

A Thermal Neutron Scattering Law for Yttrium Hydride

Michael Zerkle and Jesse Holmes
Bettis Atomic Power Laboratory

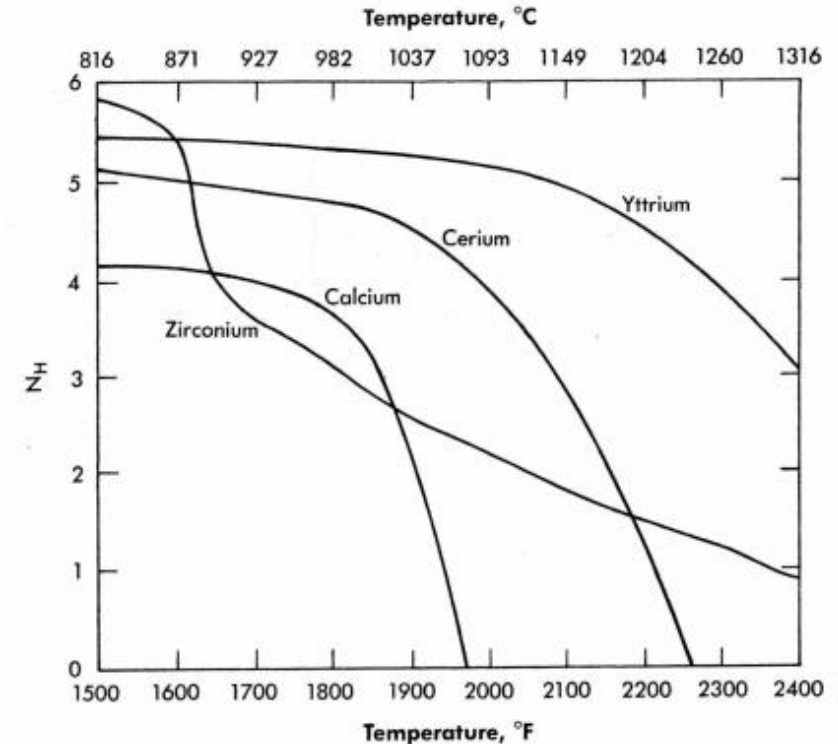
International Conference on Nuclear Data for Science and Technology (ND2016)
September 11-16, 2016 Bruges, Belgium

The Naval Nuclear Laboratory is operated for the U.S. Department of Energy and the U.S. Department of the Navy by Bechtel Marine Propulsion Corporation,
a wholly owned subsidiary of Bechtel National, Inc.



Background

- Metal hydrides (ZrH_x) used as moderator in nuclear reactors
- Yttrium hydride (YH_x) has superior hydrogen density (N_H) at elevated temperatures
- Keinert (1971) proposed H- YH_2 TSL based on simple analytic frequency distributions
 - Debye-type for acoustic mode
 - Gaussian-type for optical mode
- In this work we develop TSLs for H- YH_2 and Y- YH_2 using first-principles calculations
 - 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)



Hydrogen in metallic Zr, Ce, Y and Ca in equilibrium with 1 atm H_2 at various temperatures.

(Source: *Metal Hydrides*, Academic Press, p. 442, 1968)

N_H is number of hydrogen atoms/cc x 10^{-22}

Yttrium – Hydrogen Phase Diagram

- H has limited solubility in α -Y at low temperatures
- $H/Y < 2.0$ is region of practical interest for nuclear applications
 - Mixture of α -Y + YH_2 phases at low temperatures (next slide)
 - YH_{2-x} at elevated temperatures
- YH_2 phonon DOS needed to develop TSL for YH_x

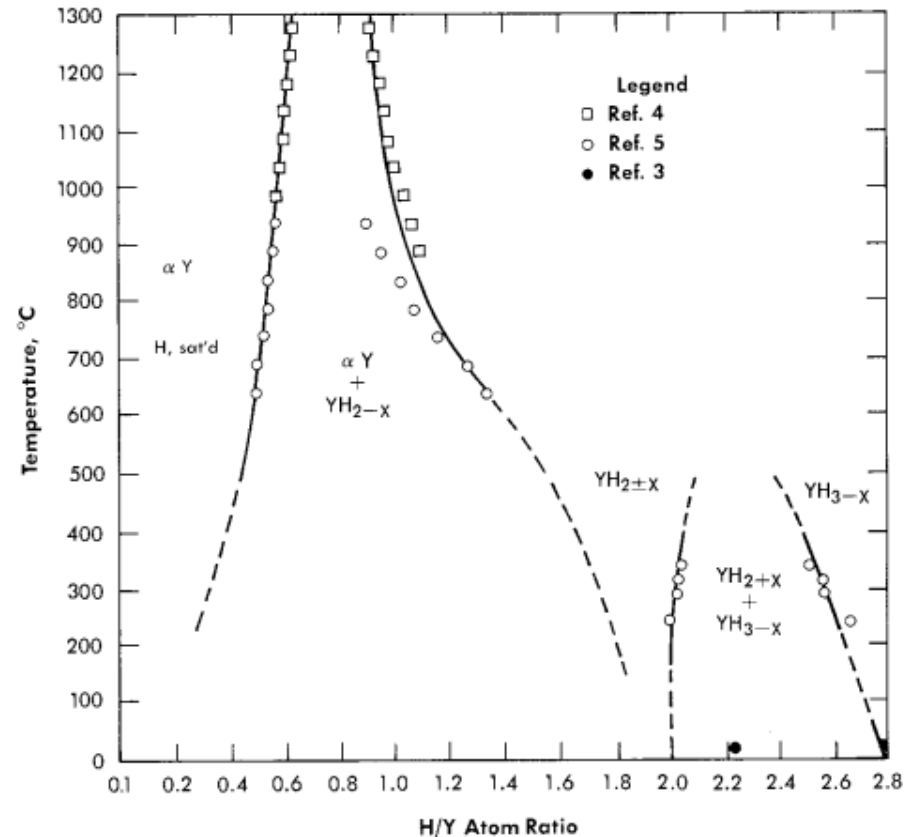
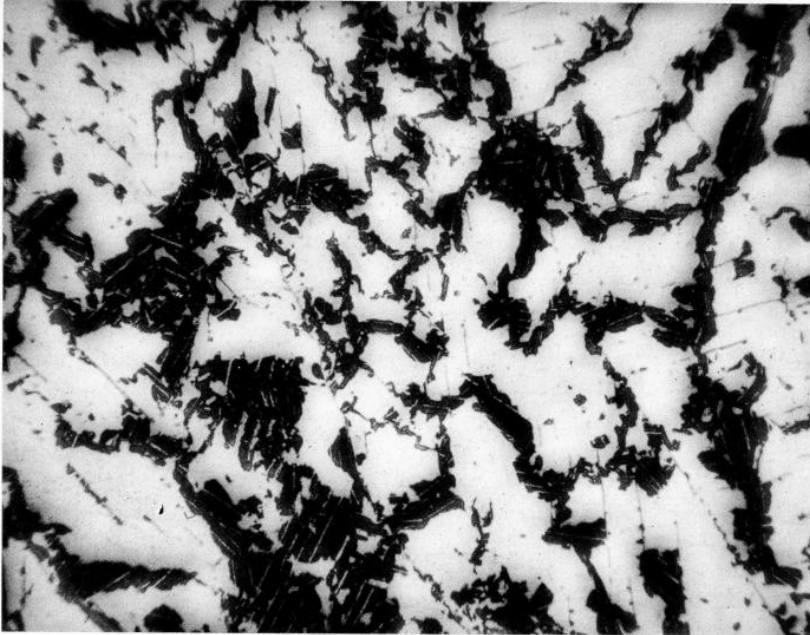


FIG. 10.2 Partial phase diagram of the yttrium-hydrogen system.

(Source: *Metal Hydrides*, Academic Press, p. 443, 1968)

Microstructure of Yttrium Hydride



Y-1.29% H alloy ($H/Y = 1.15$). Structure consists of 50% α -yttrium solid solution (dark regions) and 50% hydride phase (light regions)

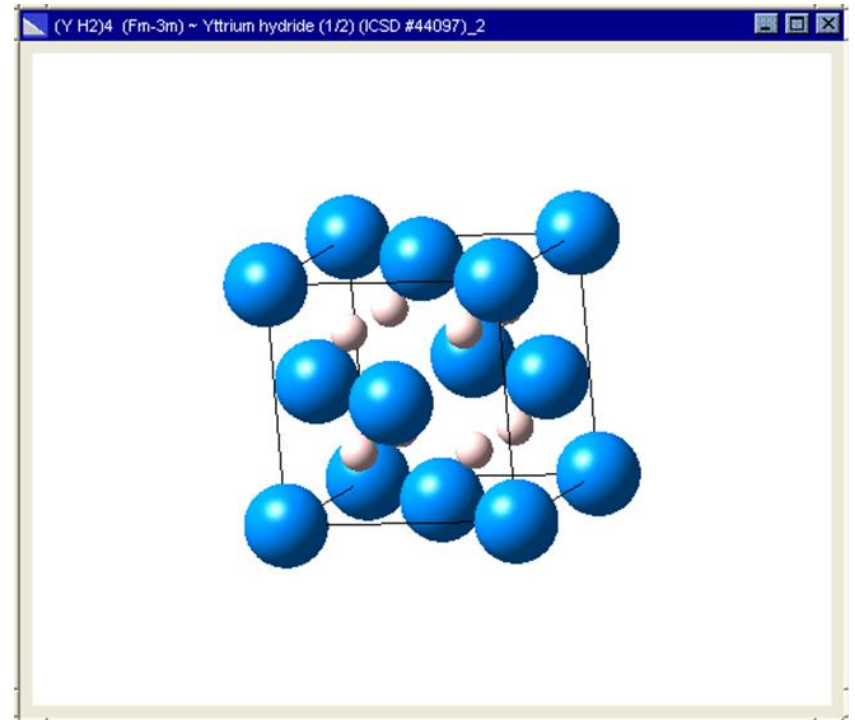


Y-2.17% H alloy ($H/Y = 1.96$). The structure is nearly all hydride phase.

(Source: *Metal Hydrides*, Academic Press, pp. 447-448, 1968)

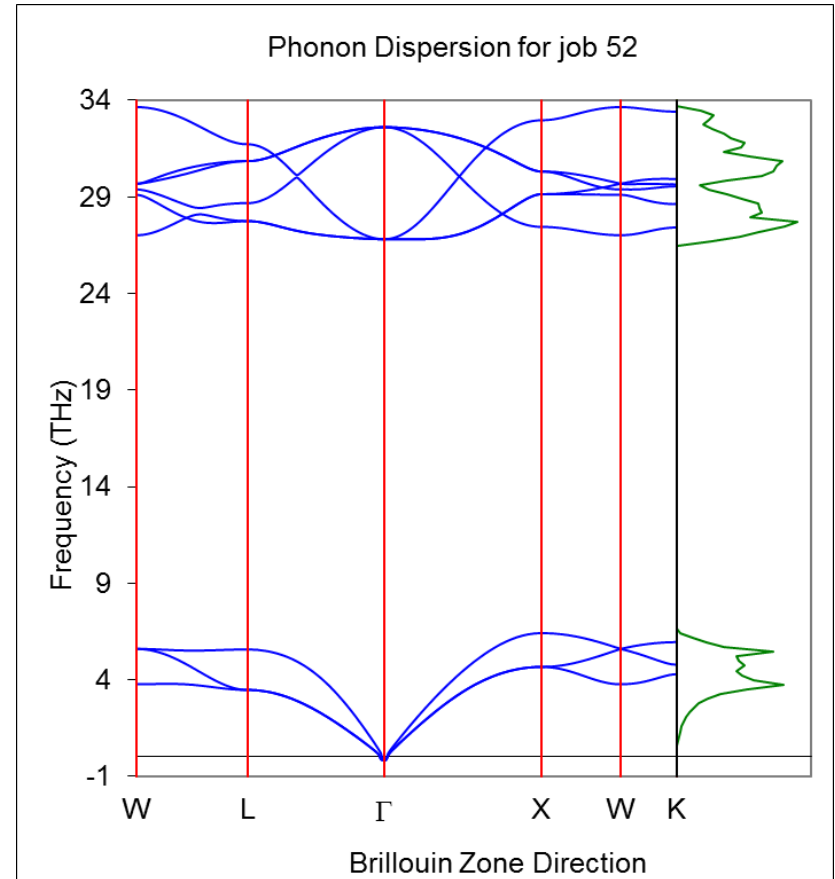
YH₂ Crystal Structure and Optimization

- YH₂ has a CaF₂ type FCC structure
 - 12 atoms
 - 4 Y atoms (blue) at vertices and faces of unit cell
 - 8 H atoms (grey) in tetrahedral holes between Y atoms
- DFT structure optimization using VASP
 - 10⁻⁶ total energy threshold
 - 0.11 1/Å k-point spacing (19×19×19 k-point mesh)
 - 500 eV planewave cutoff energy
 - GGA-PBE pseudopotential
- VASP optimized lattice constant at 0K is $a = 5.2032 \text{ \AA}$
- In excellent agreement with Daou & Vajda X-ray diffraction measurement of $5.2032 \pm 0.003 \text{ \AA}$ at 90 K

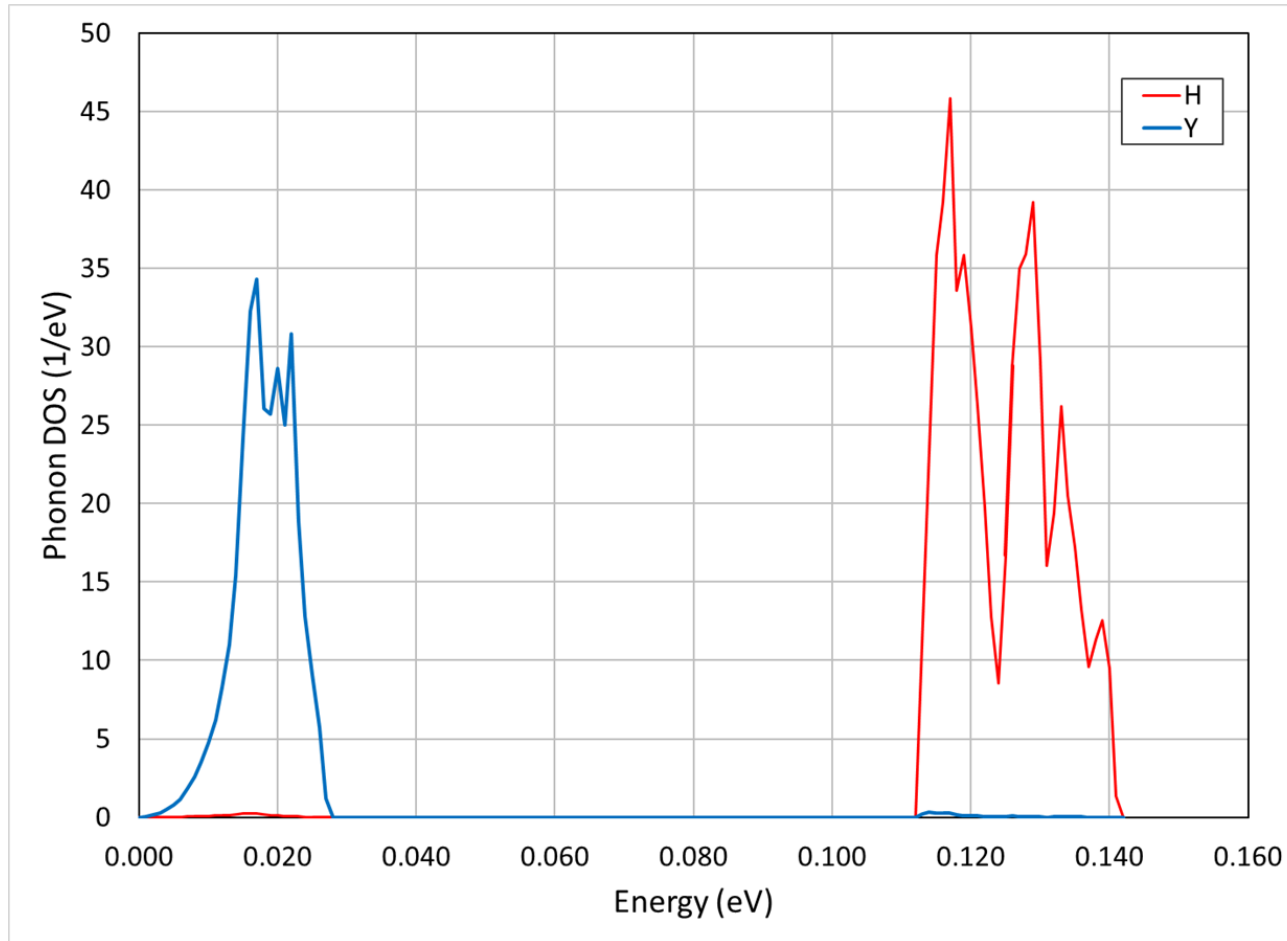


YH₂ Lattice Dynamics

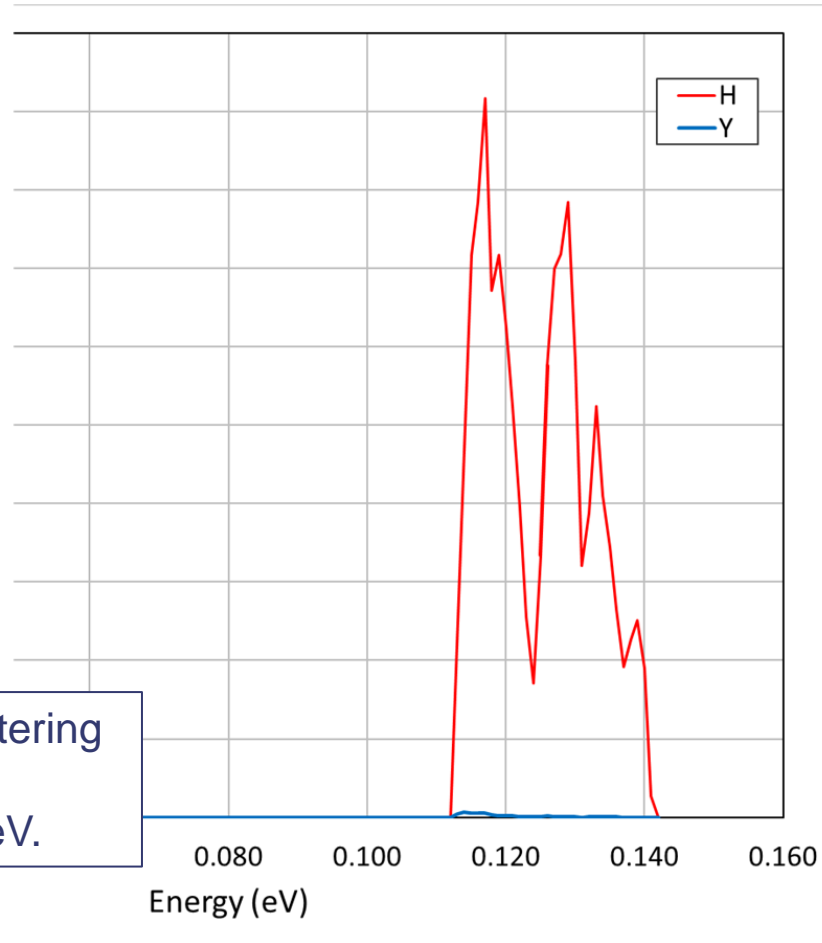
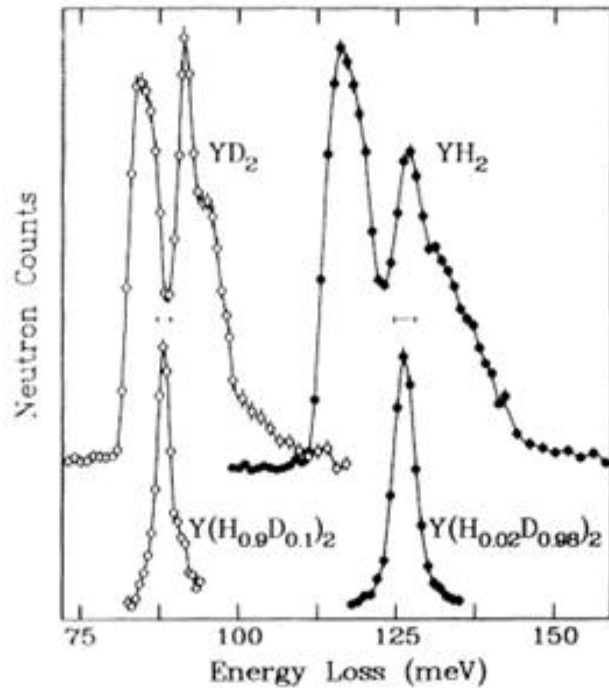
- LD calculations performed using PHONON
 - Interatomic forces calc. by VASP
 - 2×2×2 supercell (96 atoms)
 - ±0.02 Å atom displacements
 - GGA-PBE pseudopotential
 - 500 eV planewave cutoff energy
 - 0.086 1/Å k-point spacing (7×7×7 k-point mesh)
- Dispersion relations (at right)
 - Well separated acoustical and optical modes
 - Lower branches are acoustical modes mainly due to heavy Y atom vibrations
 - Higher branches are optical modes mainly due to light H atom vibrations
- Phonon DOS (next slide)



YH₂ Phonon Density of States

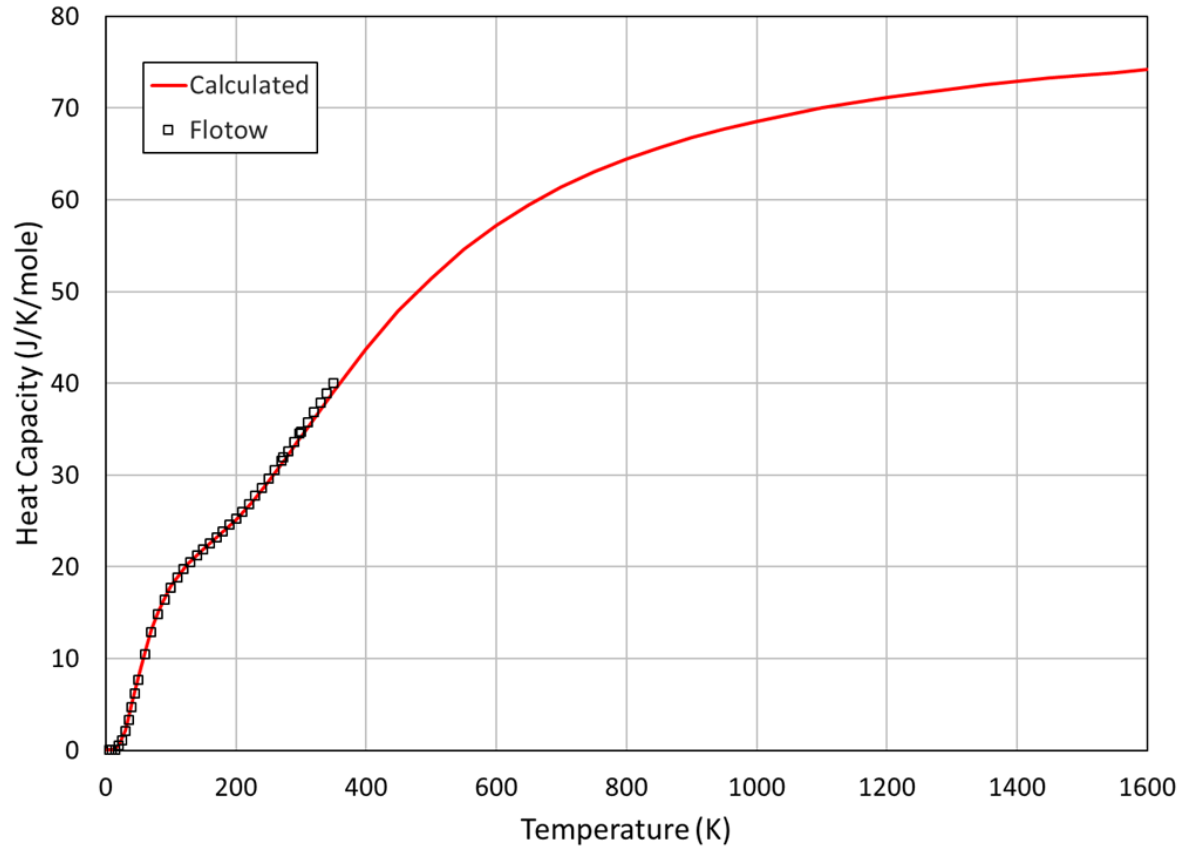


YH₂ Phonon Density of States



High resolution inelastic neutron scattering spectra from Udovic shows similar structure for YH₂ centered on 0.127 eV.

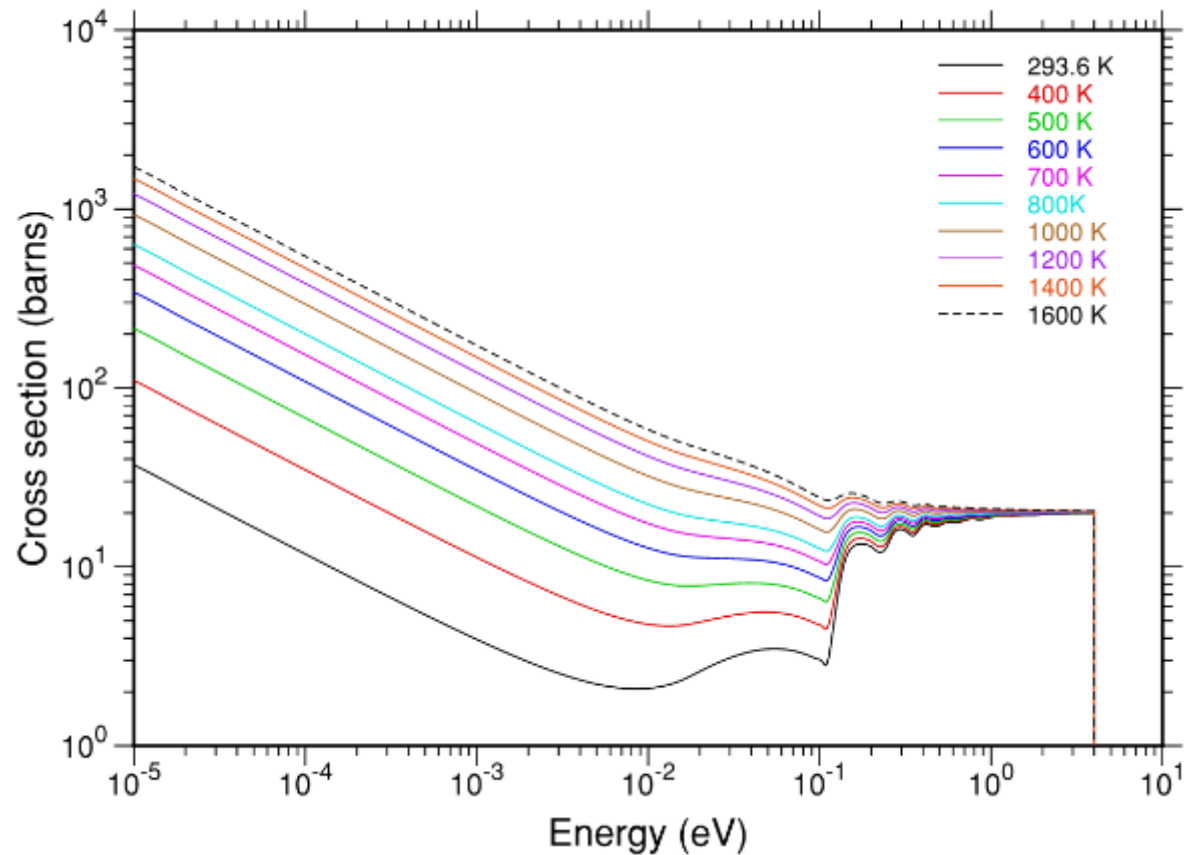
Heat Capacity Based on YH_2 Phonon DOS Agrees with Measurement



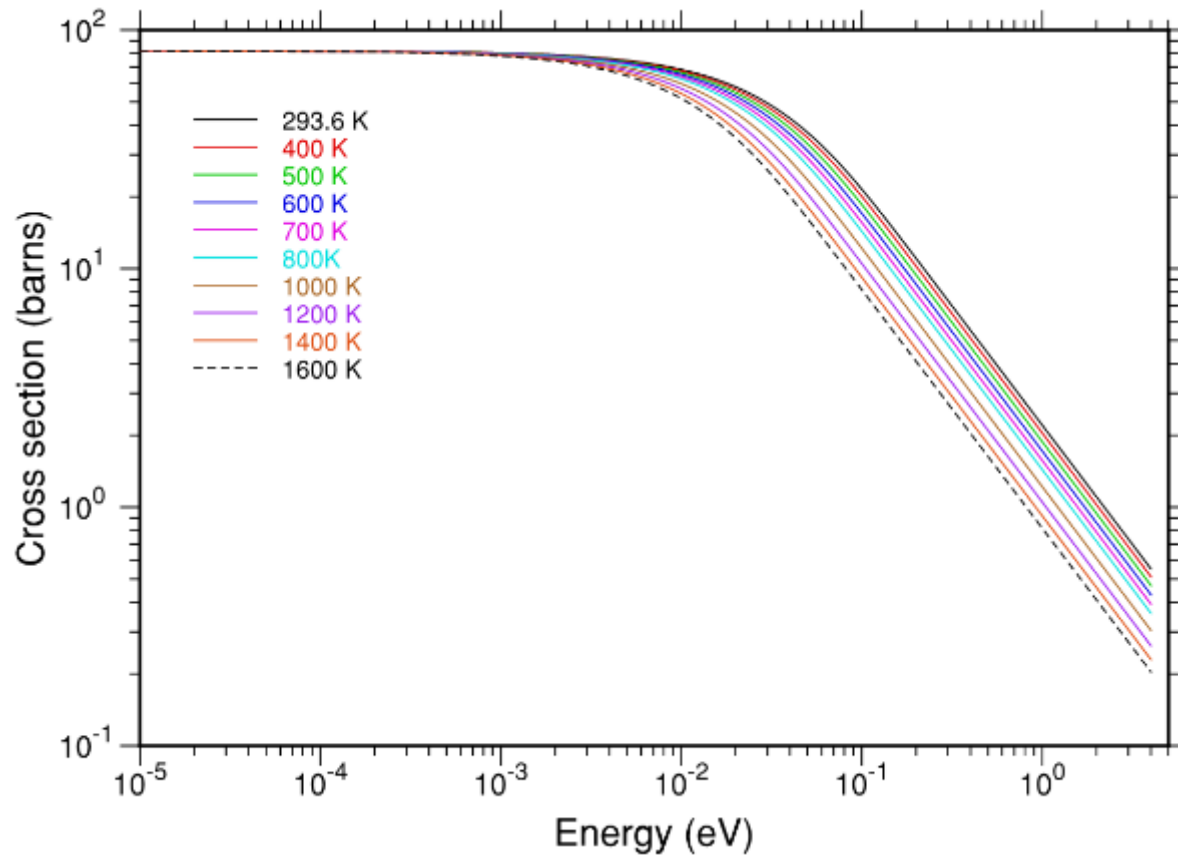
LEAPR Models of H-YH₂ and Y-YH₂ TSL

- H-YH₂ and Y-YH₂ TSLs generated using LEAPR
- Phonon DOS from PHONON calculations
- Incoherent approximation
- Atomic mass ratios and free atom scattering cross sections from ENDF/B-VII.1 ¹H and ⁸⁹Y evaluations
- α and β grids optimized to treat scattering up to 4 eV
- 10 temperatures (293.6, 400, 500, 600, 800, 1000, 1200, 1400, 1600 K)

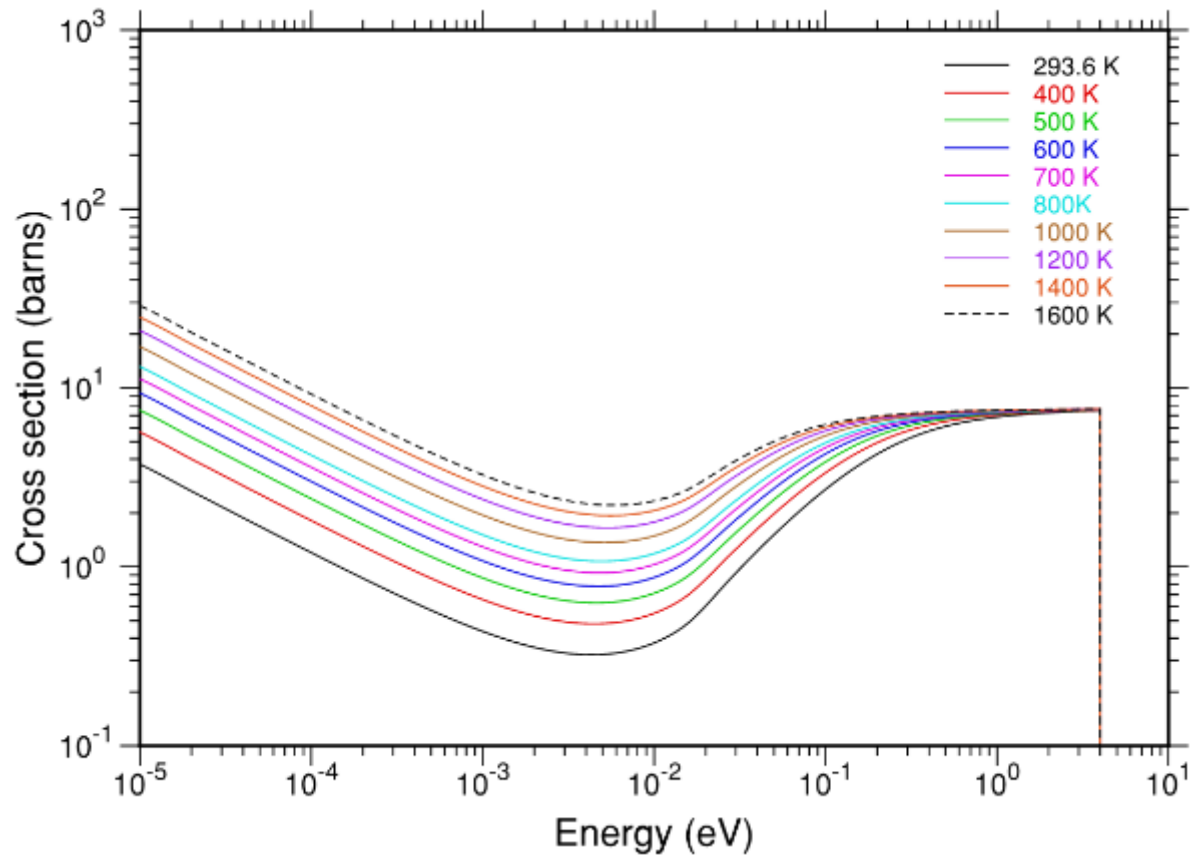
H-YH₂ Inelastic Scattering Cross Section



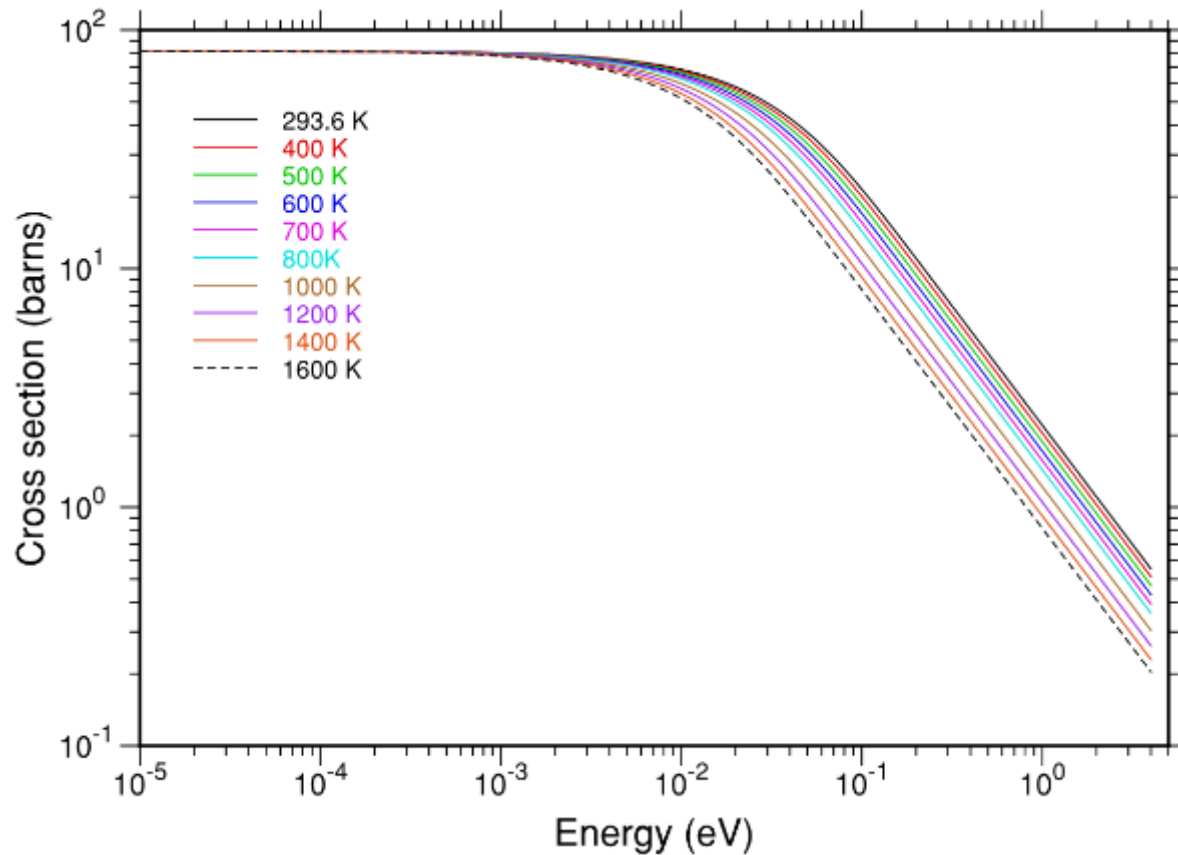
H-YH₂ Elastic Scattering Cross Section



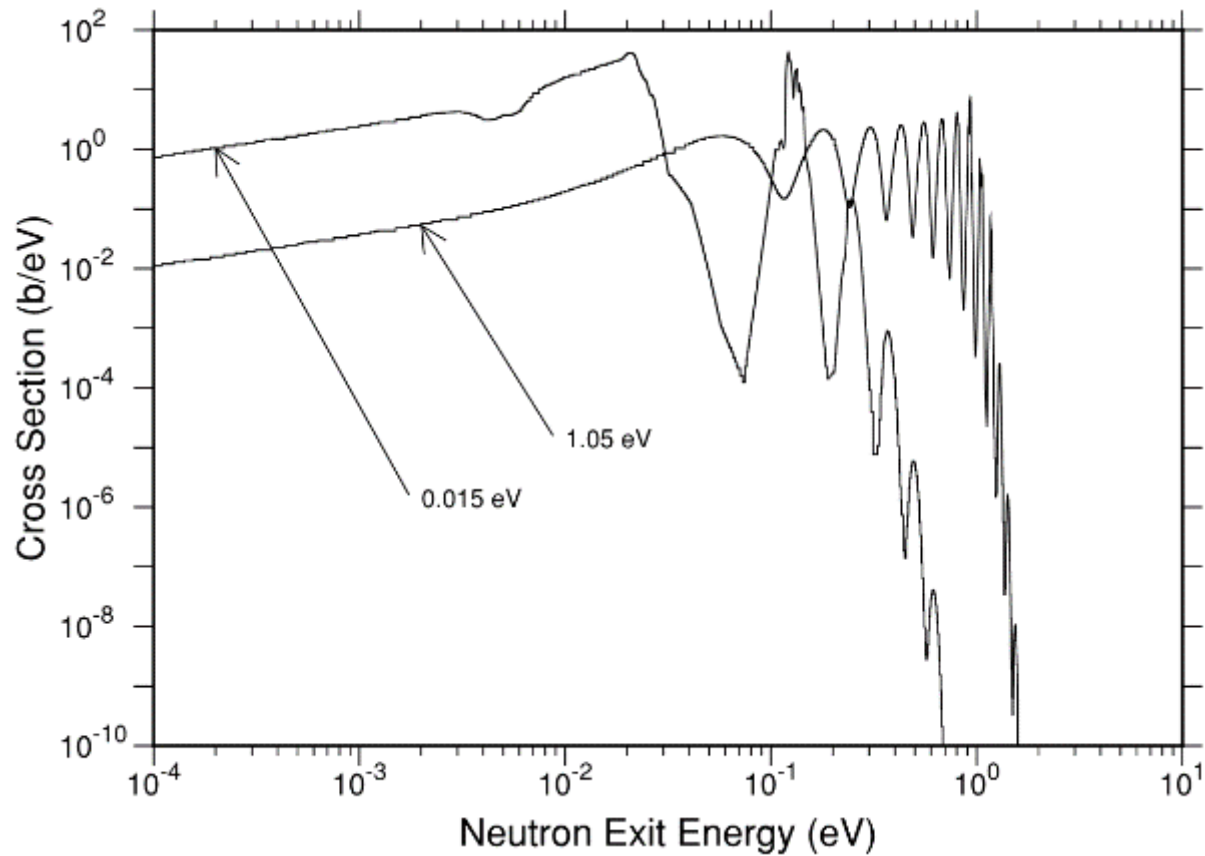
Y-YH₂ Inelastic Scattering Cross Section



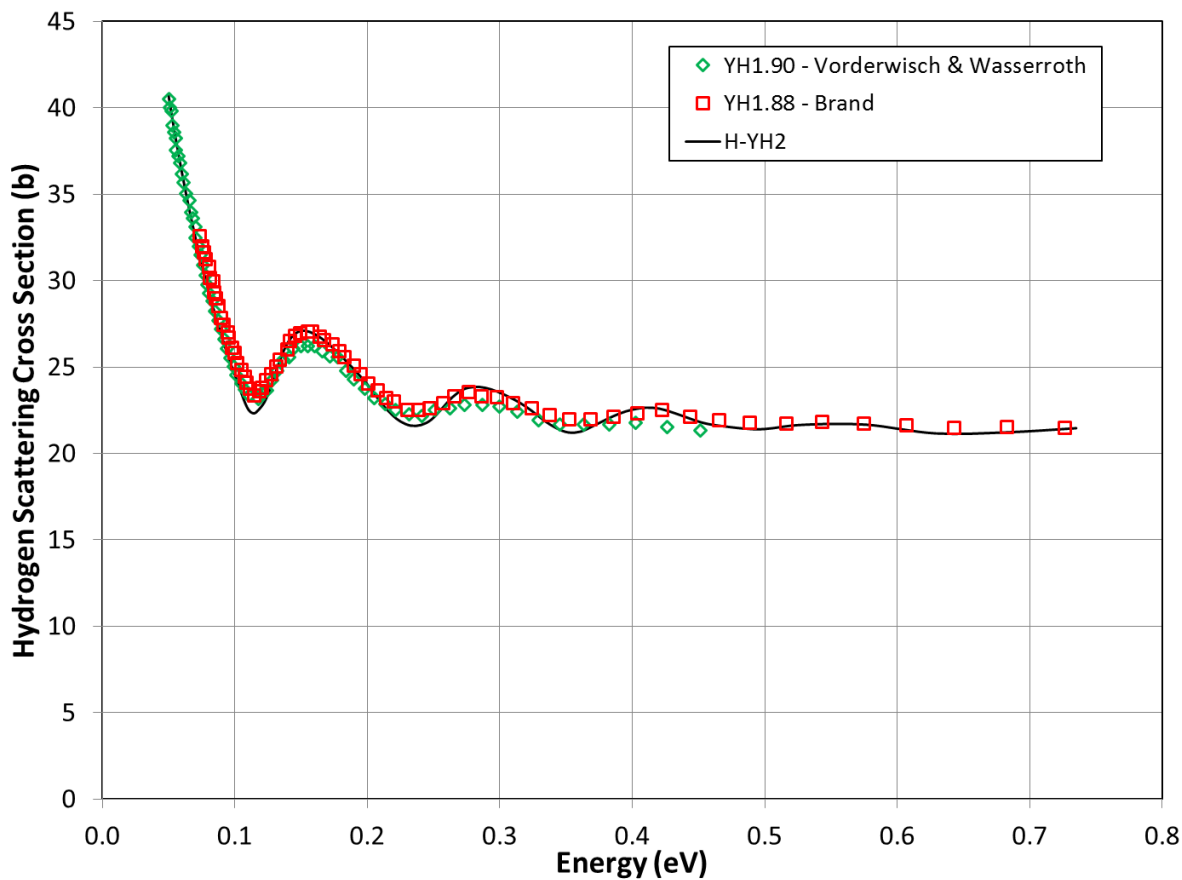
Y-YH₂ Elastic Scattering Cross Section



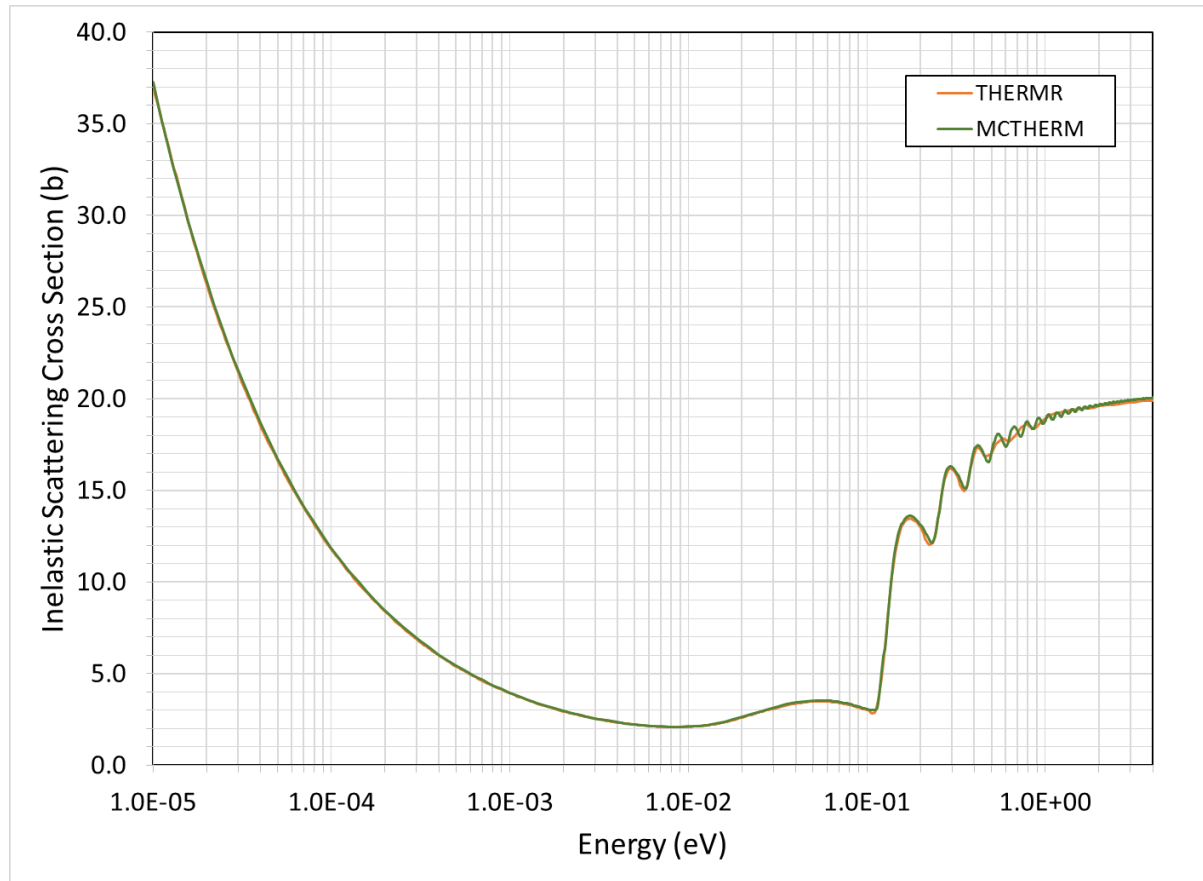
H-YH₂ Differential Inelastic Scattering Cross Section



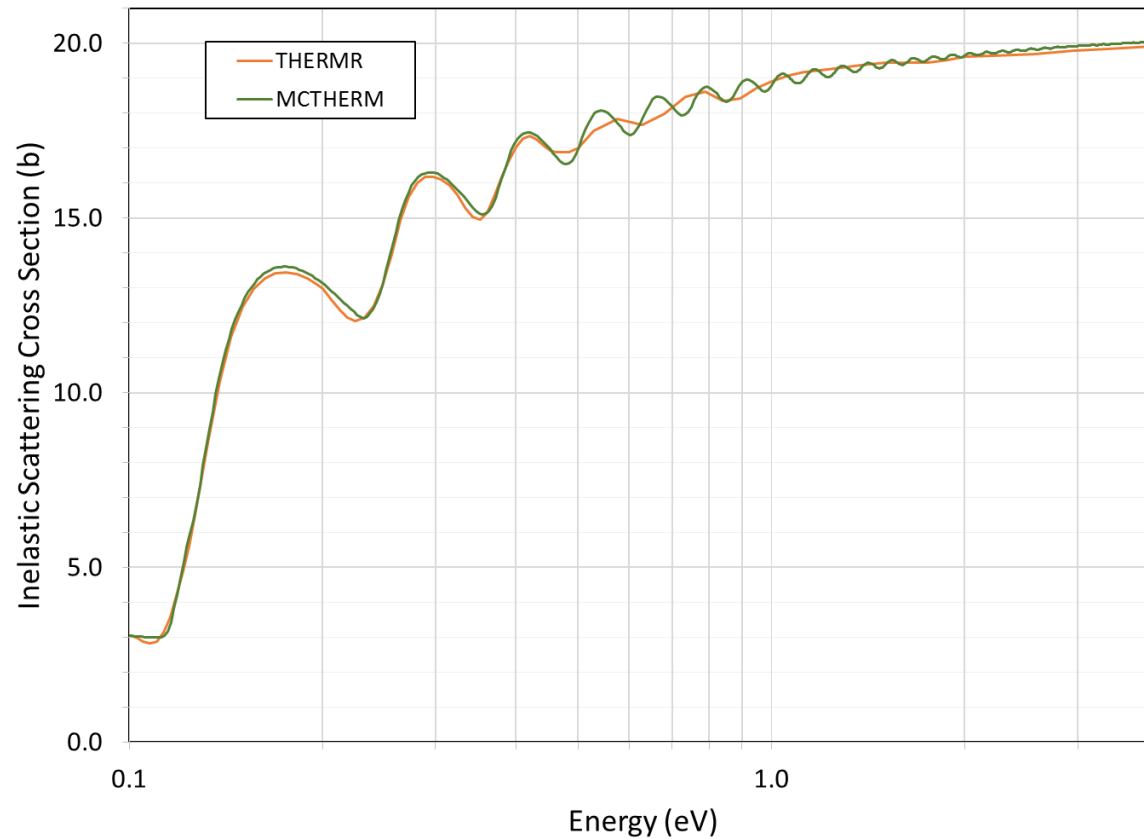
H-YH₂ Total Scattering Cross Section Agrees with Measurements



THERMR Metal Hydride Processing Issue



THERMR Metal Hydride Processing Issue



Summary

- H-YH₂ and Y-YH₂ TSLs developed using phonon DOS calculated by first principles DFT and LD calculations
- α and β grids optimized to treat scattering up to 4 eV
 - β grid can resolve 1-phonon scattering effects
- Validation
 - Measured lattice parameter
 - Phonon DOS consistent with high-resolution inelastic neutron scattering spectra
 - Measured heat capacity
 - Measured H scattering cross section for YH_{1.90} and YH_{1.88}