

# Generation of Thermal Scattering Laws for YH<sub>2</sub> using Ab Initio Methods

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# Outline

- Motivation
- Yttrium Hydrogen Phase Diagram
- YH<sub>2</sub> Crystal Structure
- Ab Initio Approach using MedeA
  - Structure Optimization
  - Lattice Dynamics
- YH<sub>2</sub> Thermal Scattering Laws
  - Phonon Frequency Distributions
  - Inelastic Scattering Cross Section
  - Incoherent Elastic Scattering Cross Section
- Conclusions



#### Motivation

- Metal hydrides (ZrH<sub>x</sub>) 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)

 $N_{\rm H}$  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 + δ-YH<sub>2</sub> phases
  - $\delta$ -YH<sub>2</sub> phonon frequency
- δ-YH<sub>2</sub> phonon frequency distribution needed to develop thermal scattering law (TSL) for YH<sub>2-x</sub>



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

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



# YH<sub>2</sub> Crystal Structure

- $YH_2$  has a CaF<sub>2</sub> 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 lattice constant compares **extremely well** with recent X-ray diffraction measurements by Daou and Vajda [1992]
  - a = 5.2032(3) Å (90 K)
  - a = 5.2085(3) Å (295 K)





#### **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



May 18-19, 2015



### **Optimization of the YH<sub>2</sub> 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  $\sim 0.05 \text{ } 1/\text{\AA}$
  - 42×42×42 final k-mesh
  - Optimized lattice constant a = 5.20319995 Å
- Optimized lattice structure then used in PHONON calculations

		Con	vergen	ce Job	48 (fini	shed) o	n serve	r local			
Title : (Y H2	!)4 (Fm-3n	n) ~ Yttrium	hydride (1)	(2) (ICSD #	44097)_2(	VASP 5.2 Co	nvergenc	e)			
Performing threshold	converge 0.0001 eV	nce on 'Tol	al Energy'	with							
E Display onl	ly stops from	Final	Phes	e							
Rafresh Step	Job Columns	Display									
Step	Ferenter indices	Convergence phase	Plane Wave Cutoff (eV)	KSpecieg (tiAng)	Actual Mesh	Conputation time	Tdal energy (6v)	Det: E (e'v)	ictai energylatom	Deta Eletan (6V)	Π
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34	0.17	K-mesh only	290	0.0686386	31 31 31	1 m 9.962 s	45.2735	0	-5.09117	0	
35	18 0	PV/C only	475	0.697	44.4	3.938 s	45.3118	0.0003	-5.10893	0.0001	
	19 0	PV/C only	487.5	0.697	44.4	3.875 s	45.3125	0.0007	-5.10017	0.00024	
	20 0	PV/C only	500	0.697	44.4	3.859 s	45.3125	0	-5.10417	0	
	10.16	Final	497.6	0.0762661	28 28 28	1 m 43.060 o	46.3372	-	-6.1124	-	
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	21.18	Panel	512.5	0.0617747	34 34 34	3 m 2,516 s	45.3378	0.0003	-0.11253	0.0001	
	22.19	Final	525	0.05559*2	38 38 38	5 m 46 328 s	-15.3382	0.0003	-5.11273	0.0001	
42	23 20	Final	537.5	0.0500375	424242	7 m 46 422 s	-15,3383	0.0001	-5.11277	4e-005	
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-15.28 -15.28 -15.3 -15.30		-15.2 -15.2 -15.3	6 - 1 8 - 1 3 - 1 2 - 1		-15.30 -15.32 -15.32	L	-				
			1								



### **YH<sub>2</sub> Dispersion Relations**

- PHONON calculation use
  - 2×2×2 supercell (96 atoms)
  - Planewave cutoff energy of 250 eV
  - k-spacing of  $0.5/\text{\AA}$  (5×5×5 k-mesh)
  - $\pm 0.02$  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







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#### **Thermodynamic Properties Agree with Measurements**





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# LEAPR Models for YH<sub>2</sub> TSLs

- H-YH<sub>2</sub> and Y-YH<sub>2</sub> 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 <sup>1</sup>H and <sup>89</sup>Y evaluations
- $\alpha$  and  $\beta$  mesh borrowed from ENDF/B-VII.0 ZrH TSLs
- 10 temperatures (293.6, 400, 500, 600, 800, 1000, 1200, 1400, 1600 K)





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#### Y-YH2 Incoherent Elastic Scattering Cross Section 10<sup>1</sup> 293.6 K 400 K 500 K Cross section (barns) 600 K 700 K 10<sup>0</sup> 800K 1000 K 1200 K <mark>1400 K</mark> 1600 K 10 10<sup>-2</sup> -10<sup>-5</sup> 10<sup>-3</sup> 10<sup>0</sup> 10<sup>-4</sup> 10<sup>-2</sup> 10<sup>-1</sup> Energy (eV)

May 18-19, 2015



# Conclusions

- *ab initio* tools like MedeA (VASP, PHONON) have sufficiently matured and can be used to generate TSLs for new moderator materials
- H-YH<sub>2</sub> and Y-YH<sub>2</sub> TSLs produced using the *ab initio* approach
- Results consistent with available measurements
- Currently reviewing  $\alpha$  and  $\beta$  mesh to ensure it adequately resolves the S( $\alpha$ , $\beta$ ,T) functions for H-YH<sub>2</sub> and Y-YH<sub>2</sub>
- Working to submit H- $YH_2$  and Y- $YH_2$  evaluations to CSEWG