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SOURCE

Thermal neutron scattering evaluation for polyethylene from RPI/ORNL/ESS

2021 CSEWG Meeting

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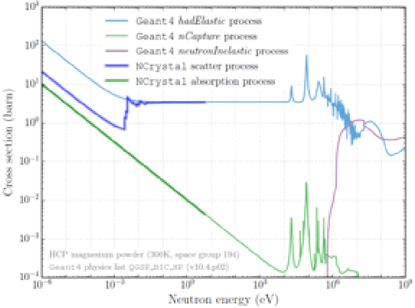
11-17-2021

Introduction

- In the absence of phonon spectrum (VDOS) based scattering kernels, thermal neutron scattering can be sampled using:

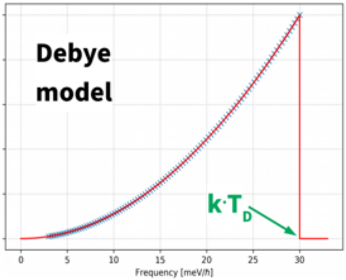
a) Free gas model:



b) Debye model:

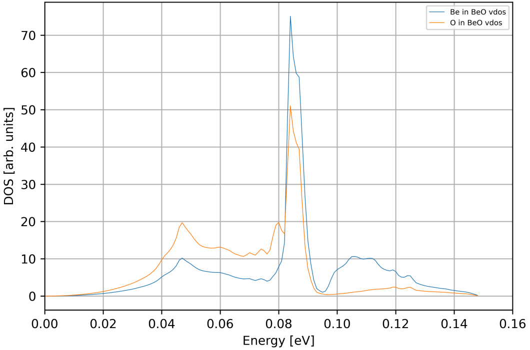
Idealized DOS (Debye Model) is constructed and fed into same infrastructure as any other DOS.

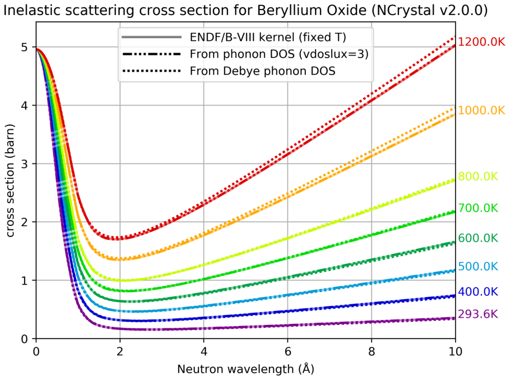
Lacks details of course, but gives consistent kinematics and handles multi-phonon physics rather well.



Introduction

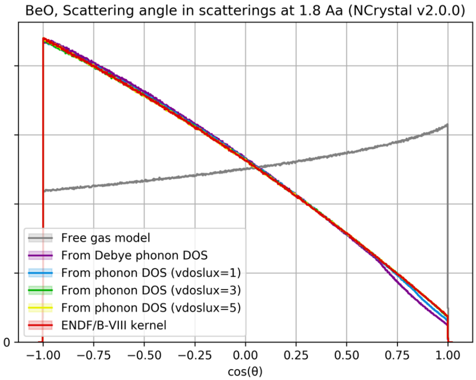
- But with the availability of the phonon spectrum:

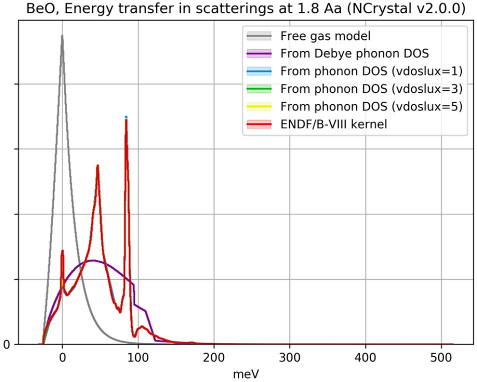




Introduction

- But only phonon based scattering kernels correctly calculate microscopic quantities:





- **The scattering kernel with best predictive capabilities will have an accurate calculation of integral and microscopic quantities as well.**

Inelastic Physics

In the incoherent and Gaussian approximation, the $S(\alpha,\beta)$ is:

$$S(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{i\beta\hat{t}} e^{-\gamma(\hat{t})} d\hat{t} \tag{1}$$

where:

$$e^{\gamma(\hat{t})} = \alpha \int_{-\infty}^{\infty} P(\beta)[1 - e^{i\beta\hat{t}}]e^{-\beta/2} d\beta \tag{2}$$

$$P(\beta) = \frac{\rho(\beta)}{2\beta\sinh(\beta/2)} \tag{3}$$

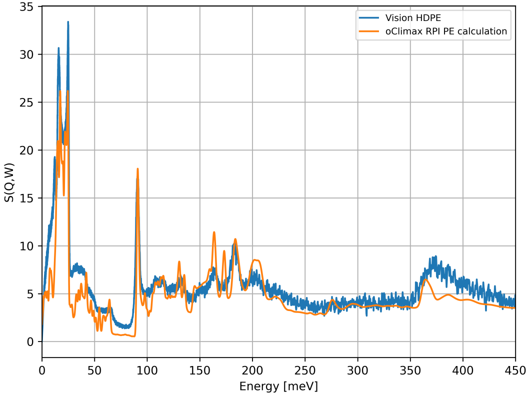
with $\rho(\beta)$ as the phonon spectrum. Equation 1 in terms of phonon expansion can be written as:

$$S(\alpha,\beta) = e^{-\alpha\lambda}\sum_{n=0}^{\infty}\frac{1}{n!}\alpha^n\frac{1}{2\pi}\times\int_{-\infty}^{\infty} e^{i\beta\hat{t}}\left[\int_{-\infty}^{\infty} P(\beta')e^{i\beta'\hat{t}}e^{-\beta'/2}d\beta'\right]^n d\hat{t} \tag{4}$$

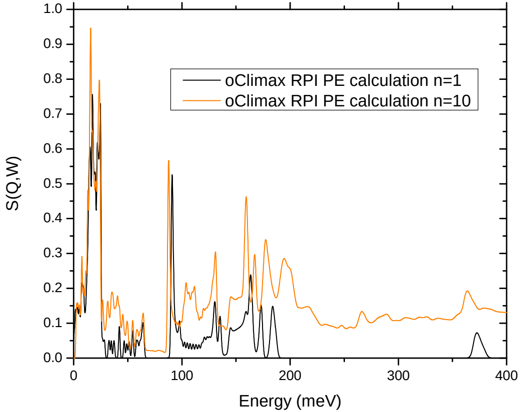
All the modeling techniques calculate only fundamental vibrational mode (n=1) phonon spectrum!!!

- **https://doi.org/10.1016/S1369-7021(05)71336-5 - "Neutron scattering and hydrogenous materials"**

Revisiting Polyethylene

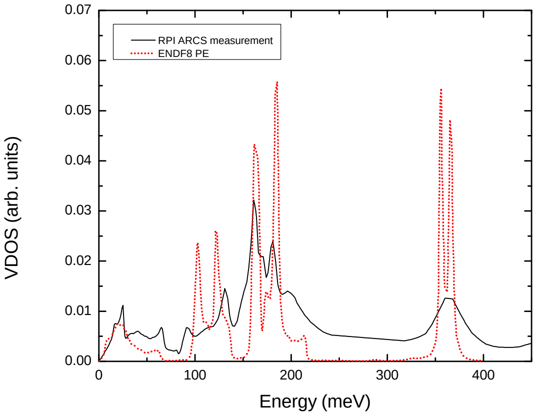


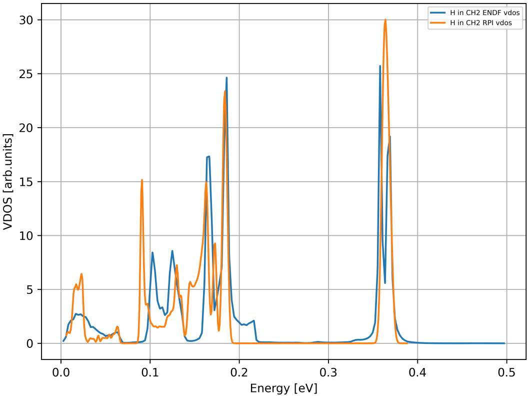
- Comparison with experimental S(Q,ω) from VISION instrument



- Fundamental vibrational mode vs multiple phonon contributions

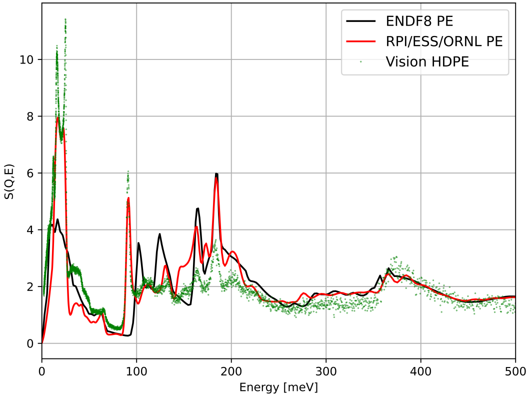
Revisiting Polyethylene





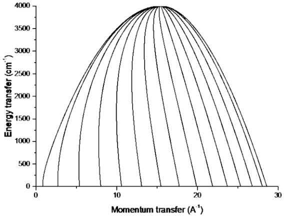
- **Experiments measure phonon spectrum with multiple phonon contributions included (n››1)!!!**

- Can ENDF phonon spectrum result in the same S(Q,ω) as measured at VISION?

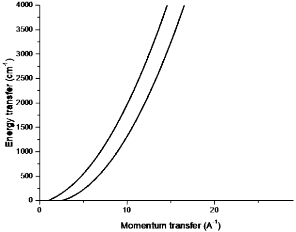


- New method by Dr. Chapman extracts $S(\alpha, \beta)$ from an ENDF file at a trajectory in $S(Q, \omega)$ phase space that corresponds to one neutrons would travel at VISION instrument at SNS at ORNL, converts it to $S(Q, \omega)$, and applies instrumental resolution.

- Direct geometry instrument S(Q,ω) phase space (ARCS or SEQUOIA):

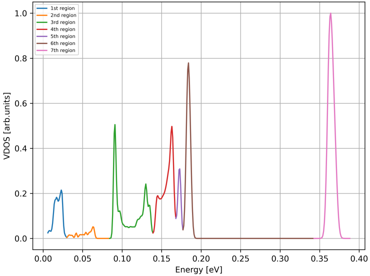


- Indirect geometry instrument S(Q,ω) phase space (VISION):



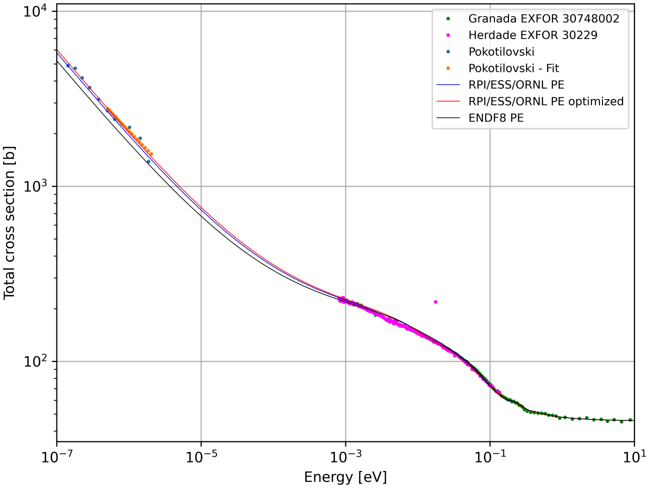
Revisiting Polyethylene

- New RPI Polyethylene total cross section data measured typical High Density Polyethyelene (HDPE).
- We can separate the VDOS into distinct regions and optimize until agreement with the cross-section matched as best as possible.

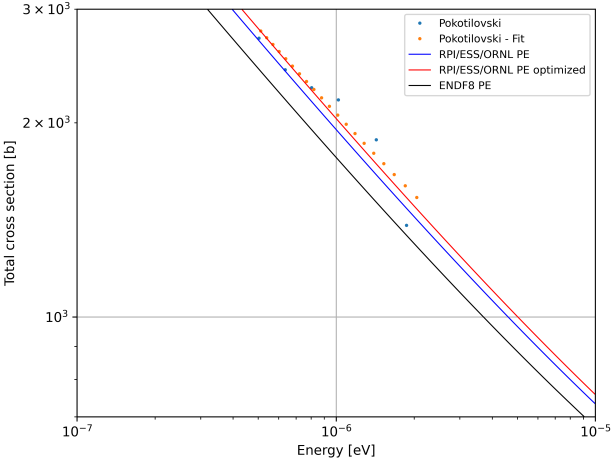


- We used NCrystal and Dakota to minimize χ^2 between the RPI new data and the evaluation. The weights for each region where changed by Dakota, total cross section is calculated by NCrystal at exactly same energy grid as the experimental cross section data, and χ^2 was calculated. The process was repeated until optimal minimal difference in χ^2 was achieved.

HDPE total cross section (1e-7 to 10 eV)

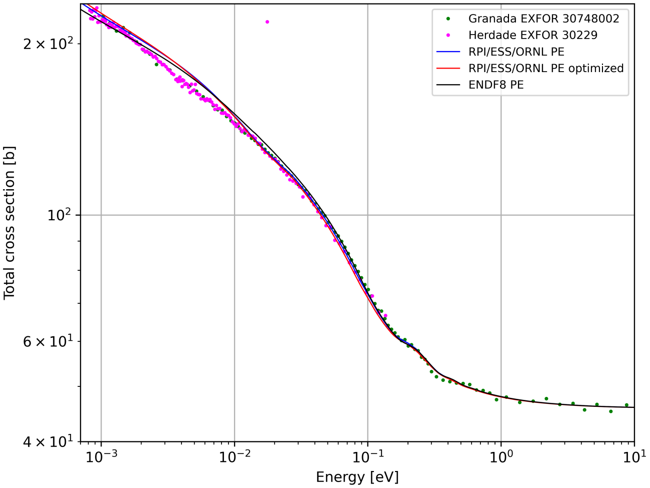


HDPE total cross section (1e-7 to 1e-5 eV)

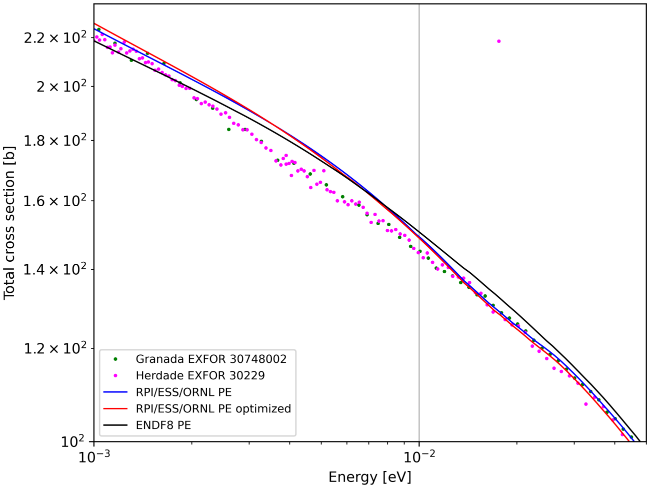


- Y. N. Pokotilovski et al., "A differential time-of-flight spectrometer of very slow neutrons", Instruments and Experimental Techniques, vol. 54, pp. 16–22, 2011

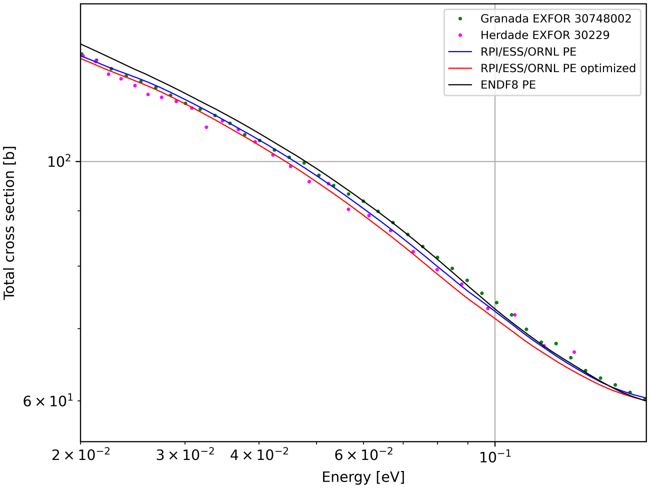
HDPE total cross section (1e-3 to 10 eV)



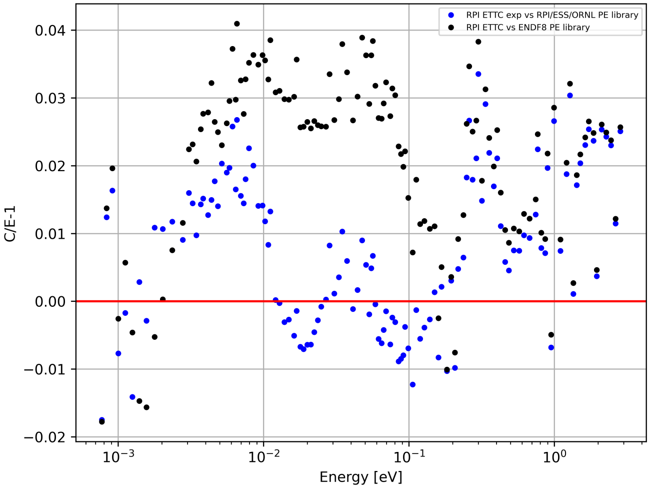
HDPE total cross section (1e-3 to 5e-2 eV)



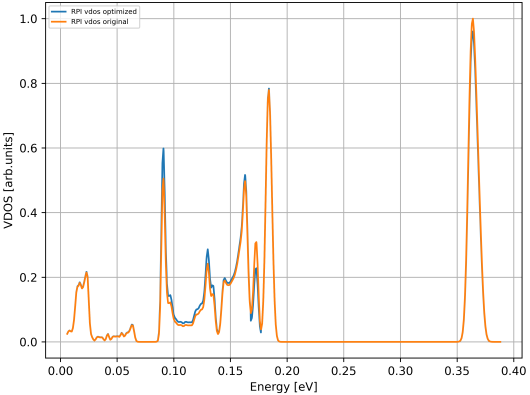
HDPE total cross section (2e-2 to 2e-1 eV)

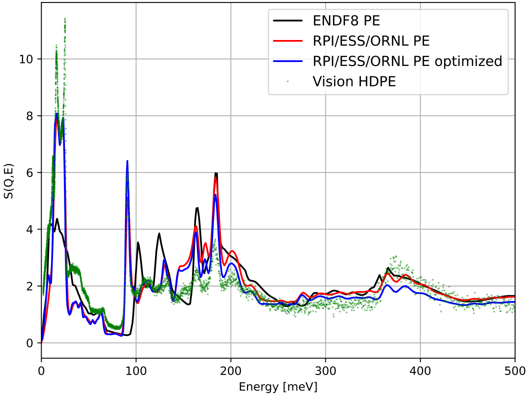


RPI experiment HDPE C/E-1



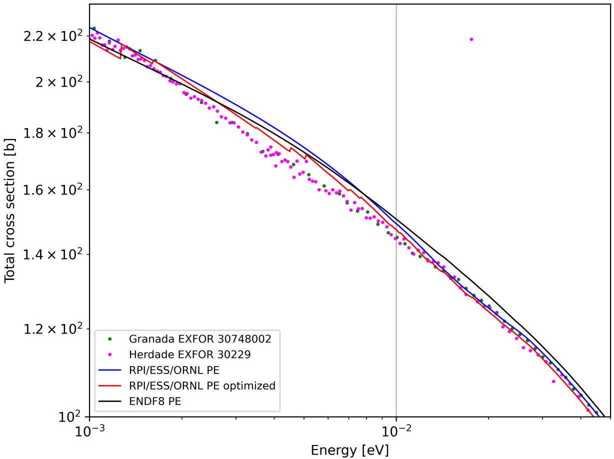
Microscopic level





- RPI/ESS/ORNL PE scattering kernel has a better predictive capabilities on both microscopical and integral level.

Polyethylene as a perfect crystal



- Polyethylene is after all crystalline material, so coherent elastic effects should be taken into account. HDPE is up 80% crystaline material.

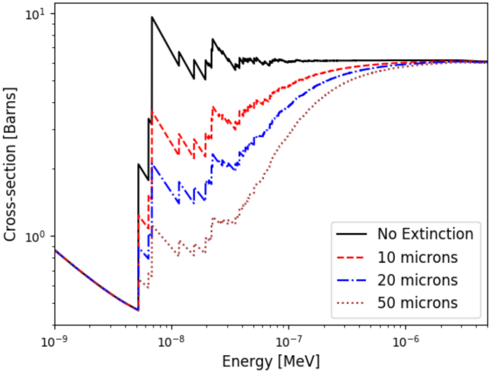
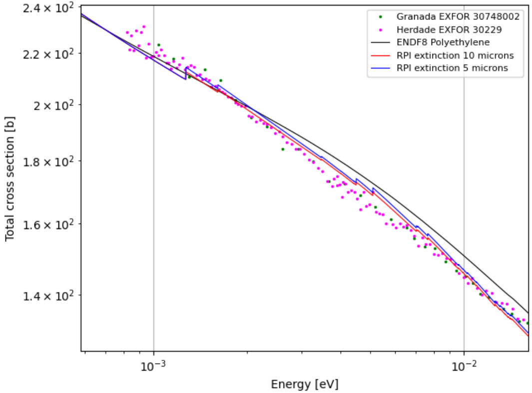


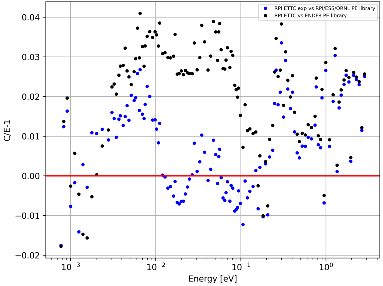
Fig. 1. Neutron cross-section for beryllium as a function of different crystallite sizes.

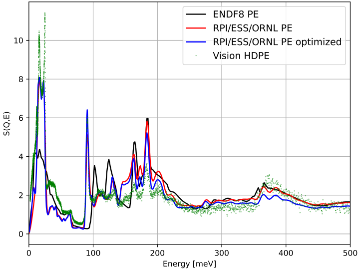


- Because Polyethylene isn't made of perfect crystals, extinction could be a possible explanation.
- D. D. DiJulio et al., "Impact of crystallite size on the performance of a beryllium reflector", Journal of Neutron Research, vol. 22, no. 2-3, pp. 275-279, 2020

Conclusions

- The importance of microscopic quantities has been demonstrated for the accurate predictability of thermalization of neutrons.
- RPI Polyethylene library has been revisited (merged into Phase 1, with C in CH2 for completness) and shown to be in the best agreement with available relevant experimental data.
- A new path for validation of tsl-ENDF files has been proposed starting from phonon measurements to total cross section measurements. Tools such as VISION, ARCS and SEQUOIA spectrometers at ORNL, should be part of validation and evaluation of thermal scattering libraries.





Thanks for your time.
Questions?