Towards a Better Thermal Neutron Scattering Law Generation: oClimax + NJOY2016

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INTRODUCTION

With continuous advancement of computational tools there is also an effort underway to improve the accuracy of nuclear data. In the past, thermal scattering law (TSL) libraries were created solely based on theoretical calculations, where almost no experimental data were available for validation of the newly created libraries. In many cases, the only experimental data used for validation were total cross section measurements.

RPI Nuclear Data research group has set out to develop a new methodology for creation of TSL libraries for different moderator materials. In the newly developed method [1], two types of the experimental data (double differential scattering cross section (DDSCS) measurements from ARCS [2] at Spallation Neutron Source at Oak Ridge National Laboratory (SNS ORNL); and S(Q,ω) measurements from VISION [3] at SNS ORNL) are used for validation and guiding of creation of new TSL. New, improved, TSL libraries were created for polyethylene, lucite, ice-1h, and quartz; and can be seen in [4].

OCLIMAX + NJOY2016 METHODOLOGY

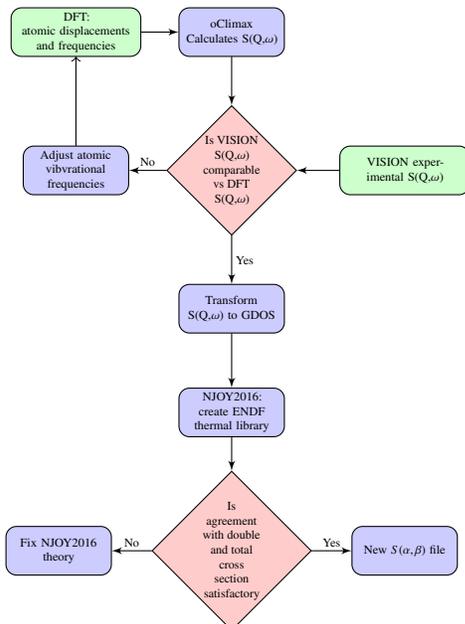The proposed method for generation of new TSL libraries can be seen in Figure 1.

Fig. 1: The proposed method for production of new thermal scattering libraries.

As it can be seen from the Figure 1, the first step is to perform a density functional theory, DFT, calculation, on the desired moderator material to obtain atomic displacements and frequencies. The details and the input files of DFT calculations, for polyethylene, lucite, ice-1h, and quartz, performed by our group can be seen in [4]. The output from the DFT calculation is processed with oClimax [5] to calculate S(Q,ω) that is comparable to S(Q,ω) measured at VISION spectrometer. It is important to note that oClimax has the ability to process the results of most popular DFT codes used today: VASP, CASTEP, QUANTUM ESPRESSO, etc. The comparison, between experimental VISION S(Q,ω) and S(Q,ω) calculated using DFT+oClimax for polyethylene, can be seen in Figure 2.
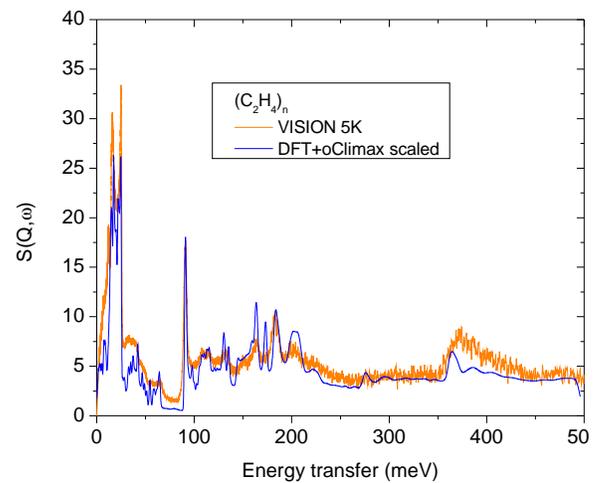

Fig. 2: Comparison of the experimental $(C_2H_4)_n$ S(Q,ω) vs. S(Q,ω) calculated using DFT and oClimax.

If the locations of the peaks match in both DFT+oClimax S(Q,ω) and the experimental one is, the next step is to transform S(Q,ω) to generalized density of states (GDOS, or sometimes referred to as phonon spectrum):

$$S(Q, \omega) = \frac{\hbar^2 Q^2}{6M\omega} exp\left(-\langle u^2 \rangle Q^2\right) G(\omega)\left[n(\omega, T) + \frac{1}{2} \pm \frac{1}{2}\right], \quad (1)$$

$$n(\omega, T) = \frac{1}{exp(\frac{\hbar\omega}{kT}) - 1}, \quad (2)$$

where $G(\omega)$ is GDOS, $\hbar\omega$ is energy of scattered neutron (in units of meV), M is the atomic mass (for mono-atomic material) of the scattering material, $n(\omega, T)$ is population Bose factor, $\langle u^2 \rangle$ is the atomic mean-square displacement (MSD), and + or - should be taken in the squared brackets for neutrons scattered with energy loss or energy gain, respectively. The comparison between fundamental vibrational mode GDOS (n=1) and GDOS containing up to ten overtones (phonon expansion) can be see in Figure 3.
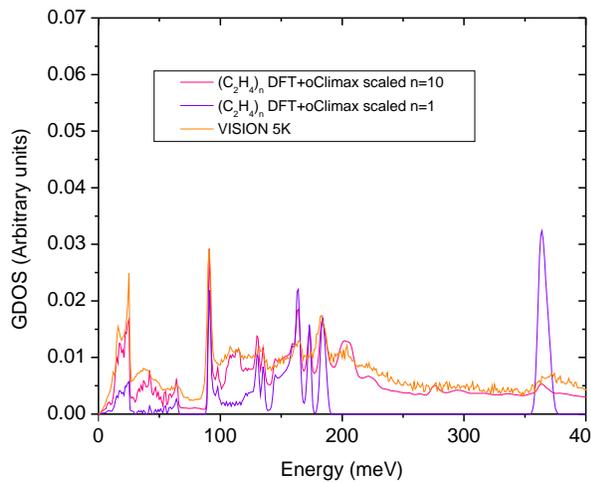
Fig. 3: Comparison of the experimentally derived $(C_2H_4)_n$ GDOS n=1 vs. GDOS with n=10 calculated using DFT and oClimax.

Fig. 4: Total cross section of $(C_5O_2H_8)_n$.

oClimax offers additional advantages due to the capability of oClimax to calculate partial contributions of each atom to the whole molecule GDOS, while at the same time calculating the fundamental vibrational mode (n=1) and multiple phonon scattering contributions (n>1 overtones), which can be seen in Figure 3. It can be also observed that the phonon expansion is responsible for creation of new peaks in the region between 200 and 300 meV. Furthermore, oClimax enables scaling of frequencies in $S(Q,\omega)$ spectra to correct the mismatch created by structural differences of the experimental sample and how the material is simulated using DFT. $S(Q,\omega)$ shown in Figure 2 has been already scaled to match the experimental frequencies.

Due to the theoretical framework of NJOY2016, which in order to create a TSL library requires fundamental vibrational mode GDOS, the next step in TSL generation is to take the n=1 GDOS for the desired material and feed it into LEAPR module of NJOY2016 [6]. The reason for this is because NJOY2016 theoretical framework has its own phonon expansion formalism to calculate n>1 vibrational modes. After the library was created it needs to be validated against the experimental total cross section measurements and newly experimentally measured DDSCS from ARCS spectrometer. To show the versatility of the proposed method and to show the importance of phonon expansion for different moderator materials, the total cross section comparison for the lucite library created by RPI Nuclear Data group can be seen in Figure 4, while the DDSCS comparison can be seen in Figure 5.

As it can be seen from Figures 4 and 5, the newly created RPI lucite library improves upon the total cross section calculation, while also improving the DDSCS calculation. From Figure 4 it can be observed that neither RPI library, nor the ENDF/B-VIII.0 library created by NCSU Nuclear Reactor Program group using their own in-house developed code FLASSH, is able to calculate the structure between 200 and 300 meV correctly. This leads us to believe that either the incoherent
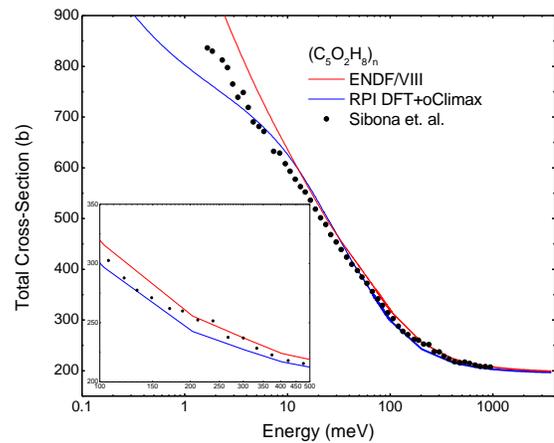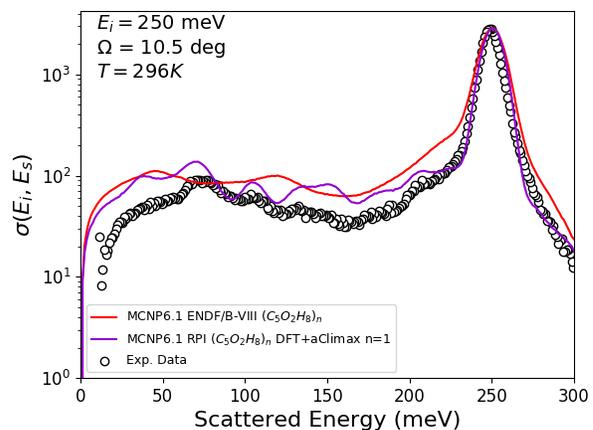

Fig. 5: DDSCS comparison for $(C_5O_2H_8)_n$ at the 250 meV incident energy at scattering angle 10.5 degree.

approximation or the phonon expansion in NJOY2016 is the cause of small discrepancies that still exist. If one looks at Figure 3 for polyethylene, which is similar to lucite, we can see that phonon expansion is indeed responsible for creation of the new extra peaks in the region between 200 and 300 meV. Hence we conclude that the phonon expansion is probably responsible for the most of the discrepancies seen in total cross section and DDSCS comparisons. The same issue is present for polyethylene too, which can be seen in Figure 7. More details on the phonon expansion issue is given in [4], where also the theory behind each code and step in the proposed methodology can be seen. It is also important to emphasize the use of the experimental data in the process of creation of new TSL libraries. Figure 6 shows comparison between polyethylene n=1 GDOS obtained from the proposed method and ENDF/B-VIII.0 library.

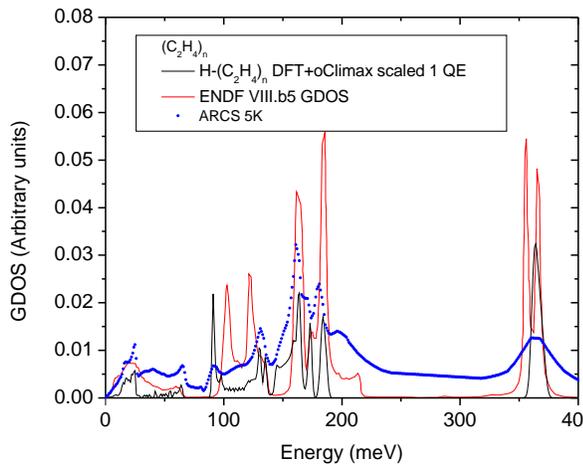The RPI ARCS 5K GDOS was derived from measurements, performed by the RPI Nuclear Data group, at ARCS

Fig. 6: Comparison of $(C_2H_4)_n$ ENDF/B-VIII.b5 GDOS, DFT+oClimax GDOS, and RPI ARCS GDOS.

spectrometer. From Figure 6, it can be observed that the agreement in locations of the peaks between ARCS 5K GDOS and RPI DFT+oCLimax GDOS is near perfect. Both of these GDOS were derived from experimental data. It can be also observed that ENDF/B-VIII.0 GDOS is arguably physically incorrect (incorrect locations of the peaks), but if one looks at the plot for total cross section of polyethylene in Figure 7, ENDF/B-VIII.0 library still calculates the total cross section correctly.
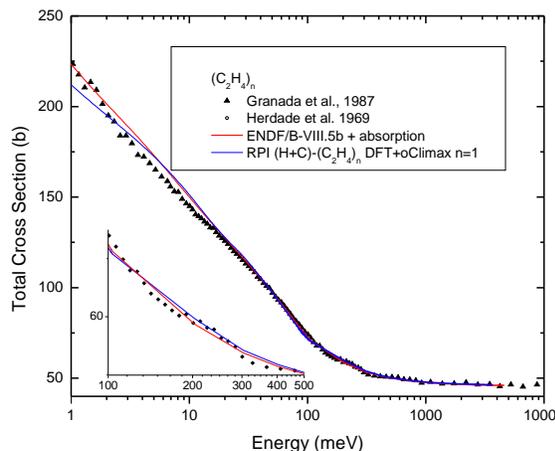

Fig. 7: Total scattering cross section of $(C_2H_4)_n$.

CONCLUSIONS

To summarize, RPI Nuclear Data group has developed a new methodology for creation of new TSL libraries for different moderator materials where the experimental data are used to validate and guide the calculation of phonon spectrum using DFT and oClimax. In practice, any output from molecular dynamics calculation can be used with oClimax to derive the phonon spectrum. The advantages of using oClimax to derive the phonon spectrum are multiple, among which are the ability to calculate partial contributions, for each atomic species, to the phonon spectrum, while also calculating fundamental vibrational mode (n=1) to be used with NJOY2016. We have also shown that phonon expansion is probably the reason for the inaccuracy in total cross section and DDSCS calculations. This work is ultimately expected to result in proposed corrections to the NJOY2016 phonon expansion implementation.

ACKNOWLEDGMENTS

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