Evaluation of Thermal Neutron Scattering Law and Cross Sections for Calcium Hydride

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INTRODUCTION

Metal hydrides are considered to be good moderator materials due to a variety of factors which include moderating ratio, hydrogen density, favorable mechanical properties, and high dissociation temperatures [1]. ZrH_2 has been of primary interest thus far in nuclear reactor designs, and has been successfully utilized in TRIGA reactors. More recently, there has been increased interest in other metal hydrides like CaH_2 and YH_2, with the former being used in the design of a <10MW microreactor [2].

Due to the increased interest in CaH_2, the purpose of this work is to evaluate its thermal scattering law (TSL) and associated thermal neutron scattering cross sections, which are of vital importance to designing moderators. There is little existing data in the literature prior to this work, with poor evaluations existing in the JEFF-3.1(3.3) libraries and none in the ENDF/B-VIII.0.

THERMAL SCATTERING THEORY

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi k_B T}\sqrt{\frac{E}{E'}}\left[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right]$$

Double-Differential Cross Section: Derived from first-principles using:

- First Born Approximation
  - First-Order Perturbation Theory; Fermi's Golden Rule
- Fermi Pseudopotential

The derived equation is shown above, where k_B is the Boltzmann constant; T is the absolute temperature (K); σ_{coh} and σ_{inc} are the bound coherent and incoherent nuclear scattering cross sections, respectively; and $S(\alpha,\beta)$ is TSL:

$$S(\boldsymbol{\kappa},\omega) = \frac{1}{2\pi\hbar}\int G(\boldsymbol{r},t)e^{i(\boldsymbol{\kappa}\cdot\boldsymbol{r}-\omega t)}\mathrm{d}\boldsymbol{r}\mathrm{d}t$$

$$S(\alpha,\beta) = k_B T\cdot S(\boldsymbol{\kappa},\omega) = S_s(\alpha,\beta) + S_d(\alpha,\beta)$$

$$\alpha = \frac{E'+E-2\mu\sqrt{E'E}}{Ak_BT};\quad \beta = \frac{E'-E}{k_BT}$$

The TSL is a material property that depends on atomic positions and describes the probability distribution of energy and momentum states within the material that are available for the neutron to interact with. In short, it describes how the neutron exchanges energy and momentum with the scattering system. The variables α and β are dimensionless momentum and energy transfer, respectively, and are defined above; μ is the cosine of the scattering angle; and A is the mass ratio between the scatterer and neutron. $G(\boldsymbol{r},t)$ is the time-dependent pair correlation function of the scattering system; $S(\boldsymbol{\kappa},\omega)$ is its Fourier transform in both space and time. The space-inverse space variables are position and scattering vector ($\boldsymbol{r}\rightarrow\boldsymbol{\kappa}$), and the time-energy variables are time and frequency ($t\rightarrow\omega$).The TSL can be divided into two parts: (1) a component that contains interatomic interference effects and (2) a component that does not; these are shown above. These are the distinct- (S_d) and self- (S_s) parts, respectively.

Approximations in Calculating TSL:

- Incoherent Approximation: $S_d(\alpha,\beta) = 0$
- Gaussian Approximation for $G(\boldsymbol{r},t)$
  - Harmonic Approximation for solid scattering systems
    - Allows use of phonon expansion with phonon Density of States (DOS)
- Cubic Approximation for calculating TSL and $\sigma^{coh}(E\rightarrow E',\mu)$

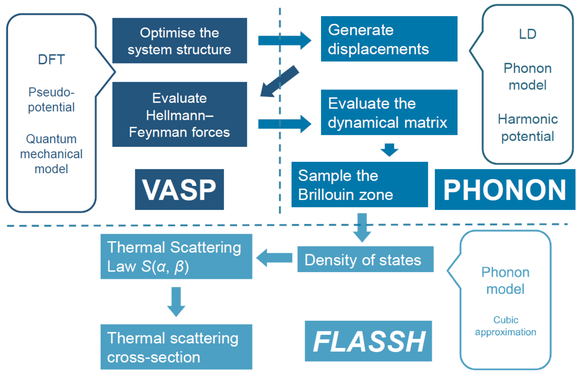


Fig. 1. LEIP Labs TSL Evaluation Process. The phonon DOS is calculated from first principles DFT using the VASP and PHONON codes. *FLASSH* [8] uses a phonon expansion of the DOS to calculate the TSL and subsequent thermal neutron scattering cross sections

COMPUTATIONAL APPROACH

The vibrational (phonon) DOS (vDOS) is calculated from first principles using Density Functional Theory (DFT), which calculates material properties using quantum-mechanical formulation of electron density and structural information. Figure 1 presents a diagram of the evaluation process used in this work by LEIP Labs; specifics regarding the DFT calculation are below. Figure 2 shows the CaH_2 unit cell.

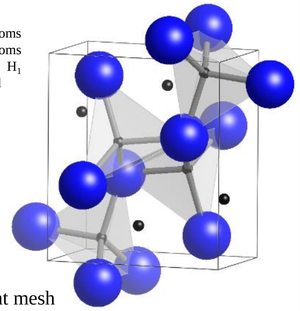


Fig. 2. CaH_2 Unit Cell. Ca atoms represented by blue spheres; H atoms represented by black spheres. H_1 coordination polyhedra are highlighted

CaH_2 Structural Information:

- Orthorhombic; Space Group #62 *Pnma*
- 3 nonequivalent atom sites: Ca, H_1, H_2

DFT Calculation:

- Vienna *ab intio* Simulation Package (VASP)
- Projector-Augmented Wave Method [3]
  - GGA-PBE exchange-correlation functional
  - 675eV plane-wave cutoff; 9x9x9 Monkhorst-Pack k-point mesh
- Hellmann-Feynman (HF) forces calculated using VASP and PHONON [4-5]; 3x3x3 supercell of 324 atoms
- vDOS calculated by PHONON, fed to *FLASSH* for calculation of TSL & cross sections [6-8]

RESULTS – STRUCTURAL DATA

Table 1. CaH_2 Lattice Constants as determined in this work via a DFT structure optimization and compared to experimental values

Lattice Constant	This Work	Experiment [9]	Error (%)
a (Å)	5.92176	5.92852	0.114
b (Å)	3.57607	3.57774	0.0468
c (Å)	6.78272	6.78956	0.1007

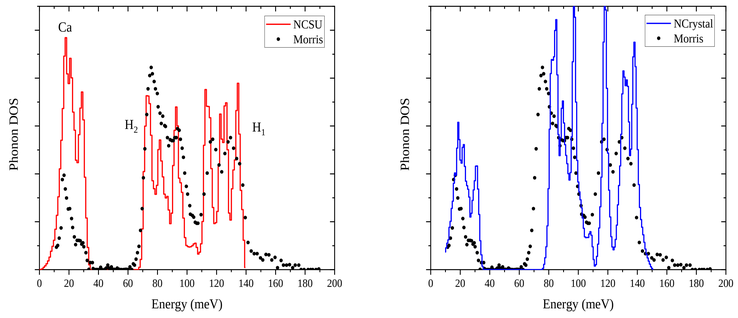


Fig. 3. Comparison of the CaH_2 vDOS calculated by LEIP Labs (current work, left) and NCrystal (right), with available experimental data

The plots shown in Figure 3 above are the vDOS (vibrational DOS) of CaH_2 as calculated in the current work (left) and by another TSL calculating code, Ncrystal (right), compared to available experimental data [10]. While the same general features are visible in the calculated DOSs, that of the current work more closely matches the experimental data. Of crucial importance to the thermal neutron scattering cross sections is the initial slope of the DOS (for each partial DOS), as phonons will always occupy this region and thus will contribute to neutron scattering. As temperature increases, so too does the phonon occupation such that more of the DOS is available to the neutron.

[1] W. M MUELLER et al., Metal *Hydrides*. New York: Academic Press, Inc. (1968).
[2] R. KIMURA and S. WADA, "Temperature Reactivity Control of Calcium Hydride–Moderated Small Reactor Core with Poison Nuclides," *Nuclear Science and Engineering*, **193**, 9, 1013 (2019).
[3] G. KRESSE and D. JOUBERT, "From ultrasoft pseudopotentials to the projector augmented-wave method," *Physical Review B*, **59**, 1785 (1999).
[4] K. PARLINSKI, "PHONON," (2013).
[5] K. PARLINSKI et al., "First-Principles Determination of the Soft Mode in Cubic ZrO 2," *Physical Review Letters*, **78**, 4063 (1997).
[6] A. I. HAWARI et al., "Ab Initio Generation of Thermal Neutron Scattering Cross Sections," *PHYSOR, The Physics of Fuel Cycles and Advanced Nuclear Systems: Global Developments*, Chicago, IL, 2004.
[7] A. I. HAWARI, "Modern techniques for inelastic thermal neutron scattering analysis," *Nuclear Data Sheets*, **118**, *1*, 172 (2014).
[8] Y. ZHU and A. I. HAWARI, "Full Law Analysis Scattering System Hub (FLASSH)," *PHYSOR – Reactor Physics Paving the Way for More Efficient Systems,"* Cancun, Mexico (2018).
[9] H. WU et al., "Structure and vibrational spectra of calcium hydride and deuteride," *Journal of Alloys and Compounds*, **436**, *1-2*, 51(2007).
[10] P. MORRIS et al., "Inelastic neutron scattering study of the vibration frequencies of hydrogen in calcium dihydride," *Journal of Alloys and Compounds*, **363**, *1-2*, 88 (2004).
[11] O. SEROT, "New results on CaH2 thermal neutron scattering cross sections," *AIP Conference Proceedings*,**769**, *1*, 1446 (2005).
[12] E. FERMI, "On the Motion of Neutrons in Hydrogenous Substances," *Ric. Scientifica*, **2**, 13 (1936).

RESULTS – TSL & CROSS SECTIONS

The only existing thermal neutron scattering cross sections for CaH_2 prior to this work were created by Serot [11] and released in the Joint Evaluated Fission and Fusion (JEFF) Nuclear Data Libraries version 3.1. However, there are two crucial shortcomings of this work: (1) the coherent elastic scattering from Ca was neglected and instead only the incoherent elastic contribution was used (and drastically overestimated); and (2) the nonequivalent H atoms were treated in an averaged manner as a single contributing atom.

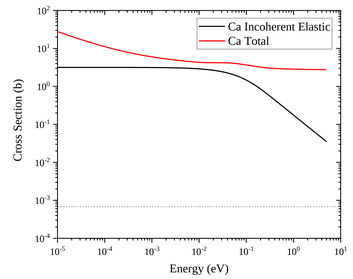
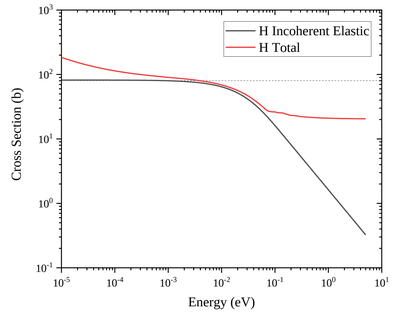


Fig. 4. CaH_2 Cross Sections in the JEFF-3.3 Database at 296K. Only the incoherent elastic component of each scatterer is considered. Ca is on the left; H is on the right. The horizontal line represents σ_b^{inc}

Table 2. Bound Scattering Cross Sections of Ca and H (NIST)

Isotope	σ_b^{coh} [b]	σ_b^{inc} [b]
Ca	2.64	0.000675
^{1}H	1.7568	80.26



In the present work, two primary improvements were made: (1) The correct components of the scattering cross section were considered for each scatterer (coherent elastic for Ca; incoherent elastic for H – see Table 2); and (2) the nonequivalent H atoms were treated separately as opposed to averaged, which allows the full physical behavior of each scatterer to be represented. The calculated cross sections are shown in Figures 5 and 6 below.

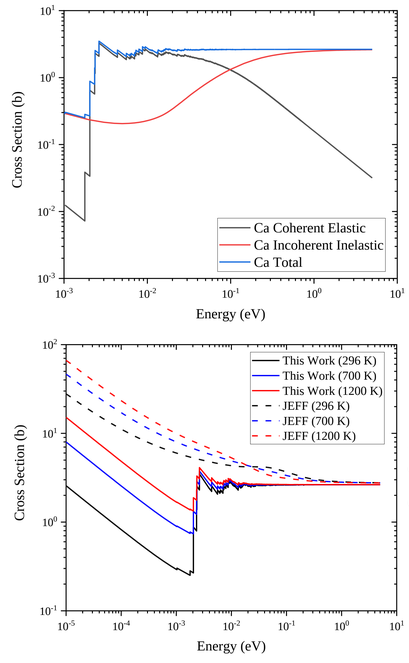


Fig. 5. (Top) Contributions to the total Ca in CaH_2 thermal neutron scattering cross section as calculated in this work. (Bottom) Comparison of total Ca scattering cross sections as calculated in this work (solid) and in JEFF (dashed)

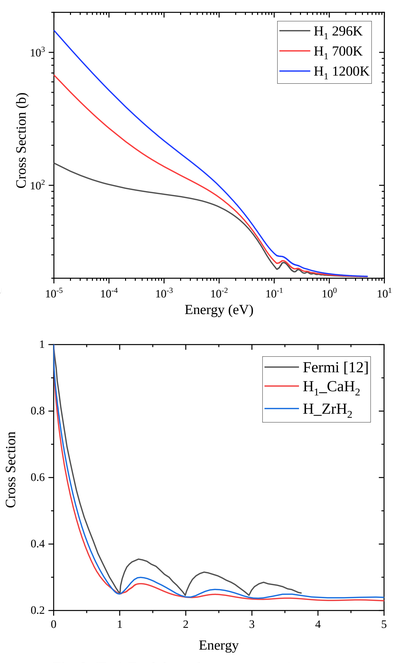


Fig. 6. (Top) Total thermal neutron scattering cross section of H_1 as calculated in this work; H_2 has similar (but shifter) data, which is not shown here. (Bottom) Comparison of H_1 in CaH_2 scattering cross section to the cross section of H bounded by an infinite mass as predicted by Fermi [12]