Naval Nuclear Laboratory

Thermal Scattering Physics Methods with Modeling Tools and Experiments

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Outline of Topics

- Pulsed-Neutron Die-Away (PNDA) theory and advantages as a validation and benchmark tool for thermal scattering laws (TSLs)

- Pulsed-Neutron Die-Away (PNDA) experiments, modeling, and physics analyses for ice, liquid water, and other materials.

- Coherent interference calculations in S(α,β) with FLASSH compared to experimental results.

- Future evaluation strategies and concerns.

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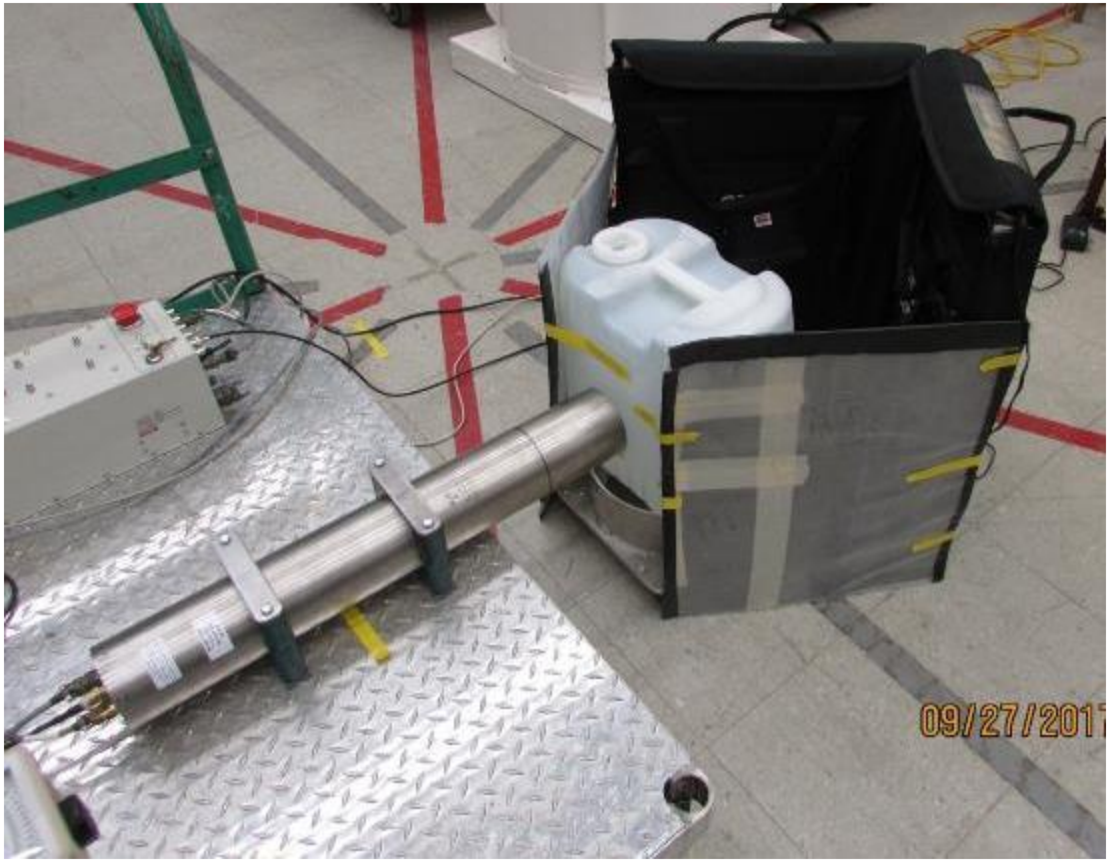
Lawrence Livermore National Laboratory (LLNL) Proof-of-Principle PNDA Experiment



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Validating Thermal Scattering Laws with Pulsed-Neutron Die-Away Diffusion Benchmarks

- A neutron generator (D+D or D+T) is used to target a pure material sample with a short mono-energetic neutron pulse (14 MeV for D+T).

- ~ 10^{-5} to 10^{-3} seconds following the pulse, the neutron population is in thermal equilibrium in the fundamental spatial mode with characteristic flux decay time eigenvalue α.

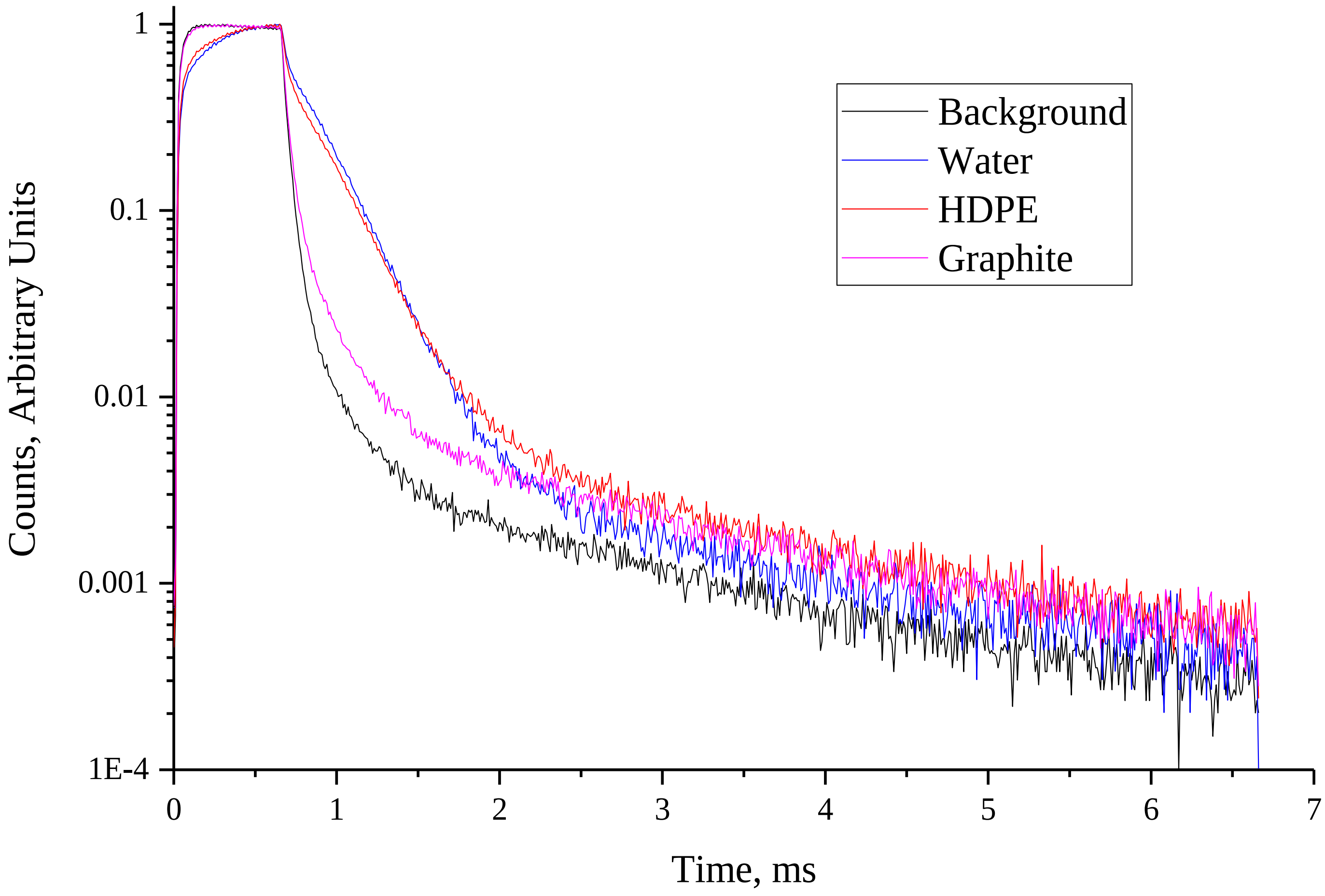
$$\varphi(\mathbf{r},t) = \varphi_0(\mathbf{r}) \exp(-\alpha t) + R(t)$$

- R(t) is room return, which can be modeled or zeroed in simulations and usually made negligible in experiments through cadmium shielding.

- In the one-speed diffusion model, the eigenvalue will have the form $\alpha = v\Sigma_a + vDB^2 - CB^4$, where

$\Sigma_a \equiv$ thermally averaged macroscopic absorption cross section,
$D = 1/\Sigma_{transport} \equiv$ diffusion coefficient,
$v \equiv$ thermally averaged neutron velocity,
$B^2 \equiv$ geometric buckling, and
$C \equiv$ diffusion cooling coefficient

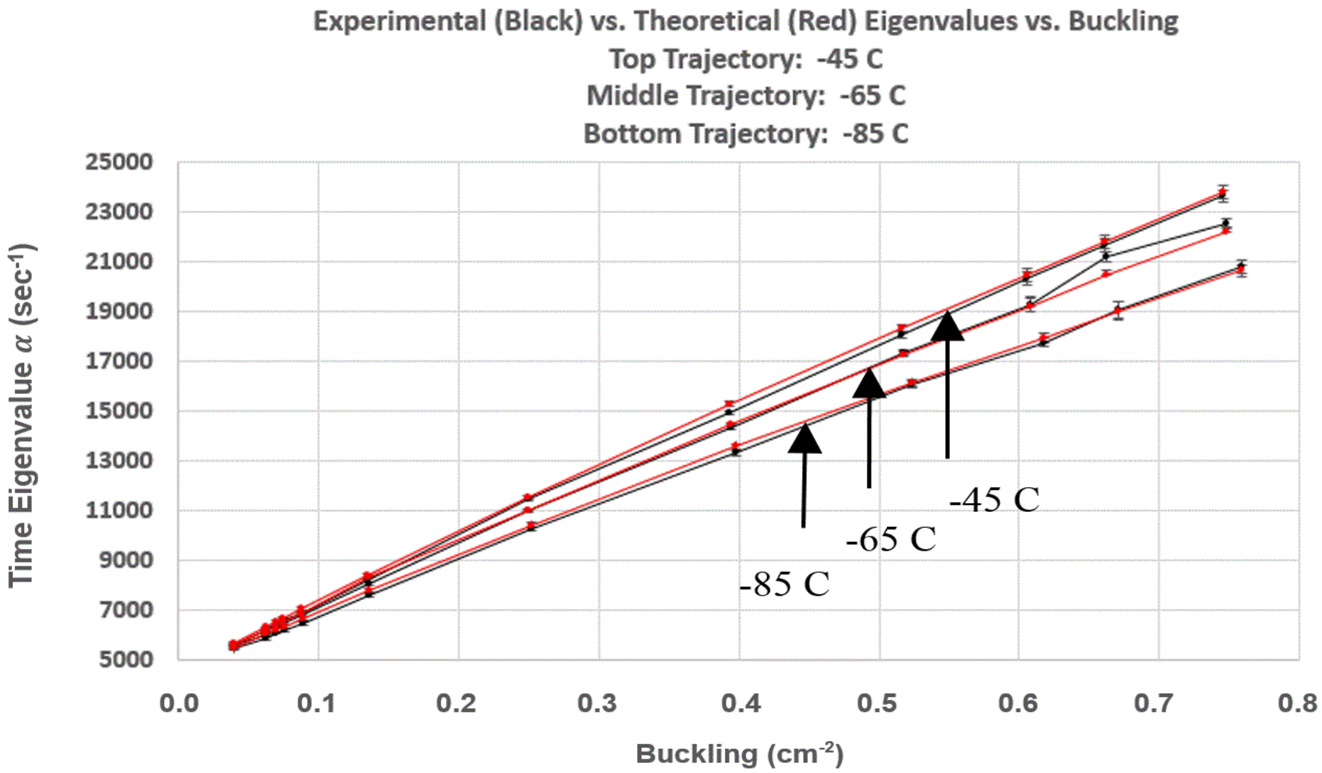
LLNL Experimental Flux Decay Profiles



Validating Thermal Scattering Laws with Pulsed-Neutron Die-Away Diffusion Benchmarks

- $\alpha = v\Sigma_a + vDB^2 - CB^4$ has quadratic form in B^2. As the cooling coefficient C is typically extremely small, α is nearly a linear function of geometric buckling with slope characteristic of the material cross sections.
- For very large systems, where B^2 is very small, α primarily depends on absorption. This is physically true independent of any model. By calculating α over a range of buckling values, the zero-buckling pure-absorption term can be extrapolated.
- For very small systems, where B^2 is very large, α primarily depends on D, which is strongly dependent on the physics of thermal scattering.
- Physically, α always depends on the particular details of integral and differential thermal scattering cross sections (and on absorption). The sensitivity of α to thermal scattering increases with geometric buckling (as sample size decreases).
- The neutron flux decay following the pulse is measured over a period of time, allowing a comparison of α values calculated experimentally and by Monte Carlo simulation.

2016 MC21 PNDA Results for Ice Cylinders with ENDF/B-VIII.0 Ice I*h* TSLs Compared to Experiment



Experimental Data: E. G. Silver, NSE Vol. 34 (1968)

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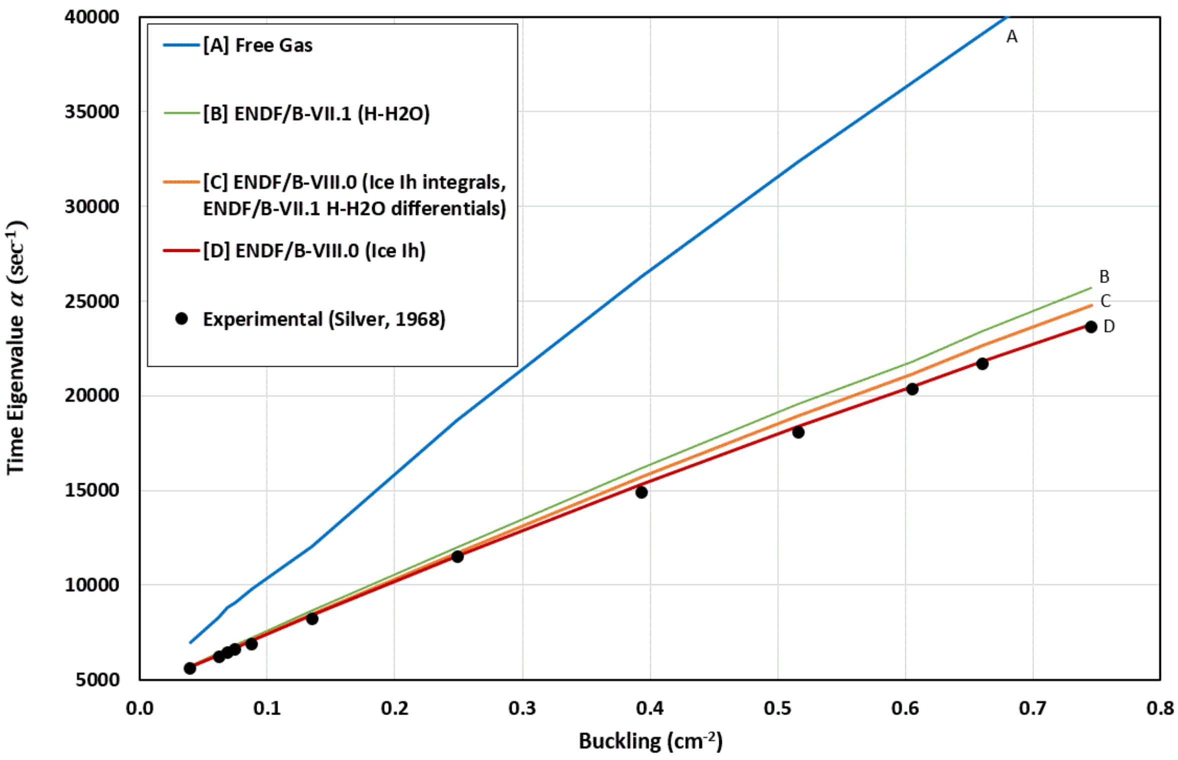
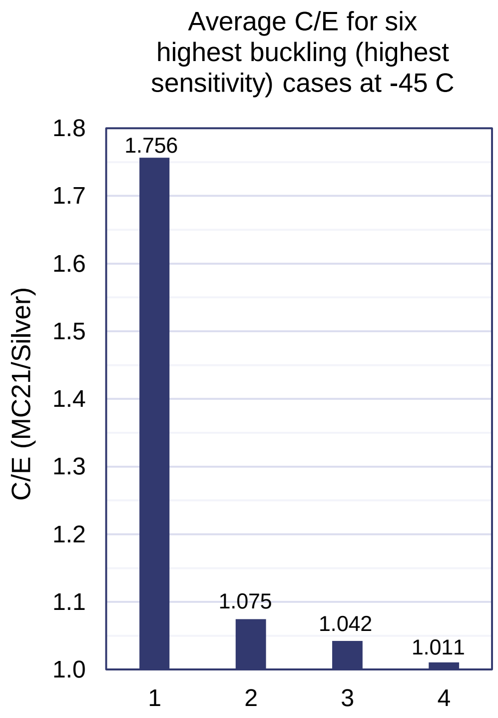
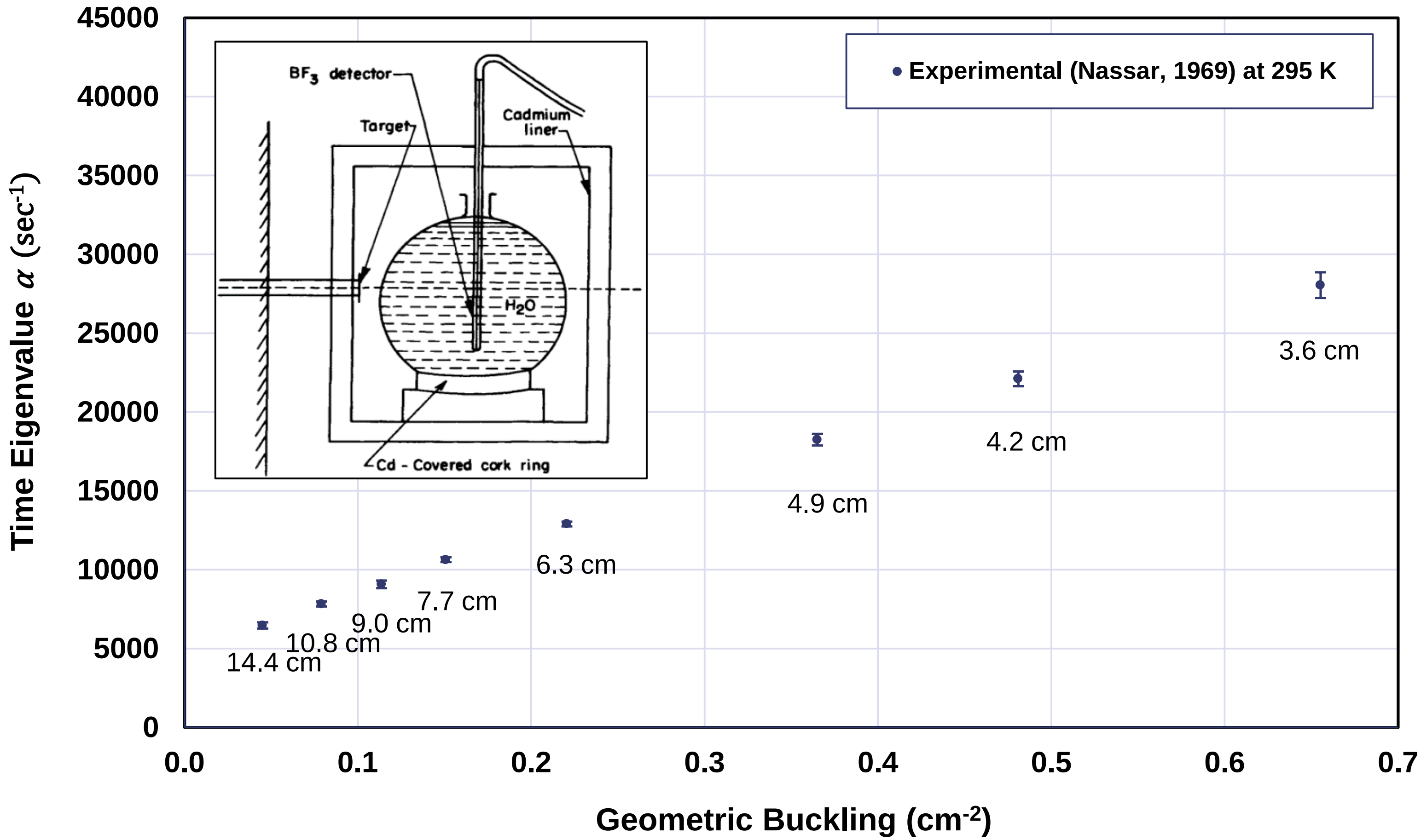
2016 MC21 Test for PNDA Sensitivity to Integral and Differential Cross Sections (Ice I*h* at -45 C)





Experimental Data: E. G. Silver, NSE Vol. 34 (1968)

Thermal Flux Time Decay Eigenvalues vs. Buckling for Liquid Water Spheres Measured by Nassar and Murphy, NSE Vol. 35 (1969)



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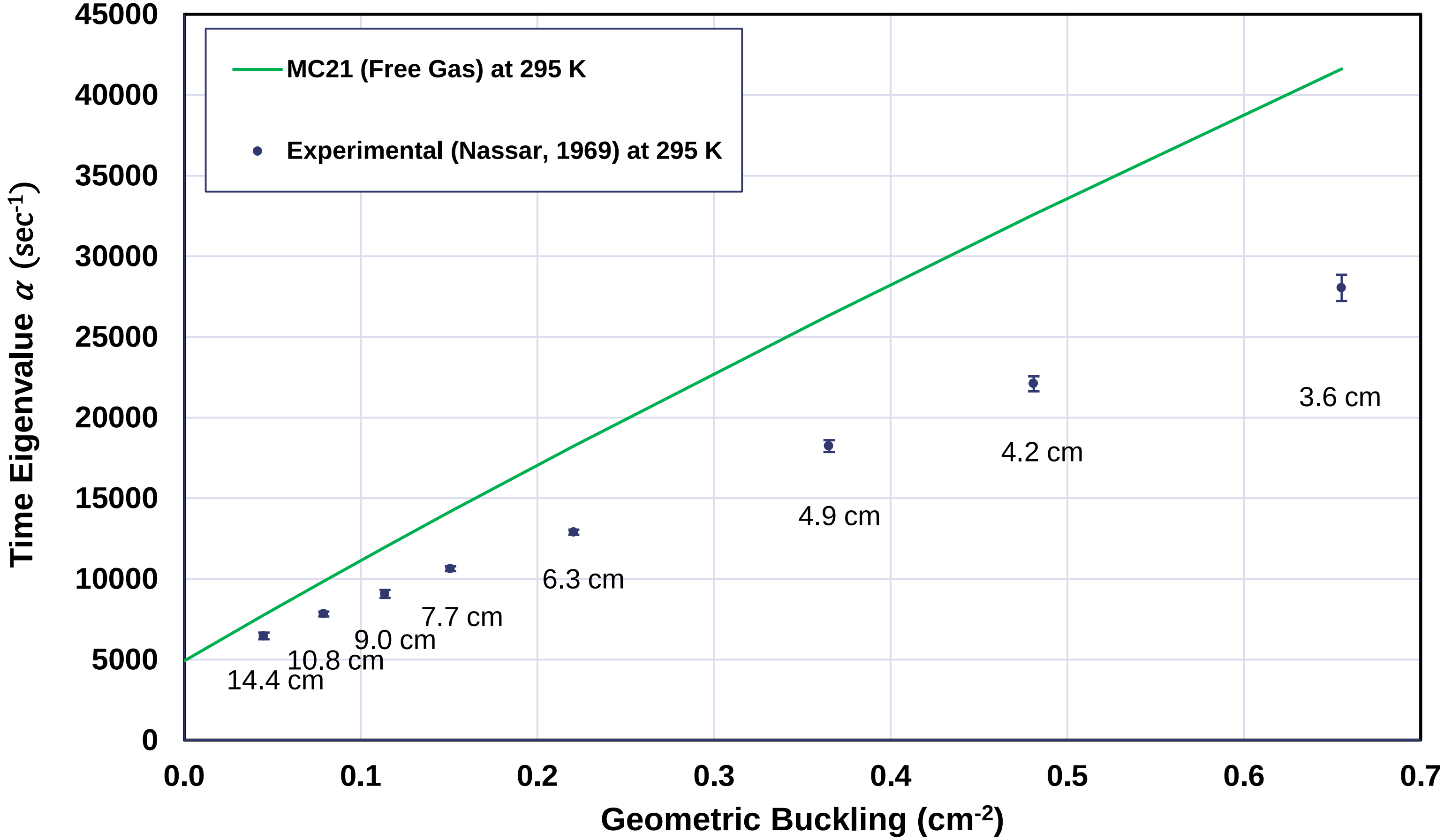
Thermal Flux Time Decay Eigenvalues vs. Buckling for Liquid Water Spheres Compared to MC21 Using Free Gas Thermal Scattering



Experimental Data: Nassar and Murphy, NSE Vol. 35 (1969)

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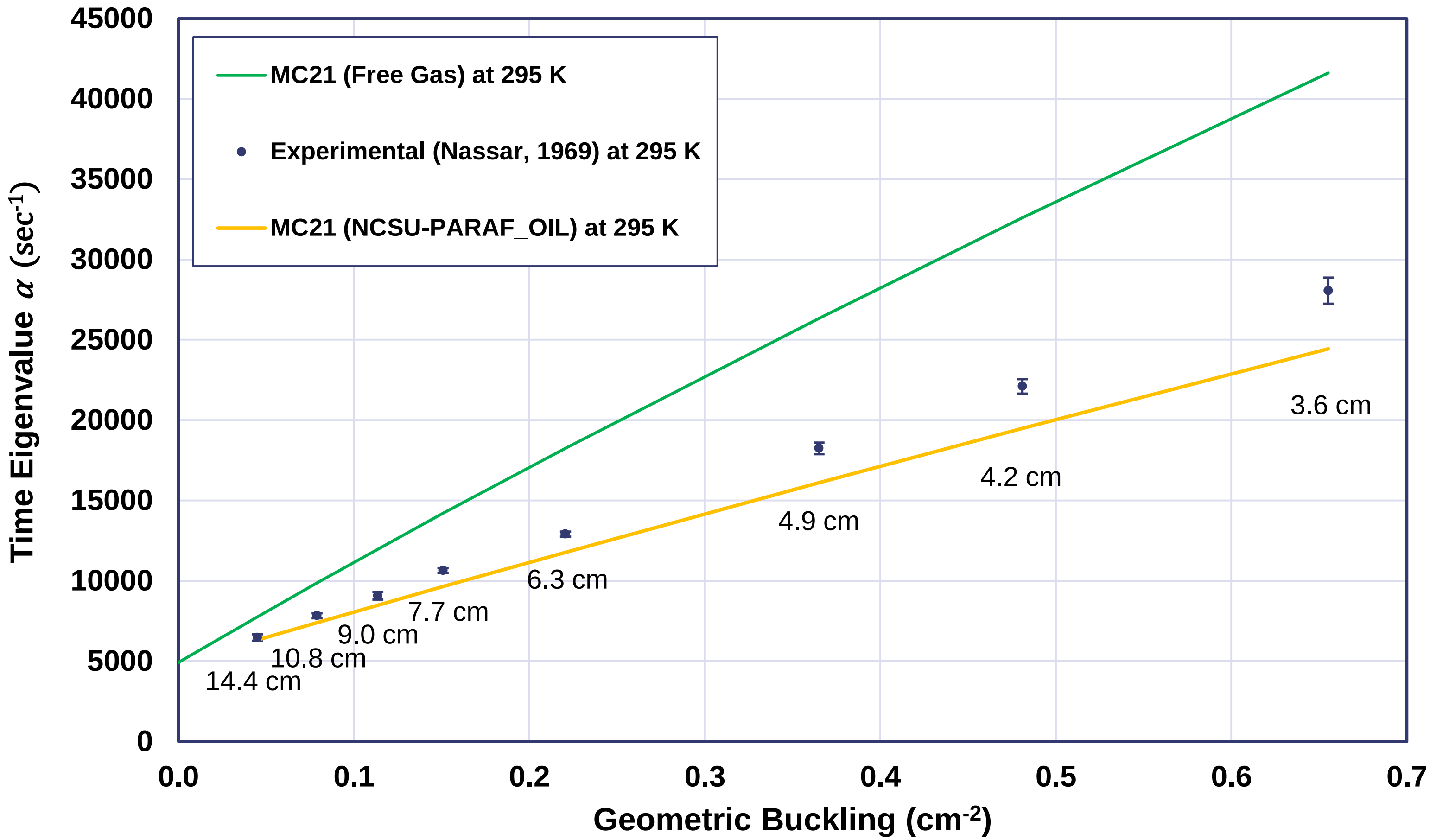
Thermal Flux Time Decay Eigenvalues vs. Buckling for Liquid Water Spheres with Selected Thermal Neutron Scattering Kernels



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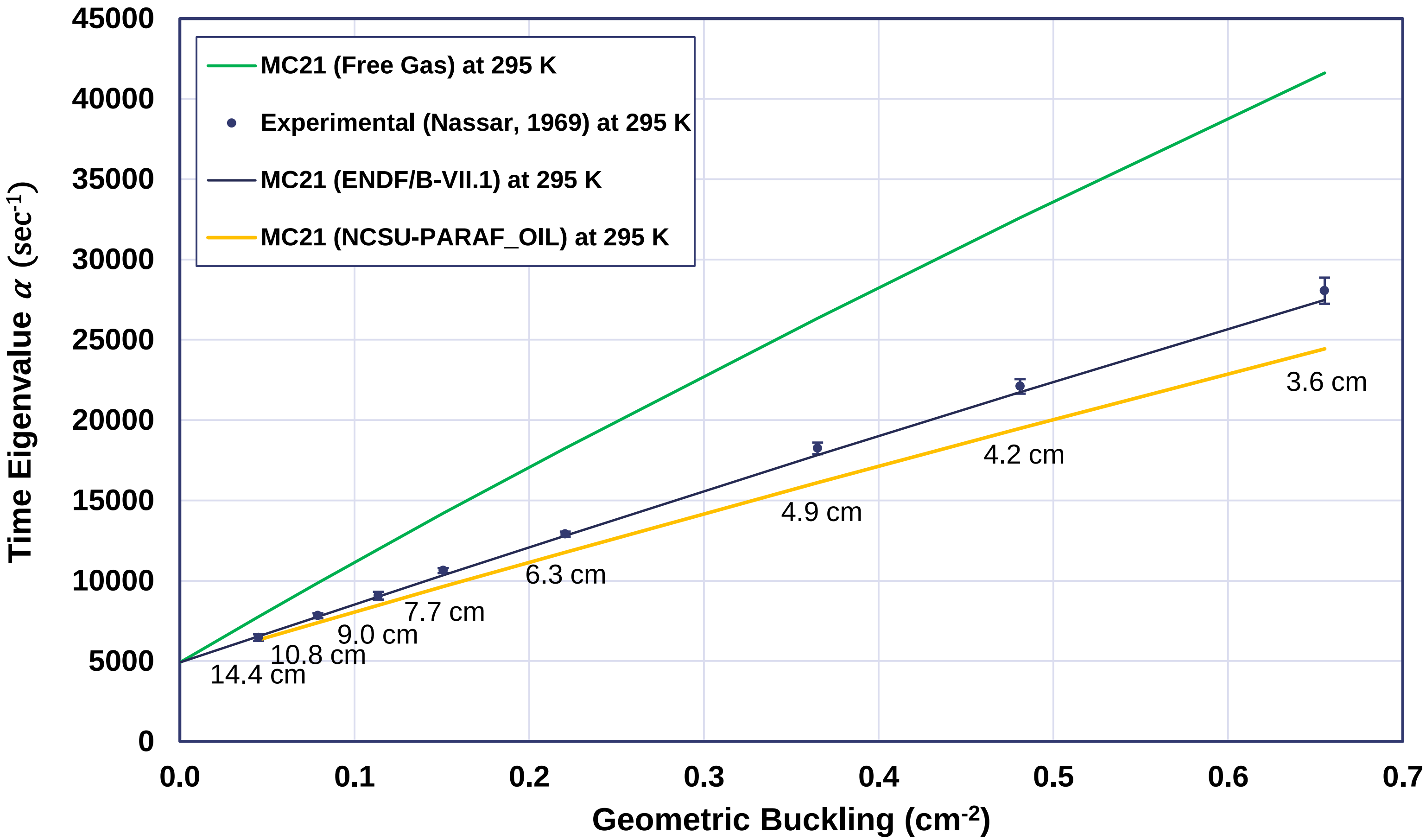

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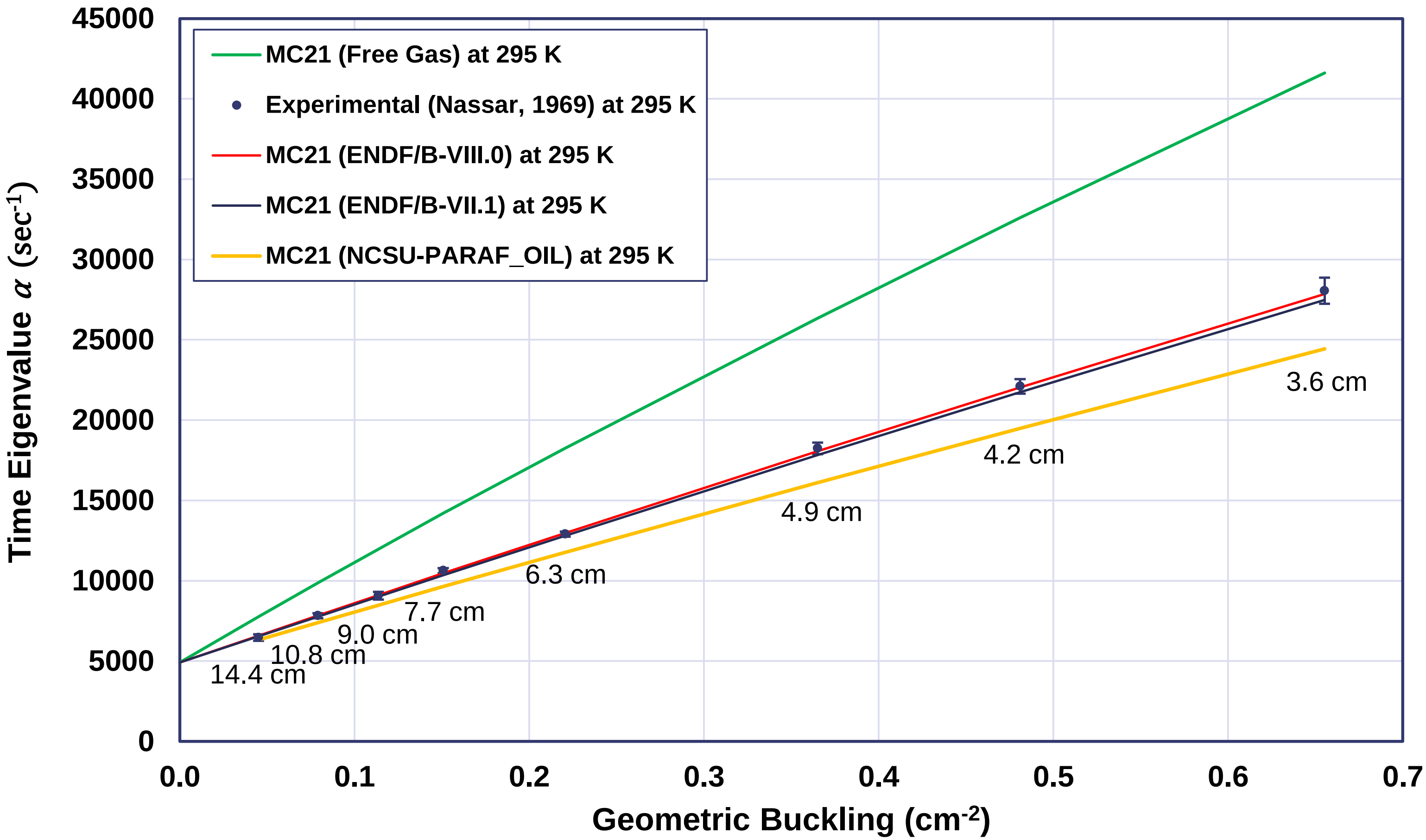

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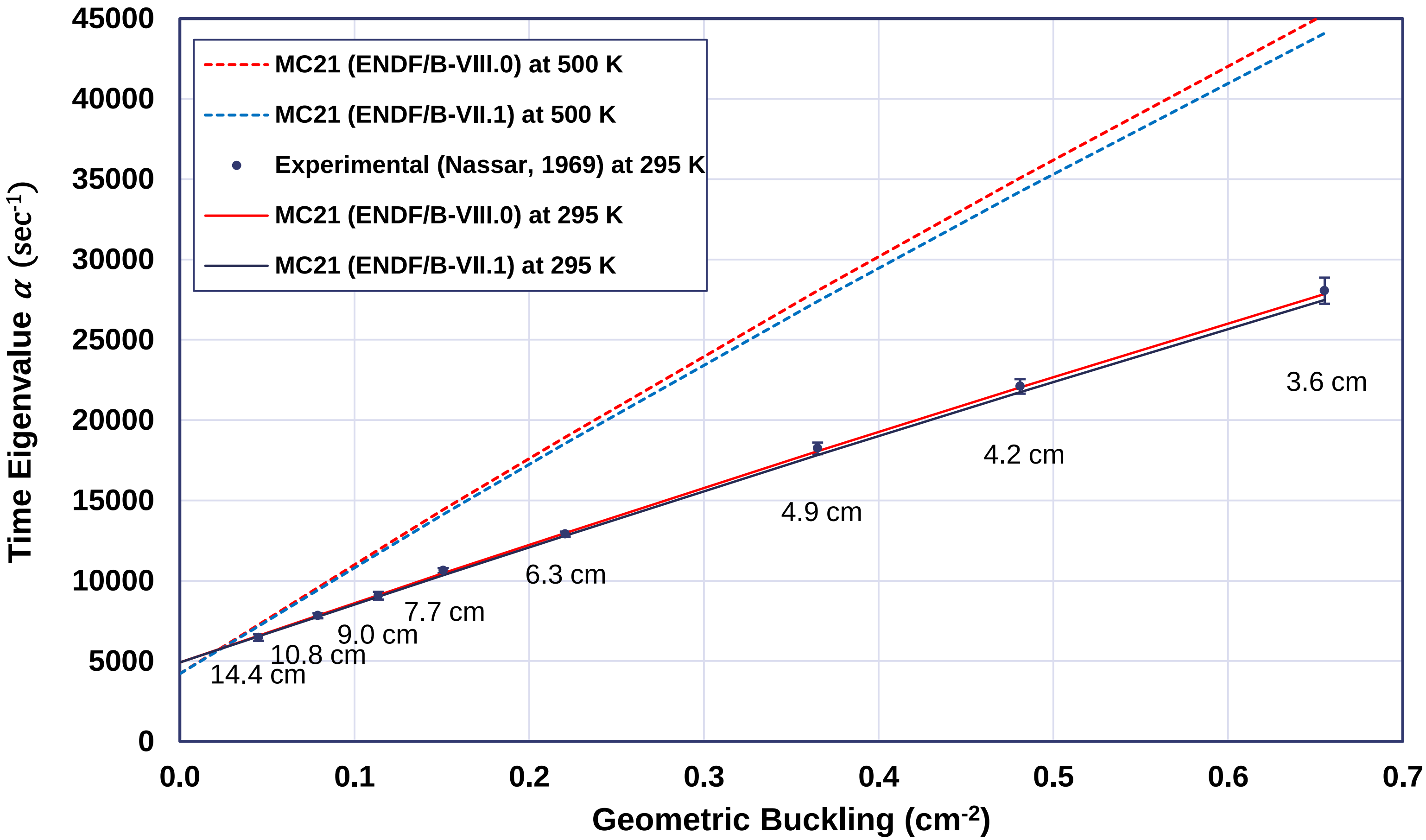
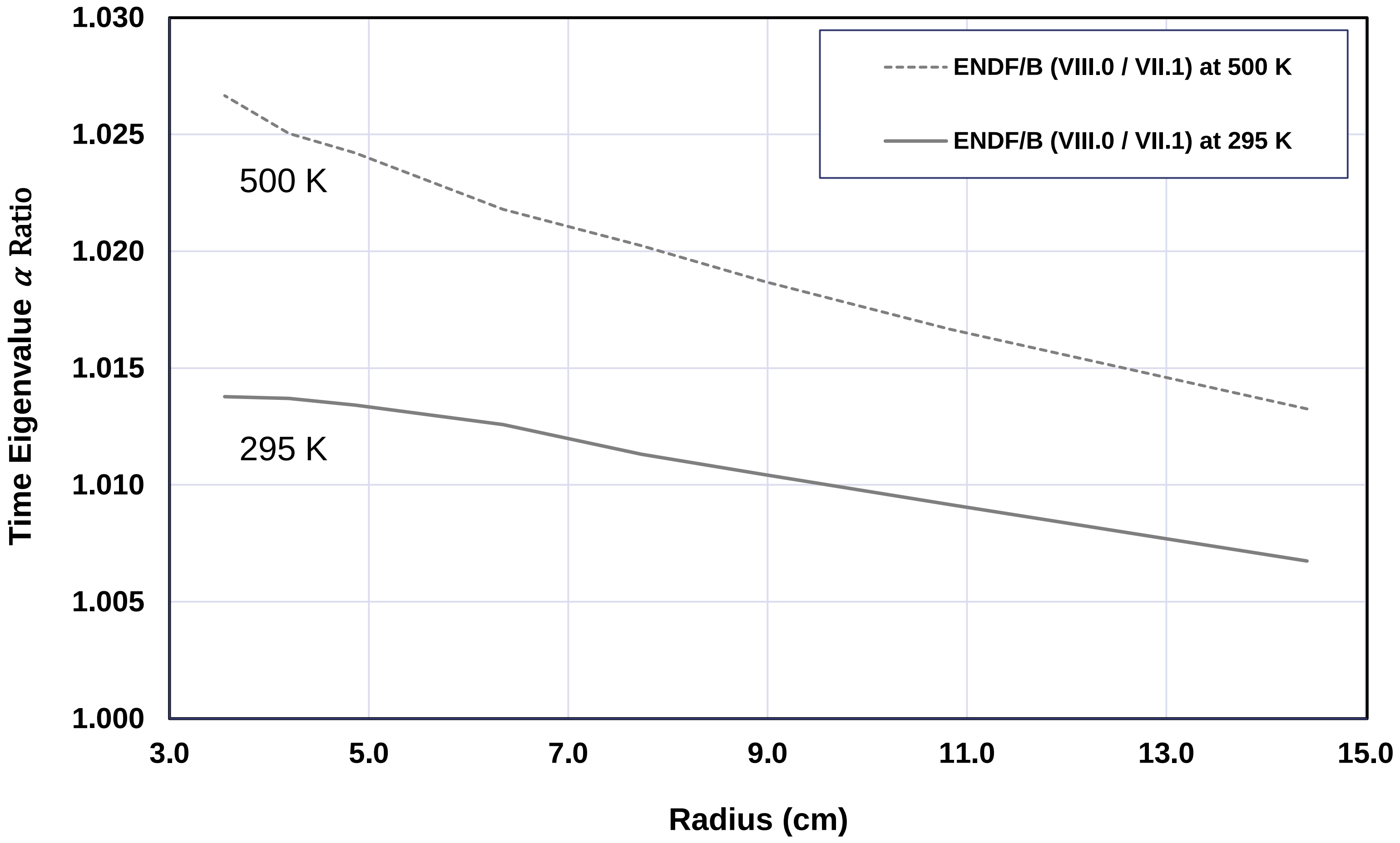
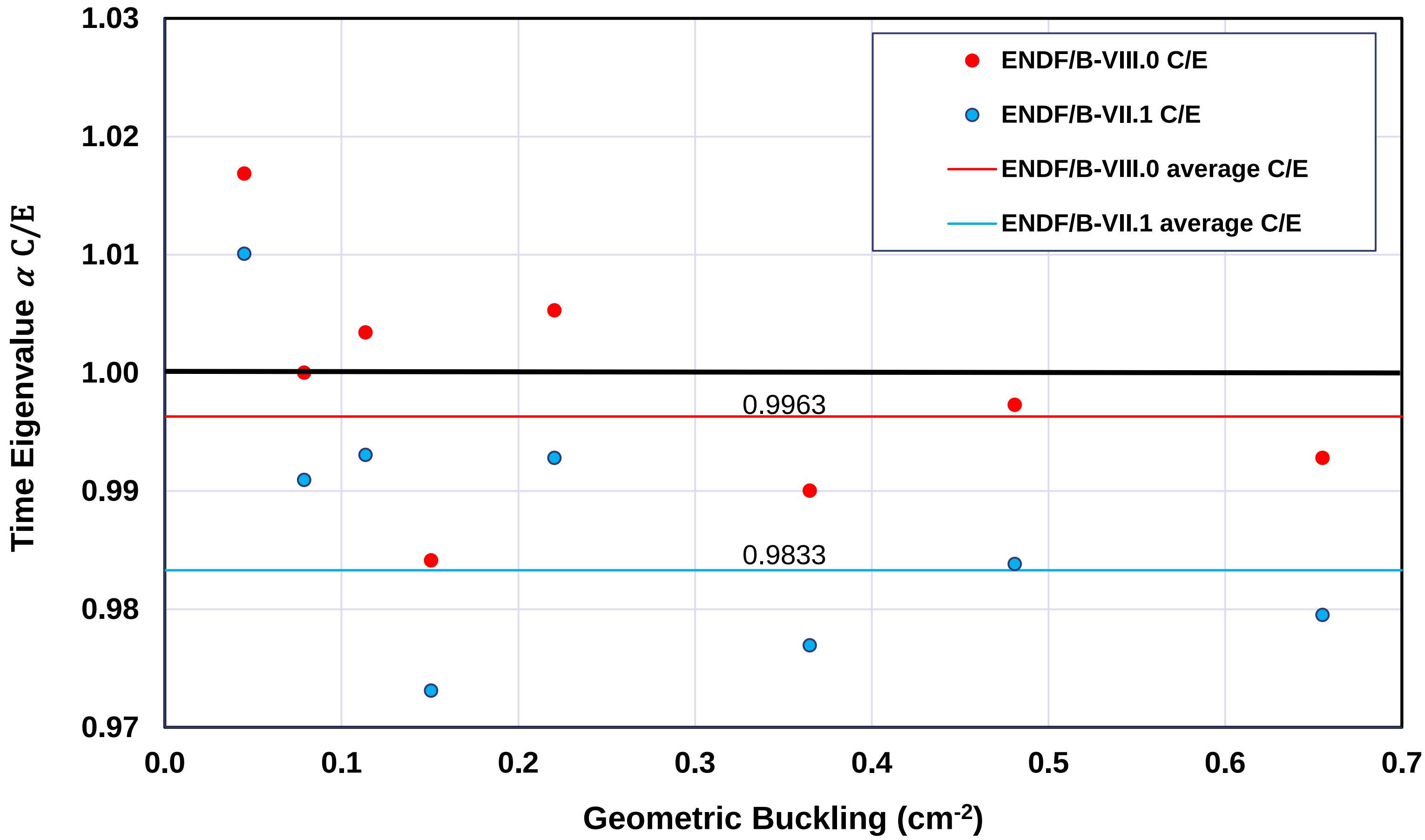


Experimental Data: Nassar and Murphy, NSE Vol. 35 (1969)

Thermal Flux Time Decay Eigenvalue Ratios vs. Buckling for Liquid Water Spheres (ENDF-VIII.0 / -VII.1 at 500 K and 295 K)



Thermal Flux Time Decay Eigenvalue C/E vs. Buckling for Liquid Water Spheres at 295 K (ENDF/B-VIII.0 vs. -VII.1)



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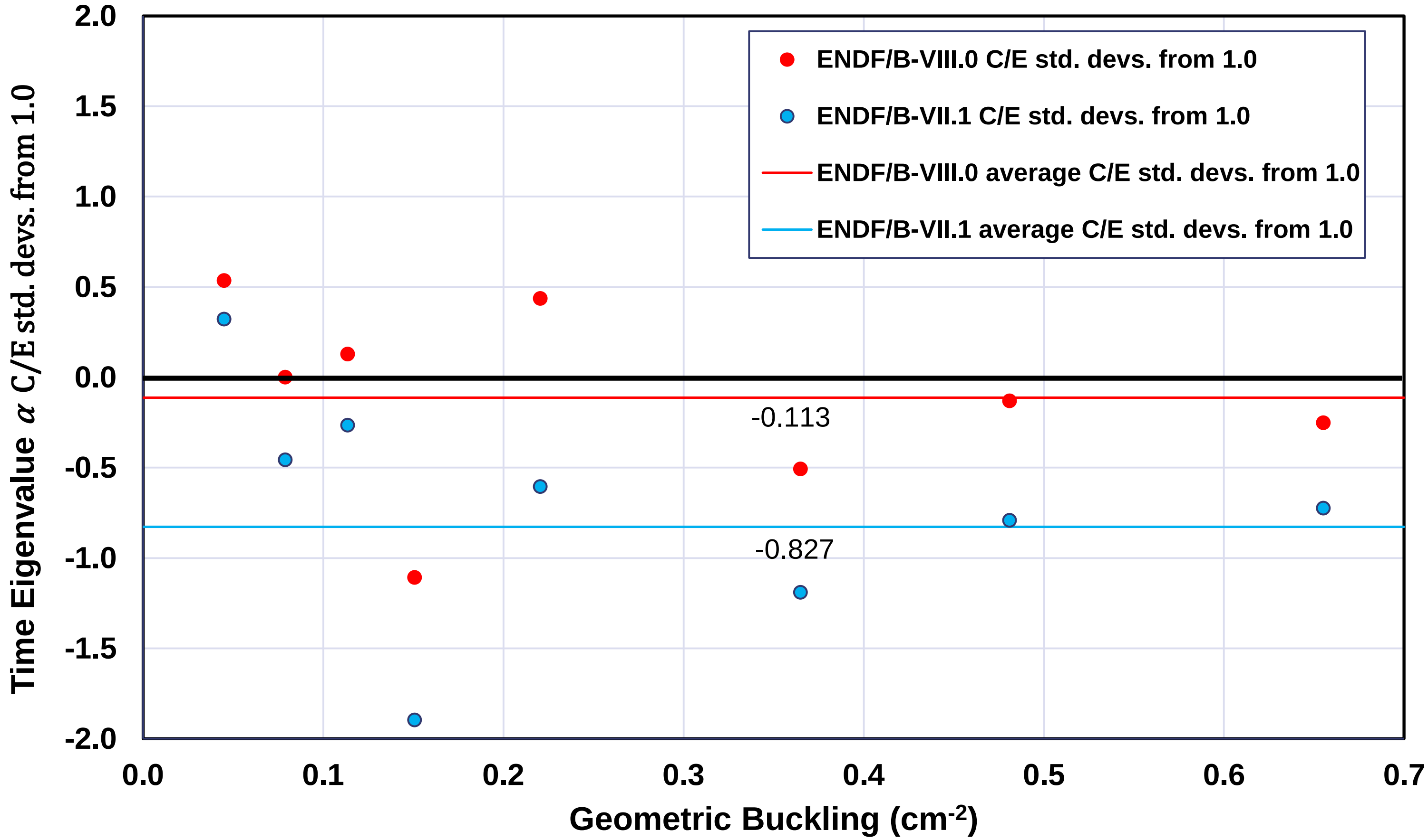
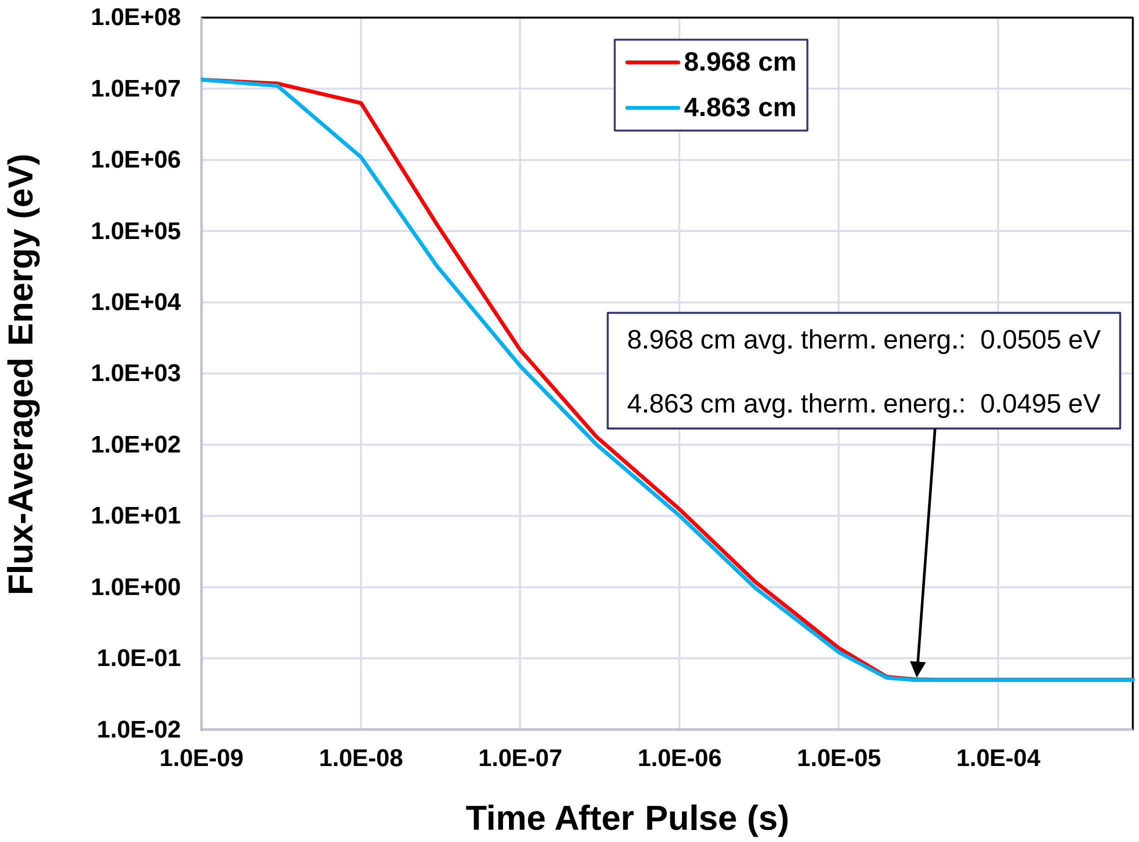
Thermal Flux Time Decay Eigenvalue C/E std. devs. from 1.0 vs. Buckling for Liquid Water Spheres at 295 K (ENDF/B-VIII.0 vs. -VII.1)



MC21 Average Energy Slowing-Down Profile for Liquid Water Spheres at 295 K



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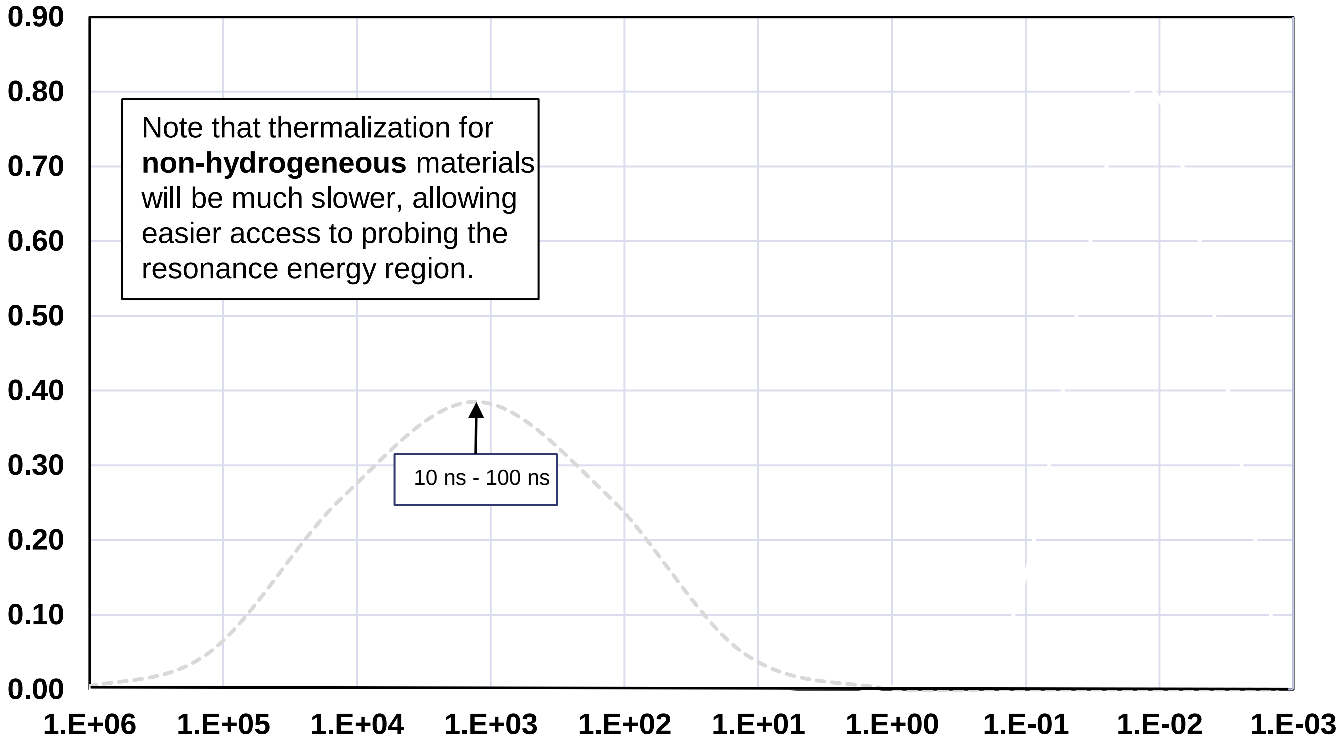
MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



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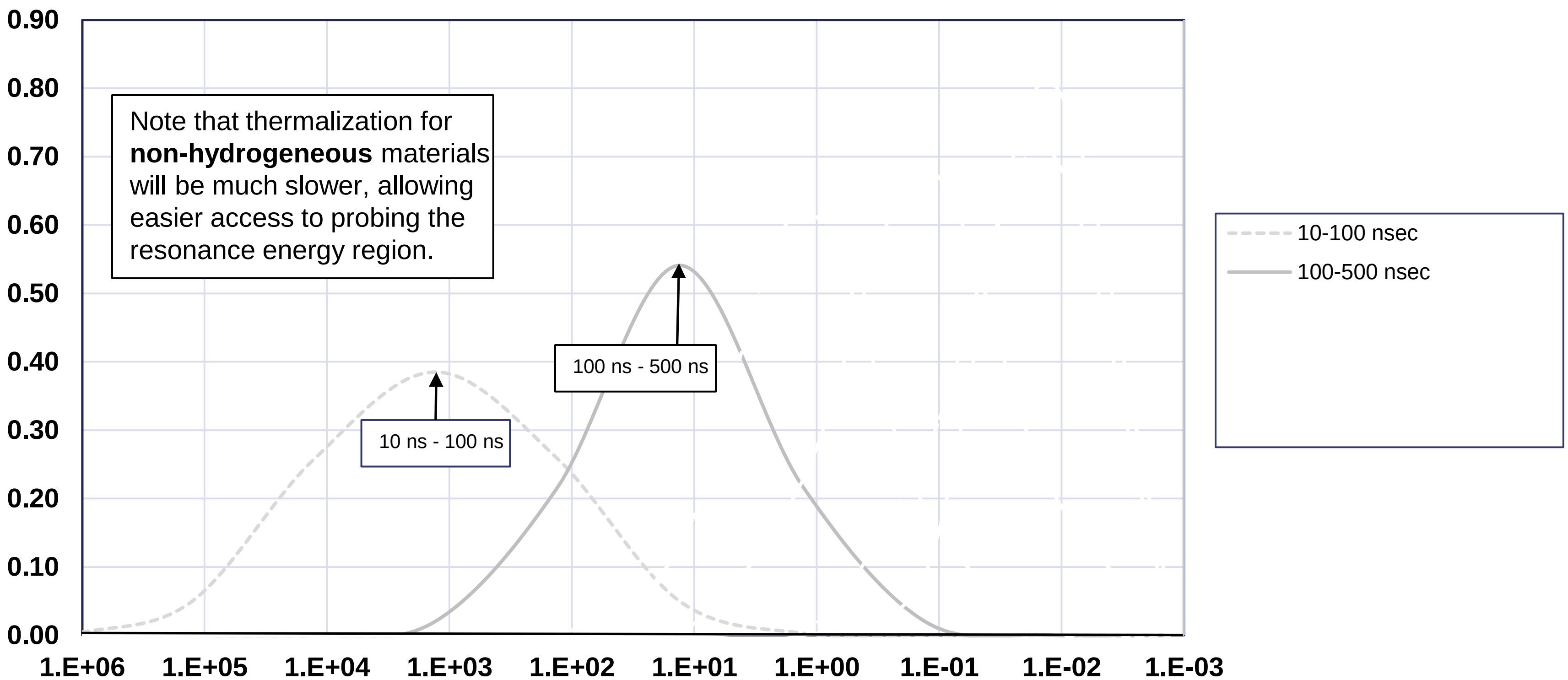
MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



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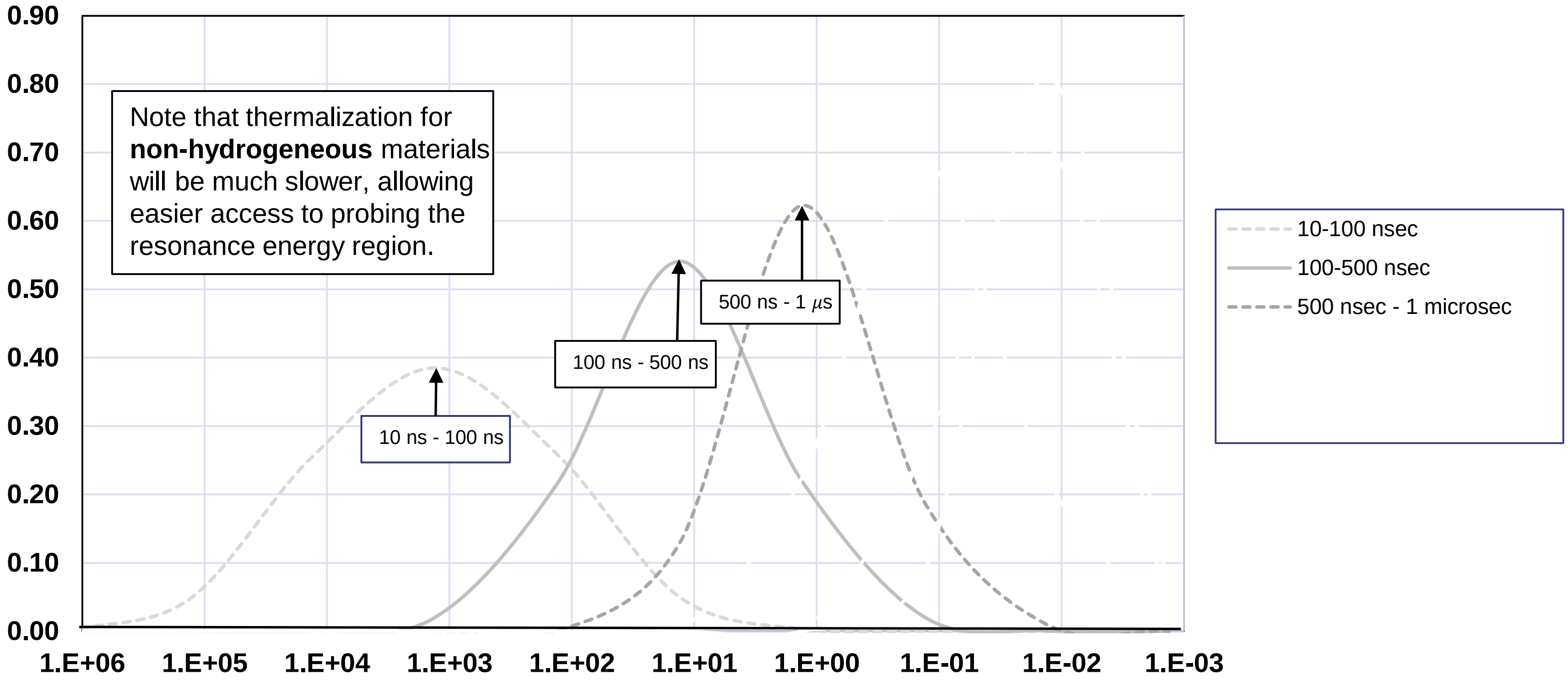
MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



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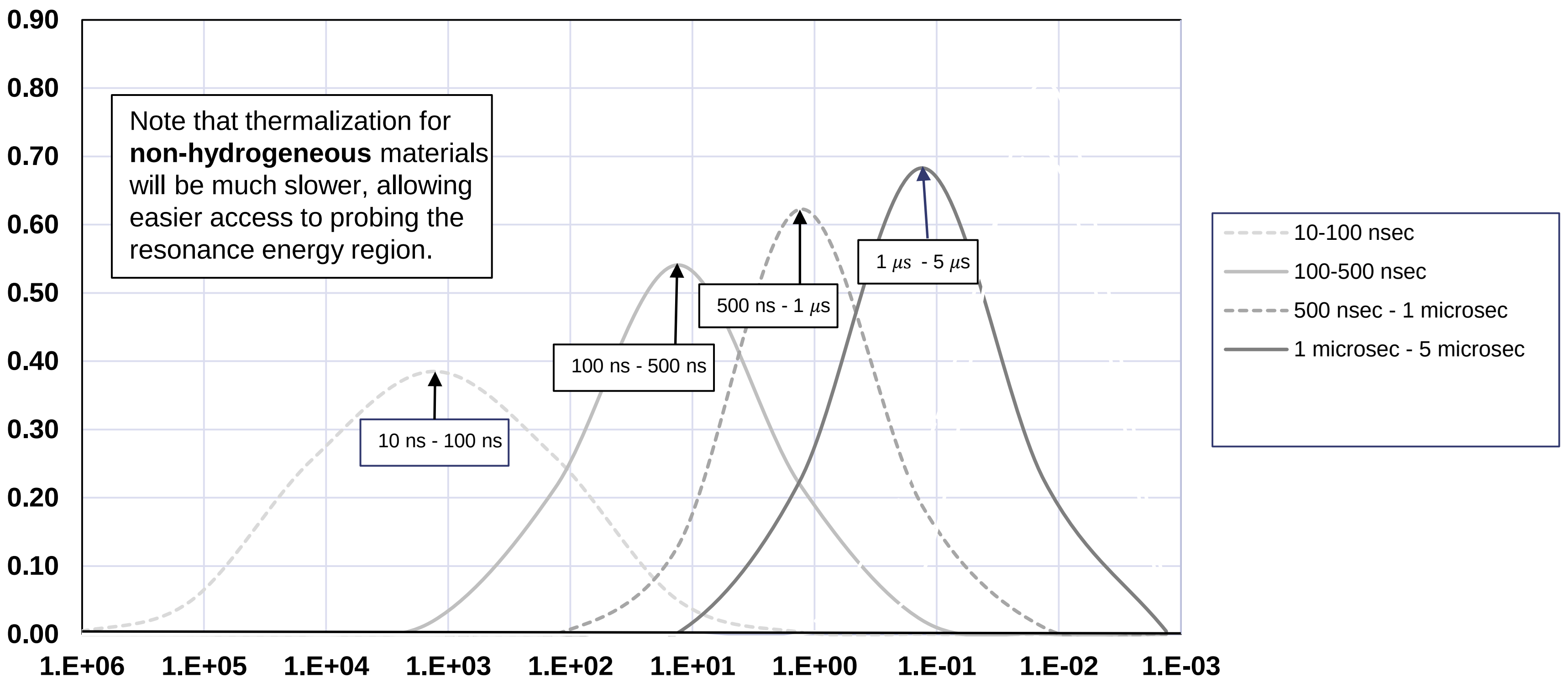
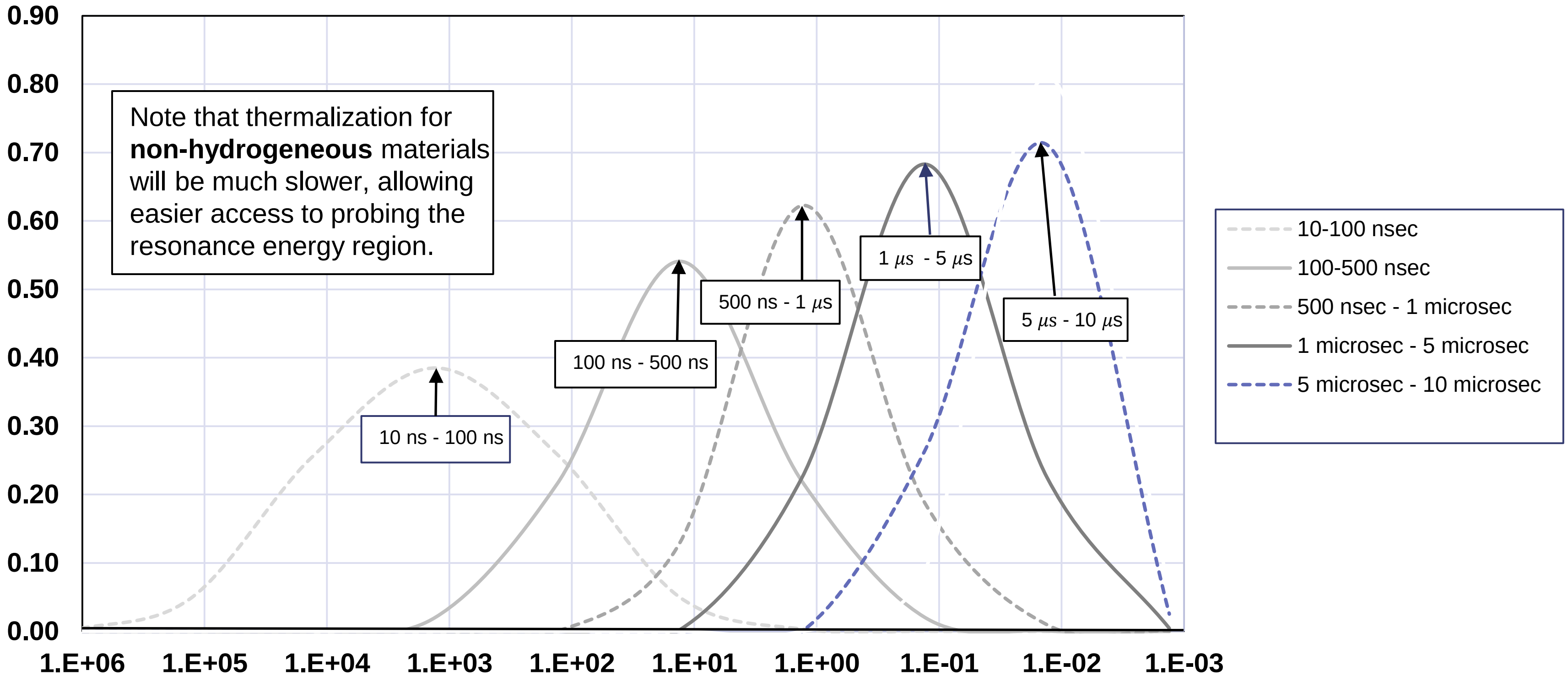
MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



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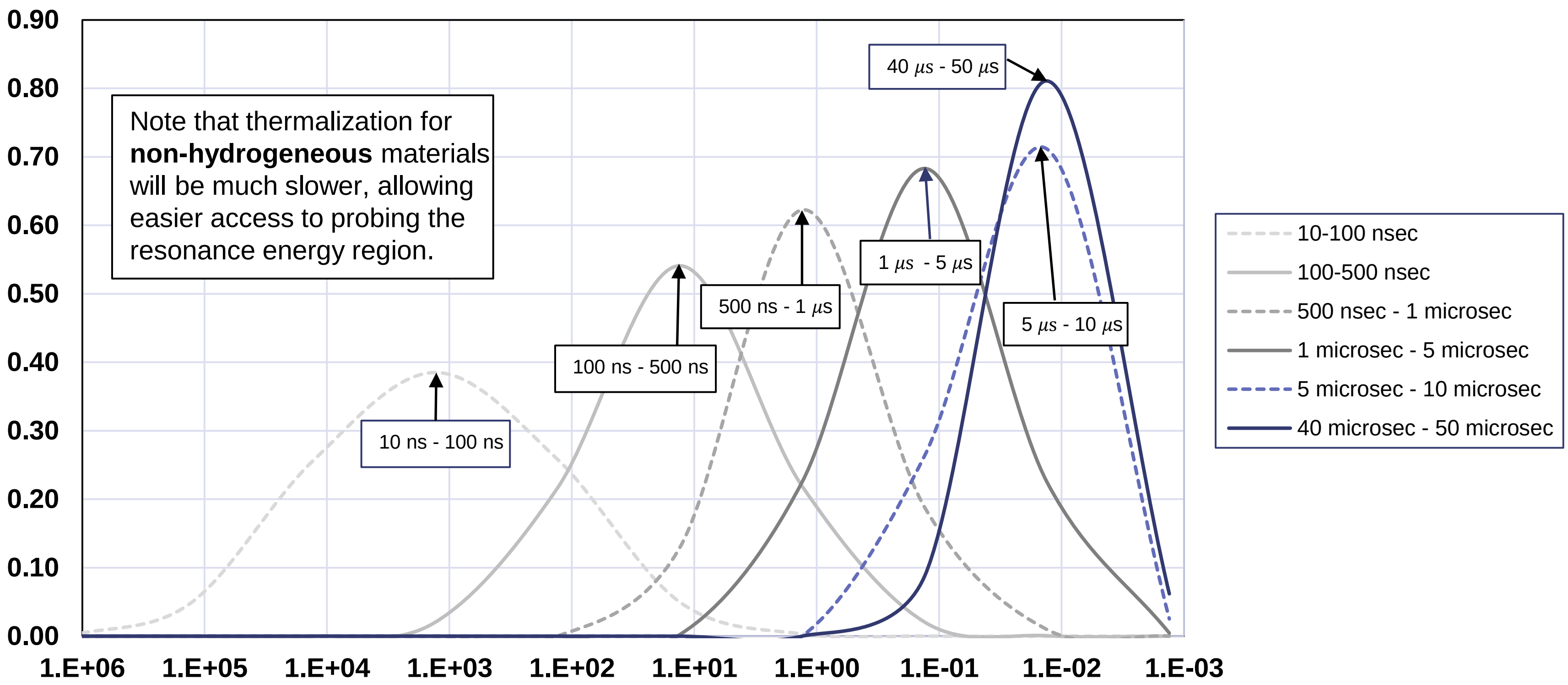
MC21 Time-Dependent Energy Distribution Profile for Liquid Water Spheres at 295 K



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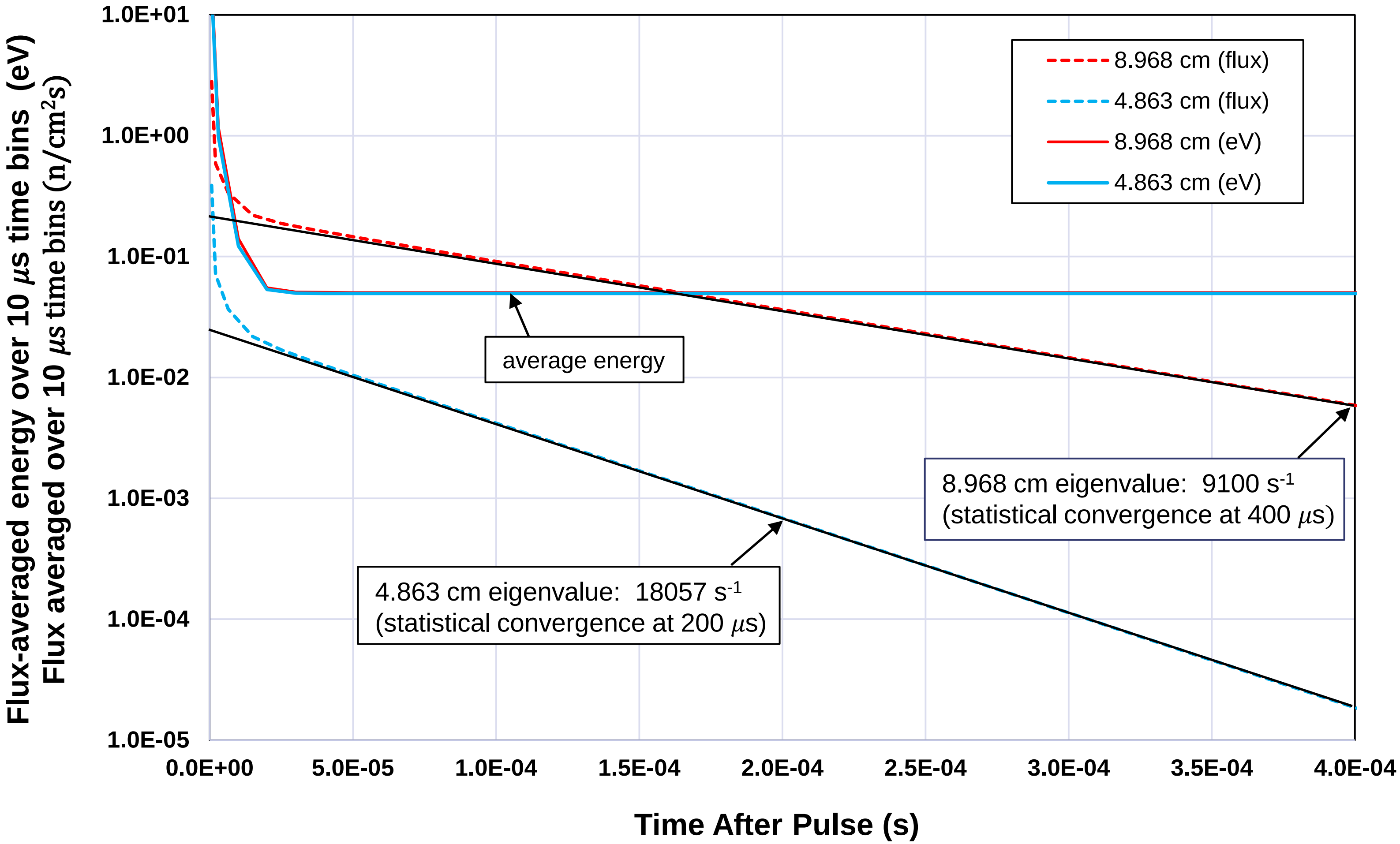
MC21 Flux Decay Profile for 10 μ-sec Time Bins for Liquid Water Spheres at 295 K



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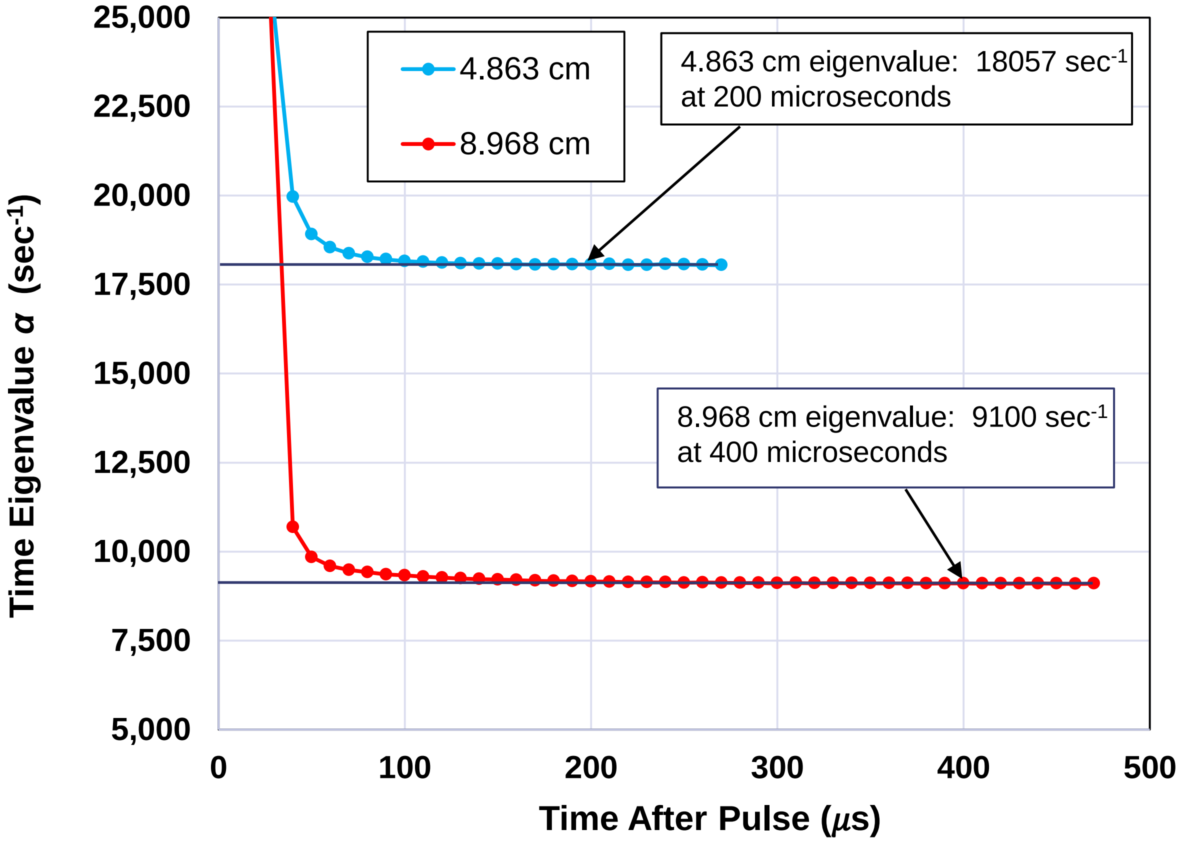
MC21 Eigenvalue Convergence for Liquid Water Spheres at 295 K



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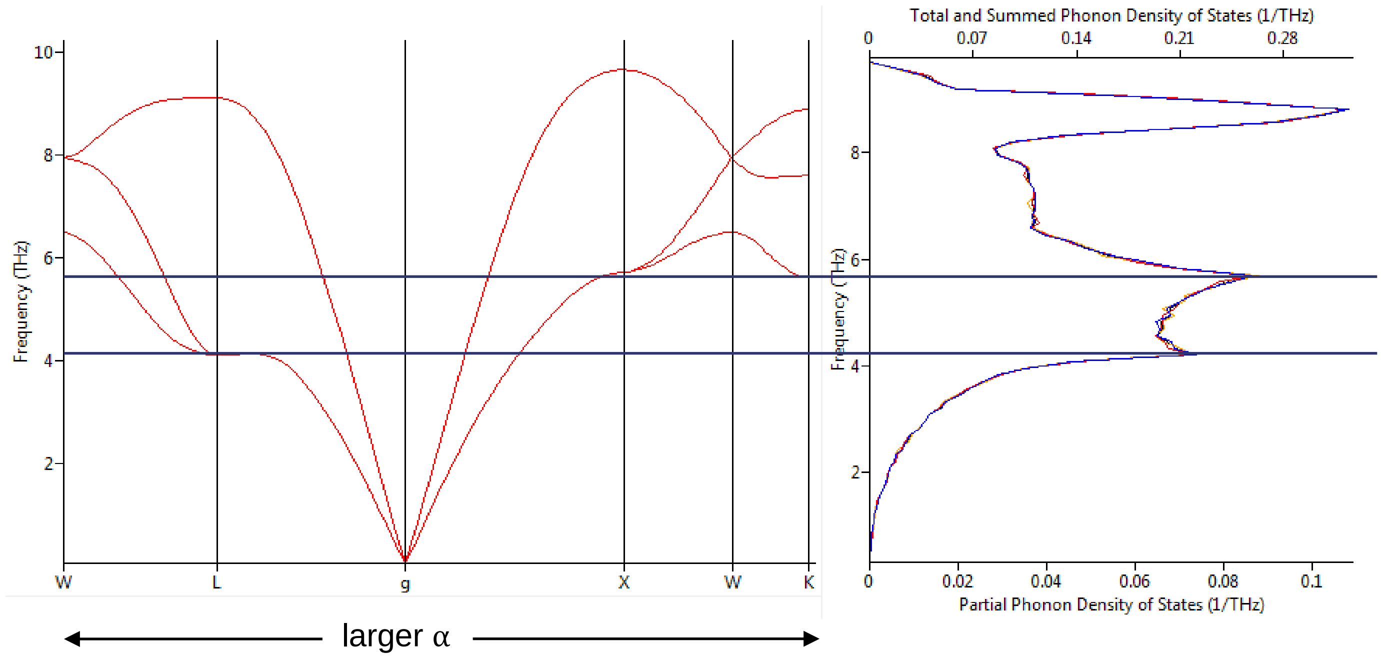
Validating Thermal Scattering Laws with Pulsed-Neutron Die-Away Diffusion Benchmarks

Unique advantages of pulsed-neutron die-away diffusion benchmarks

- Models are geometrically simple, and the only material present is the material being tested.
- The only neutron reactions are thermal scattering and absorption. In a simulation, there is no concern about nuclear data uncertainties from other materials or from other reactions.
- For small samples, eigenvalues are highly sensitive to details of both integral and differential thermal scattering cross sections, allowing clear qualification of TSL physics models.
- PNDA collectively probes the physics of the entire thermal spectrum of integral and differential cross sections in one sensitive quantity.
- Measured eigenvalues in well-designed experiments have uncertainties in the neighborhood of 0.1% to 0.5%.
- Samples can be easily heated or cooled to study thermal scattering temperature dependence.
- PNDA requires much less material than critical benchmarks, is much less expensive, and can be easily "tuned" by buckling to vary absorption and thermal scattering sensitivity.

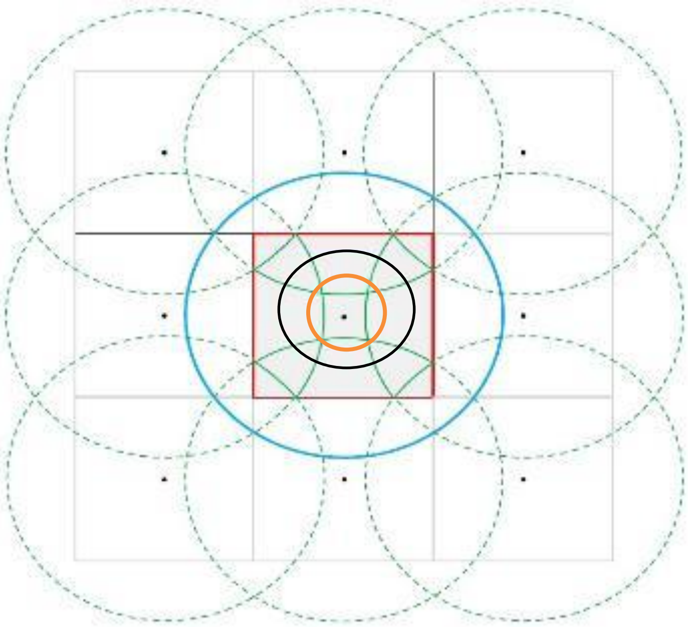
Aluminum Dispersions Relations and Total Phonon Spectrum from MedeA VASP/PHONON



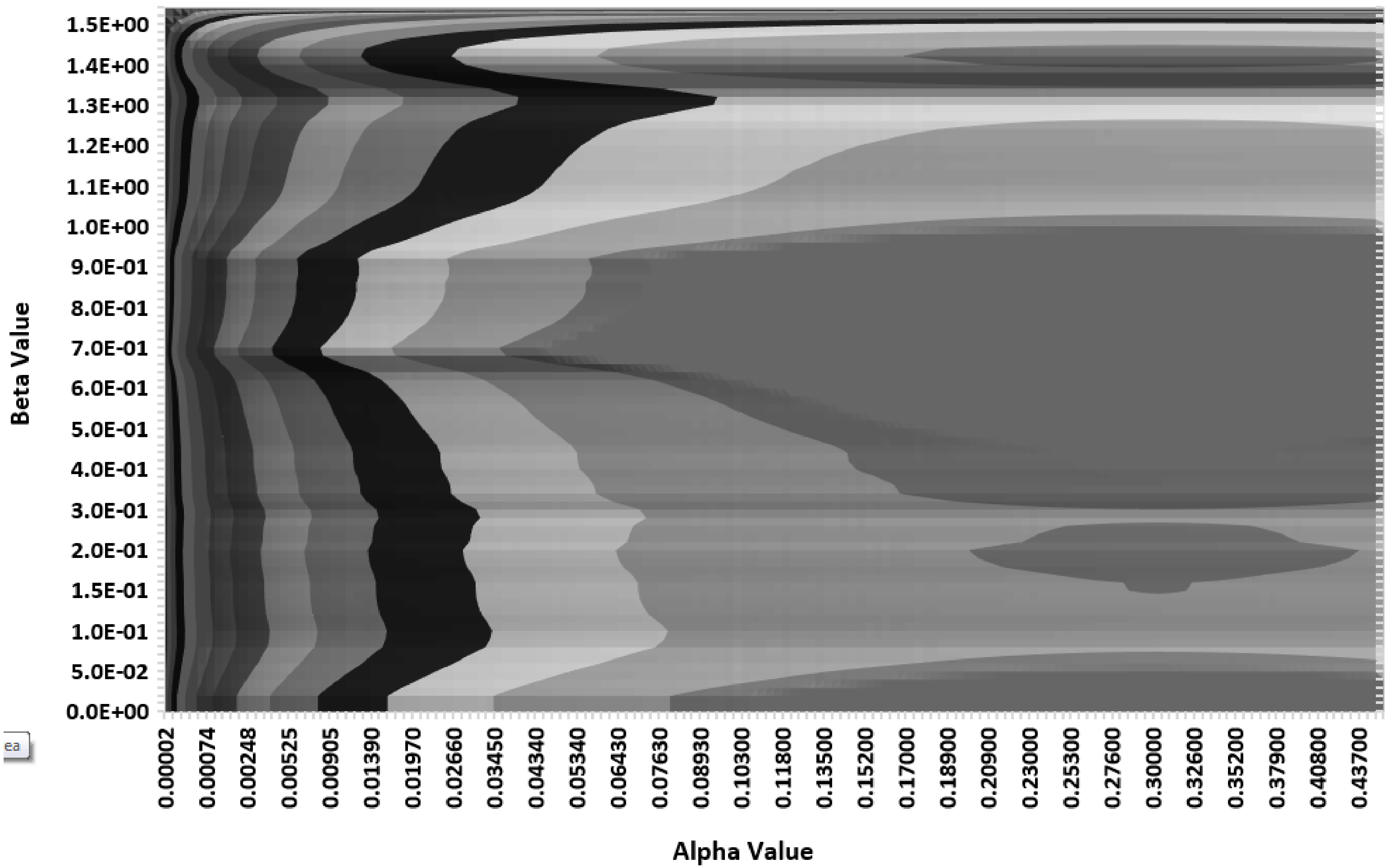
- In the incoherent approximation, all k-points (or vectors from gamma) in the 1st Brillouin zone are sampled randomly sampled and the associated frequencies are collected in bins to give the total phonon spectrum.
- The assumption is made that for any *α* (equivalent to a k-point vector magnitude), the accessible phonon frequencies for one-phonon scattering are determined only by the total phonon spectrum.
- **Physically**, the available phonon frequencies for one-phonon scattering at particular *α* depend on the isofrequency surfaces that intersect spherical *α*-radius shells centered on the gamma-point.

Illustrative Schematic of a Simple 2-D First Brillouin Zone (BZ)

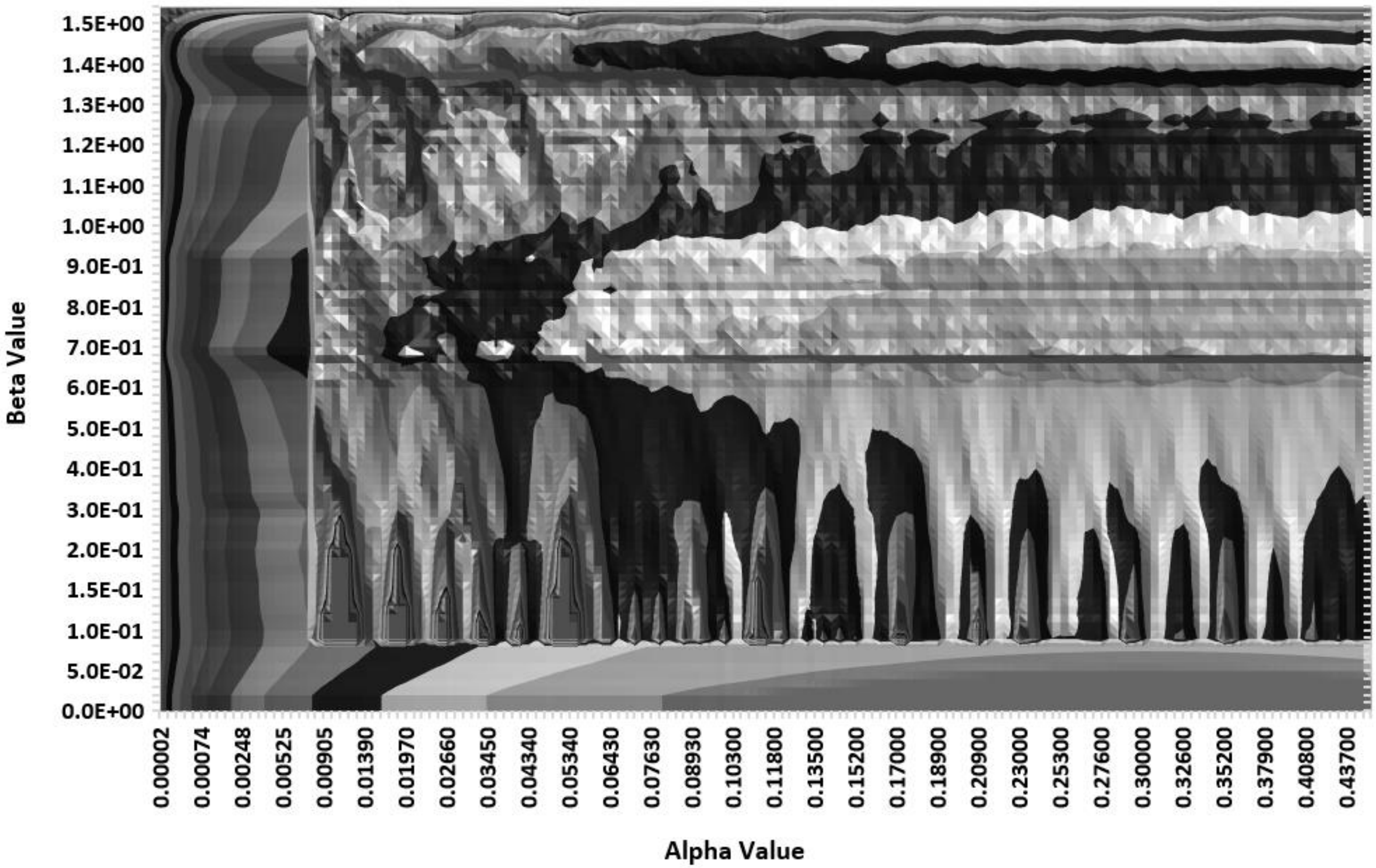
- The red box represents the first BZ. Neighboring gray boxes represent neighboring BZs.
- Only phonon modes defined at k-points in the first BZ are physically unique. Phonon modes defined at k-points in neighboring BZs (by translation in k-space by integer multiples of lattice constants) are identical to their first BZ analogs at symmetric k-points.
- For an α value given by the radius of the yellow or black circle, there is no coherent interference due to the lattice.
- The green circles represent lattice-translated blue circles, upon which the phonon modes will be identical to the blue circle.
- For an α value given by the blue circle, portions of the green circles lying within the red box represent symmetrically equivalent k-point locations within the 1st BZ. This is **coherent interference.**
- In 3-D, for an α-shell lying outside the 1st BZ, the equivalent α-shell in the 1st BZ will have a non-constant k-radius.



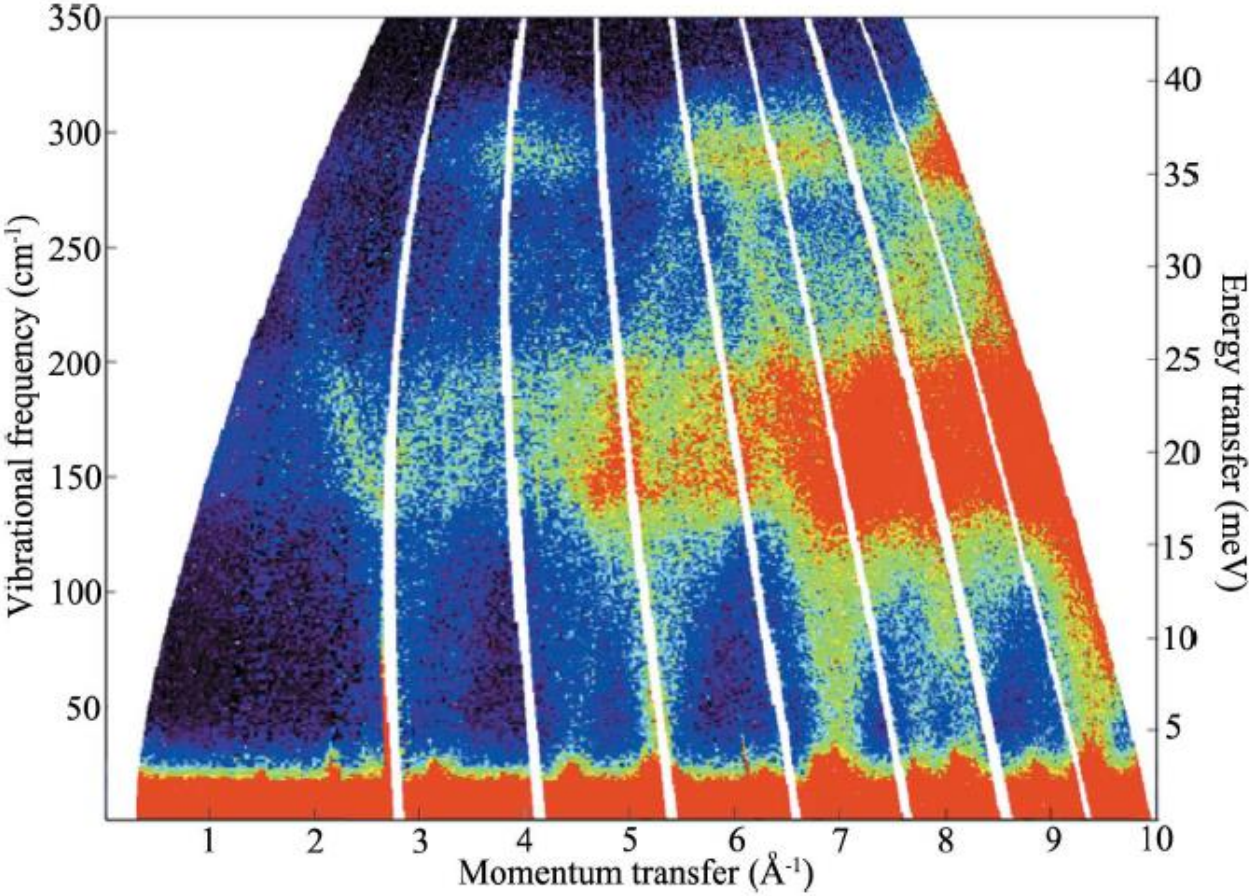
One-Phonon S(α,β) for Aluminum in Incoherent Approximation (calculated with MedeA and FLASSH)



One-Phonon S(α,β) for Aluminum with Coherent Interference (calculated with MedeA and FLASSH)



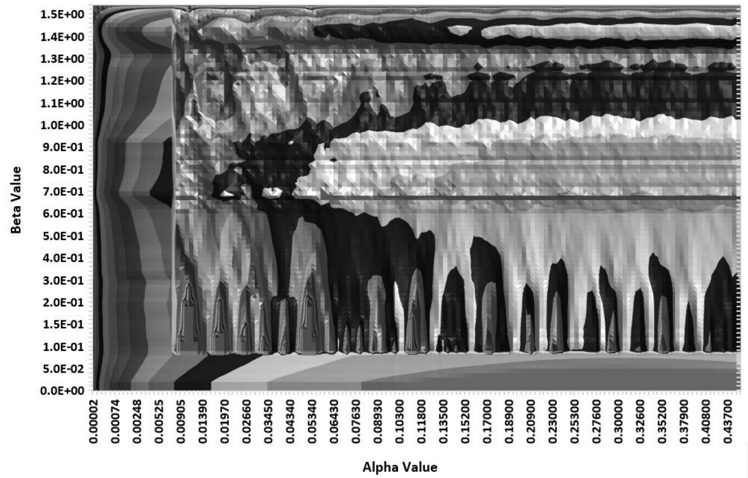
Experimental One-Phonon S(α,β) for Aluminum with Coherent Interference



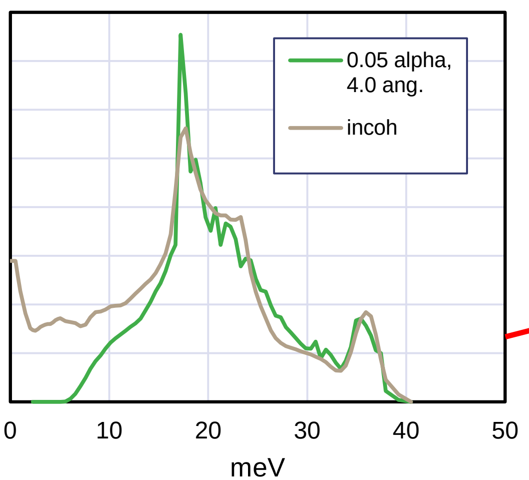
Experimental data from: Roach et al., J. Appl. Cryst. (2003)

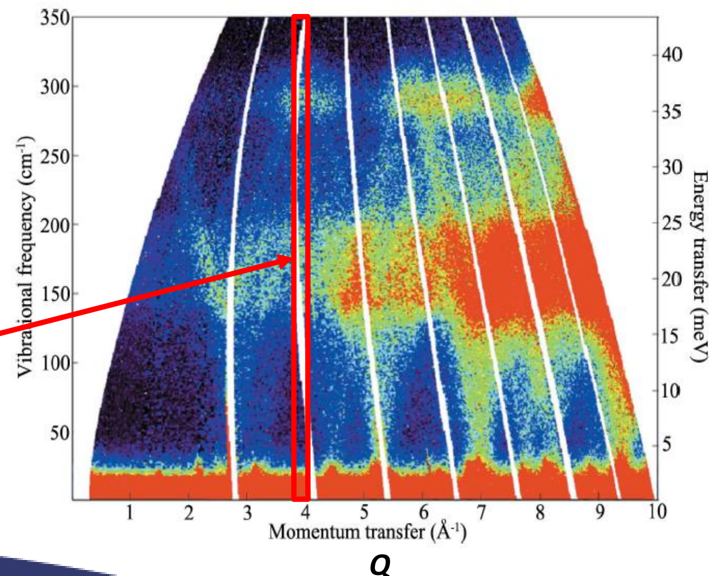
One-Phonon S(α,β) for Aluminum Comparisons

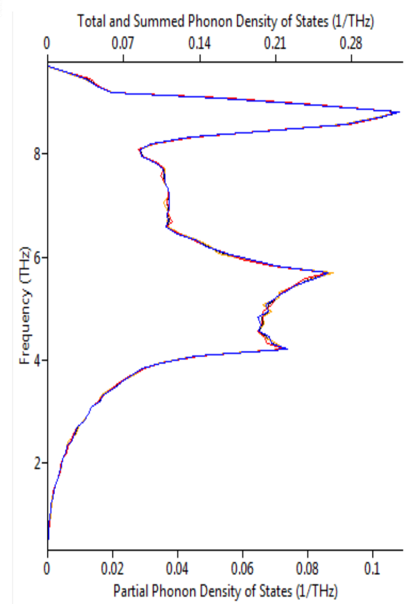
$$\alpha = \frac{\hbar^2 Q^2}{2 M k_B T}$$



One-phonon S(α,β) at α = 0.05 with and without coherent interference correction







Experimental data from: Roach et al., J. Appl. Cryst. (2003)

Evaluation Considerations for the Future

TSL evaluations developed using modern atomistic simulation methods (DFT, MD) and advanced codes outside of NJOY will become more common. As this capability progresses, several considerations should be addressed.

- For advanced TSL evaluation methods involving atomistic simulations and multiple types of experimental data:

  (atomic energy potentials, crystallographic and thermodynamic information, dispersion relations, differential and double-differential scattering data, PNDA data, etc.),

  The proper determination of covariances may be very difficult and is certainly not standardized.

  However, the evaluation process should not be tailored or degraded for the sake of simplifying covariance calculations. It is more important to have rigorously evaluated data than rigorously evaluated covariances.

- With the introduction of coherent one-phonon interference effects, $S(\alpha,\beta)$ will no longer be smooth as a function of α. This could impact File 7 post-processing methods and normalization checks.
- Recommend developing pulsed-neutron die-away diffusion benchmarks as an inexpensive and more sensitive method of validating the physics of TSL evaluations.
- Multiple types of experimental data, when available, should be used to qualify TSLs. In many cases, integral critical benchmarks may be the most obfuscated experimental method of qualification.

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