Thermal Neutron Scattering Measurements and Analysis

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Overview

- Performed thermal scattering experiments (at various temperatures and different instruments – listed next page) at Spallation Neutron Source for water, high density polyethylene (HDPE), and quartz (SiO_2).

- Comparative and integrative study of MCNP (including the evaluations), molecular dynamics simulations, and thermal neutron experiments.
  - ✓ Comparative study between thermal scattering measurements and MCNP (evaluations).
  - ✓ Comparative study between atomistic simulations and measurements.

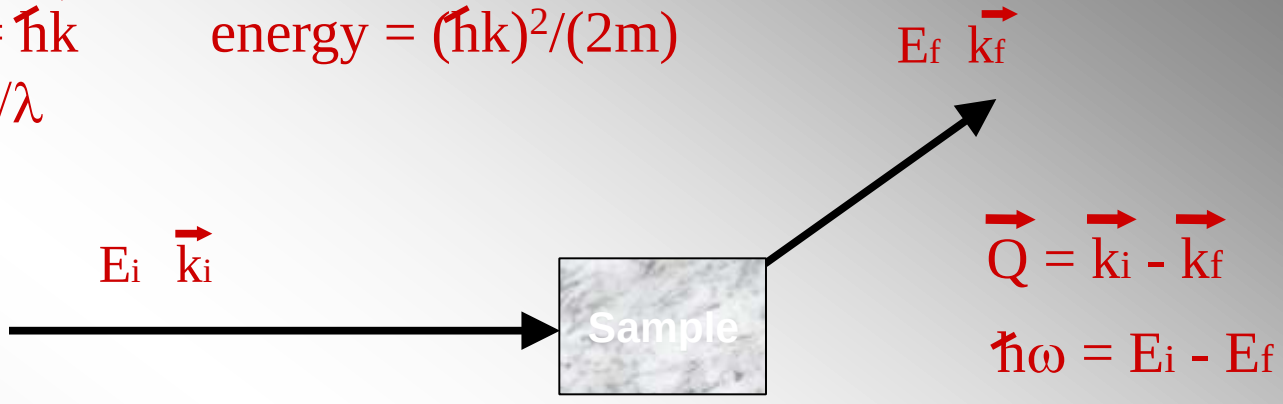
- Integration, suggestions, and improvements of nuclear data based on current study.

Thermal Scattering Measurements

Materials	The instruments utilized in Spallation Neutron Source (SNS)	
	Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA)	Wide Angular-Range Chopper Spectrometer (ARCS)
Light Water (H_2O)	E_I: 55, 160, 250, 600, 1000, 3000, 5000 meV Ω: 3-58° in 1° increments Temp = 300 K	-
Polyethylene (CH_2)	E_I: 55, 160, 250, 600, 1000, 2000 meV Ω: 3-58° in 1° increments Temp = 300 K	E_I: 50, 100, 250, 700 meV Ω: 3-125° in 1° increments Temp = 5, 295 K

Thermal Neutron Scattering Geometry

momentum = $\hbar\vec{k}$ $\quad k=2\pi/\lambda$ $\qquad$ energy = $(\hbar k)^2/(2m)$



Measure the number of scattered neutrons as a function of Q and ω

=> $S(Q,\omega)$ (the scattering function for inelastic scattering)

$I(Q)$ (elastic scattering, $E_i=E_f$, $\omega=0$)

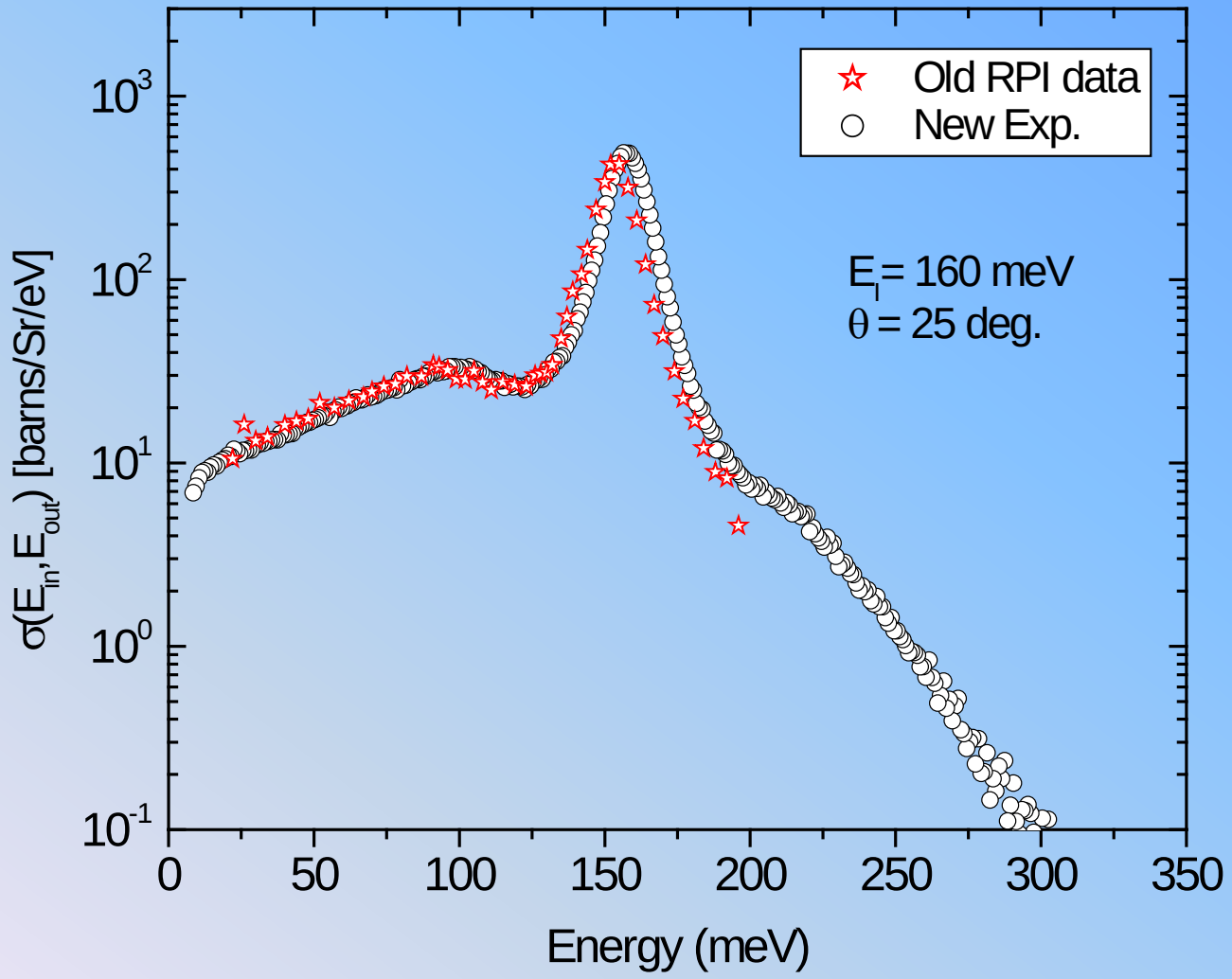
depends ONLY on the sample

$$\frac{d^2\sigma_H}{d\Omega d\omega} = 2N\frac{\sigma_H}{4\pi}\frac{k_f}{k_i}S_{inc}(Q,\omega)\otimes R(\omega)$$

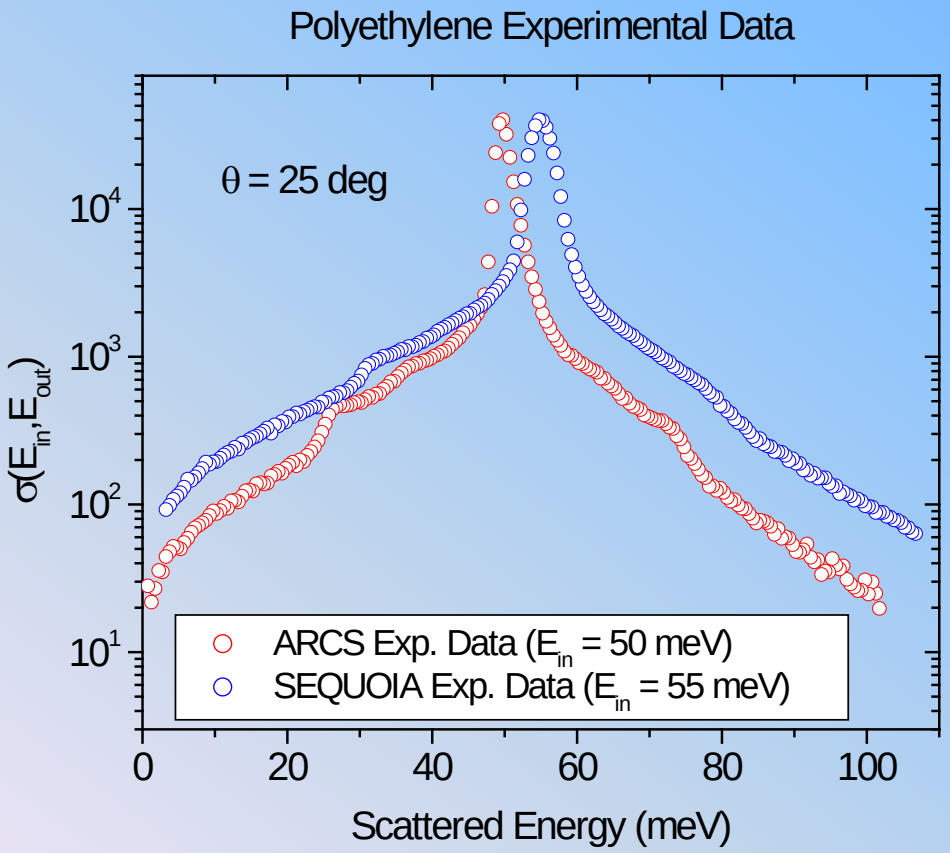
In our study:

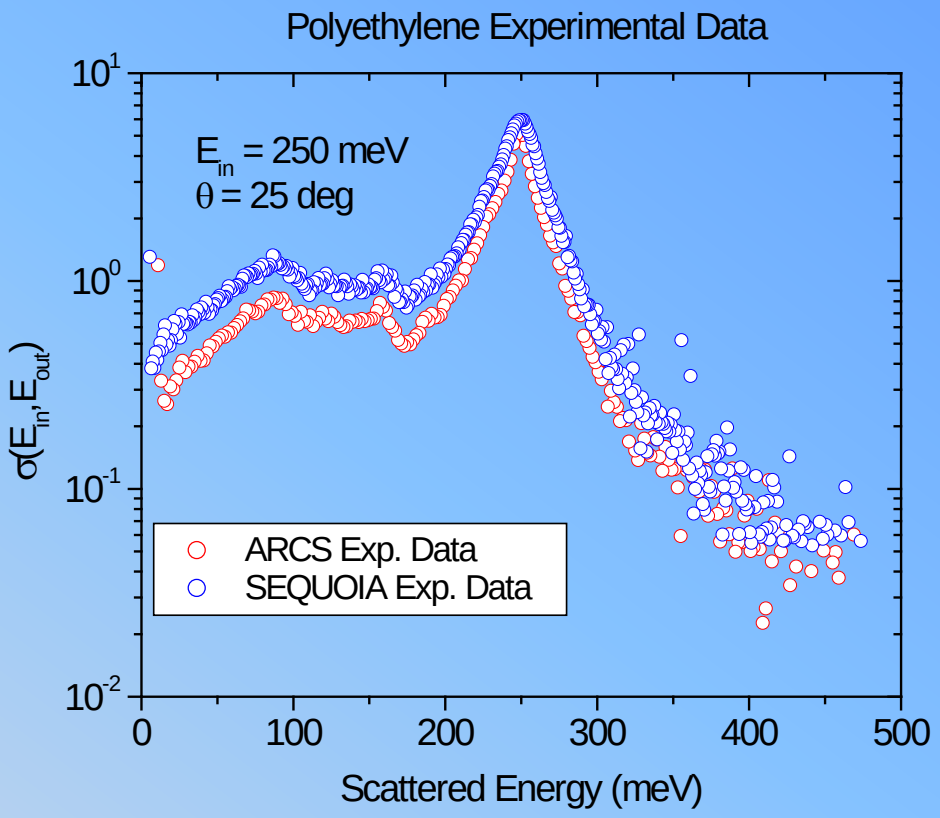
- Neutron Scattering experimental plot: Double differential cross section (instead of S(Q,ω)) vs. scattered energy (E_f, instead of ω)
- Molecular Dynamics simulation: directly calculate S(Q,ω) or S(α,β)

New SEQUOIA light water data vs. old RPI data from 1969

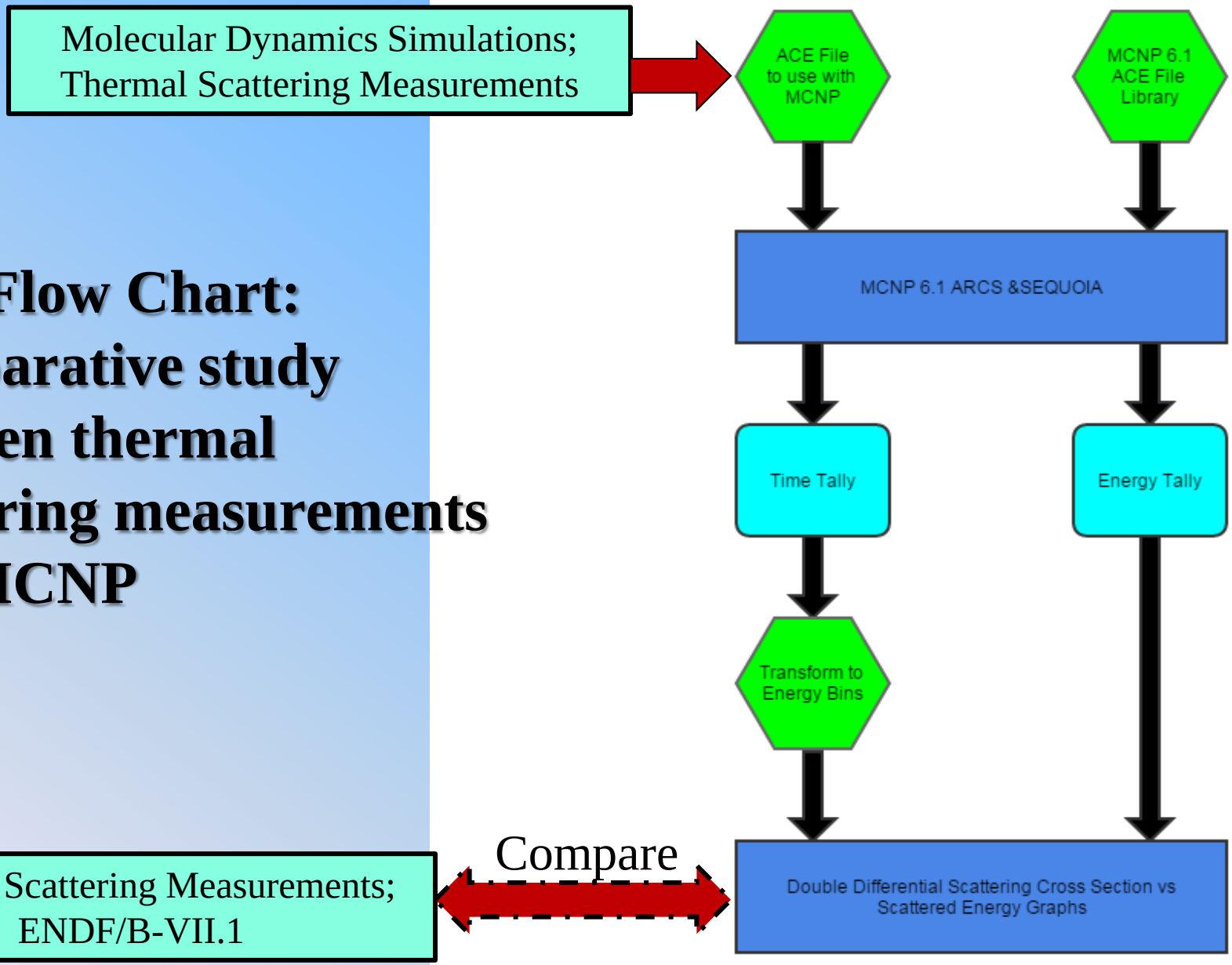


PE: SEQUOIA vs. ARCS
E_{in} = 55/50 meV, 250 meV





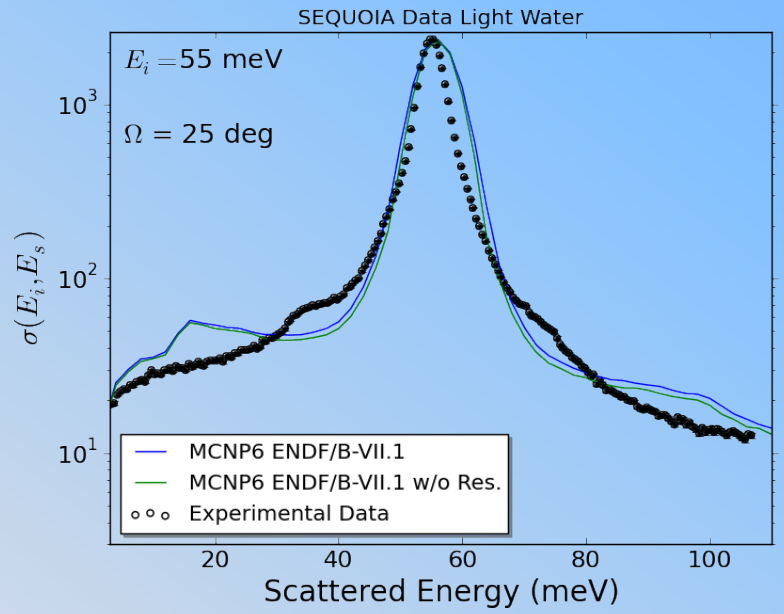
Data Flow Chart: Comparative study between thermal scattering measurements and MCNP



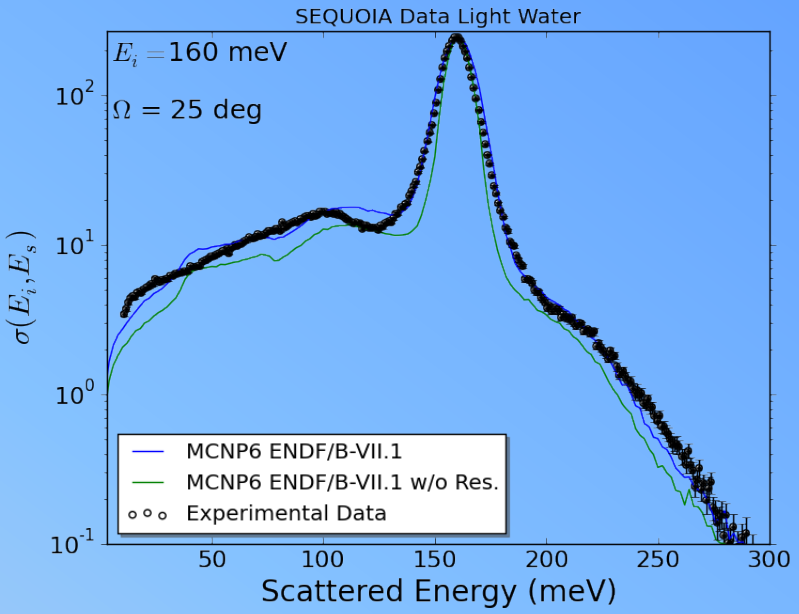
Exp/MCNP model differences and the possible issues to address those differences

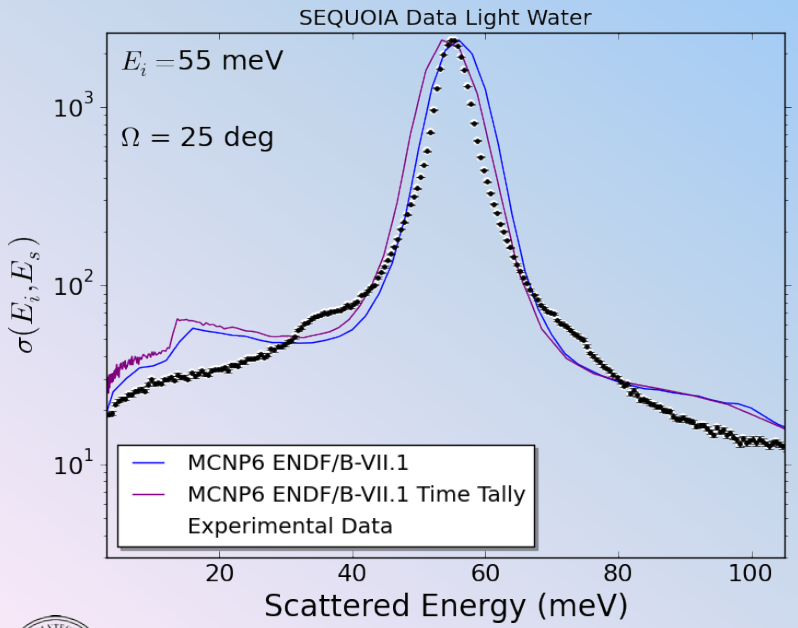
- MCNP file is an idealized version of experiment
  - Need to add spectrometer resolution: Broadening the mono-energetic neutron source to a Gaussian spectra results in a better fit of the elastic peak
  - Using a F5 energy tally to measure the DDSCS is not technically a perfect representation of the experiment: SEQUOIA and ARCS are TOF Spectrometers; changing the Tallies to F5 Time Tallies is the first step
  - Realize that the protons at the SNS do not hit the mercury target instantaneously. Proton pulse at the SNS is 1 microsecond long; this time spread is increasing the farther the neutrons have to travel; add a 1 us (100 shakes) spread to the MCNP model initial time of the neutron to compensate spread; Time and Energy Resolutions are treated as independent even though they are really coupled. Using the Proton pulse width caused the energy resolution to change as well to fit the elastic peak: $\Delta E/E \approx 1.5$ %; it agrees with the Instrument Scientist's measurement (for PE 55 meV, and specific chopper setting)
- The detailed reduction of thermal scattering experimental results of solids
  - Need to be normalized carefully based on the total cross section (ENDF)
- H_2O
  - MCNP Quasi-elastic peak is narrower than experiment
- CH_2
  - MCNP Elastic peak is a delta function while the experiment's is a near symmetrical curve
  - MCNP Inelastic regions do not match experiment very accurately

H_2O Light Water

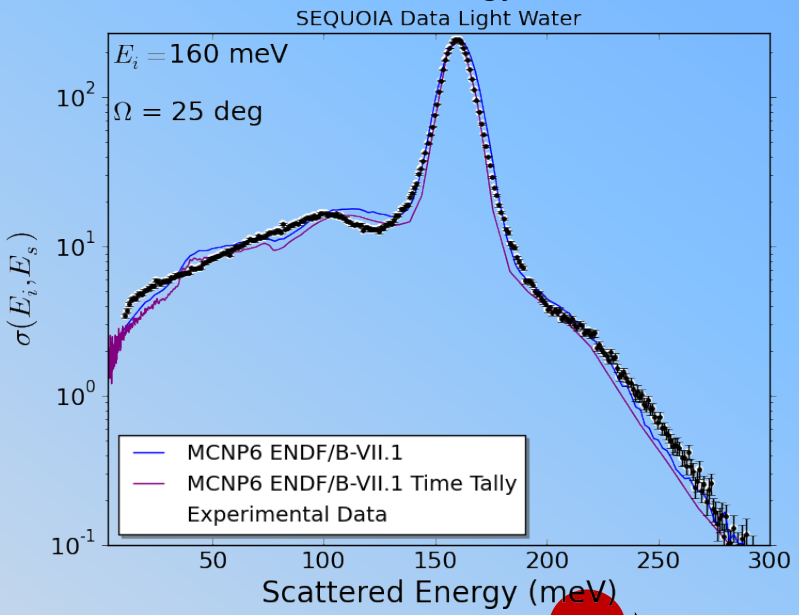


With vs. without resolution

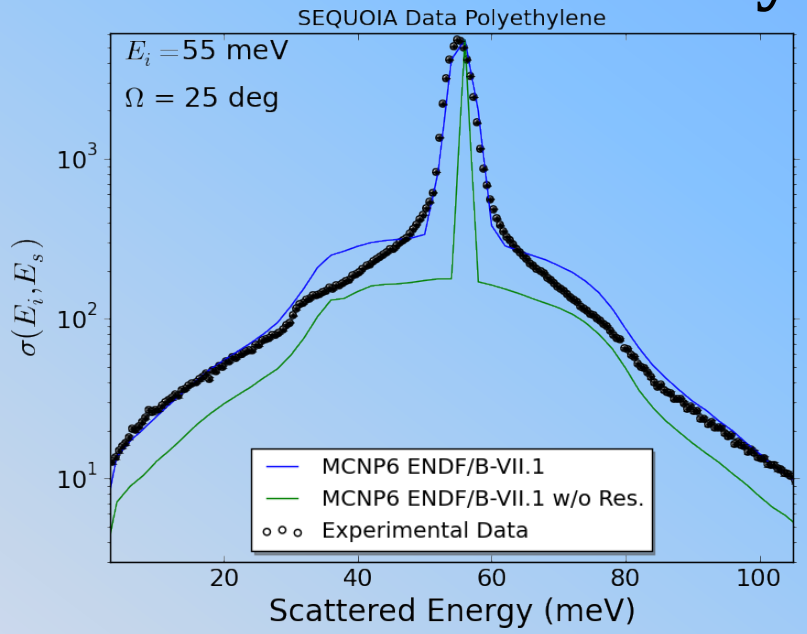




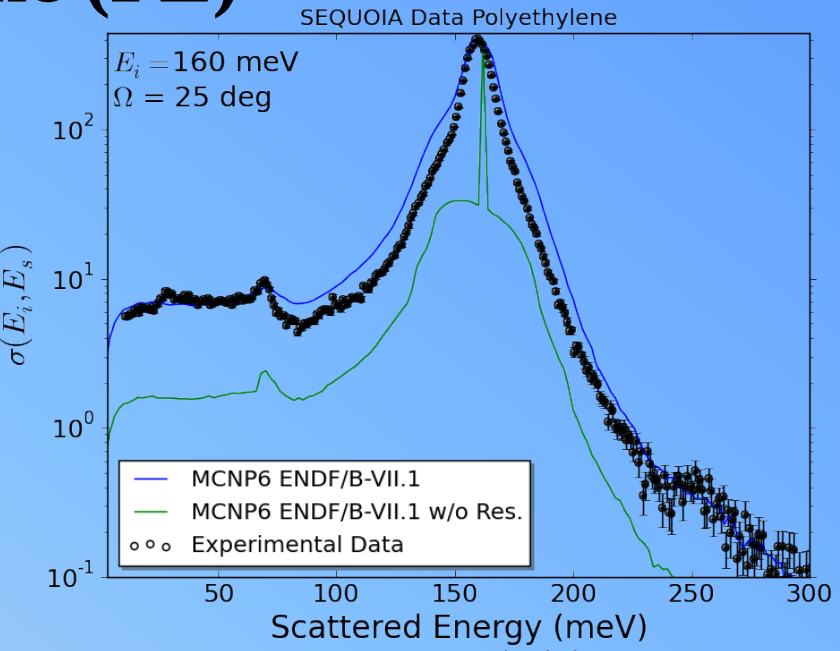
Energy vs. time tally

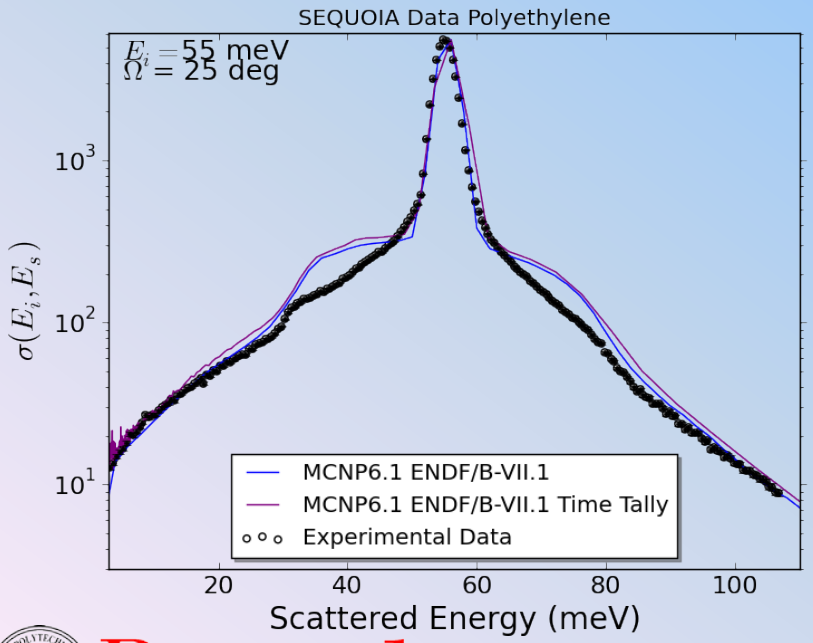


Polyethylene (PE)

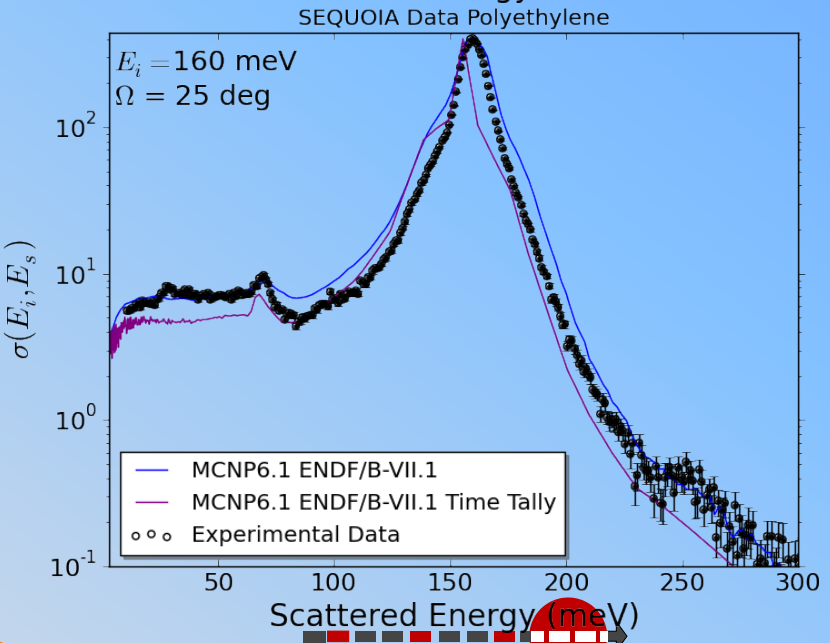


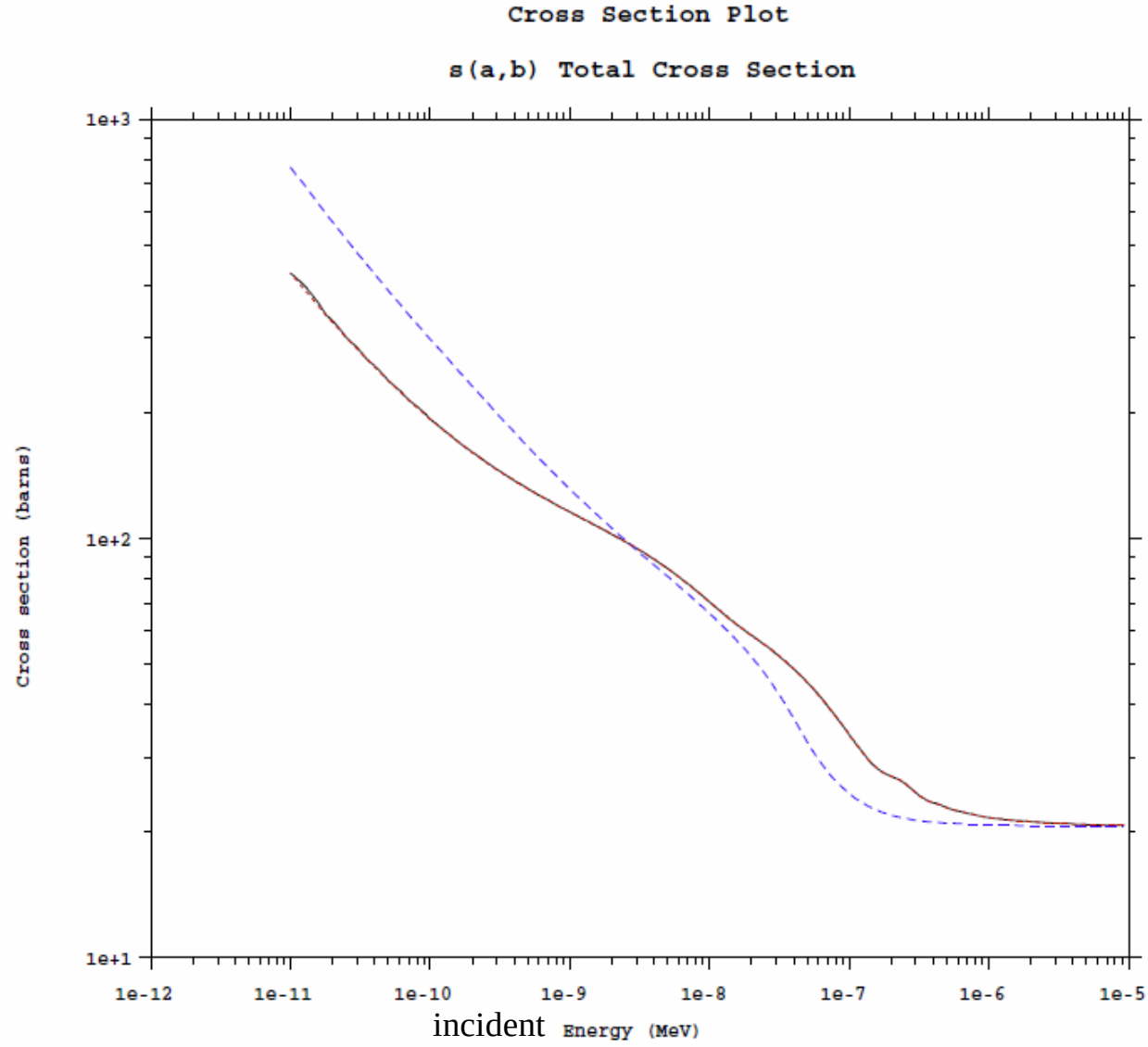
With vs. without resolution





Energy vs. time tally





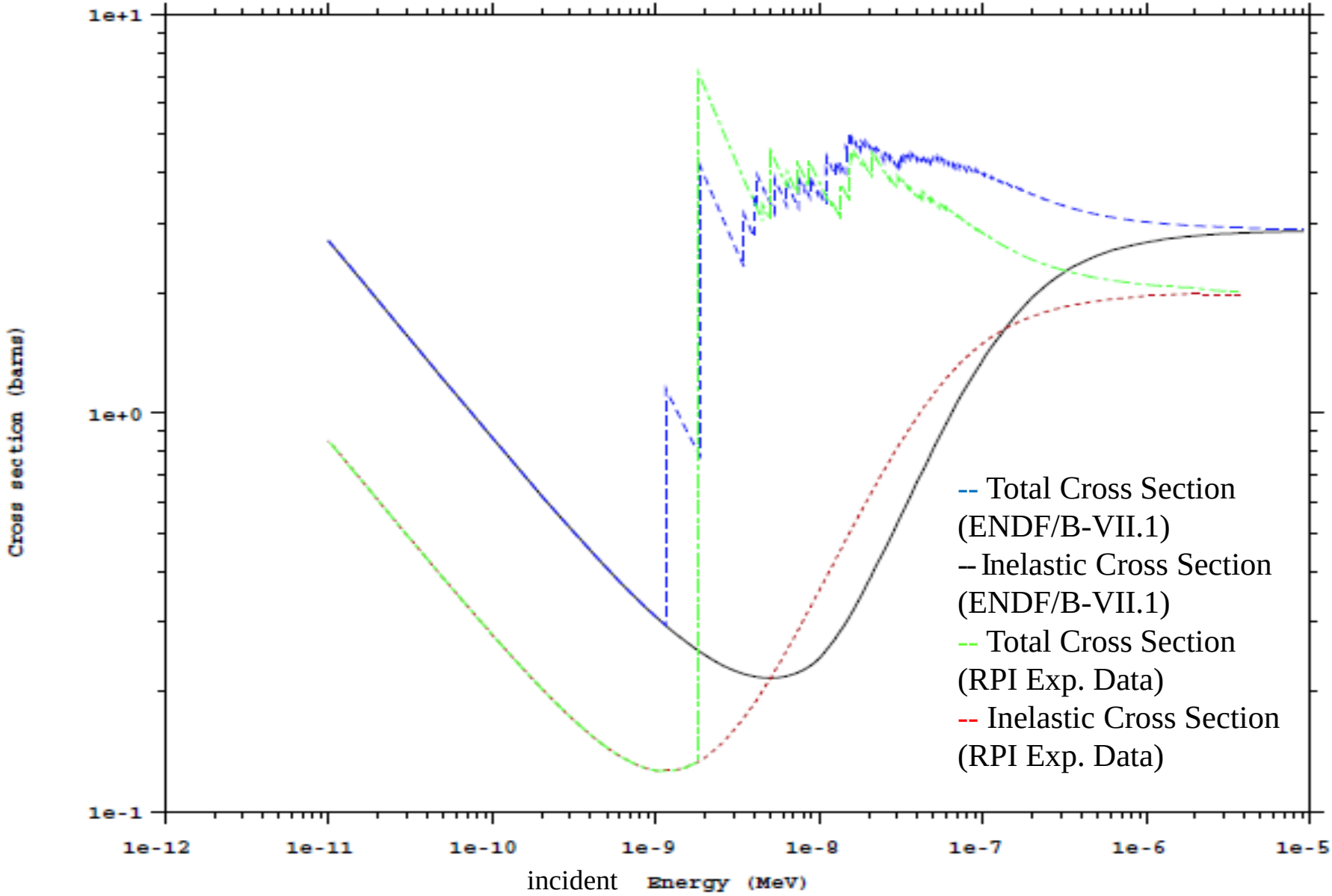
poly.20t-- is the Cross Section that comes from the MCNP 6.1 library (the poly version of lwtr.20t)

poly12.07t-- is a total Cross Section from Kemal's work and is made from the phonon spectrum of the experimental data and ran through the LEAPR module of NJOY to create our file (it corresponds to the ARCS GDOS line in our legends)

poly12,01t-- is the cross section of the ENDF/B-VII.1 S(α,β) file that the students have run through our NJOY 2012 input file. It was the file we used to confirm we can recreate the file that comes with MCNP i.e the poly.20t cross section.

Also, the experimental sample is never perfect (amorphous structure - incoherent and regular repeating structure - coherent).

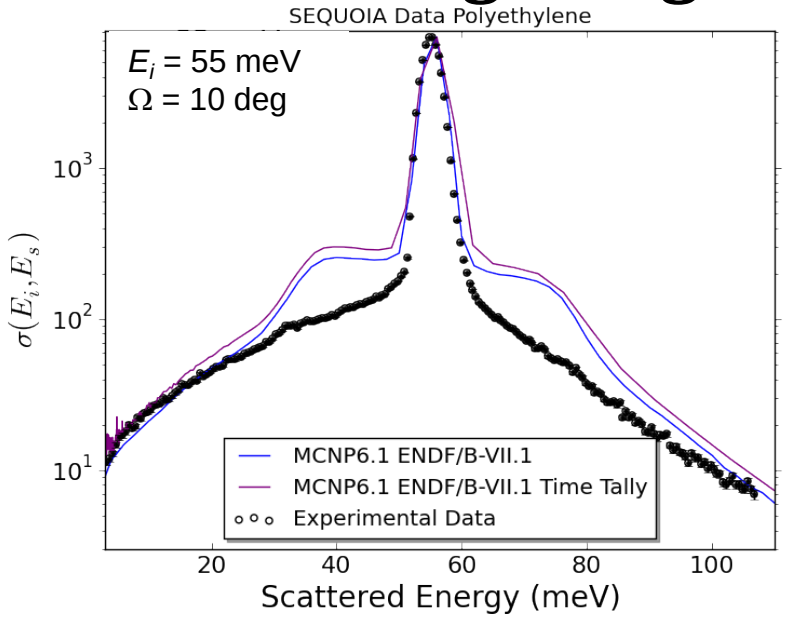
SiO_2 Total Cross section

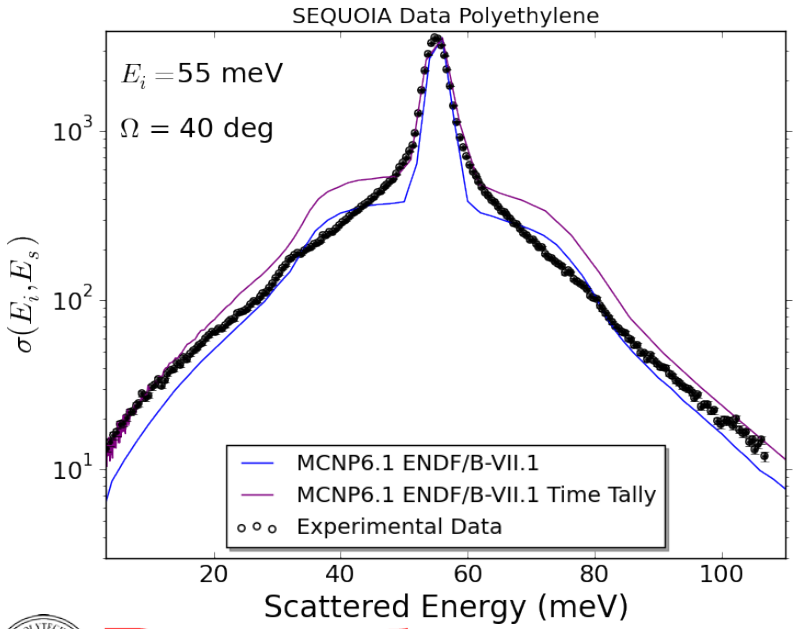


Integration, suggestions, and improvements of nuclear data based on current study

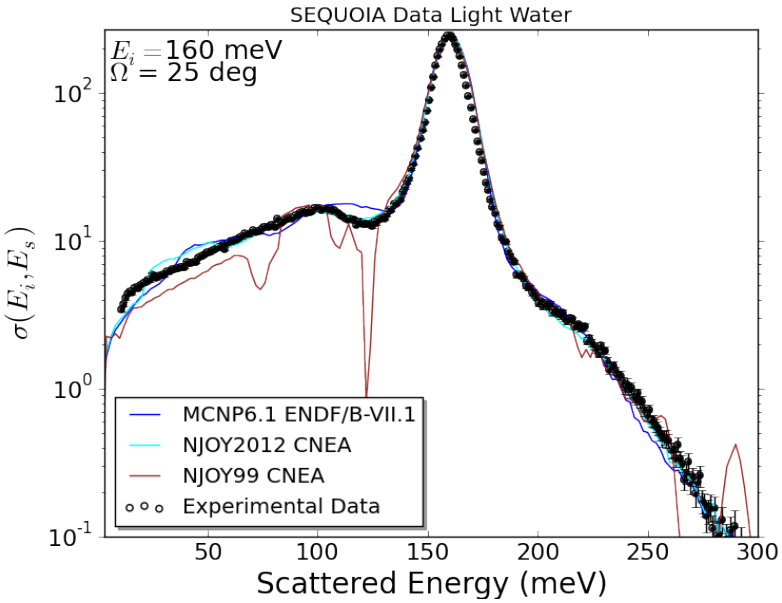
- Thermal scattering measurements (validation and prediction)
  - ✓ As is stated in the NJOY manual there are issues with small angles and thin samples in the library.
  - ✓ NJOY2012 doesn't need certain extra tweaks that NJOY99 needs for quasi-elastic scattering part in H_2O.
  - ✓ Quasi-elastic *vs.* inelastic scattering. For the case of liquids (e. g., water), we don't see a problem with how it is done and represented now. But for solids the worst places of disagreement in the DDSCS *vs.* Scattered Energy plots are around the elastic peak in the place where quasi-elastic scattering is happening. For the most part the solid ENDF evaluation over-estimates the quasi-elastic region, compared with experimental data.

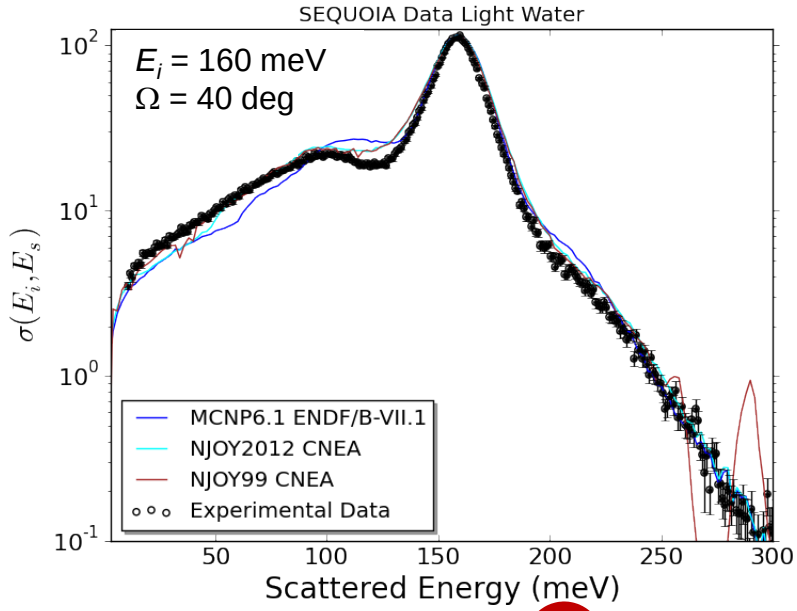
Small vs. Large Angles



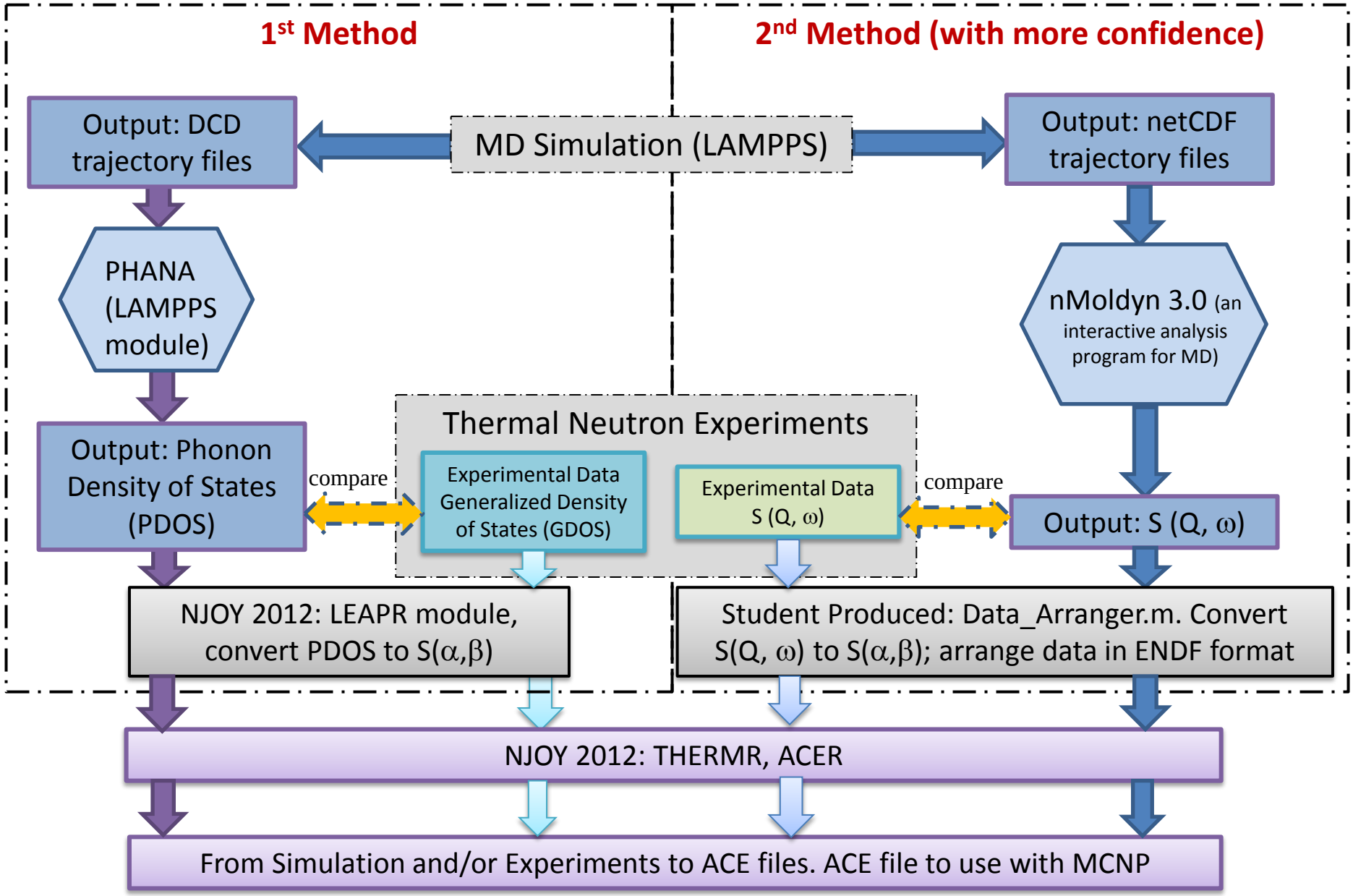


NJOY99 vs. NJOY2012





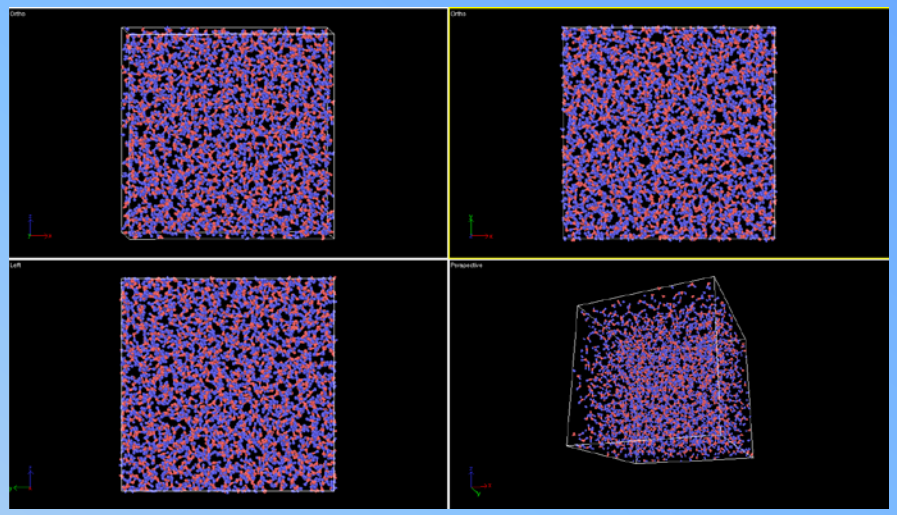
Comparative study between atomistic simulations and measurements



Water - LAMMPS Simulation

- Extended Simple Point Charge (SPC/E) potential

- Water simulation parameters:
  - 4500 atoms.
  - 300 K temperature.
  - 0.1 fs (femtoseconds) time step
  - 10,000,000 steps = 1 ns
    (← This is the time simulated)



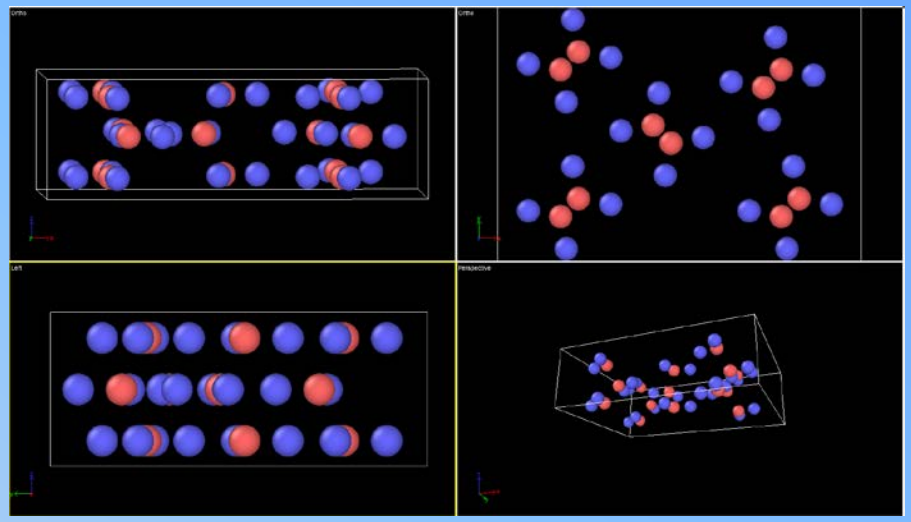
- Output: the location and velocity trajectory files which can be transferred into the phonon density of states (PDOS) or dynamic structure factor $S(q,\omega)$ where q is the wave vector, and ω the frequency.

- Reference: *lammps.sandia.gov/bench*

Polyethylene - LAMMPS Simulation

- Adaptive Intermolecular Reactive Empirical Bond Order (AIREBO) potential for systems of carbon and hydrogen atoms.

- Polyethylene (-CH_2-)n chain, orthorhombic unit cell.
  - 45 atoms in unit cell, 1215 atoms.
  - 296 K temperature.
  - 0.1 fs (femtoseconds) time step
  - 20,000,000 steps = 2 ns
    (← This is the time simulated)



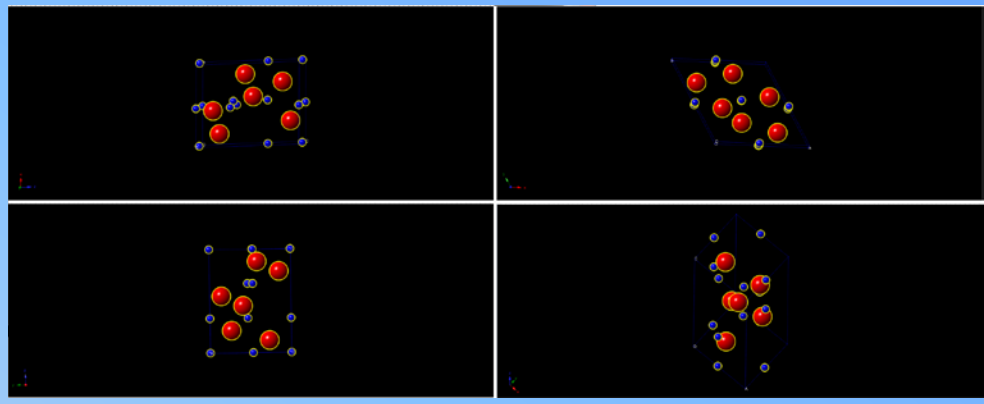
- Output: the location and velocity trajectory files which can be transferred into the phonon density of states (PDOS) or dynamic structure factor S(q,ω) where q is the wave vector, and ω the frequency.

- Reference: *Handbook of Polyethylene: Properties, and Applications by Andrew Peacock*

Quartz - LAMMPS Simulation

- Charge-Optimized Many Body (COMB) potential for Si/SiO2 systems

- Alpha-quartz simulation:
  - 20 atoms in unit cell, 2000 atoms.
  - 293 K temperature.
  - 0.1 fs (femtoseconds) time step
  - 15,000,000 steps = 1.5 ns
    (← This is the time simulated)

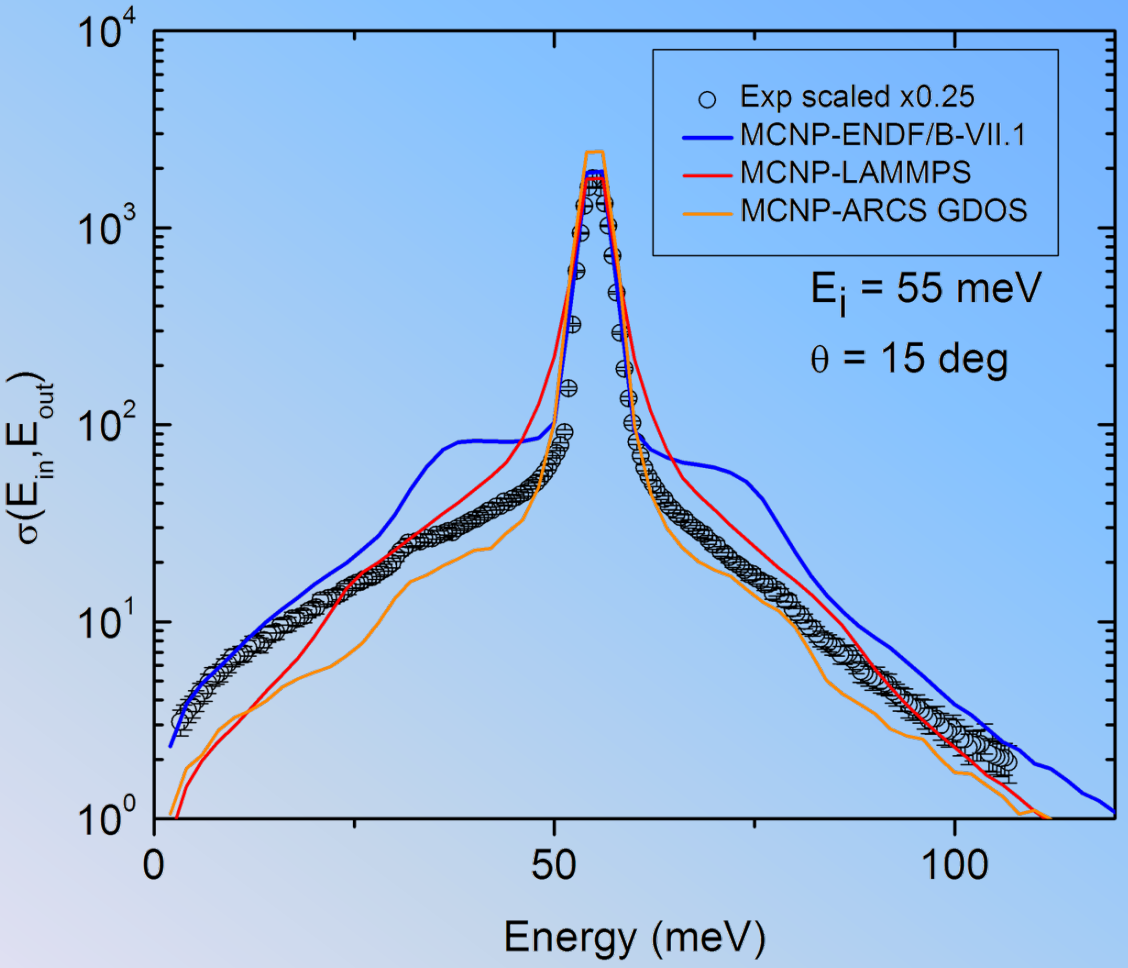


- Output: the location and velocity trajectory files which can be transferred into the phonon density of states (PDOS) or dynamic structure factor S(q,ω) where q is the wave vector, and ω the frequency.

- Reference: *CrystalMaker software, Structure Type 097*

1st Method: PDOS and The Scattering Kernel - S(α,β)

MD generated phonon spectrum can be converted to a scattering kernel S(α,β) Example: Polyethylene



2nd and New Method: S(Q,ω) → S(α,β)

- Transformation from S(Q,ω) to S(α,β):

$$\alpha = \hbar^2 Q^2 / 2mkT$$

$$\beta = \hbar\omega / kT$$

$$S(\alpha,\beta) = kTS(Q,\omega)$$

where kT is the temperature in eV.

- In the process of calculating results.

Integration, suggestions, and improvements of nuclear data based on current study

- Molecular Dynamics (understanding the Physics and help with prediction)
  - ✓ Employ MD simulations to improve S(α,β) scattering kernel for different materials.
  - ✓ Current Evaluated Nuclear Data Files (ENDF) contain S(α,β) only at specific temperatures, interpolation for other temperatures.
  - ✓ Using predictive capabilities of MD the errors produced by interpolation could be removed by simulating and calculating S(α,β) values at all needed temperatures.
  - ✓ Continue collaboration with Dr. Arbanas at ORNL with regards to error propagation for S(α,β) files.

Acknowledgement

- Key collaborators at ORNL (Goran Arbanas, Luiz Leal, Mike Dunn).
- Special thanks to the help from scientists at SNS:

  Alexander Kolesnikov, Doug Abernathy, and others
- The project is funded by DOE NCSP.

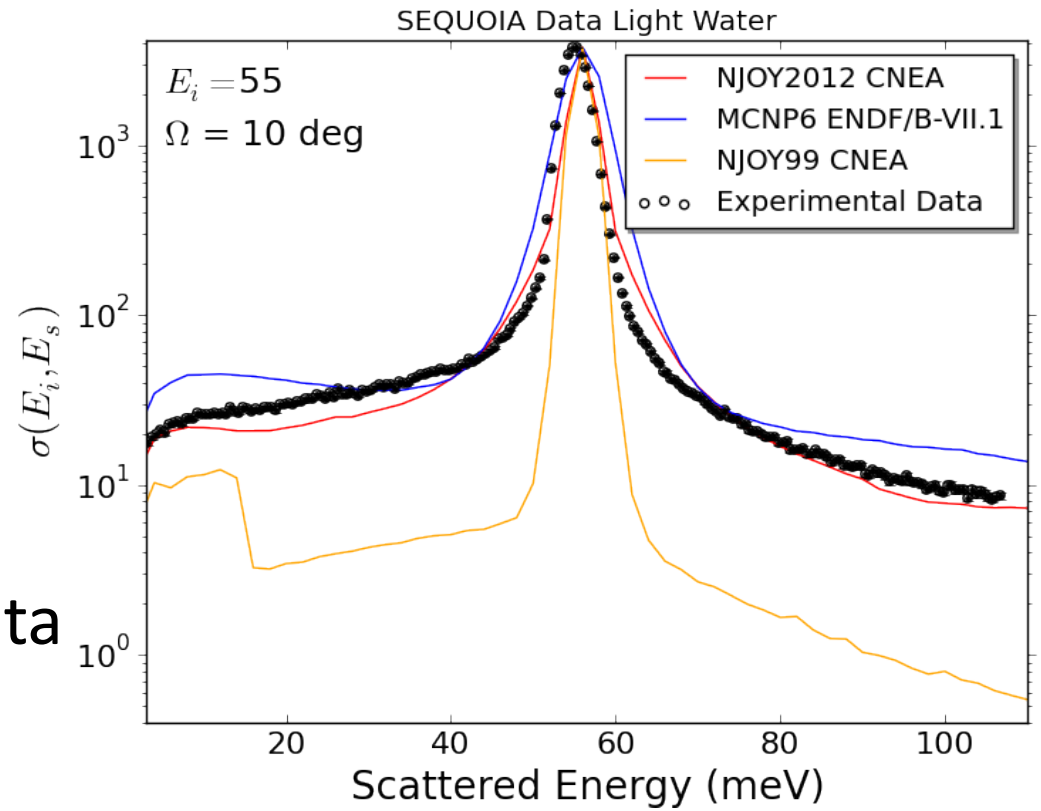
Questions?

Important Extra Slides - Plot Color Outline

- MCNP 6.1 ENDF/B-VII.1
  - F5 (ring detector) Energy Tally Output from MCNP simulation with energy resolution
- MCNP 6.1 ENDF/B-VII.1 w/o Res.
  - F5 (ring detector) Energy Tally Output from MCNP simulation without energy resolution
- MCNP 6.1 ENDF/B-VII.1 Time Tally
  - F5 (ring detector) Time Tally Output from MCNP simulation with energy resolution
- MCNP 6.1 Lammps
  - MCNP Energy Tally Output where the S(α,β) data was created using MD Simulation
- MCNP 6.1 ARCS GDOS
  - MCNP Energy Tally Output where the S(α,β) data was created using ARCS phonon spectrum data and processed through NJOY (Using Leapr, Thermr, & ACER)
- MCNP 6.1 RPI ACE
  - MCNP Energy Tally Output where the S(α,β) data was created by taking Experimental data and converting it to S(α,β) and putting it into ENDF format manually and processing it through NJOY (Only Thermr & ACER used)
- NJOY99 CNEA
  - MCNP Energy Tally Output where the S(α,β) data was created by the Comision Nacional Energia Atomica (Argentinian) research group, data processed using NJOY99
- NJOY2012 CNEA
  - MCNP Energy Tally Output where Argentinian evaluation is processed through NJOY 2012
- ENDF/B-VI.0
  - [illegible]

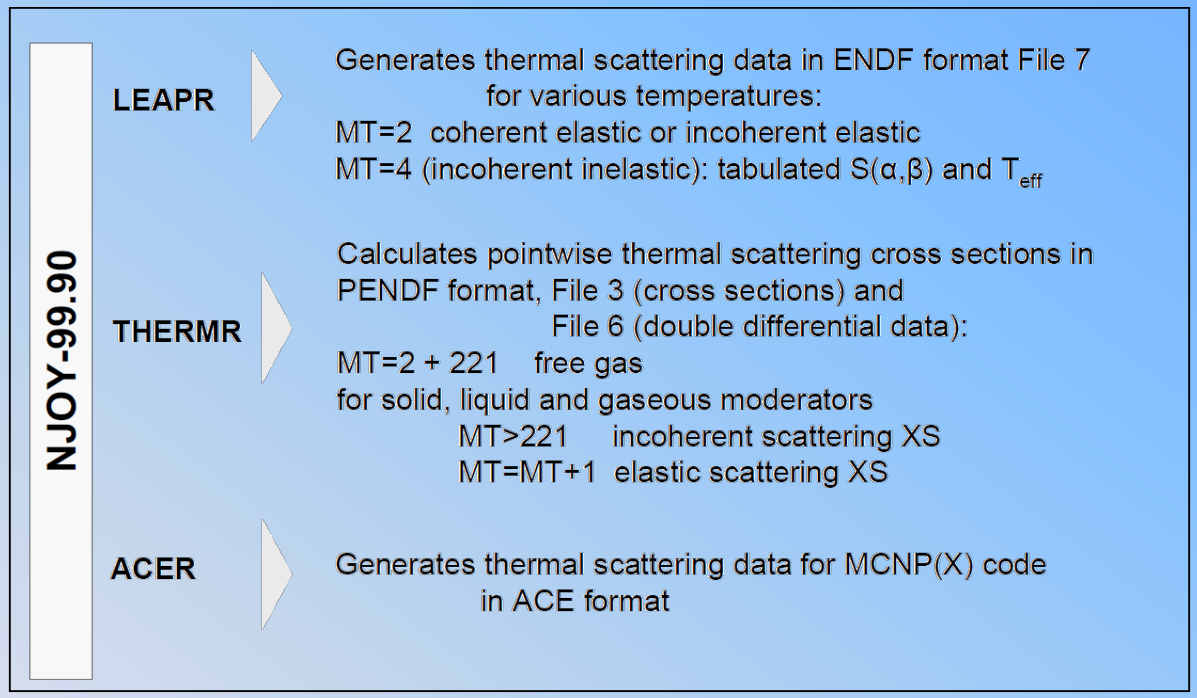
Error Analysis

- The error on the experimental data is transformed from counts to DDSCS like the data itself.
- Currently the simulations are normalized to the data so no other transformation is required



Polyethylene discussion – S(α,β)

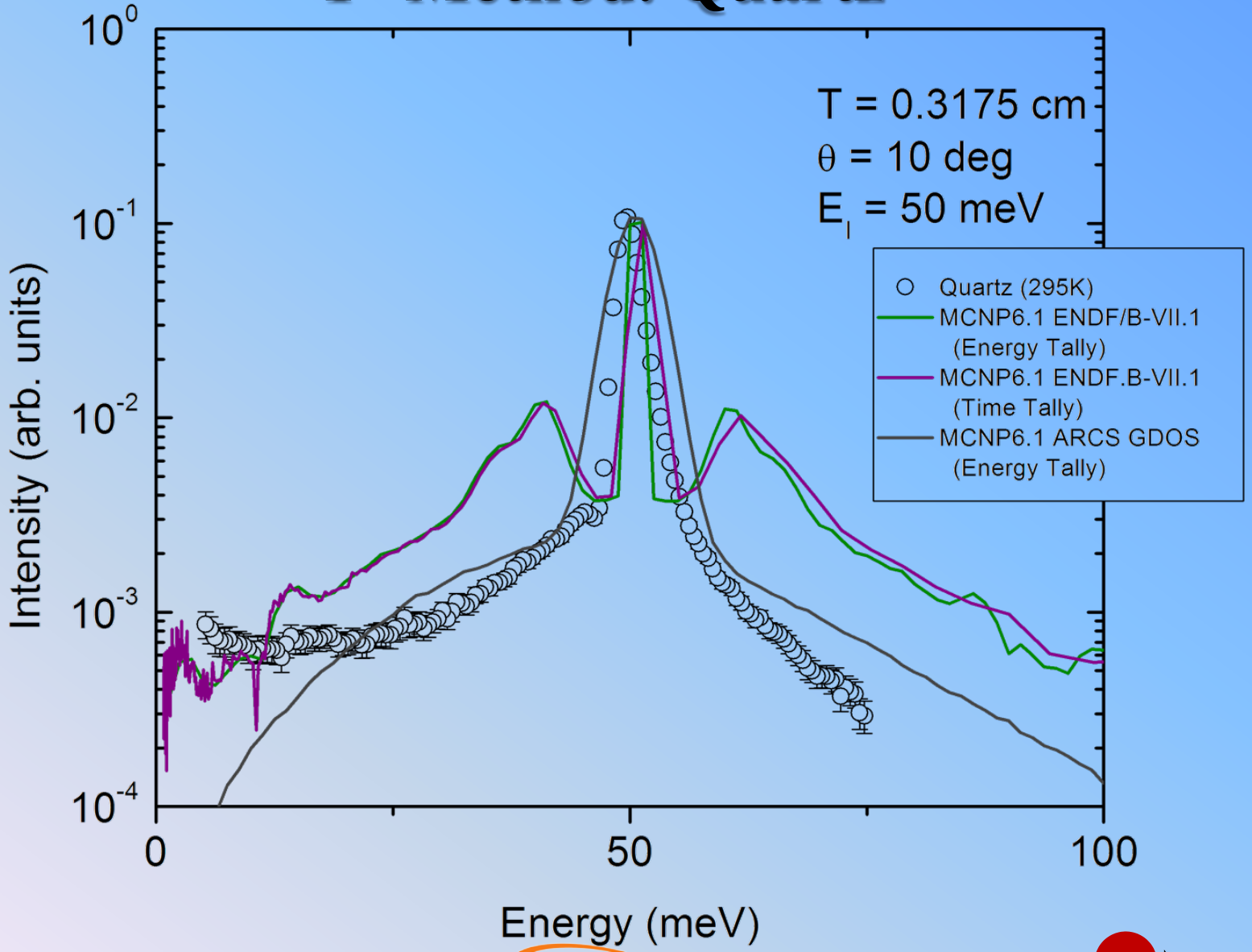
- PDOS →NJOY99 (LEAPR)



- ACE files → MCNP6.1 to generate double differential scattering cross section (DDSCS) to compare with the experiment.

*M. Mattes and J. Keinert, Thermal Neutron Scattering Data for the Moderator Materials H2O, D2O and ZrHx in ENDF-6 Format and as ACE Library for MCNP(X) Codes, IAEA INDC(NDS)-0470, 2005.

1st Method: Quartz



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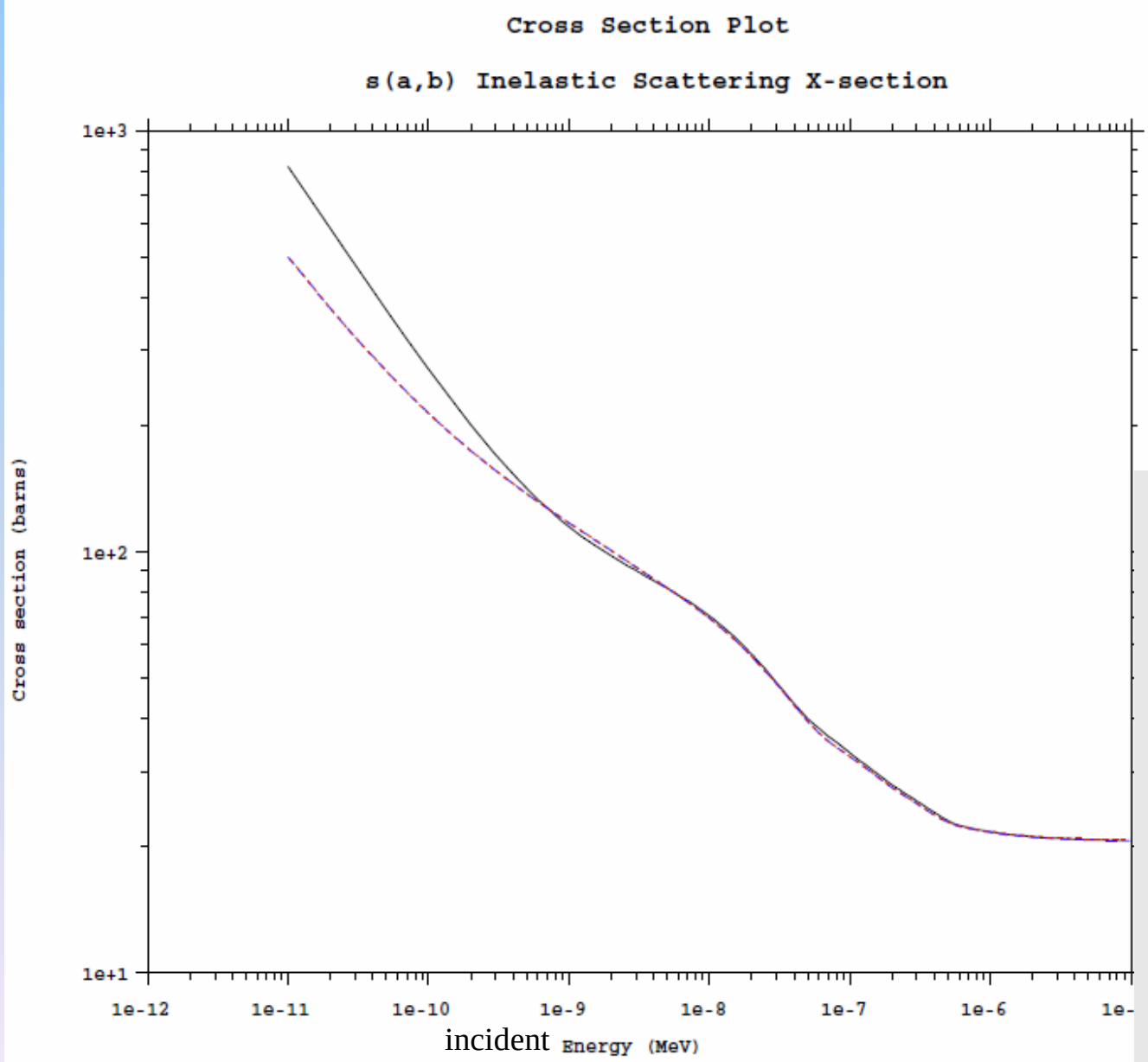

lwtr.20t--is the Cross Section from ENDF/B-VII.1 S(α,β) ACE file that comes with the MCNP 6.1 release library or in other words it is the current standard.

lwtr00.00t--is the Cross Section from the Argentinian ACE file, it was created being run through NJOY 99

lwtr00.01t--is the Cross Section from the Argentinian data that I ran through our NJOY 2012 input file to create an ACE file.

1meV cold neutron agreement may be good enough.

However, NJOY 99 and NJOY 2012 are different…

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