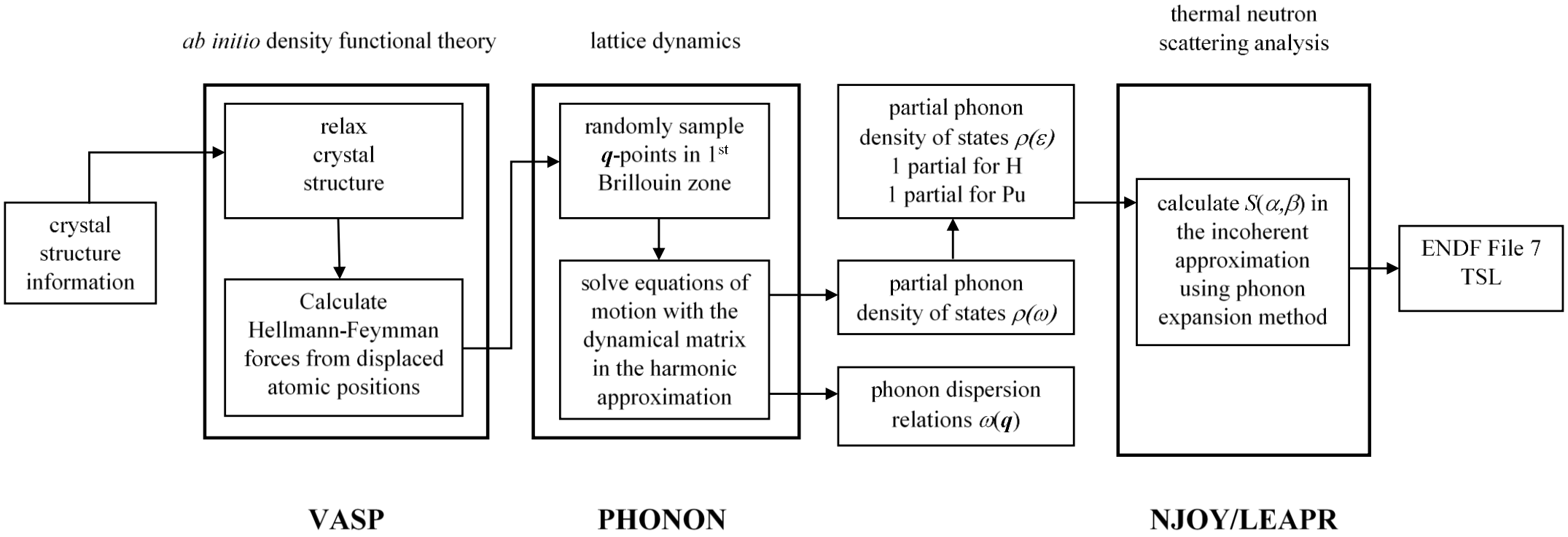
NAVAL NUCLEAR LABORATORY

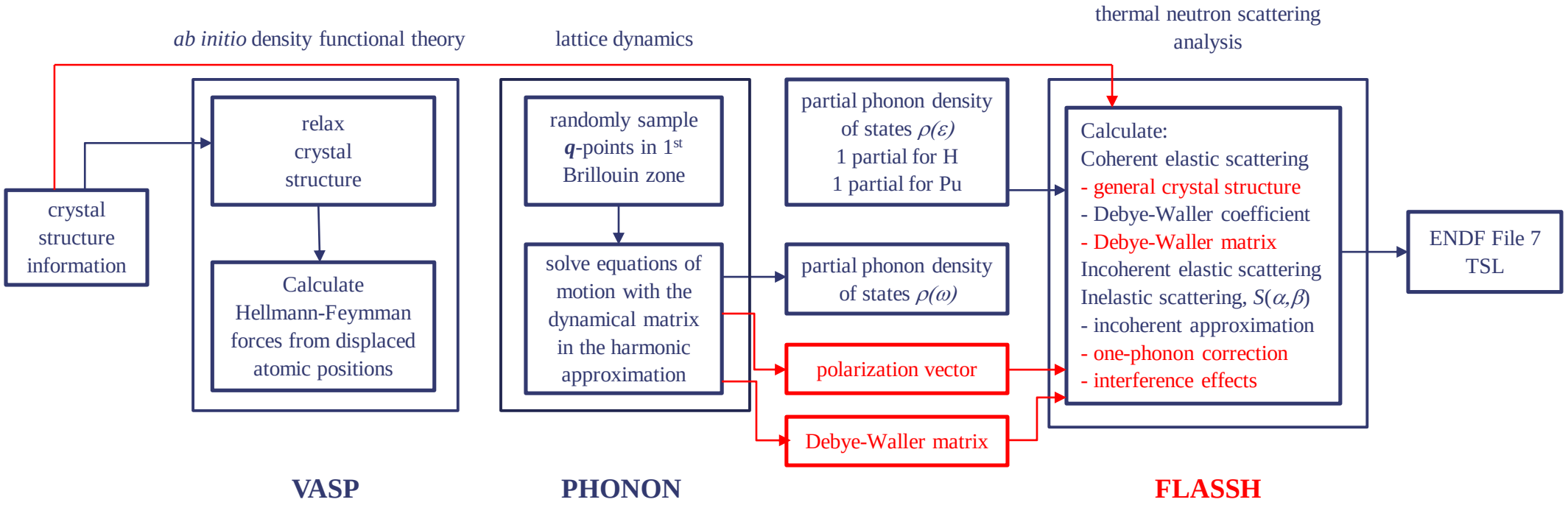
Outline

- NNL TSL Evaluation Process
  - Current
  - New
- NNL Evaluations in Progress
  - H-PuH_2 (2017 NCSD Topical Meeting)
  - H-UH_3 (2018 ANS Annual Meeting)
- Summary

Current TSL Evaluation Process (Crystalline Moderators)

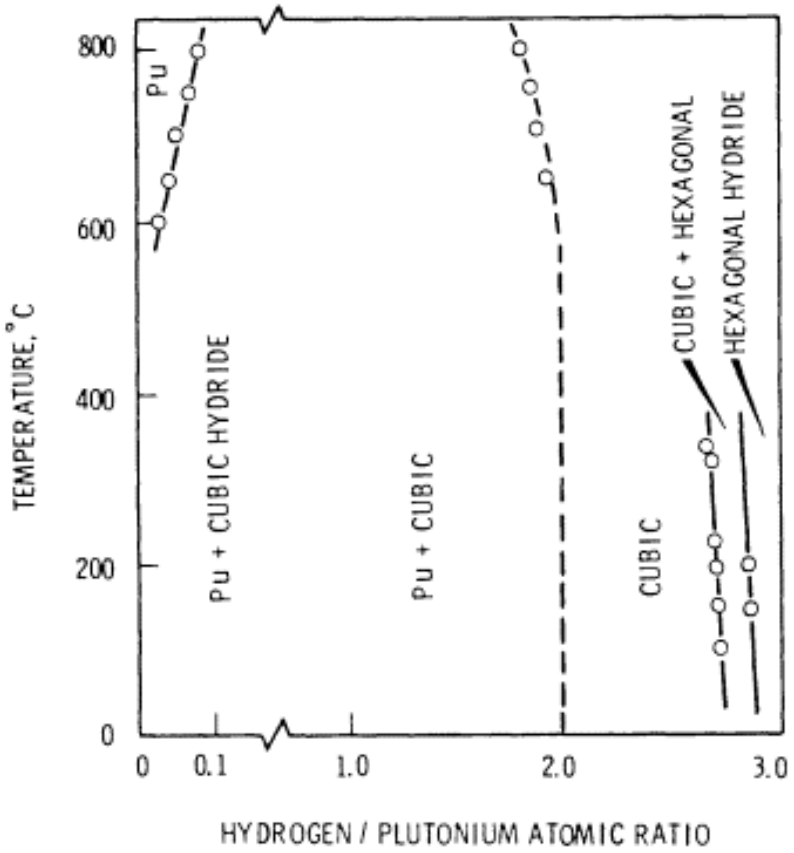


New TSL Evaluation Process (Crystalline Moderators)



Plutonium-Hydrogen Phase Diagram

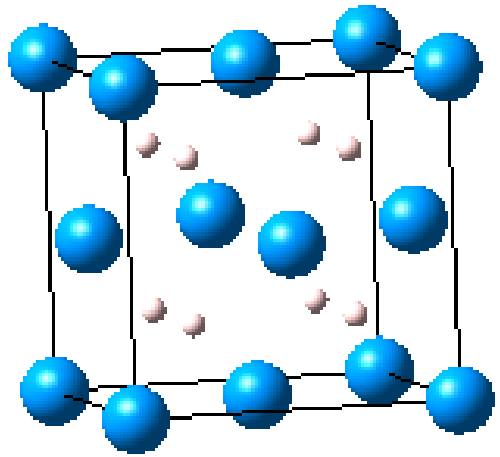
- H_2 (gas) reacts with Pu (metal) to form plutonium hydrides (PuH_x)
- Three PuH_x phases present
  - PuH_2 (FCC) for H/Pu ≤ 2.0
    - Two phase solid solution of Pu (metal) + PuH_2 (FCC)
  - PuH_{2+x} (FCC) for 2.0 < H/Pu < 3
    - Single phase solid solution for 2.0 < H/Pu < 2.75
    - Two phase solid solution, PuH_{2+x} (FCC) + PuH_3 (Hex) for 2.75 < H/Pu < 3-ε
  - PuH_3 (Hex) for 2.75 < H/Pu ≤ 3.0
    - Single phase solid solution for 3-ε < H/Pu < 3.0
- This work concentrates on PuH_2
  - PuH_{2+x} and PuH_3 to be evaluated later



Phase diagram for the plutonium-hydrogen system. From R. N. R. Mulford and G. E. Sturdy, *J. Am. Chem. Soc.*, **78**, 3899 (1956).

PuH_2 Structure

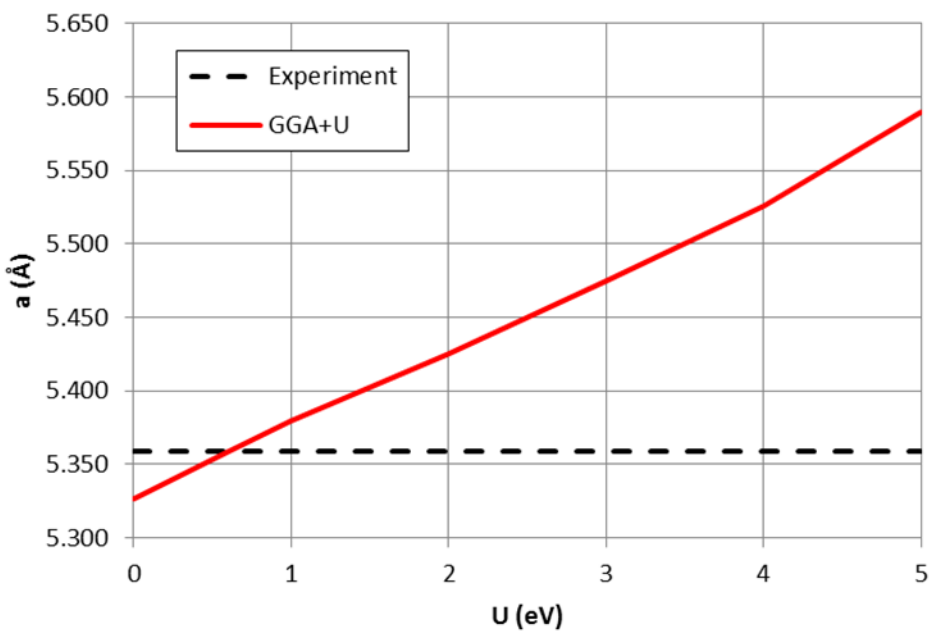
- PuH_2 has a CaF_2 type FCC structure
  - 12 atoms per unit cell
  - 4 Pu atoms (blue) at vertices and faces of unit cell
  - 8 H atoms (grey) in tetrahedral holes between Pu atoms
  - Mass density of 10.40 g/cm^3
- Measured lattice parameter (X-ray diffraction)
  - *a* = 5.359 ± 0.002 Å, Mulford and Sturdy (1955)
  - *a* = 5.359 ± 0.001 Å, Coffinberry and Ellinger (1956)
  - *a* = 5.359 ± 0.002 Å, Muromura et al. (1972)
  - *a* = 5.3593 Å, Willis et al. (1985)
- H-PuH_2 TSL developed using first-principles 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 (DOS)
  - H-PuH_2 TSL evaluated in incoherent approximation using NJOY/LEAPR



PuH_2 Unit Cell

PuH_2 Structure Optimization

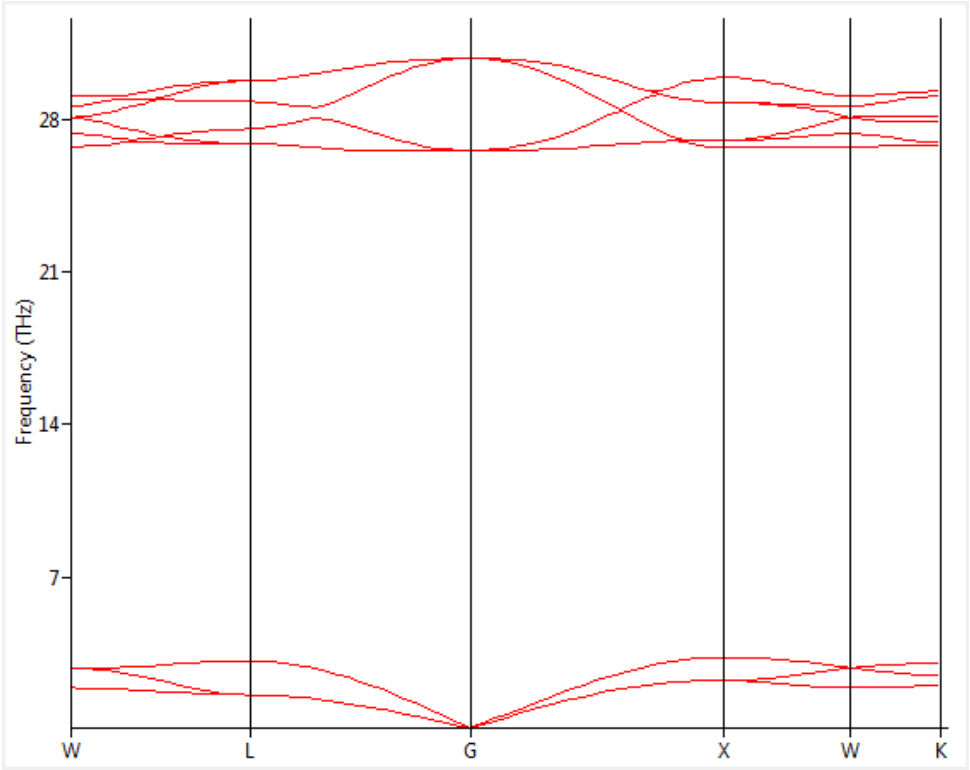
- DFT structure optimization using VASP (*Vienna Ab-Initio Simulation Package*)
  - GGA exchange and correlation functional
  - Hubbard *U* parameter correction applied to Pu 5*f* electrons
    - Account for effect of strong correlation of 5*f* electrons on chemical binding of U and Pu molecules
  - Spin-polarized magnetism
  - 500 eV planewave cutoff
  - *k*-point spacing of 0.2 $Å^{-1}$ (11×11×11 k-mesh)
  - 10^{-6} eV total electronic energy threshold
- **Hubbard U = 0.6 eV reproduces the measured lattice parameter of *a* = 5.359 Å**



VASP structure optimization of PuH_2 using GGA+U.

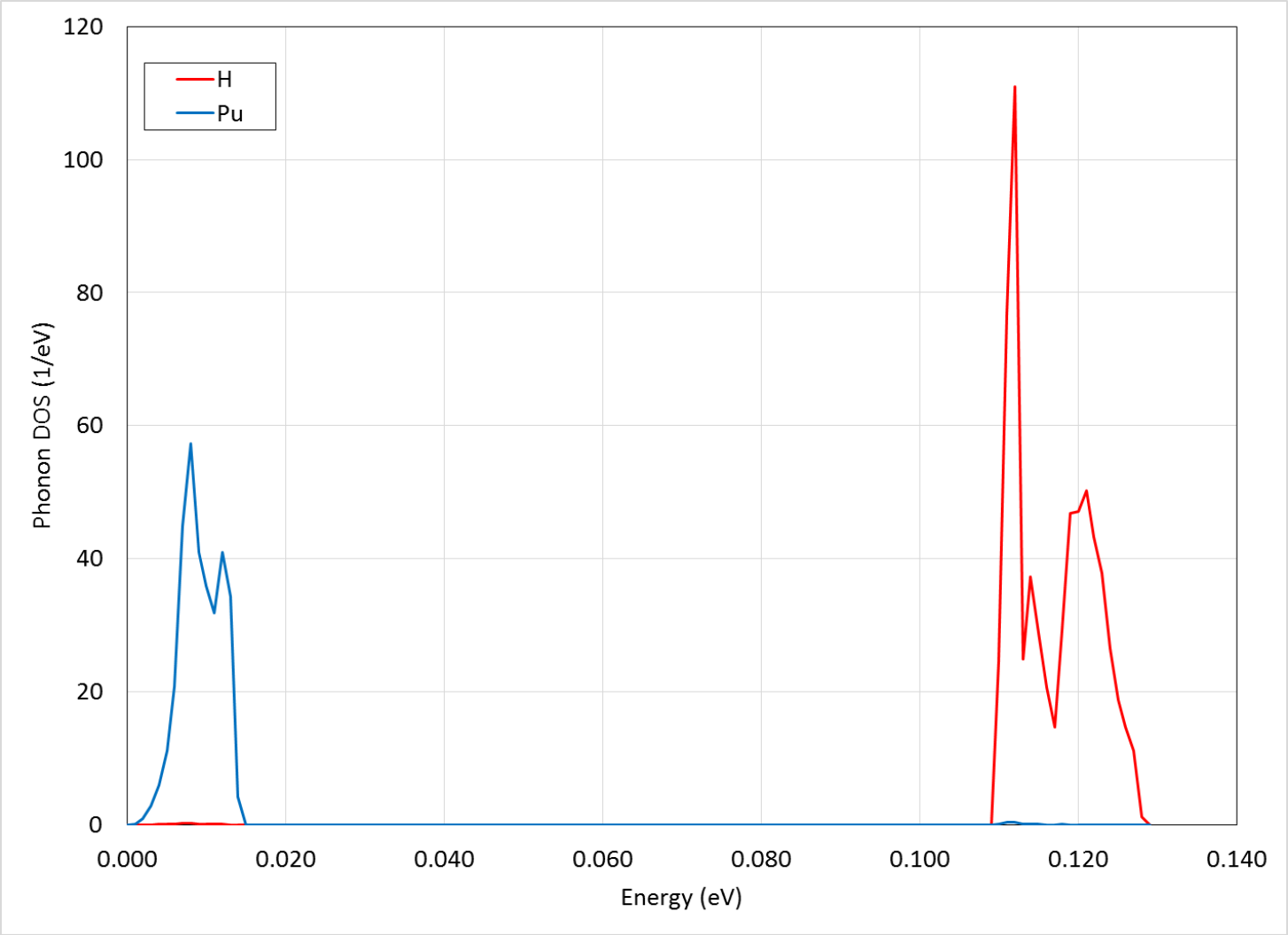
PuH_2 Lattice Dynamics

- LD calculations performed using PHONON
  - Interatomic forces calculated by VASP
  - 2×2×2 supercell (96 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 Pu atom vibrations
  - Higher branches are optical modes mainly due to light H atom vibrations
- Phonon DOS (next slide)
- No published INS measurements are available to verify calculated dispersion relations & phonon DOS



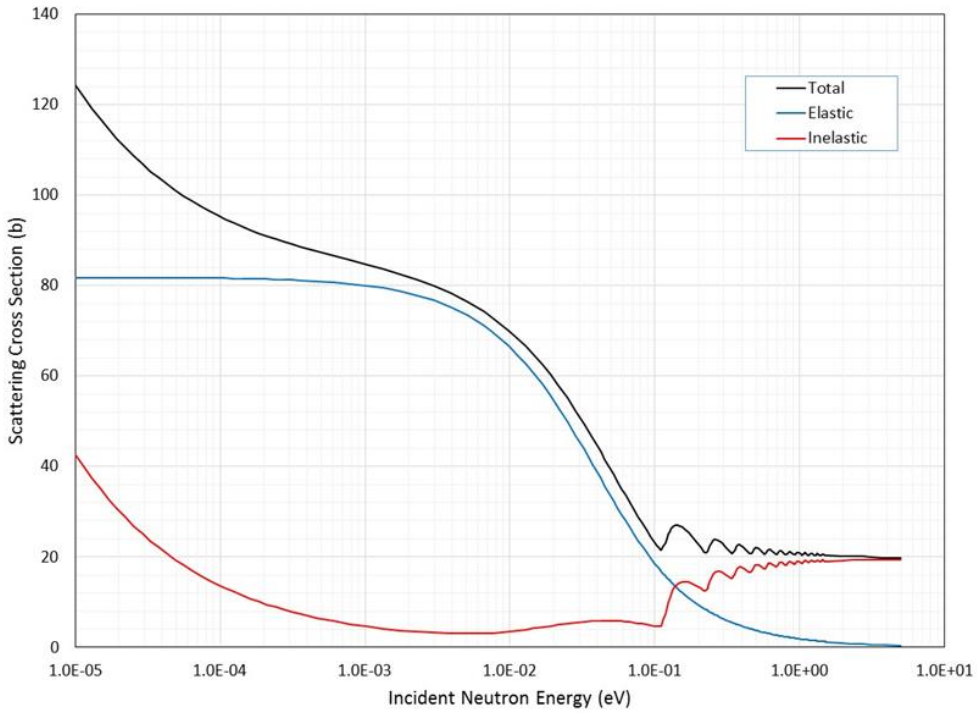
Calculated dispersion relation for PuH_2 along the highest-symmetry points of the Brillouin zone.

Calculated Phonon DOS for PuH_2



H-PuH_2 TSL Evaluation

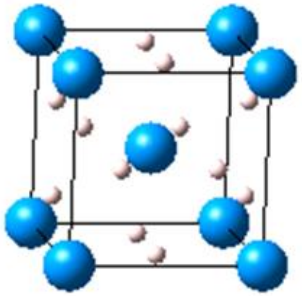
- H-PuH_2 TSL generated using NJOY/LEAPR
  - H-PuH_2 phonon DOS from PHONON calculation
  - Incoherent approximation
  - Atomic mass ratio and free atom scattering cross section for ^{1}H from ENDF/B-VII.1
  - α and β grids optimized to treat scattering up to 5 eV without SCT approximation
  - Temperature evaluated at 293.6 K (room temp.)
- Pu-PuH_2 TSL not evaluated at this time
  - LEAPR can't properly treat Pu coherent elastic scattering in PuH_2 without extensive modifications
  - Deferring evaluation of Pu-PuH_2 until new beta version of FLASSH code is available
  - Proper treatment of coherent elastic scattering and relax incoherent approximation
  - Eventually account for nuclear excitations
- Use free gas approximation for Pu (interim)
  - Small approximation for PuH_2 since H scattering dominates



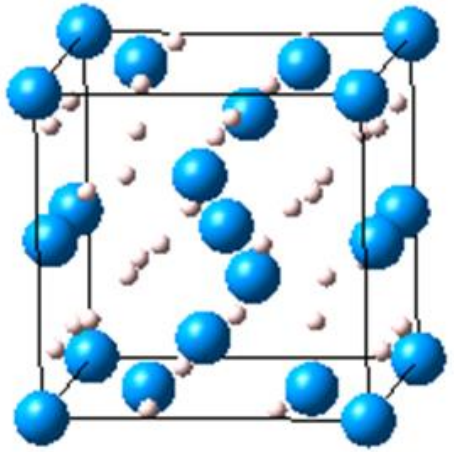
Total, elastic, and inelastic scattering cross section for H-PuH_2 at 293.6 K generated by NDEX

UH_3 Structure

- UH_3 has two allotropes, both cubic
  - Pm3n symmetry group
- α-UH_3 metastable, only found at low (cryogenic) temps
  - 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 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 (DOS)
  - H-UH_3 TSL evaluated in incoherent approximation using NJOY/LEAPR



α-UH_3 Unit Cell

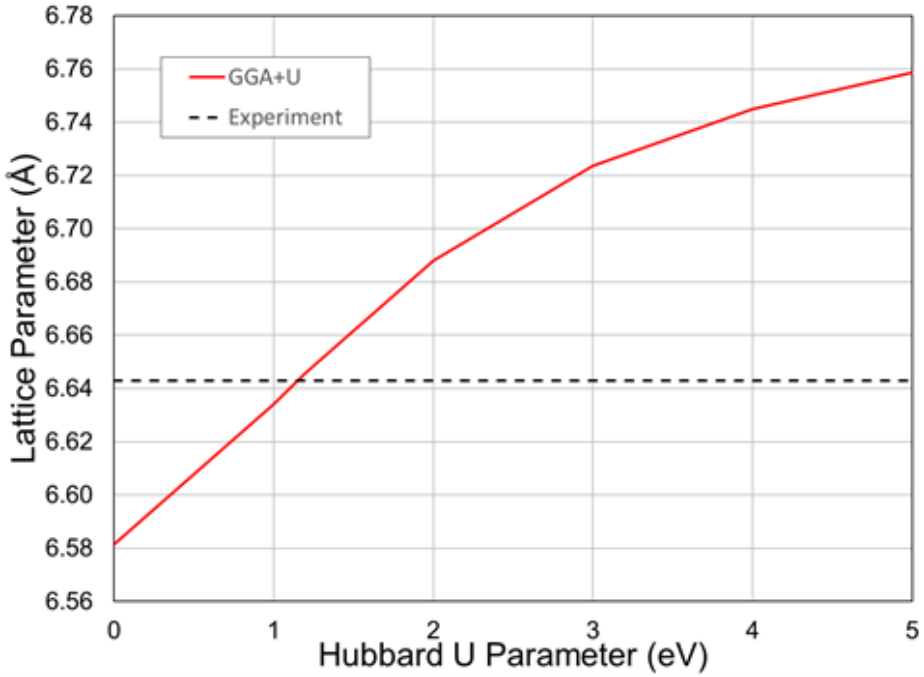


β-UH_3 Unit Cell

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 and Pu 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
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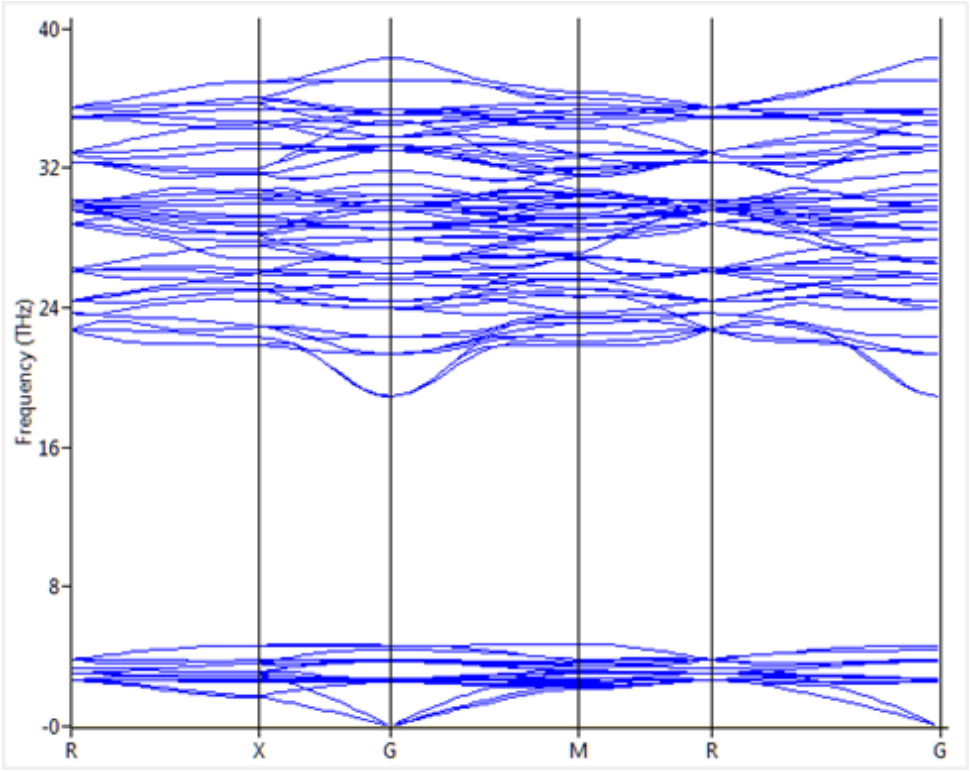
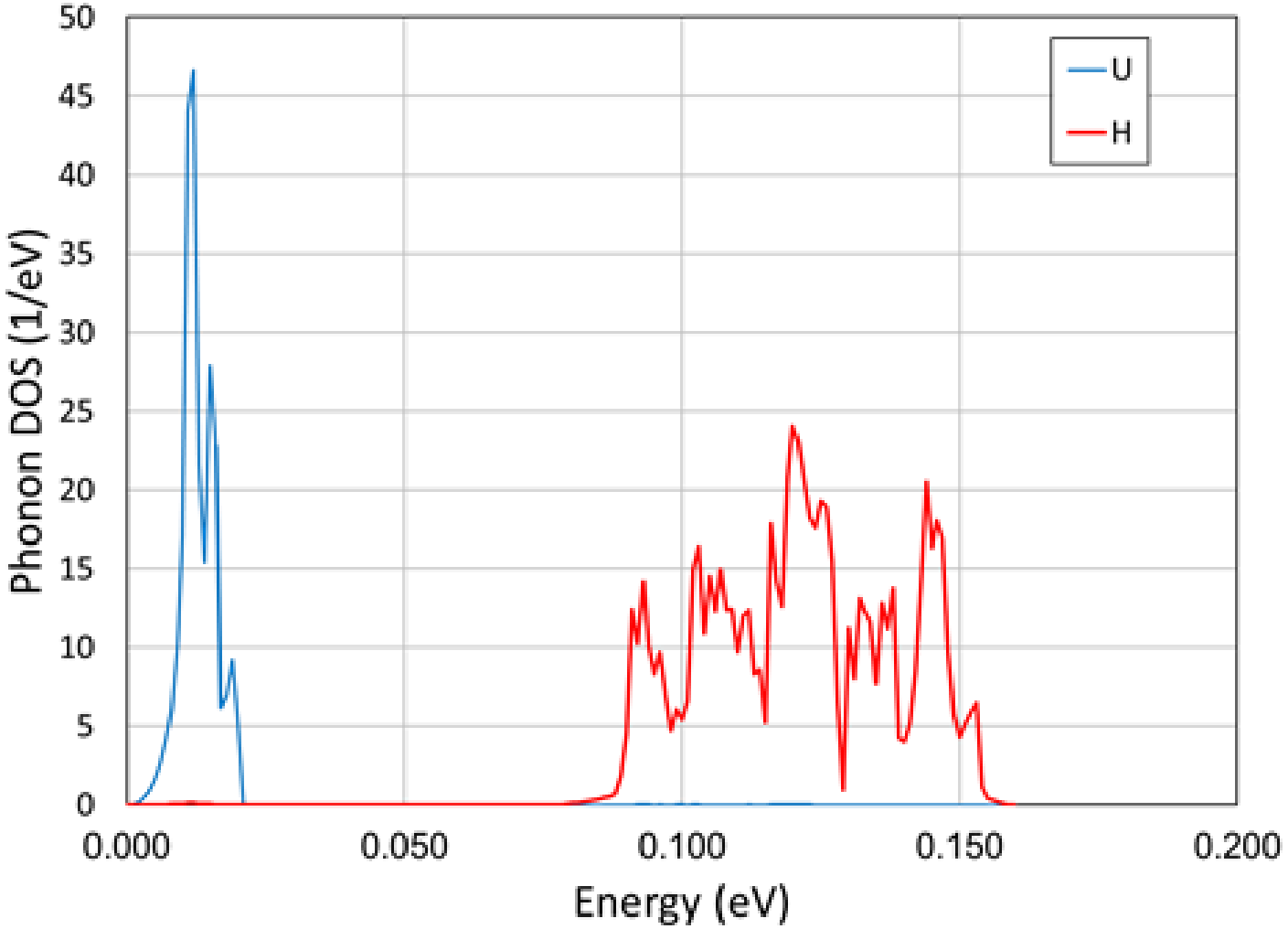
  - 2×2×2 supercell (256 atoms)
  - ±0.02 Å atom displacements
  - 0.02 $Å^{-1}$ *k*-point spacing (3×3×3 *k*-point mesh)
- Dispersion relations (at right)
  - Well separated acoustic and optical modes
  - Lower branches are acoustic modes mainly due to heavy U atom vibrations
  - Higher branches are optical modes mainly due to light H atom vibrations
    - Relatively wide and dense optical mode
- Phonon DOS (next slide)
- UH_3 optical peak measured by Glogolenko *et al.* consistent with calculated phonon DOS

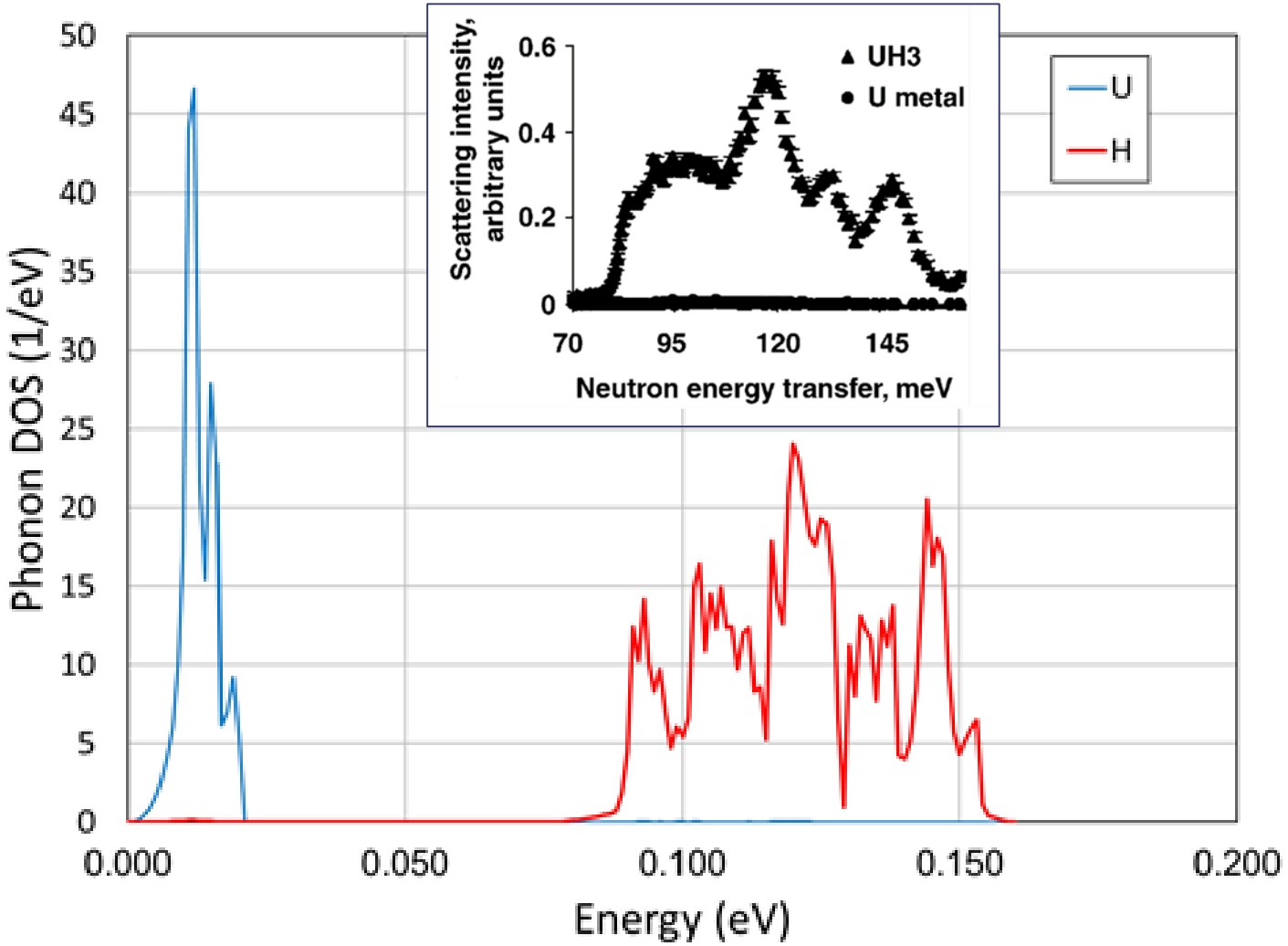


Calculated dispersion relation for UH_3 along the highest-symmetry points of the Brillouin zone.

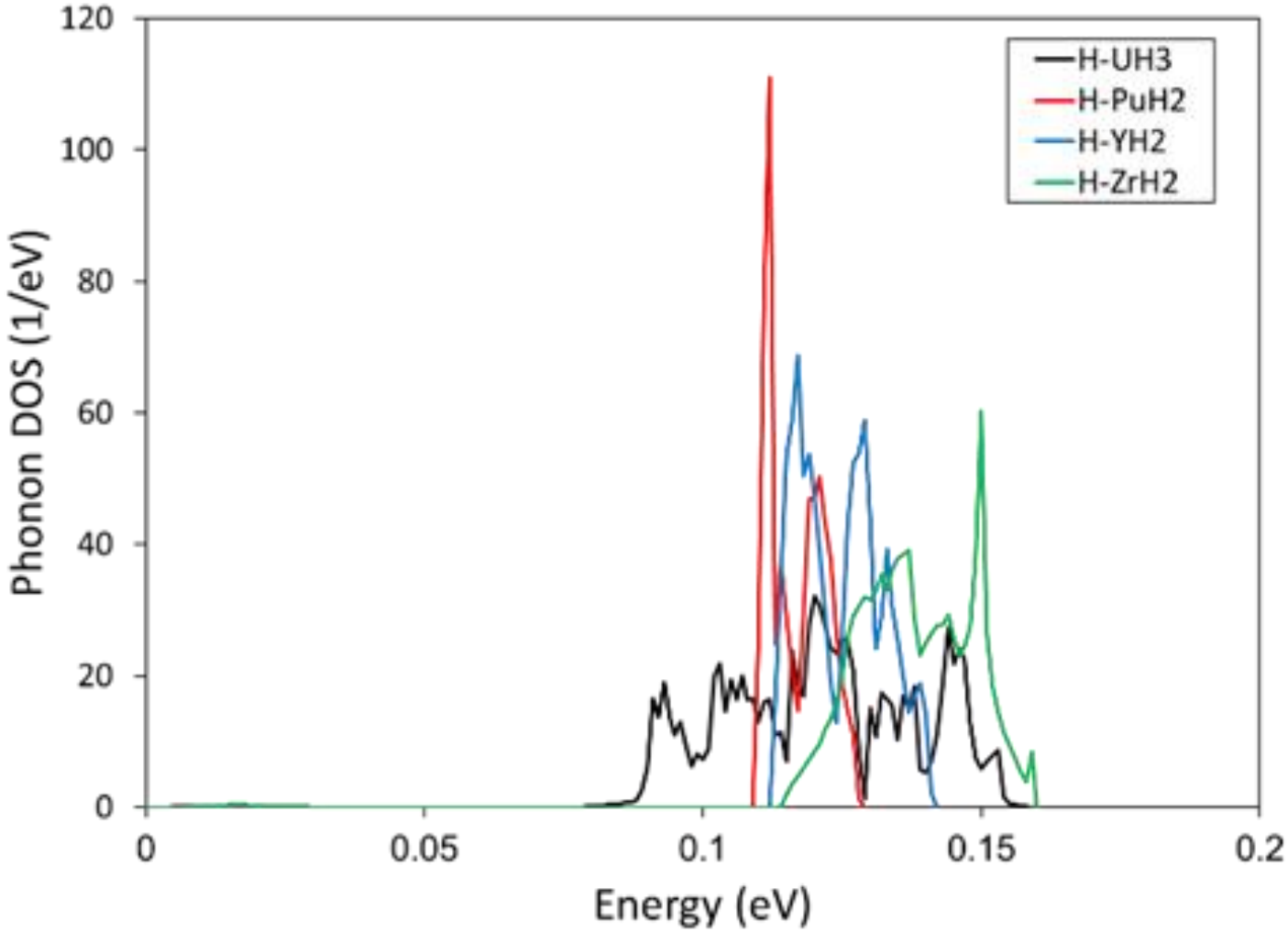
Calculated Phonon DOS for UH_3



Calculated Phonon DOS for UH_3



Comparision of H Phonon DOS for Several Metal Hydrides



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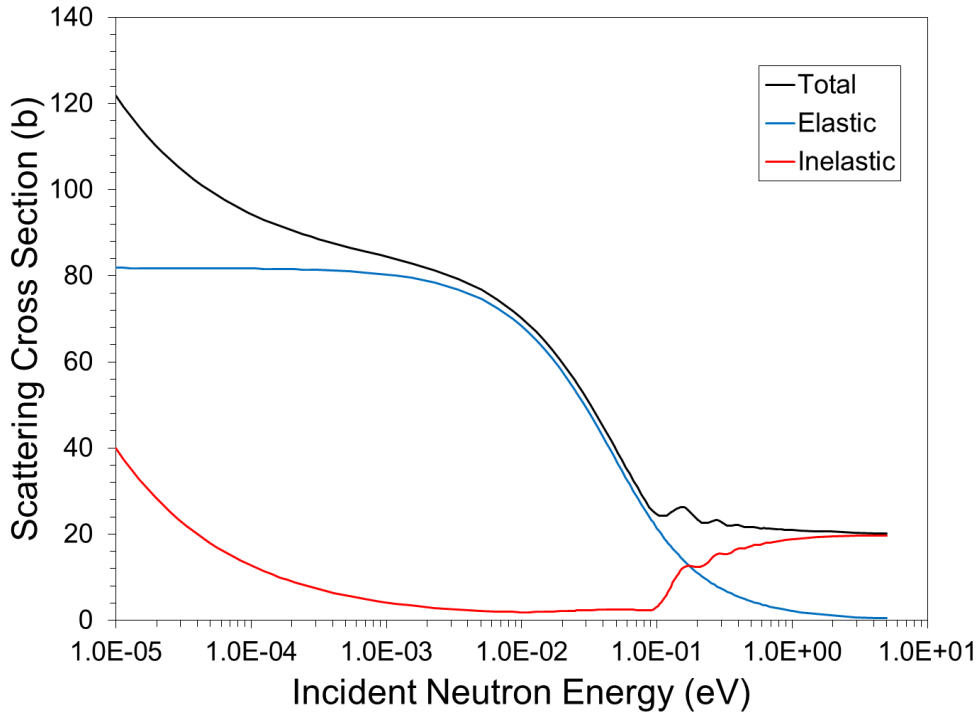
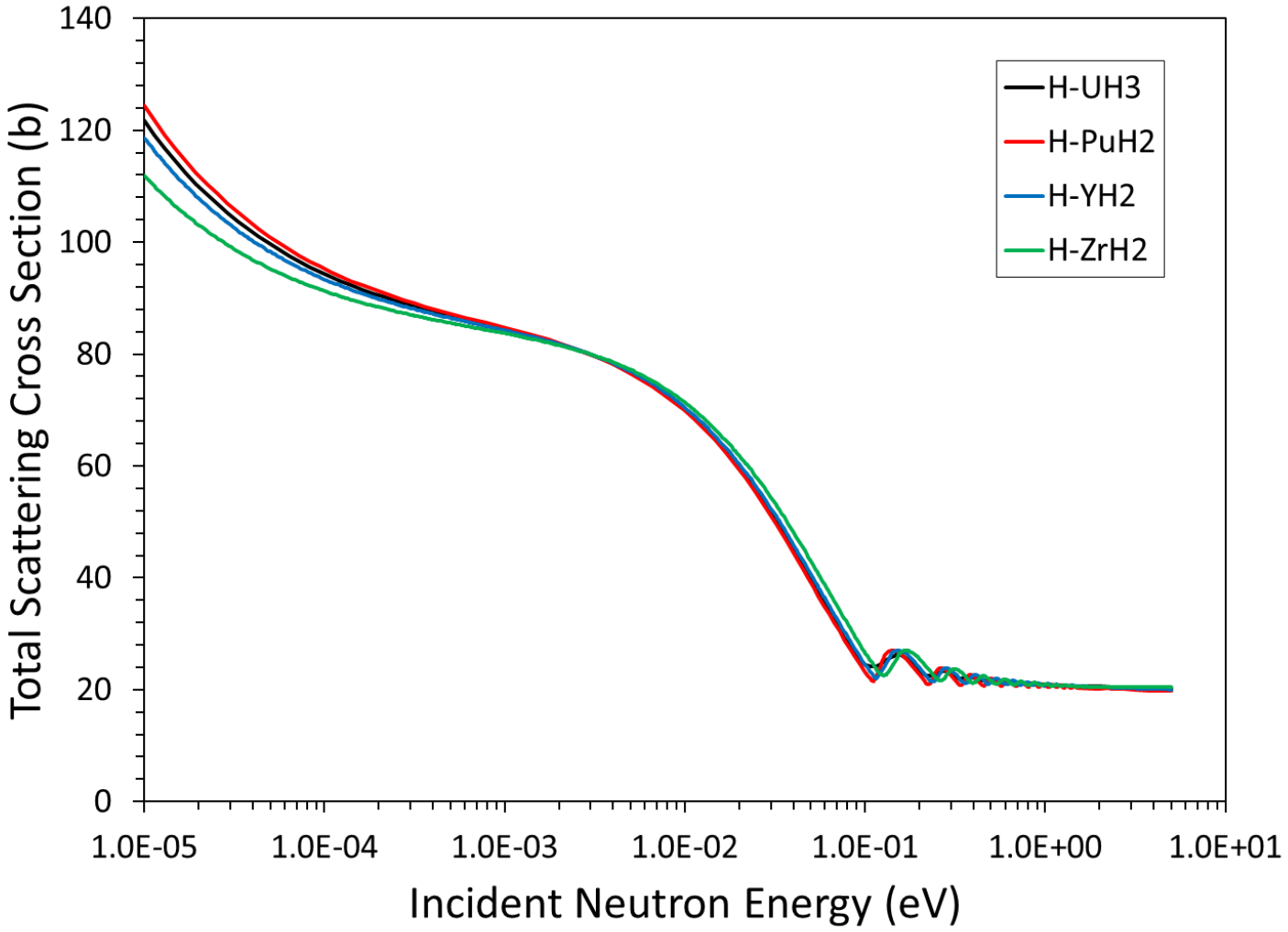
  - Plan to evaluate U-UH_3 using FLASSH
  - Proper treatment of coherent elastic scattering and relax incoherent approximation



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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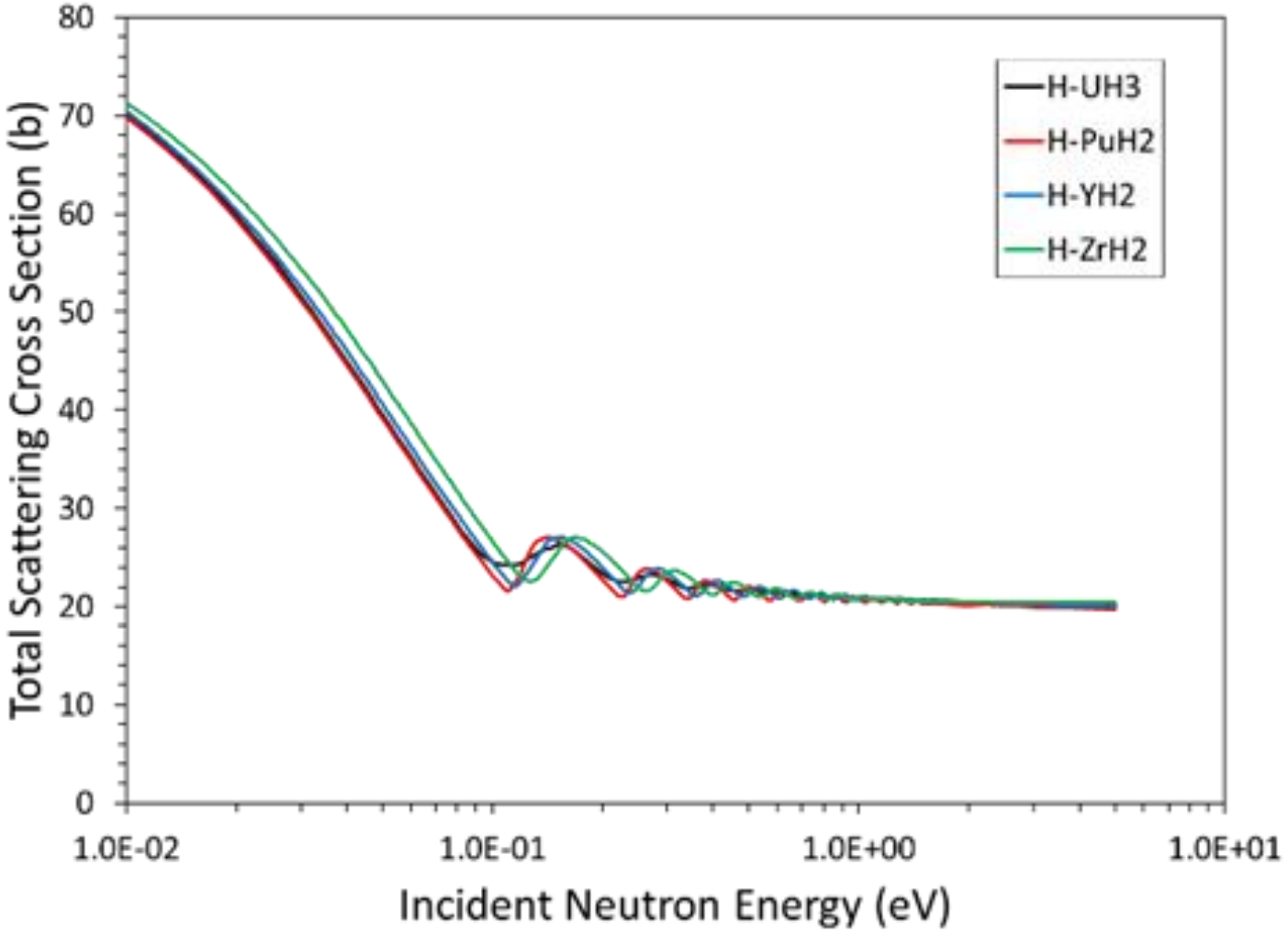

Summary

- NNL transitioning from NJOY/LEAPR to FLASSH based TSL evaluation methodology to leverage improved physics
- TSL Evaluations in progress for ENDF/B-VIII.1
  - H-PuH_2
  - H-UH_3
- Additional TSL evaluations being considered for ENDF/B-VIII.1
  - Y-YH_2 (improve elastic scattering treatment)
  - U-UH_3
  - H-PuH_{2+x}
  - Pu-PuH_2 (maybe)
  - Pu-PuH_{2+x} (maybe)