Evaluation Strategies and Pitfalls for Thermal Neutron Scattering Data

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Motivation

1. The availability of accurate thermal neutron scattering data is very important in many reactor, nuclear criticality safety, and materials physics applications.

2. Different applications can be sensitive to different aspects of integral, differential, and/or double-differential scattering cross sections. Not all applications are integral!

3. Condensed matter physics is well understood. Thermal neutron scattering depends on electromagnetic forces within a material’s interatomic structure. For a well-characterized material, accuracy limitations in thermal neutron scattering calculations are primarily computational in nature.

4. There are many different types of material structures (e.g., crystals, amorphous solids, liquids, multi-element compounds, compositions/alloys, polymorphs, etc.). Evaluation strategies for these different cases can widely vary.

5. The role and character of experimental data in thermal neutron scattering calculations differs greatly from resonance scattering. Thermal scattering cross sections may be calculated from first-principles and fundamental scattering physics models which do not contain empirical nuclear model parameters.

Questions we should ask…

1. What are our goals in the evaluation process? To provide the best S(α,β) data possible? To allow for the best calculations of differential and integral cross sections? Should the ability to provide covariances factor into decisions about the evaluation process?

2. When is the incoherent approximation appropriate? How should experimental phonon spectra be used? When is an MD calculation or a DFT / lattice dynamics calculation appropriate?

3. What metric(s) should be used to verify calculations? What are appropriate uses for experimental double-differential, differential, integral and total scattering data? Experimental thermophysical data? In the incoherent approximation, should the phonon spectrum be adjusted/fitted to any of this data?

4. What α and β grid structure and resolution should be provided in an evaluation? How is it determined to be adequate? Should the inclusion of covariances influence these choices? Should we provide the phonon spectrum to users?

5. Will different non-standardized processes for generating covariances be acceptable to users? If the generation of covariance data reduces the quality of the evaluated data, does it have value? Should the format of thermal scattering covariance data be dictated by practical limitations in generating and storing the data, or by practical limitations in processing and utilizing the data?

$\hbar$

Thermal Scattering Theory

- **The double-differential thermal neutron scattering cross section may be written in general form as:**

$$\frac{d^2\sigma(E)}{d\Omega dE'} = \frac{1}{4\pi}\left(\frac{k'}{k}\right)[\sigma_{\mathrm{coh}}S(\boldsymbol{Q},\omega) + \sigma_{\mathrm{incoh}}S_{\mathrm{s}}(\boldsymbol{Q},\omega)] = \frac{\sigma_{\mathrm{b}}}{4\pi}\left(\frac{k'}{k}\right)F(\boldsymbol{Q},\omega)$$
$$F(\boldsymbol{Q},\omega) = S_{\mathrm{s}}(\boldsymbol{Q},\omega) + \frac{\sigma_{\mathrm{coh}}}{\sigma_{\mathrm{b}}}S_{\mathrm{d}}(\boldsymbol{Q},\omega)$$

$$S(\boldsymbol{Q},\omega) = S_s(\boldsymbol{Q},\omega) + S_d(\boldsymbol{Q},\omega)$$
$$\boldsymbol{Q} = \boldsymbol{k} - \boldsymbol{k}'$$
$$\omega = (E - E')\,/\,\hbar$$

- **Randomly oriented microstructure applies for most nuclear engineering applications:**

$$\longrightarrow$$

$$S(Q,\omega) = S_{\mathrm{s}}(Q,\omega) + S_{\mathrm{d}}(Q,\omega)$$
$$F(Q,\omega) = S_{\mathrm{s}}(Q,\omega) + \frac{\sigma_{\mathrm{coh}}}{\sigma_{\mathrm{b}}}S_{\mathrm{d}}(Q,\omega)$$

$$S(\alpha,\beta) = F(Q,\omega) \times k_{\mathrm{B}}T$$

$$\alpha = \hbar^2Q^2/2Mk_{\mathrm{B}}T$$
$$\beta = -\omega/k_{\mathrm{B}}T$$

- **The impact of coherent interference on INTEGRAL inelastic scattering cross sections is negligible for many materials and temperatures of concern, and so the incoherent approximation has been traditionally applied:**

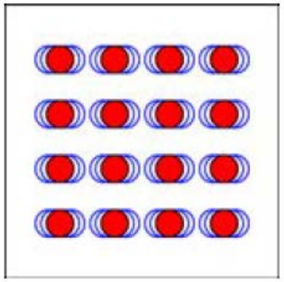
$$\boxed{S(Q,\omega) \approx S_{\mathrm{s}}(Q,\omega)} \longrightarrow \boxed{F(Q,\omega) \approx S_{\mathrm{s}}(Q,\omega)} \longrightarrow \boxed{S(\alpha,\beta) = S_{\mathrm{s}}(Q,\omega) \times k_{\mathrm{B}}T}$$

- **S_{s} is a function of only $\rho(\omega)$, or the phonon DOS, and T. In general, this is appropriate for (1) liquids, (2) amorphous solids, and (3) crystals for which the isotopically weighted average of $\sigma_{\mathrm{coh}}/\sigma_{\mathrm{b}}$ << 1. Randomly oriented microstructure does NOT eliminate inelastic coherent interference in crystals.**

- **For crystals for which the isotopically weighted average of $\sigma_{\mathrm{coh}}/\sigma_{\mathrm{b}}$ is NOT << 1, coherent interference will ALWAYS be significant in one-phonon cross sections. In this case, the phonon spectrum available for one-phonon scattering is DIFFERENT for each α (or scattering angle θ).**

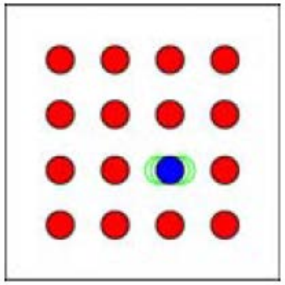
Inelastic Scattering in Crystals

Coherent Inelastic



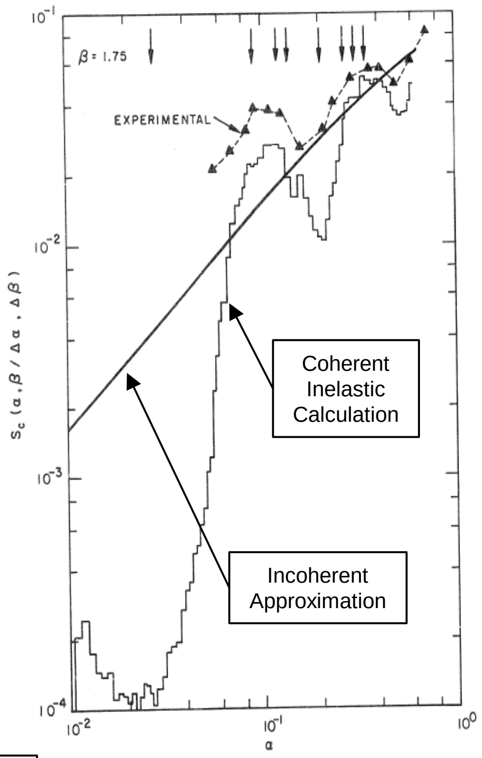
Phonon vibrations. Spatially correlated atomic motion. One-phonon interference is always significant when there is little to no spin or isotopic variation over lattice sites. This means $\sigma_{\mathbf{coh}}/\sigma_{\mathbf{b}} \approx 1$.

Incoherent Inelastic



Local vibrations or internal molecular vibrations. Also, when spin or isotopic variation over lattice sites is significant ($\sigma_{\mathbf{coh}}/\sigma_{\mathbf{b}} << 1$), spatially correlated atomic motion will not result in significant interference.

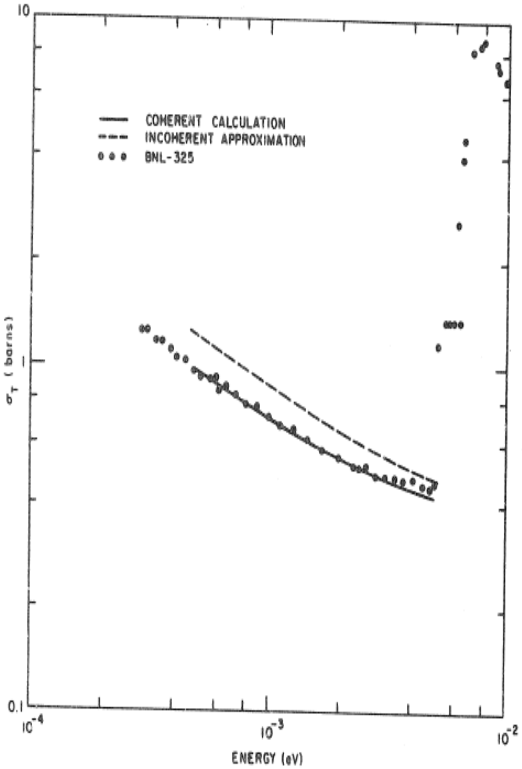
$S(\alpha,\beta)$



Coherent Inelastic Calculation

Incoherent Approximation

Total XS

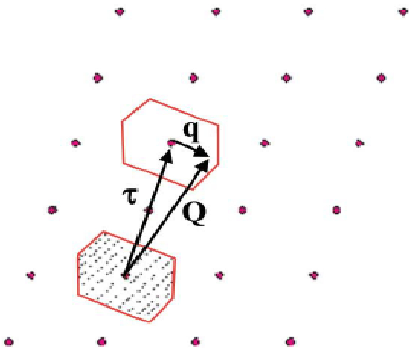


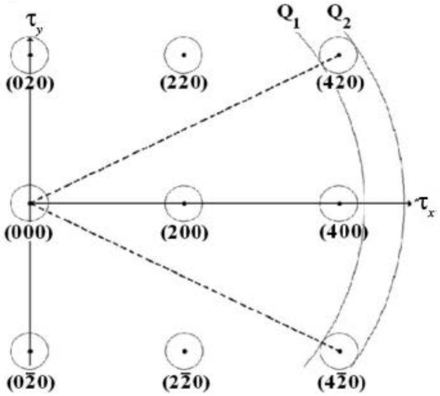
In many cases, DFT with lattice dynamics is ideal for calculating phonon spectra and dispersion relations (which can account for coherent interference) with high accuracy for well-characterized crystals.

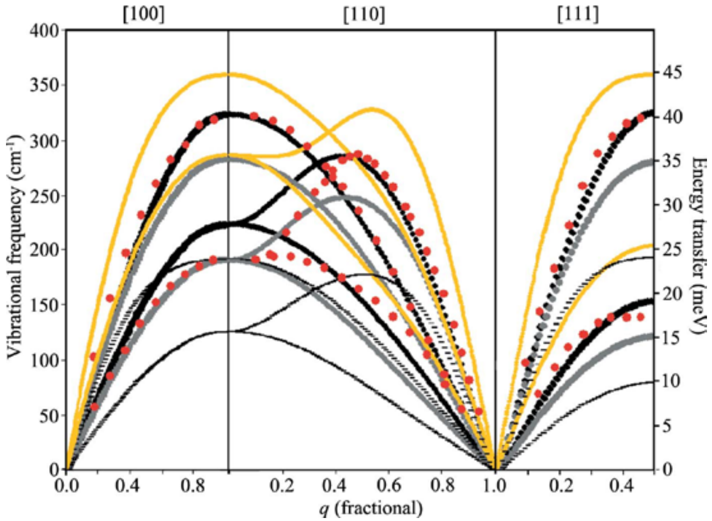
Borgonovi, G. M., “Coherent Scattering Law for Polycrystalline Beryllium,” GA-9364, Gulf General Atomic, Inc. (May 16, 1969).

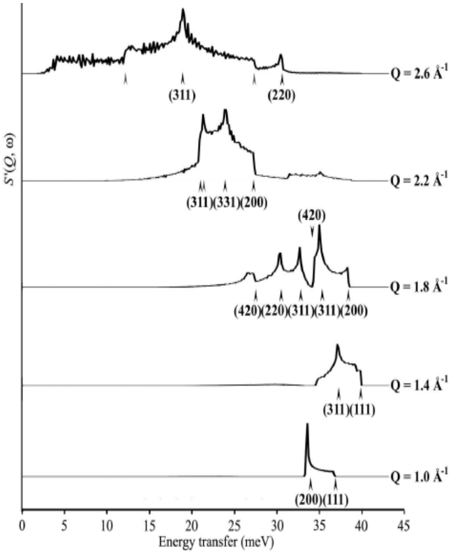
Inelastic Scattering in Crystals

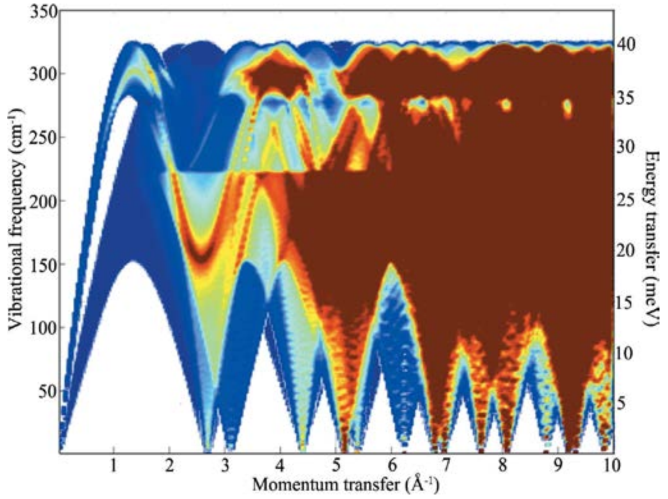
Roach, D. L. et al., “The Interpretation of Polycrystalline Coherent Inelastic Neutron Scattering from Aluminum,” *Applied Crystallography*, **46** (2013).

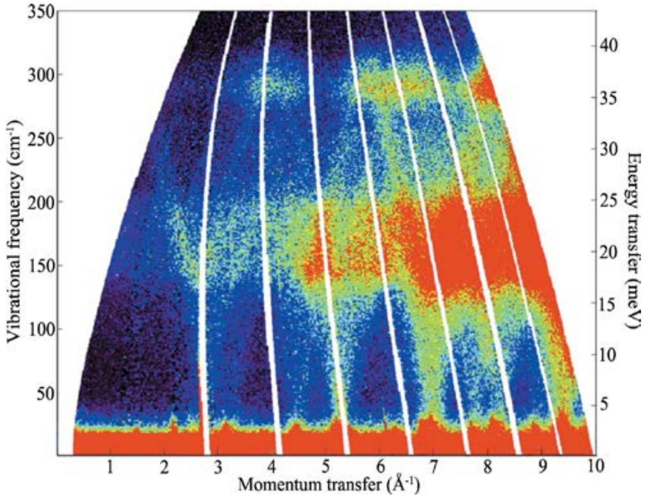




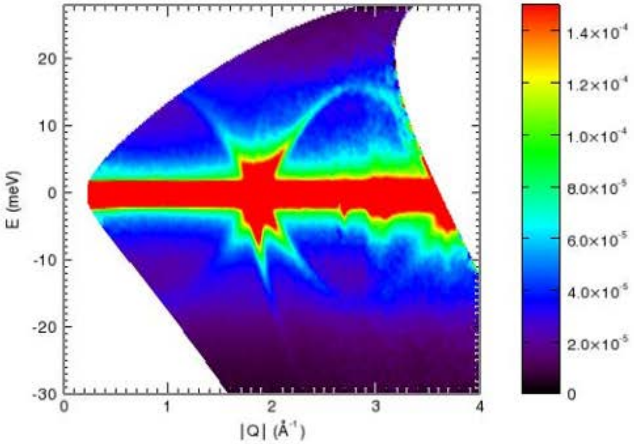


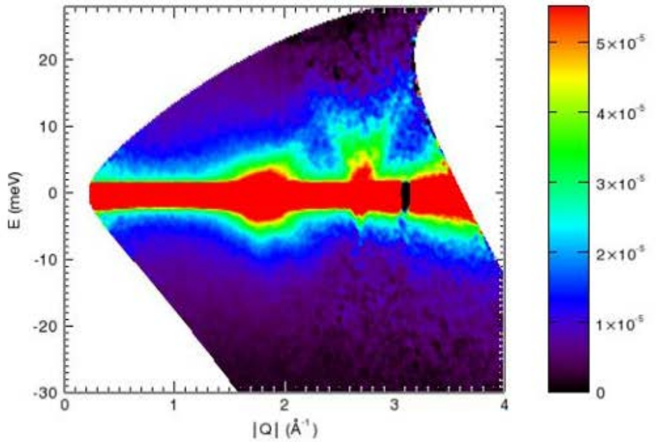






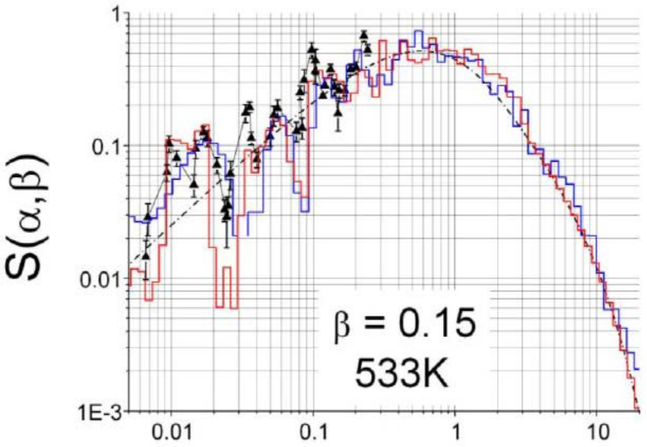
Inelastic Scattering in Crystals

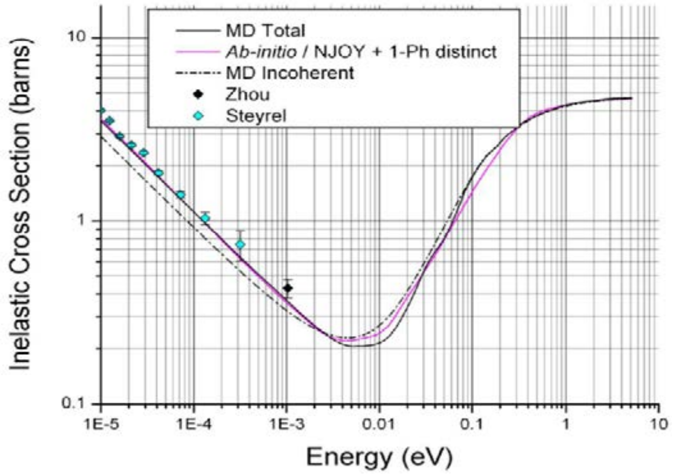




Experimental data from Sequoia, Spallation Neutron Source, Oak Ridge National Laboratory (2011).

Hawari, A. I. et al., “Inelastic Neutron Scattering Analysis of Reactor-Grade Graphite,” *Transactions of the American Nuclear Society*, **110** (2014).





Hehr, B. D., “Development of Thermal Neutron Scattering Cross Sections of Graphitic Systems using Classical Molecular Dynamics Simulations,” Ph.D. dissertation, North Carolina State University (2010).

Inelastic Scattering in Multi-Element Crystals, Amorphous Solids, and Liquids

- For a multi-element crystal, or a crystal with multiple basis positions per unit cell, the double-differential cross section is:

$$\frac{d^2\sigma(E)}{d\mu dE'} = \sum_{i=1}^{N} \frac{(\sigma_\mathrm{b})_i}{2}\sqrt{\frac{E'}{E}}\,F_i(Q_i,\omega), \qquad \text{where} \qquad F_i(Q_i,\omega) = S_{\mathrm{s},i}(Q_i,\omega) + \frac{(\sigma_\mathrm{coh})_i}{(\sigma_\mathrm{b})_i} S_{\mathrm{d},i}(Q_i,\omega)$$

- If the incoherent approximation applies, then:

$$\frac{d^2\sigma(E)}{d\mu dE'} = \sum_{i=1}^{N} \frac{(\sigma_\mathrm{b})_i}{2k_\mathrm{B}T}\sqrt{\frac{E'}{E}}\,S_i(\alpha_i,\beta)$$

- In either case, each basis position has its own vibrational spectrum. Even if the incoherent approximation applies, no single phonon spectrum can be used in a theoretical calculation to reproduce the experimental cross sections (although in certain cases the result may be close).

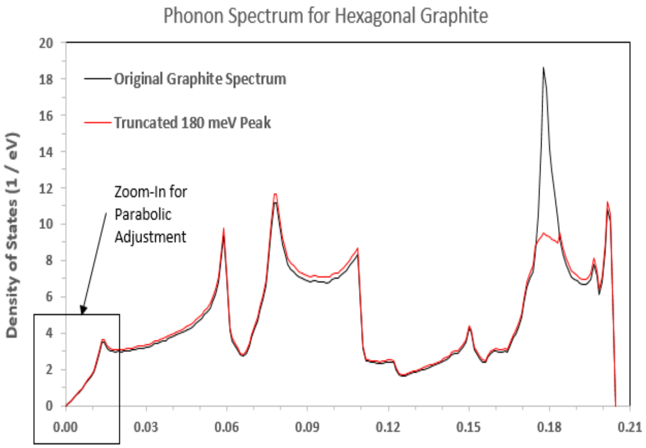
- An experimentally determined phonon spectrum will be an “effective” spectrum averaged over all basis positions. (Neutrons cannot be told to scatter with only one element or basis position.)

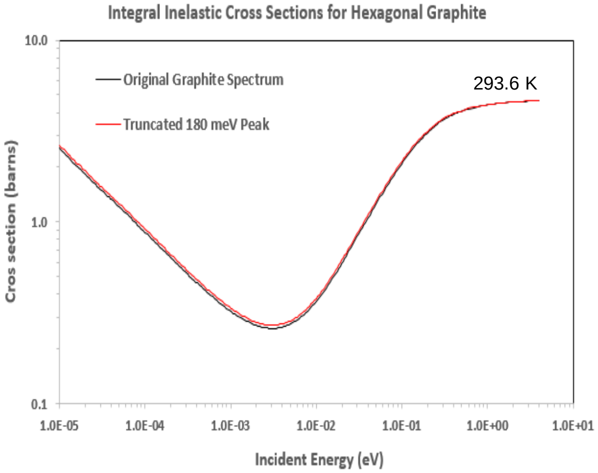
- Similarly, in the case of amorphous solids, where the incoherent approximation is typically adequate, an “effective” phonon spectrum (whether determined experimentally OR theoretically) will not reproduce experimental cross sections. HOWEVER, depending on the particular material, a very good approximation can sometimes be made.

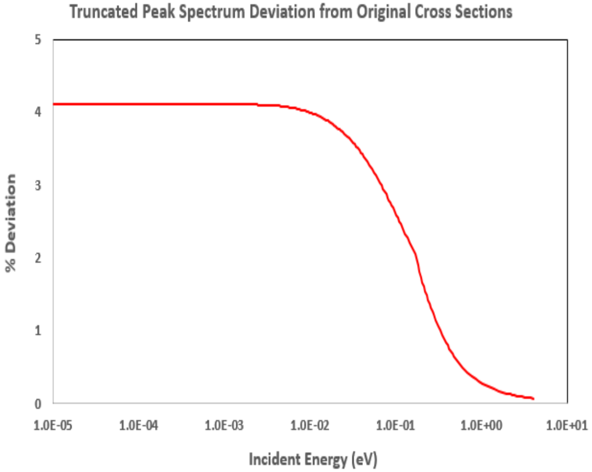
- In principle, a molecular dynamics simulation of an amorphous solid can bypass the problem of variable vibrational spectra and output the thermal scattering law for the “effective” material structure directly.

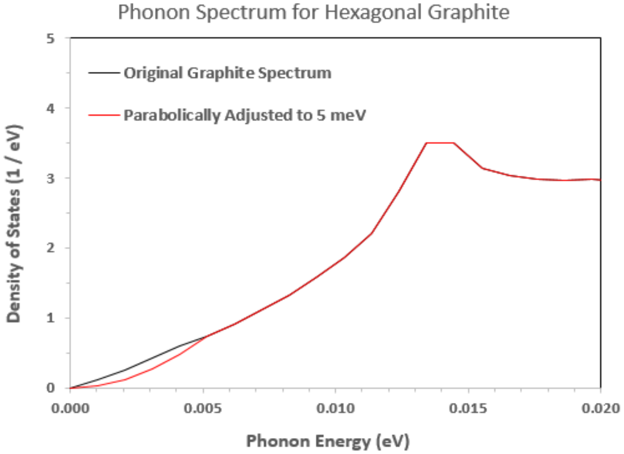
- For liquids, the phonon spectrum for each basis position in a molecule applies, and molecular diffusion and rotations are possible. In this case, a molecular dynamics simulation is likely ideal for calculations of the thermal scattering law.

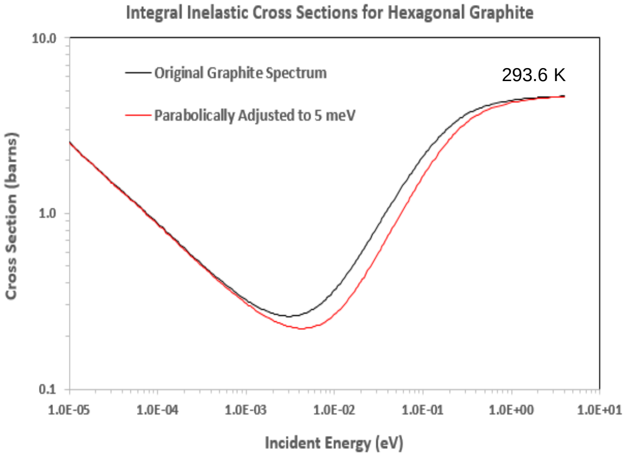
Cross Section Sensitivity to the Phonon Spectrum

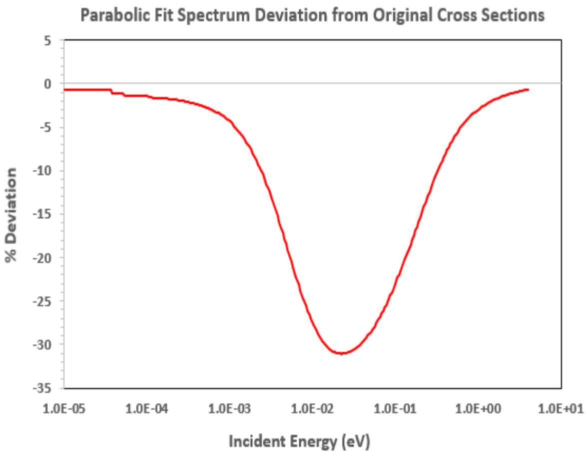








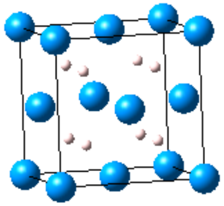


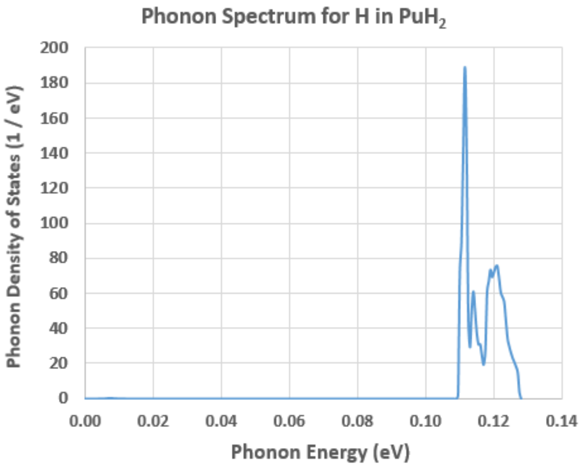


Holmes, J. C., “Monte Carlo Calculation of Thermal Neutron Inelastic Scattering Cross Section Uncertainties by Sampling Perturbed Phonon Spectra”, Ph.D. dissertation, North Carolina State University (2014).

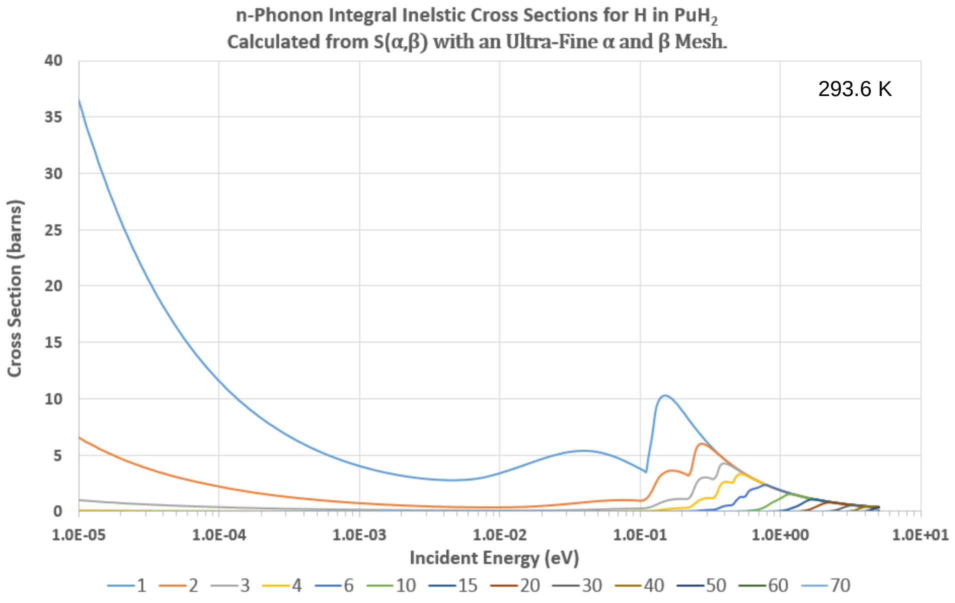
Unique Properties of Metal Hydrides

PuH_2

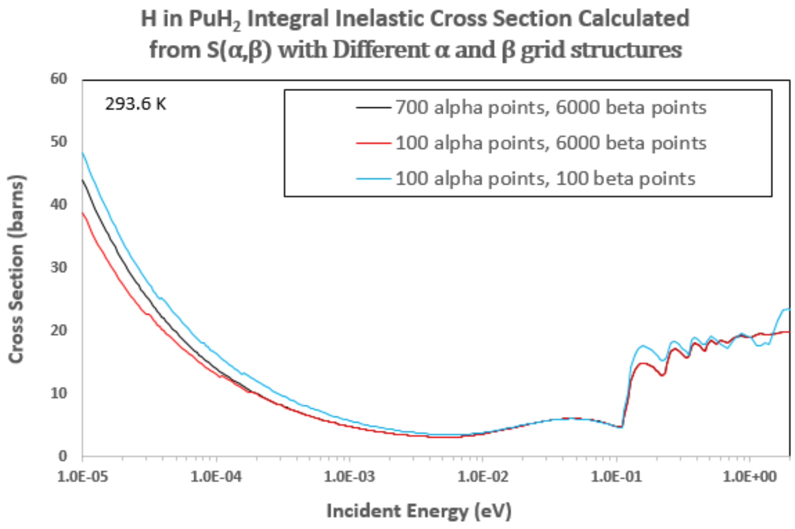




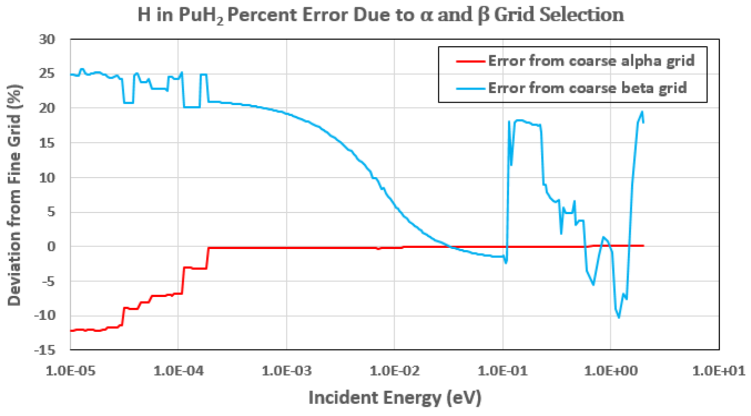
Phonon Spectrum for H in PuH_2



n-Phonon Integral Inelstic Cross Sections for H in PuH_2 Calculated from $S(\alpha,\beta)$ with an Ultra-Fine α and β Mesh.



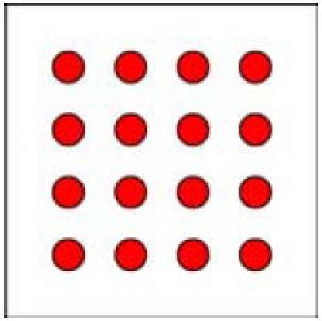
H in PuH_2 Integral Inelastic Cross Section Calculated from $S(\alpha,\beta)$ with Different α and β grid structures



H in PuH_2 Percent Error Due to α and β Grid Selection

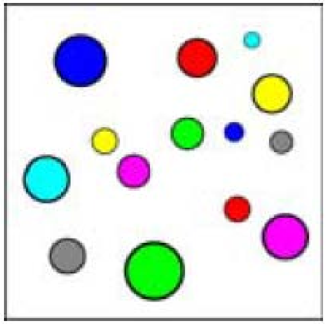
Elastic Scattering in Solids

Coherent Elastic

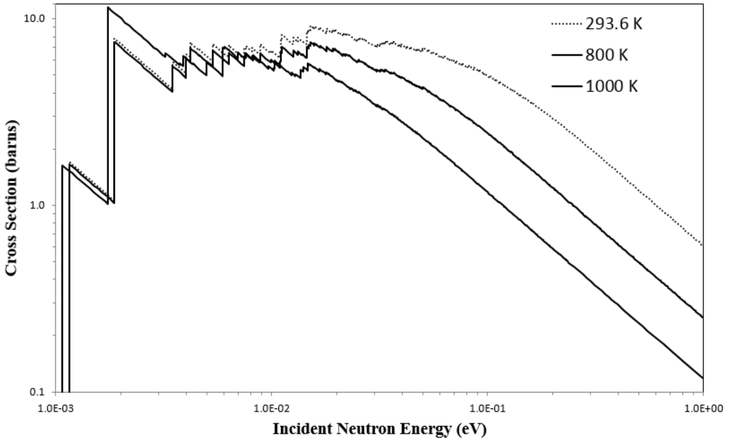


Spatially correlated structure. Significant coherent interference occurs when there is little to no spin or isotopic variation over lattice sites, or when $\sigma_{\mathrm{coh}}/\sigma_{\mathrm{b}} \approx 1$. Prohibited below Bragg cutoff energy.

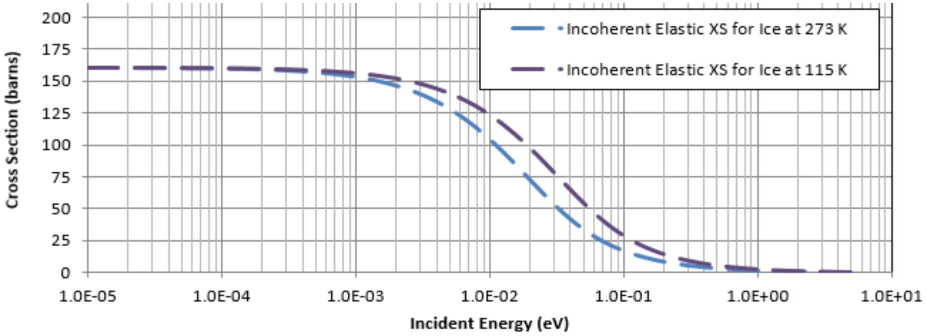
Coherent Inelastic



Spatially uncorrelated structure largely cancels out interference effects. (Also, for spatially correlated structures, significant spin or isotopic variation over lattice sites has the same effect.)



Holmes, J. C., “Development of ENDF Thermal Neutron Scattering Libraries for Silicon Dioxide and MCNP Criticality Testing with an ICSBEP Benchmark,” Masters project, North Carolina State University (2011).



Holmes, J. C. et al., “Production of the Thermal Scattering Law and Neutron Scattering Cross Sections for Ice from First Principles,” *Proceedings of the 10th International Conference on Nuclear Criticality Safety (ICNC 2015)*, September 2015.

Elastic Scattering in Solids

Coherent

$$\frac{d\sigma_{\mathrm{el}}(E)}{d\mu} = \frac{\sigma_{\mathrm{coh}}}{E}\sum_{i,\,(E_i<E)} \frac{f_i}{\tau_i} e^{-4WE_i}\delta\left[\mu - \left(1-\frac{E_i}{E}\right)\right]$$

Incoherent

$$\sigma(E) = \frac{\sigma_{\mathrm{incoh}}}{2}\left(\frac{1-e^{-4WE}}{2WE}\right)$$

- The given forms for coherent and incoherent elastic scattering assume that the bound nuclear scattering cross section is either entirely coherent or entirely incoherent. While often this is nearly true, there are exceptions.

- Some elements have significant coherent and incoherent bound nuclear scattering cross sections. Additionally, for multi-element crystals (such as metal hydrides), an accurate treatment of elastic scattering may require separate calculation of coherent elastic and incoherent elastic cross sections for the different element types to be consistent with total measured cross sections.

- Currently, the NJOY code does not allow for separate treatment of coherent and incoherent elastic scattering in a single evaluation.

- Elements which have significant coherent AND incoherent bound nuclear scattering cross sections are: Li, B, Na, Cl, Ar, Sc, Ti, Cr, Mn, Co, Ni, Cd, In, La, Nd, Eu, Gd, Dy, Yb, Hf, W, and Hg.

- For amorphous solids, the incoherent form for elastic scattering applies regardless of the actual coherent or incoherent fractions of the total bound nuclear scattering cross sections.

Some Thoughts About Thermal Scattering Covariance Data

- For resonance scattering data, the evaluation process does not **fundamentally** change across nuclides. **Generally speaking**, the experimental process is the same, the empirical model fitting process is the same, the generation of covariance data follows from this fitting process, and data testing and verification is the same from nuclide to nuclide.

- For thermal neutron scattering, different material structures can require different models, different experimental and calculation methods, different data testing and verification methods, and different evaluation strategies (and philosophies). Consequently, for thermal scattering data, there may never be a single standardized method for generating covariances.

- Getting to a standardized covariance data format is a practical goal. Format decisions should be informed by:

  (1) practical compatibility of the format with the range of calculation and evaluation methods that exist for different materials,
  (2) practical compatibility with current and future formats for storing thermal scattering data, and
  (3) practical compatibility with processing codes and uncertainty / sensitivity analysis codes which will read and use the data.

- For resonance scattering, covariances inherently provide information about the relationship between theoretical and experimental scattering data. **This is often not true for thermal scattering.**

  Reasons are:
  (1) Thermal scattering data can be calculated by various first-principles methods which do not contain empirical parameters. In many cases, uncertainties may not be able to be clearly parameterized but may be due to approximations or limitations, whether computational or physical in nature, in a complex simulation process.
  (2) Experimental scattering data, such as S(α,β), may be limited or have higher uncertainty than other measured thermophysical material properties which directly informed the calculation process.

- **Most importantly, should the thermal scattering evaluation process be influenced by the desire to provide covariances?**

Summary / Conclusions

- Thermal neutron scattering is the process of neutrons interacting with the interatomic structure and dynamics of a material. Specifically, correlations in the positions of atoms in space and time within a material structure influence neutron scattering probability and scattered neutron wave spectra.

- The interatomic structure and dynamics of a material is controlled by the electromagnetic force. The electromagnetic force is well understood and, consequently, information about the interatomic structure and dynamics of a material may be accessed through first-principles calculations.

- Accessing information about nuclear resonances **requires** experimentally interacting with the resonances.

  For thermal neutron scattering, the measurement of thermophysical material properties can inform first-principles calculations without the need to experimentally probe the interatomic structure and excitation modes (the material analog of resonances).

  Neutron scattering experiments do access information about material structure and excitation modes and can **further** inform first-principles calculations. However (unlike single-nuclide resonance scattering experiments), for materials with multiple basis positions, multiple elements, and/or multiple isotopes, scattering experiments only provide **material-averaged** information.

- Consequently, the evaluation process for thermal neutron scattering data can involve the interpretation of many different sources and types of information, experimental and theoretical, each having distinct uncertainties and/or limitations. Any material evaluation must duly consider and weigh the merits of all available information.