Generation of Thermal Scattering Laws for YH_2 using Ab Initio Methods

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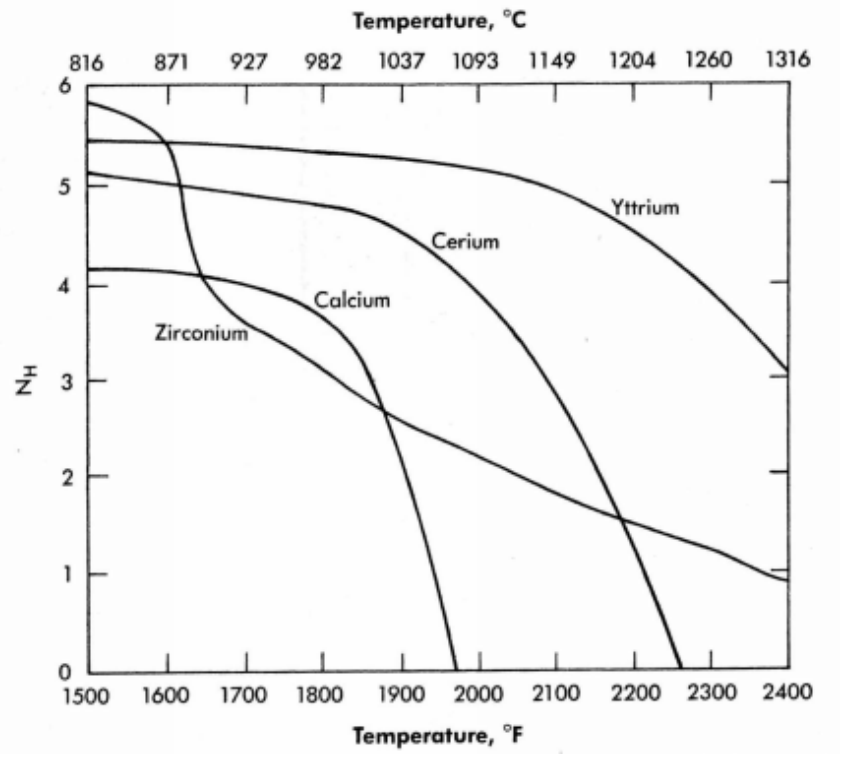
Outline

- Motivation
- Yttrium – Hydrogen Phase Diagram
- YH_2 Crystal Structure
- Ab Initio Approach using MedeA
  - Structure Optimization
  - Lattice Dynamics
- YH_2 Thermal Scattering Laws
  - Phonon Frequency Distributions
  - Inelastic Scattering Cross Section
  - Incoherent Elastic Scattering Cross Section
- Conclusions

Motivation

- Metal hydrides (ZrH_x) used as high temperature moderator/reflector in nuclear reactors
  - TRIGA
  - SNAP-10
  - TOPAZ
- Yttrium hydride (YH_x) has superior hydrogen density (N_H) at elevated temperatures
- Test improvements to MedeA software, use *ab initio* approach to generate TSLs for new moderator materials



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 lower temperatures (see next slide)
  - YH_{2-x} at elevated temperatures
- YH_2 phonon frequency distribution needed to develop thermal scattering law (TSL) for YH_{2-x}

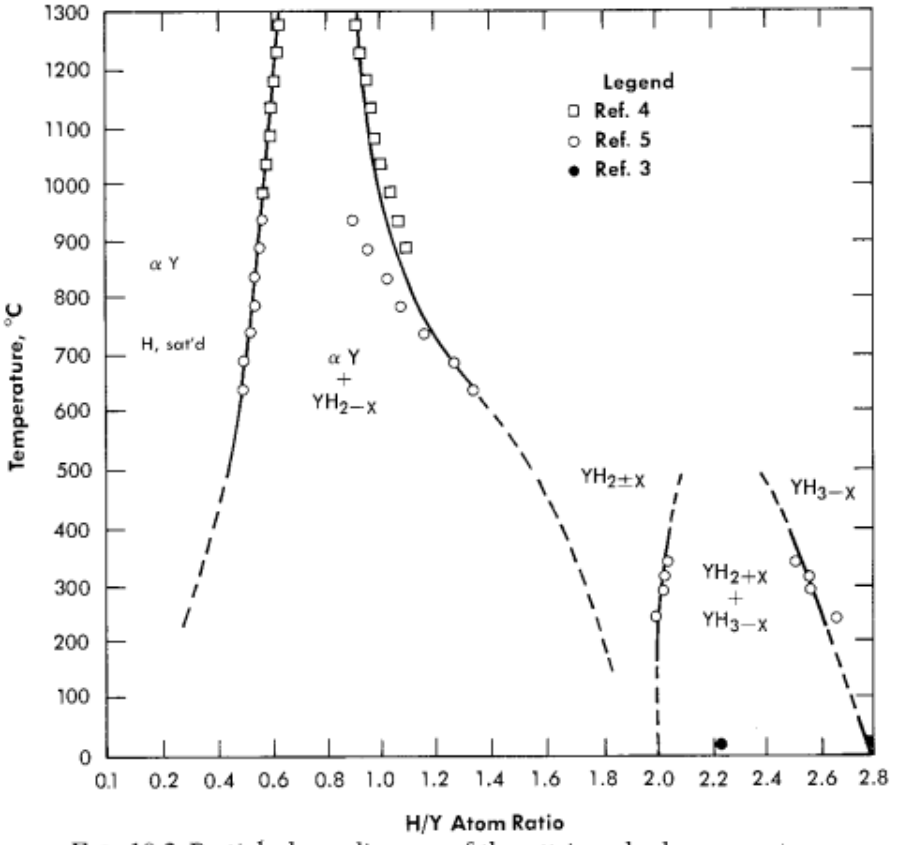
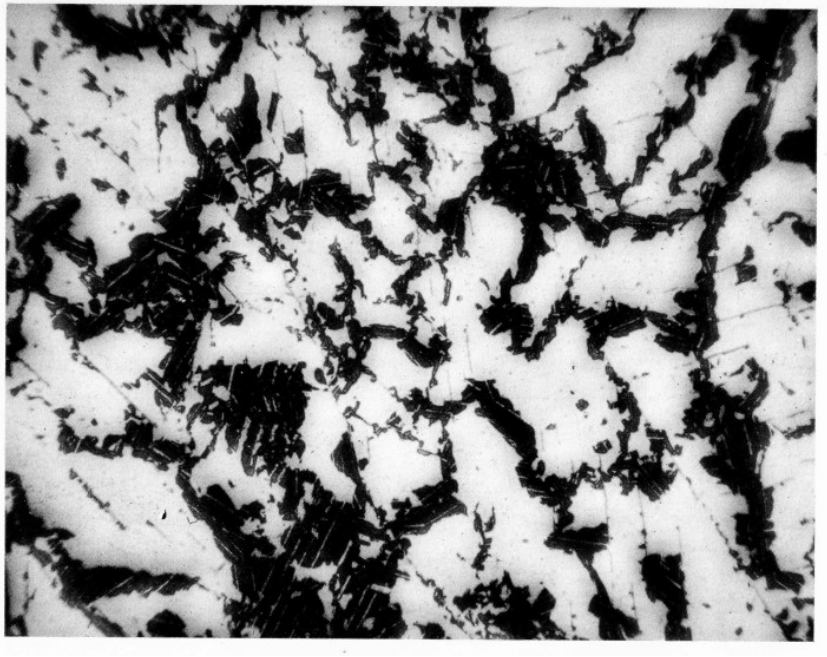


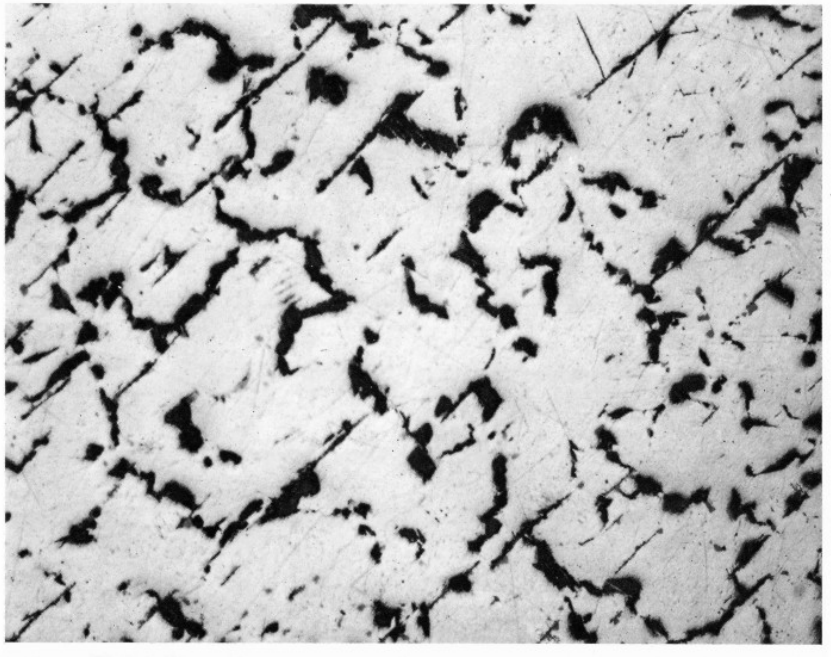
FIG. 10.2 Partial phase diagram of the yttrium–hydrogen system.

(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)

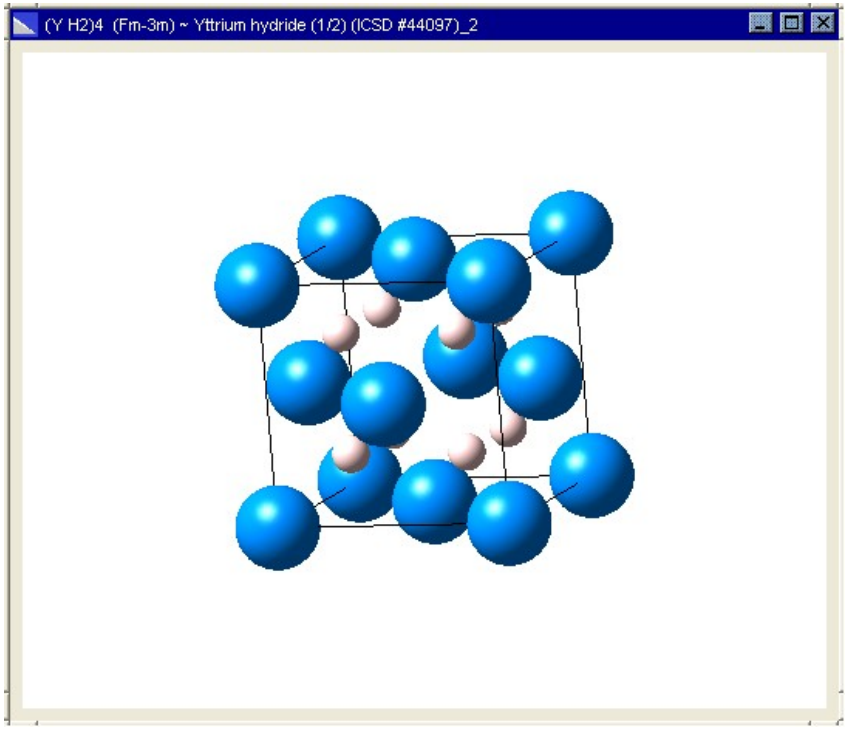


Y-2.17% H alloy (H/Y = 1.96). The structure is nearly all hydride phase.

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

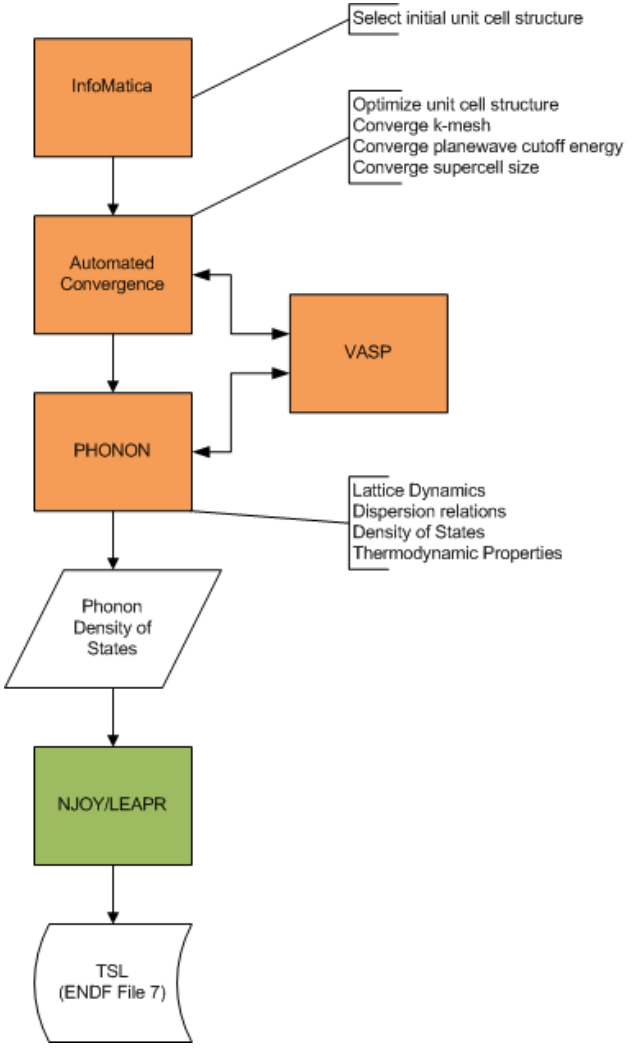
YH_2 Crystal Structure

- YH_2 has a CaF_2 type FCC structure
  - 12 atoms
  - 4 Y atoms (blue) at vertices and faces of cubic unit cell
  - 8 H atoms (grey) located in tetragonal holes between Y atoms
- Lattice constant a = 5.2032 Å
- MedeA calculated value compares well with average experimental value of a = 5.204 Å from X-ray diffraction measurements.



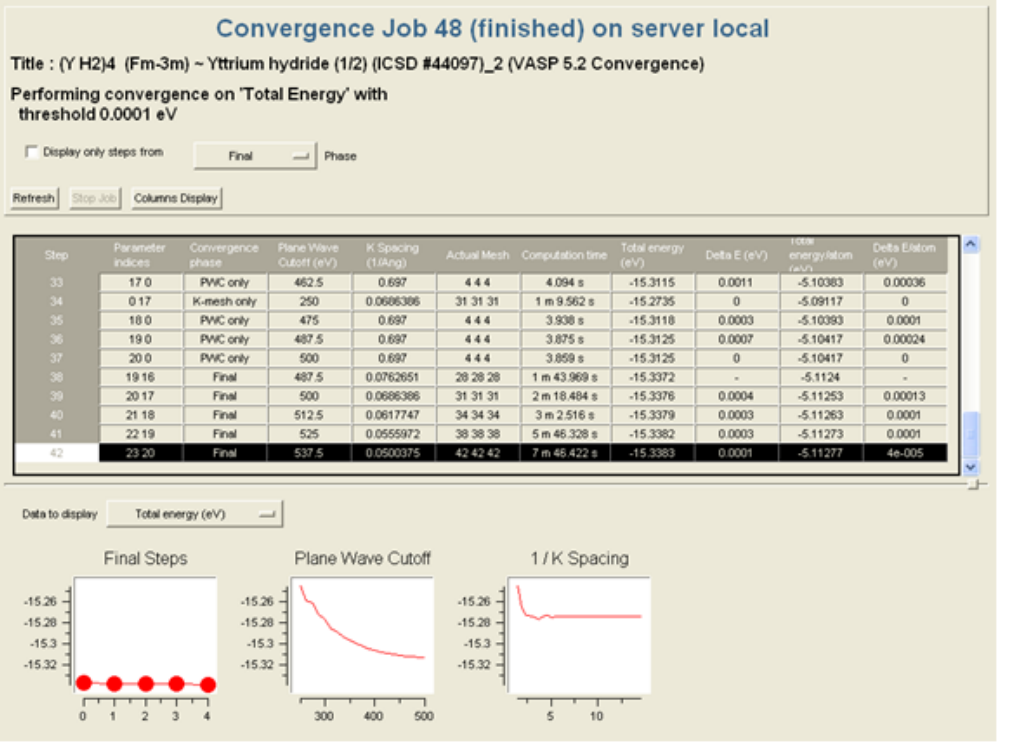
Ab Initio Approach Using MedeA

- MedeA software used to perform the *ab initio* calculations
- Informatica tool used to select the initial unit cell structure from the ICSD (crystal structure) database
- Automated Covergence tool used to optimize unit cell structure
  - Converge k-mesh
  - Converge planewave cutoff energy
  - Converge supercell size
  - Iterative VASP *ab initio* calculations
- PHONON tool performs the lattice dynamics calculations on optimized structure
  - Dispersion relations
  - Phonon density of states
  - Thermodynamic properties
- NJOY/LEAPR used to generate TSL from phonon frequency distribution



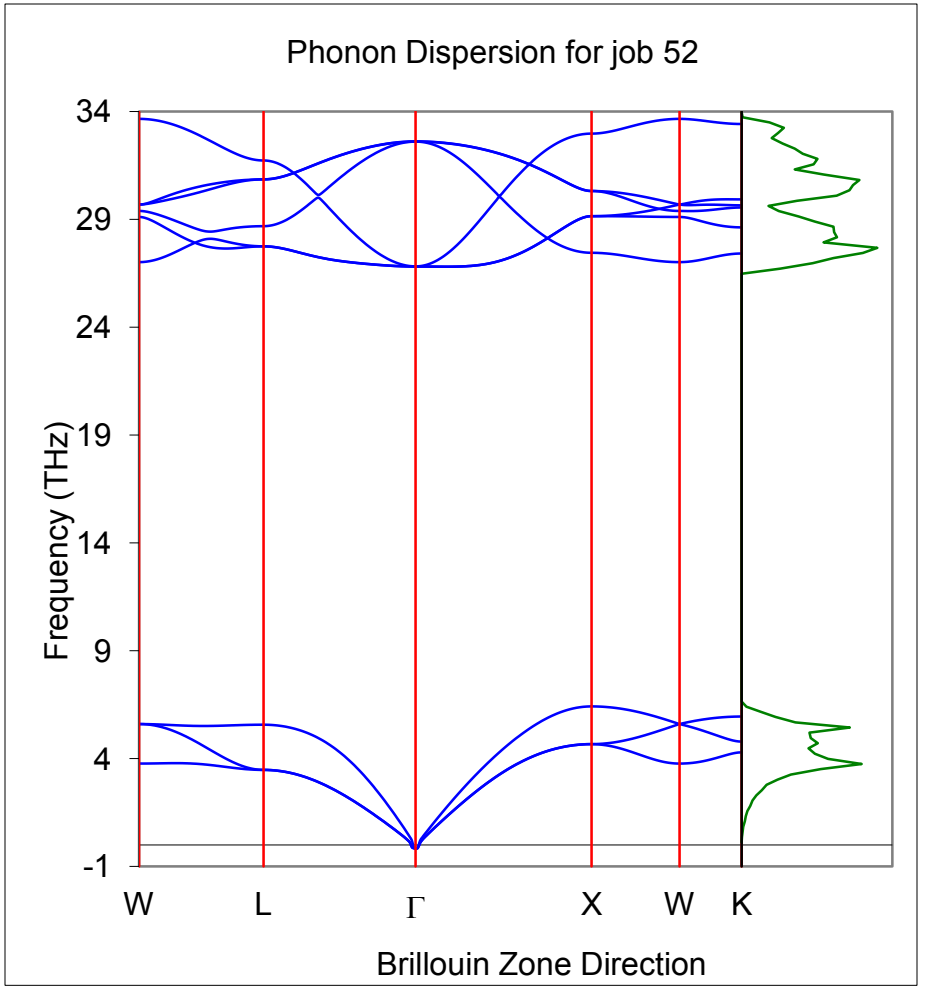
Optimization of the YH_2 Structure

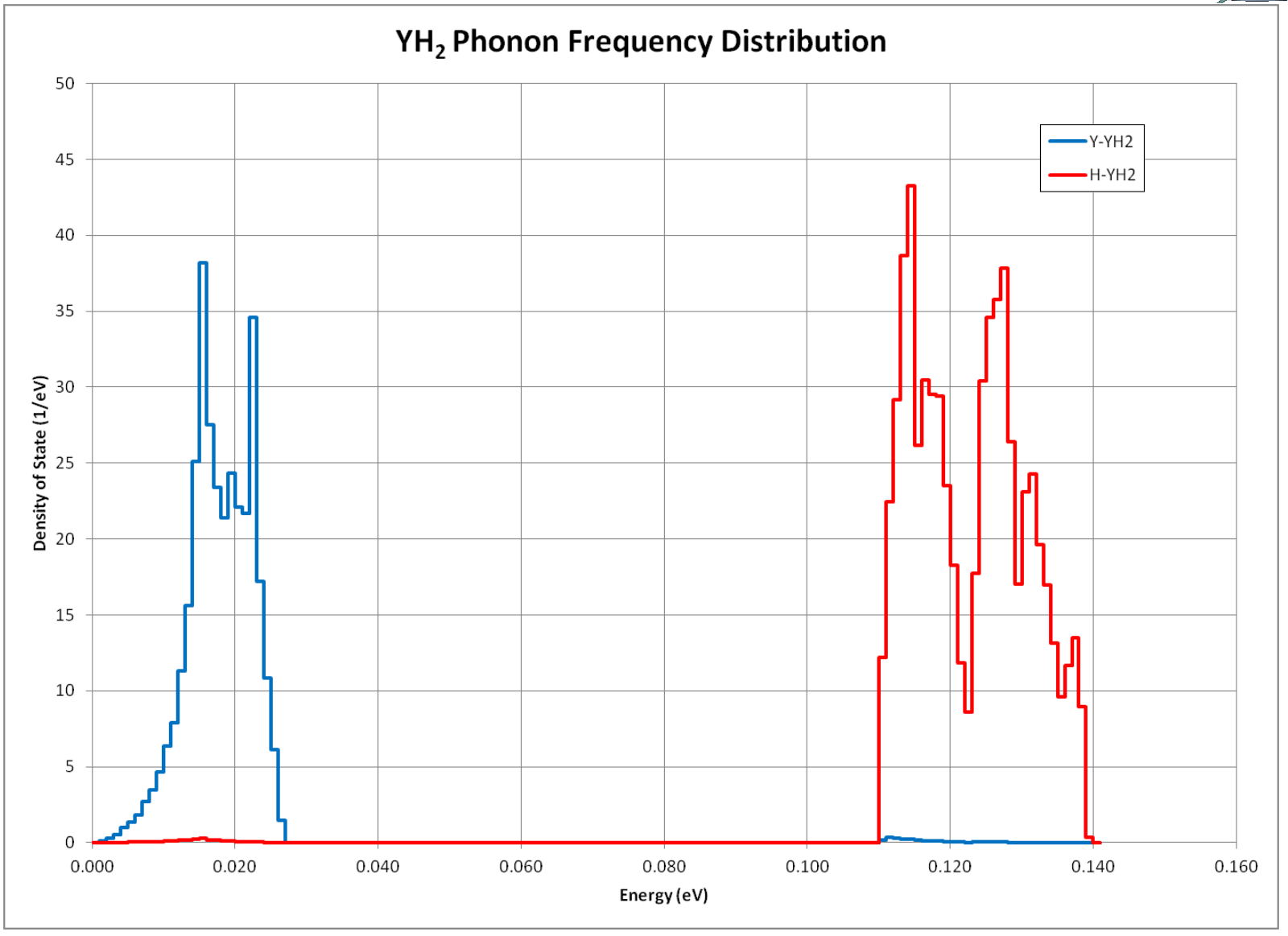
- VASP calculations
  - Detailed quantum mechanical first principle simulation of crystal lattice using density functional theory
  - Generalized Gradient Approximation (GGA)
  - Projector Augmented Wave (PAW) pseudopotential
- Optimized structure to a total energy threshold of 0.1 meV
  - Required 42 VASP calculations
  - 537.5 eV planewave cutoff energy
  - k-spacing of ~0.05 1/Å
  - 42×42×42 final supercell size
  - Optimized lattice constant a = 5.20319995 Å
- Optimized lattice structure then used in PHONON calculations

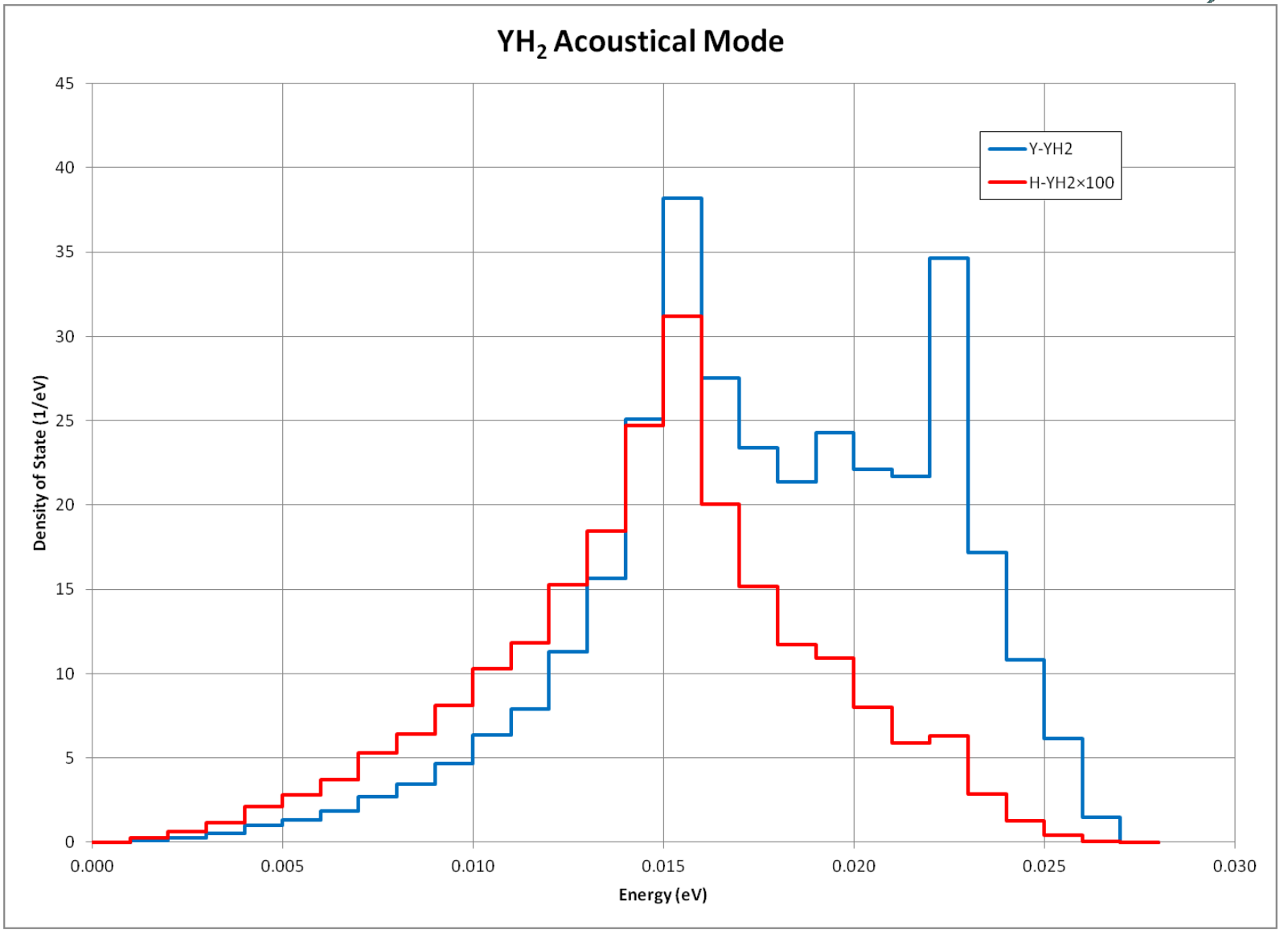


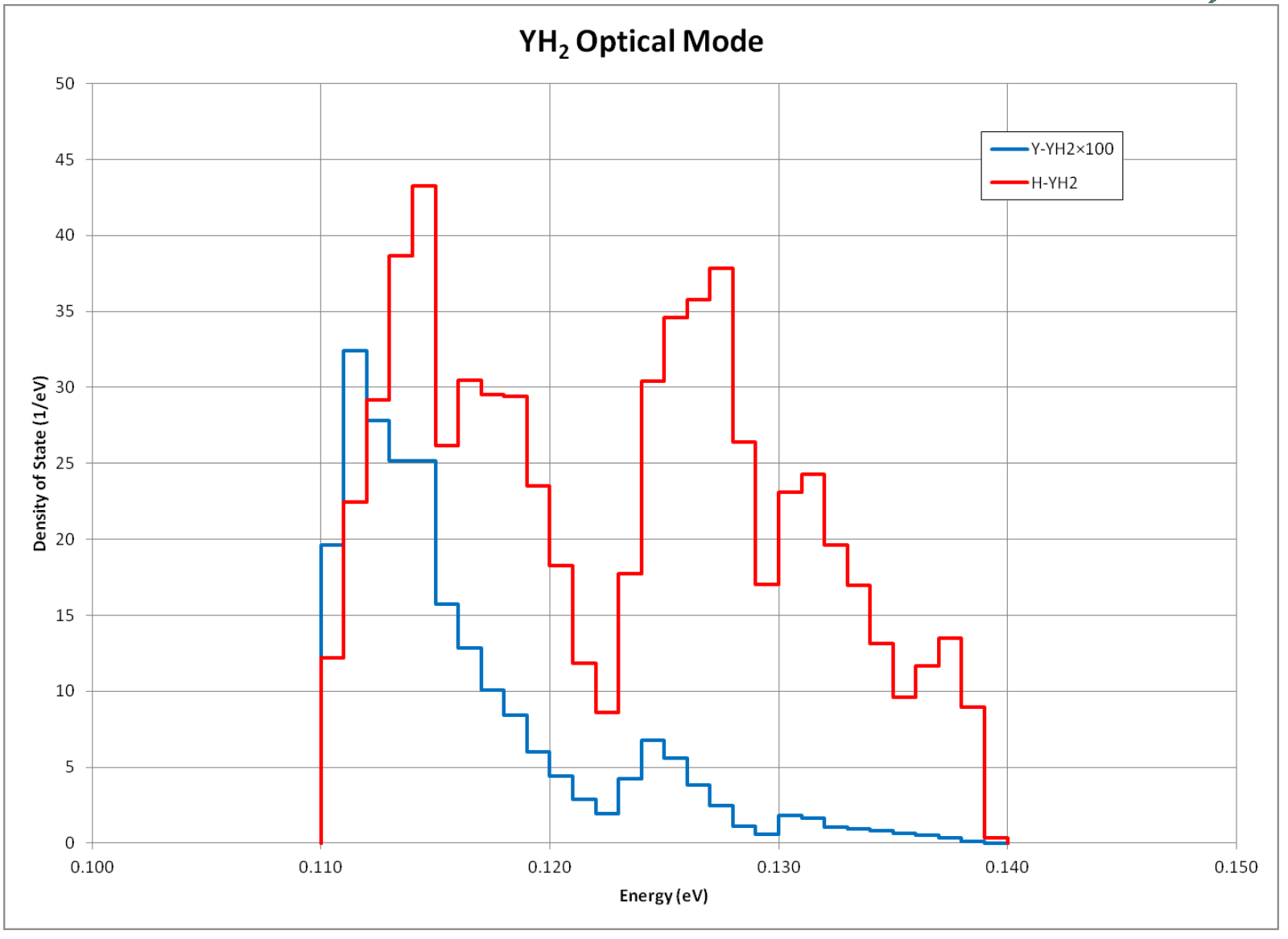
YH_2 Dispersion Relations

- PHONON calculation use
  - 2×2×2 supercell (96 atoms)
  - Planewave cutoff energy of 250 eV
  - k-spacing of 0.5/Å (5×5×5 k-mesh)
  - ± 0.02 Å displacement
- Dispersion relations (at right)
  - Well separated acoustical and optical modes (as expected)
  - Lower branches are acoustical modes mainly due to heavy Y atom vibrations
  - Higher branches are optical modes mainly due to light H atom vibrations

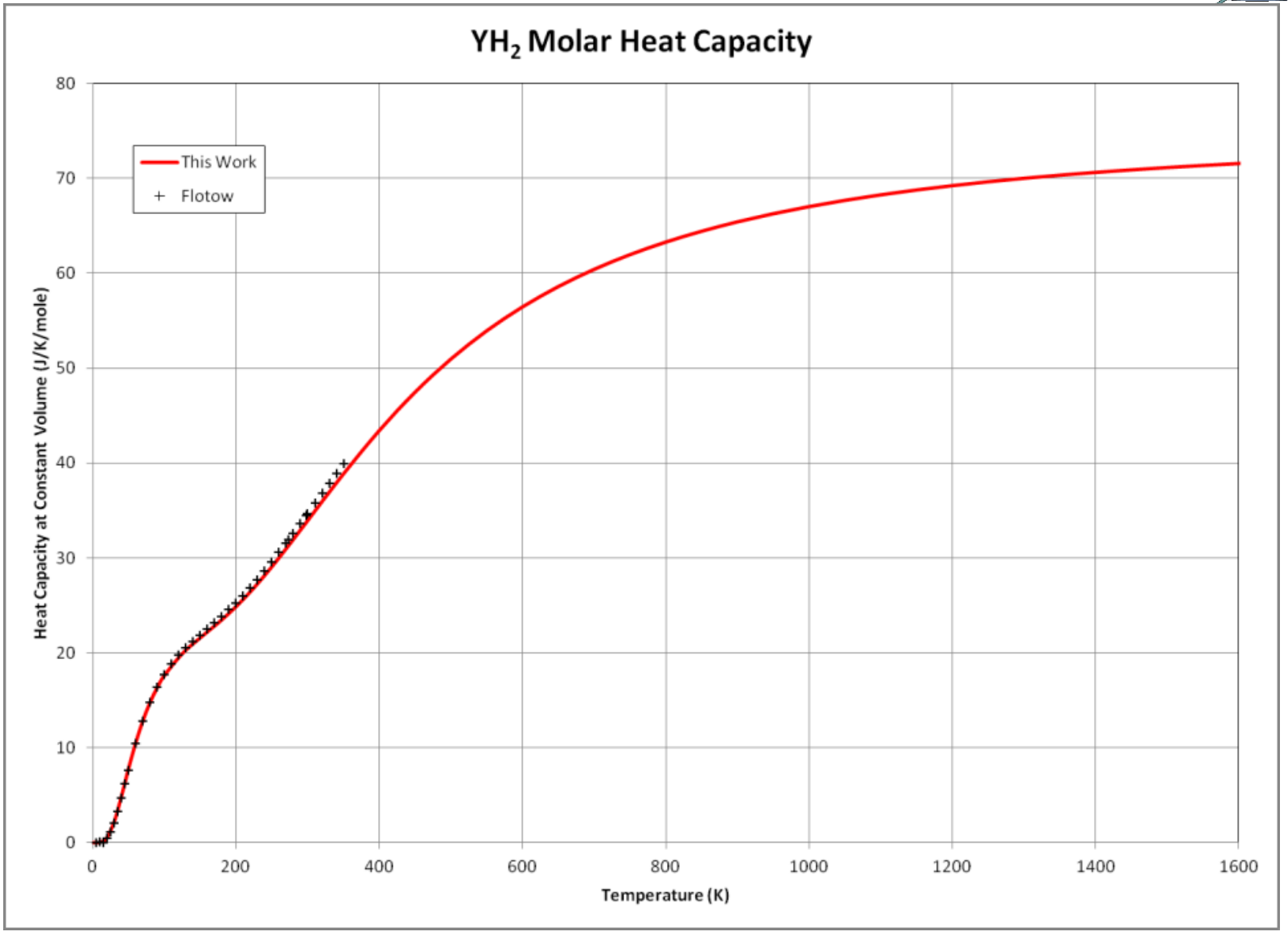



YH2 Phonon Frequency Distribution
Density of State (1/eV)
Energy (eV)
Y-YH2
H-YH2
0
5
10
15
20
25
30
35
40
45
50
0.000
0.020
0.040
0.060
0.080
0.100
0.120
0.140
0.160

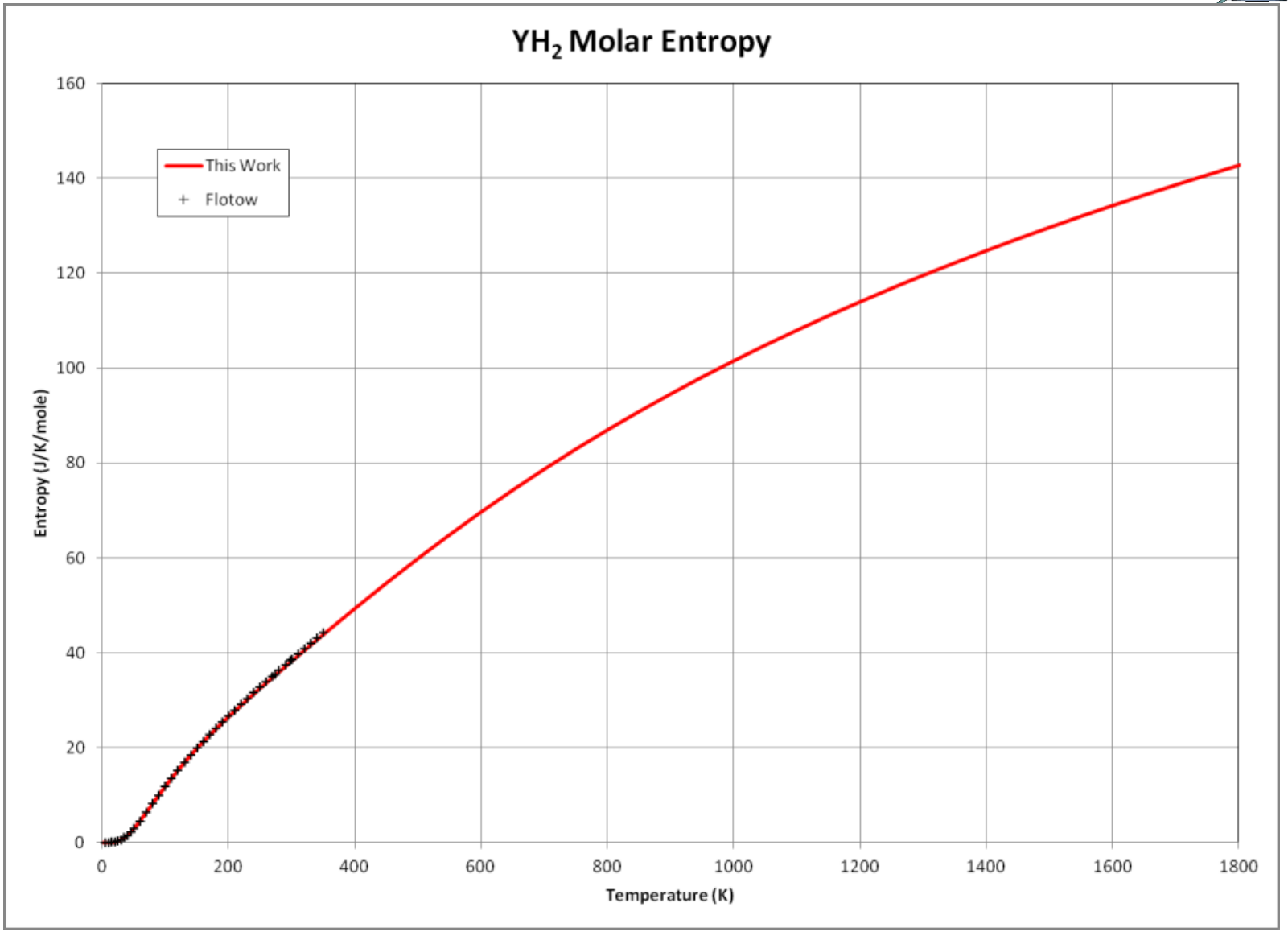

YH2 Acoustical Mode
Y-YH2
H-YH2×100
Density of State (1/eV)
Energy (eV)
0
5
10
15
20
25
30
35
40
45
0.000
0.005
0.010
0.015
0.020
0.025
0.030


YH2 Optical Mode
Y-YH2×100
H-YH2
Density of State (1/eV)
Energy (eV)
0
5
10
15
20
25
30
35
40
45
50
0.100
0.110
0.120
0.130
0.140
0.150

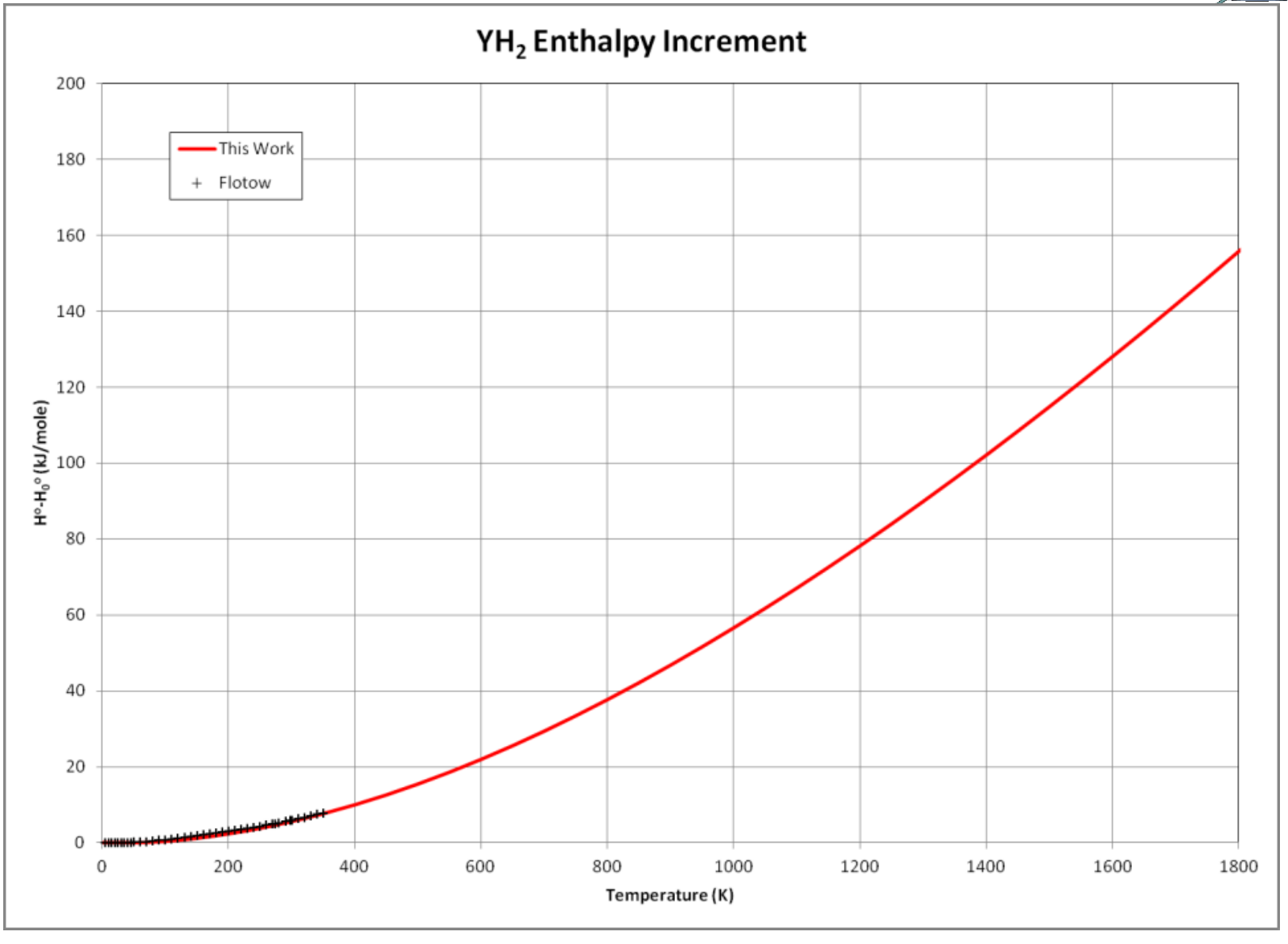
Thermodynamic Properties Agree with Measurements



Thermodynamic Properties Agree with Measurements

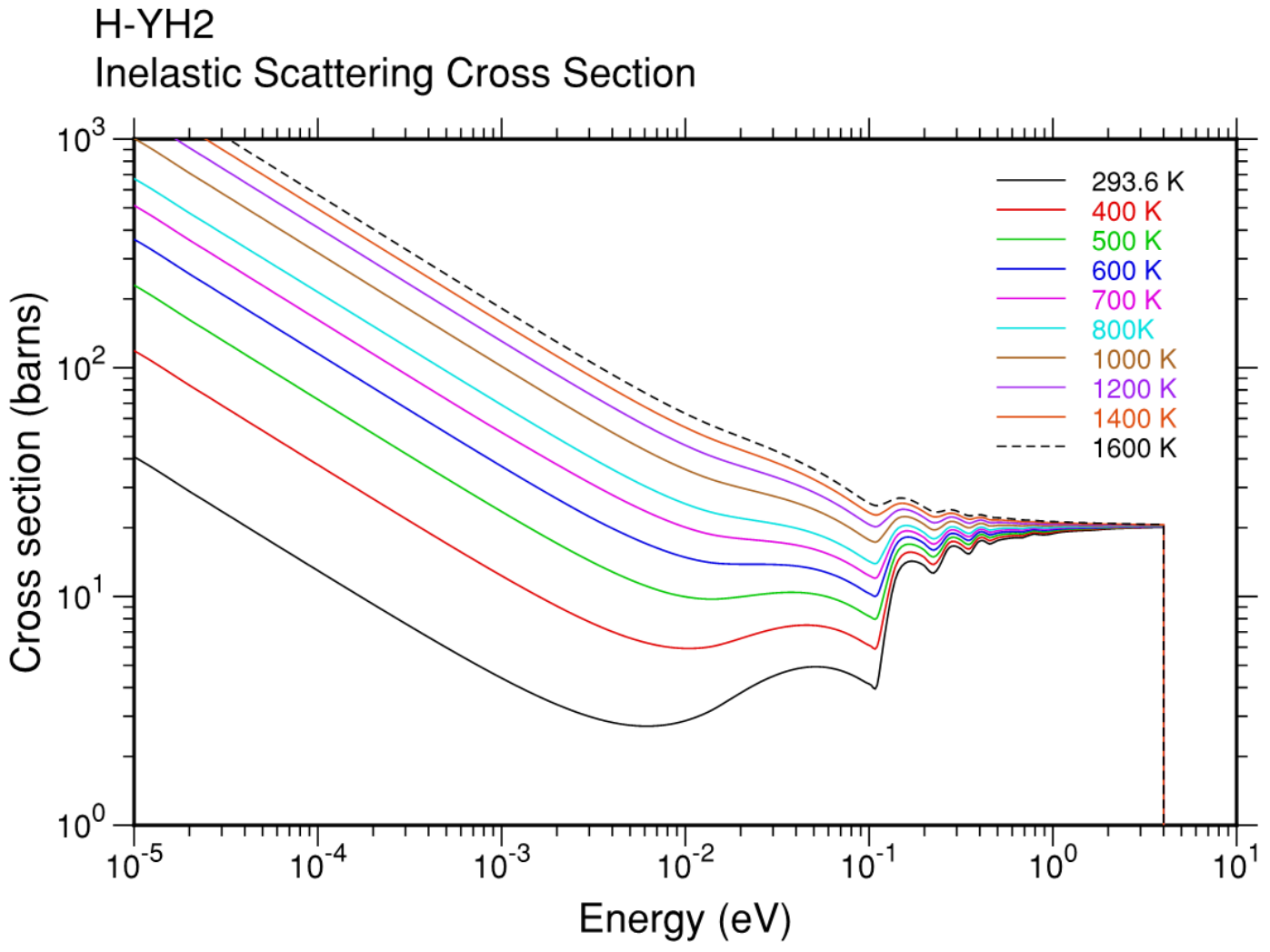


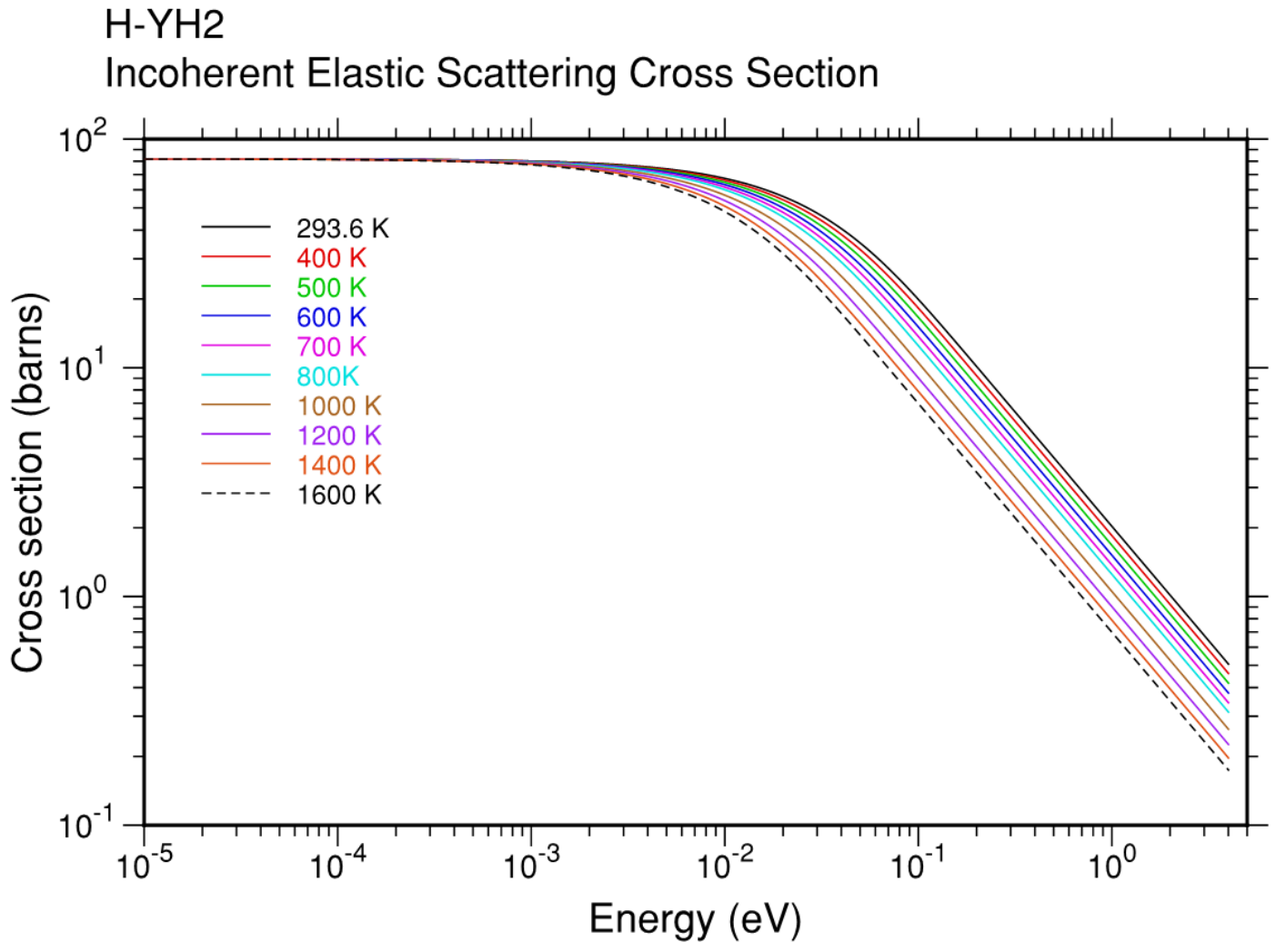
Thermodynamic Properties Agree with Measurements

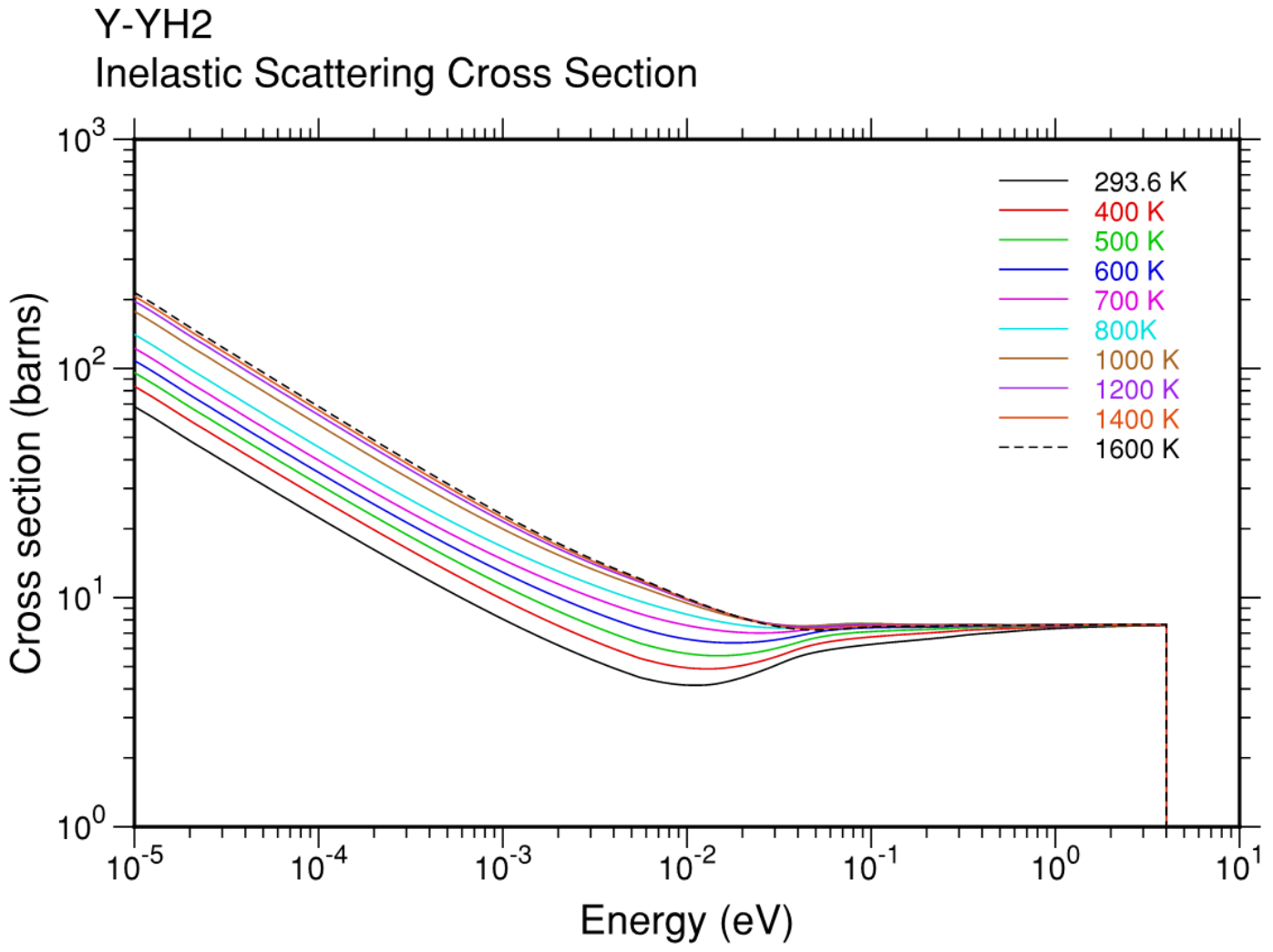


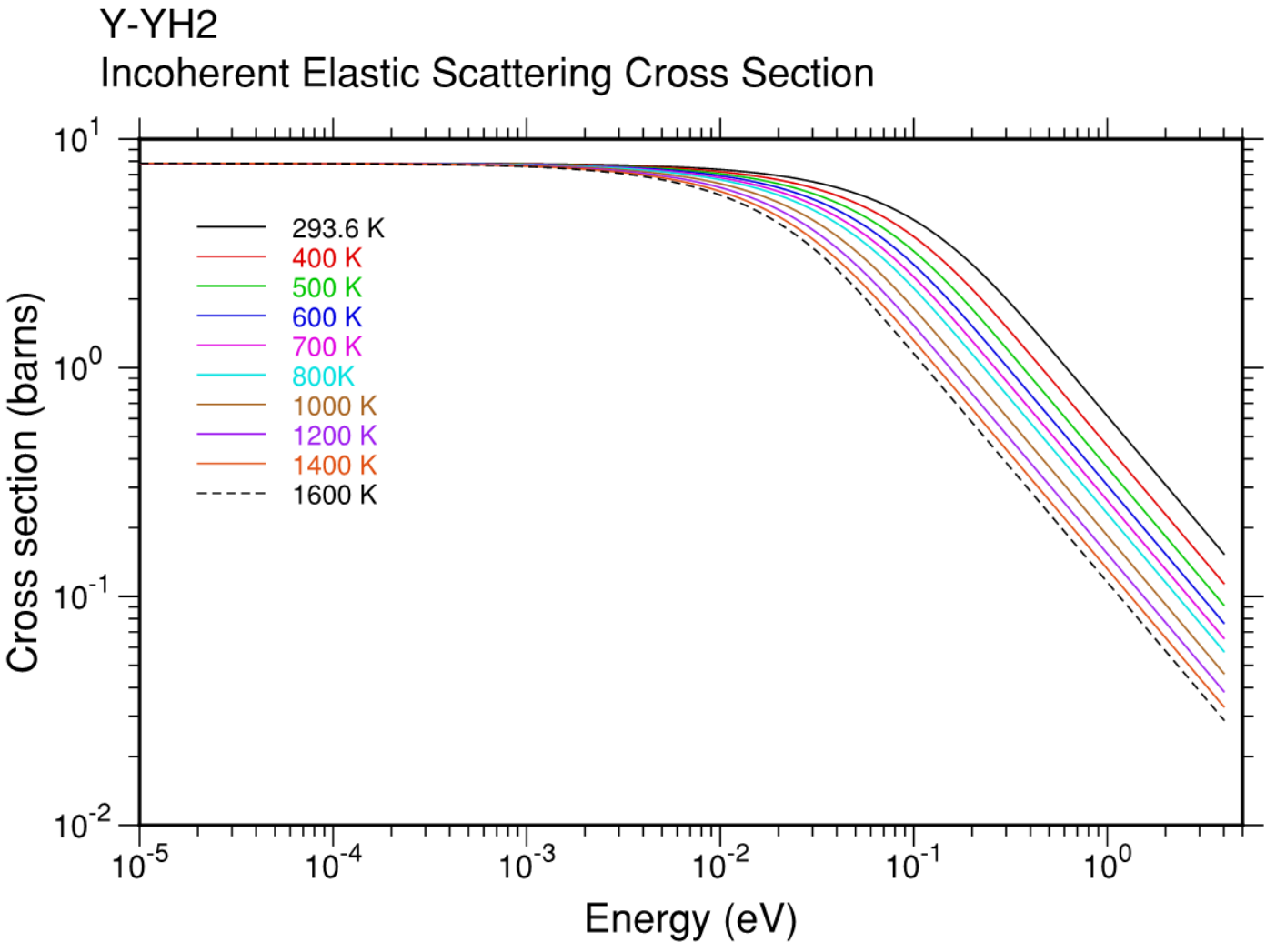
LEAPR Models for YH_2 TSLs

- H-YH_2 and Y-YH_2 TSL generated using LEAPR
- Phonon frequency distributions from PHONON calculation
- Incoherent approximation
- Atomic mass ratios and free atom scattering cross sections from ENDF/B-VII.1 ^{1}H and ^{89}Y evaluations
- α and β mesh borrowed from ENDF/B-VII.0 ZrH TSLs
- 10 temperatures (293.6, 400, 500, 600, 800, 1000, 1200, 1400, 1600 K)


H-YH2
Inelastic Scattering Cross Section
Cross section (barns)
Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800K
1000 K
1200 K
1400 K
1600 K


H-YH2
Incoherent Elastic Scattering Cross Section
Cross section (barns)
Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800K
1000 K
1200 K
1400 K
1600 K


Y-YH2
Inelastic Scattering Cross Section
Cross section (barns)
Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800K
1000 K
1200 K
1400 K
1600 K


Y-YH2
Incoherent Elastic Scattering Cross Section
Cross section (barns)
Energy (eV)
293.6 K
400 K
500 K
600 K
700 K
800K
1000 K
1200 K
1400 K
1600 K

Conclusions

- *ab initio* tools like MedeA have sufficiently matured and can be used to generate TSLs for new moderator materials
- H-YH_2 and Y-YH_2 TSLs produced using the *ab initio* approach
- Results consistent with available measurements
- Need to review α and β mesh to confirm it adequately resolves the S(α,β,T) functions for H-YH_2 and Y-YH_2
- Cross section measurements would be of use to more completely validate TSLs for ENDF/B