

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

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**Abstract.** Presented here are the calculated thermal scattering law (TSL) and thermal neutron scattering cross sections for Calcium Hydride, hereafter referred to by its chemical symbol CaH<sub>2</sub>. The only other such data prior to this evaluation are thermal neutron scattering libraries in the JEFF database, which suffer from nonphysical features and inaccuracies. The data in this evaluation are calculated from first principles; Density Functional Theory (DFT) is used to calculate the phonon density of states (DOS), which is the primary input required to calculate the TSL. The TSL and cross sections have been evaluated for the three non-equivalent atom sites in the CaH<sub>2</sub>: Ca, H<sub>1</sub>, and H<sub>2</sub>. Each evaluation has been submitted to the NNDC for consideration in the next ENDF release.

## 1 Introduction

CaH<sub>2</sub> is being considered for use as a moderator in new reactor designs, including a small modular microreactor [1]. Metal hydrides have long been known to be great moderators based on various properties; ZrH<sub>2</sub> has been successfully used in TRIGA reactors for decades. Of crucial importance to the moderating behavior of any material, especially for use in thermal reactors, is the thermal neutron scattering mechanism. Unlike higher-energy cross sections which are determined empirically, thermal neutron scattering cross sections can be derived from first principles [2], [3]. This means that these cross sections can be calculated for any material, and are not limited by the availability of experimental data. This paper describes the process of evaluating new thermal scattering data for CaH<sub>2</sub>.

## 2 Thermal Scattering Theory

The double-differential scattering cross section represents the probability of a neutron scattering from incident energy  $E$  into outgoing energy  $E'$  about  $dE'$  through solid angle  $\Omega$  about  $d\Omega$ . The derived form is shown in Eq. 1, where  $k_B$  is the Boltzmann constant;  $T$  is the absolute temperature (K);  $\sigma_{coh}$  and  $\sigma_{inc}$  are the bound coherent and incoherent neutron scattering cross sections (b), respectively; and  $S(\alpha, \beta)$  is the TSL.

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

The TSL is inherently a material property; it is the Fourier transform of the time-dependent pair correlation function in both space and time. It describes the probability distribution of energy and momentum states

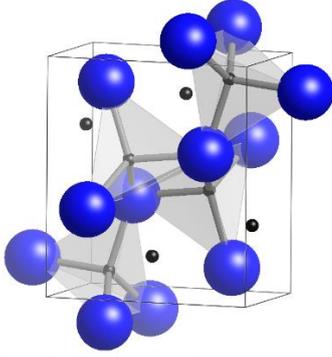
of the material, and thus defines how a thermal neutron may interact with the scattering system. It is comprised of two components, as shown in Eq. 2; the distinct component ( $S_d$ ) contains interatomic interference effects while the self-component ( $S_s$ ) does not.

$$S(\alpha, \beta) = S_s(\alpha, \beta) + S_d(\alpha, \beta) \quad (2)$$

The incoherent approximation assumes that the contribution of  $S_d$  is negligible to the total TSL ( $S_d = 0$ ), and thus the TSL is entirely incoherent. For crystalline materials the harmonic approximation allows the TSL to be calculated via a phonon expansion, which requires the phonon DOS. DFT is a method of calculating material properties, including its DOS, using a quantum-mechanical model of electron density.

## 3 Computational Method

CaH<sub>2</sub> has an orthorhombic crystal structure with symmetry described by the  $Pnma$  space group (#62). Its unit cell contains three non-equivalent (not related by symmetry) atom sites, labelled in this work as Ca, H<sub>1</sub>, and H<sub>2</sub>. Figure 3 shows the CaH<sub>2</sub> unit cell. The structural information is fed to the Vienna *ab initio* Simulation Package (VASP) [4], [5] which performs the DFT calculations. The MedeA-VASP [6] platform is used to perform a structure optimization; the *ab initio* Lattice Dynamics (AILD) DFT calculation is performed using the projector-augmented wave (PAW) method [7] and a GGE-PBE exchange-correlation functional. The structure optimization informed a plane-wave cut-off energy of 675eV and a 9x9x9 Monkhorst-Pack k-point mesh. VASP and PHONON [8], [9] are used to calculate the Hellmann-Feynman forces and subsequently the phonon dispersion curves and DOS using the dynamical matrix method.



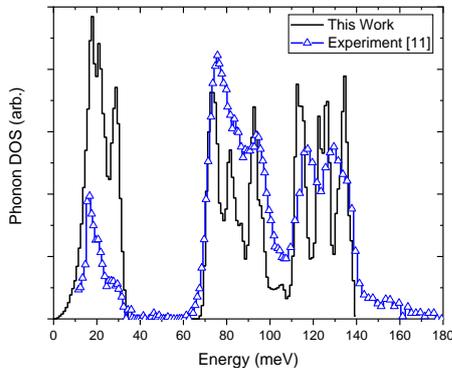
**Fig. 1.** CaH<sub>2</sub> unit cell. The large spheres are Ca; the small spheres H. The H<sub>1</sub> coordination polyhedra are shown.

## 4 Results

The results of the structure optimization are shown below in Table 1. The lattice constants show excellent agreement with experimental data [10]. This data, passed to DFT, was used to calculate the phonon DOS shown in Figure 2. The DOS has three peak clusters corresponding to each of the non-equivalent atoms; the low-energy acoustic modes belong to the high-mass Ca atom and the high-energy optical modes to the H atoms (the more tightly-bound H<sub>1</sub> modes are the higher of the two optical groups). These are compared on a one-to-one basis, and to experimental data [11]. The experimental data is naturally weighted towards the energy states of the H atoms; no attempt was made to undo the experimental weighting or apply it to the calculated data. The portion of the DOS which has the highest impact on thermal neutron scattering is the beginning of each mode, which phonons will always occupy regardless of temperature (according to the Debye temperature,  $\Theta_T$ ). As temperature increases, so does the phonon occupation and the energies available to the thermal neutrons. These crucial regions of the DOS well match the experimental curves.

**Table 1.** Comparison of calculated to experimental CaH<sub>2</sub> lattice constants.

Lattice Constant	This Work	Experiment	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

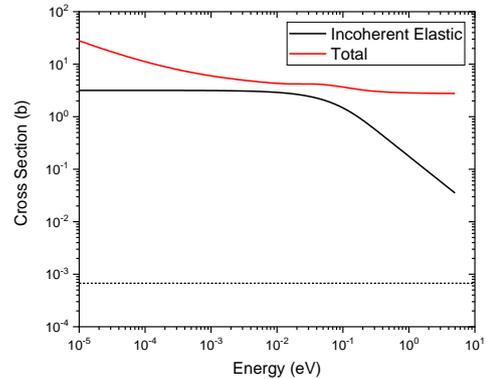


**Fig 2.** Comparison between the calculated phonon DOS and experiment.

The thermal neutron scattering cross sections in the JEFF libraries suffer from several severe problems, specifically in the evaluation for Ca in CaH<sub>2</sub>. Table 2 shows the bound coherent and incoherent neutron scattering cross sections of Ca and H, taken from the NIST database. It is clear based on these data that Ca is almost entirely a coherent scatterer while H is almost entirely an incoherent scatterer. However, the JEFF evaluation only includes the incoherent component of the Ca cross section and overestimates it by several orders of magnitude [12]. This can be seen in Figure 3 below, where the horizontal line is the bound incoherent scattering cross section. Additionally, the behavior of the unique H sites are averaged in the JEFF evaluation, which results in lost information.

**Table 2.** Bound coherent ( $\sigma_b^{coh}$ ) and incoherent ( $\sigma_b^{inc}$ ) neutron scattering cross sections for Ca and H from NIST

Isotope	$\sigma_b^{coh}$ [b]	$\sigma_b^{inc}$ [b]
Ca (natural)	2.64	0.000675
<sup>1</sup> H	1.7568	80.26

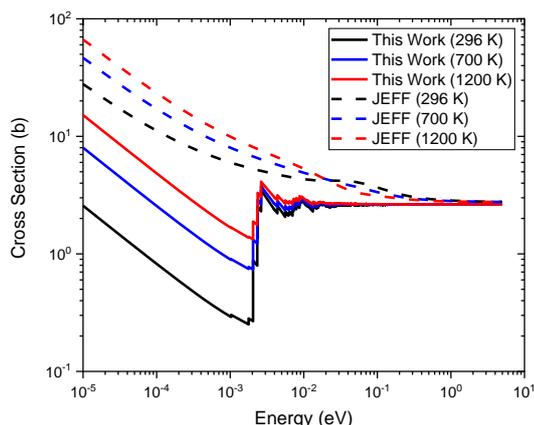


**Fig 3.** Components of the Ca in CaH<sub>2</sub> cross section in the JEFF libraries. The dashed horizontal line is the bound incoherent cross section, to which the incoherent elastic cross section should asymptote at low energies.

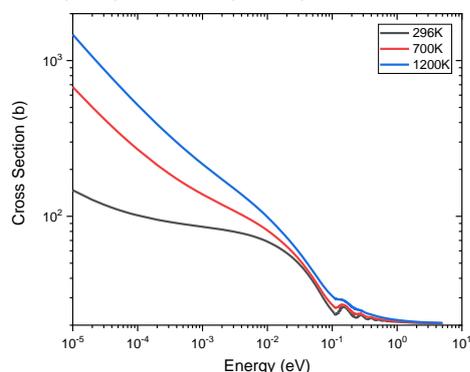
The phonon DOS was passed to the *FLASSH* code [13], where a 500-order phonon expansion was used to calculate the TSL using the cubic approximation. The code also calculated the incoherent inelastic cross section for all three non-equivalent atoms, the coherent elastic (cubic approximation) for Ca, and the incoherent elastic for H<sub>1</sub> and H<sub>2</sub>. Figure 4 is a comparison between the total Ca cross sections of this work and of the JEFF libraries. Figure 5 shows the total cross section of H<sub>1</sub> at various temperatures as calculated in this work (the H<sub>2</sub> cross sections have similar features shifted to lower energies). Finally, figure 6 is an analysis of the calculated H<sub>1</sub> cross section with what Fermi predicted for the interaction between a neutron and H in a hydrogenous substance [14]. There is shown to be great agreement with the predicted phenomena; as the mass of the binding atom increases, the data approaches Fermi's values for an effectively infinite binding mass. The ZrH<sub>2</sub> data is was generated for this comparison.

## 5 Conclusions

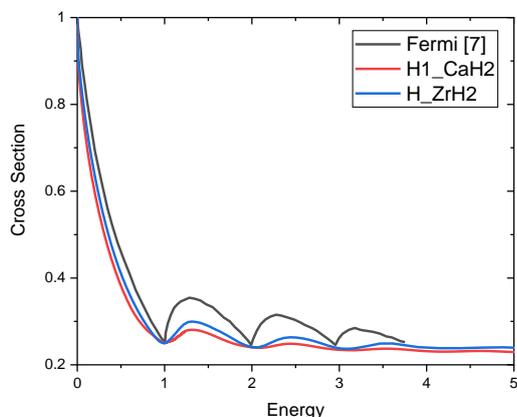
The thermal scattering law and cross sections of CaH<sub>2</sub> were calculated using *ab initio* methods. The data were generated for each of the three non-equivalent sites in CaH<sub>2</sub>: Ca, H<sub>1</sub>, and H<sub>2</sub>. The cross sections for each are consistent with the physics of the scattering system – the contributions to Ca in CaH<sub>2</sub> are incoherent inelastic and coherent elastic, while the contributions for H<sub>1</sub> and H<sub>2</sub> are incoherent inelastic and incoherent elastic. This is an improvement over the existing data in the JEFF-3.3 database, particularly for the Ca cross section. The TSLs and cross sections generated in this work have been submitted to the NNDC for consideration in the next ENDF database release, ENDF/B-VIII.1.



**Fig 4.** Comparison of total Ca in CaH<sub>2</sub> cross sections from this work (solid) and JEFF (dashed).



**Fig 5.** Total cross sections for H<sub>1</sub> in CaH<sub>2</sub> as calculated in this work for a range of temperatures.



**Fig 6.** Comparison between the thermal neutron scattering cross section in a hydrogenous material as derived by Fermi [14], and the calculated cross section for H<sub>1</sub> in CaH<sub>2</sub> at 296K and H in ZrH<sub>2</sub> at 293.6K.

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