Implementation of TSL Evaluations Beyond the Incoherent Approximation

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Outline

- Motivation
- Thermal Scattering Theory
- Generalized Thermal Scattering Law
- Relaxing the Incoherent Approximation
- Application & Validation
- Summary & Conclusions

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Motivation

- Production of TSL libraries that contain high fidelity physics including coherent inelastic effects
  - Graphite and beryllium
    - Highly non-cubic forces
    - Non-negligible structure in TSL
- High fidelity TSL libraries are of interest in various application, e.g.
  - Neutron scattering instrument response analysis
  - Directional biases (reflection) in neutronic analysis

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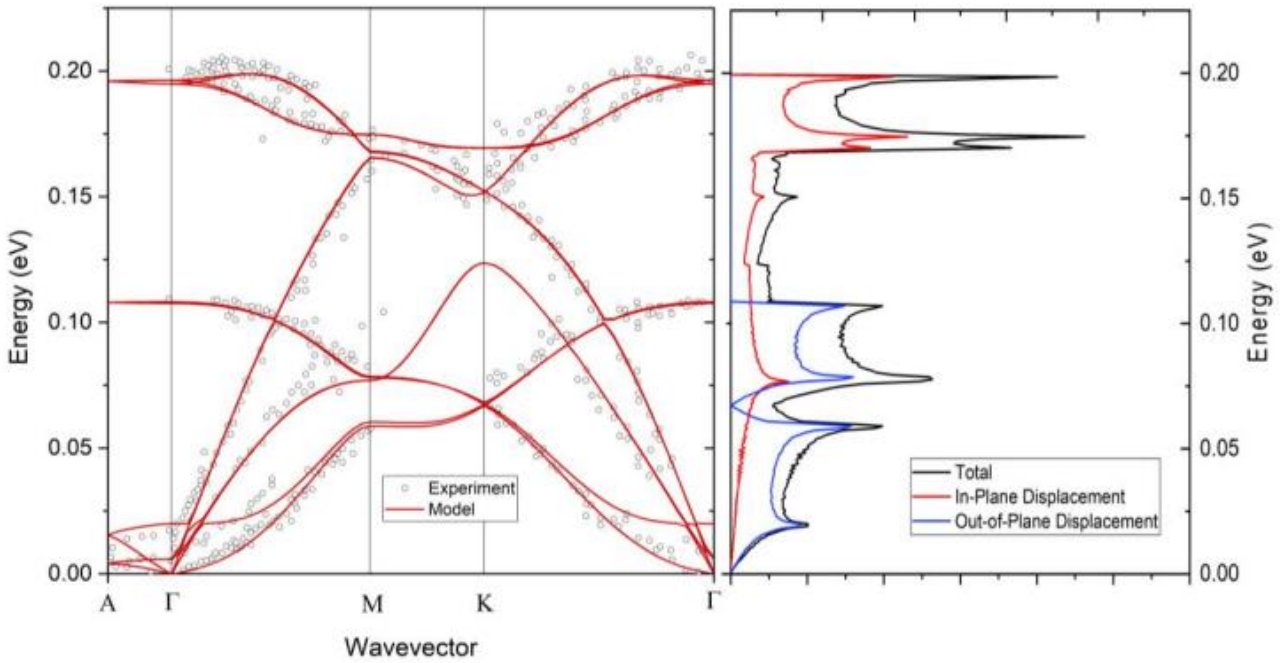
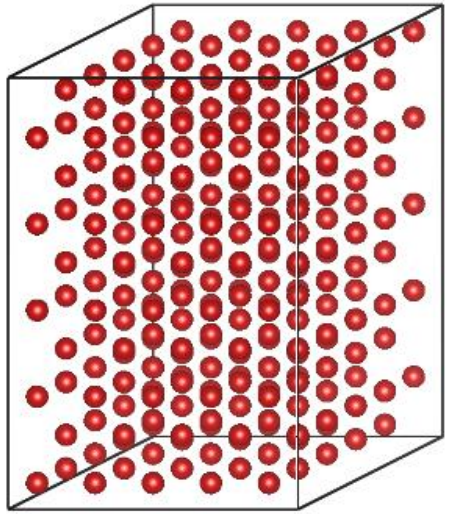
Graphite & Beryllium Structures

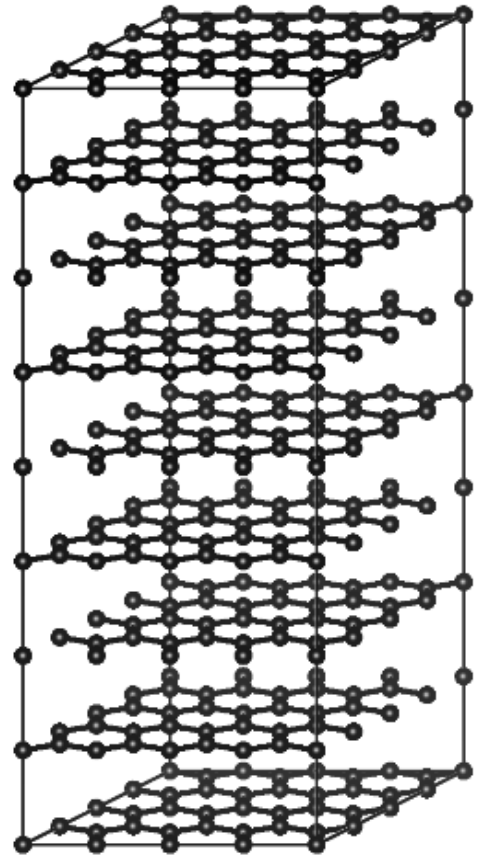
- HCP crystals
- Graphite has highly directional bonding
  - Covalent in-plane
  - Van der Waals out-of-plane
  - Preferred Orientation



Phonon dispersion curves and vibrational DOS for crystalline graphite, calculated using *ab initio* methods.



Beryllium 5x5x5 supercell (250 atoms)



Crystalline Graphite 4x4x4 supercell (256 atoms)

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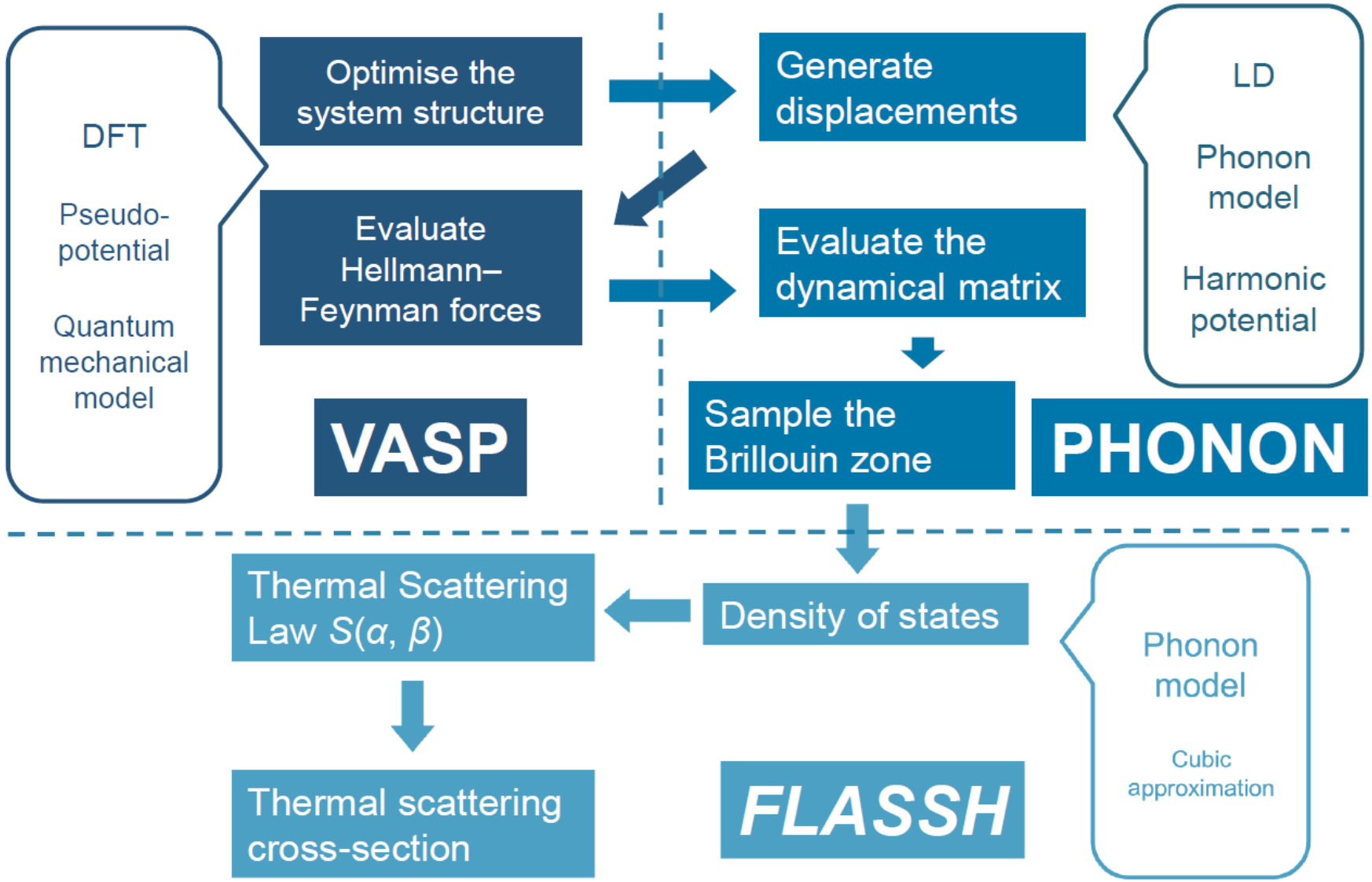
LEIP Evaluation Process



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Thermal Scattering Theory

- Thermal Scattering Law (TSL)
  - Material property
  - Energy & Momentum States
  - $S(\alpha,\beta) = S(\boldsymbol{\kappa},\omega)\cdot k_B T$

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'} = \frac{1}{4\pi}\sqrt{\frac{E'}{E}}\left[\sigma_{coh}S(\alpha,\beta) + \sigma_{inc}S_s(\alpha,\beta)\right]$$

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

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

Correlations in Scattering Theory

- Thermal neutron scattering is defined by atomic correlations

$$S(\alpha,\beta) = \cdots \int G(\boldsymbol{r},t) \ldots \mathrm{d}\boldsymbol{r}\mathrm{d}t$$

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

- The self-component
  - S_s
  - Correlates an atom with itself
- The distinct-component
  - S_d
  - Correlates an atom with the entire system
  - Lattice (Structure) effects

Typical TSL Approximations

- Incoherent Approximation ($S_d = 0$)
  - Assumes the distinct component is negligible
- Cubic Approximation
  - Assumes isotropic forces (non-directional)
  - Impacts inelastic and elastic scattering
- Atom Site Approximation
- Harmonic & Gaussian Approximations
  - Allows for use of the phonon expansion
  - Simplifies correlation functions

Removing the Cubic Approximation: Generalization

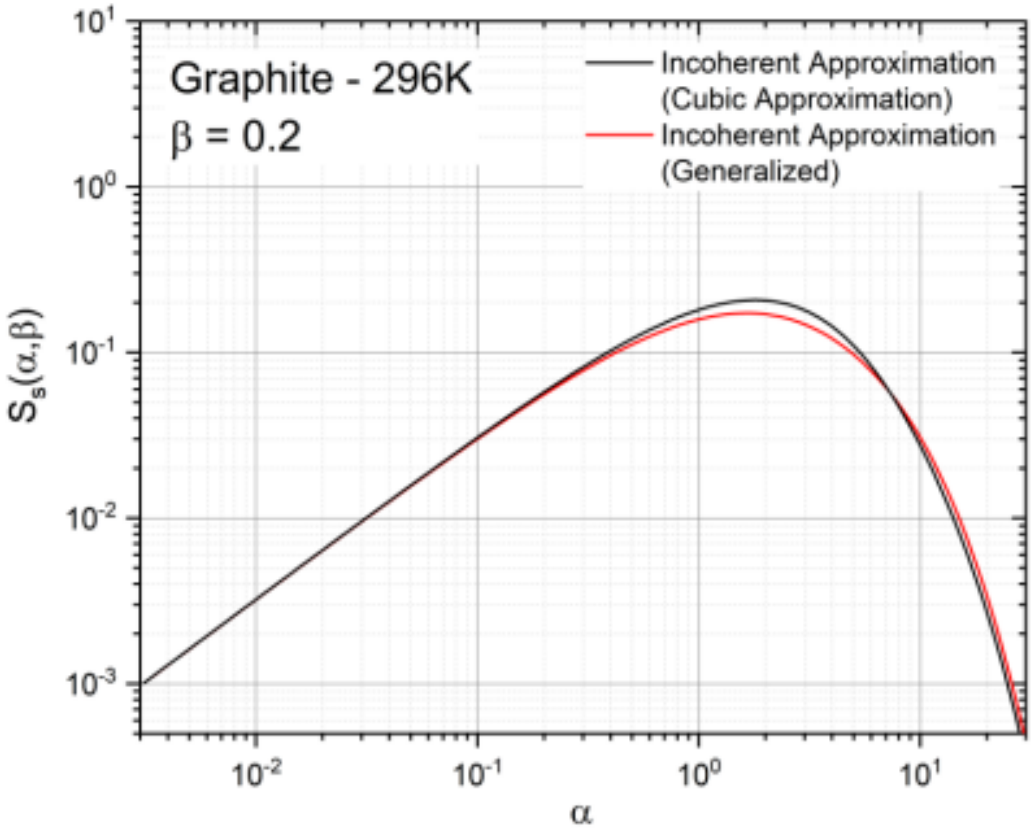
- This approximation directionally-averages the forces in a crystal
  - Application of the approximation must be consistent throughout the evaluation (inelastic, elastic)

$$\sum_s (\boldsymbol{\kappa} \cdot \boldsymbol{e}_s)^2 \Rightarrow \frac{1}{3} k^2$$

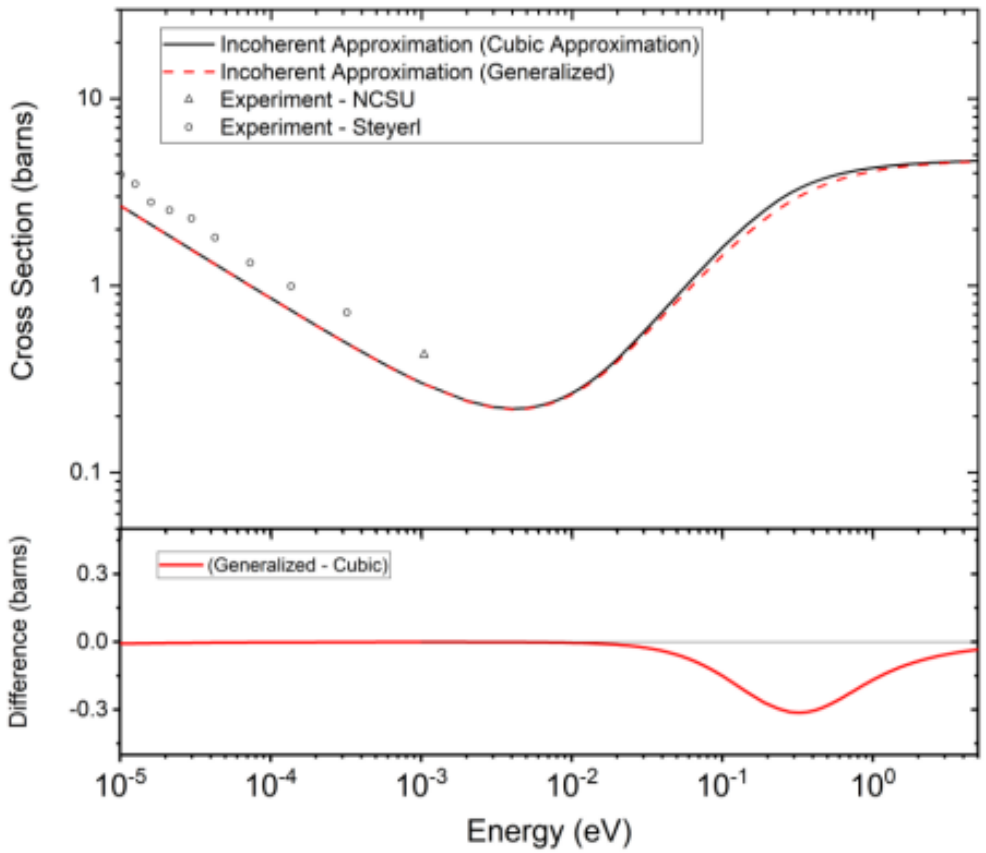
Generalized (exact): $$\langle U^2 \rangle_m = -2W_m = -\frac{\hbar}{2N} \sum_s \frac{1}{\omega_s} \frac{|\boldsymbol{\kappa} \cdot \boldsymbol{e}_s^m|^2}{M_m} \coth\left(\frac{\hbar\omega_s}{2k_B T}\right)$$

Approximated: $$\langle U^2 \rangle = -\frac{\hbar k^2}{2M} \int_0^{\omega_m} \frac{\rho(\omega)}{\omega} \coth\left(\frac{\hbar\omega_s}{2k_B T}\right) \mathrm{d}\omega$$

Removing the Cubic Approximation: Generalization

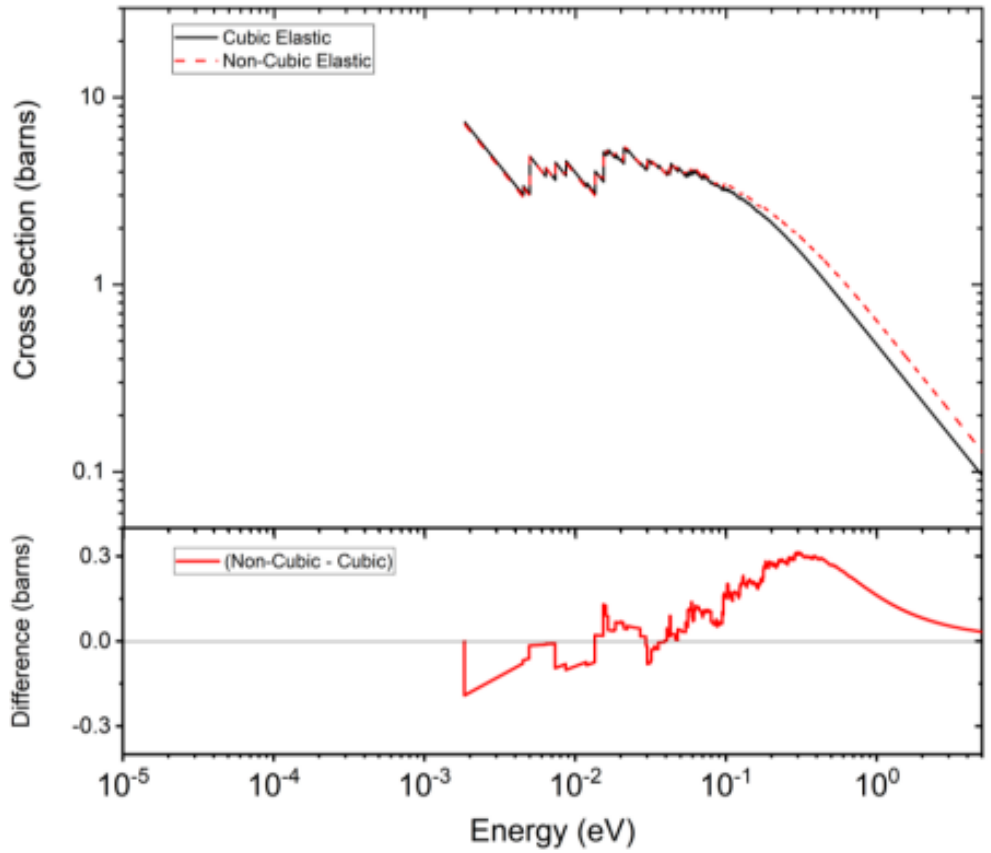


Self-component of the TSL of crystalline graphite at a specific β value under the cubic and generalized treatments

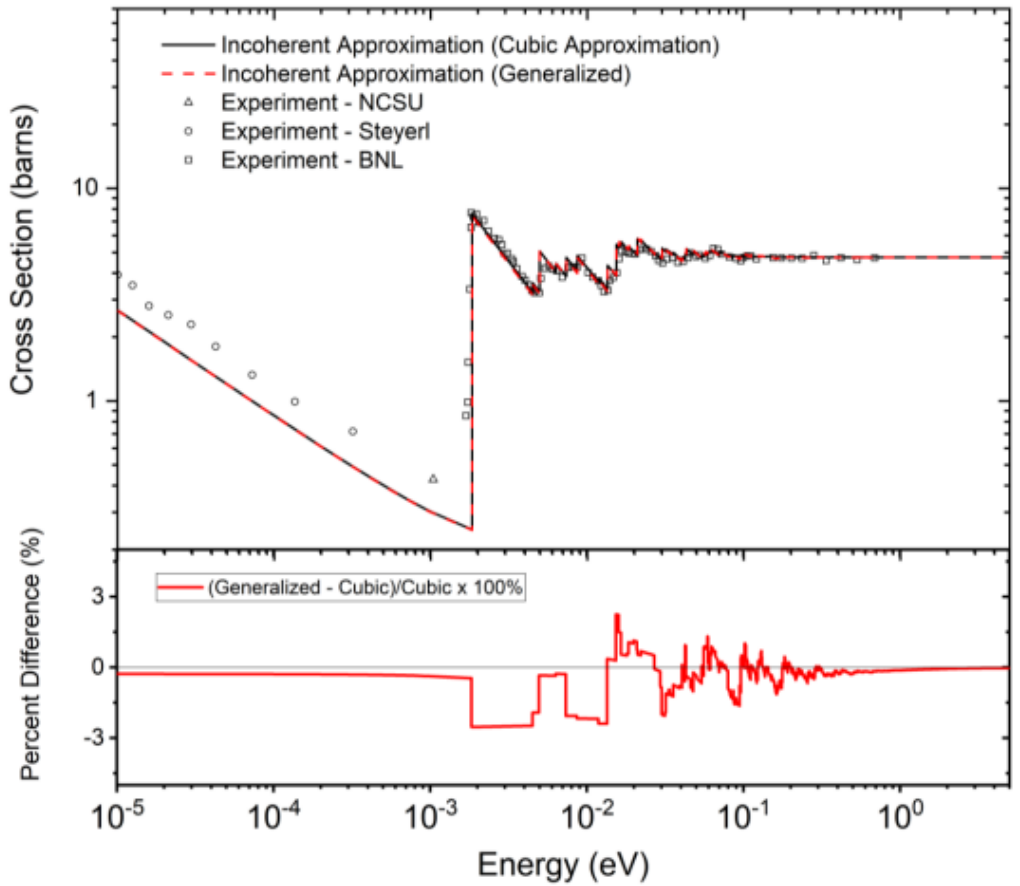


Inelastic elastic scattering cross section of crystalline graphite under the cubic and generalized treatments

Removing the Cubic Approximation: Consistency



Coherent elastic scattering cross section of crystalline graphite under the cubic and generalized treatments



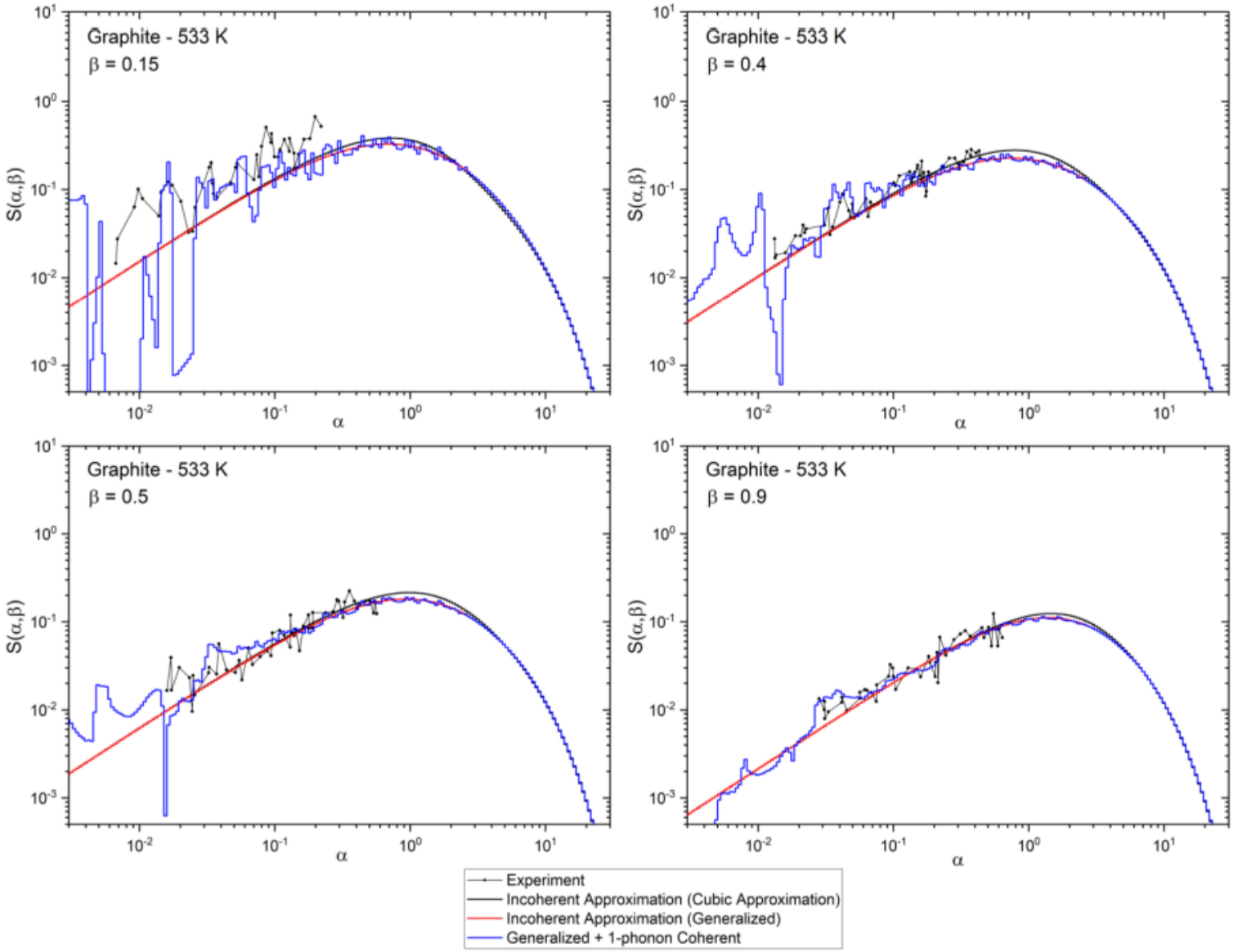
Total scattering cross section of crystalline graphite under the cubic and generalized treatments

Beyond the Incoherent Approximation

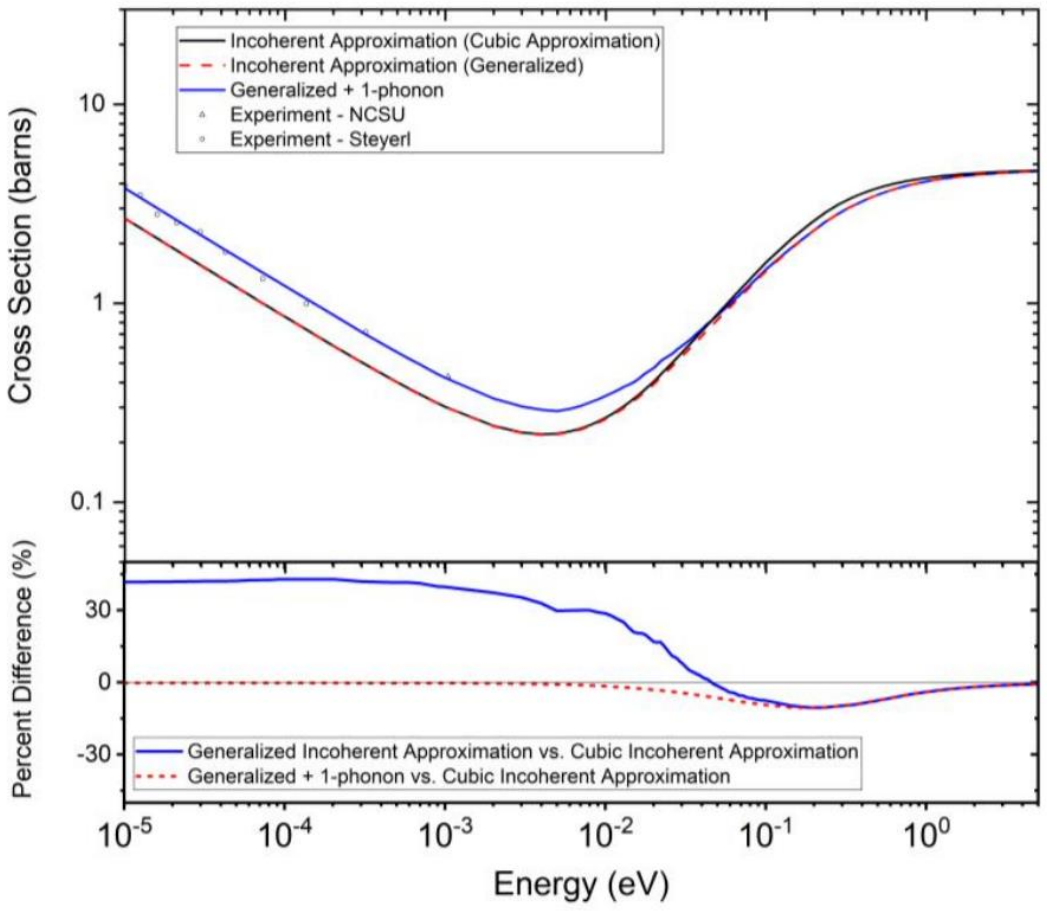
	Incoherent	Atom Site
1-Phonon / Generalized:	$S(\alpha,\beta) = S_s(\alpha,\beta) + S_d^1(\alpha,\beta)$ $\left(\frac{\partial^2\sigma}{\partial\Omega\partial E'}\right)_{inelastic} = \frac{1}{4\pi}\frac{k'}{k}([\sigma_{inc}+\sigma_{coh}]S_s + \sigma_{coh}[S^1 - S_s^1])$	$S(\alpha,\beta) = \sum_m S_m(\alpha,\beta)$
Approximation:	$S(\alpha,\beta) = S_s(\alpha,\beta)$ $\left(\frac{\partial^2\sigma}{\partial\Omega\partial E'}\right)_{inelastic} = \frac{1}{4\pi}\frac{k'}{k}(\sigma_{coh}+\sigma_{inc})S_s$	$S(\alpha,\beta) = mS(\alpha,\beta)$

Beyond the Incoherent Approximation

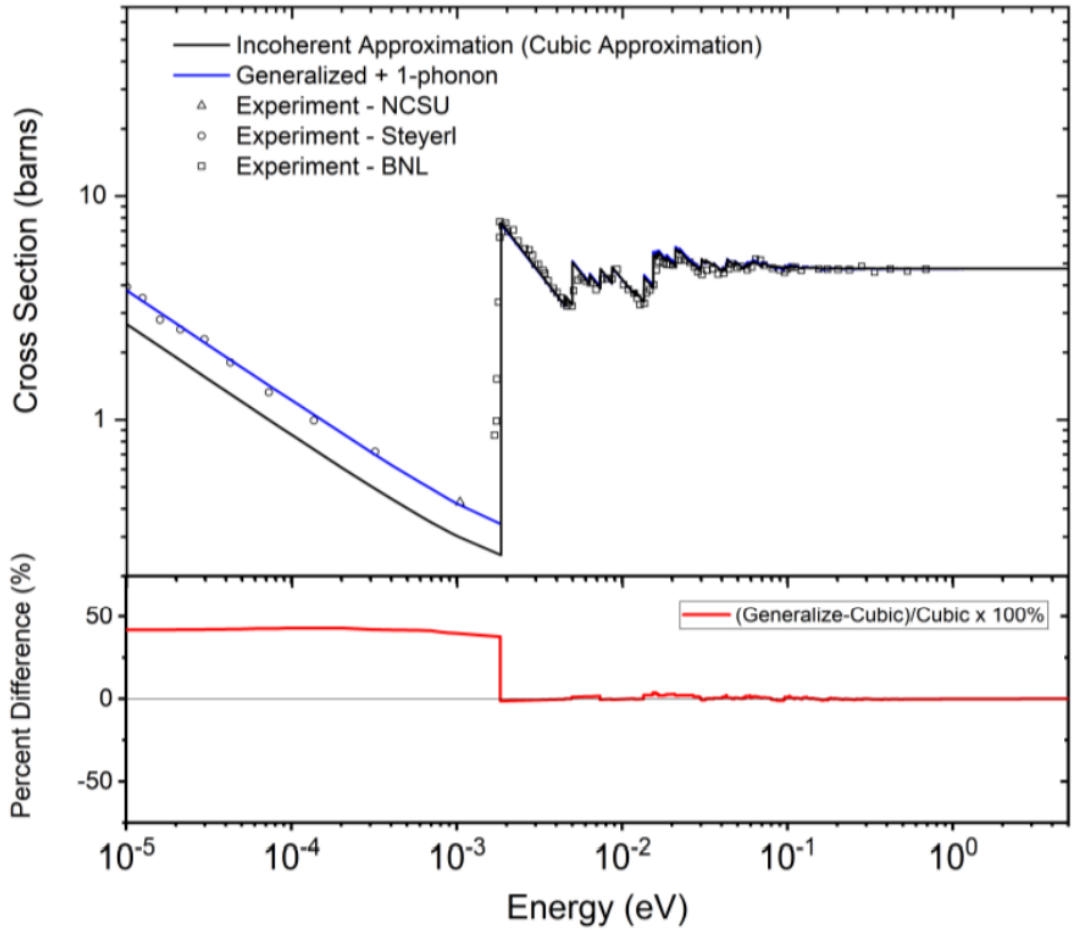
TSL of crystalline graphite at various β values



Beyond the Incoherent Approximation



Inelastic cross section of crystalline graphite under the cubic, generalized, and generalized + 1-phonon treatments



Total cross section of crystalline graphite under the cubic, generalized, and generalized + 1-phonon treatments

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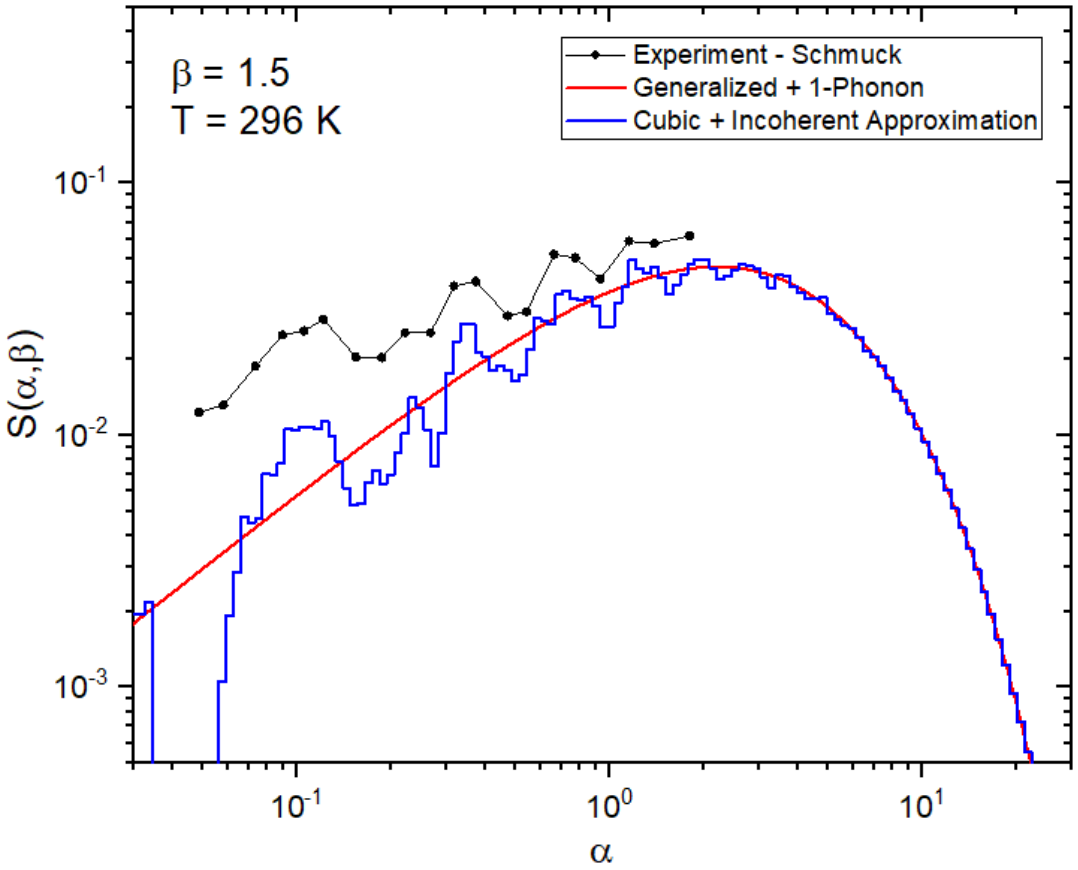
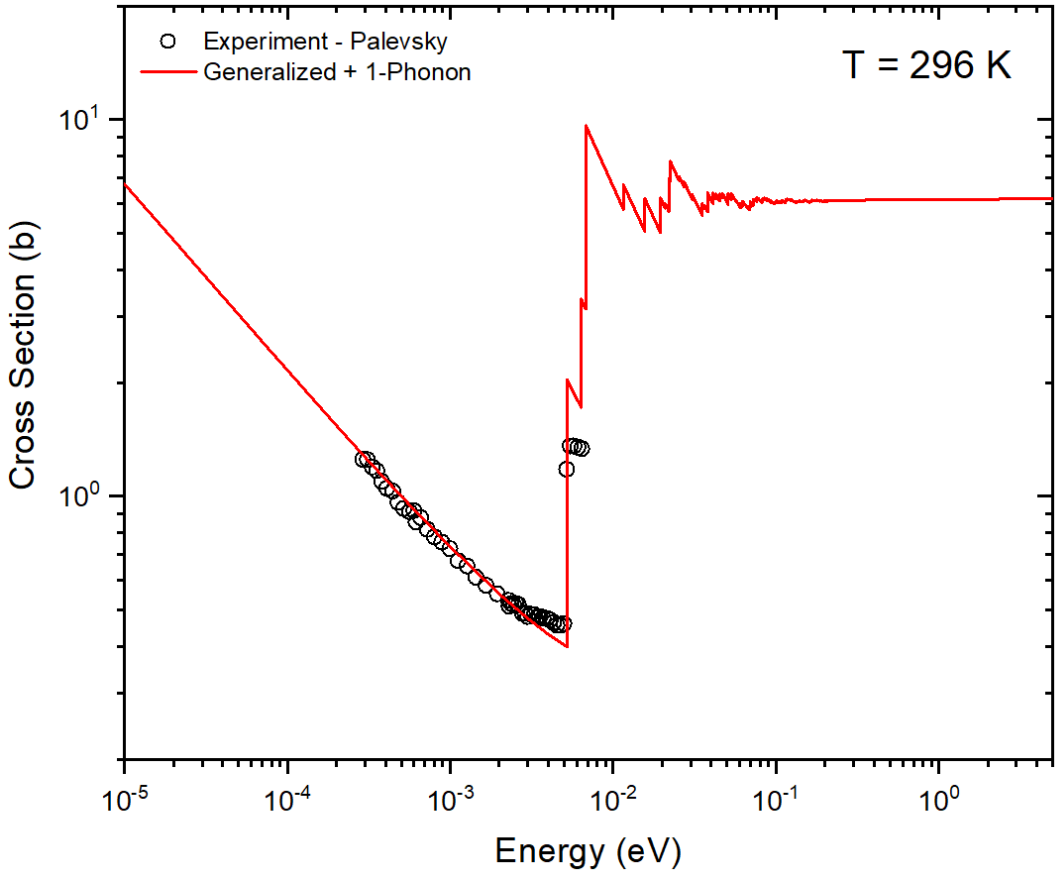
Application to Beryllium



Inelastic cross section of beryllium under the cubic and generalized + 1-phonon treatments



Total cross section of beryllium under the cubic and generalized + 1-phonon treatments

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Generation of File 7

- File 7 formatting remains the same
  - Physics changes embedded in the usual data
    - e.g., $S(\alpha,\beta) = S_s(\alpha,\beta) + S_d^1(\alpha,\beta)$

File 7s for beryllium + S_d crystalline graphite + S_d as submitted to ENDF/B-VIII

```
 1.250000+2 8.934780+0          -1           0           0           0
 0.000000+0 0.000000+0           0           0           0           6
 1.000000+0 5.000000+0           0           0          12           8
 0.000000+0 0.000000+0           0           0          38           3
Be-metal+Sd LEIP LAB  EVAL-Dec20 N.C. Fleming, A.I. Hawari
                      DIST-
----ENDF/B-VIII       MATERIAL  25
-----THERMAL NEUTRON SCATTERING
------ENDF-6 FORMAT

 Temperatures = 77 100 296 400 500 600 700 800 1000 1200 K

HISTORY
-------
This library was produced by the Low Energy Interaction Physics
(LEIP) group at North Carolina State University, USA. The
thermal scattering law data for Be-metal was developed using ab
initio lattice dynamics (AILD) [1, 2]. The coherent elastic
cross sections were calculated based on the lattice constants
obtained from the AILD beryllium structure. Distinct effects
are included in this evaluation using directional information
derived from the Be-metal AILD structure. Ten temperatures are
available in this library. The Full Law Analysis Scattering
```

```
                                                                  1 0  0
 1.280000+2 1.189800+1          -1           0           0           0  28 1451
 0.000000+0 0.000000+0           0           0           0           6  28 1451
 1.000000+0 5.000000+0           0           0          12           8  28 1451
 0.000000+0 0.000000+0           0           0          39           3  28 1451
Graphite+Sd LEIP LAB  EVAL-Dec20 N.C. Fleming, A.I. Hawari           28 1451
                      DIST-                                          28 1451
----ENDF/B-VIII       MATERIAL  28                                   28 1451
-----THERMAL NEUTRON SCATTERING                                     28 1451
------ENDF-6 FORMAT                                                 28 1451
                                                                    28 1451
 Temperatures = 296 400 500 600 700 800 1000 1200 1600 2000 K       28 1451
                                                                    28 1451
HISTORY                                                             28 1451
-------                                                             28 1451
This library was produced by the Low Energy Interaction Physics     28 1451
(LEIP) group at North Carolina State University, USA. The           28 1451
thermal scattering law data for crystalline graphite developed      28 1451
using ab initio lattice dynamics (AILD) [1, 2]. The coherent        28 1451
elastic cross sections were calculated based on the lattice         28 1451
constants obtained from the AILD graphite structure. Distinct       28 1451
effects are included in this evaluation using directional           28 1451
information derived from the crystalline graphite AILD              28 1451
structure. Ten temperatures are available in this library. The      28 1451
Full Law Analysis Scattering System Hub (FLASSH) system was         28 1451
used to produce File 7 MT = 2, 4 data for graphite [3]. The         28 1451
coherent elastic data were prepared using the cubic                 28 1451
approximation. MAT=28 and ZA=128 are used for crystalline           28 1451
graphite with distinct effects.                                     28 1451
                                                                    28 1451
REFERENCES                                                          28 1451
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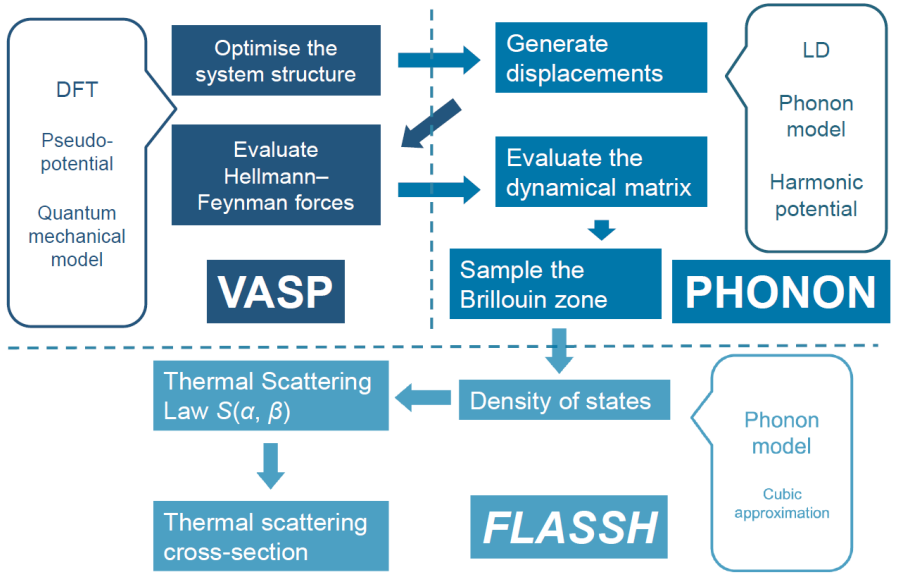
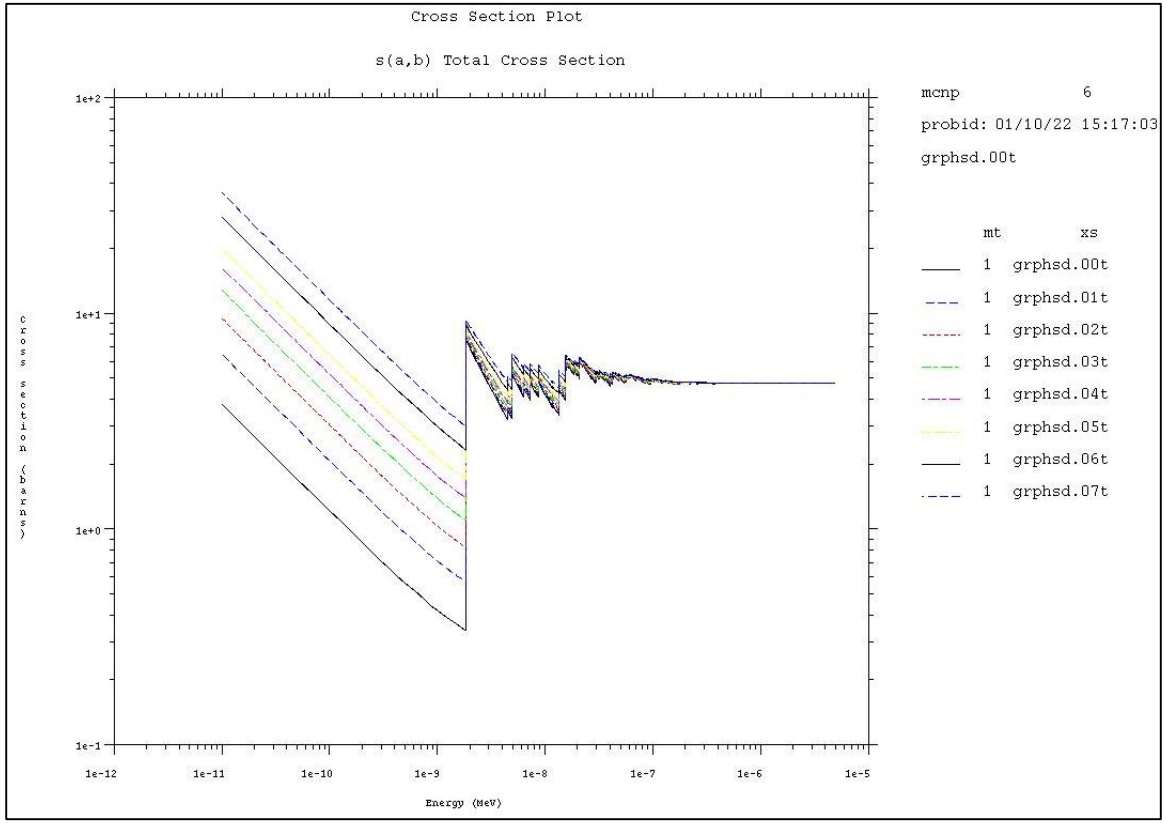
Overall Evaluation Validation

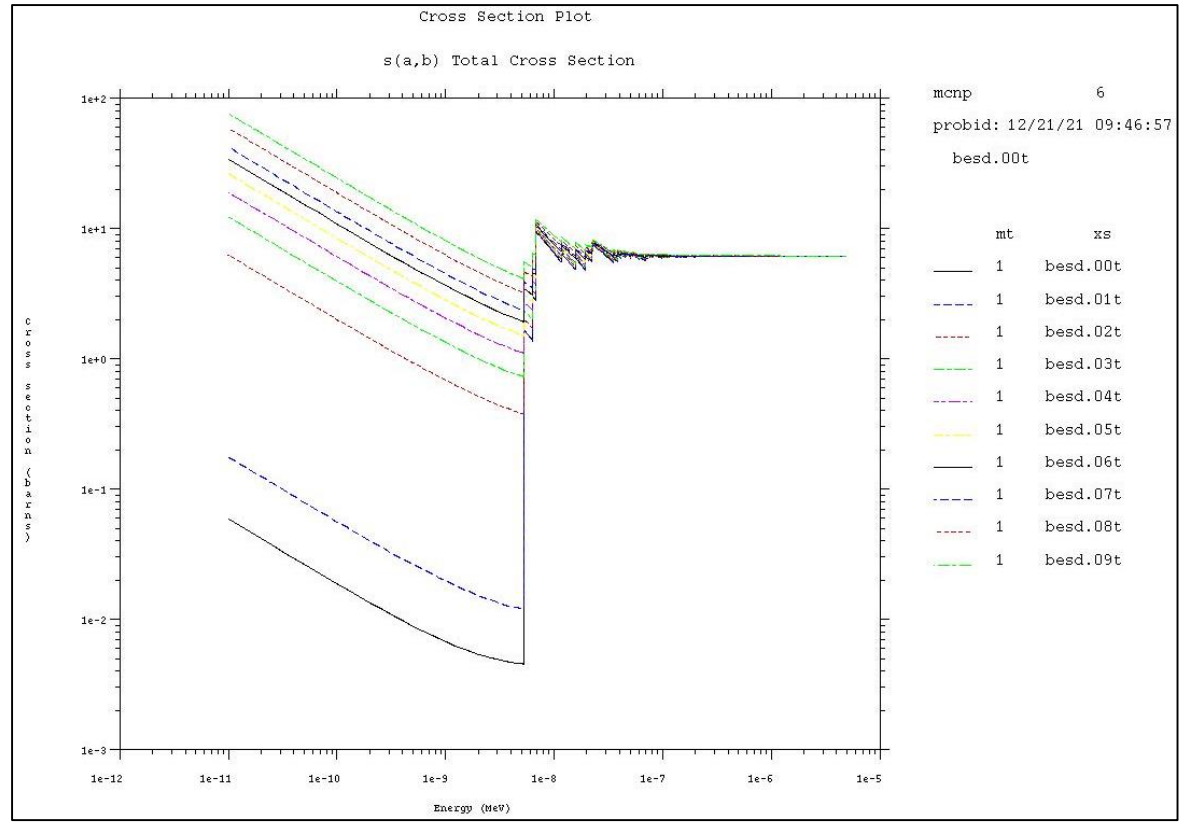
- Physically-informed gridding α- and β-space
- Natural isotopic mass
  - Obtained from ENDF/B-VIII.0 tape20
- Natural isotopic free atom cross section
  - Obtained from ENDF/B-VIII.0 tape20 (MF=3 MT=2 E=1e-5eV)
- Phonon DOS from *ab initio* DFT
- Experimental unit cell (coherent elastic)



Validation & Impact on File 7



Total cross section plots for beryllium + S_d (right) and crystalline graphite + S_d (left). Plots generated using MCNP and ACE files



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Summary

- Removal of typical approximations
  - Incoherent
  - Cubic
  - Atom Site
- Application of generalized equations to crystalline graphite and beryllium TSL evaluations
  - Both evaluations submitted to NNDC
    - Crystalline graphite + S_d^1
    - Beryllium + S_d^1
- Validation of data and data formats

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Thank You!

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