Thermal Neutron Scattering Measurements and Analysis

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Overview

❖ Performed some analysis of measured samples of high density polyethylene (HDPE), and quartz (SiO_2) at various temperatures and different instruments (SEQUOIA and ARCS of SNS).

❖ Comparative and integrative study of MCNP (including the evaluations), molecular dynamics simulations, and thermal neutron experiments are in progress.

✓ Comparative study between measurements and MCNP (evaluations)

✓ Comparative study between atomistic simulations and measurements

Thermal Scattering Overview

- Analyzed experimental data for High Density Polyethylene (HDPE) and Quartz (SiO_2) using the Wide Angular-Range Chopper Spectrometer (ARCS) at SNS.
  - Quartz measurements were done at 20, 300, 550, 600 °C
  - HDPE was measured at 295 K and 5 K.
  - Incident energies at 50, 100, 250, and 700 meV
- ARCS has an angular range of -28° – 135°, Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) has an angular range of -30° – 60°.
- In the progress of addressing issues in discrepancies between the thermal scattering experiments and MCNP calculations.
- LAMMPS code is utilized to calculate the phonon spectrum, scattering function $S(Q, \omega)$ and scattering kernel $S(\alpha,\beta)$ for HDPE and SiO_2.
- In the progress of addressing issues in discrepancies between the thermal scattering experiments and atomistic simulations.

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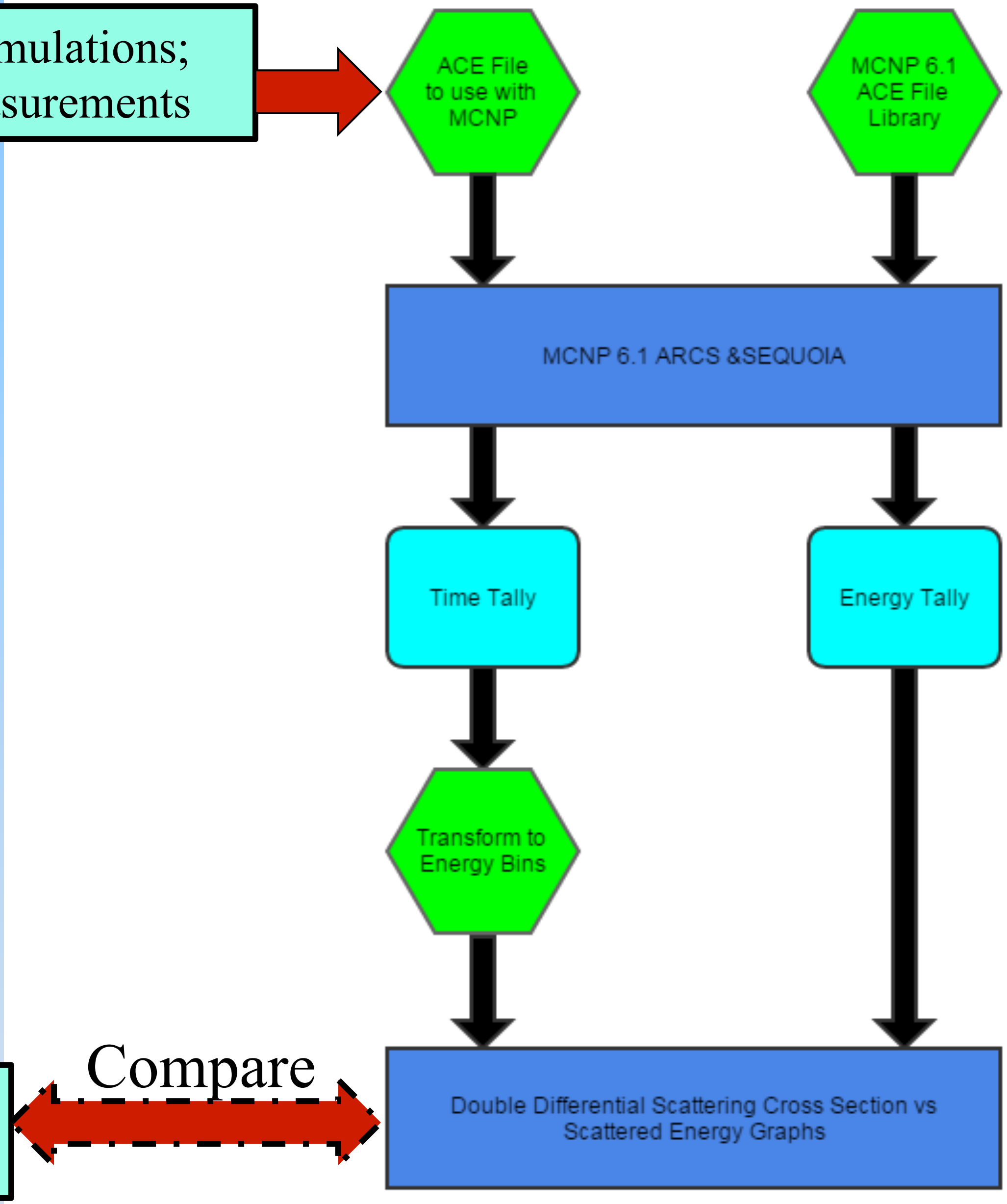
Molecular Dynamics Simulations;
Thermal Scattering Measurements

Data Flow Chart: Compare Experiments with MCNP Models

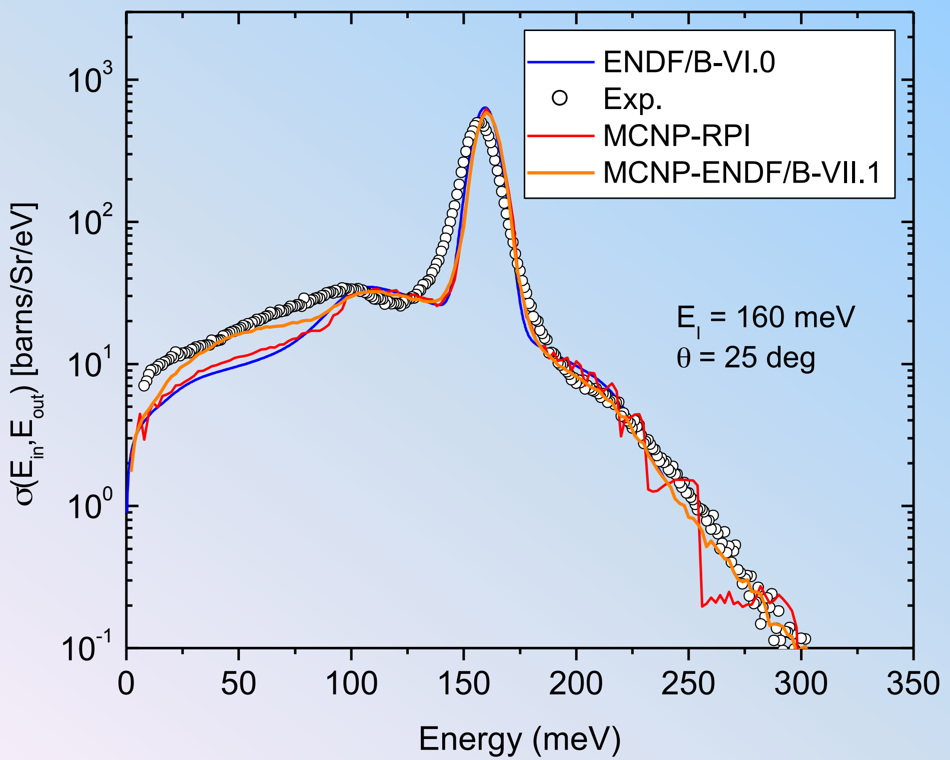


Thermal Scattering Measurements;
ENDF/B-VII.1

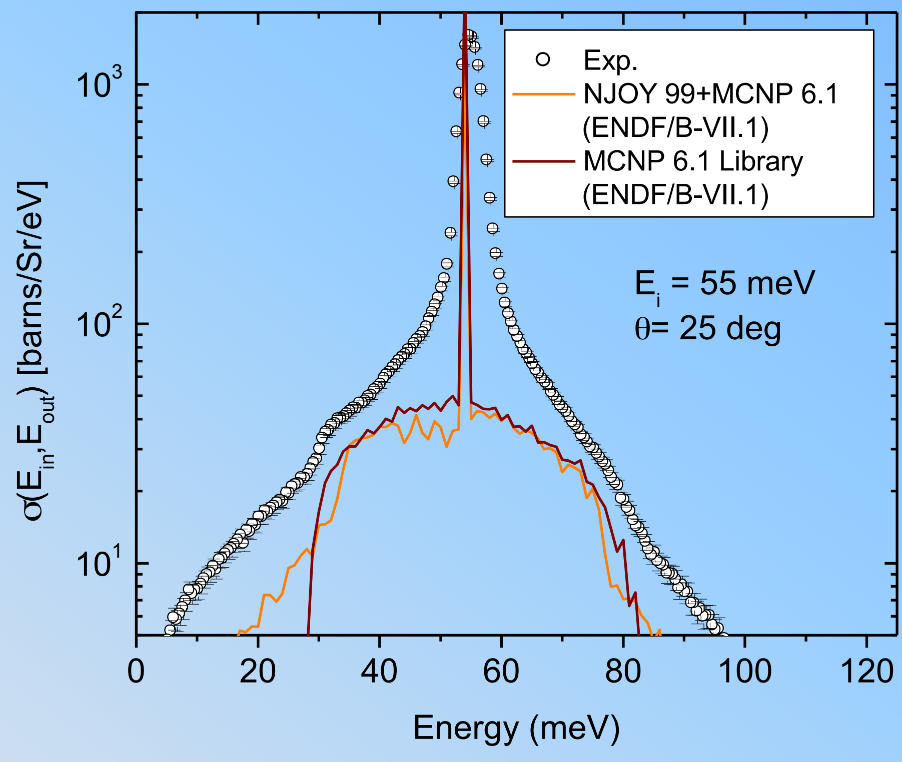
SEQUOIA H_2O and CH_2 with MCNP models

- Use MCNP simulation of the incident energy spread.
- Comparison with several evaluations.
- More structure and larger differences in forward angles.
- Possible issues with how we run NJOY/MCNP.

H_2O



CH_2



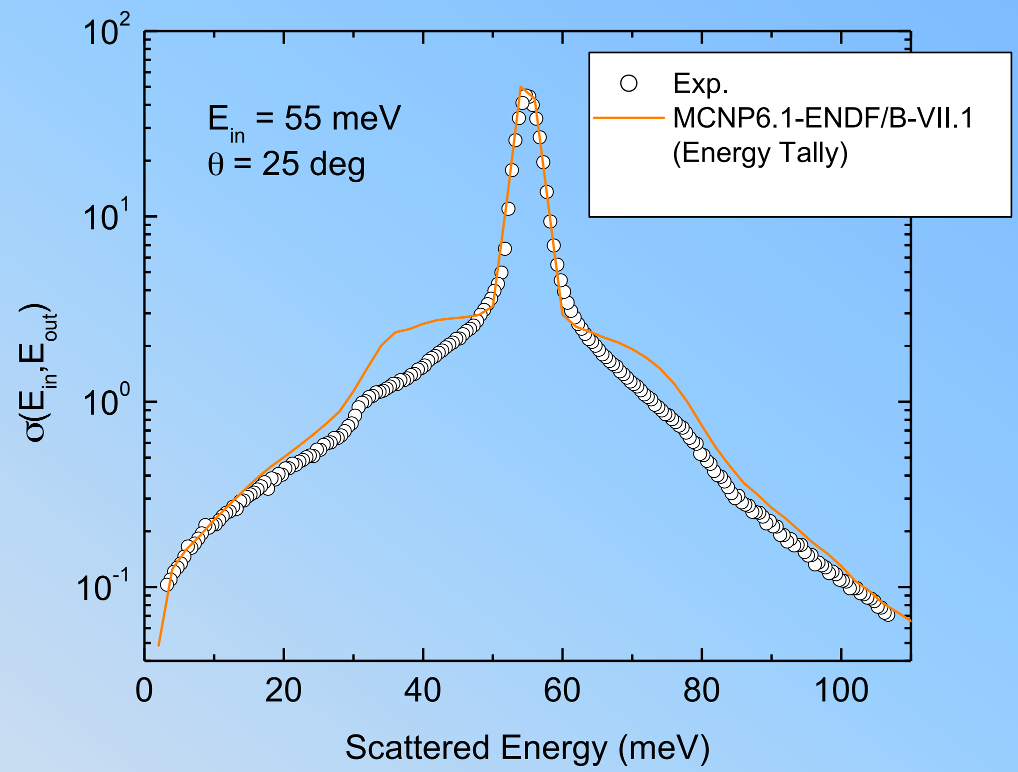
Exp/MCNP model differences and the possible issues to address those differences

- 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
- The detailed reduction of thermal scattering experimental results
  - Need to be re-visited and be normalized carefully based on the total cross section (ENDF)
- MCNP file is an idealized version of experiment
  - Need to add spectrometer resolution
  - Add sensitivity/uncertainty

Step 1: Add spectrometer resolution

- Broadening the mono-energetic neutron source to a Gaussian spectra results in a better fit of the elastic peak

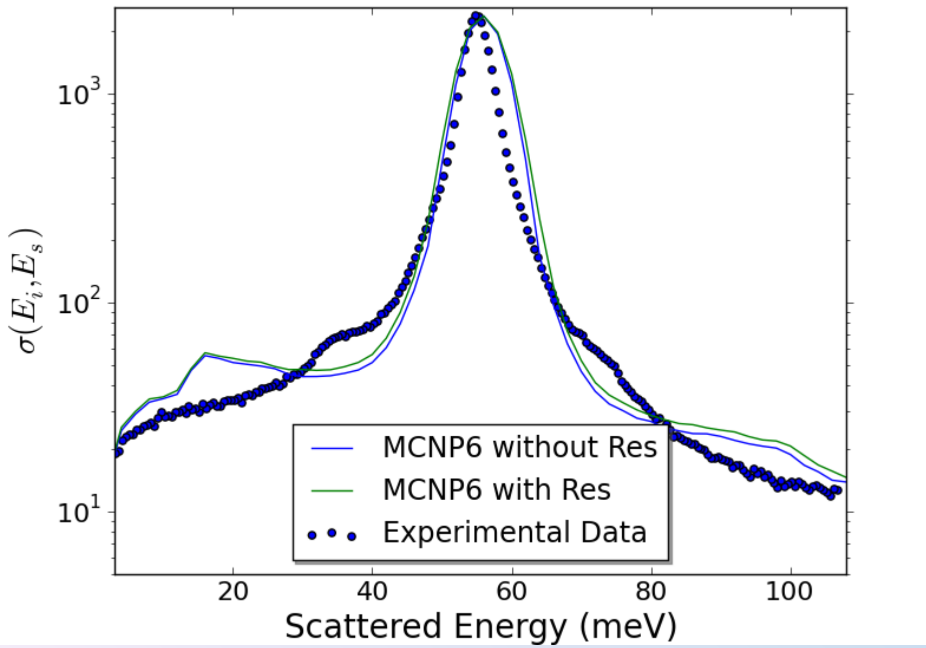
Polyethylene

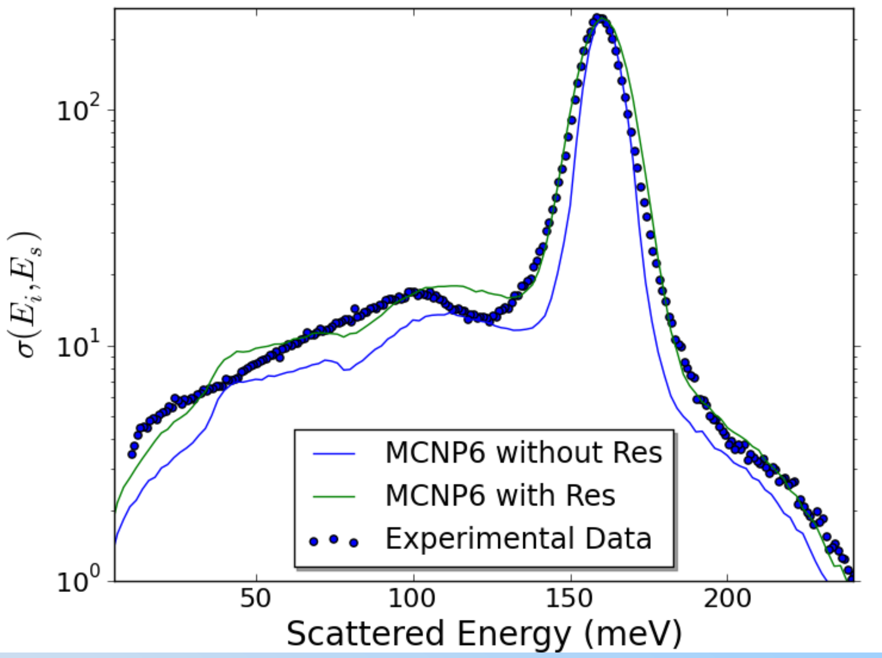


Check the change on H_2O

Improvement at higher incident energies

- 55 meV, 25 deg
- 160 meV, 25 deg





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Step 2: Further Energy Resolution Issues

- Using trial and error, best resolution fit occurs around $\Delta E/E = 3\%$ (FWHM) for the MCNP simulation at 55 meV
- The energy resolution measured by the instrument scientists at SEQUOIA is about half that, ~1.5%

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Step 3: Energy Tally Limitations

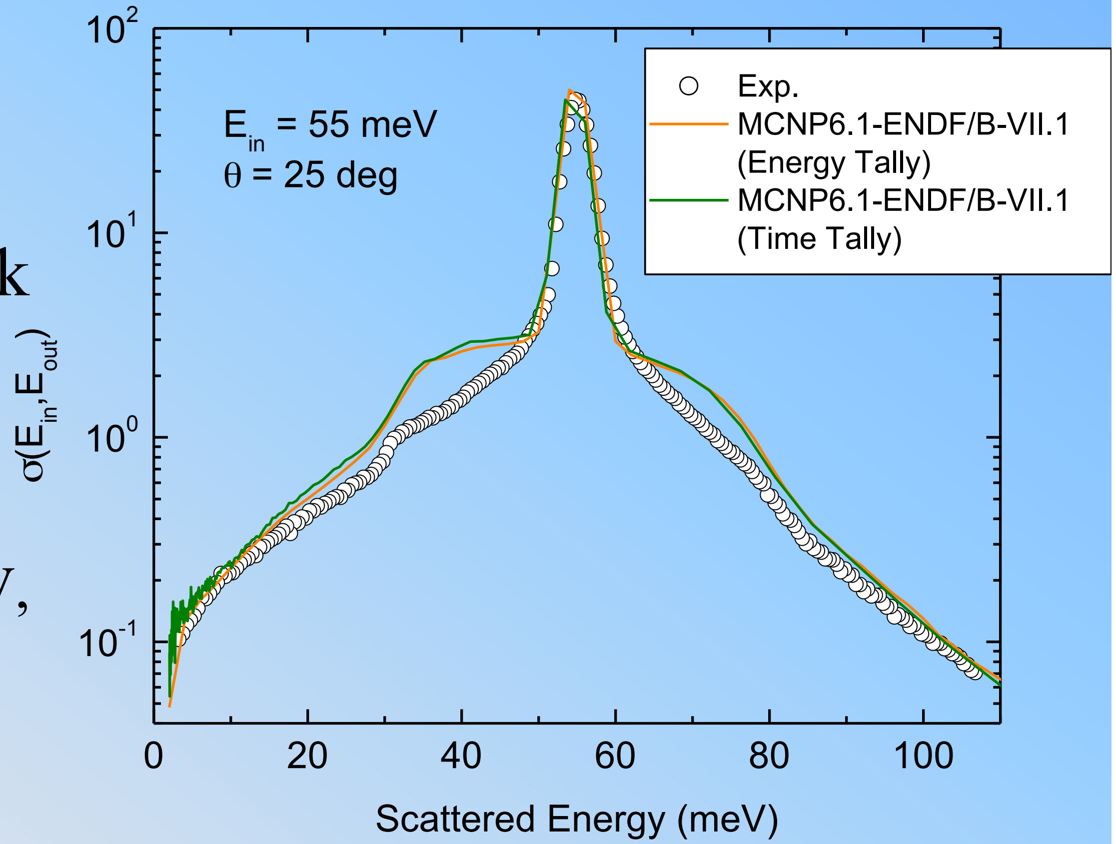
- Using a F5 energy tally to measure the DDSCS is not technically a perfect representation of the experiment
- SEQUOIA and ARCS are TOF Spectrometers
- Changing the Tallies to F5 Time Tallies is the first step
- Next is realize that the protons at the SNS do not hit the mercury target instantaneously

Step 4: Time Resolution

- 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

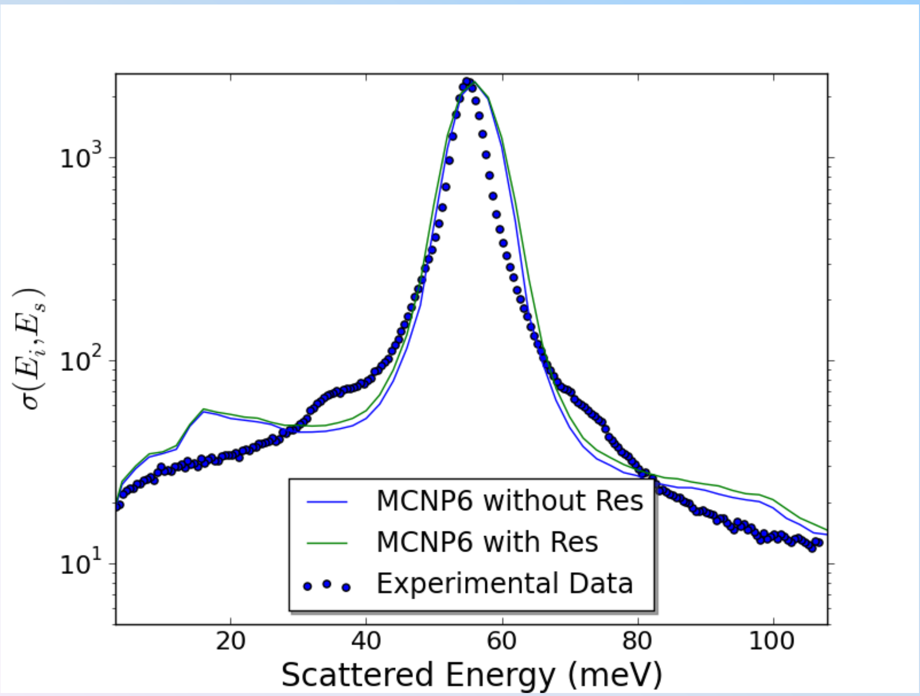
Polyethylene Time Tally

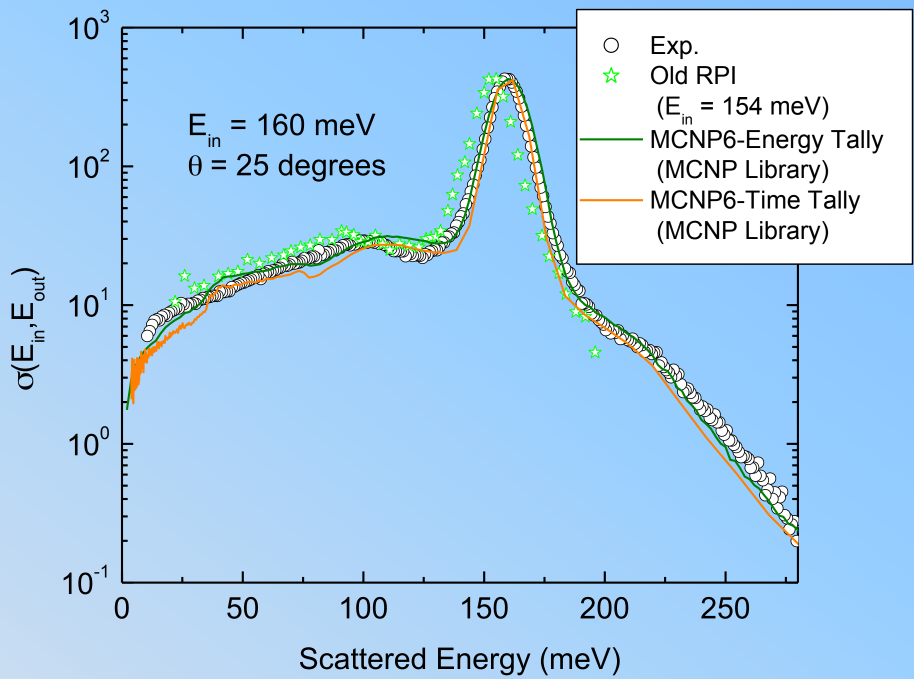
- Using the Proton pulse width caused the energy resolution to change as well to fit the elastic peak
  - $\Delta E/E \approx 1.5$ %
  - Which agrees with the Instrument Scientist's measurement (for 55 meV, and specific chopper setting)



Return to Light Water

- 55 meV, 25 deg





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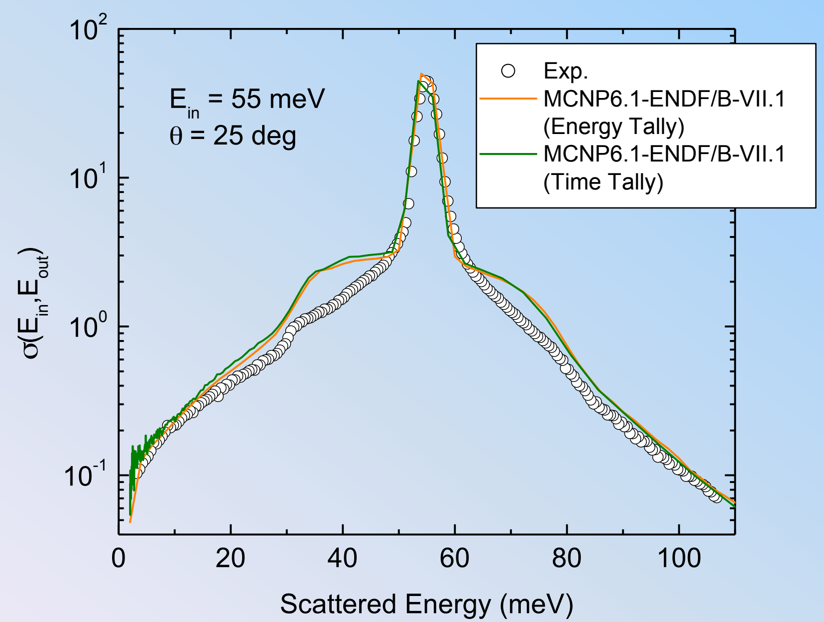
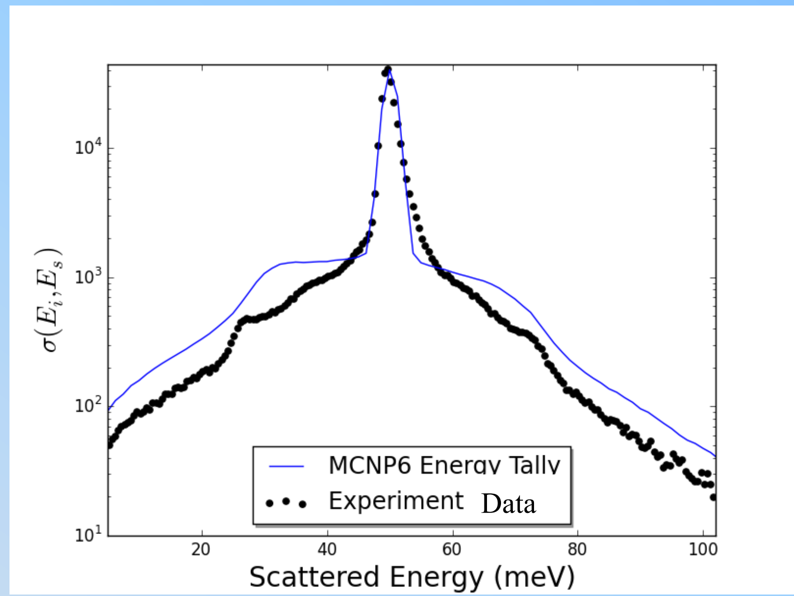
SEQUOIA vs ARCS Poly (1)

Energy Tally & Time Tally

- SEQUOIA: 55 meV, 25 deg
- ARCS: 50 meV, 25 deg





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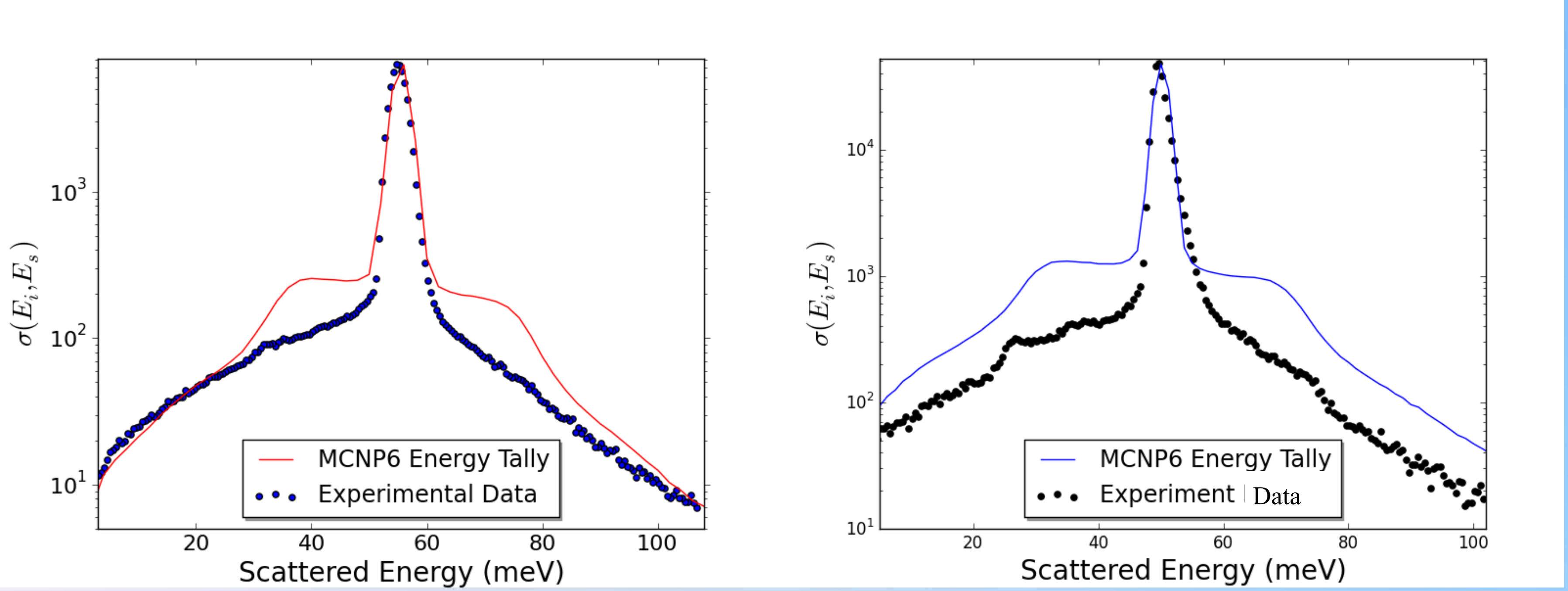
SEQUOIA vs ARCS Poly (2)

- SEQUOIA: 55 meV, 10 deg
- ARCS: 50 meV, 10 deg



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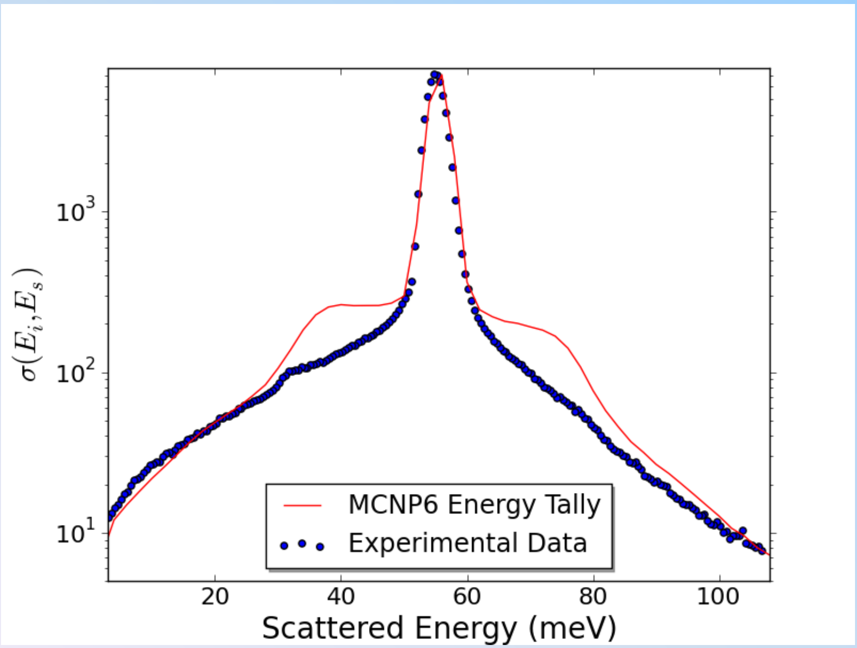
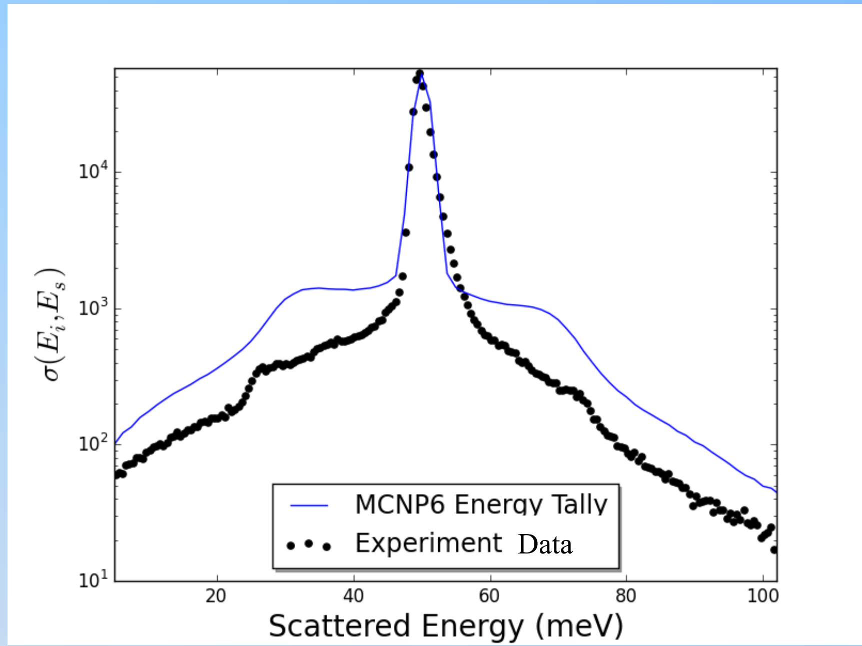
SEQUOIA vs ARCS Poly (3)

SEQUOIA: 55 meV, 15 deg

ARCS: 50 meV, 15 deg





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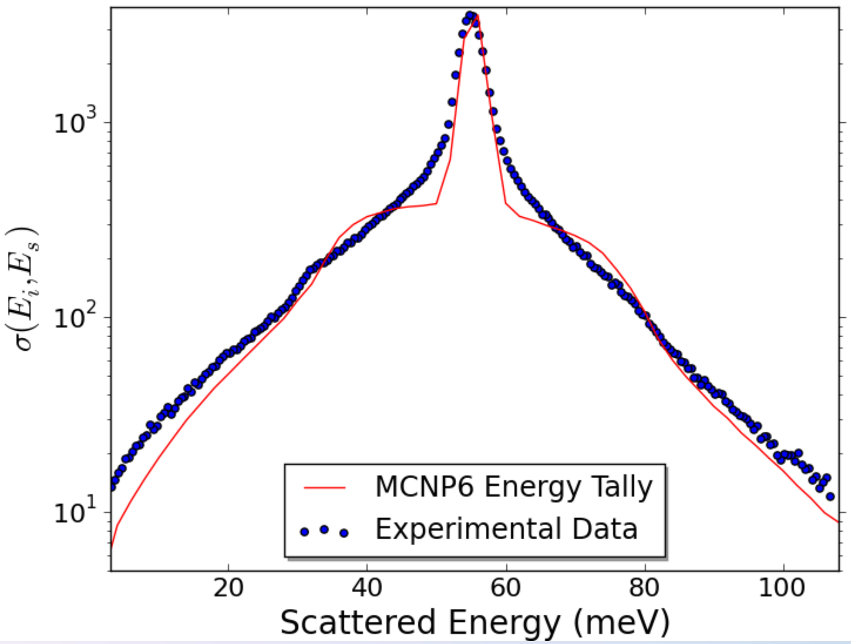
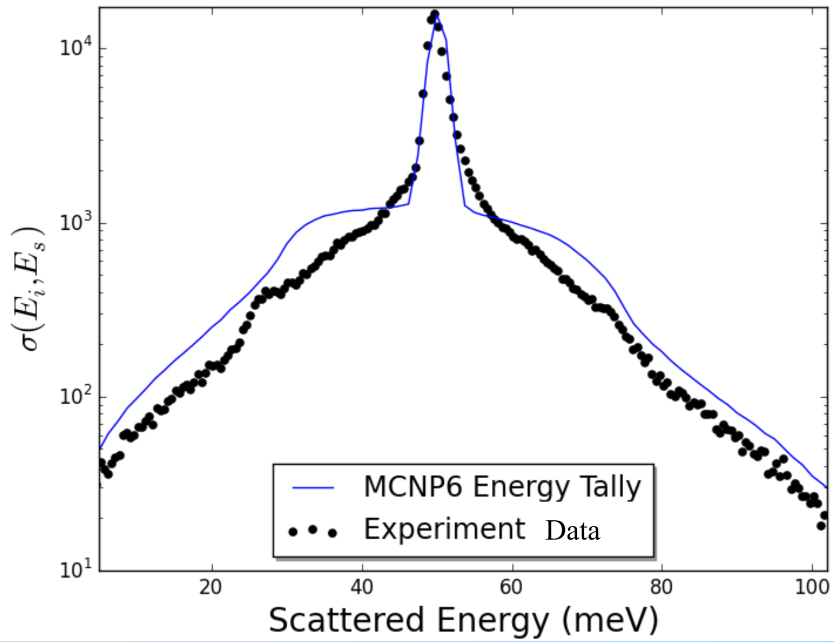
SEQUOIA vs ARCS Poly (4)

- SEQUOIA: 55 meV, 40 deg
- ARCS: 50 meV, 40 deg





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Research Goals – Atomistic Simulation: Molecular Dynamics (MD)

- 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.
- Work in progress for calculation of scattering kernels for water, polyethylene, and quartz.

Polyethylene - LAMMPS Simulation

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

- Polyethylene $(-CH_2-)n$ chain.
  - 400 molecules in chain, 1200 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 $S(q,\omega)$ where q is a wave vector, and ω a frequency.

Polyethylene discussion – S(α,β)

- PDOS → NJOY99 (LEAPR)

NJOY-99.90

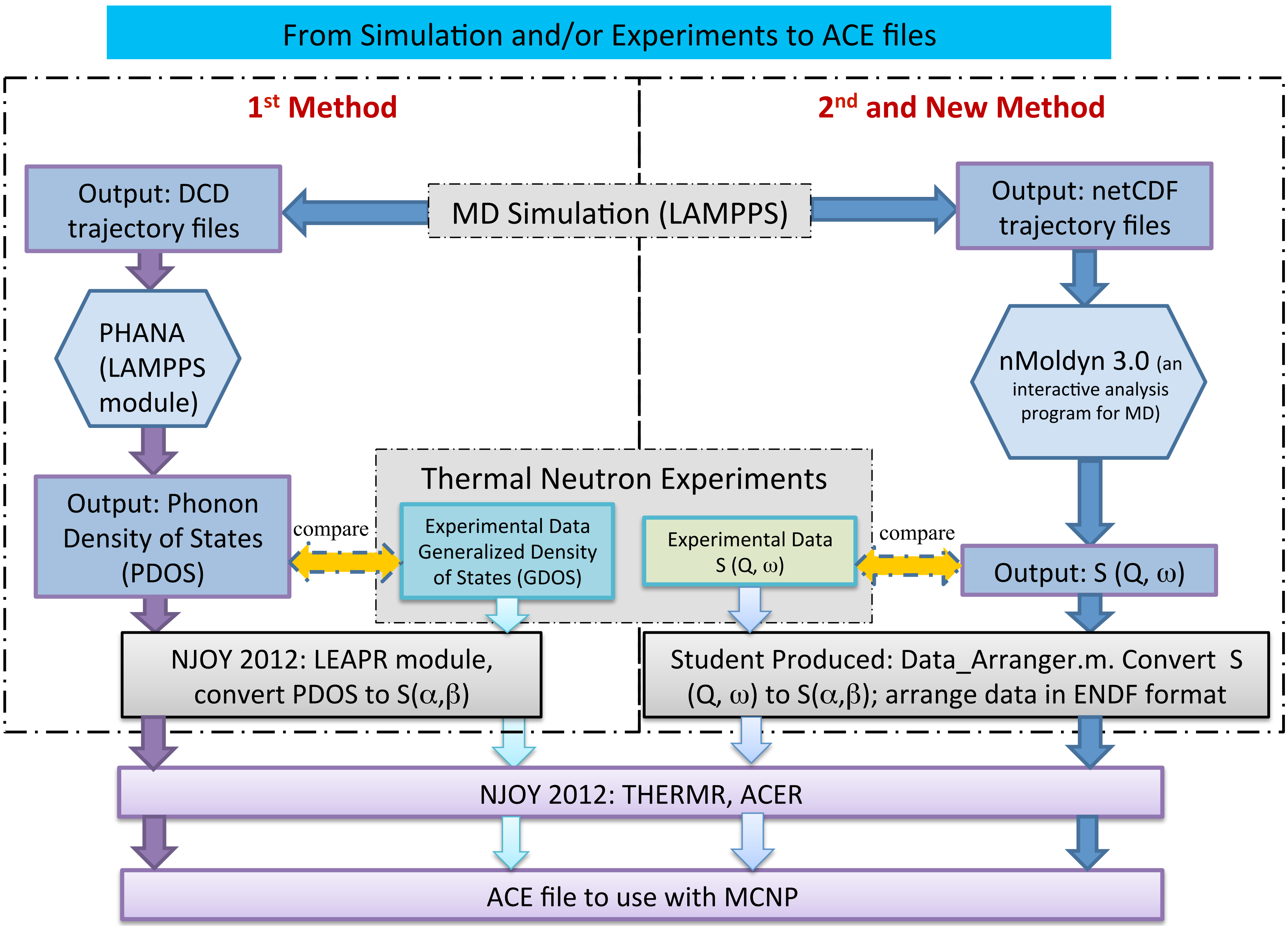
LEAPR → Generates thermal scattering data in ENDF format File 7 for various temperatures:
MT=2 coherent elastic or incoherent elastic
MT=4 (incoherent inelastic): tabulated S(α,β) and T_{eff}

THERMR → Calculates pointwise thermal scattering cross sections in PENDF format, File 3 (cross sections) and File 6 (double differential data):
MT=2 + 221 free gas
for solid, liquid and gaseous moderators
MT>221 incoherent scattering XS
MT=MT+1 elastic scattering XS

ACER → Generates thermal scattering data for MCNP(X) code in ACE format

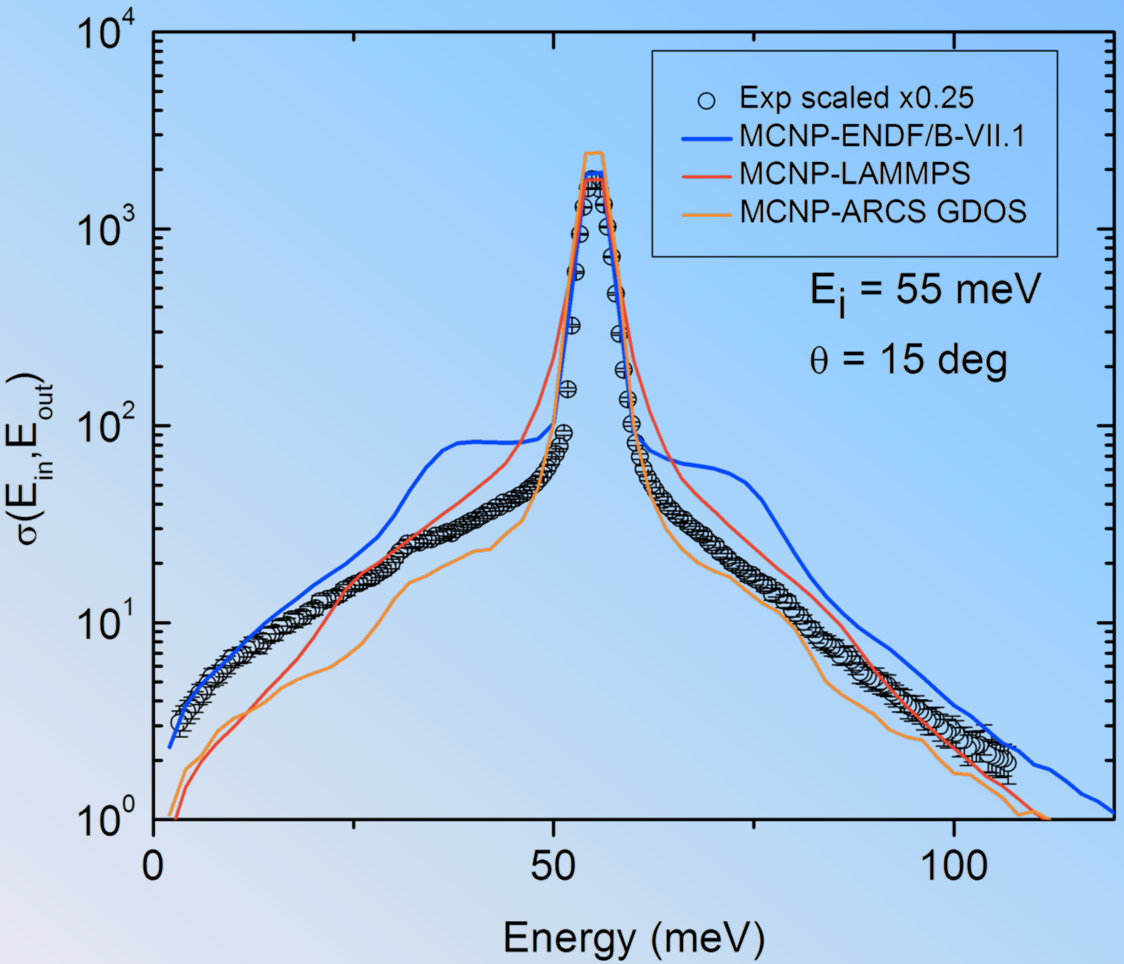
- ACE files → MCNP6.1 to generate double differential scattering cross section (DDSCS) to compare with the experiment.

*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.


From Simulation and/or Experiments to ACE files
1st Method
2nd and New Method
Output: DCD trajectory files
MD Simulation (LAMPPS)
Output: netCDF trajectory files
PHANA (LAMPPS module)
nMoldyn 3.0 (an interactive analysis program for MD)
Thermal Neutron Experiments
Output: Phonon Density of States (PDOS)
compare
Experimental Data Generalized Density of States (GDOS)
Experimental Data S (Q, ω)
compare
Output: S (Q, ω)
NJOY 2012: LEAPR module, convert PDOS to S(α,β)
Student Produced: Data_Arranger.m. Convert S (Q, ω) to S(α,β); arrange data in ENDF format
NJOY 2012: THERMR, ACER
ACE file to use with MCNP

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



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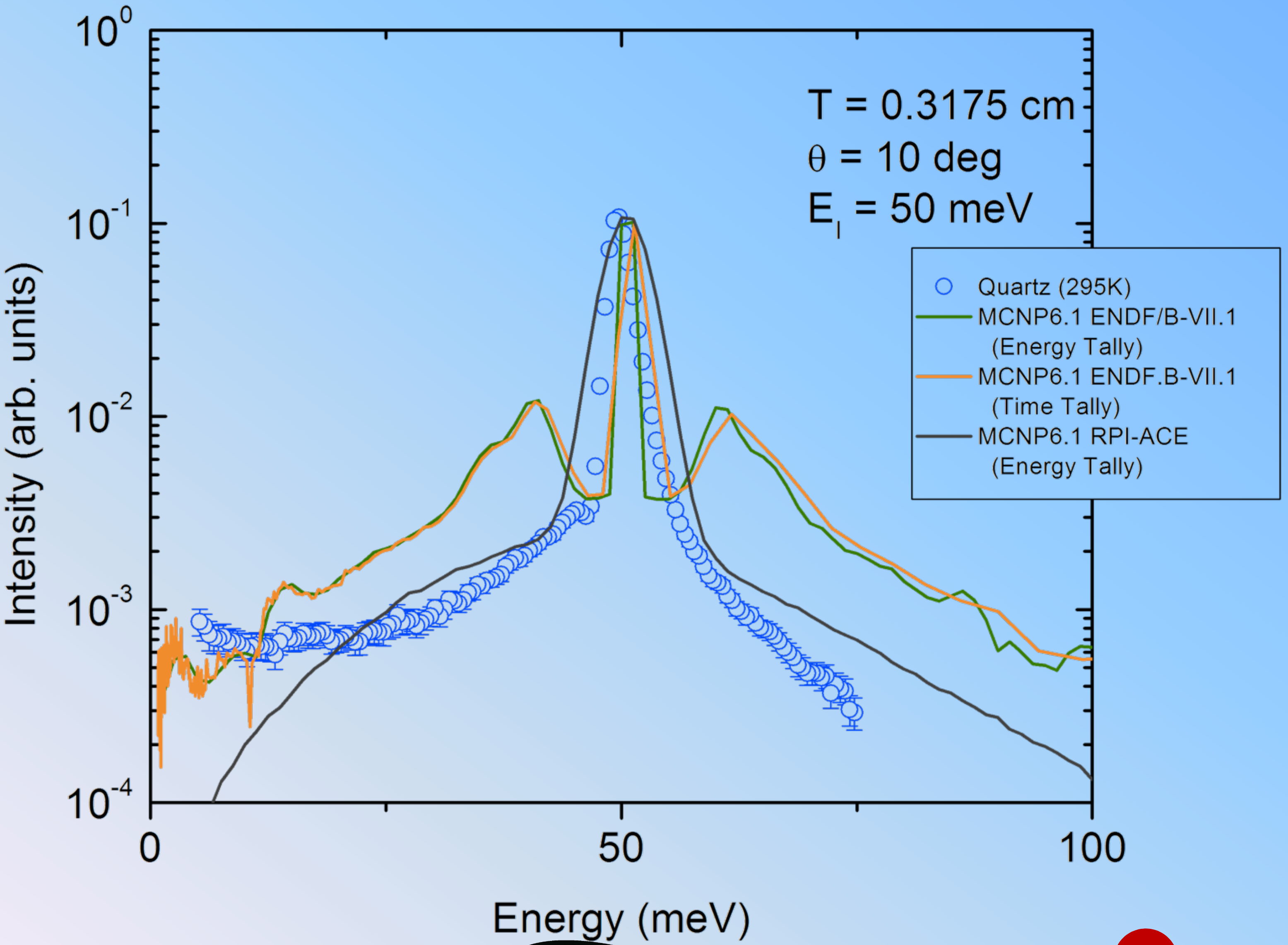
1st Method: Quartz



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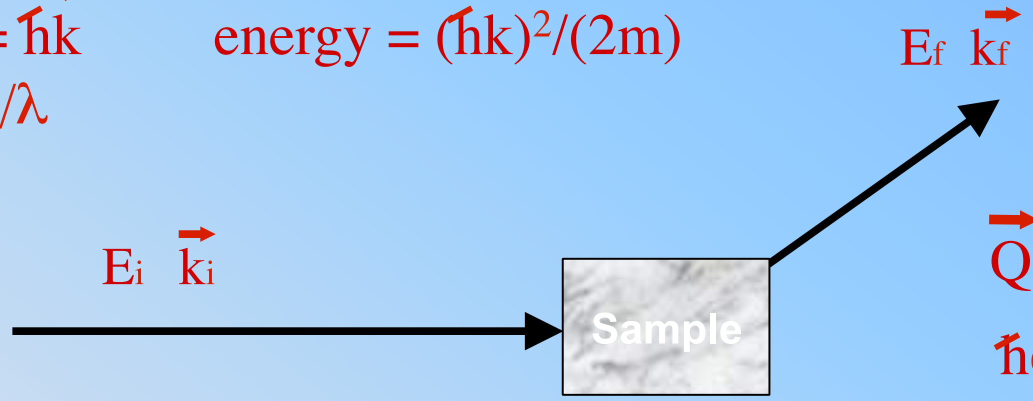
Geometry of Inelastic Neutron Scattering Experiments

momentum = $\hbar \vec{k}$

$k = 2\pi/\lambda$

energy = $(\hbar k)^2/(2m)$



$\vec{Q} = \vec{k}_i - \vec{k}_f$

$\hbar\omega = E_i - E_f$

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

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

depends ONLY on the sample

2nd and New Method: $S(Q,\omega) - S(\alpha,\beta)$

- Transformation from $S(Q,\omega)$ to $S(\alpha,\beta)$:

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

$$\beta = h\omega / kT$$

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

where kT is the temperature in eV.

- In the process of calculating results.

Future Studies

- Proper identification of the reasons associated with the discrepancies.
- Finish the 2nd and new method to calculate ACE file from MD simulation.
- Better understanding of PDOS physics.
- Perform MD simulations using different force models and try to improve upon the current ones.

Recent Accomplishments

- Completed measurements of thermal scattering at SNS
  - Quartz measurements were done at 20, 300 550, 600 °C at ARCS instrument of SNS
  - Both SEQUOIA and ARCS instruments of SNS were utilized for HDPE studies
  - HDPE was measured at 295 K and 5 K.

- Data analysis, MD simulations, and MCNP (with evaluations) calculations are in progress

Acknowledgement

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Questions?