Thermal Neutron Scattering Research and Development at Oak Ridge National Laboratory

Chris W. Chapman

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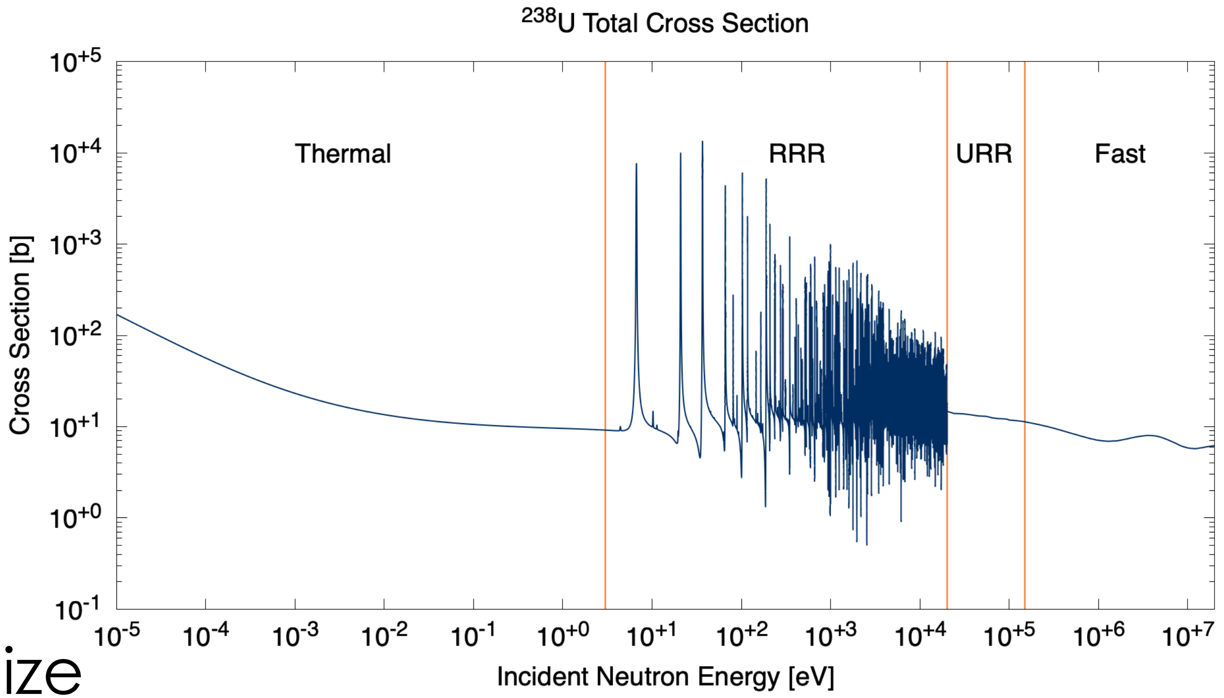
Outline

- Overview
- Experimental Capabilities
- Evaluations
  - Polyethylene
  - Yttrium Hydride
- Uncertainty Quantification
- Conclusions

Overview

- Neutron cross sections can be broken up into several regions
  - Thermal, resolved resonance, unresolved resonance, fast
- Thermal scattering is unique
  - Neutron wavelengths approach size of molecules and spacings in crystal lattices
  - Energies are on the order of excitation energy of the materials



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Overview

- Thermal neutron double differential scattering cross sections are defined by:

$$\frac{d^2\sigma}{dE_f d\Omega} = \frac{\sigma_b}{4\pi k_B T}\sqrt{\frac{E_f}{E_i}}\, e^{\frac{-\beta}{2}} S(\alpha,\beta)$$

$$\alpha = \frac{q^2\hbar^2}{2Mk_BT} \qquad \beta = \frac{-E}{k_BT}$$

- Thermal Scattering Law (TSL) $S(\alpha,\beta)$ related to dynamic structure factor $S(q,E)$

$$S(\alpha,\beta) = k_BTe^{\frac{-E}{2k_BT}}S(q,E)$$

Overview

- $S(q, E)$ broken up into 2 primary components:
  - Incoherent
    - Occurs when there is no interference between scattering neutron wavefunctions
    - More prevalent in hydrogenous materials
  - Coherent
    - Occurs when neutrons scatter with different nuclei and their wavefunctions interfere with one another
    - More prevalent in solids; specifically crystals
- ENDF currently stores coherent elastic (Bragg edges), incoherent elastic and incoherent inelastic

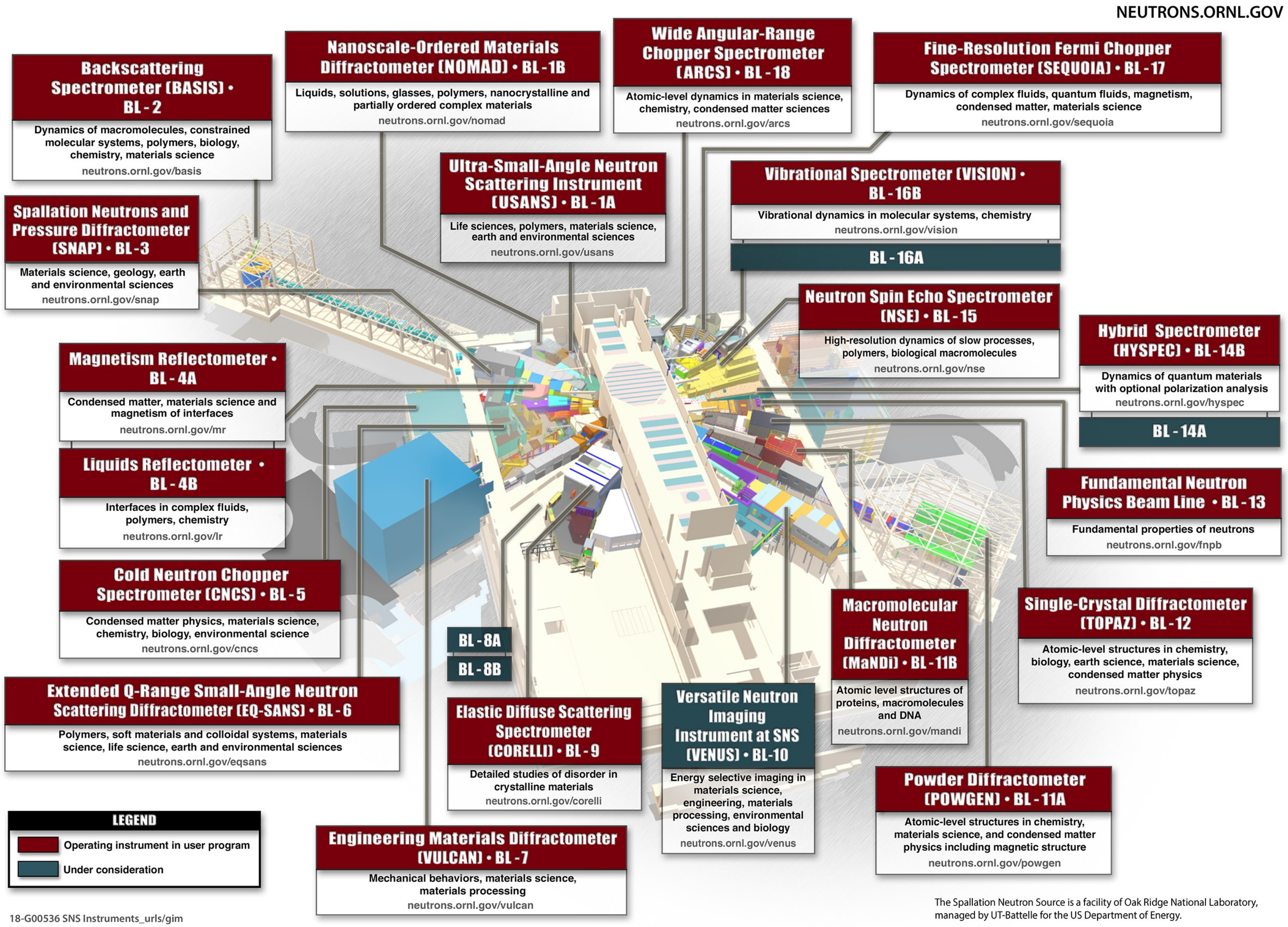
Overview

- $S(q, E)$ can be calculated several ways:
  - Density Functional Theory (DFT)
    - Simulate small cluster of atoms (~10s – 100s of atoms) in repeated structure
    - Atomic forces calculated from first principles by approximating the Schrödinger equation
  - Molecular Dynamics (MD)
    - Large cluster of atoms (thousands to millions of atoms)
    - Atomic forces calculated using classical potentials

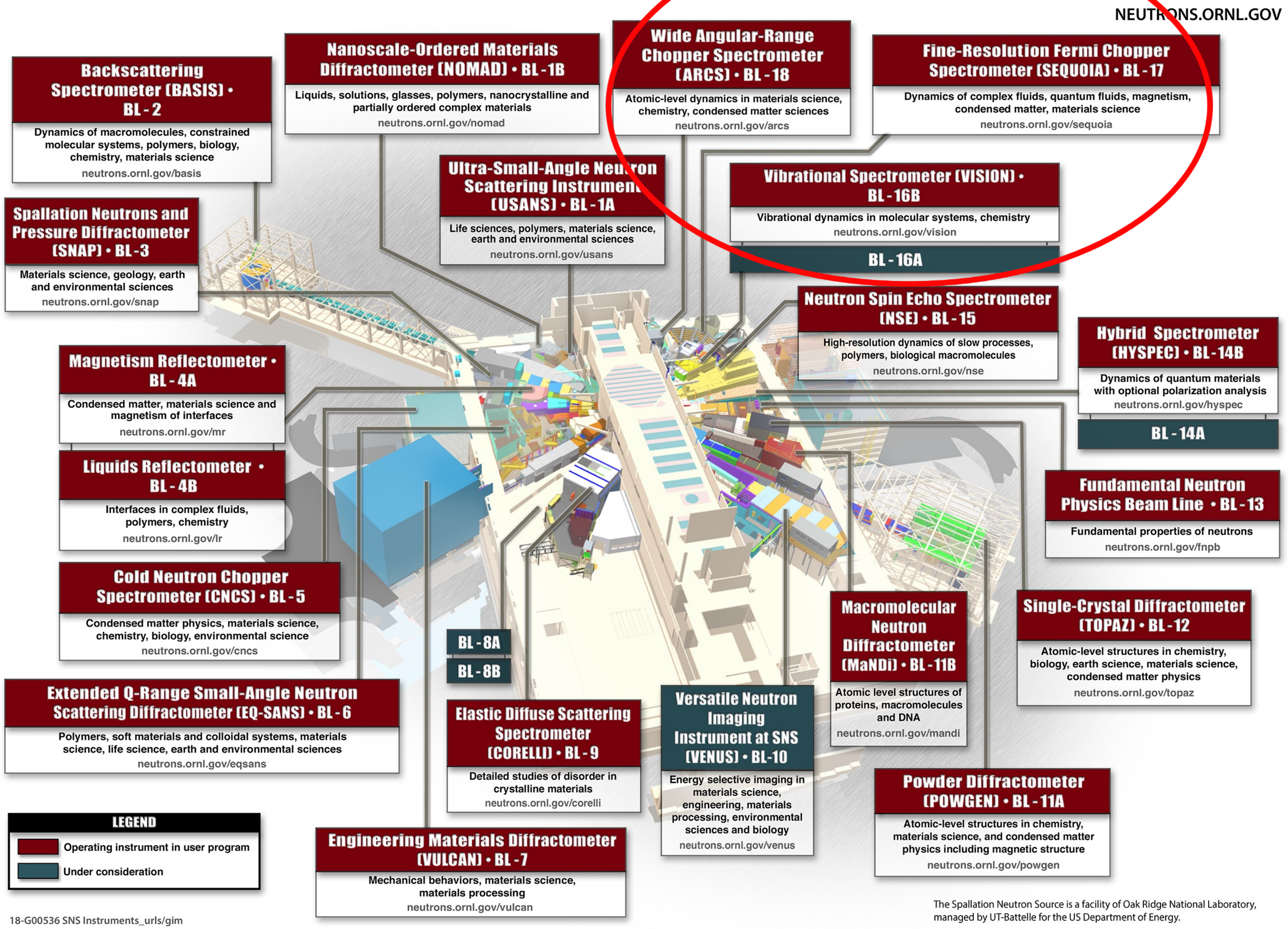
Experimental Capabilities – SNS

- High-energy protons accelerated to 1 GeV
  - 1.4 MW LINAC
- Neutron produced by spallation with mercury target
  - Pulsed neutrons produced at 60 Hz
- Neutrons thermalized by passing through room temperature water or 20 K liquid hydrogen moderators
- Peak brightness: ~1×10^{13} n/cm^2/sr/Å/s

Experimental Capabilities – SNS

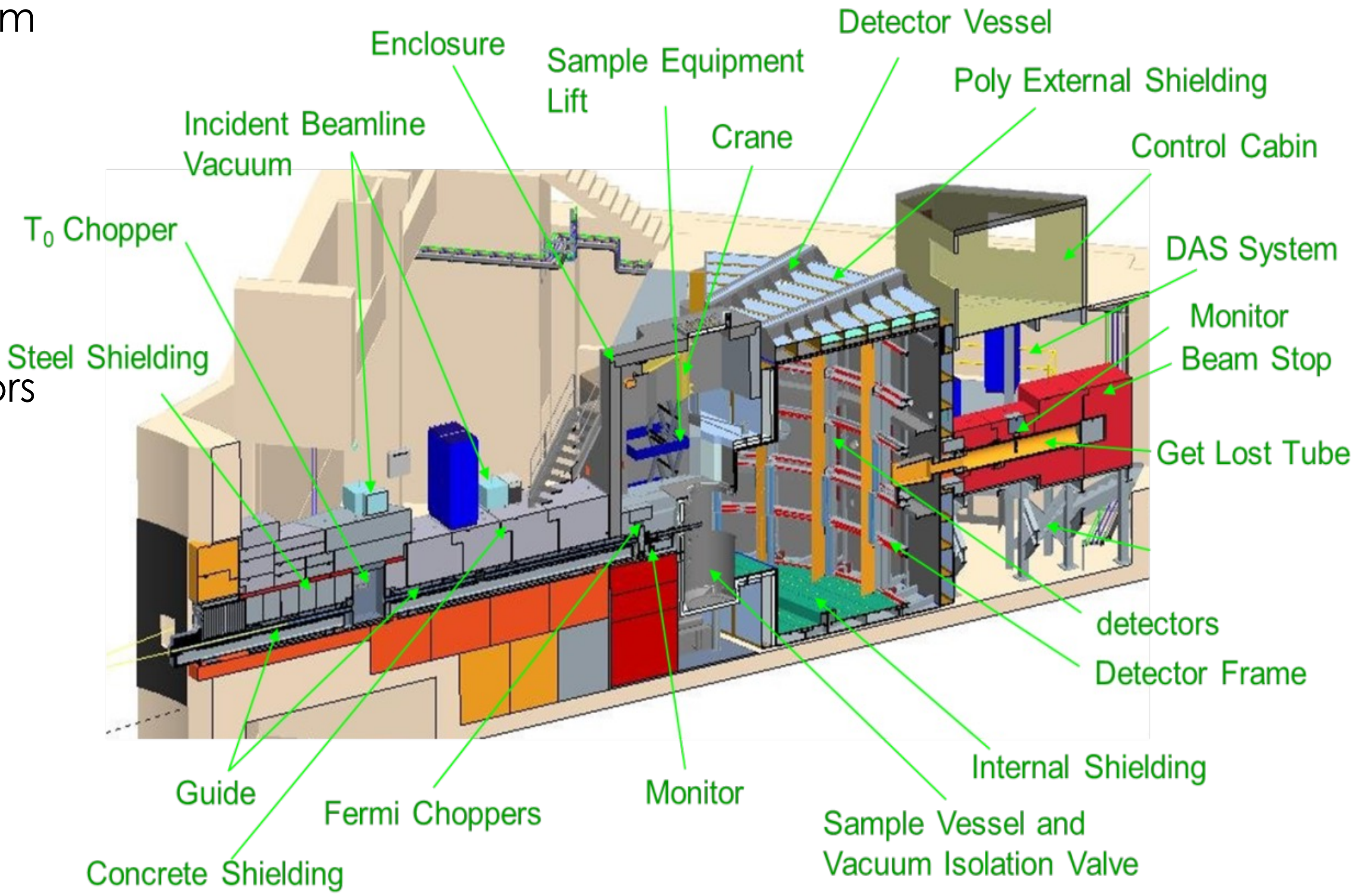


Experimental Capabilities – SNS



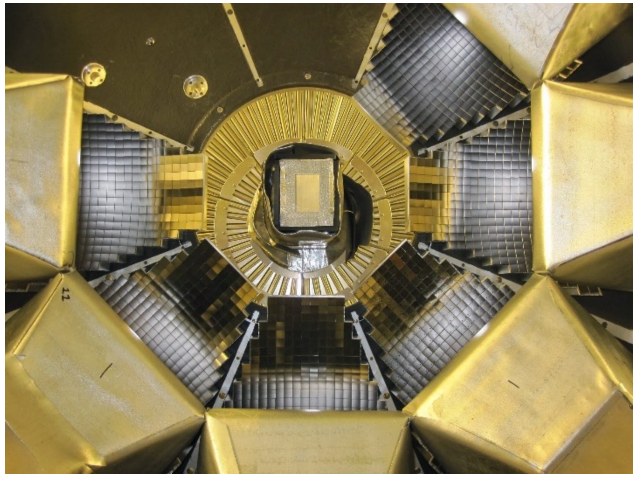
Experimental Capabilities: ARCS & SEQUOIA

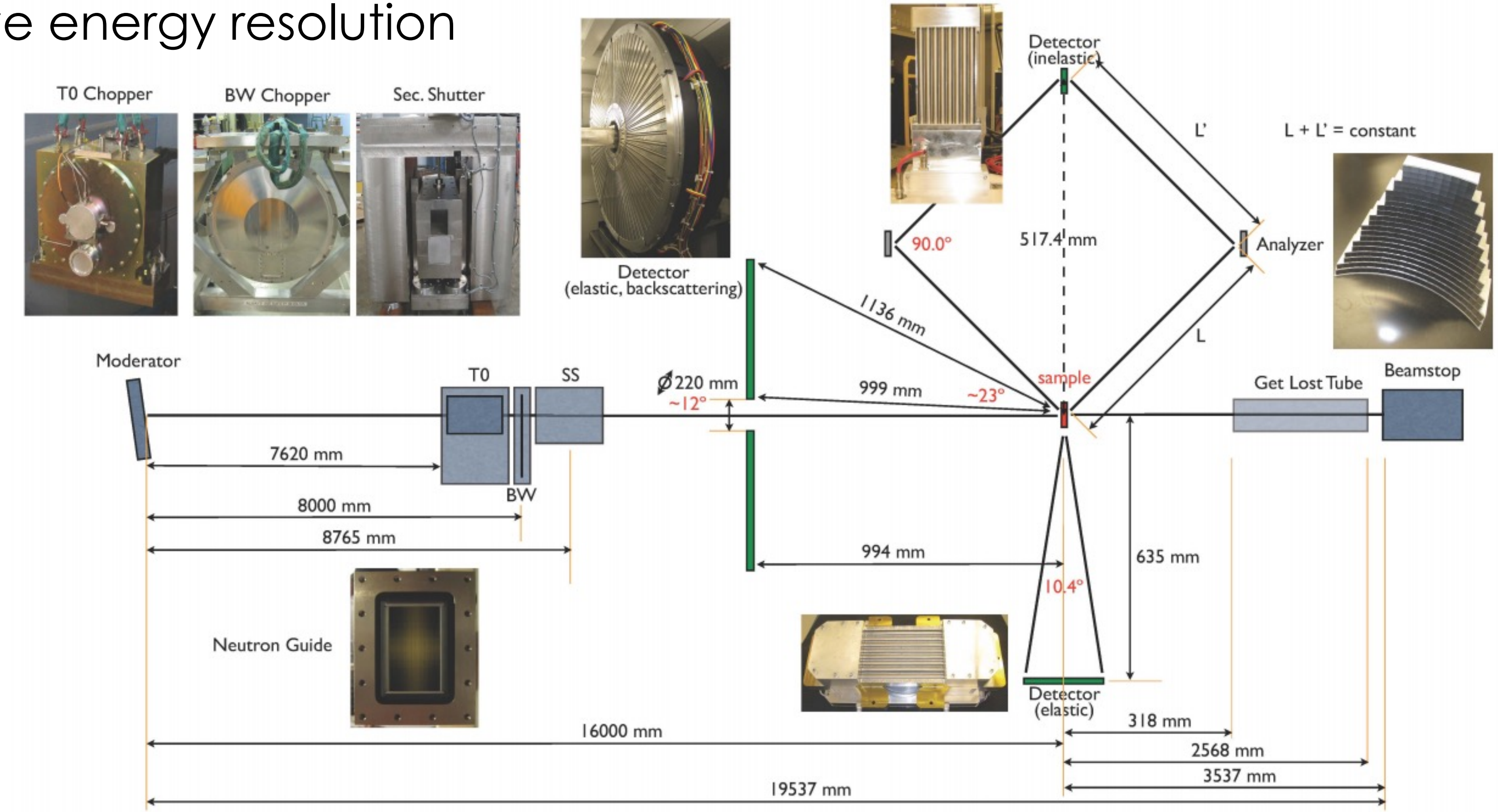
- Time-of-flight direct geometry spectrometer
- User chooses incident energy; Fermi choppers rotate to select energy from white beam
- Detector & data acquisition system (DAS) setup measures final energy and scattering angle
  - SEQUOIA: better energy resolution
  - ARCS: larger angular range
- Selection of furnaces and refrigerators for temperatures range 5 – 1800 K



Experimental Capabilities: VISION

- Indirect geometry vibrational spectrometer
- White beam of neutrons hits sample
- Scattered neutrons reflected off graphite blocks to two detectors for forward- and backward-scattering angles
- Graphite blocks configured to scatter neutrons at 4 meV
- Constant relative energy resolution

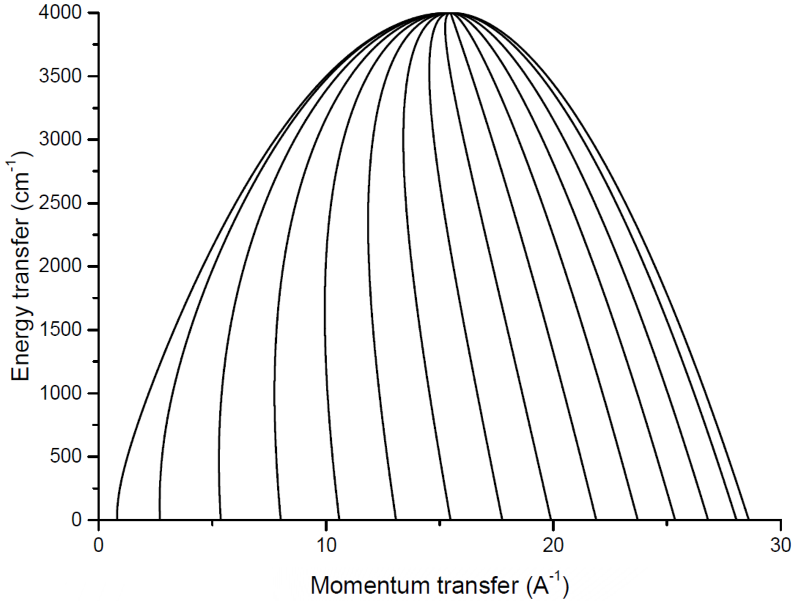




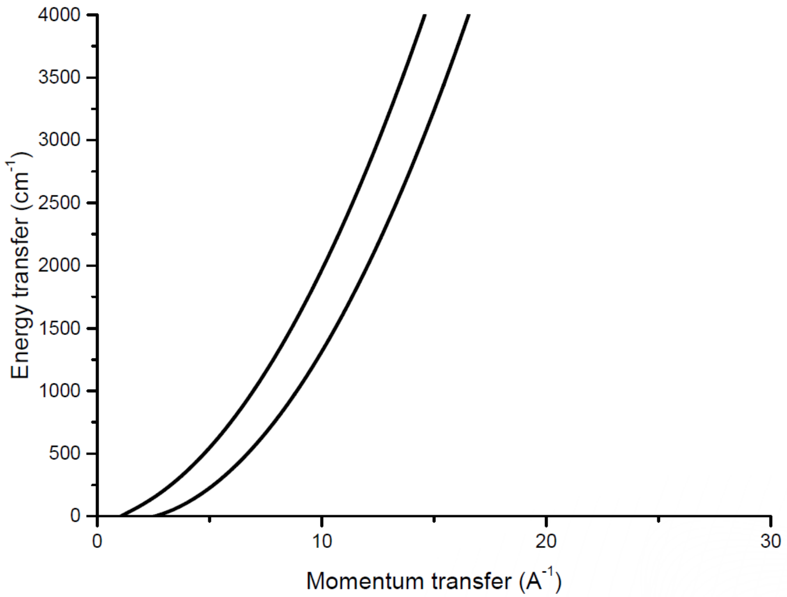
Experimental Capabilities: Comparison

Direct Geometry
(ARCS/SEQUOIA)

Indirect Geometry
(VISION)



Complimentary
measurement techniques

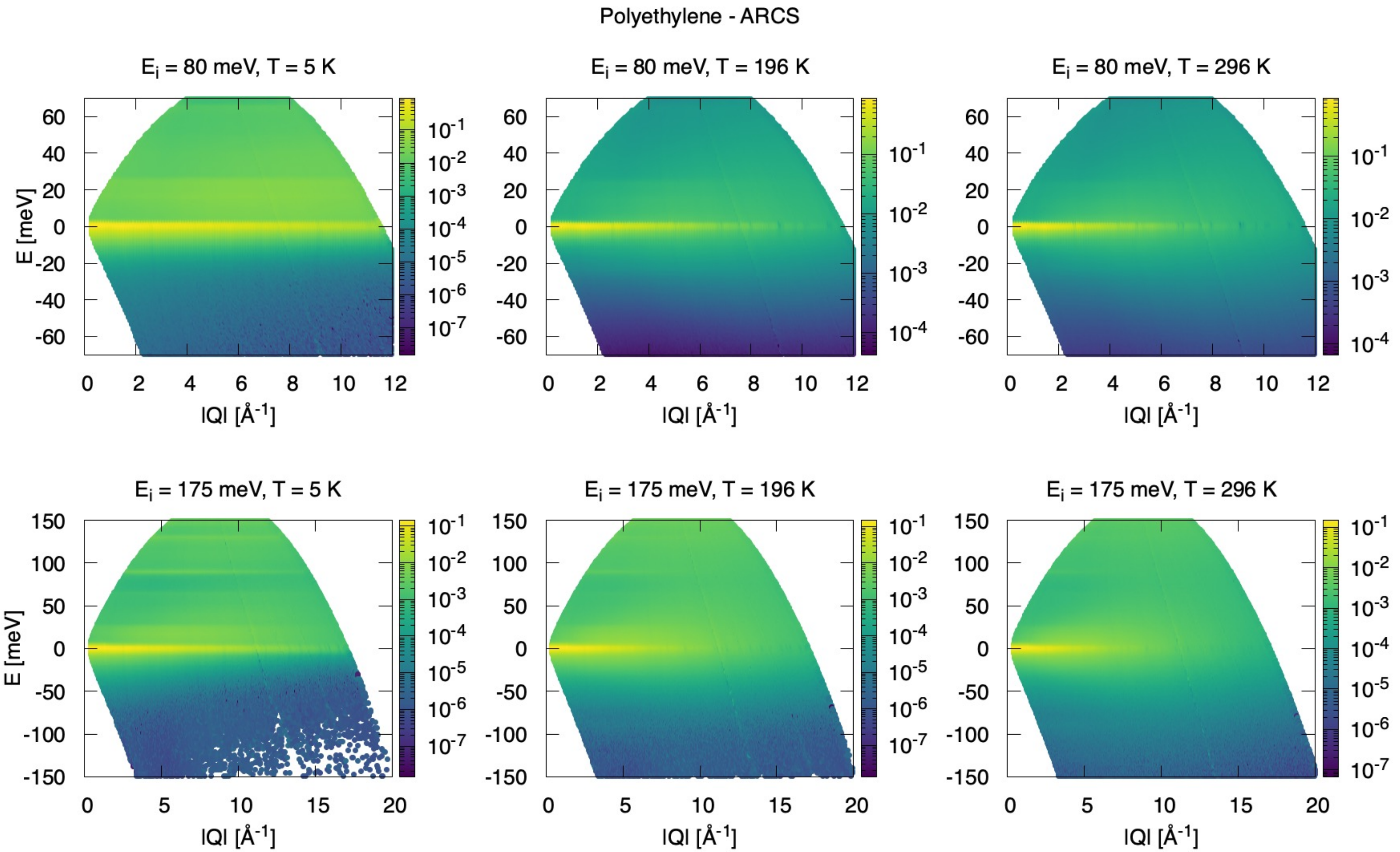


Evaluations – Polyethylene

- Ubiquitous moderator material
- Carried out in collaboration with RPI as a follow up on [1]
- Interested in changes in the inelastic spectra as a function of temperature

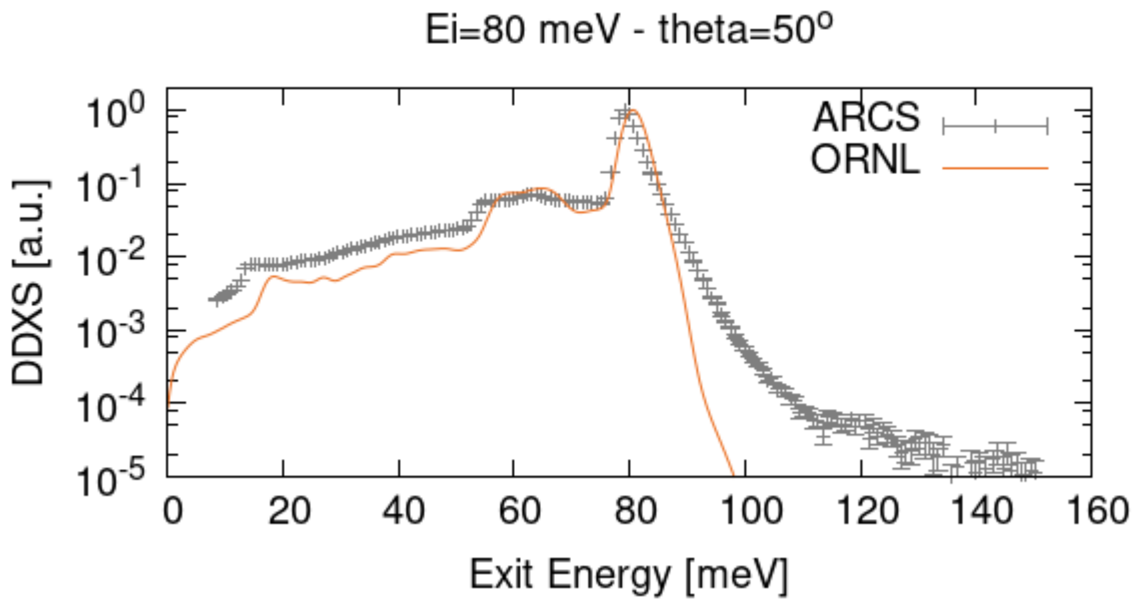
Instrument	Incident neutron energy [meV]	Temperature [K]
ARCS	80, 175, 400	5, 100, 196, 268, 295, 313
	250	5, 295
VISION	N/A	5, 20, 40, 60, 77, 100, 120, 140, 160, 180, 196, 220, 233, 240, 263, 283, 293.6, 300, 303, 313, 323, 333, 343, 350

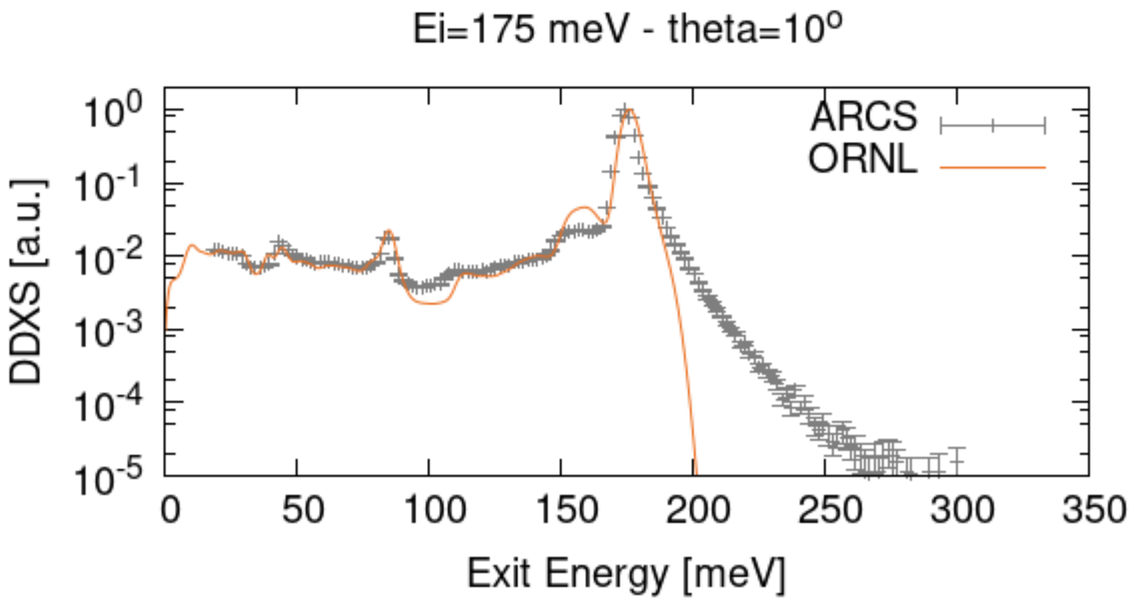
Evaluations – Polyethylene – ARCS

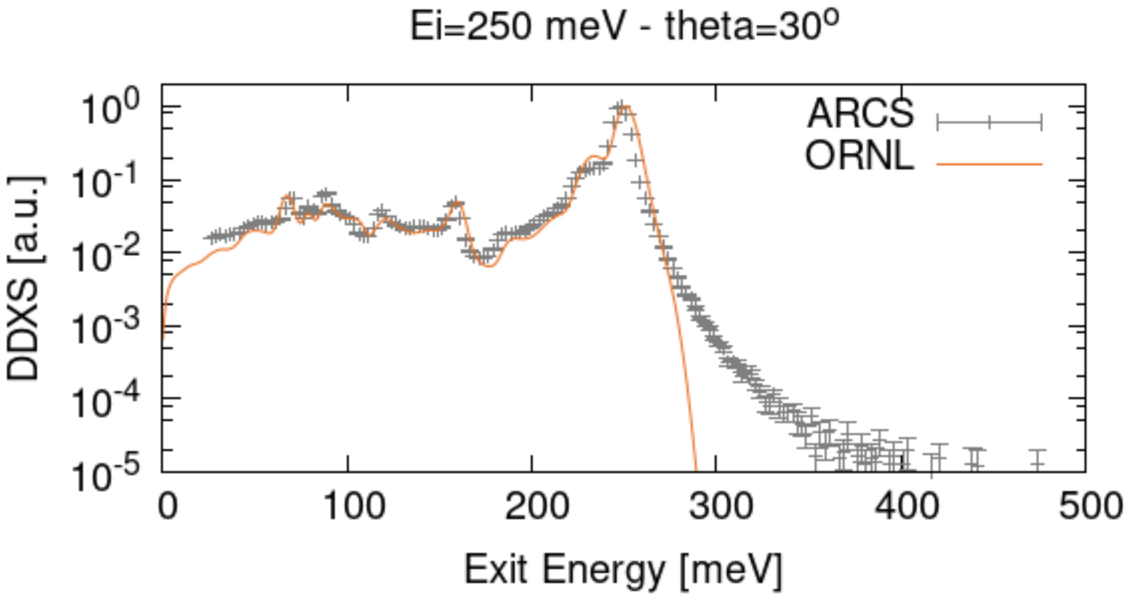


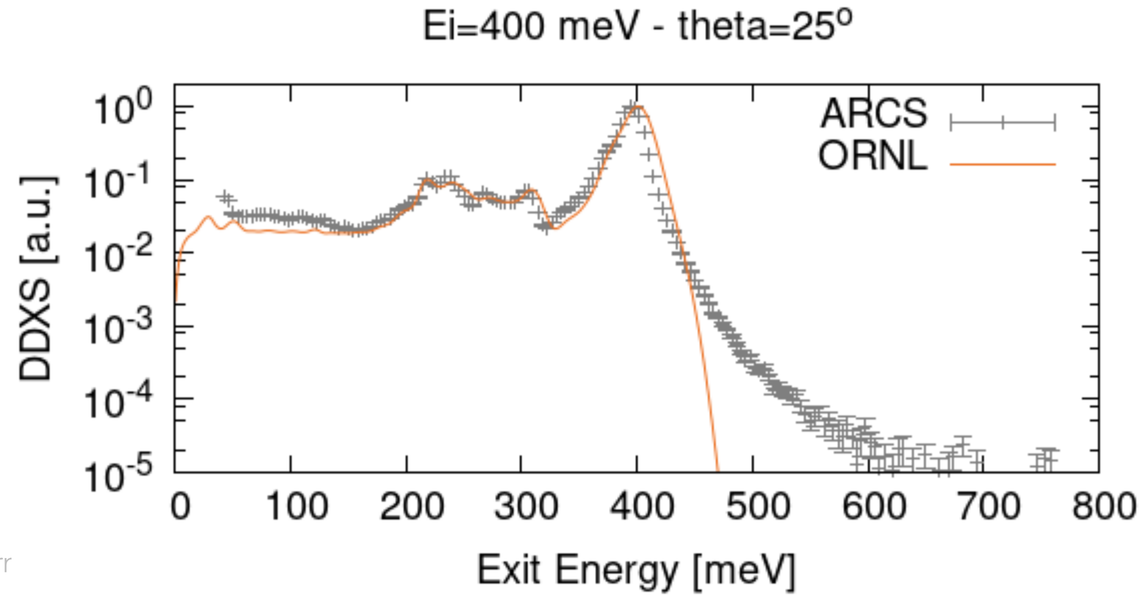
Loss of fine-resolution features at increasing temperatures

Evaluations – Polyethylene – ARCS









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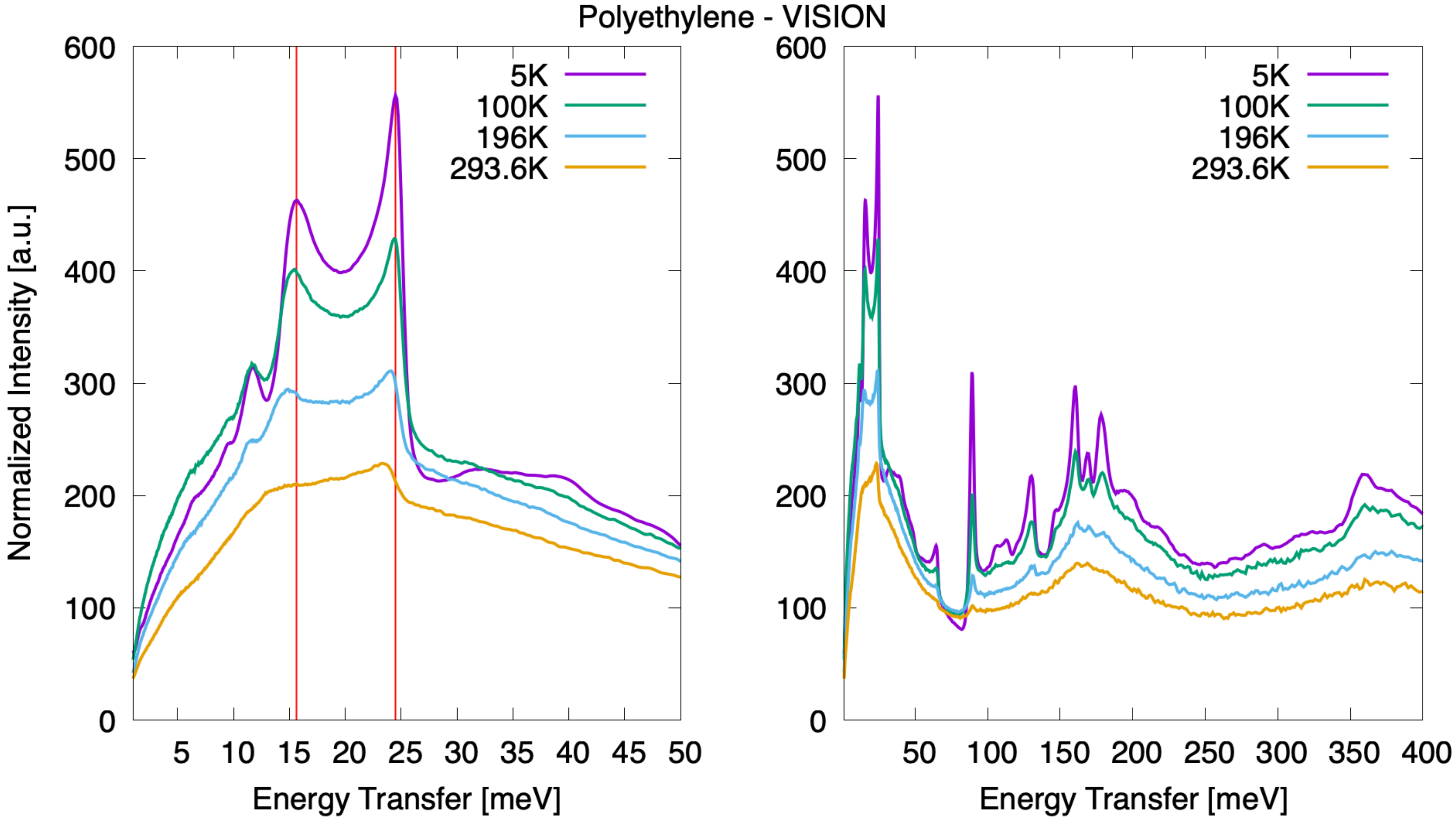
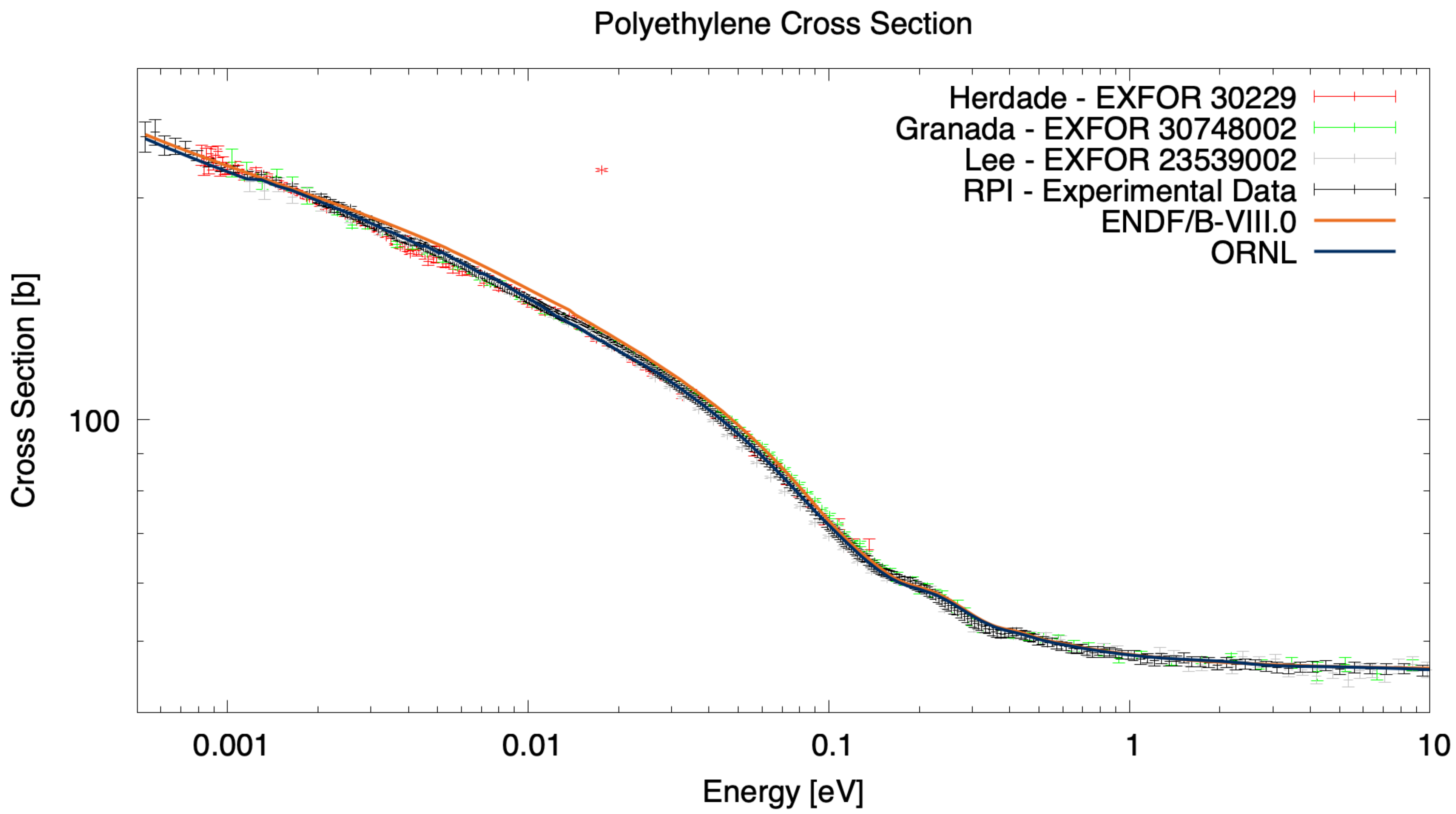
Evaluations – Polyethylene – VISION



Shifted peaks represent changes in lattice parameters

Broadening at higher temperatures & loss of fine-resolution features

Evaluations – Polyethylene – Total XS

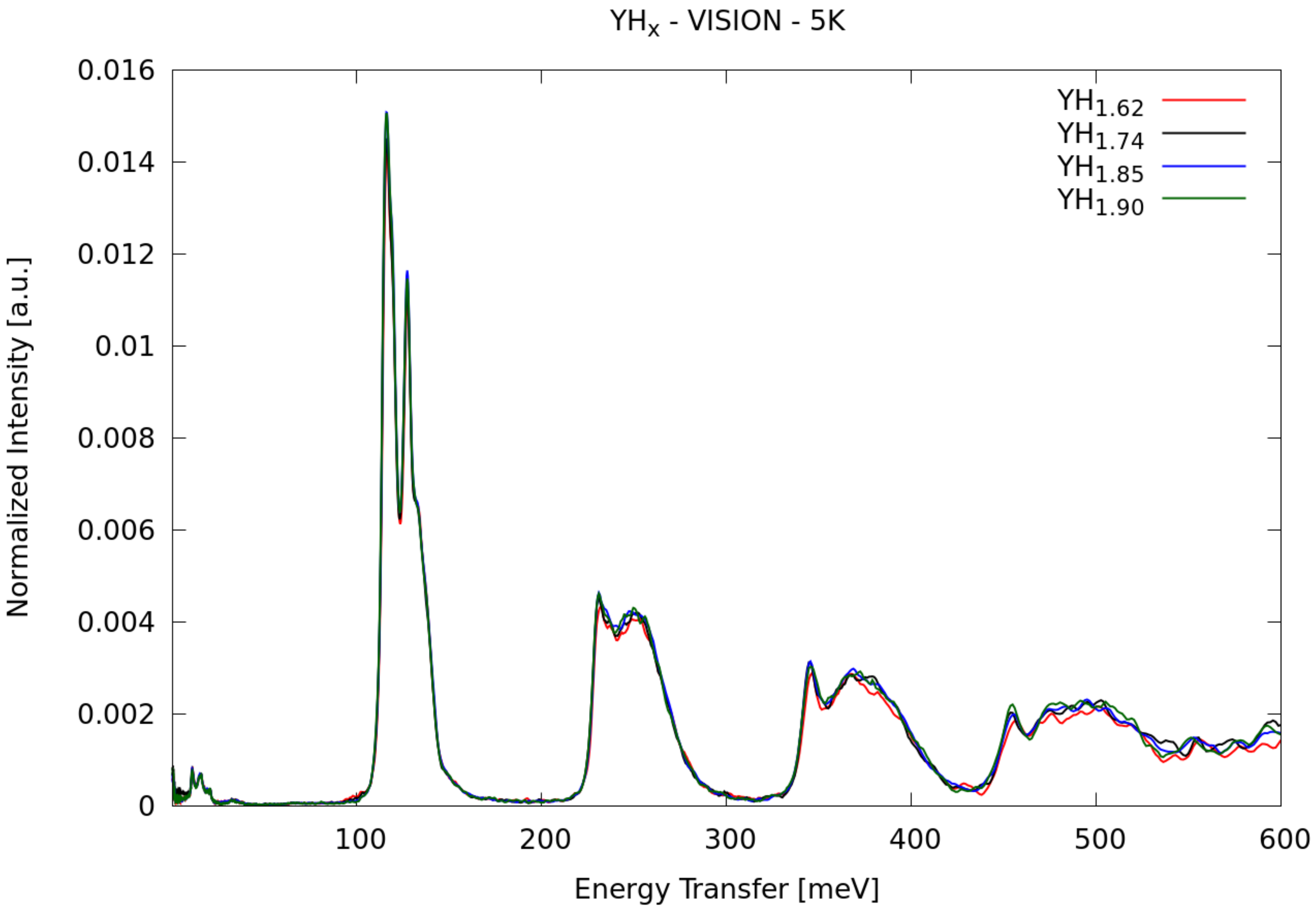


Evaluations – Yttrium Hydride

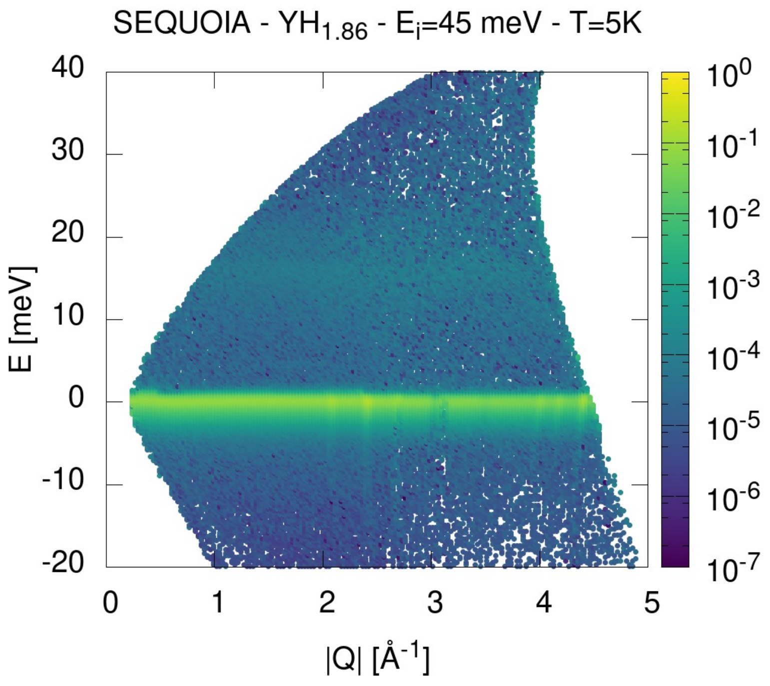
- Moderator of choice for Oak Ridge National Laboratory (ORNL) Transformational Challenge Reactor (TCR)
- Measurement is two-fold:
  - Hydrogen concentration
  - Anharmonicities at high temperatures
- Full report available here [2]

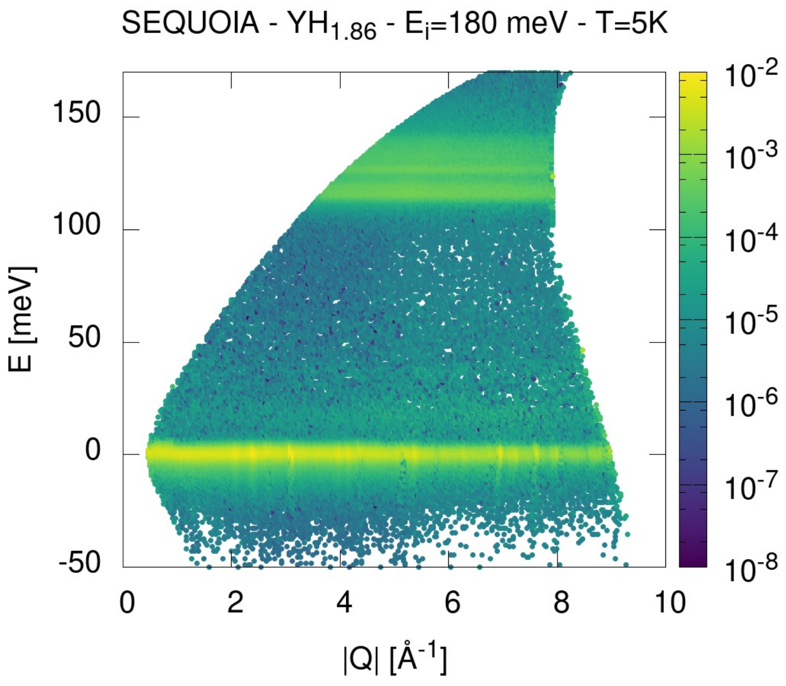
SNS Instrument	Samples	Sample Form	Sample Fixture	Mass [g]	Incident neutron energy [meV]	Temperature [K]
SEQUOIA	$YH_{1.62}$	Powder	Aluminum plate and cover	0.6966	45, 180, 600	5
	$YH_{1.86}$			0.702		5, 295, 550, 800
VISION	$YH_{1.62}$			0.6966	N/A	5, 293
	$YH_{1.74}$			0.6684		
	$YH_{1.86}$			0.702		
	$YH_{1.90}$			0.7696		
ARCS	$YH_{1.68}$	0.1 mm hydride foil	Thin-wall quartz tube	1.6681	45, 180, 600	295, 550, 800, 900, 1,000, 1,100, 1,200
	$YH_{1.87}$			1.8937		

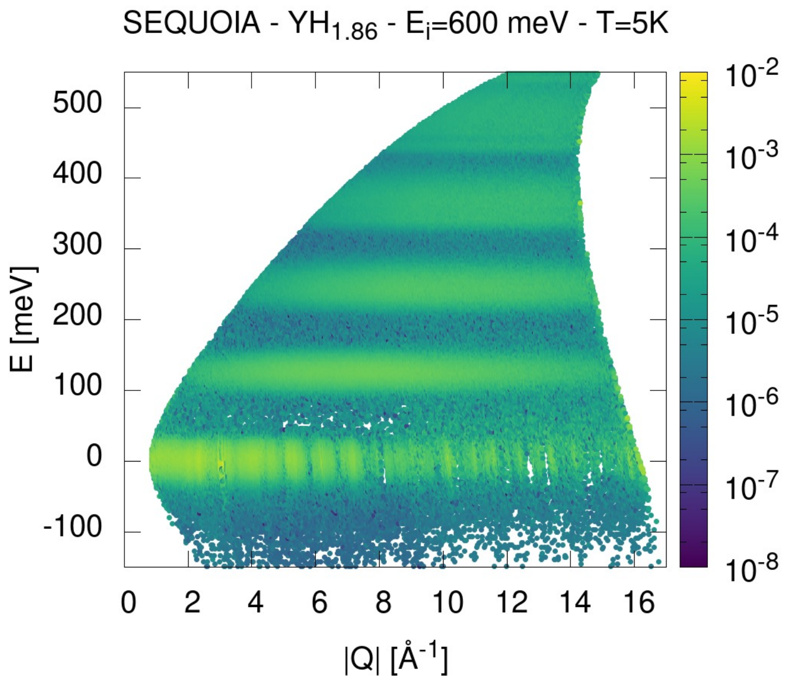
Evaluations – Yttrium Hydride – VISION



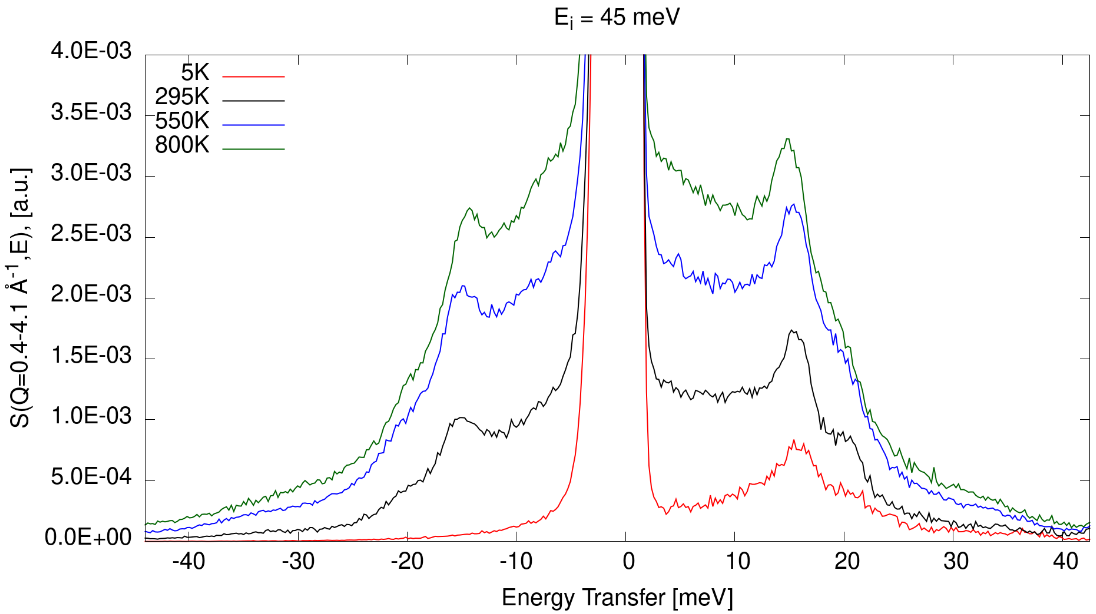
Evaluations – Yttrium Hydride – SEQUOIA

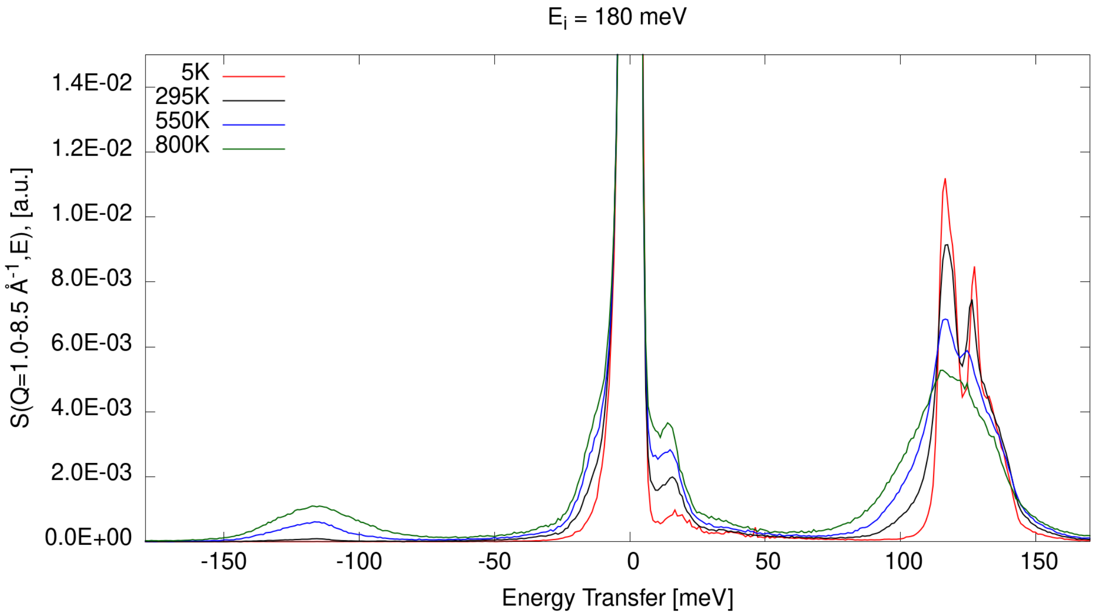


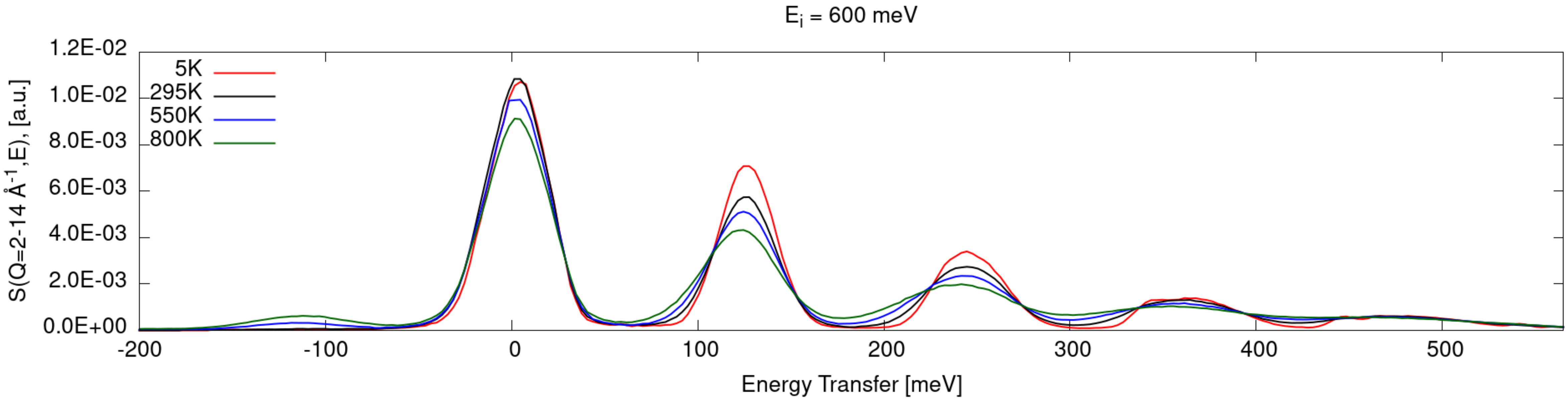




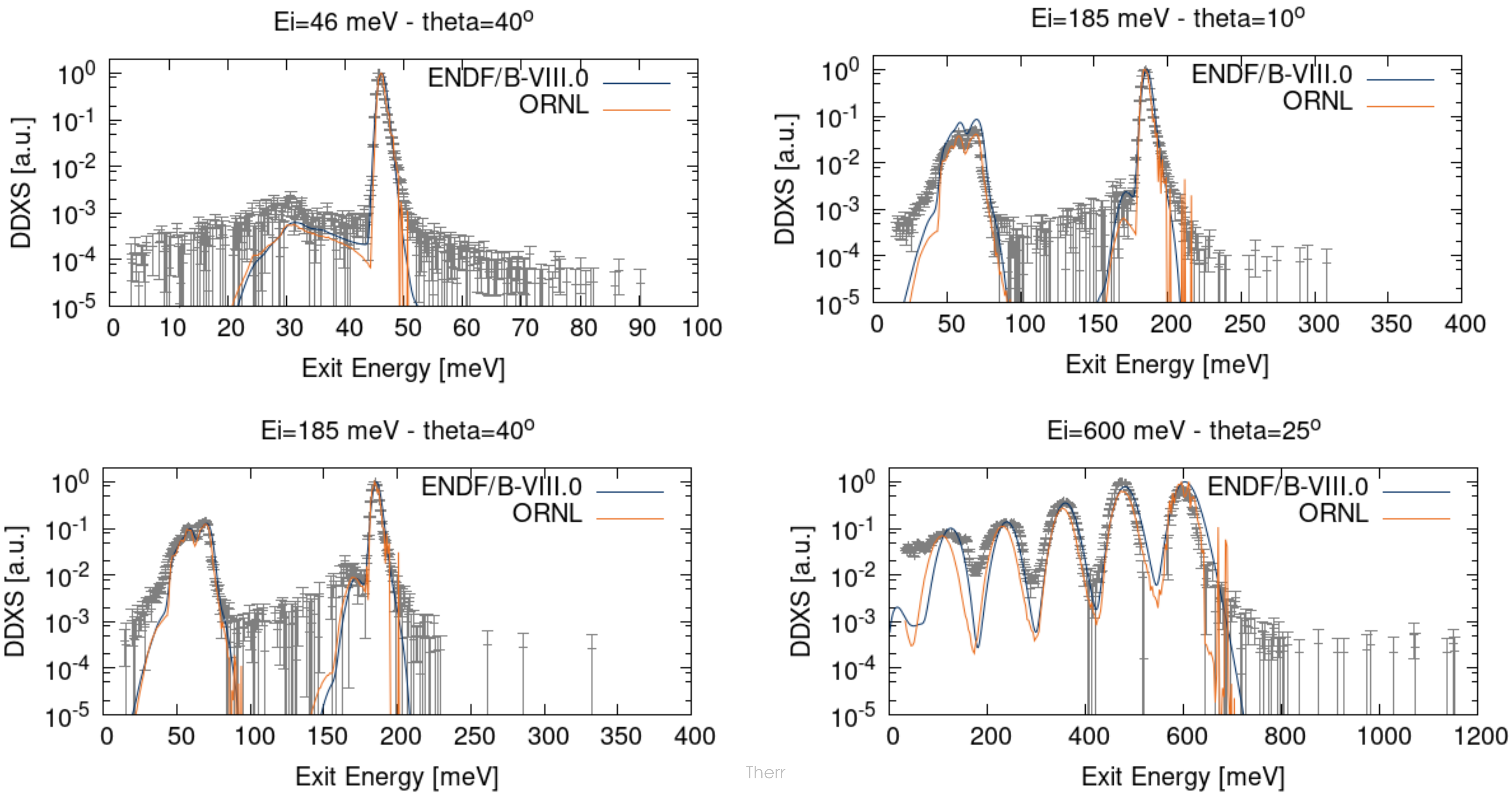
Evaluations – Yttrium Hydride – SEQUOIA







Evaluations – Yttrium Hydride – SEQUOIA



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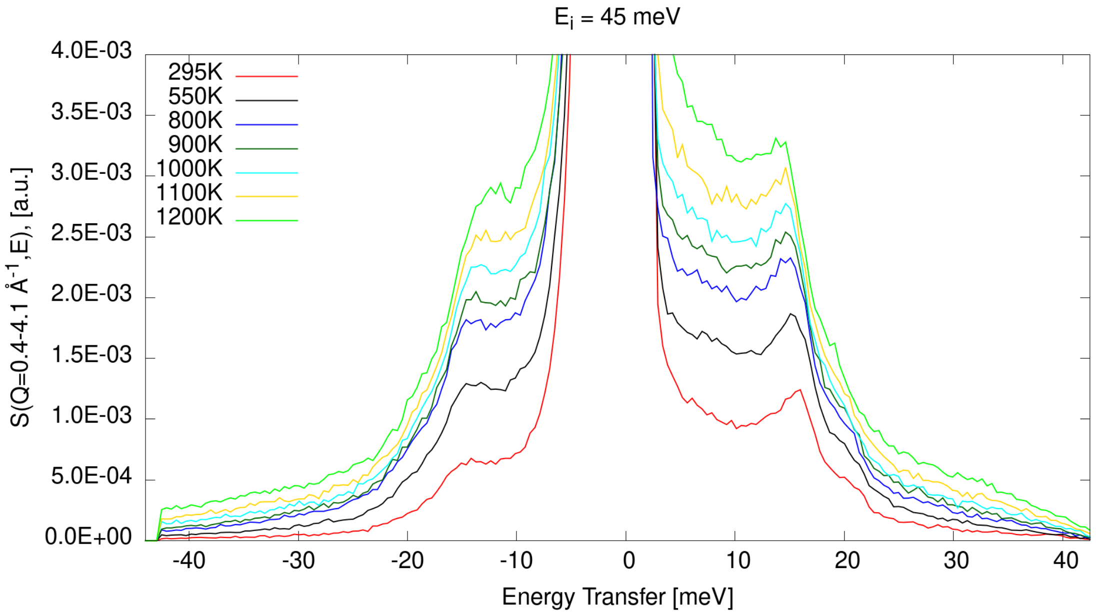
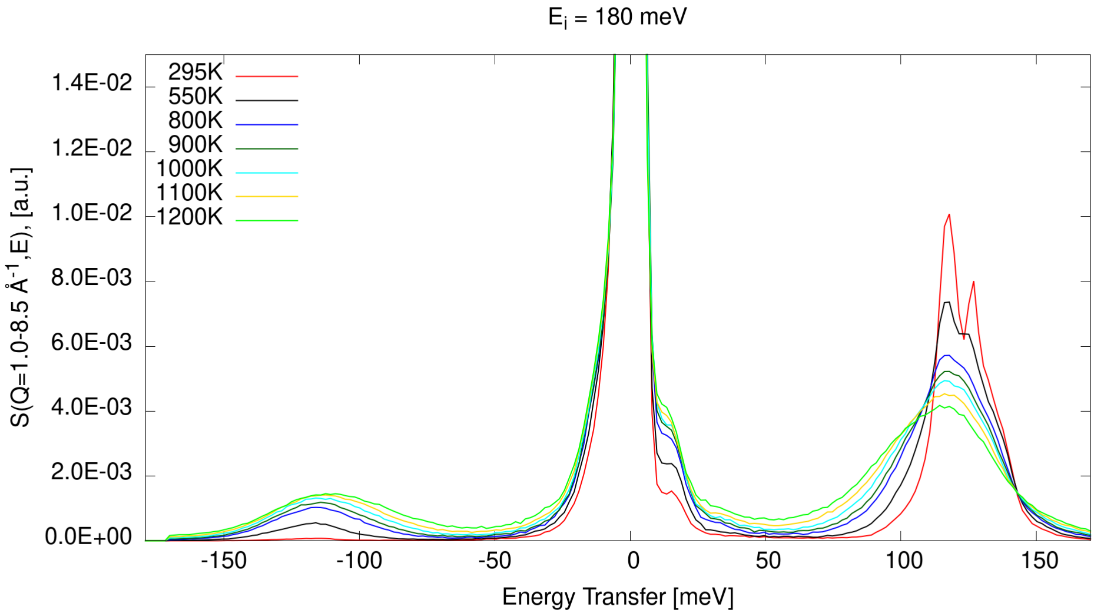
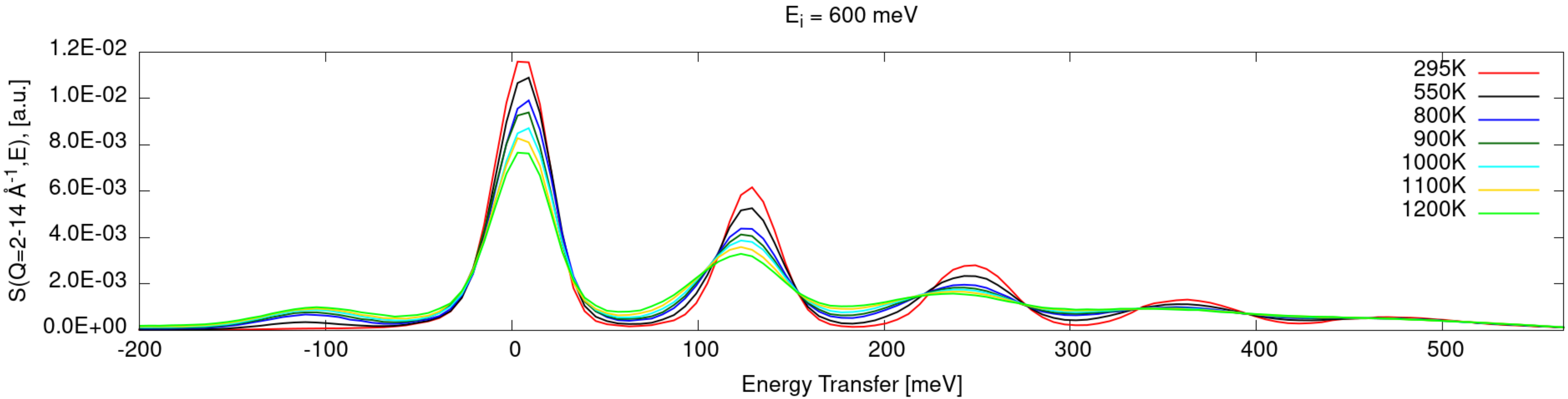
Evaluations – Yttrium Hydride – ARCS







Uncertainty Quantification

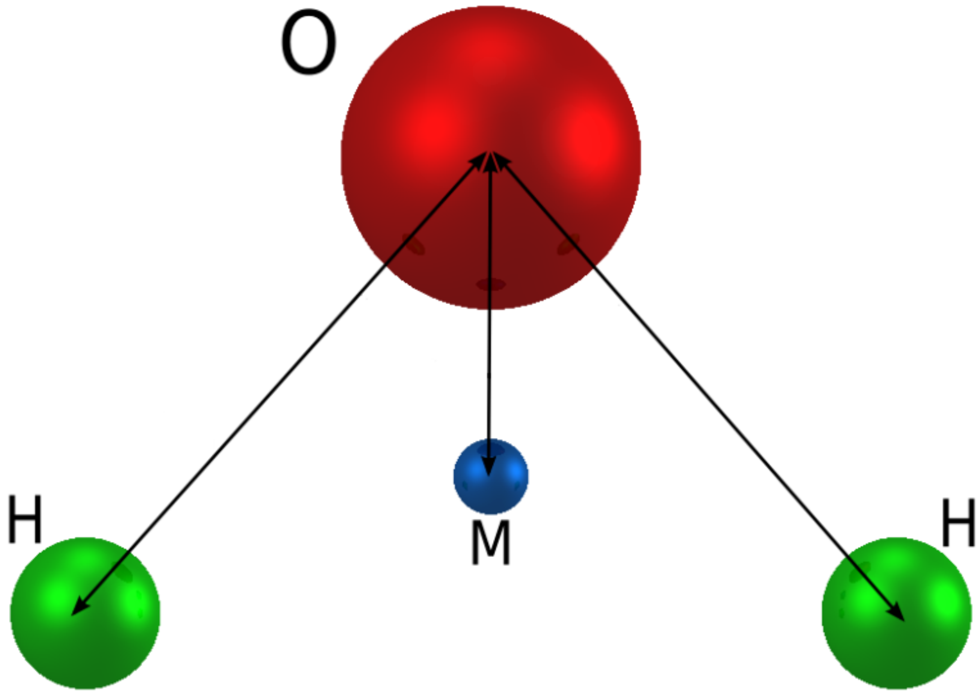
- Goal was to develop a procedure to generate a covariance matrix for $S(\alpha, \beta)$ data that incorporated both computation simulations and experimental data
- Experimental data and computer simulation fit is achieved using the Unified Monte Carlo (UMC) method [3]
- Framework should not rely on how the TSL file is generated:
  - Classical MD vs. DFT
  - Underlying assumptions used to calculate TSL from computational simulation methods
- Demonstrated using light water

Uncertainty Quantification

- Active area of research, as evident by:
  - WPEC Subgroup 44
  - Monte Carlo perturbations of phonon density of states [4]
  - Generalized least-squares uncertainty quantification of LEAPR [5] and molecular dynamics parameters [6] to data

Uncertainty Quantification

- Water is difficult to model computationally
- Properties are calculated using molecular dynamics (MD)
- Models categorized by the number of ‘sites’
- 3-6 sites, extra sites are ‘dummy’ particles
- Over 30 different models of water exist



Uncertainty Quantification

- Used the TIP4P/2005f [7] parameter set and varied 8 model parameters (7 in red below plus spacing between oxygen and 'dummy' atoms) using Latin hypercube sampling (±5%) to ensure representative sampling of phase space

$$V_{i,j}(r_{i,j}) = \frac{1}{4\pi\epsilon_o}\frac{q_i q_j}{r_{ij}} + 4\epsilon\left(\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right) + D_r\left(1 - e^{-\beta(r_{ij}-b_o)^2}\right) + \frac{1}{2}k_\theta(\theta_{ijk} - \theta_o)^2$$

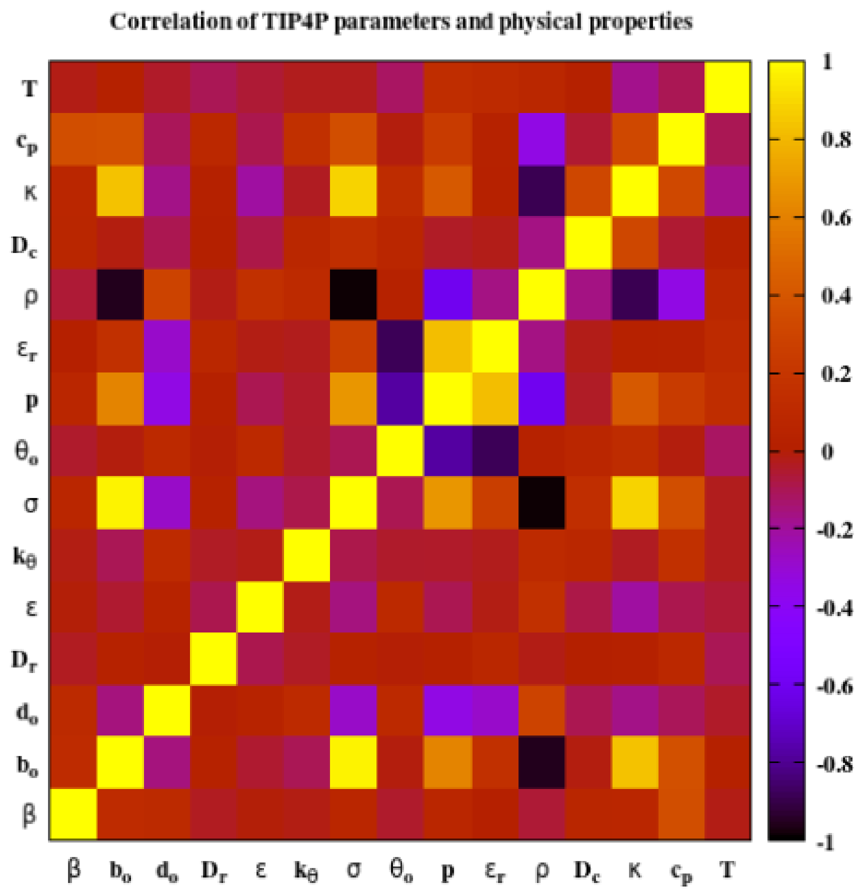
- 2048 randomly generated parameter samples were generated, of which 1615 successfully completed (job failures due to unphysical combination of parameters)

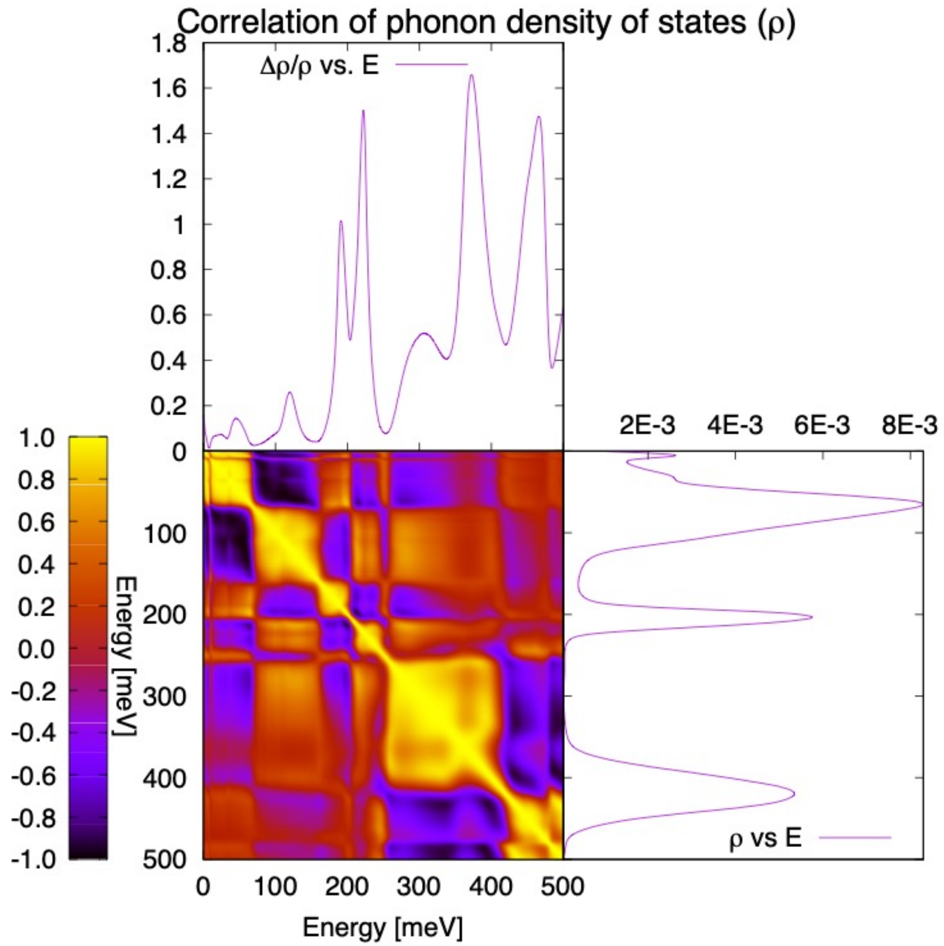
- From those 1615, the 250 simulations with the diffusion coefficient and density closest to their experimental values were chosen

Uncertainty Quantification

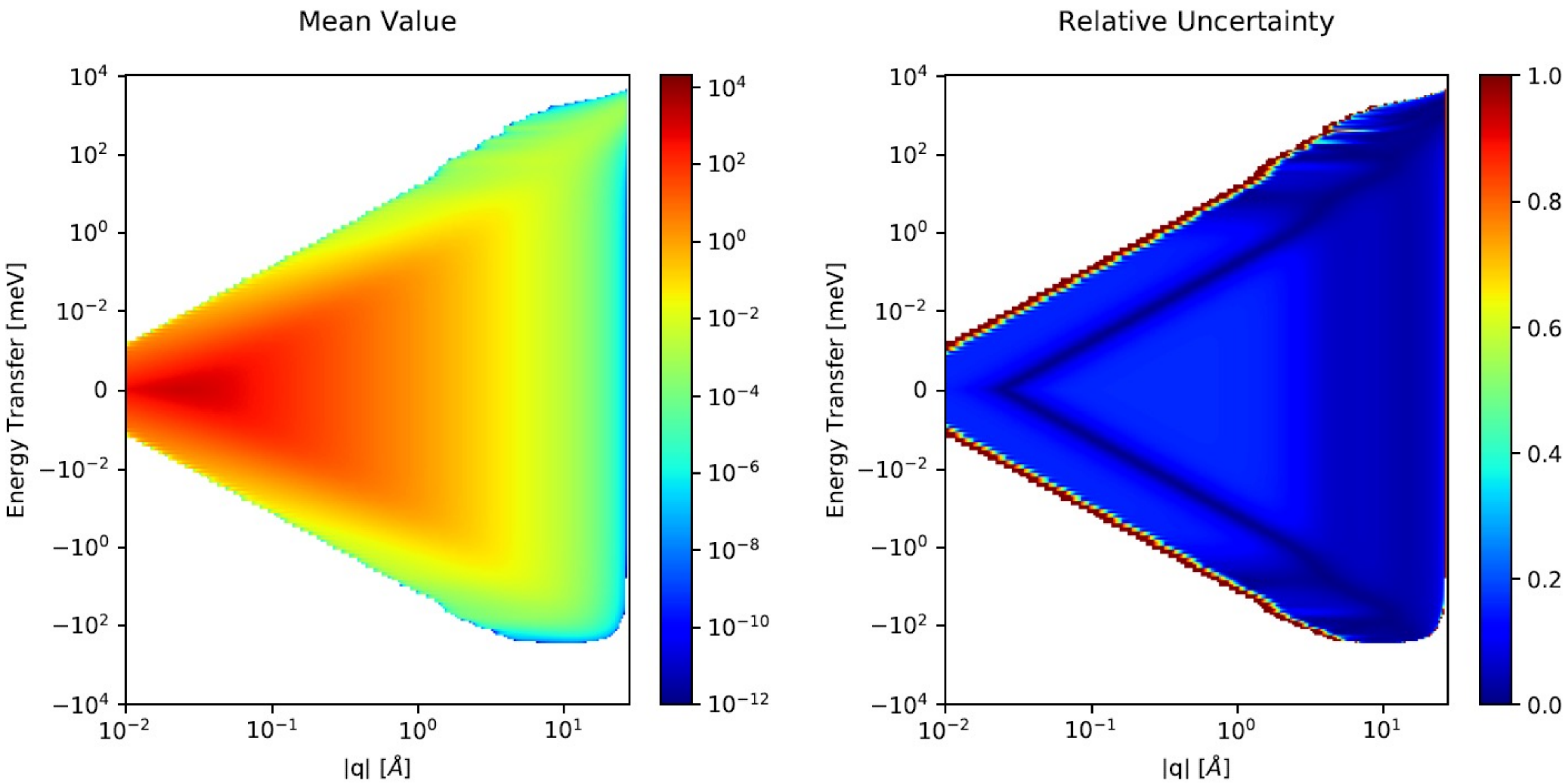
- These 250 accepted results were used to compare against experimental data gathered at the SNS
- 300K measurement performed by RPI in 2011 at SEQUOIA beamline
- 55, 160, 250, 600, 1000, 3000, and 5000 meV incident neutron energies
- Phonon density of states (pDOS) calculated from trajectory information of simulations, which were then used to calculate $S(\alpha, \beta)$
- Simplified model of SEQUOIA detector in MCNP used to include experimental effects

Uncertainty Quantification – Correlation Matrices

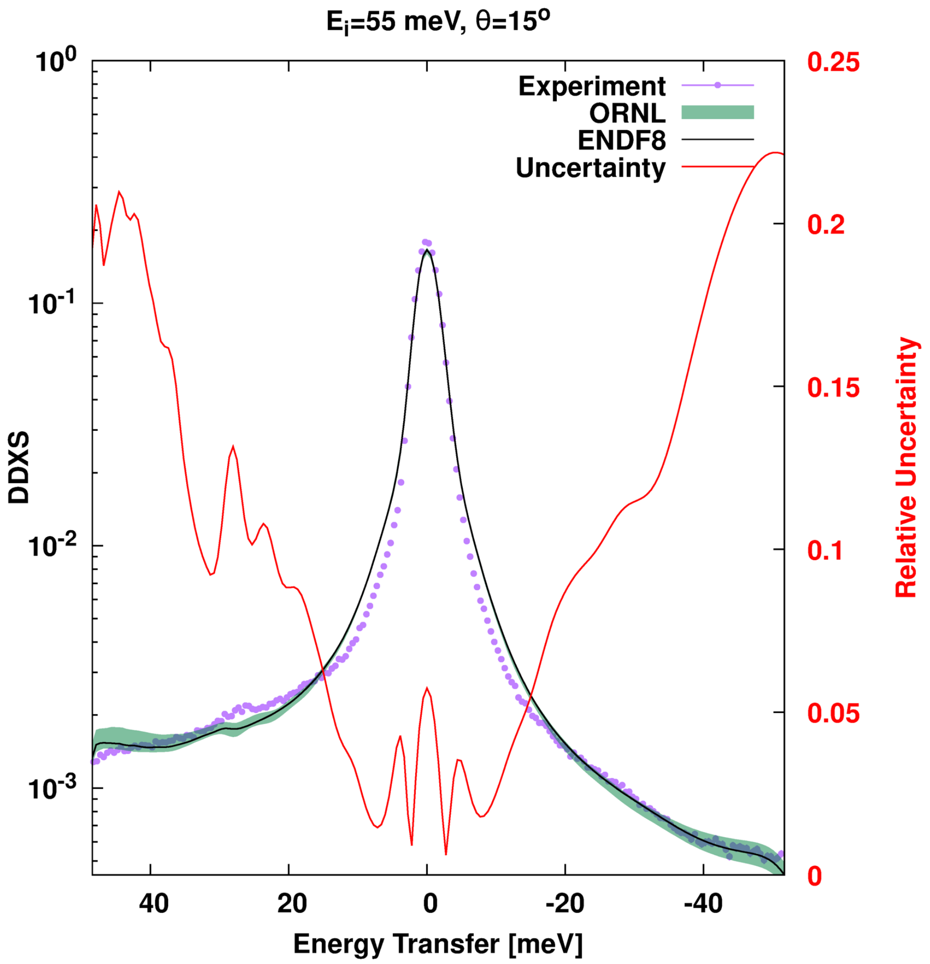


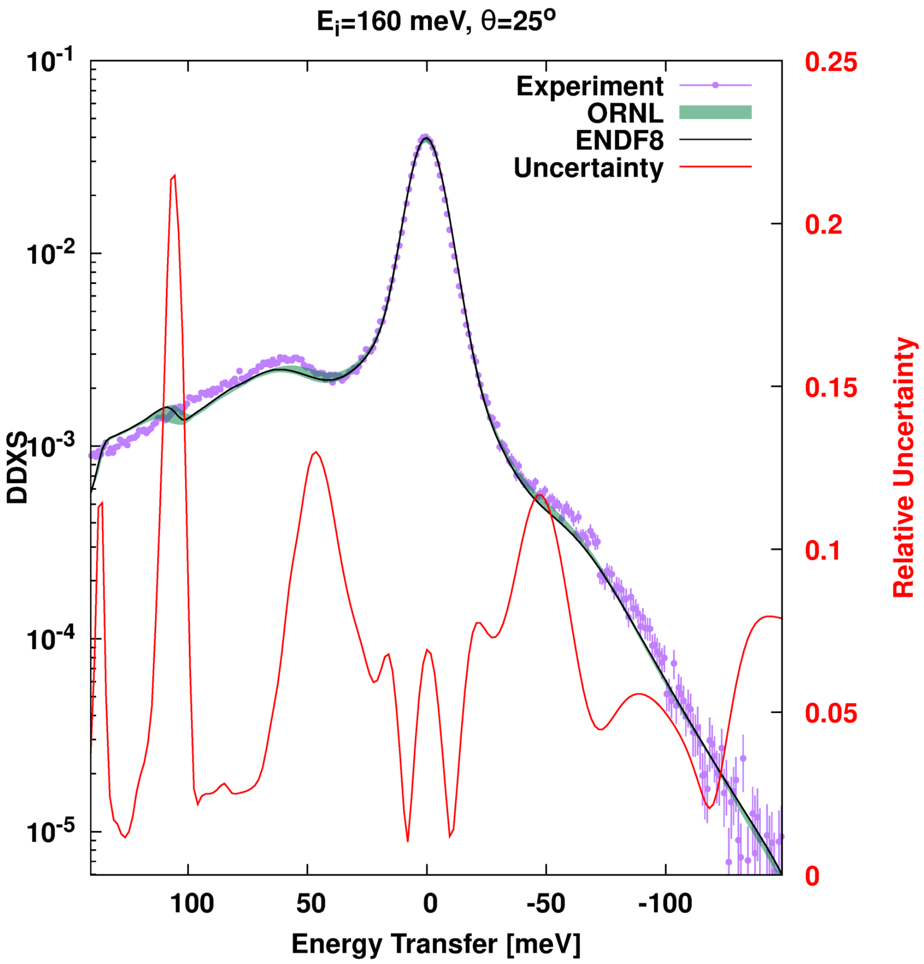


Uncertainty Quantification – $S(Q, E)$ and Uncertainties

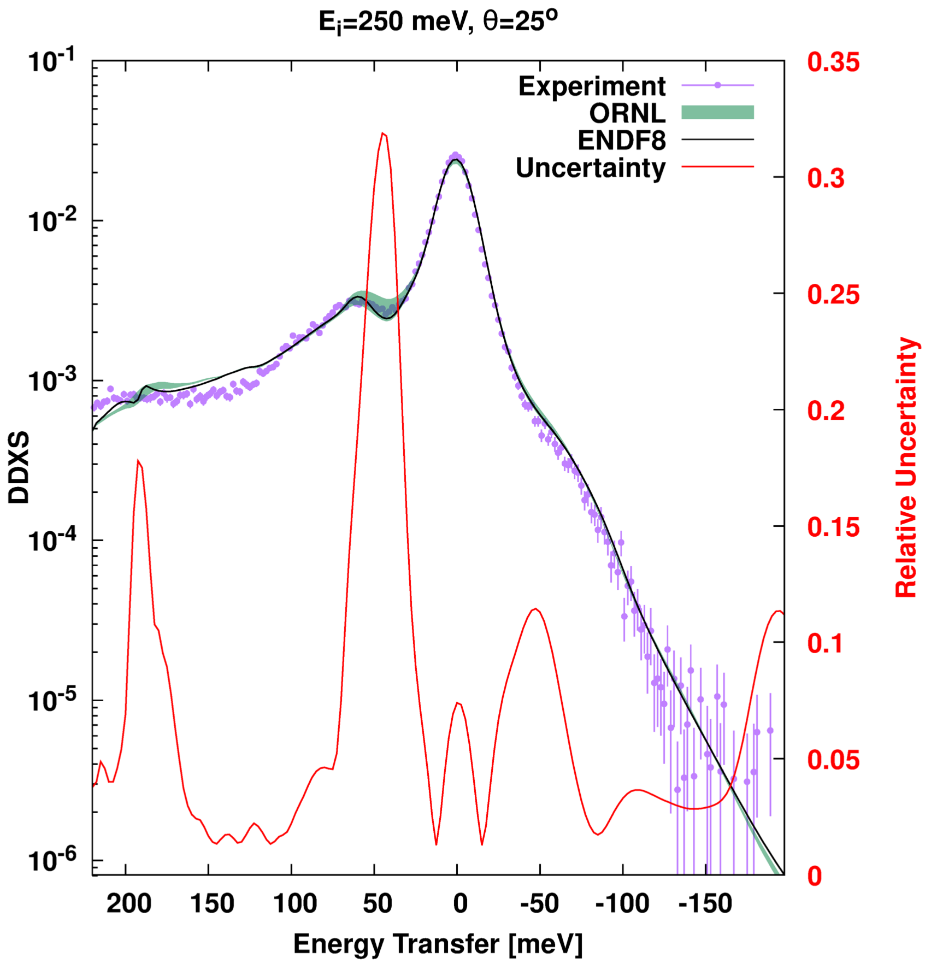


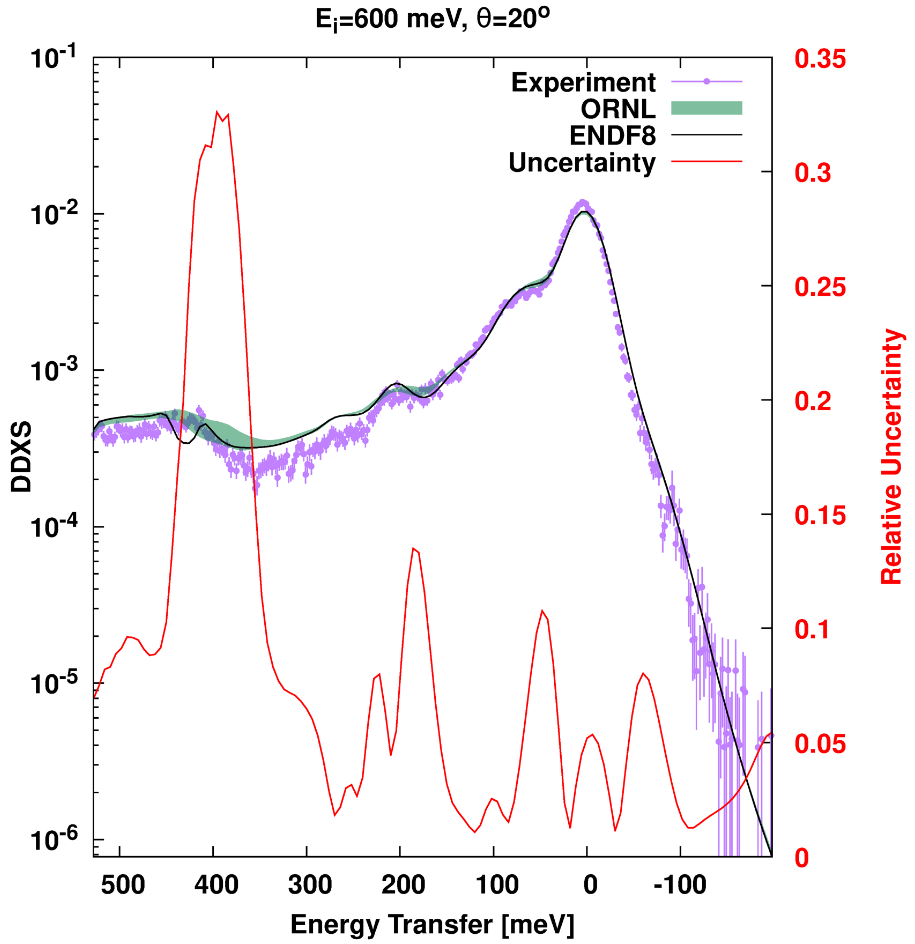
Uncertainty Quantification – DDXS





Uncertainty Quantification – DDXS (cont.)





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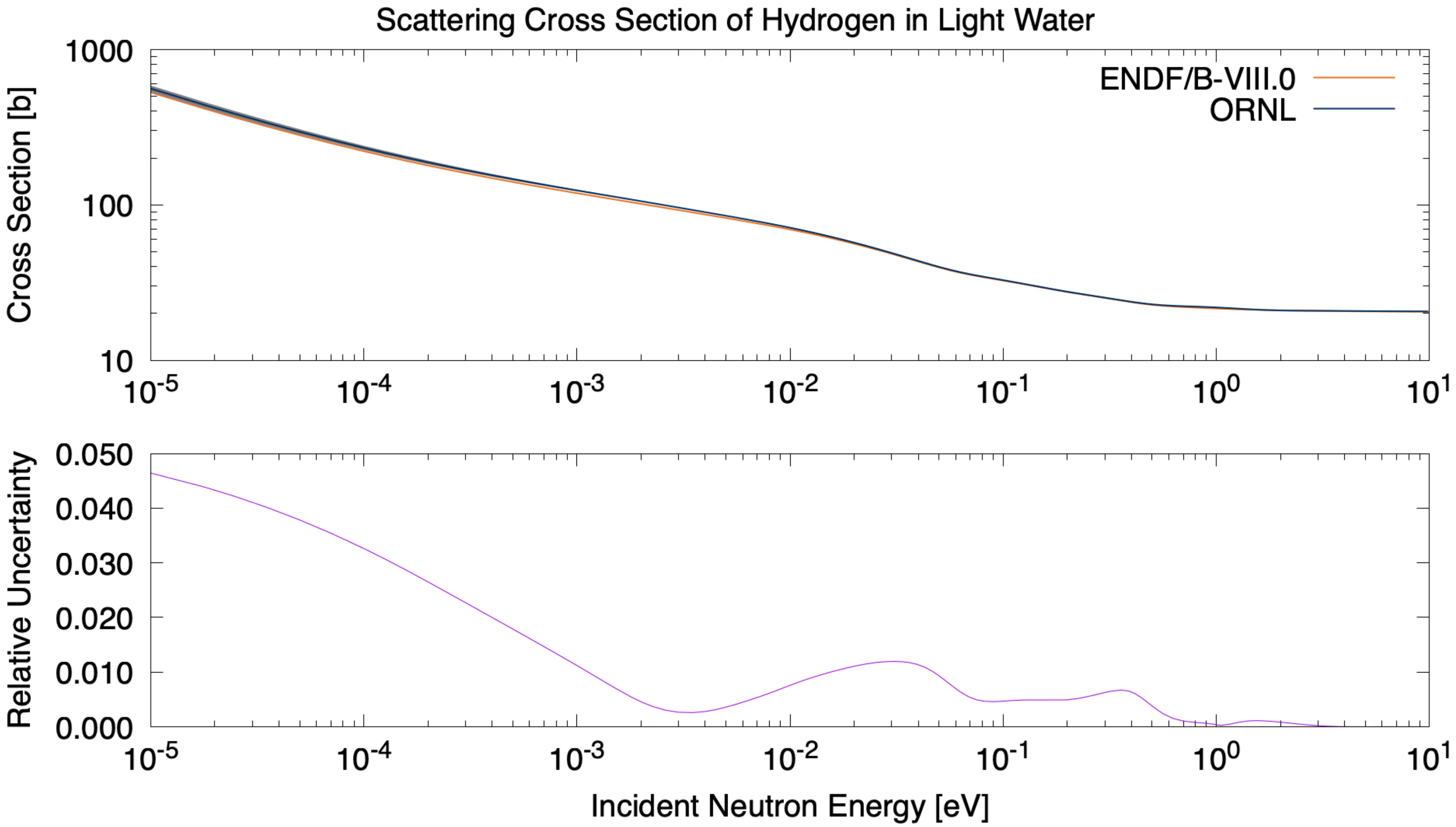
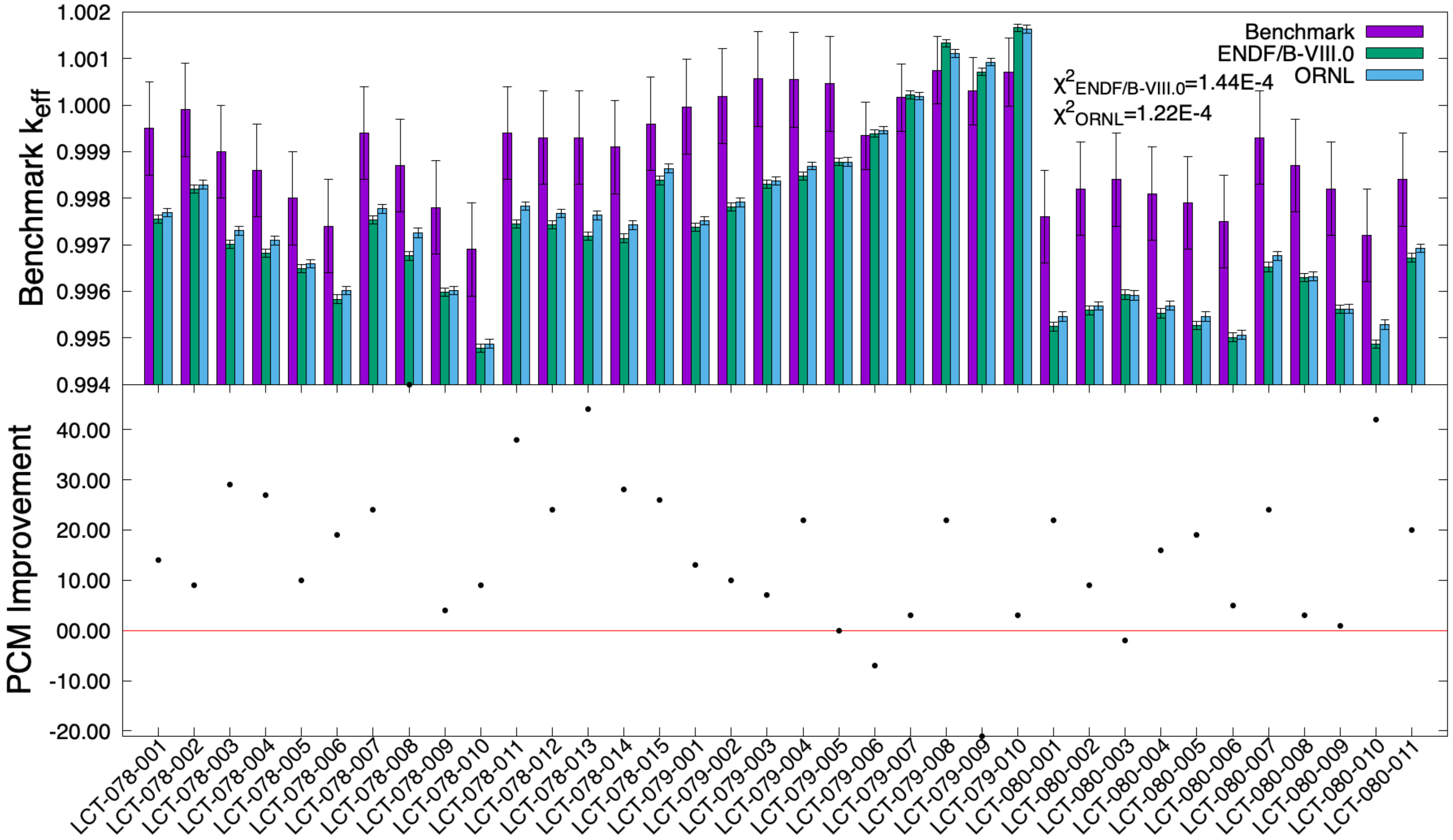
Uncertainty Quantification – Total XS



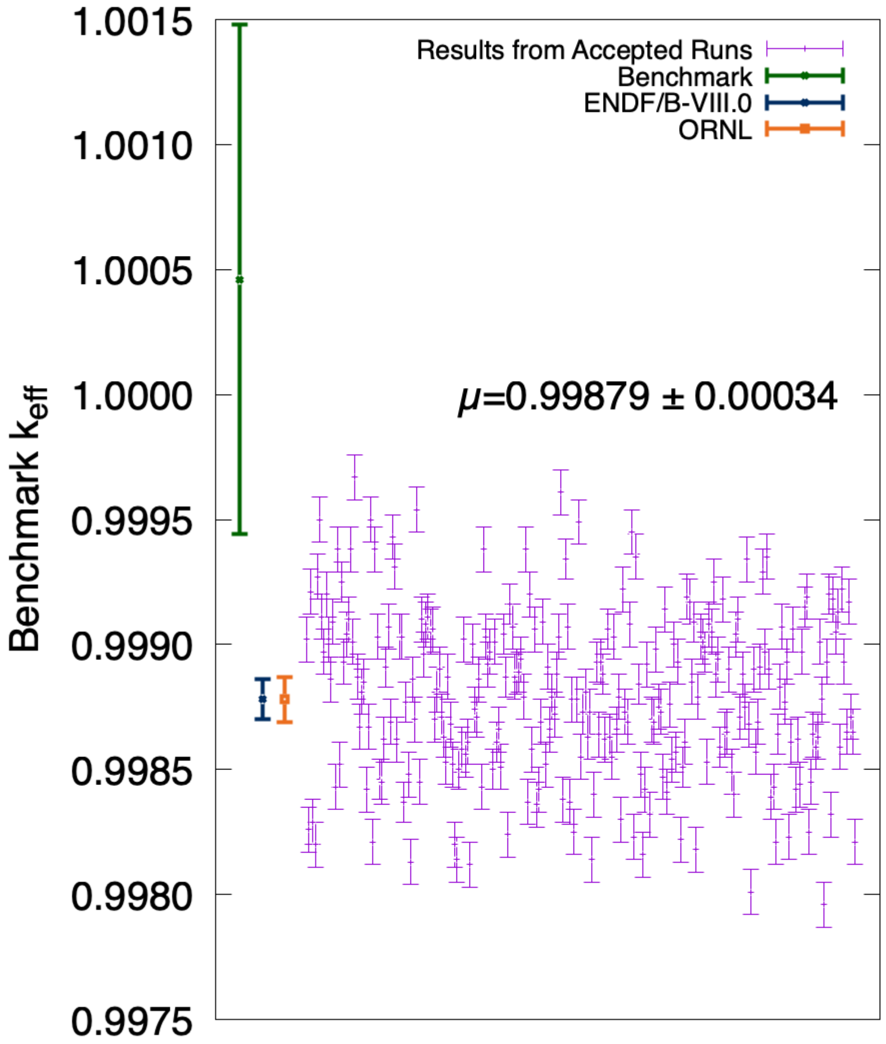
Uncertainty Quantification – Benchmarks

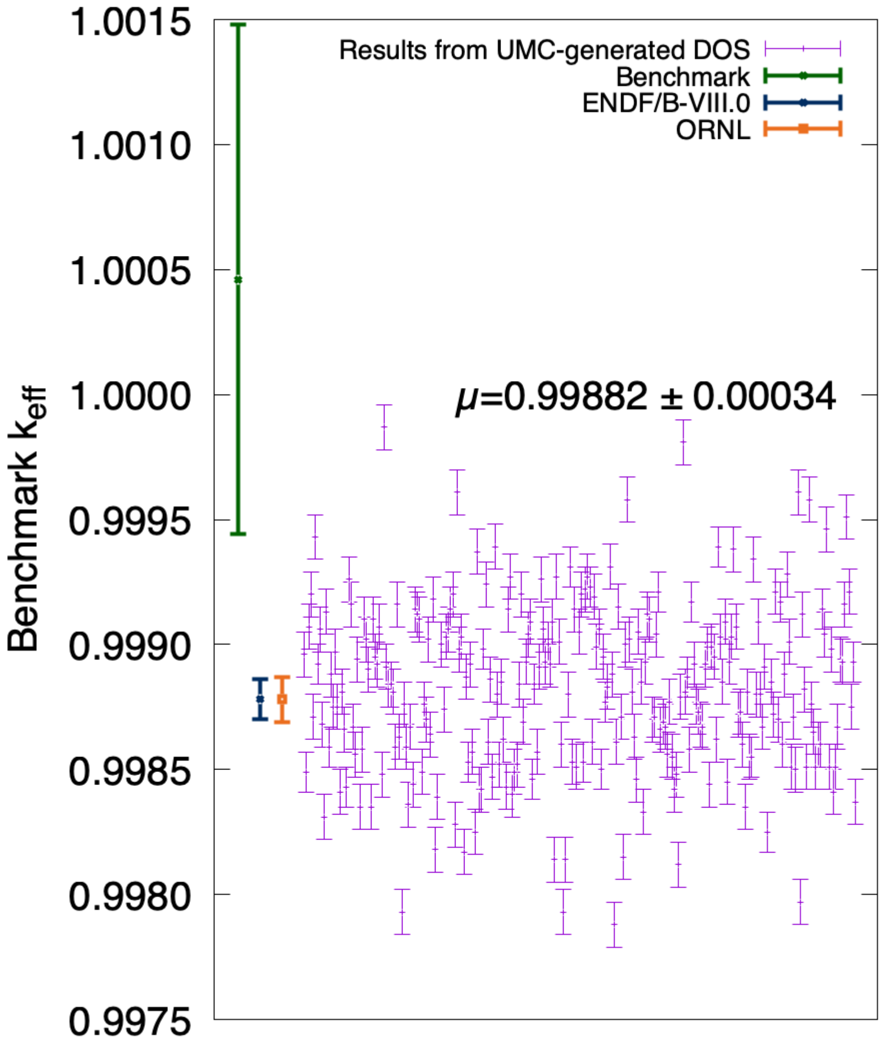
- Compared averaged $S(\alpha,\beta)$ against ENDF/B-VIII.0 evaluation for 3 sets of benchmarks: LCT-078 (298K), LCT-079 (300K), LCT-080 (298K)
  - Benchmarks were chosen because they are all at temperatures close to the experimental temperature (300K) and are well characterized
- Need to ensure covariance matrix reproduces similar spread to those generated by ensemble of Monte Carlo runs
- Covariance matrix of pDOS used to generate 250 pDOS

Uncertainty Quantification – LCT-078, -079, & -080 Validation



Uncertainty Quantification – LCT-079-005 Validation





Conclusions

- Nuclear data is used in a wide variety of application
- Experimental data necessary for both evaluation and validation of thermal scattering libraries
- Still work to be done regarding uncertainty propagation
  - Storing covariances
  - Propagating 2-D covariance information

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Questions?