Thermal Neutron Scattering Evaluation Methods Development at ORNL

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Overview

- Thermal Scattering Law Covariance Methodology
  - Overview
    - Light Water
    - Framework Theory
  - Results and Validation
    - Differential Data
    - Integral Benchmarks
- Temperature-Dependence of Polyethylene Thermal Scattering law
- Conclusions

TSL Covariance - Overview

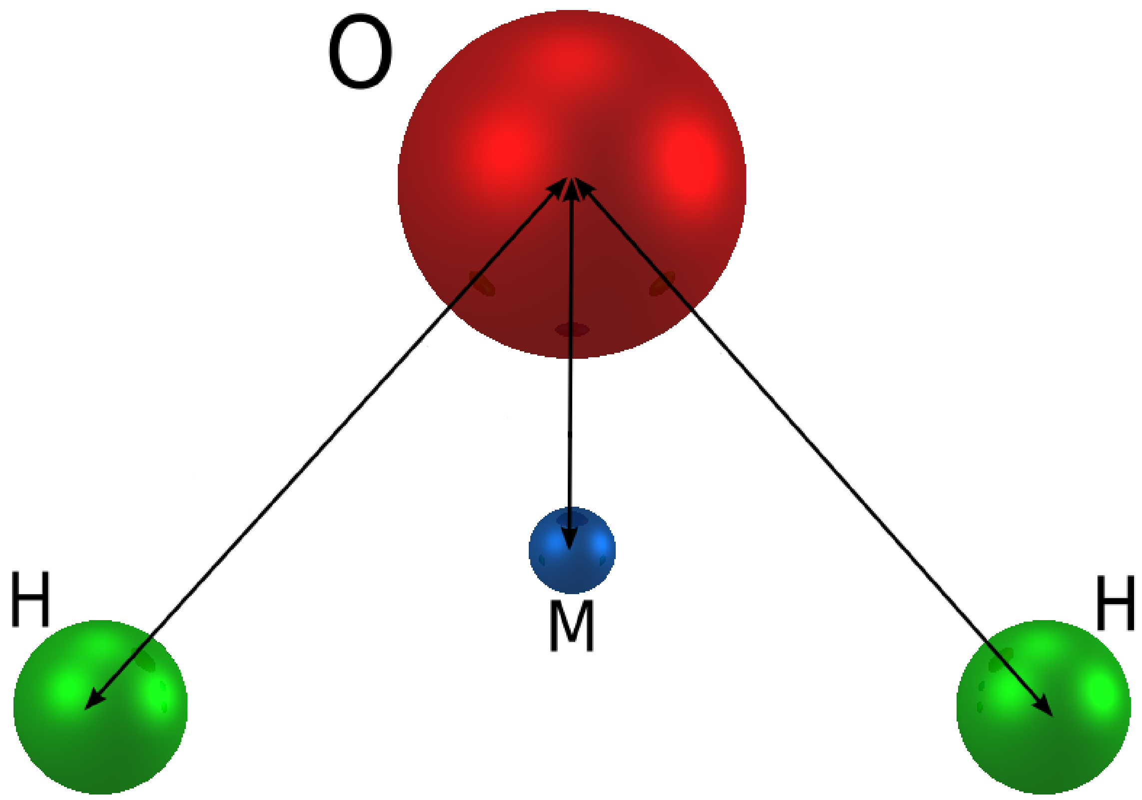
- Goal was to develop a procedure to generate covariance matrix for $S(\alpha, \beta)$ data that incorporated both computational simulations and experimental data
- Experimental data and computer simulation fit is achieved using the Unified Monte Carlo (UMC) method [1]
- Framework is material & simulation method independent
- Demonstrated using light water

TSL Covariance - Previous Work

- Active area of research, as evident by:
  - WPEC Subgroup 44 (Vlad’s talk)
  - Monte Carlo perturbations of phonon density of states [2]
  - Generalized least-squares uncertainty quantification of LEAPR [3] and molecular dynamics parameters [4] to data

TSL Covariance - Light Water

- 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



TSL Covariance - Simulation

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

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

- 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

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TSL Covariance - Simulation

- 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

TSL Covariance - UMC

- Results fit to data using UMC method, where simulations are assigned a weight used to calculate mean and covariance values
- Weight for the k^{th} simulation is calculated by:

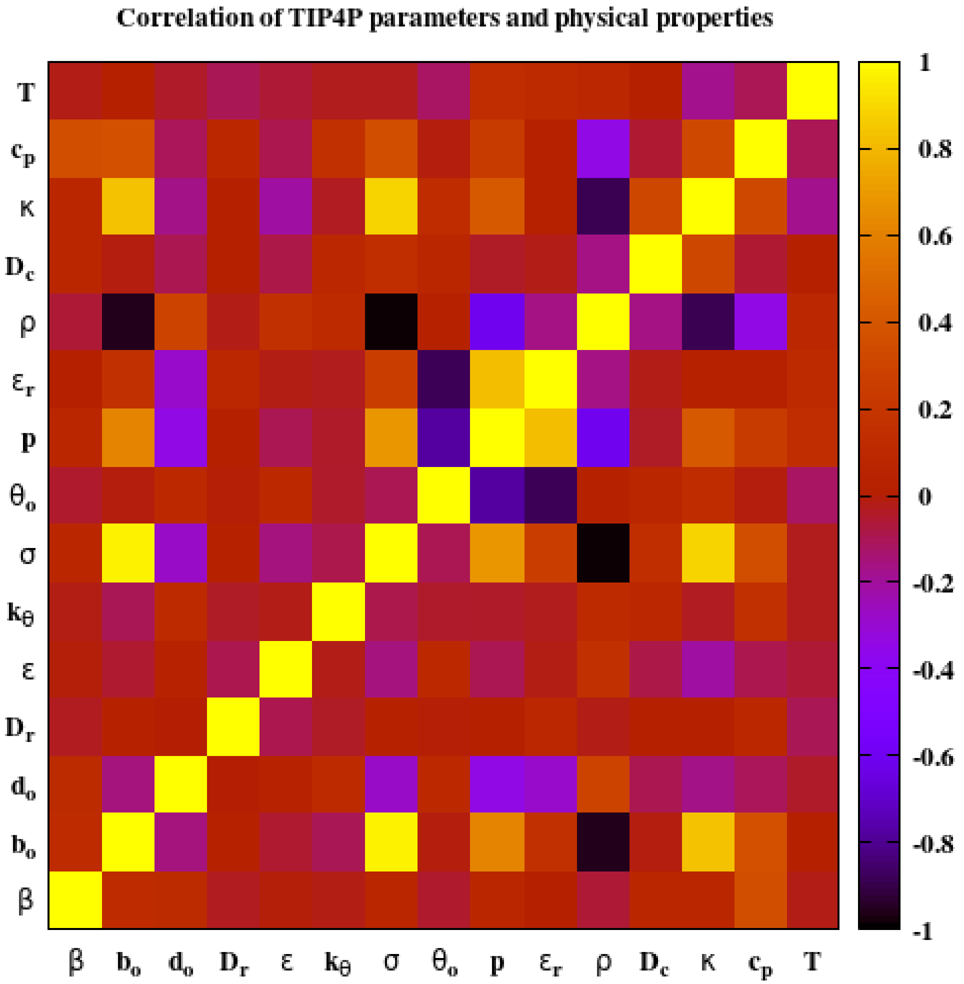
$$\omega_k = \exp\left\{-\frac{1}{2}\left[(\mathbf{y_k} - \mathbf{y_E})^\intercal \cdot \mathbf{V_E}^{-1} \cdot (\mathbf{y_k} - \mathbf{y_E})\right]\right\}$$

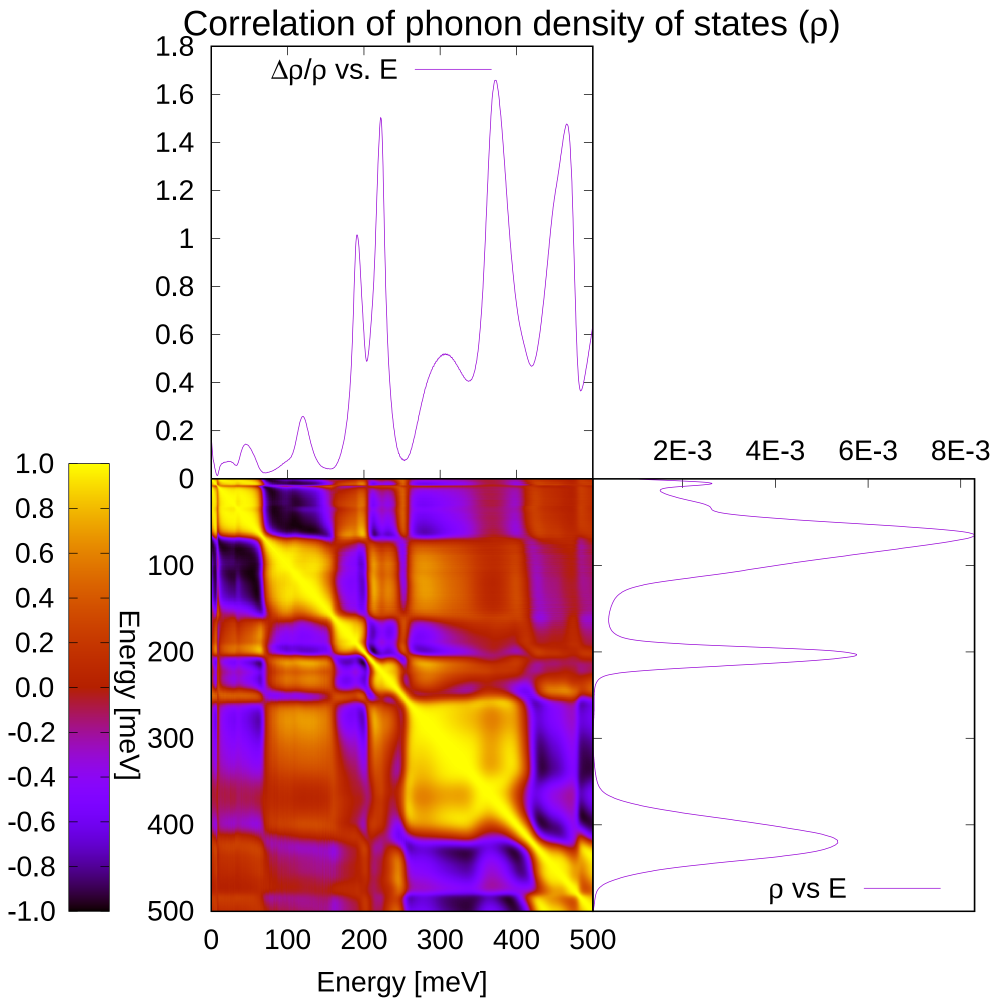
$$\langle x_i \rangle = \lim_{M\to\infty} \frac{\sum_{k=1}^{M} x_{ik}\omega_k}{\sum_{k=1}^{M} \omega_k}, \qquad \langle \mathbf{V} \rangle_{i,j} = \lim_{M\to\infty} \frac{\sum_{k=1}^{M} x_{ik}x_{jk}\omega_k}{\sum_{k=1}^{M} \omega_k} - \langle x_i \rangle \langle x_j \rangle$$

TSL Covariance - UMC

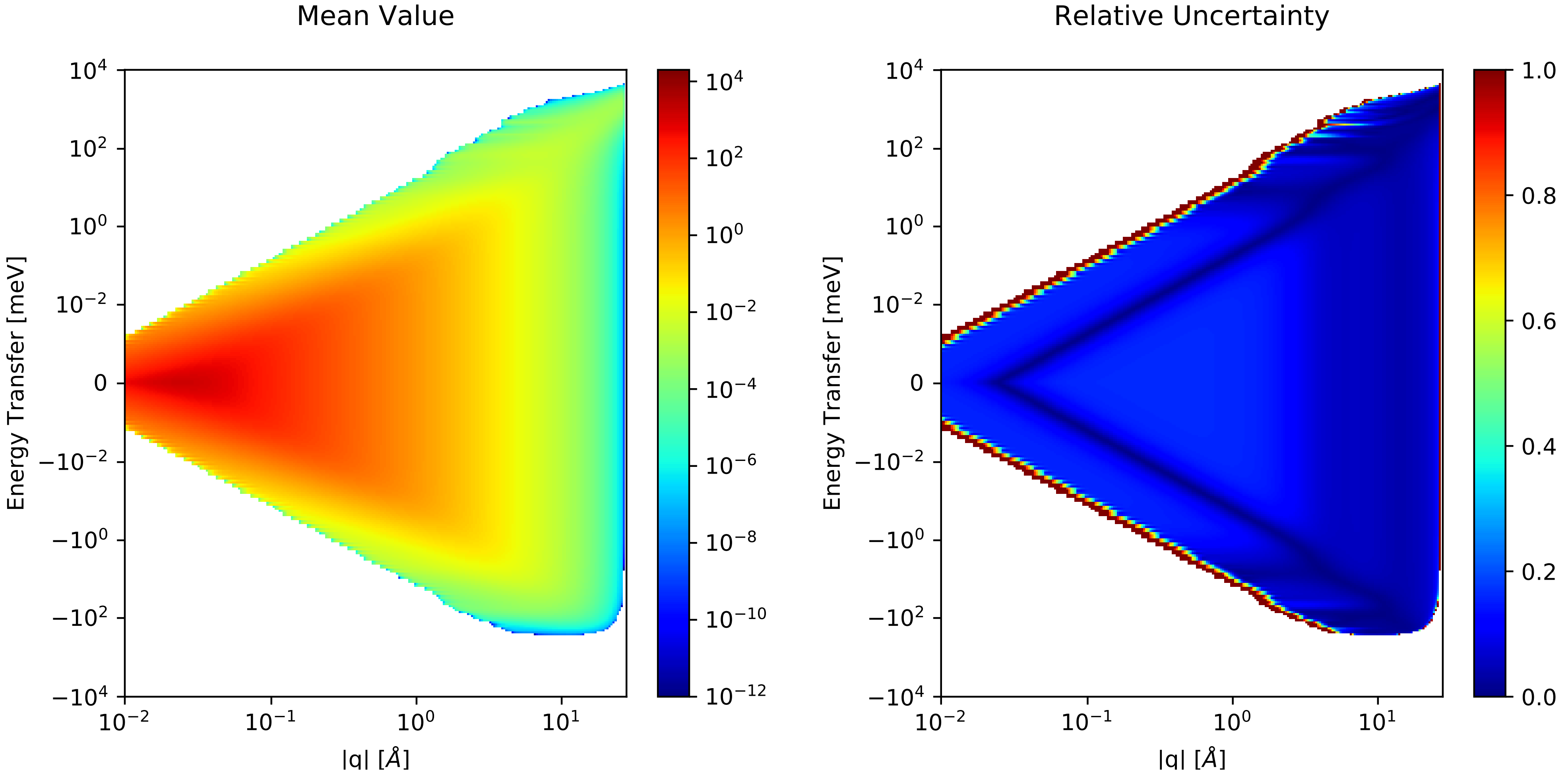
- UMC weights then used to calculate mean values and covariance matrices of:
  - TIP4P/2005f parameters
  - Thermophysical properties
  - pDOS
  - Double differential & total scattering cross section
- Mean values and variance of thermal scattering law $S(\alpha, \beta)$ also generated
  - Methods of storing $S(\alpha, \beta)$ covariance matrix not studied here

TSL Covariance - Correlation Matrices

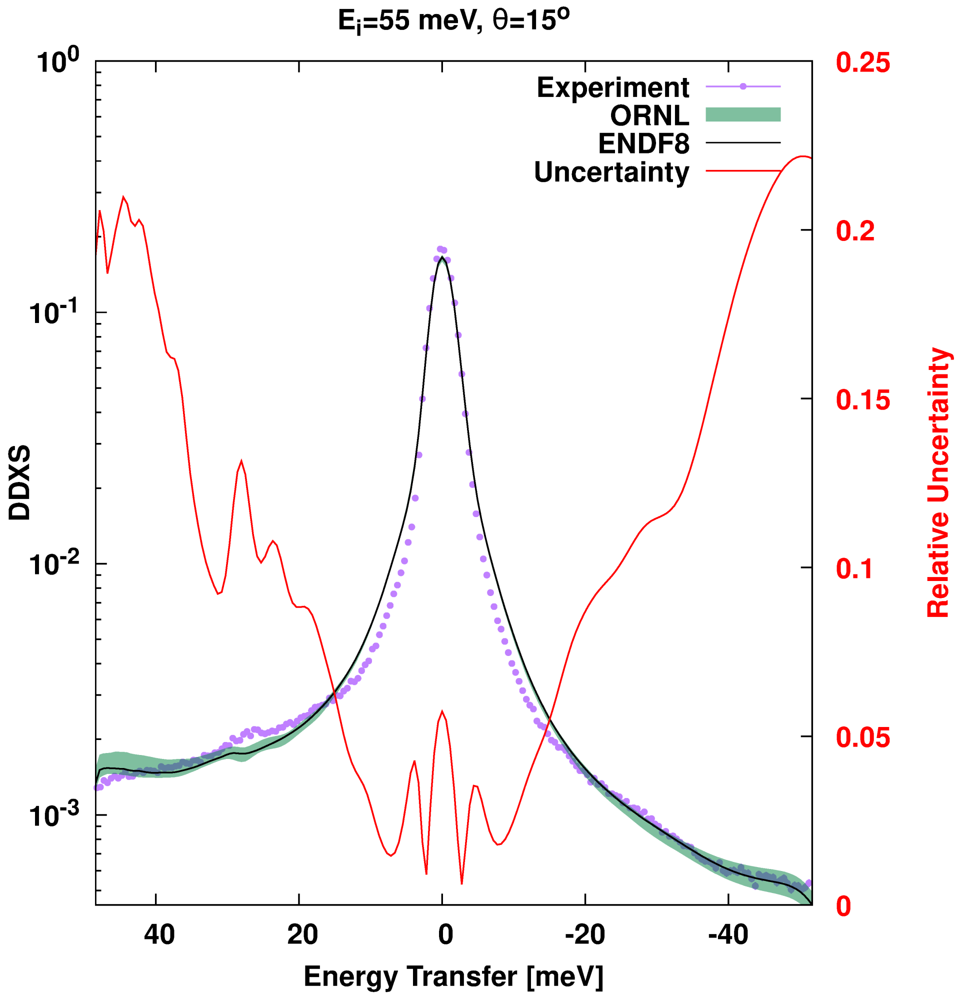


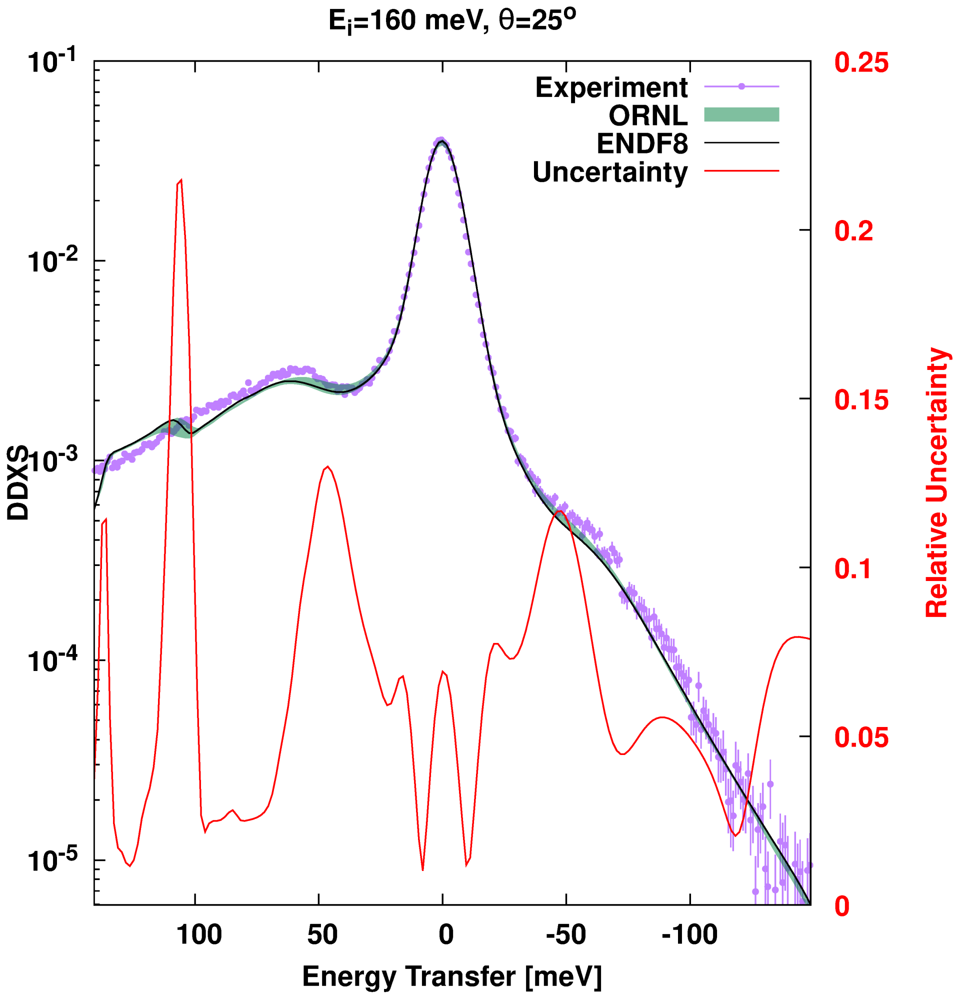


TSL Covariance - $S(q, E)$ & Uncertainties

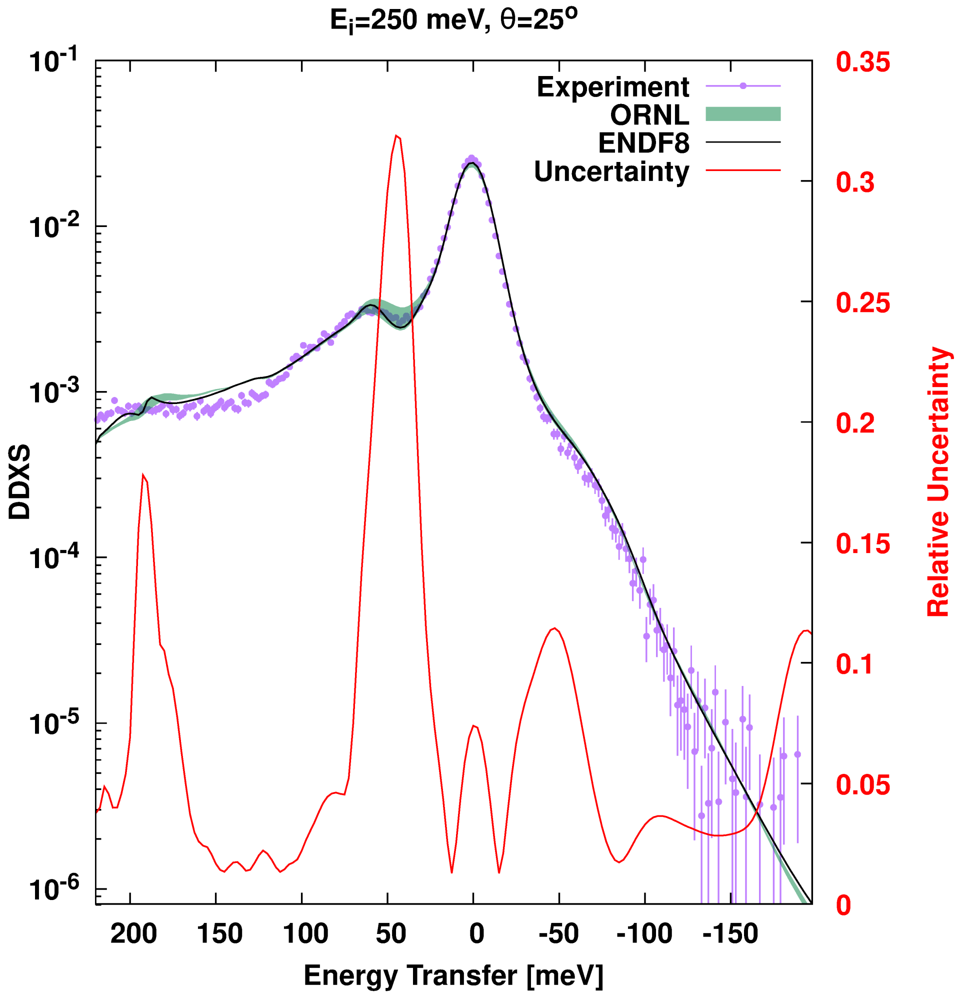


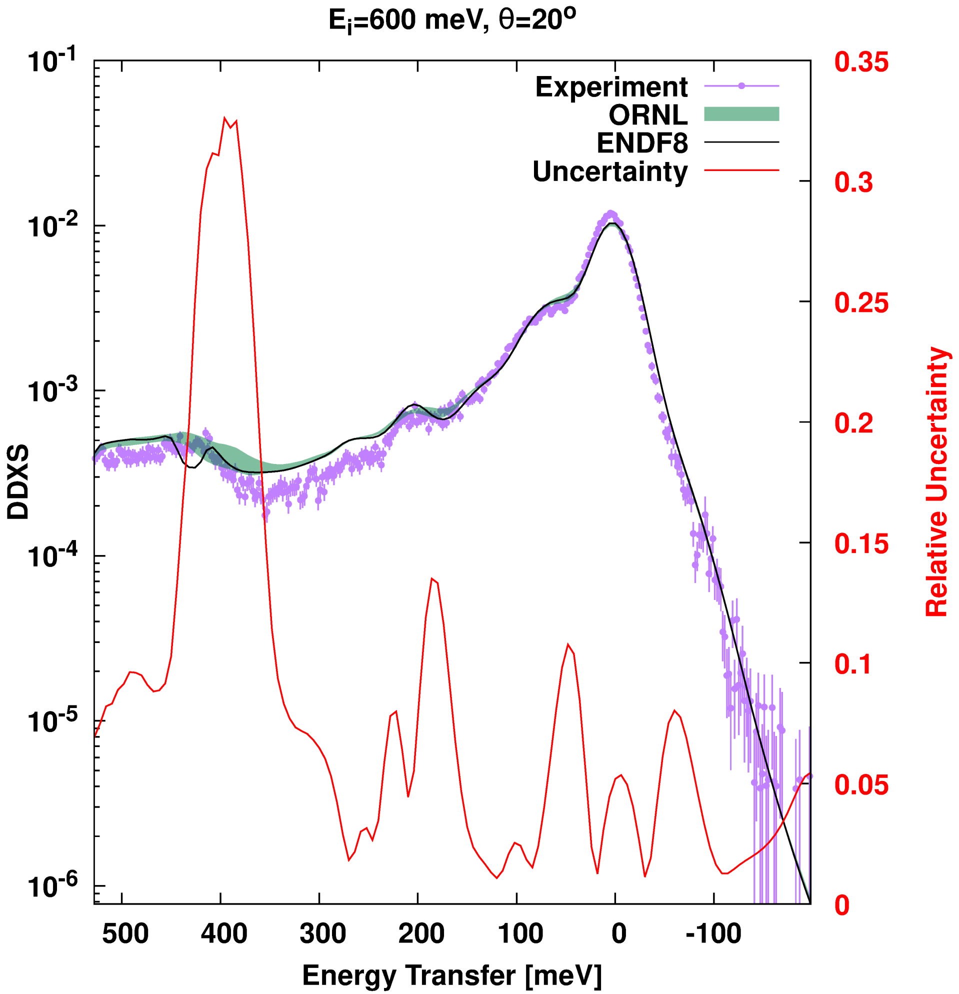
TSL Covariance - DDXS



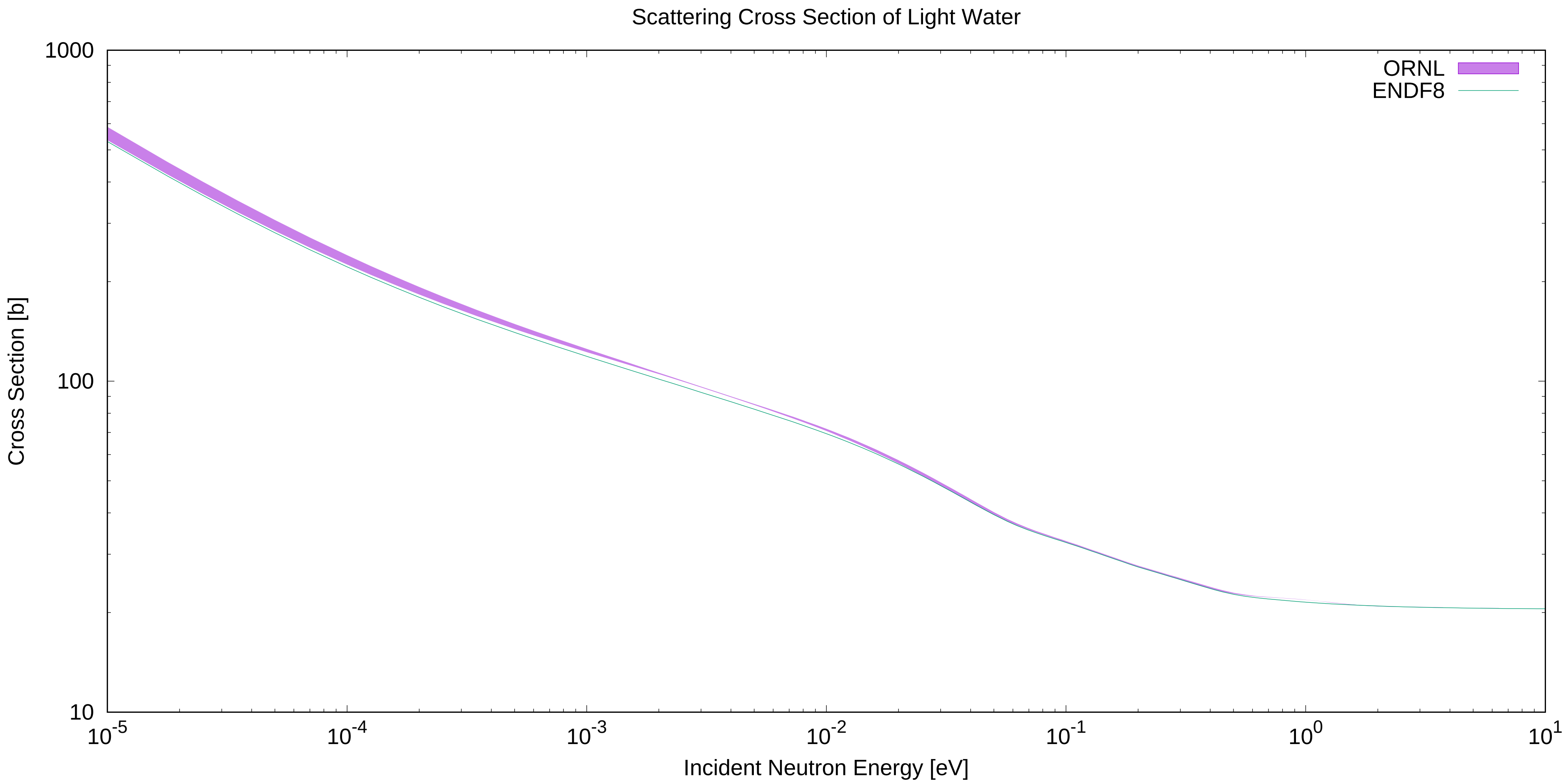


TSL Covariance - DDXS (cont.)





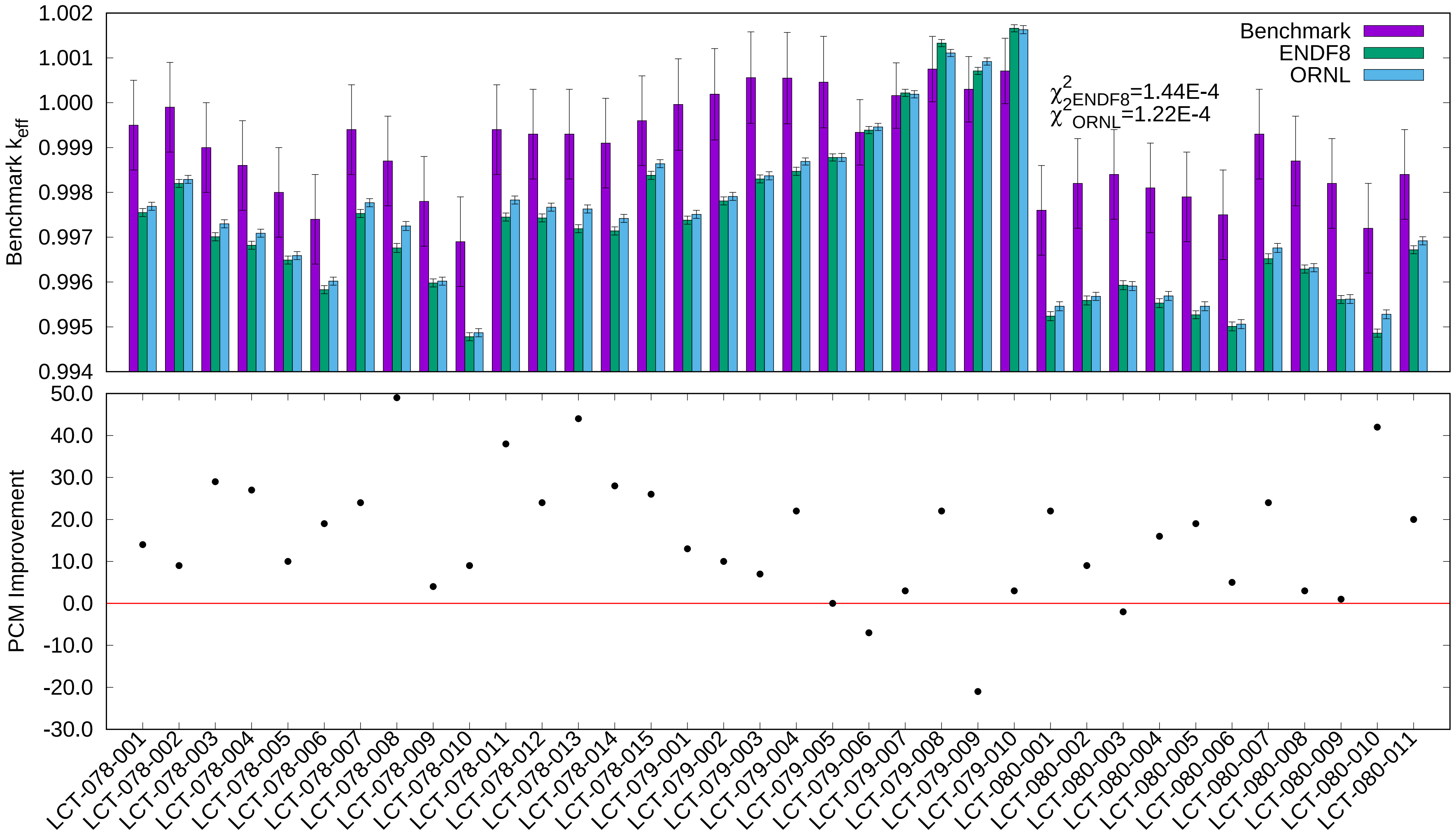
TSL Covariance - Total XS



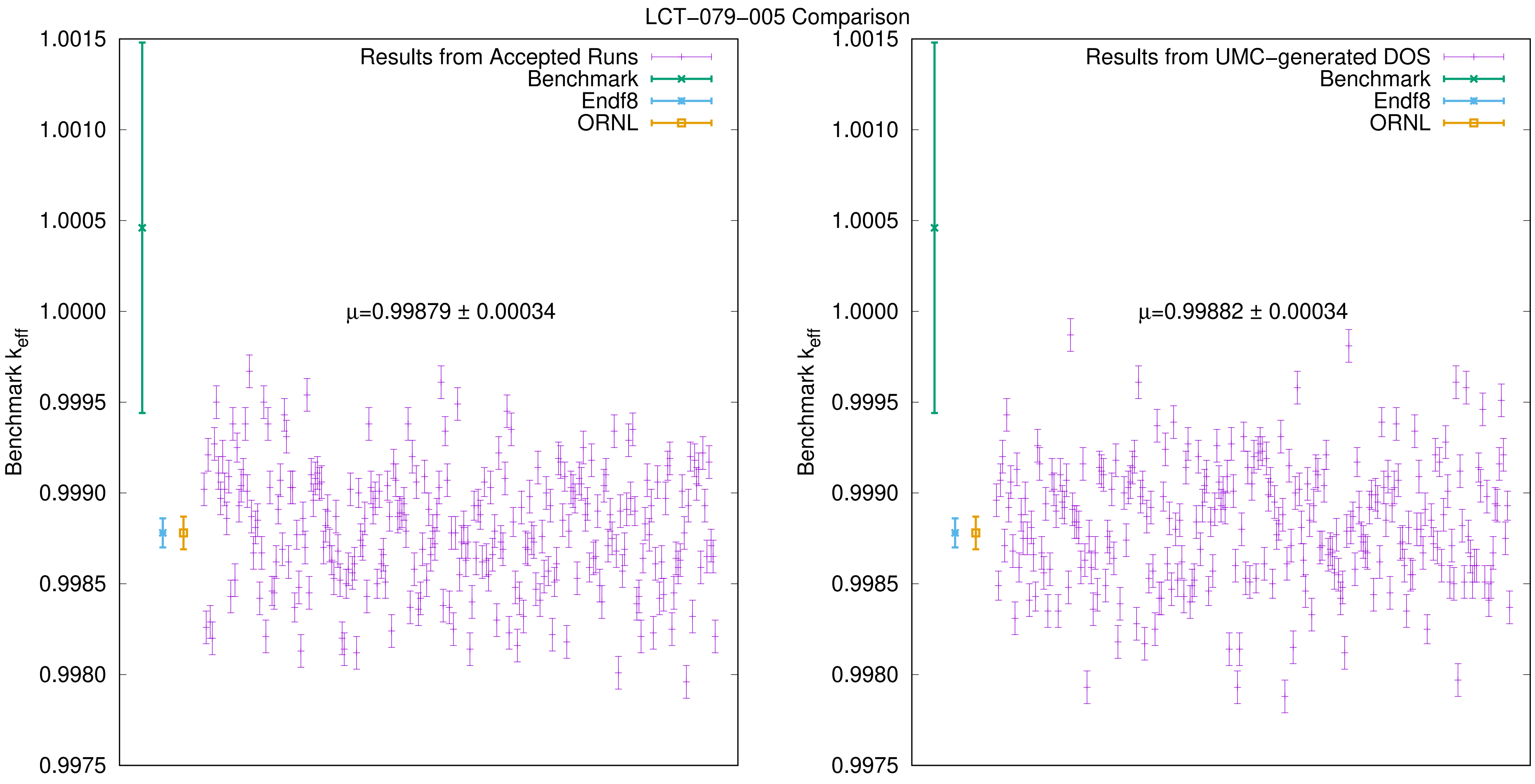
TSL Covariance - Benchmarks

- 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

TSL Covariance - LCT-078, -079, & -080 Validation



TSL Covariance - LCT-079-005 Validation

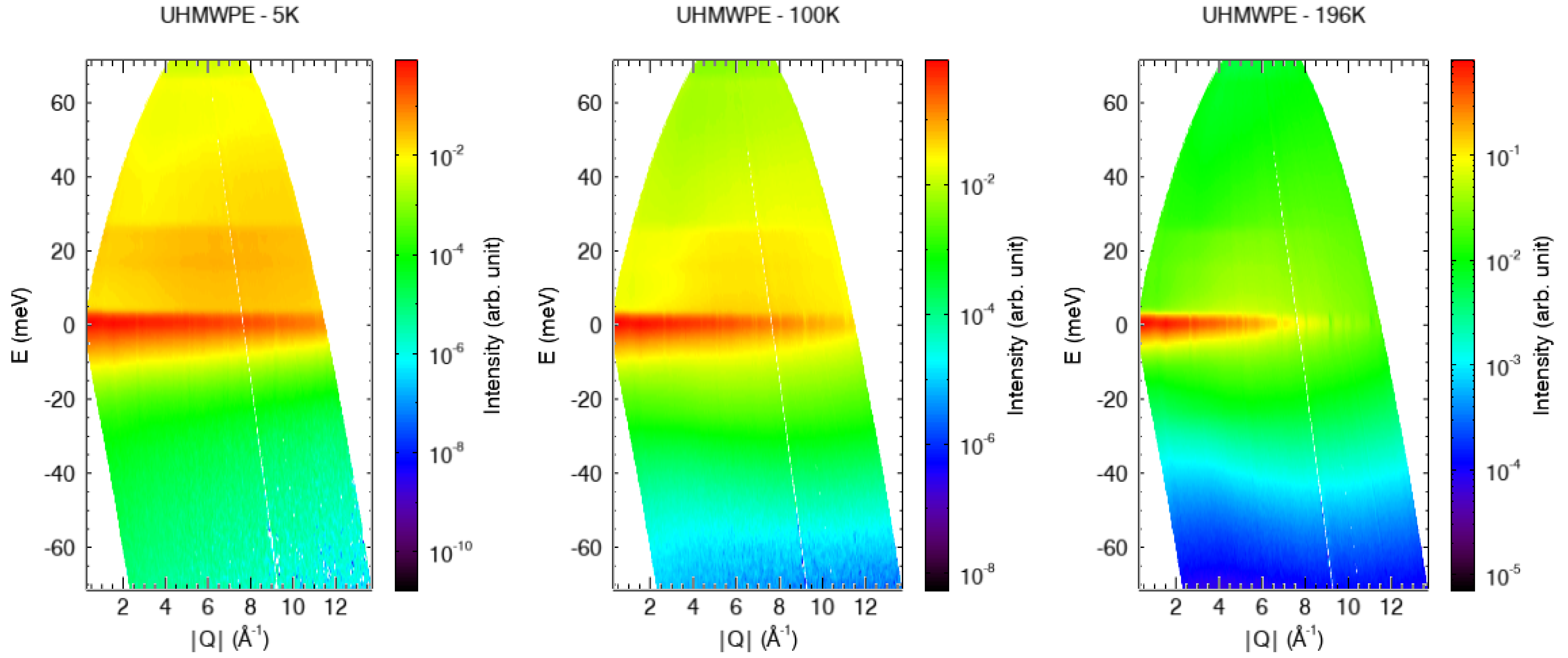


- Error bars represent either benchmark or Monte Carlo uncertainty

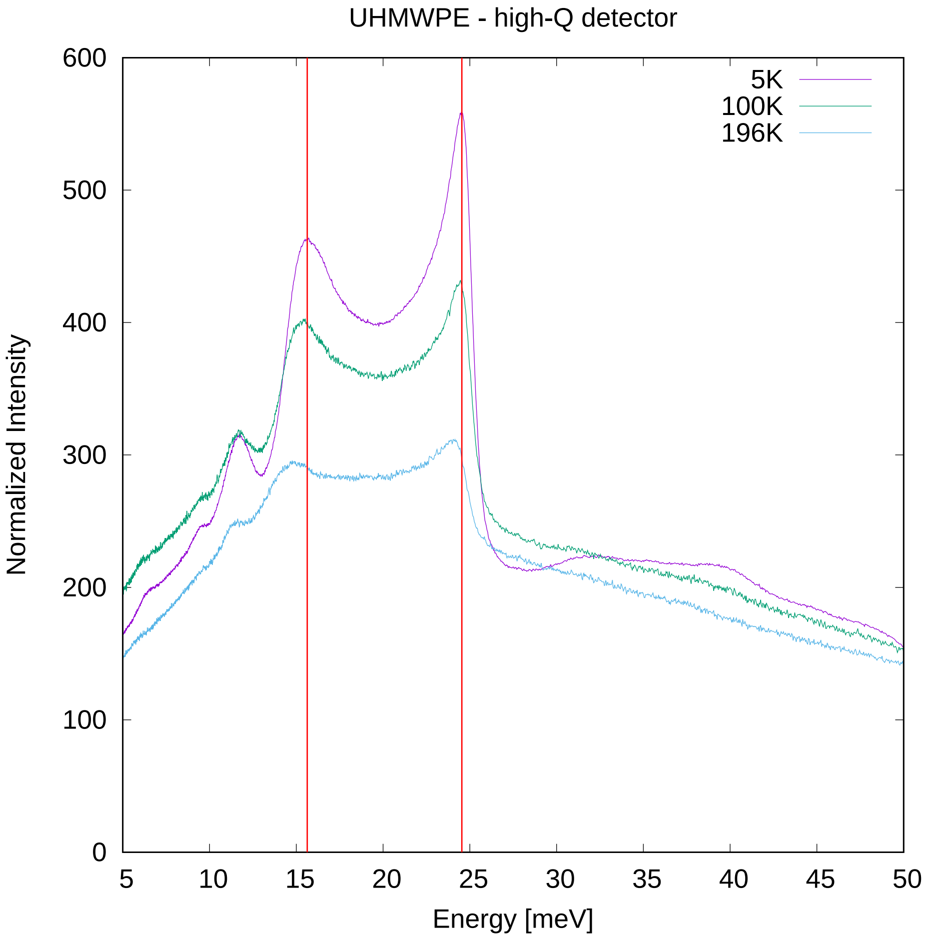
Temperature Dependence of Polyethylene

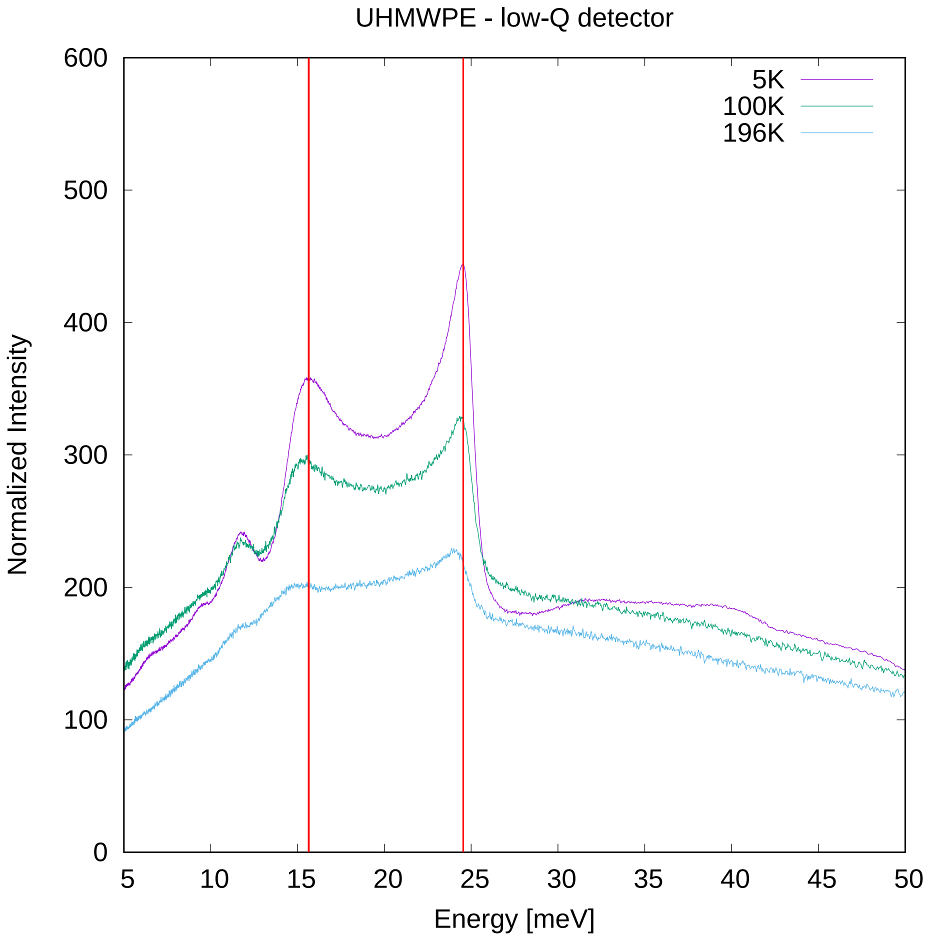
- Aim is to determine if there are any temperature-dependent effects of polyethylene not present in the current ENDF evaluation
- Carried out in collaboration with RPI as a follow up on [6]
- Experiments carried out at 2 different beamlines
  - ARCS: Direct geometry configuration at T=5, 100, 196, 268, 295, 313K
  - VISION: Indirect geometry at T=5, 20, 40, 60, 77, 100, 120, 140, 160, 180, 196, 220, 233, 240, 263, 283, 293.6, 300, 303, 313, 323, 333, 343, 350K

Temperature Dependence of Polyethylene - ARCS



Temperature Dependence of Polyethylene - VISION





- Shifted peaks represent changes in lattice parameters

Conclusions

- Methodology developed for generalized generation of covariances for thermal scattering files
- Framework subsequently tested on light water by generating co-variance of pDOS with encouraging results
- Preliminary results suggest temperature effects of polyethylene may warrant further investigation

Acknowledgments

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Questions?

Auxiliary Slides - TIP4P/2005f Parameters Results

Parameter	UMC mean value	UMC uncertainty	Published Value	Relative Error
β	2.291E+01	6.672E-01 (2.91%)	2.287E+01	0.15%
b_0	9.441E-02	2.707E-03 (2.87%)	9.419E-02	0.23%
d_0	1.545E-02	4.664E-04 (3.02%)	1.546E-02	0.08%
D_r	4.34152E+02	1.23567E+01 (2.85%)	4.32581E+02	0.36%
ε	7.717E-01	2.141E-02 (2.77%)	7.749E-01	0.42%
K_θ	3.6695E+02	1.0209E+01 (2.78%)	3.6781E+02	0.23%
σ	3.1679E-01	7.6056E-03 (2.40%)	3.1644E-01	0.11%
θ_0	1.068E+02	3.087E+00 (2.89%)	1.074E+02	0.54%

- Precision of each value chosen to match precision of published value

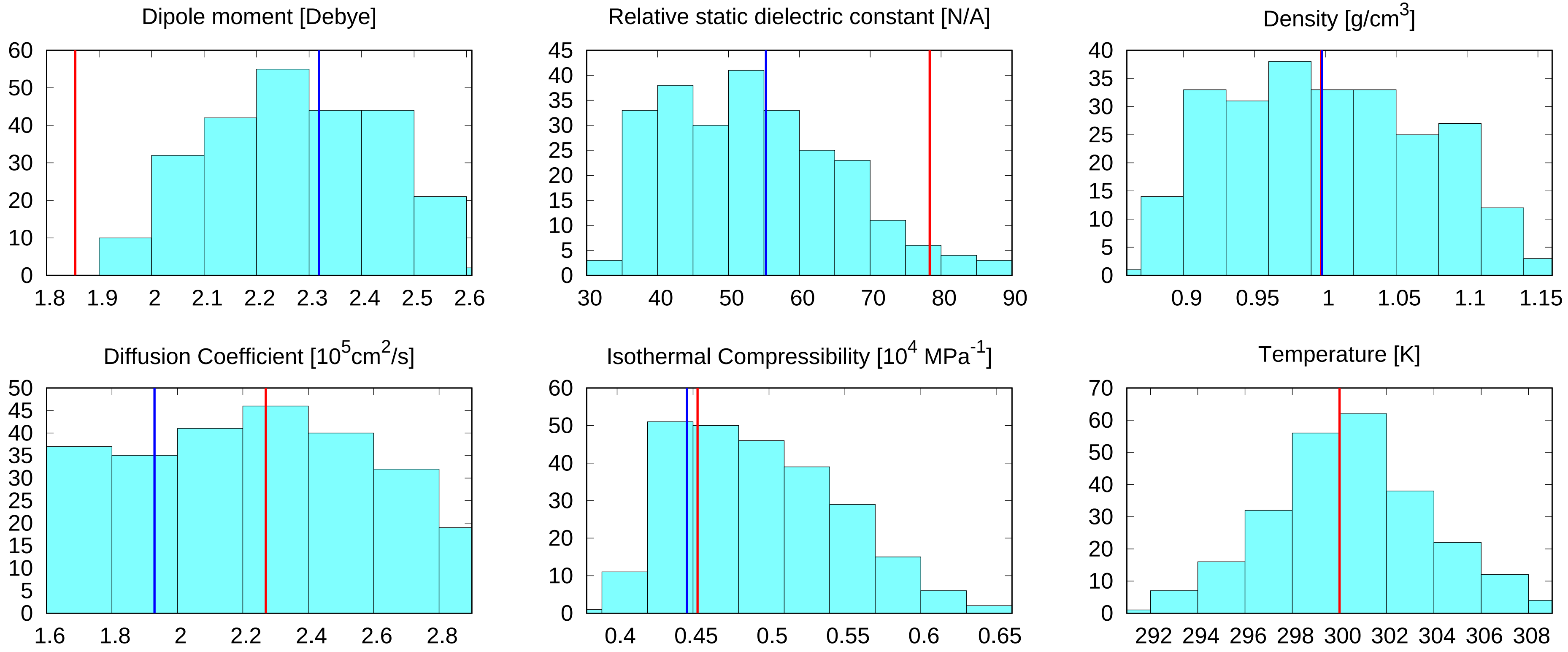
Auxiliary Slides - Thermophysical Properties Results

Property	UMC mean value	UMC uncertainty	Published Values	Exp. Value
Dipole Moment (p)	2.299E+00	1.597E-01 (6.949%)	2.319E+00	1.8546E+00
Dielectric Constant (ε_r)	5.518E+01	1.230E+01 (22.29%)	5.53E+00	7.84E+01
Density (ρ)	9.961E-01	7.165E-02 (7.193%)	9.977E-01	9.97E-01
Diffusion Coefficient (D_c)	2.241E+00	3.644E-01 (16.26%)	1.93E+00	2.27E+00
Isothermal Compressability (κ)	4.953E-01	5.402E-02 (10.91%)	4.46E-01	4.95E-01
Heat Capacity (c_p)	7.080E+01	1.471E+00 (2.078%)	N/A	7.441E+01

Auxiliary Slides - Correlation of Thermophysical Properties

Frequency plots of thermophysical properties

Blue lines are reference values from [5], Red lines are experimental values



Auxiliary Slides - Total XS with experimental data