Naval Nuclear Laboratory Thermal Neutron Scattering Investigations and Perspectives

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Outline of Topics

- Temperature-dependent cross section behavior of ENDF/B-VIII.0(β4) H-H_2O, sensitivity to phonon spectra, and benchmark testing.
- Impact of NJOY procedures and modifications with respect to thermal cross section calculations.
- Quality-assurance checks requested by the US CSEWG and format issues for thermal scattering libraries.
- Validating thermal scattering laws with pulsed-neutron die-away diffusion benchmarks.
- Future evaluation strategies and concerns.

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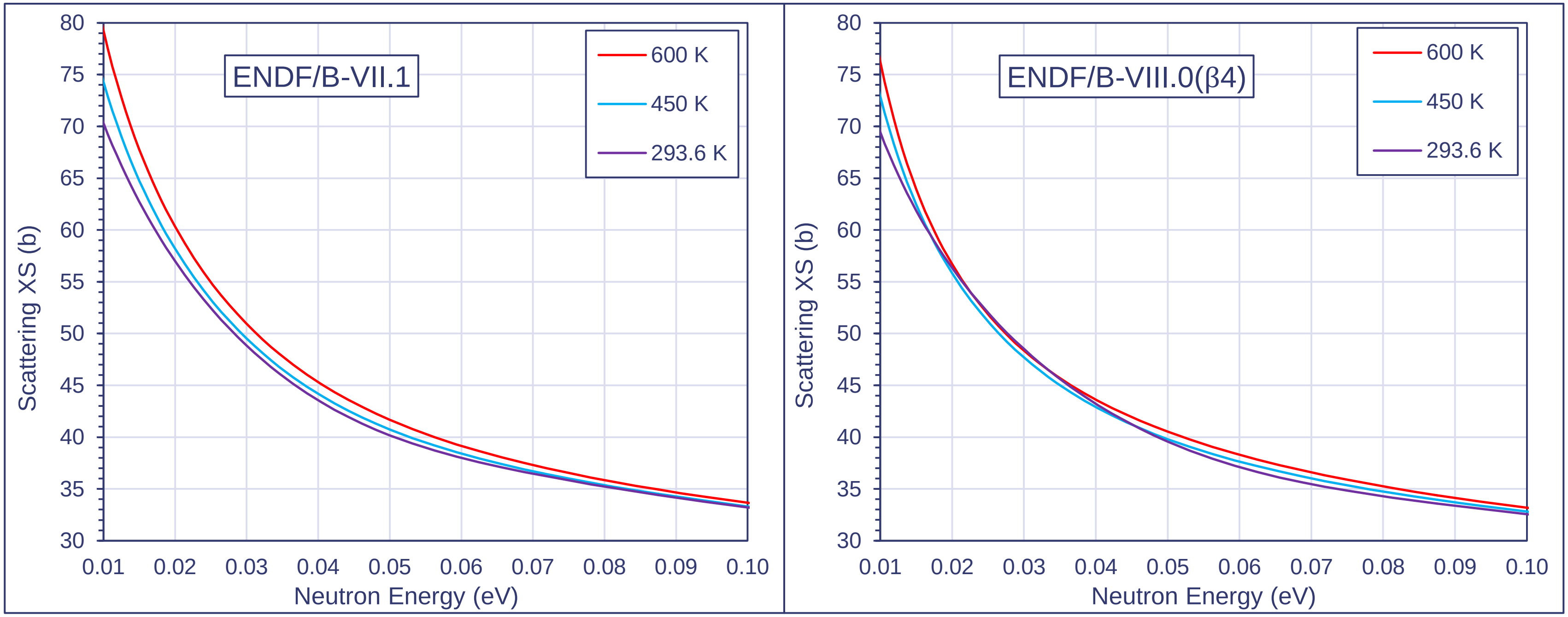
H-H_2O ENDF/B-VII.1 and ENDF/B-VIII.0(β4) Intermediate-Energy Thermal Scattering Cross Section Comparison



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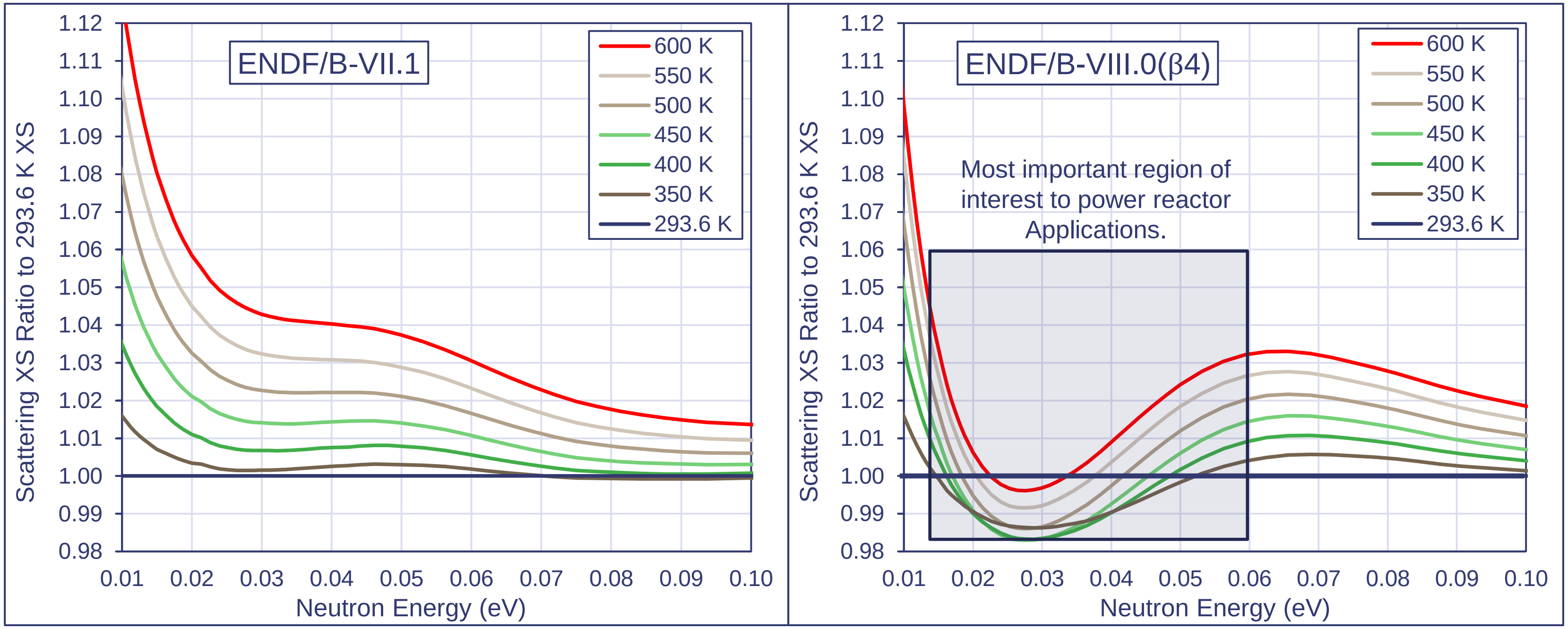
H-H_2O ENDF/B-VII.1 and ENDF/B-VIII.0(β4) Temperature-Dependence of Intermediate-Energy Thermal Scattering Cross Section Ratios to 293.6 K Cross Sections



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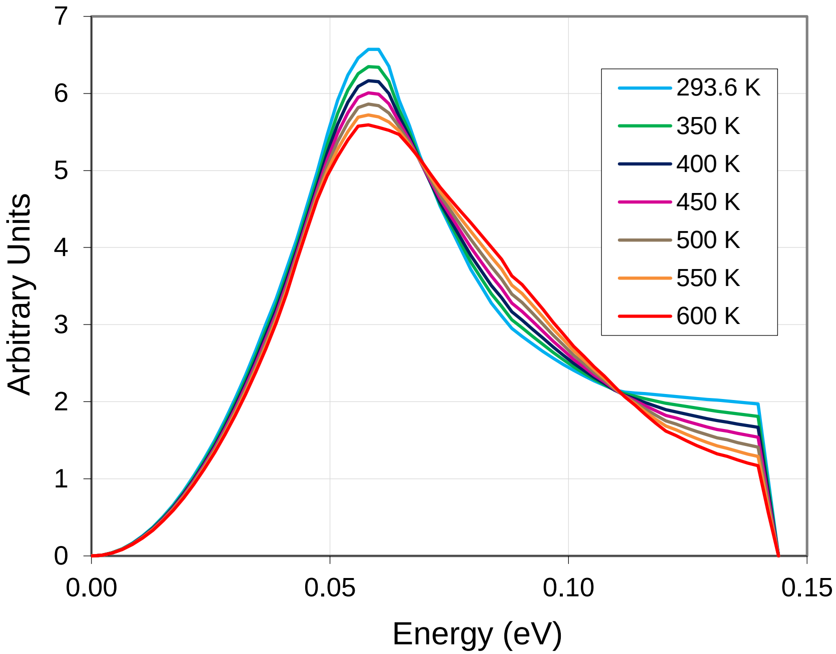
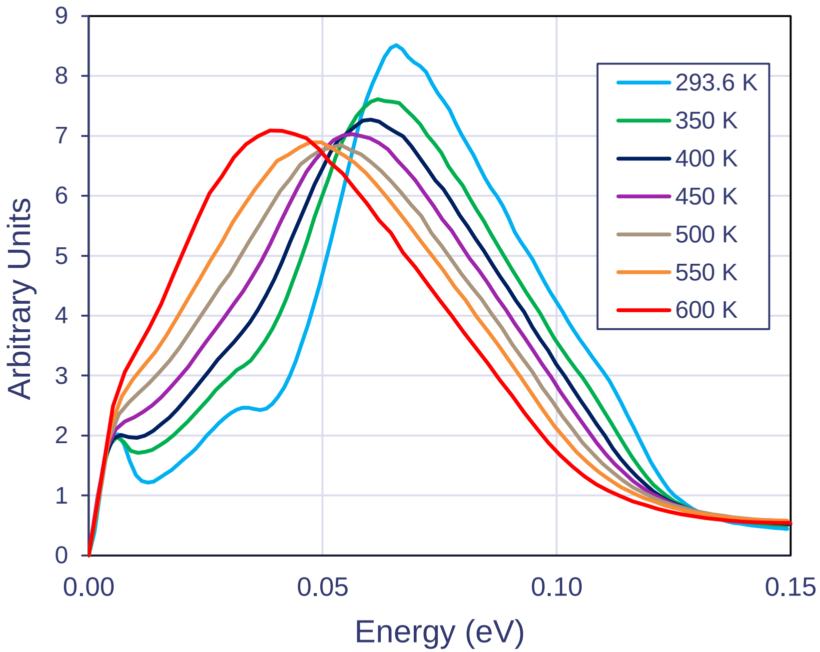
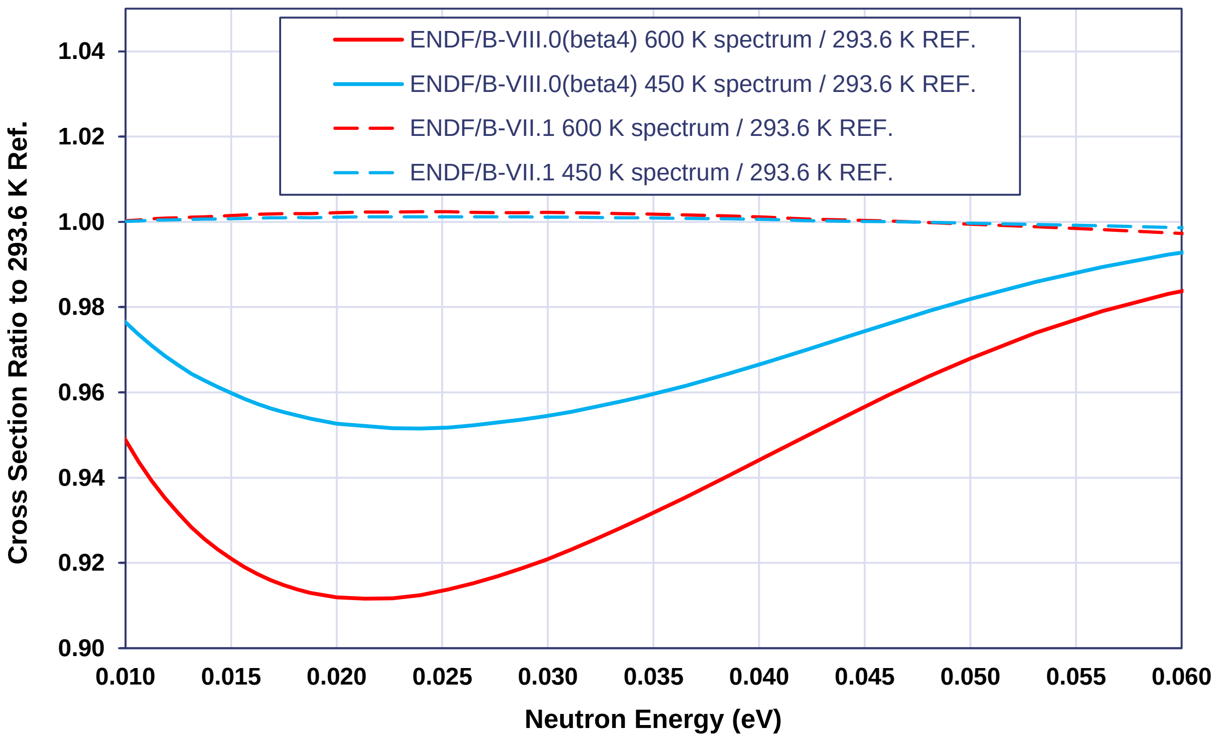
ENDF/B-VII.1 vs. ENDF/B-VIII.0(β4) Temperature-Dependent $H-H_2O$ Free/Hindered Rotational Phonon Mode Density



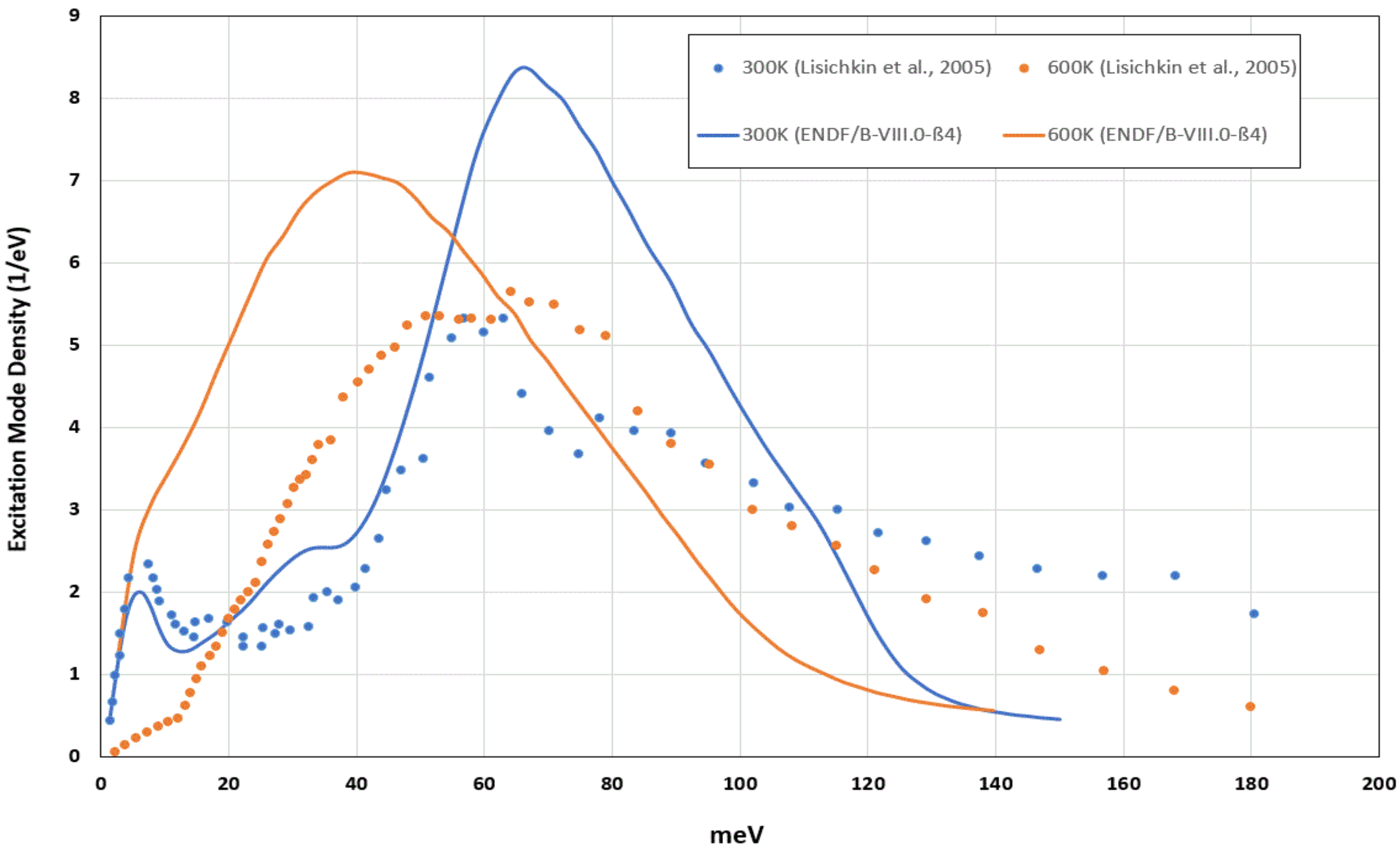


H-H_2O ENDF/B-VII.1 and ENDF/B-VIII.0(β4) Cross Section Ratio to 293.6 K Reference Changing Only Temperature-Dependent Phonon Spectra



Integral scattering cross sections for ENDF/B-VIII.0(β4) are greater than 20× more sensitive to changes in the phonon spectrum with respect to temperature than ENDF/B-VII.1.

ENDF/B-VIII.0(β4) H-H_2O Phonon Mode Density Compared to Lisichkin et al. (2005), No Renormalization



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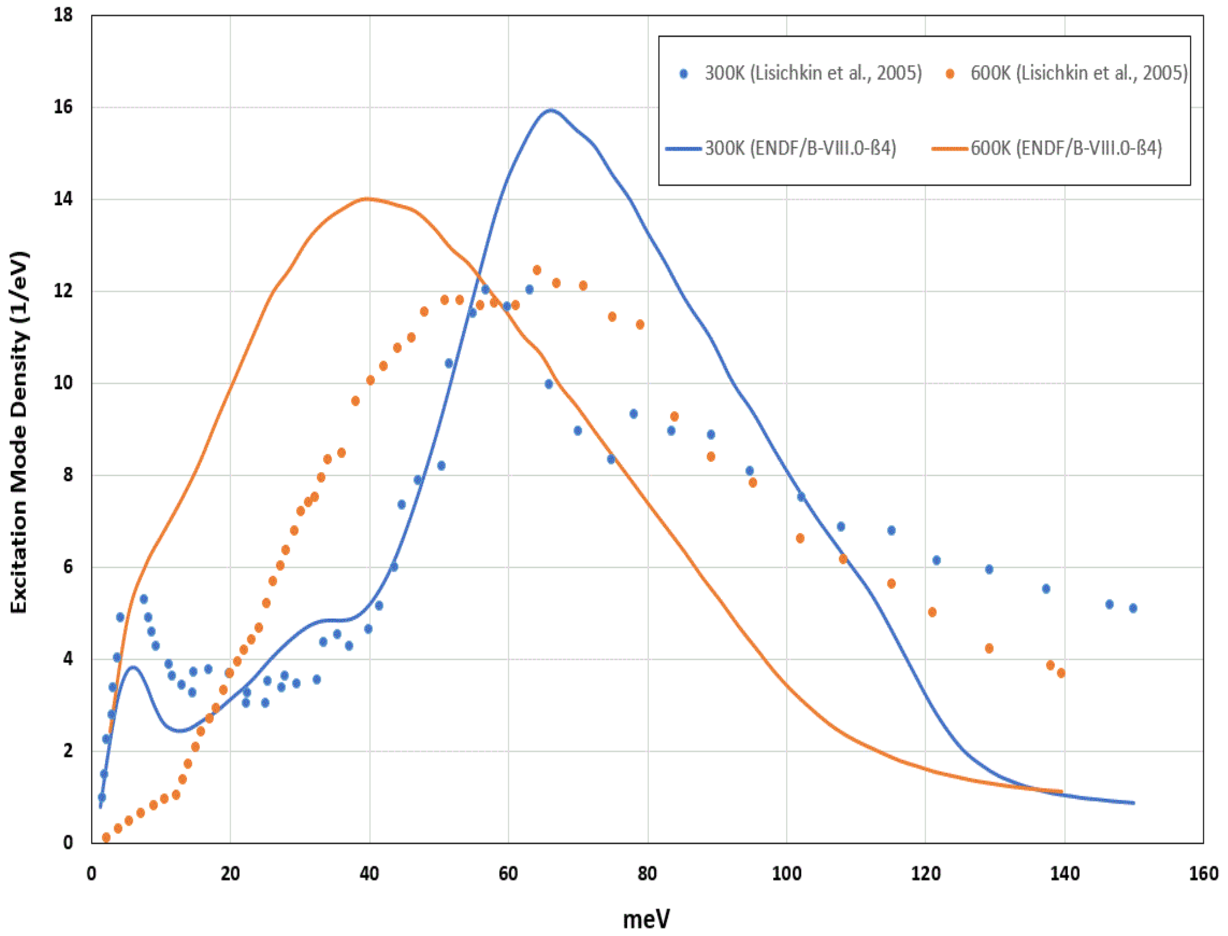
ENDF/B-VIII.0(β4) H-H_2O Phonon Mode Density Compared to Lisichkin et al. (2005), Unity Normalization



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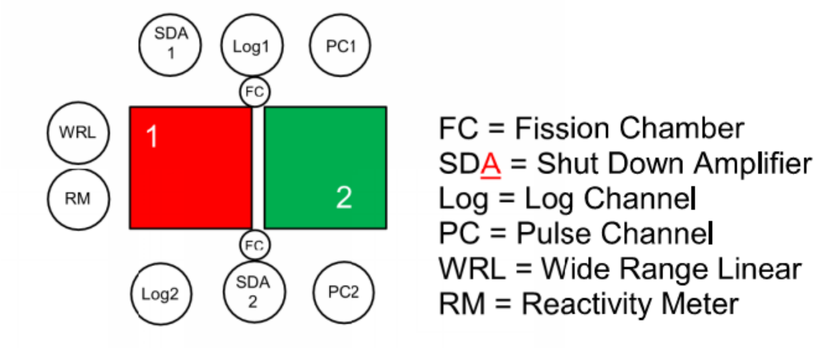
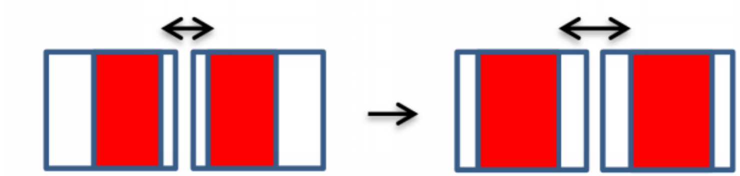
Validation of ENDF/B-VIII.0(β4) $H\text{-}H_2O$ at Elevated Temperatures with Neptune

- In 2014, NNL and Rolls-Royce collaborated on a series of critical experiments at the Neptune facility to validate our ability to predict criticality for water-isolated arrays as function of temperature.
- Configurations were neutronically similar to spent fuel storage racks without poison inserts in flux trap.
- Test was specifically designed to assess criticality safety issues for spent fuel rack configurations with water gaps.
- In this configuration, undermoderated fuel assemblies can have a positive temperature coefficient of reactivity.
- Water temperature varied from 20-60 °C

Schematic of Core and Detector Arrangement



Schematic of Fuel Arrangement Showing Increase in Effective Water Gap



Ref.: S. Walley et al., "Measurement of Positive Temperature Coefficients of Reactivity for Rack-like Arrangements of Reactor Fuel in the Neptune Zero Energy Facility," Proc. RRFM-2016, Berlin, March 13-17, 2016.

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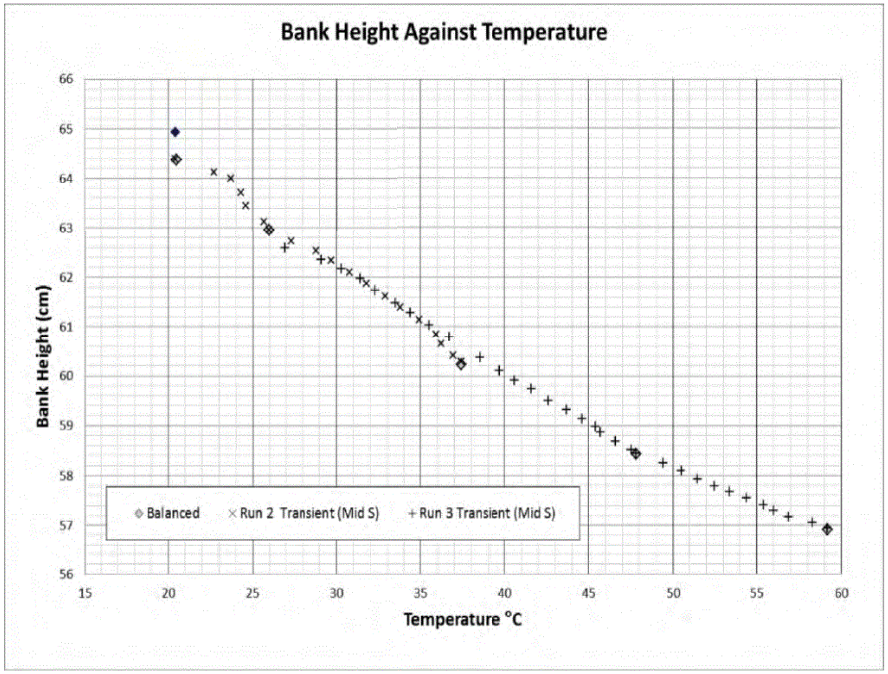
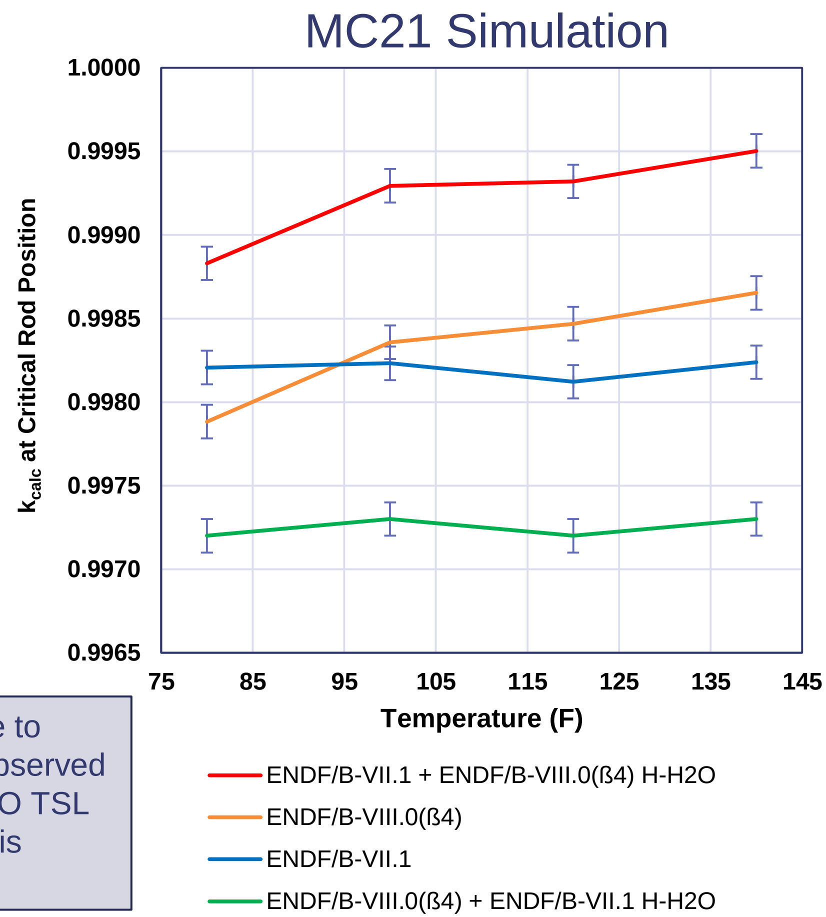
Neptune Configuration C Response to ENDF/B-VIII.0(β4) $H\text{-}H_2O$ Thermal Scattering Cross Sections vs. Temperature





This Neptune experiment is specifically designed to be sensitive to thermal scattering in water as a function of temperature. The observed reactivity bias is only present when the ENDF/B-VIII.0(β4) $H\text{-}H_2O$ TSL is used. Neither ENDF/B-VII.1 nor ENDF/B-VIII.0(β4) exhibit this behavior when the ENDF/B-VII.1 $H\text{-}H_2O$ TSL is used.

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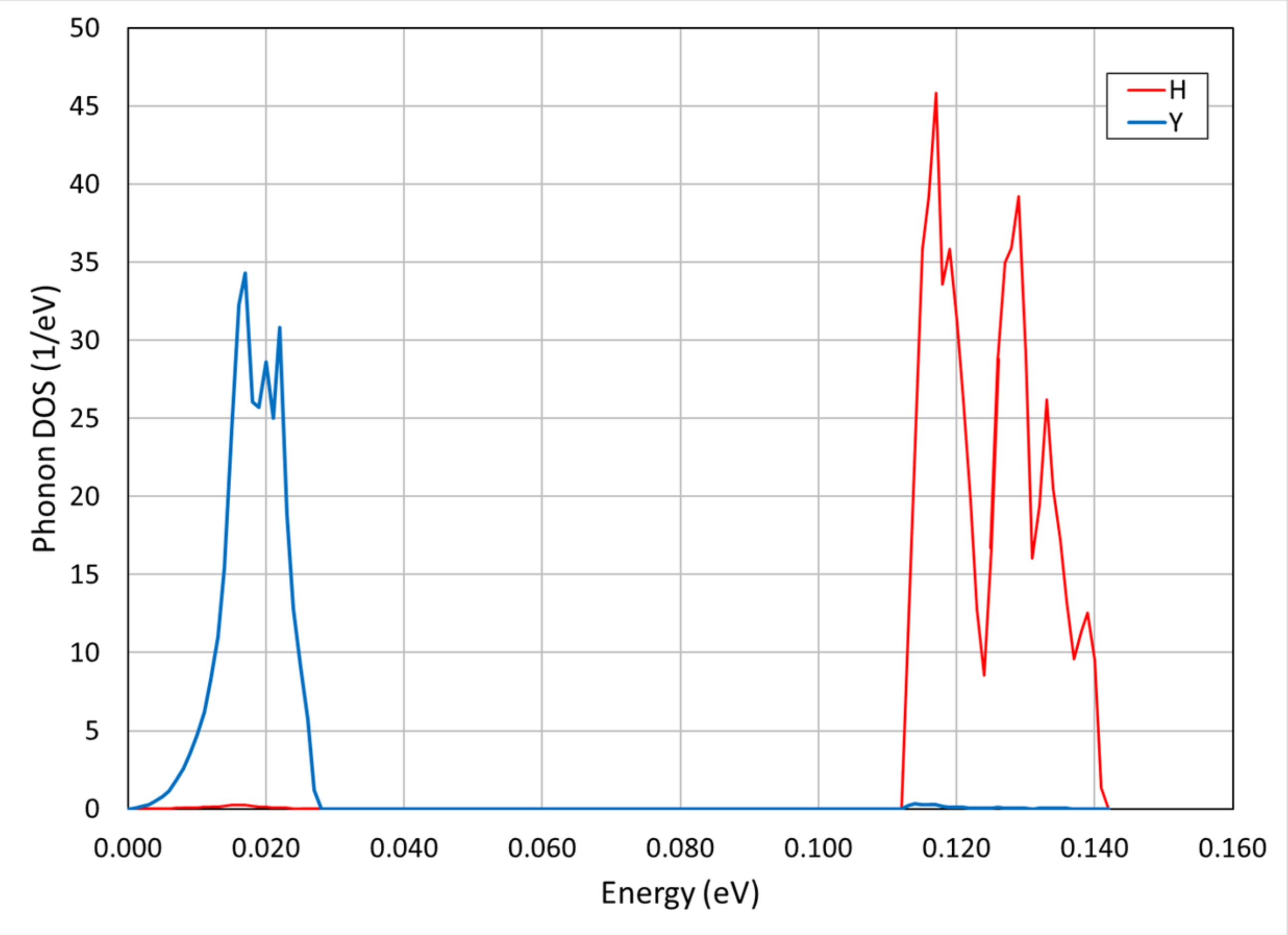
YH_2 Phonon Density of States Calculated with Density Functional Theory and Harmonic Lattice Dynamics



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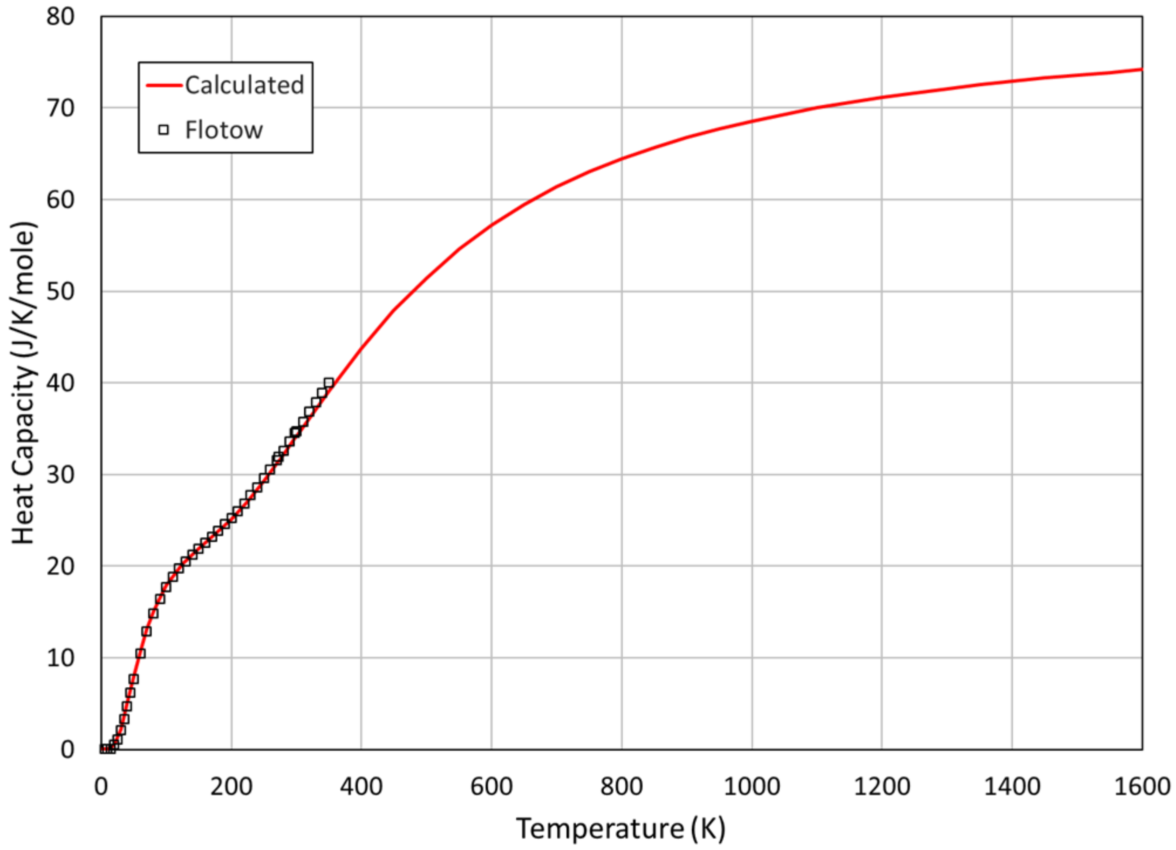
Heat Capacity Based on YH_2 Phonon Density of States Compared to Flotow et al. (1962) Experimental Data



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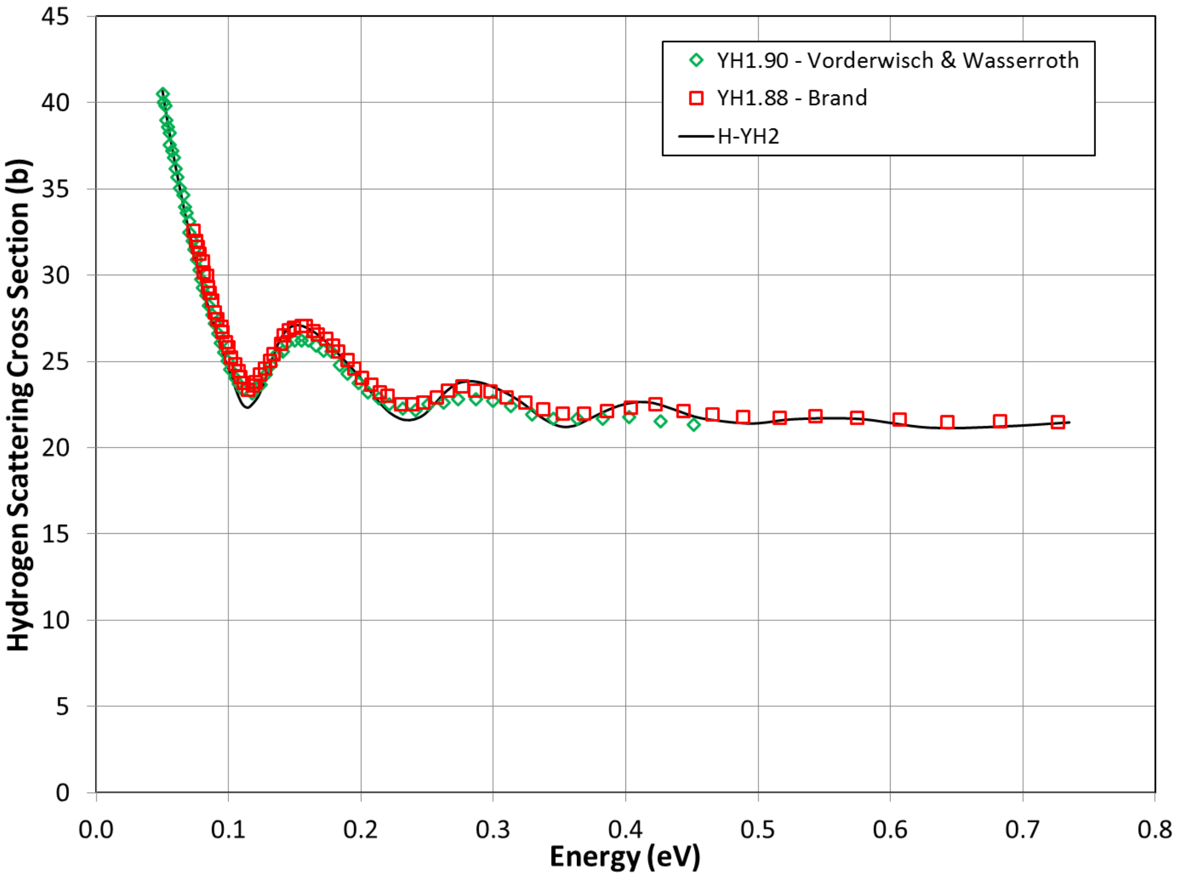
H-YH_2 Total Scattering Cross Section Compared to Vorderwisch et al. (1969) Experimental Data



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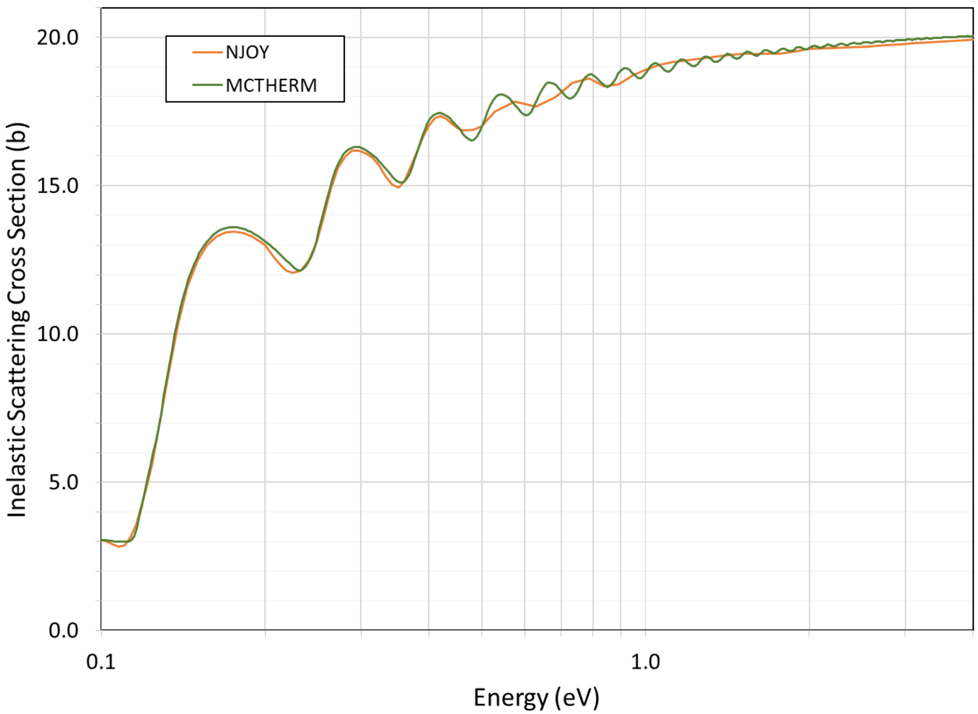
THERMR Metal Hydride Processing Issue (H-YH_2)



- The above plot was calculated with NJOY2012.50, with adaptive gridding used by THERMR which does not capture oscillations.
- Calculation with NJOY2012.82, using a fixed energy grid, produced similar results failing to capture oscillations.
- MCTHERM is a thermal scattering test code used internally by NNL.

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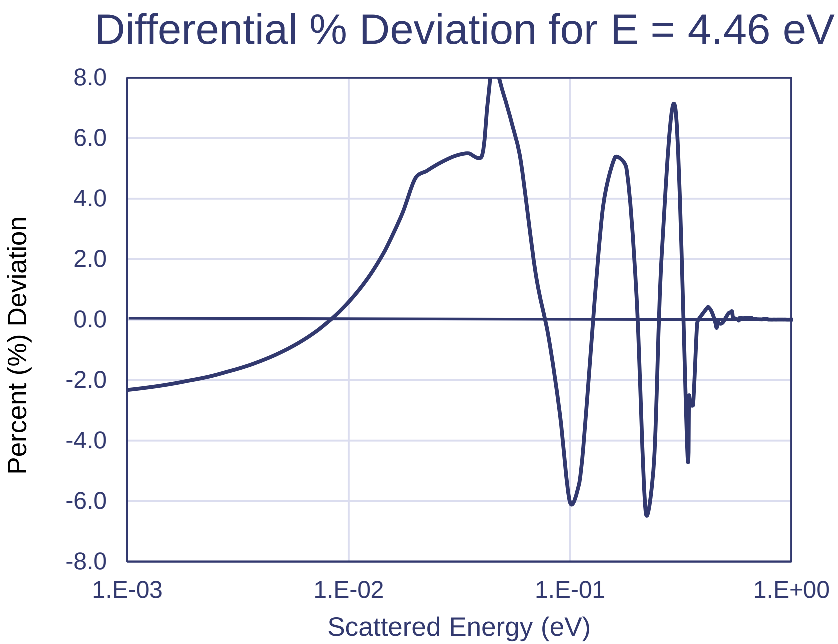
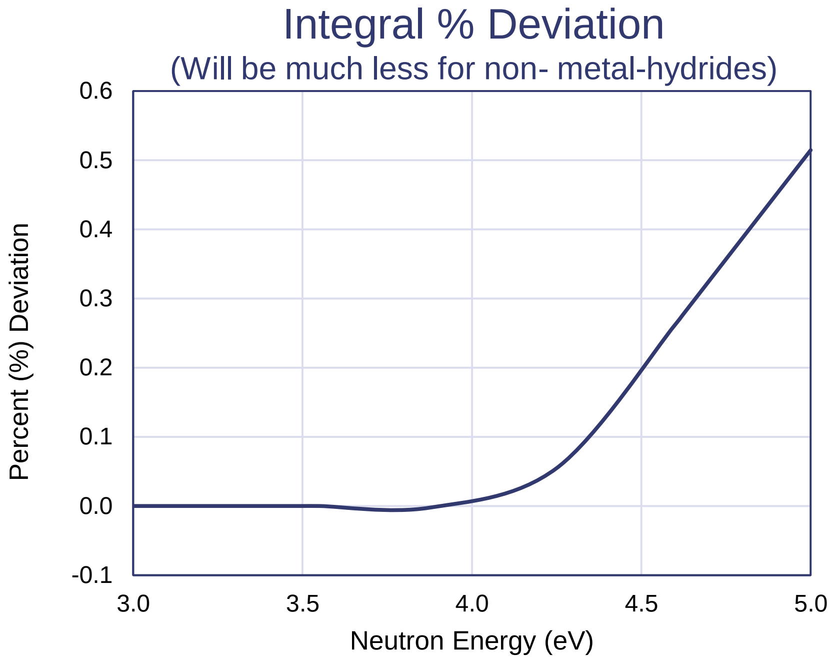
Percent Differential and Integral Cross Section Deviation for H-YH_2 at 293.6 K Due to Implementation of a 2×10^{-38} Cutoff for $S(\alpha,\beta)$ in LEAPR





- At 293.6 K, for energy transfers > 4.2 eV, or $\beta > 166$, all $S(\alpha,\beta)$ information is truncated and the SCT is used (which can overpredict integral cross sections).
- Partial truncation begins for much lower β and progressively increases with β.
- The proposed $S(\alpha,\beta)$ cutoff also adversely affects resolving higher-energy integral oscillations for metal-hydrides with large phonon band gaps.

Consistency of the Free Scattering Cross Section at the Thermal/Fast Energy Region Interface

$$\frac{d^2\sigma(E)}{d\alpha d\beta} = \sigma_{\text{bound}} * F[S(\alpha,\beta)]$$

$$\sigma_{\text{bound}} = 4\pi|b_{\text{coherent}}|^2 + 4\pi|b_{\text{incoherent}}|^2$$

b ≡ neutron scattering length (complex valued)

$$\sigma_{\text{free}} = \sigma_{\text{bound}} * \left[\frac{A^2}{(A+1)^2}\right]$$

- For commonly occurring nuclides, *b* is typically very accurately measured. Therefore, σ_{bound} is typically very well known.
- The uncertainty in *A* is exceedingly small. Therefore, σ_{free} is also typically very well known.

  EXAMPLES of relative uncertainty in σ_{bound} from Sears, *Neutron News*, **3** (1992):
  ^{1}H (0.073%), ^{9}Be (0.26%), ^{12}C (0.054%), ^{16}O (0.14%), ^{27}Al (0.27%), ^{238}U (0.12%)
- The elastic thermal scattering cross section determined from MF3/MT2 data (with RECONR/BROADR) commonly has an uncertainty 2 to 20 × greater than for σ_{free}. The two different cross sections are evaluated in completely different ways.
- Any adjustment of σ_{free} in File 7 thermal libraries – the B(1) field of MF7/MT4 – will change ALL calculated differential and integral inelastic thermal cross sections!
- Any impact on criticality of a scattering cross section discontinuity between the thermal and fast energy regions will likely be much less than due to adjusting all thermal cross sections.

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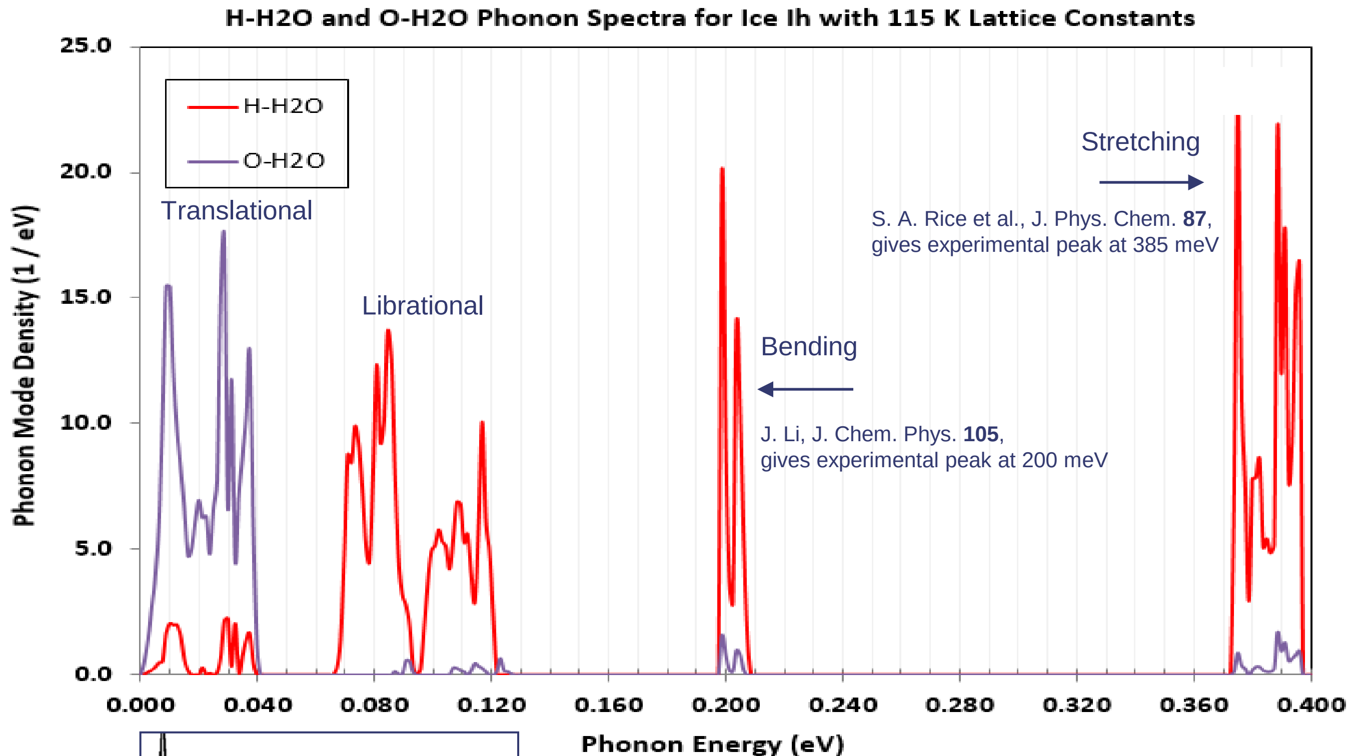
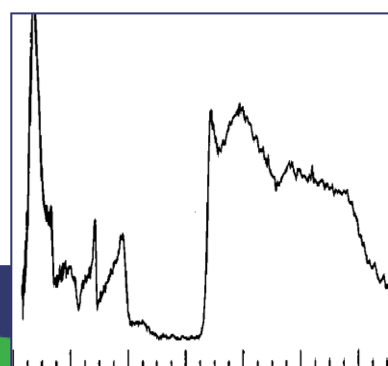
Calculated Phonon Spectrum for Ice I*h* with Density Functional Theory and Harmonic Lattice Dynamics

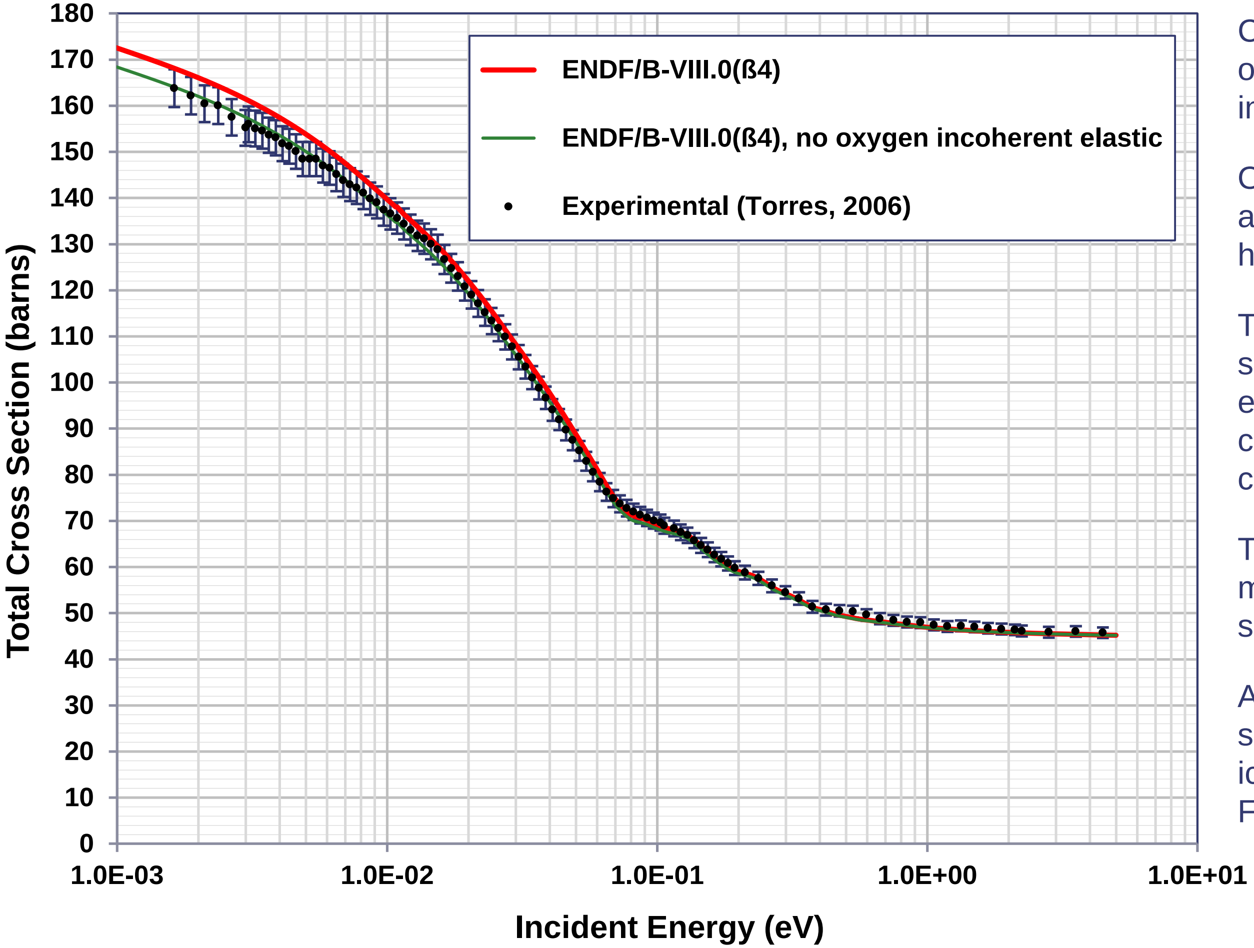




Experimental inelastic neutron scattering spectrum for polycrystalline ice I*h* (same energy scale as above).

J. Li, "Inelastic Neutron Scattering Studies of Hydrogen Bonding in Ices," *Journal of Chemical Physics* **105**, 6733-6755 (1996).

Ice I*h* Total Cross Sections



Coherent elastic scattering on oxygen was estimated in the incoherent approximation.

Oxygen scattering cross sections are very small compared to hydrogen.

The incoherent approximation somewhat overestimates oxygen elastic scattering (which is actually coherent), and below the Bragg cutoff it is zero.

The green curve gives a bounding minimum without oxygen elastic scattering.

An evaluation of coherent elastic scattering on the oxygen lattice for ice I*h* will be performed with FLASSH.

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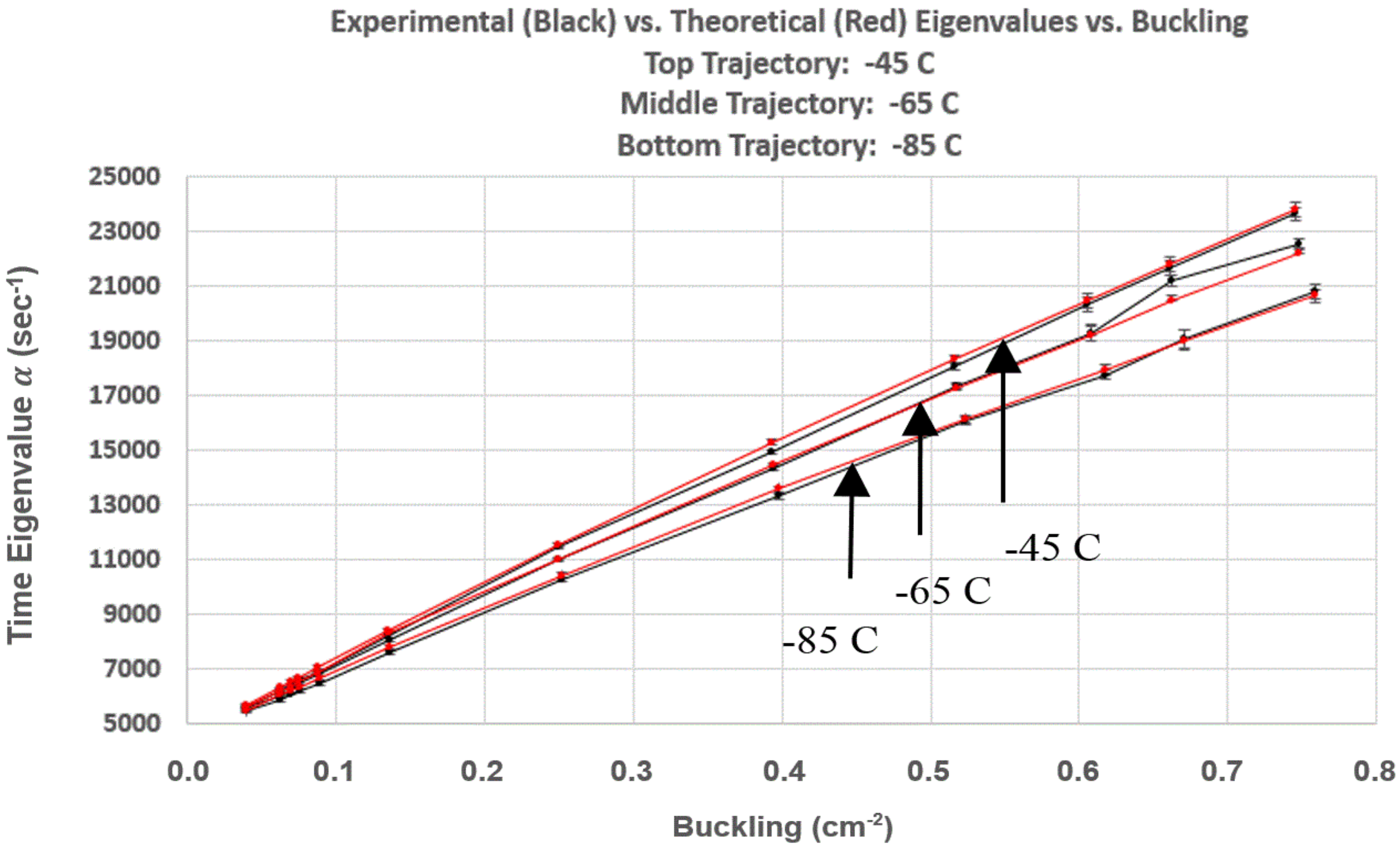
Validating Thermal Scattering Laws with Pulsed-Neutron Die-Away Diffusion Benchmarks

- A neutron generator (D+D or D+T) is used to target a pure material sample with a short mono-energetic neutron pulse.
- 10^{-4} to 10^{-3} seconds following the pulse, the neutron population is in thermal equilibrium in a fundamental spatial mode with a characteristic flux decay time eigenvalue α.

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

- 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.
- The eigenvalue is physically determined by neutron diffusion through the material.
- **Advantages of pulsed-neutron die-away diffusion benchmarks:**
  - The only reactions neutrons undergo are thermal scattering, absorption, and leakage.
  - In a simulation, there is no concern about nuclear data uncertainties from other materials.
  - Models are geometrically simple, and the only material present is the material being tested.
  - Eigenvalue sensitivity to thermal scattering is a strong function of geometric buckling. Absorption dominates in large samples. Thermal scattering dominates in small samples.
  - Diffusion is sensitive to both integral and differential scattering cross sections.
  - Samples can be easily heated or cooled to study thermal scattering temperature dependence.
  - They require much less material than critical benchmarks and are much less expensive!

MC21 results with ENDF/B-VIII.0(β4) Ice I*h* TSLs Compared to Silver (1968) Pulsed-Neutron Die-Away Flux Decay Measurements for Ice Cylinders



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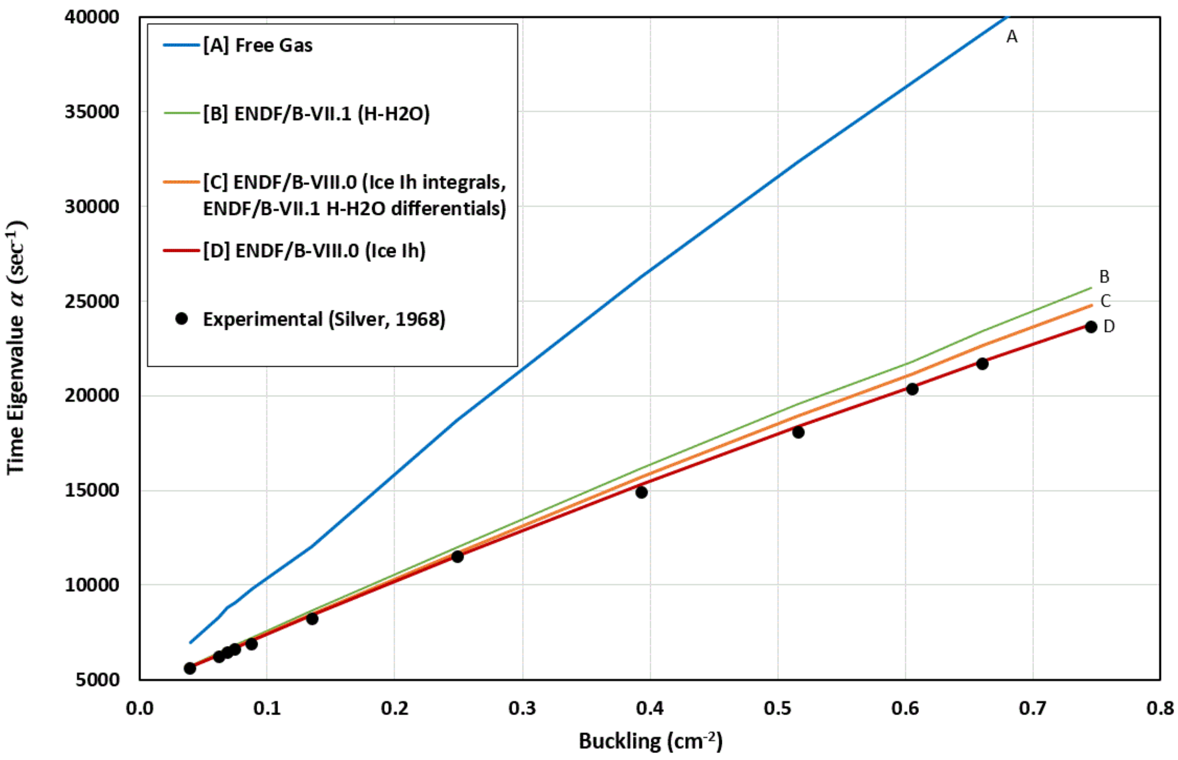
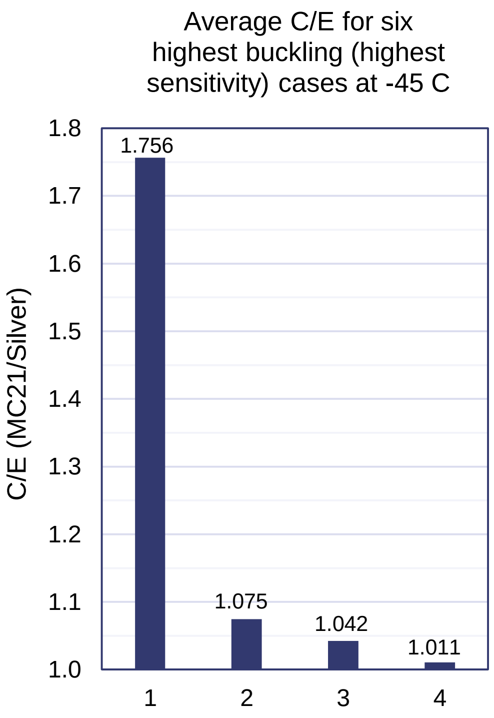
MC21 Test for Sensitivity to Integral and Differential Cross Sections for Ice I*h* Diffusion Benchmark at -45 C





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

- In the one-speed diffusion model, the flux decay of the fundamental spatial mode can be expressed as:

$$\varphi(\mathbf{r},t) = \varphi_0(\mathbf{r}) \exp[-(v\Sigma_a + vDB^2 - CB^4)t]$$

$$\text{where } \alpha = v\Sigma_a + vDB^2 - CB^4$$

Σ_a is the macroscopic absorption cross section,
$D = 1/\Sigma_{transport}$ is the diffusion coefficient,
v = the effective average neutron velocity,
B^2 = geometric buckling, and
C = the diffusion cooling parameter

- Due to the quadratic behavior of α with respect to B^2, the C/E bias in the $v\Sigma_a$ term can be determined by a quadratic fit over a range of B^2, verified for a large sample α where B^2 is nearly zero, and then subtracted out to improve theoretical α calculations for small samples.

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Ice I*h* Benchmark Results with MC21

SILVER EXPERIMENTAL									FINAL LIBRARY (-45 C)				FINAL LIBRARY (-65 C)				FINAL LIBRARY (-85 C)			
-45 C			-65 C			-85 C			MC21 α	σ	C/E	σ	MC21 α	σ	C/E	σ	MC21 α	σ	C/E	σ
B^2	α	σ	B^2	α	σ	B^2	α	σ												
0.0394	5630	52	0.0395	5579	59	0.0396	5443	35	5659	6	1.0051	0.009	5589	3	1.0018	0.009	5504	3	1.0112	0.007
0.0621	6207	38	0.0621	6031	35	0.0624	5890	61	6325	5	1.0191	0.006	6190	4	1.0264	0.006	6057	1	1.0284	0.011
0.0689	6413	66	0.0690	6266	22	0.0693	6096	53	6523	2	1.0172	0.010	6370	2	1.0166	0.010	6217	4	1.0199	0.009
0.0748	6584	70	0.0749	6455	38	0.0752	6226	66	6697	2	1.0171	0.011	6530	2	1.0117	0.011	6365	2	1.0223	0.011
0.0880	6888	49	0.0881	6825	76	0.0885	6493	72	7074	3	1.0270	0.007	6876	3	1.0075	0.007	6682	2	1.0290	0.011
0.1352	8214	70	0.1354	8052	73	0.1362	7593	72	8415	3	1.0244	0.009	8356	5	1.0377	0.009	7804	3	1.0278	0.010
									Low B^2 Avg. C/E		1.0183		Low B^2 Avg. C/E		1.0169		Low B^2 Avg. C/E		1.0231	
0.2490	11493	87	0.2494	10997	76	0.2514	10272	82	11546	12	1.0046	0.008	10985	10	0.9989	0.008	10427	8	1.0150	0.008
0.3933	14915	73	0.3941	14341	65	0.3981	13310	100	15302	56	1.0259	0.006	14458	43	1.0082	0.006	13598	33	1.0216	0.008
0.5161	18083	127	0.5174	17307	132	0.5233	16055	129	18350	59	1.0148	0.008	17259	45	0.9972	0.008	16158	47	1.0064	0.009
0.6058	20349	245	0.6078	19292	295	0.6172	17772	189	20482	52	1.0065	0.012	19232	39	0.9969	0.012	17952	30	1.0102	0.011
0.6606	21669	258	0.6624	21226	195	0.6710	19073	344	21818	104	1.0069	0.013	20449	77	0.9634	0.013	19033	57	0.9979	0.018
0.7460	23662	229	0.7482	22555	198	0.7586	20808	236	23795	105	1.0056	0.011	22199	77	0.9842	0.011	20663	57	0.9930	0.012
									High B^2 Avg. C/E		1.0107		High B^2 Avg. C/E		0.9915		High B^2 Avg. C/E		1.0074	

For the six highest buckling geometries (B^2 is in units of cm^{-2}), which are the most sensitive to the supplied thermal scattering kernel, the average C/E for *α* (units of sec^{-1}) across all three temperatures is 1.0032. When a quadratic C/E bias correction to the absorption term is performed for each temperature, the average *α* C/E is improved to 1.0001.

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Items for SG42 Review

- ENDF/B-VIII.0(β4) reverse cross section behavior at elevated temperatures in the energy region of highest flux for operating reactors warrants further testing. The 293.6 K cross sections are believed to be very good and the cross section dip at higher temperatures is believed to be physical. However, testing with the Neptune benchmark, designed to be sensitive to temperature-dependent changes in the thermal scattering kernel, indicates this cross section dip may be excessive. This assessment is consistent with observed experimental scattering cross section and inelastic neutron scattering measurements, and this library effect can have important safety consequences.

- Impact of NJOY modifications and procedures:
  - 2×10^{-38} cutoff for $S(\alpha,\beta)$ in LEAPR has an adverse impact on cross section accuracy for metal hydrides.
  - Fixed energy grid in THERMR is insufficient to resolve integral cross section details for metal hydrides. It is recommended that the user be able to supply a specific energy grid in the input deck if desired (this is especially important for experimental comparison), and that the default remains adaptive gridding.

- Quality-assurance checks requested by CSEWG and format rules for thermal scattering libraries.
  - Adjusting the free scattering cross section in MF7/MT4 will affect all thermal scattering cross sections. This quantity, when determined from neutron scattering length measurements, is often much more accurately known than elastic thermal cross sections calculated from MF3/MT2.
  - Calculation of thermal scattering cross sections up to 10 eV can be non-physical. This significantly exceeds the chemical binding energy of many atomic bonds. Care should be exercised.

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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.

- The ENDF File 7 format currently does not support the simultaneous tabulation of coherent and incoherent elastic scattering. This is an unnecessary format limitation that should be resolved.
- The MF7/MT4 B(6) field is used to identify the number of primary scatterers the B(1) free scattering cross section is associated with. Some TSLs set B(1) and B(6) to be consistent with chemical stoichiometry. This does not affect cross section calculations. However, future TSLs for oils, plastics, alloys, and composites with non-integer stoichiometry will require B(6) to always be '1'.
- 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, etc.), the proper determination of covariances may be very difficult. 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.
- Pulsed-neutron die-away diffusion and neutron slowing-down benchmarks offer an inexpensive and sensitive method of validating the performance of TSL evaluations without the presence of uncertainties and reactions from other materials.

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