Production of an S(α,β) Covariance Matrix with a Monte Carlo-Generated Set of Perturbed Phonon Frequency Spectra

Jesse C. Holmes
Ayman. I. Hawari

Department of Nuclear Engineering,
North Carolina State University,
Raleigh, NC

and

Luiz Leal

Oak Ridge National Laboratory,
Oak Ridge, TN

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Motivation

- Uncertainties in the outputs of modeling and simulation codes such as MCNP and SCALE inherently depend on nuclear data uncertainties as described by the covariance matrix of a given data library.
- No procedure exists for producing or representing covariance matrices for thermal neutron scattering data in libraries (such as ENDF format).
- Thermal neutron scattering libraries are typically produced using physics models that incorporate structural information of a given material. For inelastic scattering, the dynamic structure factor $S(\alpha, \beta)$ is the fundamental output.
- In crystalline materials, $S(\alpha, \beta)$ is a function of the phonon frequency spectrum, or vibrational density of states (DOS).
- By capturing the uncertainties of the phonon spectrum in the $S(\alpha, \beta)$ covariance matrix, covariance matrices may be produced for the secondary neutron distributions in energy and angle, and for the integrated inelastic scattering cross sections.

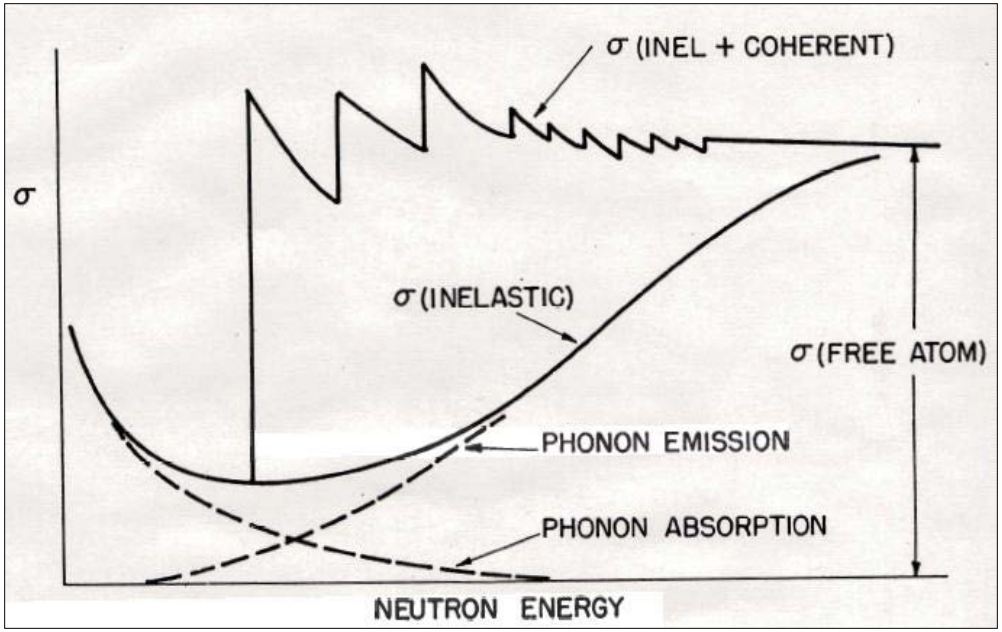
Objectives

- Investigate the uncertainty contribution of the physics model, its parameters and the methodology implemented to generate the DOS. Silicon dioxide (α-quartz) will be used as the example material in this work due to the particular features of its DOS.
- Establish probability distribution functions (PDFs) for the various energy regions of the DOS spectrum that reflect the physical and statistical attributes of the DOS.
- Explore the utilization of Monte Carlo sampling of these PDFs to generate a set of perturbed phonon spectra for production of an $S(\alpha, \beta)$ covariance matrix.
- Produce an $S(\alpha, \beta)$ covariance matrix and examine the impact of its propagation.

Thermal Neutron Scattering and Energy Transfer

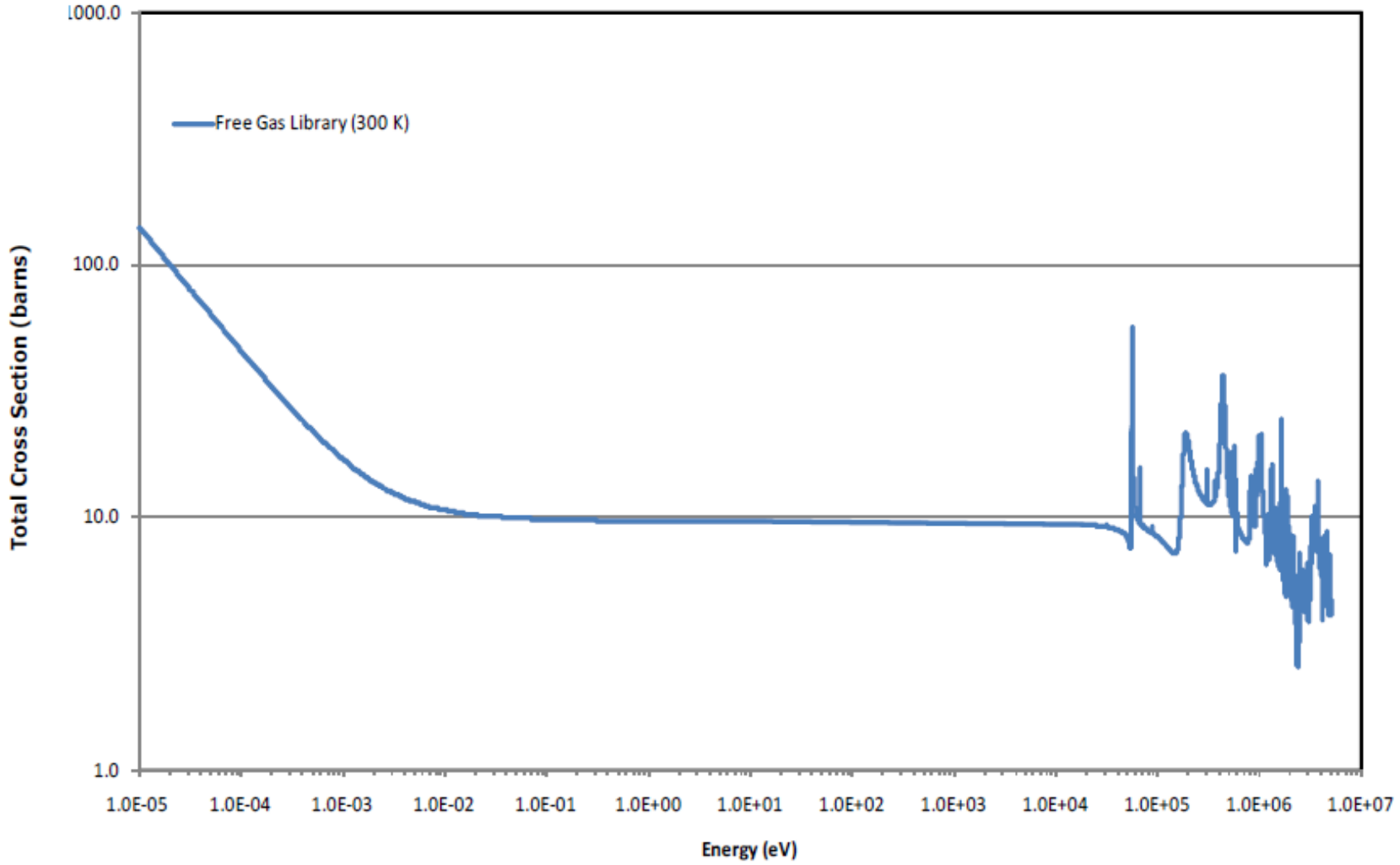
- The de Broglie wavelength ($\lambda = h / p$) of thermal neutrons (< ~ 1 eV) is on the order of the interatomic distances in crystalline solids.

- The energy of thermal neutrons is on the same order as the vibrational excitation modes (e.g., phonons) available in condensed matter.



Thermal Neutron Scattering Cross Sections

Modifying the Free-Atom Cross Section for α-quartz SiO_2



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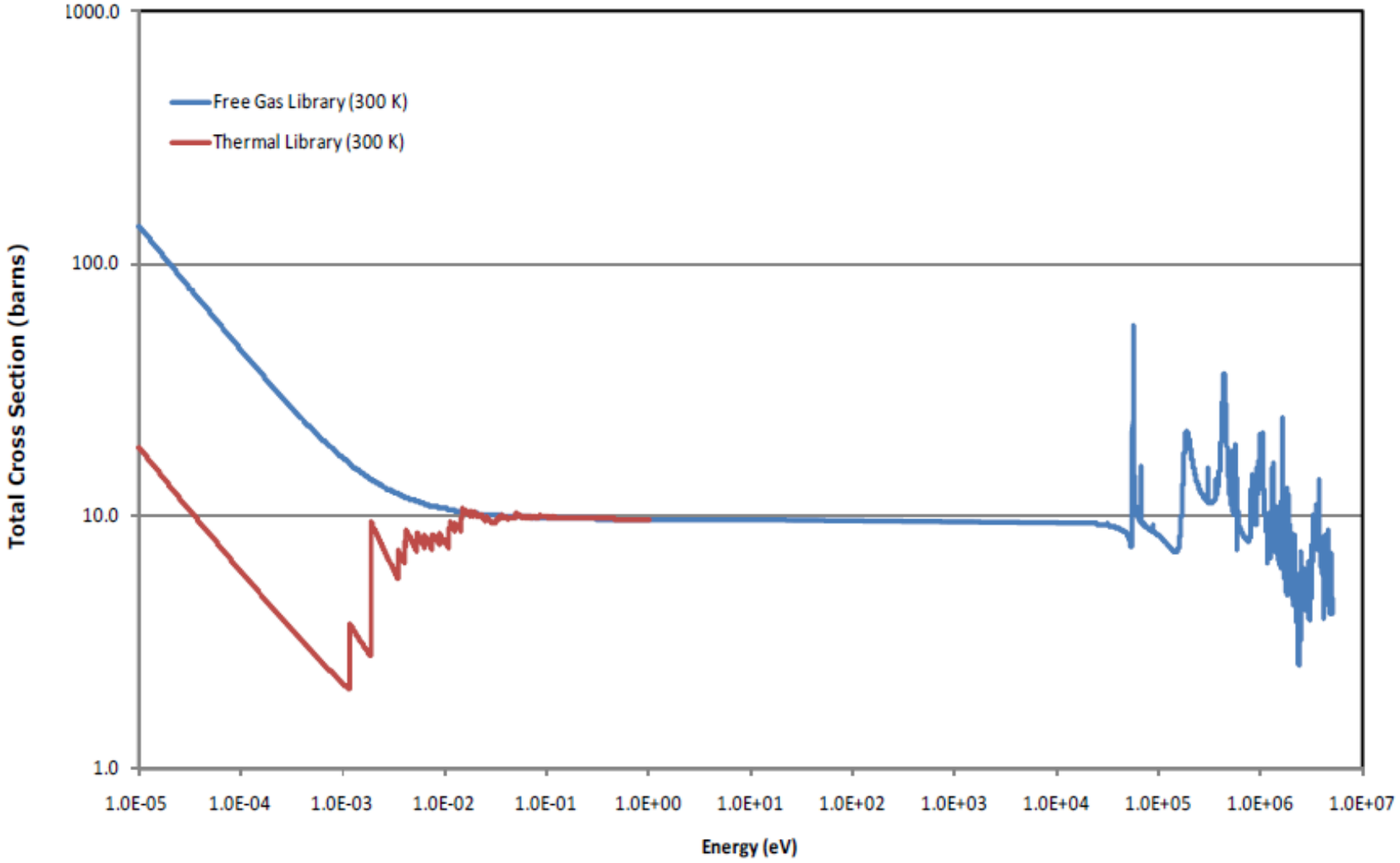


Thermal Scattering Theory

Using the first-order Born approximation and Fermi pseudopotential, the double-differential thermal neutron scattering cross section may be written as

$$\frac{d^2\sigma(E)}{d\Omega dE'} = \frac{1}{4\pi}(k'/k)(\sigma_{coh}S(\boldsymbol{Q},\omega) + \sigma_{incoh}S_s(Q,\omega)) \quad \text{with}$$

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

Applying the incoherent approximation for inelastic scattering,

$$S(\boldsymbol{Q},\omega) = S_s(Q,\omega),$$

yields

$$\frac{d^2\sigma_{inel}(E)}{d\Omega dE'} = \frac{\sigma_b}{4\pi}(k'/k)S_s(Q,\omega), \quad \text{or}$$

$$\frac{d^2\sigma_{inel}(E)}{d\Omega dE'} = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E'}{E}}S(\alpha,\beta).$$

Thermal Scattering Theory

$$\frac{d^2\sigma_{inel}(E)}{d\alpha\, d\beta} = \frac{Ak_BT\sigma_b}{4E} S(\alpha,\beta).$$

Momentum transfer factor: $\alpha = \dfrac{E' + E - 2\sqrt{EE'}\mu}{Ak_BT}$

Energy transfer factor: $\beta = \dfrac{E' - E}{k_BT}$

$$S(\alpha,\beta) = \sum_{n=1}^{\infty} S_n(\alpha,\beta)$$

For n = 1: $S_1(\alpha,\beta) = \dfrac{\alpha\, e^{-\alpha\lambda} e^{-\beta/2}\rho|\beta|}{2\beta\, sinh(\beta/2)}$ with $\lambda = \displaystyle\int_{-\infty}^{\infty} \frac{e^{-\beta/2}\rho|\beta|}{2\beta\, sinh(\beta/2)} d\beta$

(the term $e^{-\alpha\lambda}$ is the Debye-Waller factor)

For n > 1: $S_n(\alpha,\beta) = (n!)^{-1}\alpha^n e^{-\alpha\lambda} T_n(\beta)$

$$T_n(\beta) = \int_{-\infty}^{\infty} T_1(\beta') T_{n-1}(\beta - \beta')\, d\beta'$$

$$T_1(\beta) = \frac{e^{-\beta/2}\rho|\beta|}{2\beta\, sinh(\beta/2)}$$

The DOS, $\rho|\beta|$, is the primary unknown parameter

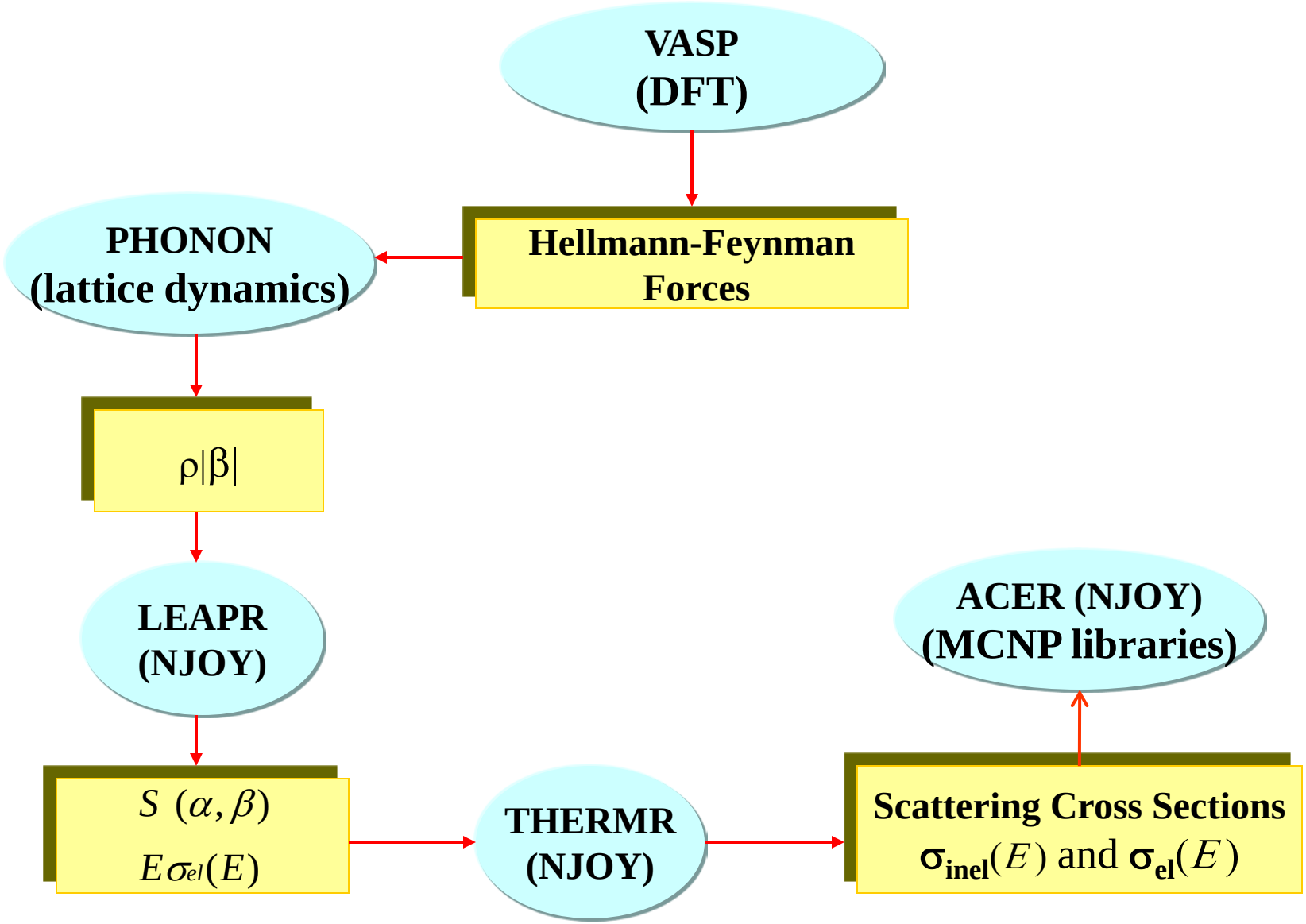
Generating the Phonon Density of States

The phonon DOS can be generated in several different ways:

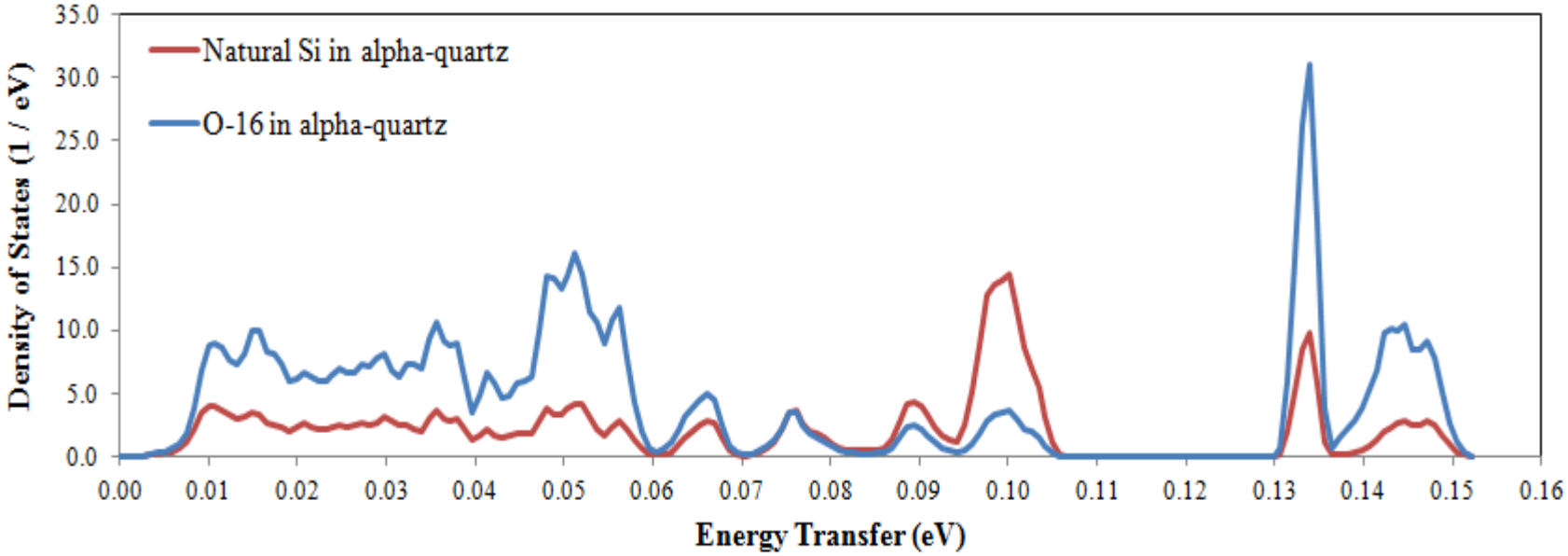
1. A double-differential scattering experiment can be performed measuring time-of-flight and scattering angle.

2. Lattice dynamics can be applied using force constant models.

   a. Force constants obtained by fitting to thermodynamic experimental data.
   b. Force constants obtained by fitting to experimental dispersion curves along symmetry directions in the first Brillouin zone.
   c. Force constants obtained theoretically from first principles using density functional theory (DFT). This will be the approach utilized in this work.

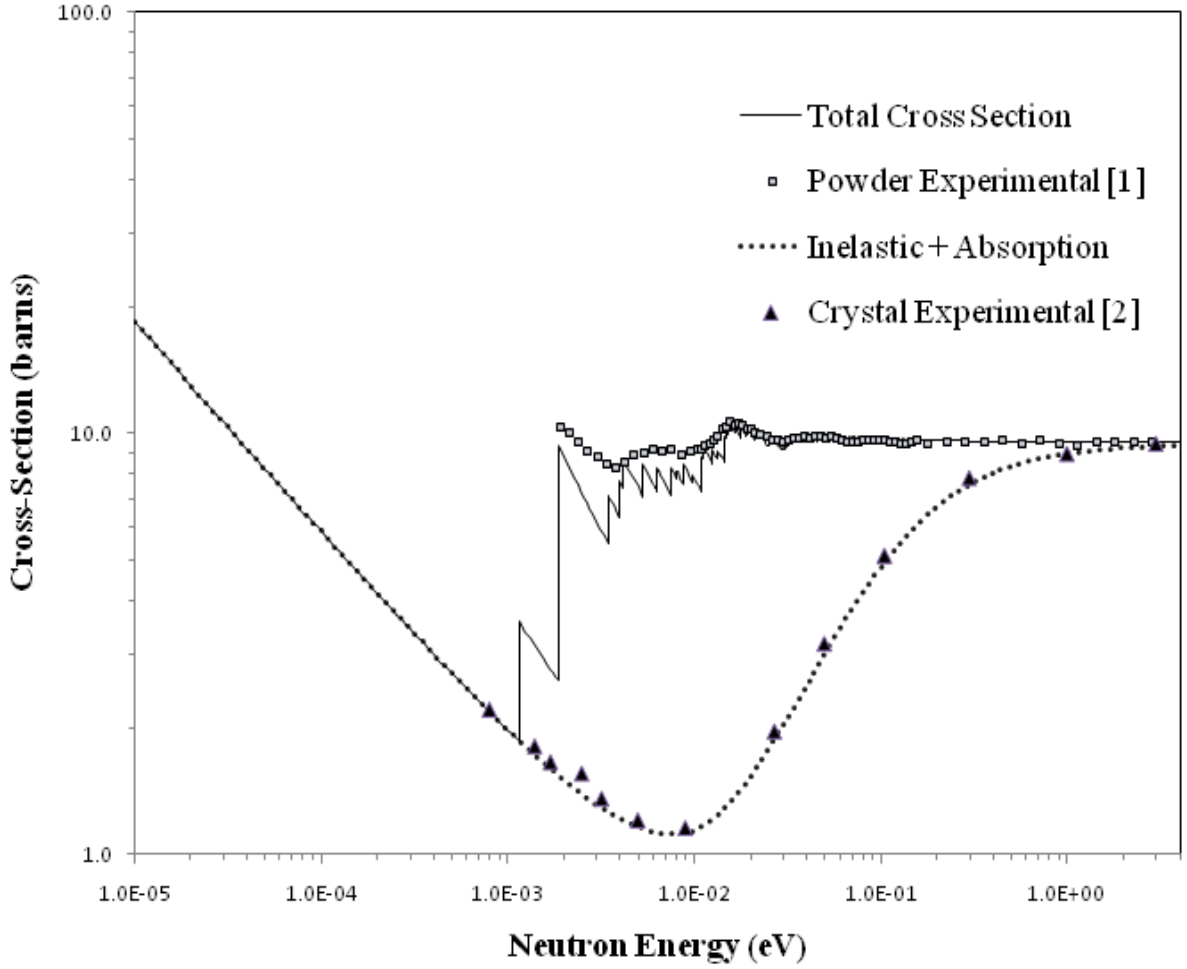

VASP
(DFT)
Hellmann-Feynman
Forces
PHONON
(lattice dynamics)
ρ|β|
LEAPR
(NJOY)
$S(\alpha,\beta)$
$E\sigma_{el}(E)$
THERMR
(NJOY)
ACER (NJOY)
(MCNP libraries)
Scattering Cross Sections
$\sigma_{inel}(E)$ and $\sigma_{el}(E)$

Reference α-Quartz DOS



Silicon is normalized to 1/3. Oxygen is normalized to 2/3.

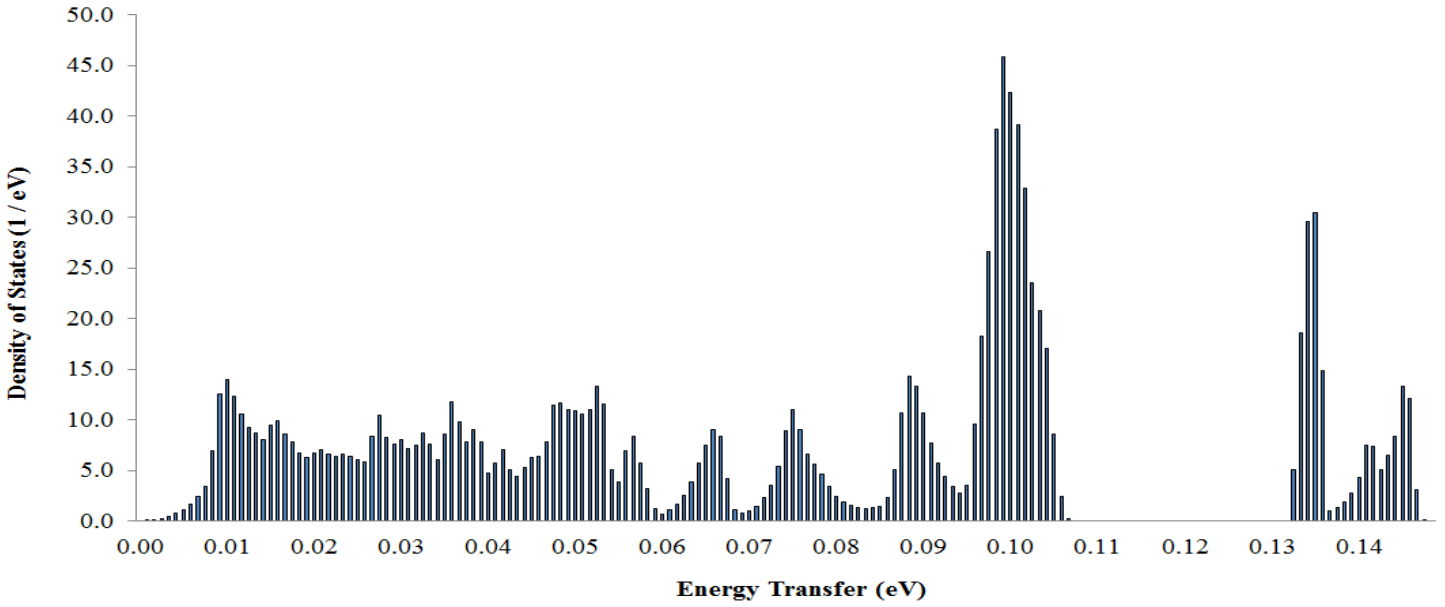
Comparison of Experimental and Calculated Cross Sections for α-Quartz



[1] J. J. BLOSTEIN and J. DAWIDOWSKI, "Experimental Neutron Data: Sigma(E) of Vitreous Silica (SiO2) Over the Thermal Energy Range," Private Communication, Inst. Balseiro y Centro Atomico Bariloche, Argentina (1999).

[2] B. M. RUSTAD, et al., "Neutron Crystal Spectrometer with Range Extended to Subthermal Energies," *Review of Scientific Instruments*, **36**, 887 (1965).

Discrete DOS for Natural Si in α-Quartz



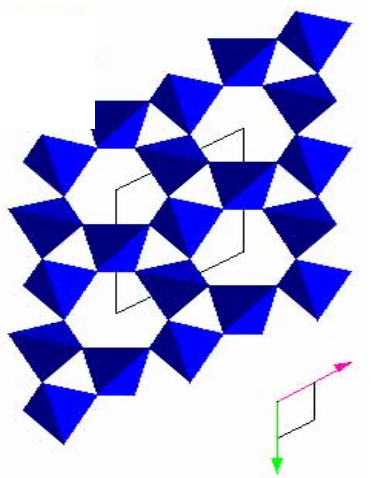
The $\rho|\beta|$ DOS is itself a PDF of available energy transfer quanta.

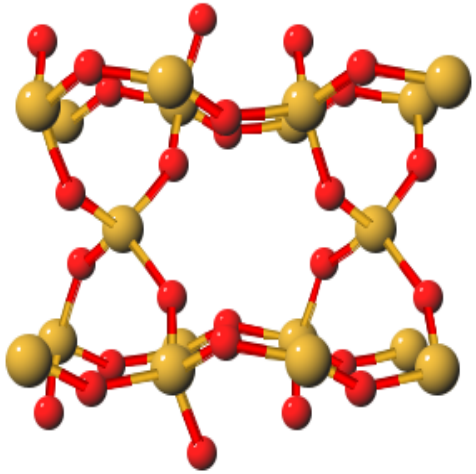
Consider the normalized DOS to consist of many random variables with possible valuations described by PDFs.

Ideally, these PDFs should represent statistical uncertainties in the phonon counting process and feature uncertainties due to uncertainties in the physics model parameters.

Hexagonal Structure of α-quartz

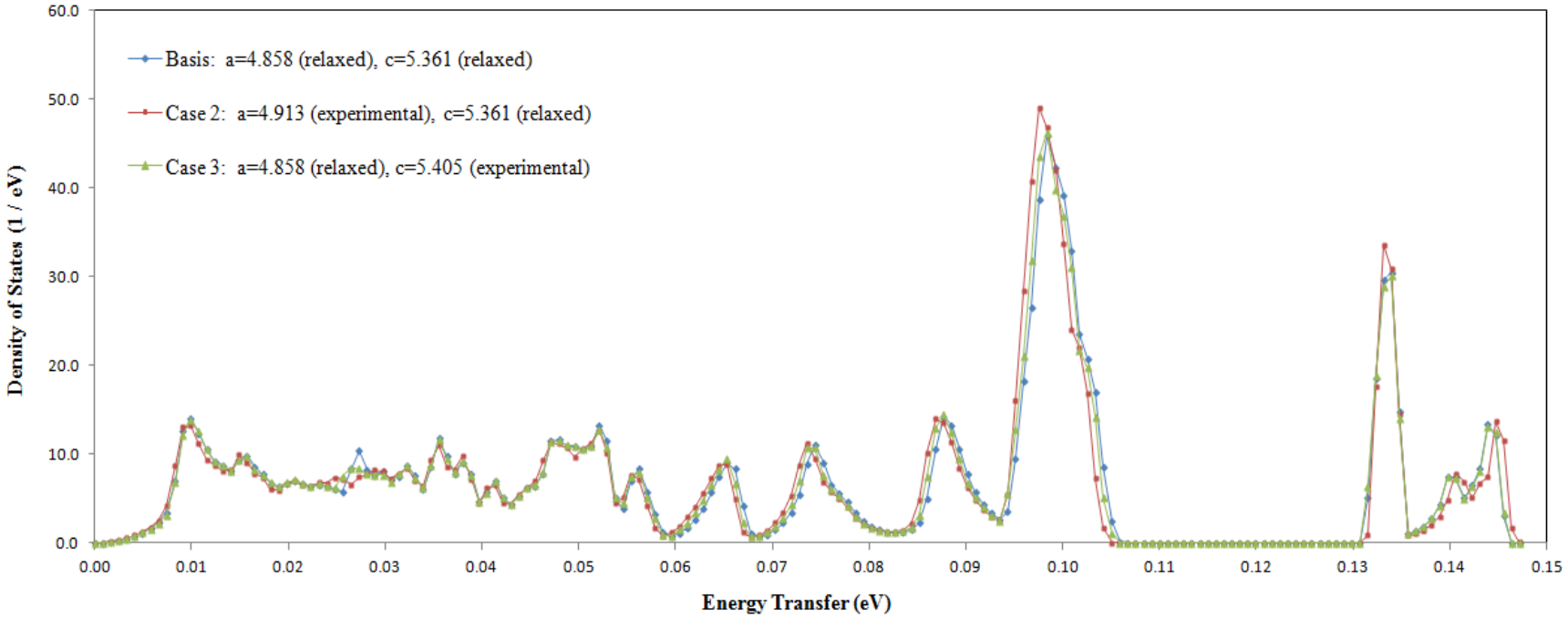
Silicons (yellow), Oxygens (red)





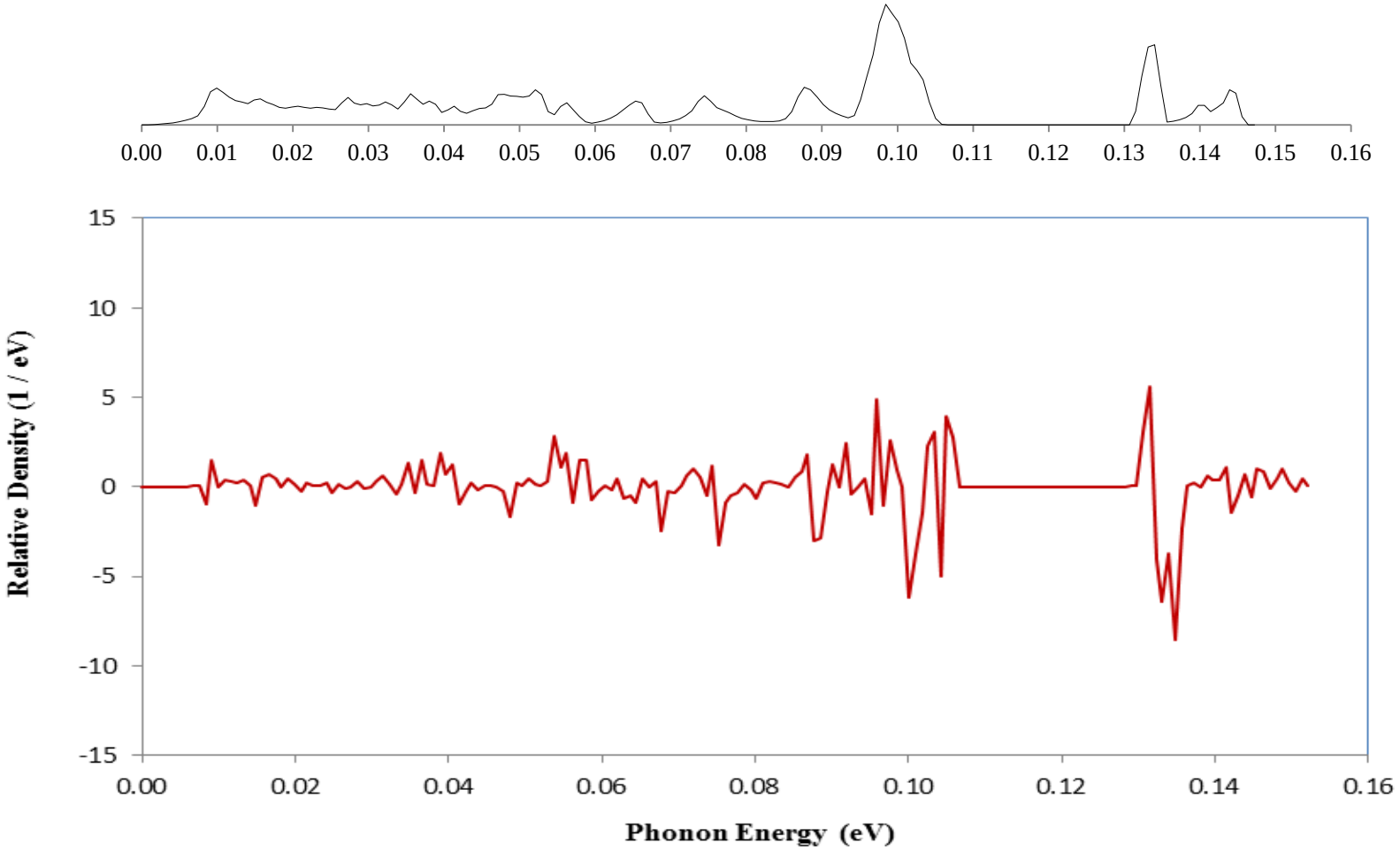
Lattice constant a: 4.913 Å
Lattice constant b: 4.913 Å
Lattice constant c: 5.405 Å

Three Examples of Parameter-Modified DOS Spectra



Maximum shift in features observed is < ~ 0.001 eV (about one bin-width). Other parameter modifications (not plotted here) yield similar results. In this work, a PDF is assumed which randomly shifts each "bin feature" up or down in energy within +/- one bin width. Renormalization follows.

Example of Perturbed DOS Difference from Reference DOS



Calculating the $S_{ENDF}(\alpha,\beta)$ Covariance Matrix

The symmetric $S_{ENDF}(\alpha,\beta)$ covariance matrix will be calculated, where $S_{ENDF}(\alpha,\beta) = e^{\beta/2} S(\alpha,\beta)$.

The terms y_i and y_j represent $S_{ENDF}(\alpha,\beta)$ terms for different α and β combinations.

Each $S_{ENDF}(\alpha,\beta)$ term is calculated for a Monte Carlo trial k with a particular perturbed DOS k for k = 1 to N with N = 330.

$$\bar{y_i} = \frac{1}{N}\sum_{k=1}^{N} y_{i,k}$$

$$\sigma^2(y_i) = \frac{1}{N}\sum_{k=1}^{N}\left(y_{i,k} - \bar{y_i}\right)^2$$

$$\mathrm{COV}(y_i, y_j) = \frac{1}{N}\sum_{k=1}^{N}\left(y_{i,k} - \bar{y_i}\right)\left(y_{j,k} - \bar{y_j}\right)$$

$$S_{ENDF}(\alpha,\beta) = \sum_{n=1}^{\infty} S_{ENDF,n}(\alpha,\beta)$$

$$S_{ENDF,n=1}(\alpha,\beta) = \alpha e^{-\alpha\lambda}\frac{\rho|\beta|}{2\beta\ sinh(\beta/2)}$$

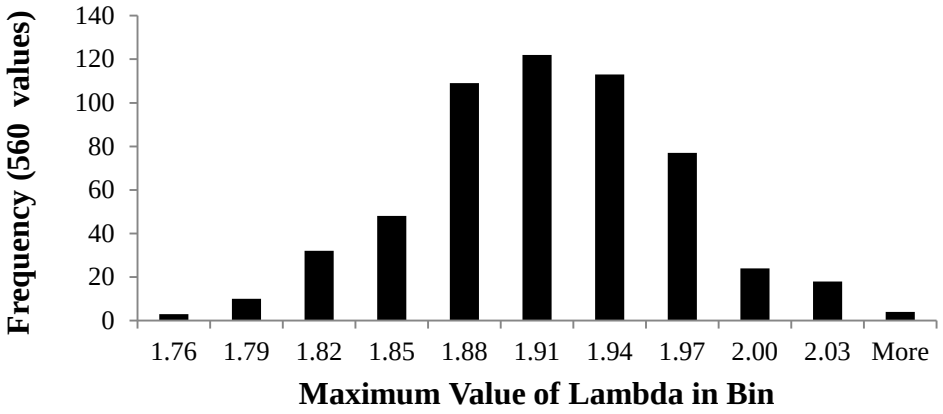
$$S_{ENDF,n>1}(\alpha,\beta) = (n!)^{-1}\alpha^n e^{\beta/2} e^{-\alpha\lambda} T_n(\beta)$$

$$\lambda = \int_{-\infty}^{\infty} \frac{e^{-\beta/2}\rho|\beta|}{2\beta\ sinh(\beta/2)}\, d\beta$$

Distribution of λ Values

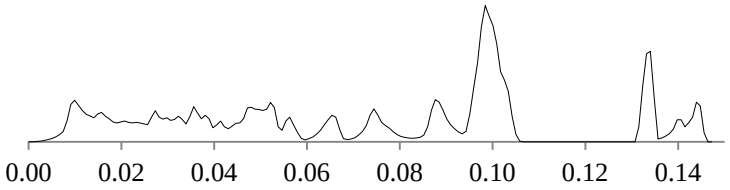
$$\lambda = \int_{-\infty}^{\infty} \frac{e^{-\beta/2}\rho|\beta|}{2\beta\ sinh(\beta/2)}\, d\beta$$

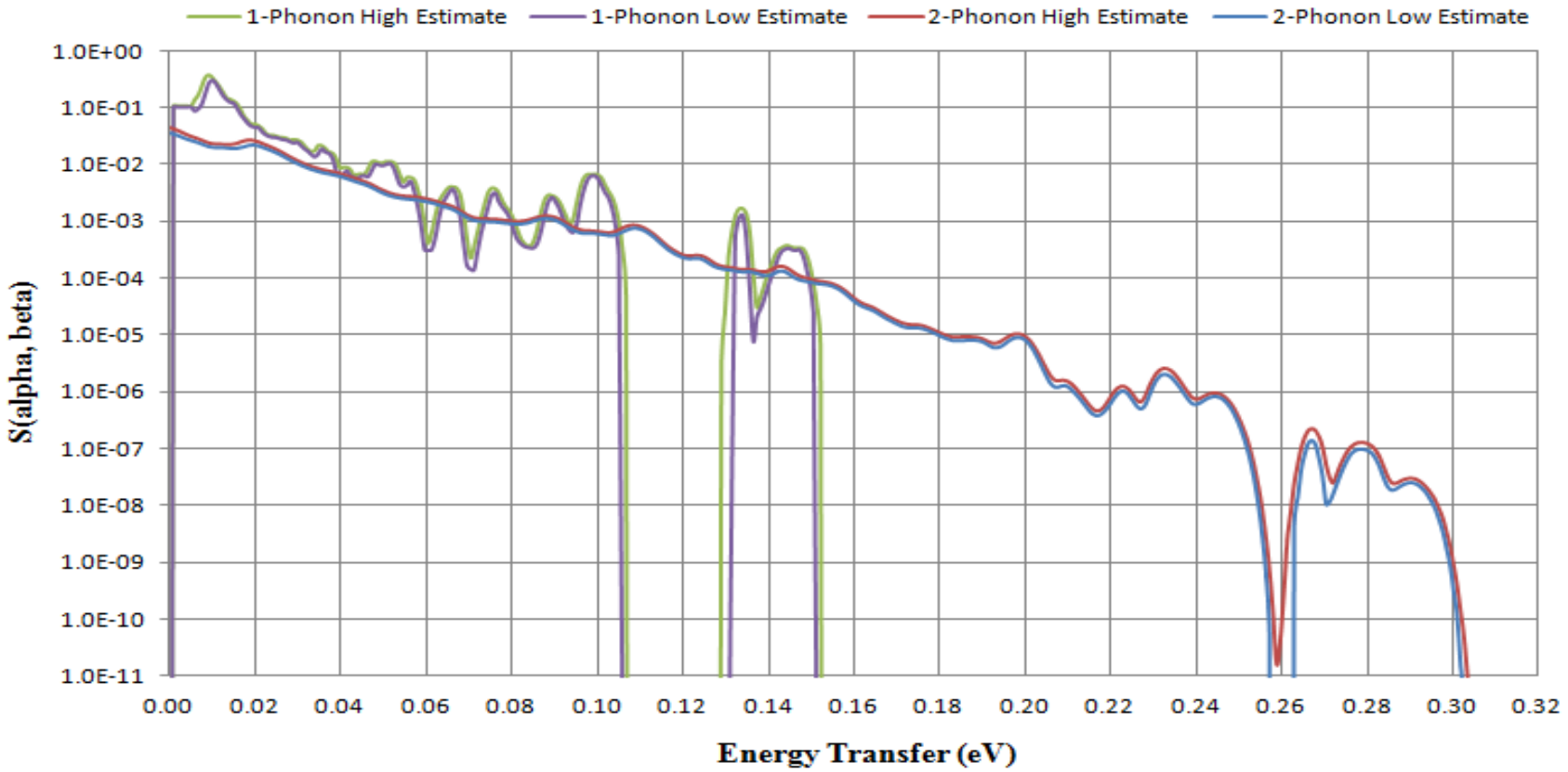
alpha values	low PERCENT change	high PERCENT change
0.010	-0.053723	0.053752
0.015	-0.080574	0.080639
0.023	-0.120837	0.120983
0.034	-0.181201	0.181530
0.051	-0.271678	0.272418
0.076	-0.407240	0.408905
0.114	-0.610237	0.613984
0.171	-0.913958	0.922388
0.256	-1.367800	1.386768
0.384	-2.044668	2.087347
0.577	-3.051271	3.147303
0.865	-4.541813	4.757908
1.297	-6.734768	7.221092
1.946	-9.930104	11.024887
2.919	-14.519021	16.985089
4.379	-20.967771	26.530658
6.568	-29.740291	42.329083
9.853	-41.107563	69.801091
14.779	-54.805115	121.263977



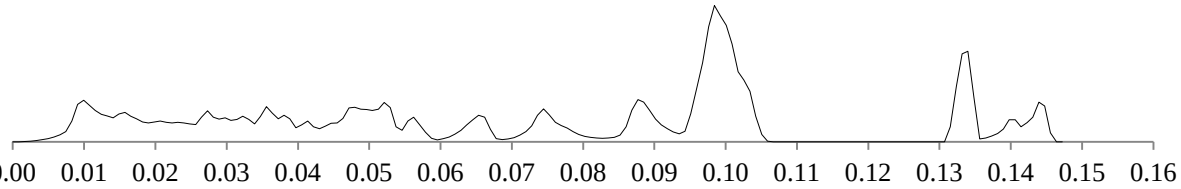
Standard Deviation	0.053738
Monte Carlo Mean	1.899448
Basis Mean	1.893759

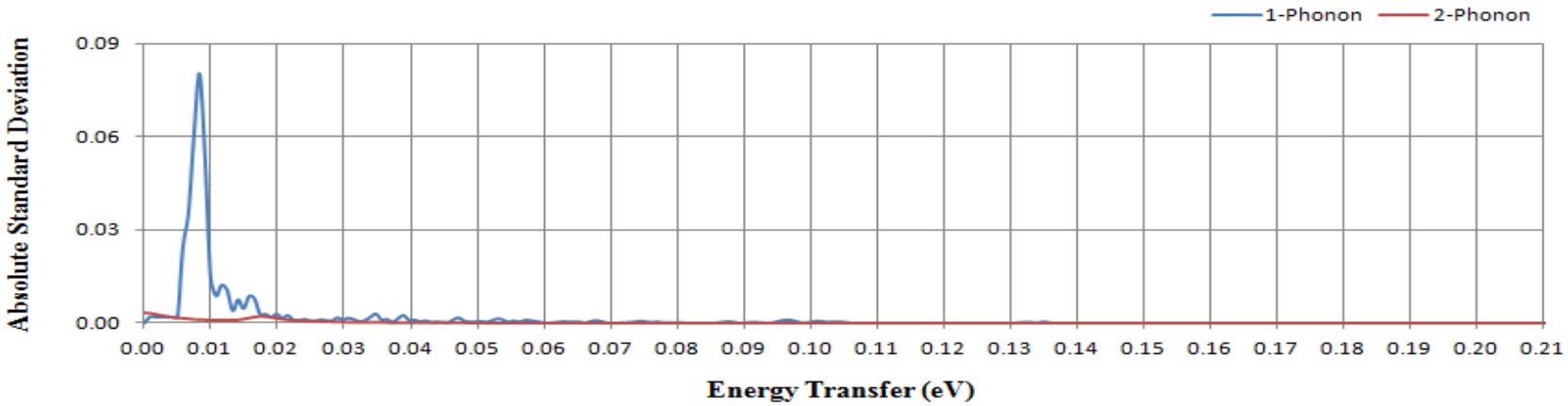
Uncertainty Bands for $S_{ENDF,n}(\alpha=0.3,\beta)$ with n = 1, 2

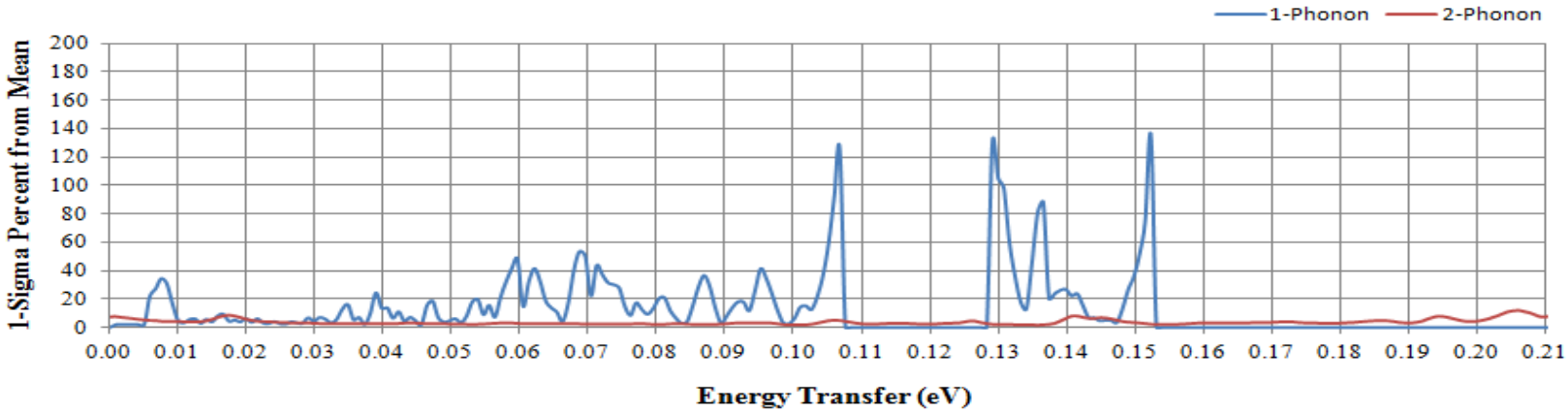




Absolute and Relative Uncertainty in $S_{ENDF,n}(\alpha=0.3,\beta)$ with n = 1, 2

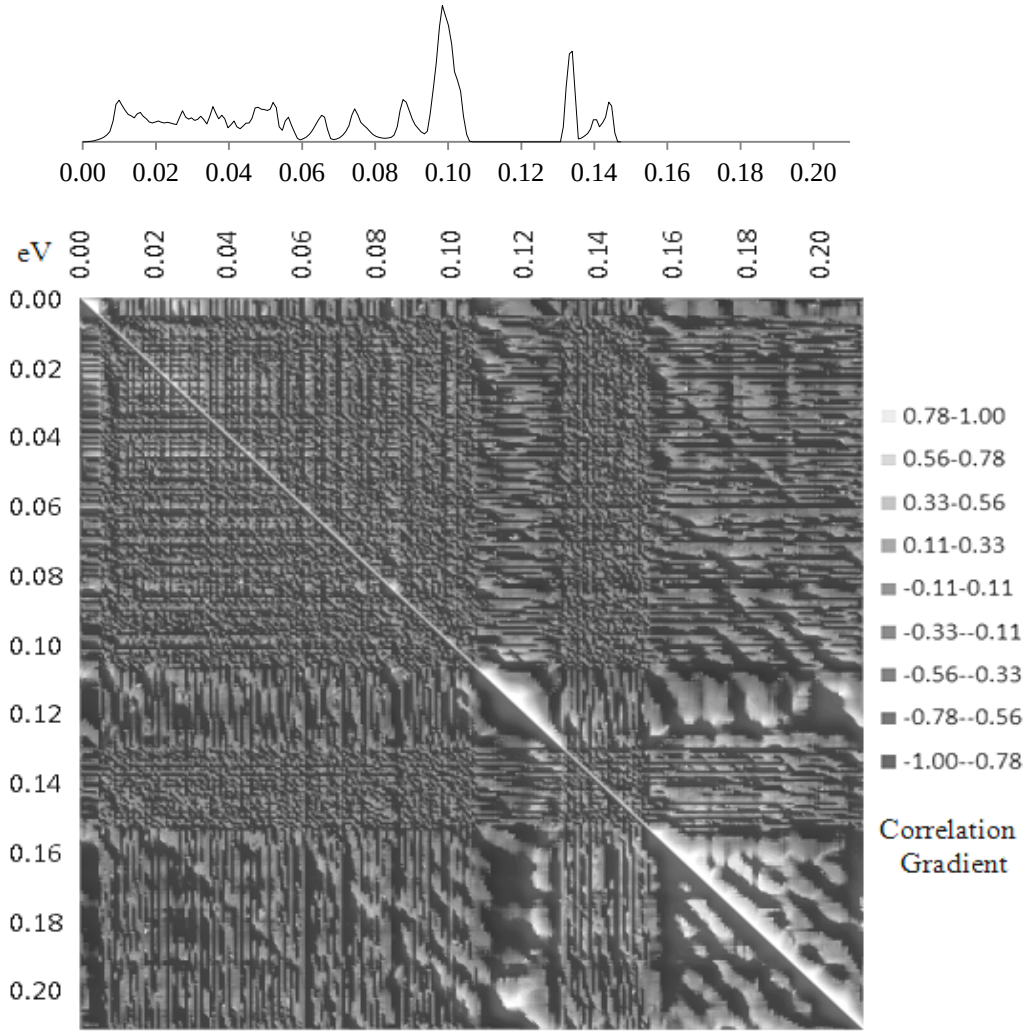






Correlation Matrix for $S_{ENDF,n}(\alpha=0.3,\beta)$ with n = 1+2

$$CORR(y_i, y_j) = \frac{COV(y_i, y_j)}{\sigma(y_i)\sigma(y_j)}$$



Propagating an $S_{ENDF}(\alpha,\beta)$ Covariance Matrix

The general matrix formula for first-order propagation of uncertainty and covariance is

$$COV(z_m, z_n) = \boldsymbol{V}_{z_{mn}} = \boldsymbol{M}_{zy}^{T} \boldsymbol{V}_{y_{ij}} \boldsymbol{M}_{zy}.$$

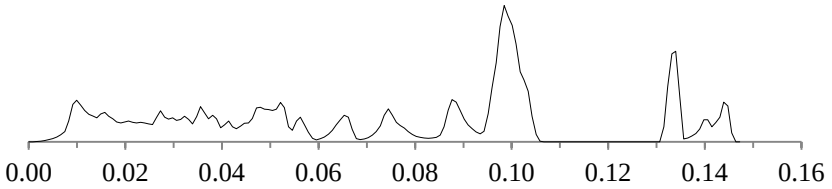
The variables z represent integrated cross sections (across any range of α and/or β), the variables y represent $S_{ENDF}(\alpha,\beta)$ terms, and $\mathbf{M}_{zy}$ is the sensitivity matrix with entries

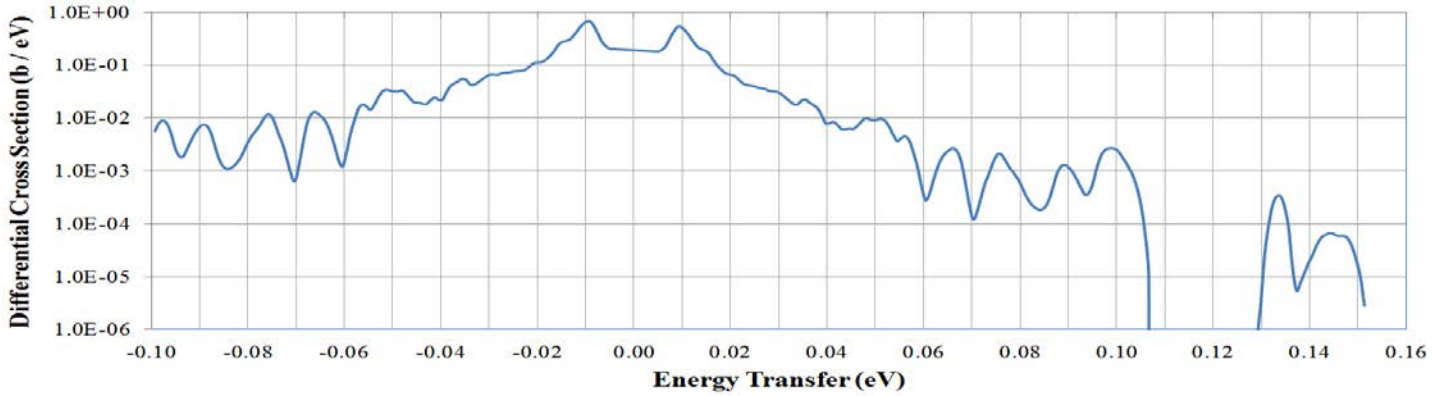
$$\boldsymbol{M}_{im} = \frac{\partial z_m}{\partial y_i}.$$

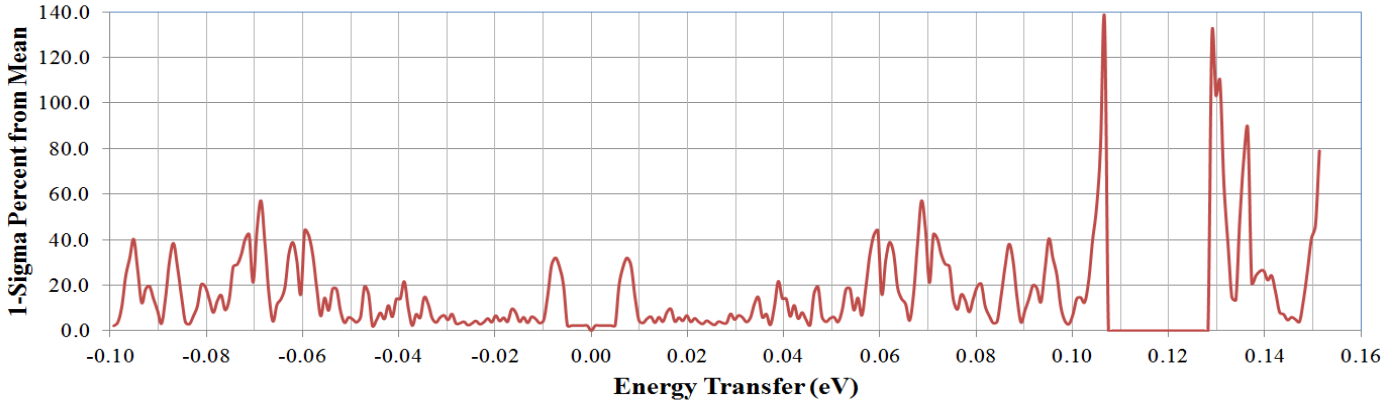
Given the relationship $\frac{d^2\sigma_{inel}(E)}{d\alpha\, d\beta} = \frac{Ak_BT\sigma_b}{4E} e^{\beta/2} S_{ENDF}(\alpha,\beta)$, the sensitivity matrix can be computed analytically and need not be stored in file space.

The $S_{ENDF}(\alpha,\beta)$ covariance matrix can be propagated to yield covariance matrices for secondary neutron energy distributions, coupled energy-angle bin distributions, or integrated inelastic scattering cross sections.

Impact on Secondary Neutron Energy Distributions
(1-Phonon Distribution for 1.0 eV Incident Energy in 293.6 K Material)







Summary / Conclusions

- Development of an S(α,β) covariance matrix would allow construction of covariance matrices for secondary neutron energy distributions, coupled energy-angle distributions and integrated inelastic cross sections.
- For the current methodology of producing thermal cross sections from a physics model, covariances among S(α,β) data must reflect uncertainties in the DOS input parameter, ρ|β|.
- Uncertainties in the DOS can be addressed through discretization of the DOS into energy-dependent PDFs.
- At room temperature, the majority of uncertainty in thermal neutron scattering arises from uncertainty in the one-phonon terms of S(α,β).
- Large uncertainties in particular S(α,β) values and in secondary neutron energy distributions at corresponding β values can exist while integrated inelastic cross section uncertainties may be small.