OECD/NEA Kick-off Meeting: WPEC SG42
"Thermal Scattering Kernel S(α,β): Measurement, Evaluation and Application"
May 18 – 19, 2015 • Paris, France

Implementation of Atomistic Simulation Techniques in Thermal Neutron Scattering Data Generation

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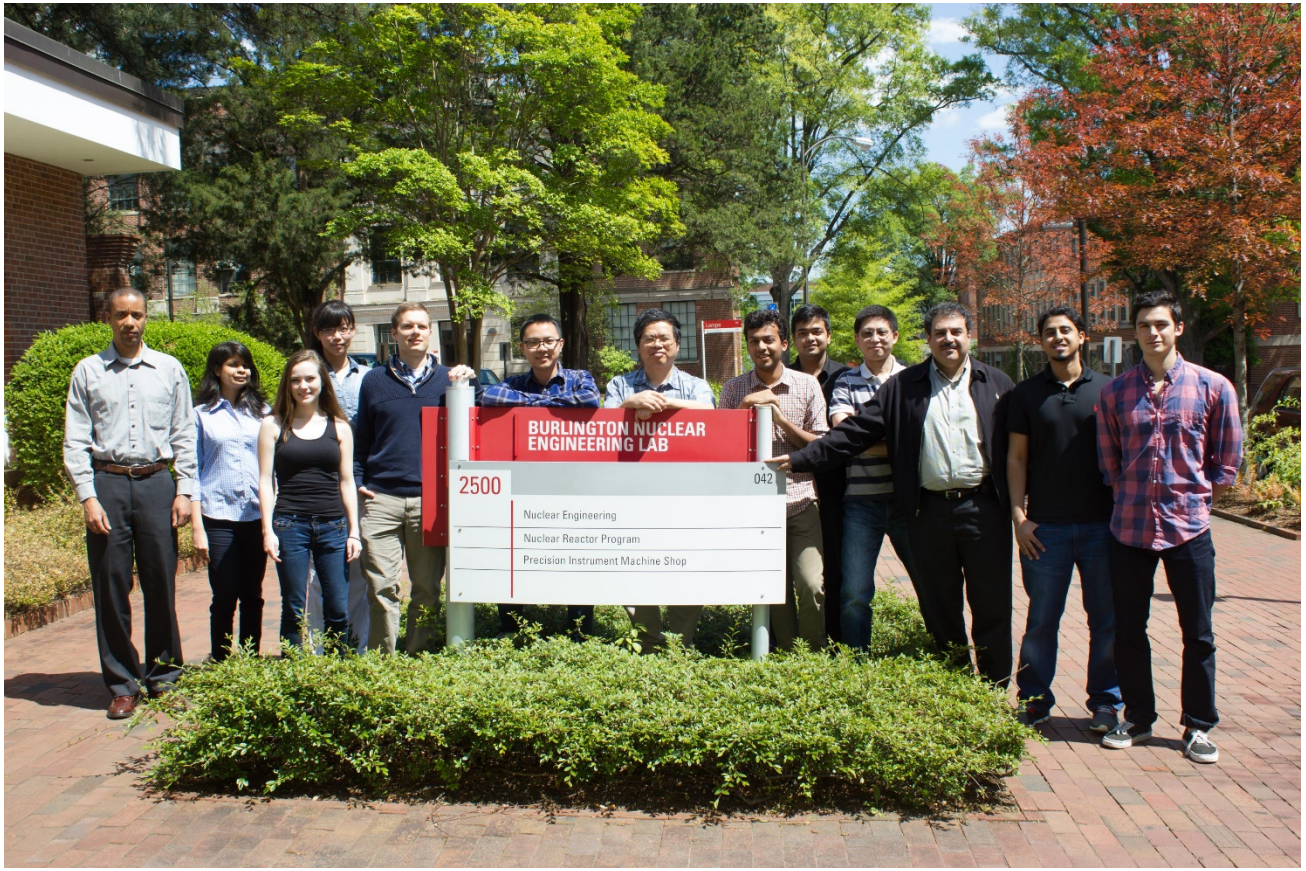
Acknowledgement

- The many graduate students and postdocs at North Carolina State University

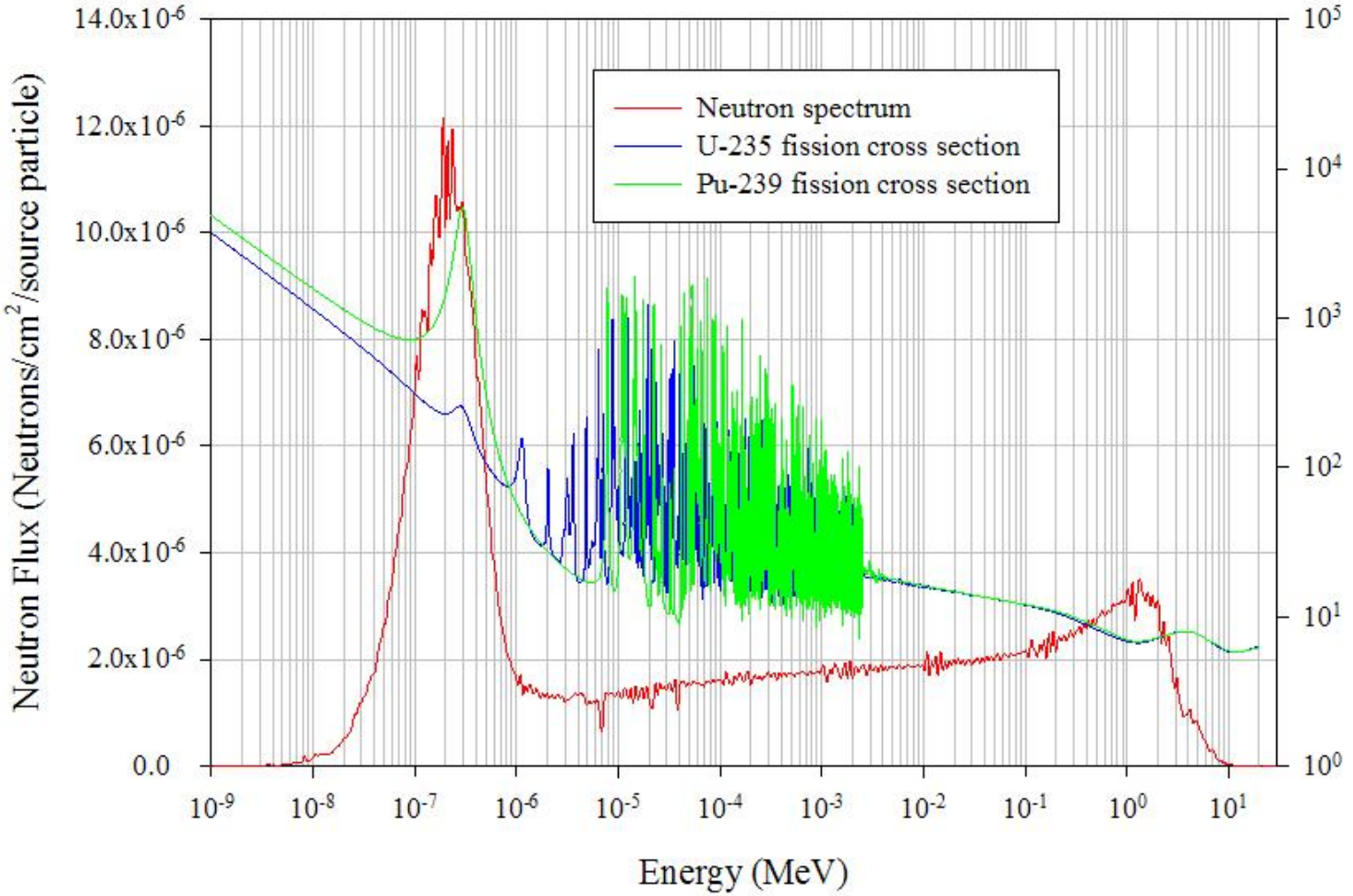


Acknowledgement

- Collaboration with LLNL and Bettis labs

- Current funding by the NCSP and NR programs and past funding by DOE NE through NERI and NEUP programs

Motivation


Neutron spectrum
U-235 fission cross section
Pu-239 fission cross section
Neutron Flux (Neutrons/cm^2/source particle)
14.0x10^{-6}
12.0x10^{-6}
10.0x10^{-6}
8.0x10^{-6}
6.0x10^{-6}
4.0x10^{-6}
2.0x10^{-6}
0.0
10^5
10^4
10^3
10^2
10^1
10^0
10^{-9} 10^{-8} 10^{-7} 10^{-6} 10^{-5} 10^{-4} 10^{-3} 10^{-2} 10^{-1} 10^0 10^1
Energy (MeV)

Example

CONF-880911--23

DE89 003625

THE EFFECT OF CARBON CRYSTAL STRUCTURE ON TREAT REACTOR PHYSICS CALCULATIONS

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Work Supported by the U. S. Department of Energy, Nuclear Energy Programs, under Contract W-31-109-Eng-38.

submitted to:

1988 International Reactor Physics Conference
Jackson Hole, Wyoming
18-21 September 1988

Table II

Calculated Values of k-effective

Calculational Description	k-effective
Loading 1341 100% Graphite	0.9724 ± 0.0021
Loading 1341 59% Graphite	0.9921 ± 0.0012
Loading 1343 100% Graphite	0.9707 ± 0.0024
Loading 1343 59% Graphite	0.9922 ± 0.0017

Table III

Calculated Fission Density Ratios

Calculational Description	Calculated Fission Density Ratio	Calc.-Exp. / Exp.
100% Graphite	43.2 ± 1.4	+6.9%
59% Graphite	39.4 ± 1.2	-2.5%

Vision

- Establish a predictive approach for generating the needed data (cross sections) to describe the energy exchange of thermal neutrons in matter
- Various applications:
  - Nuclear criticality safety
  - Nuclear reactor design
  - Neutron beam spectral shaping (i.e., filtering)
  - Neutron source (cold, ultracold, etc.) characterization

Thermal Neutron Scattering

Using Born approximation, it can be shown that the double differential scattering cross section has the form

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{1}{4\pi}\frac{k'}{k}\left\{\sigma_{coh}S(\vec{\kappa},\omega) + \sigma_{incoh}S_s(\vec{\kappa},\omega)\right\}$$

The scattering law $S(\vec{\kappa},\omega)$ is composed of two parts

$$S(\vec{\kappa},\omega) = S_s(\vec{\kappa},\omega) + S_d(\vec{\kappa},\omega)$$

Van Hove's formulation

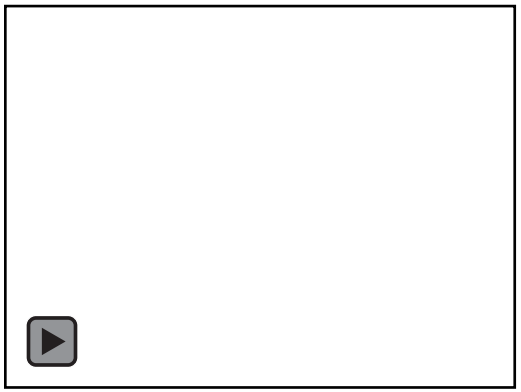
$$I(\vec{\kappa},t) = \int G(\vec{r},t)\exp(i\vec{\kappa}\cdot\vec{r})d\vec{r}$$

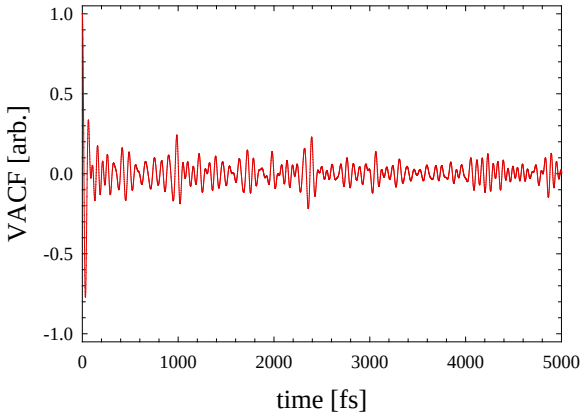
$$S(\vec{\kappa},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} G(\vec{r},t)e^{i(\vec{\kappa}\cdot\vec{r}-\omega t)}d\vec{r}dt$$

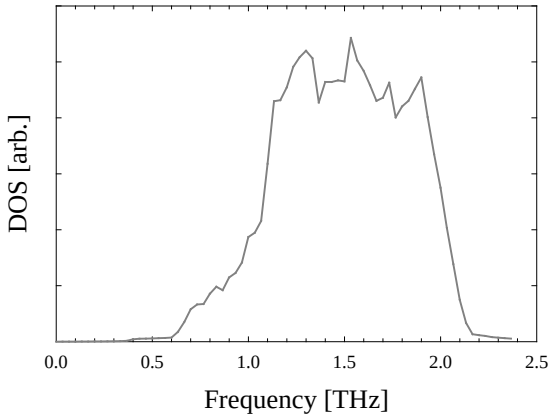
where G(r,t) is the *dynamic pair correlation function* and is expressible in terms of the atomic positions.

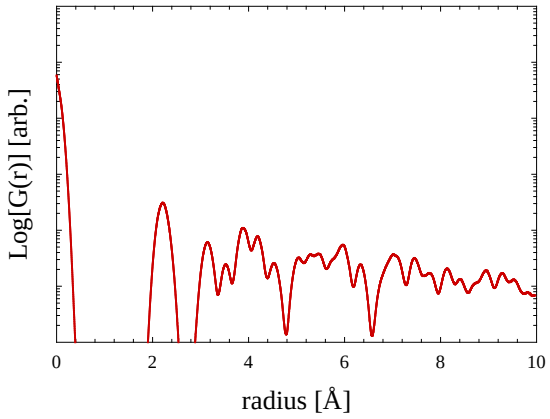
Correlation Functions

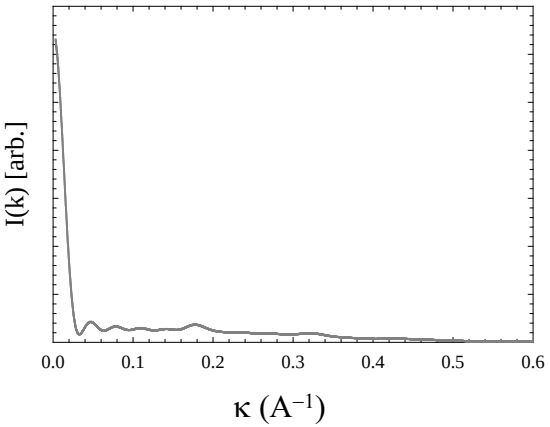
- Fourier transforms of correlation functions in space and time reveal the energy and momentum states available within the system





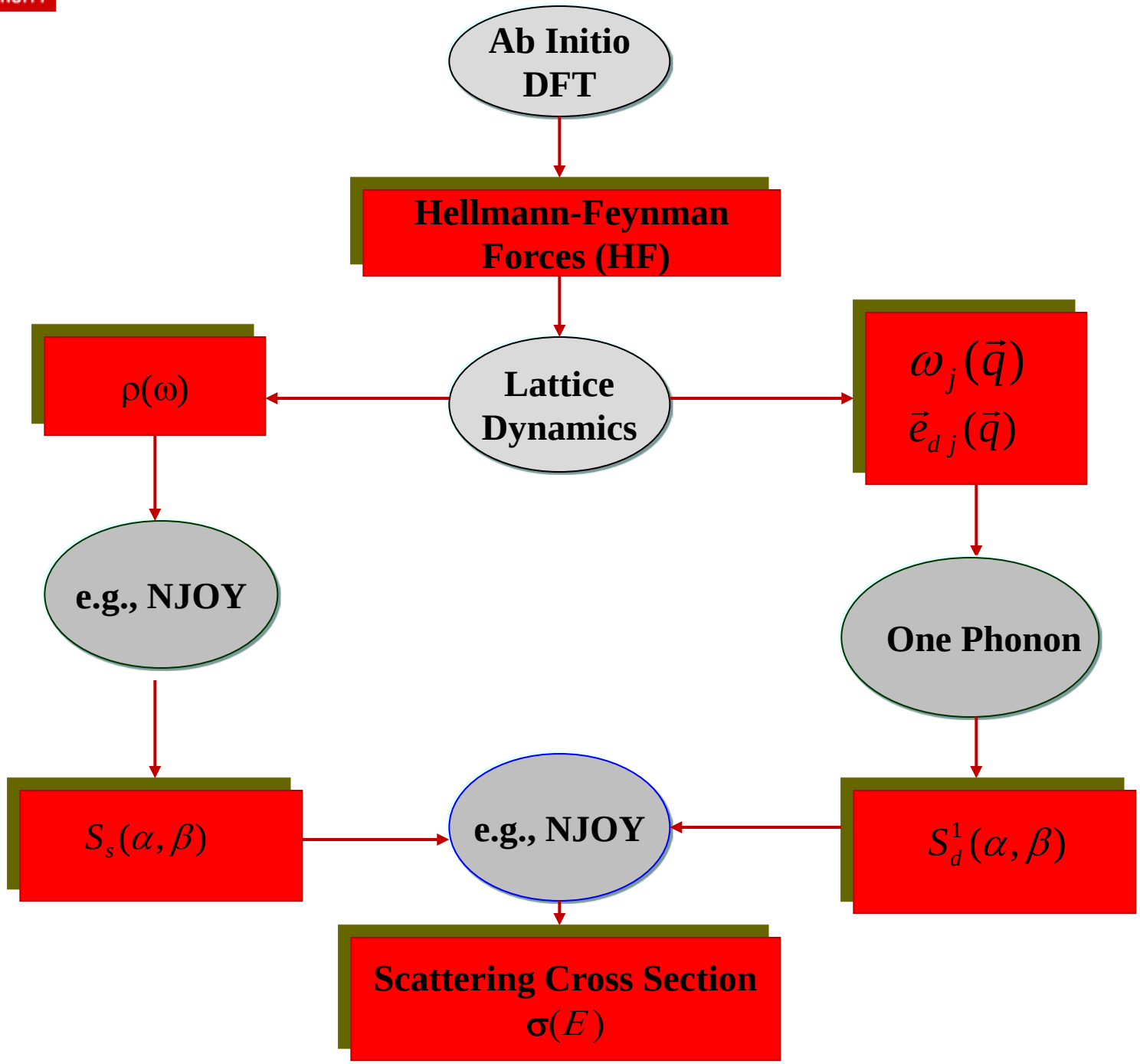


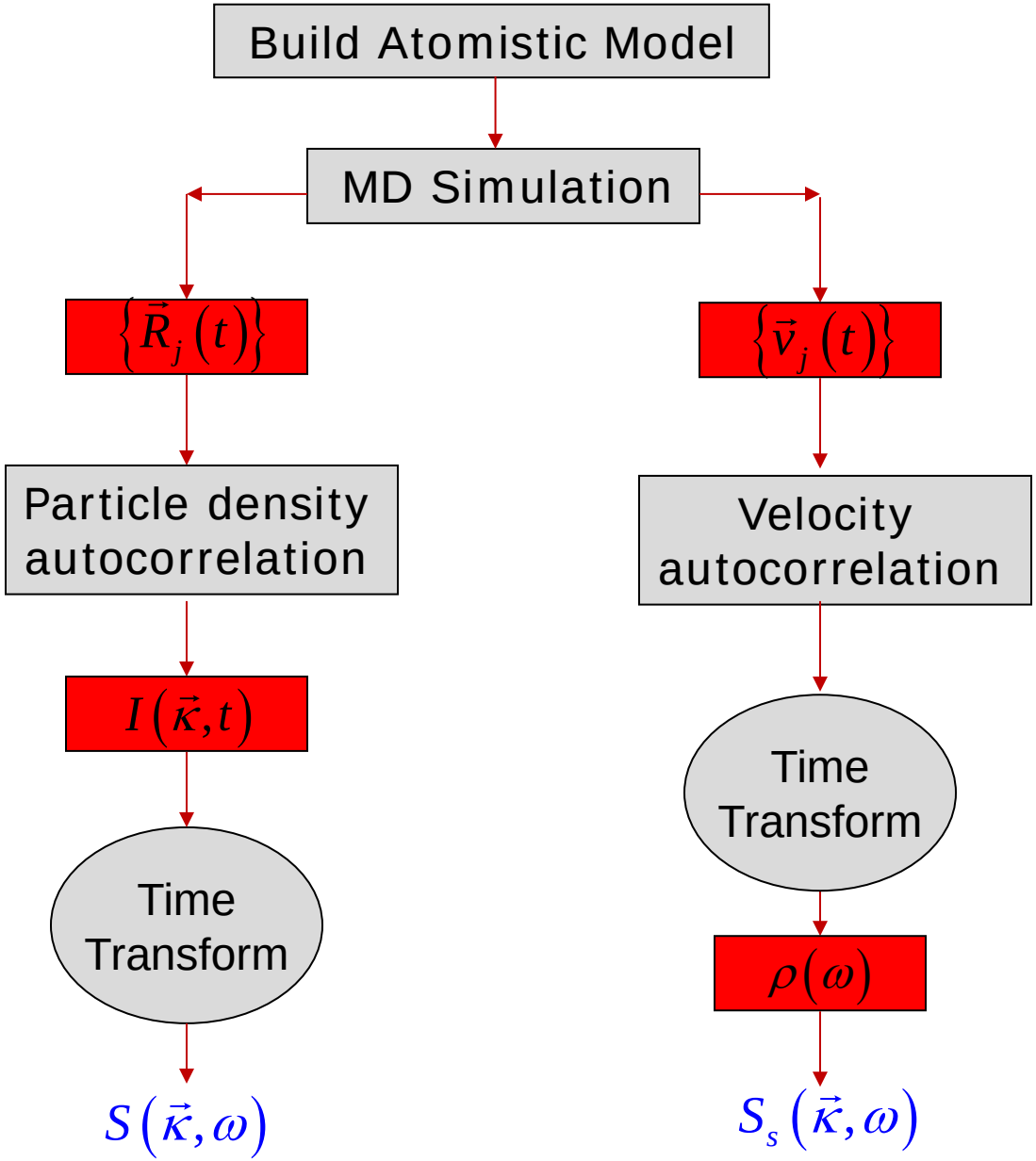




Methods

- Several approaches can be used to extract the fundamental information for calculating the scattering law and eventually the cross sections
  - Empirical atomic force analysis combined with dynamical matrix calculations
    - Basis of current ENDF/B libraries
  - Ab initio Quantum (DFT) methods combined with dynamical matrix calculations
  - Molecular Dynamics (ab initio MD or classical MD) methods combined with correlation function analysis


Ab Initio DFT
Hellmann-Feynman Forces (HF)
Lattice Dynamics
ρ(ω)
$\omega_j(\vec{q})$
$\vec{e}_{dj}(\vec{q})$
e.g., NJOY
One Phonon
$S_s(\alpha,\beta)$
e.g., NJOY
$S_d^1(\alpha,\beta)$
Scattering Cross Section
σ(E)

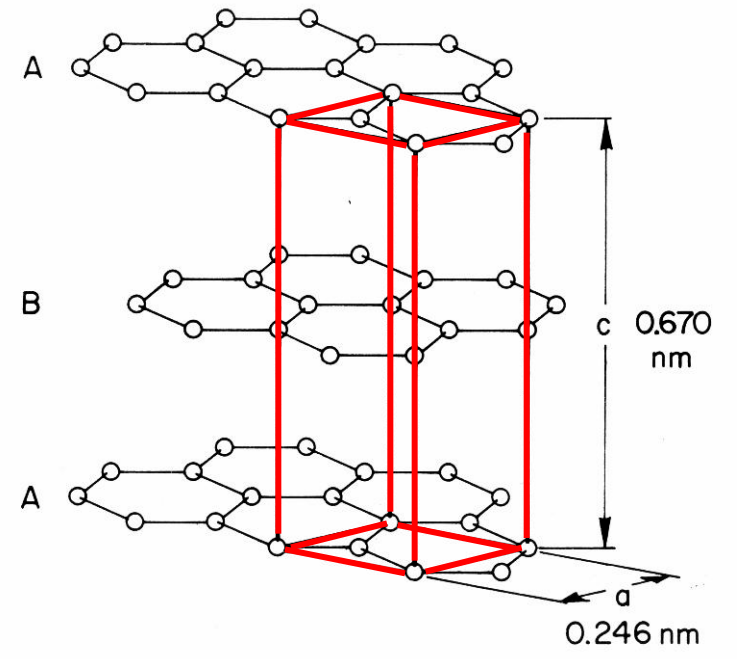

Build Atomistic Model
MD Simulation
$\{\vec{R}_j(t)\}$
$\{\vec{v}_j(t)\}$
Particle density autocorrelation
Velocity autocorrelation
$I(\vec{\kappa},t)$
Time Transform
Time Transform
$\rho(\omega)$
$S(\vec{\kappa},\omega)$
$S_s(\vec{\kappa},\omega)$

Materials Studied at NCSU

- Graphite, Beryllium (PHYSOR 2004 & 2008)
  - Treatment of nuclear graphite (porous system)
  - Including coherent inelastic for both materials
- Silicon dioxide (New, contributed to NNDC/ENDF, PHYSOR 2008)
  - Support criticality safety analysis
- Silicon carbide (New, contributed to NNDC/ENDF)
  - Support advanced fuel cycle applications (e.g., FCM fuels)
- Thorium hydride, uranium-zirconium hydride, calcium hydride (PHYSOR 2004)
- Sapphire and bismuth (PHYSOR 2006)
  - Thermal neutron filters
- Solid methane (predictive analysis – AccApp 2011)
  - Cold neutron moderator
  - Captured phase I to II transformation upon cooling below 22 K
- Lucite ($C_5O_2H_8$) and polyethylene (C_2H_4)
  - Of interest as moderators

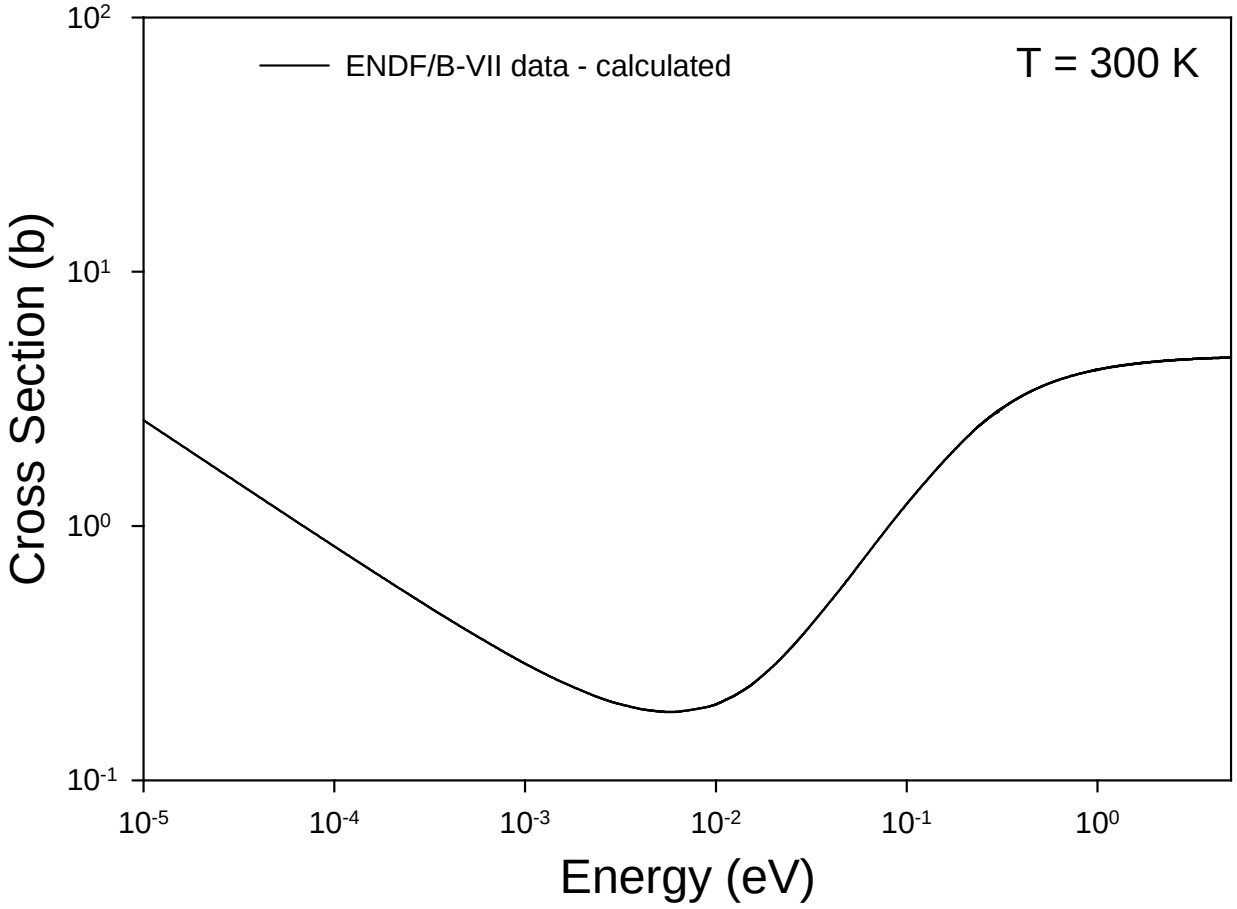
Graphite

Ideal graphite consists of planes (sheets) of carbon atoms arranged in a hexagonal lattice. Covalent bonding exits between intraplaner atoms, while the interplaner bonding is of the weak Van der Waals type. The planes are stacked in an "abab" sequence.

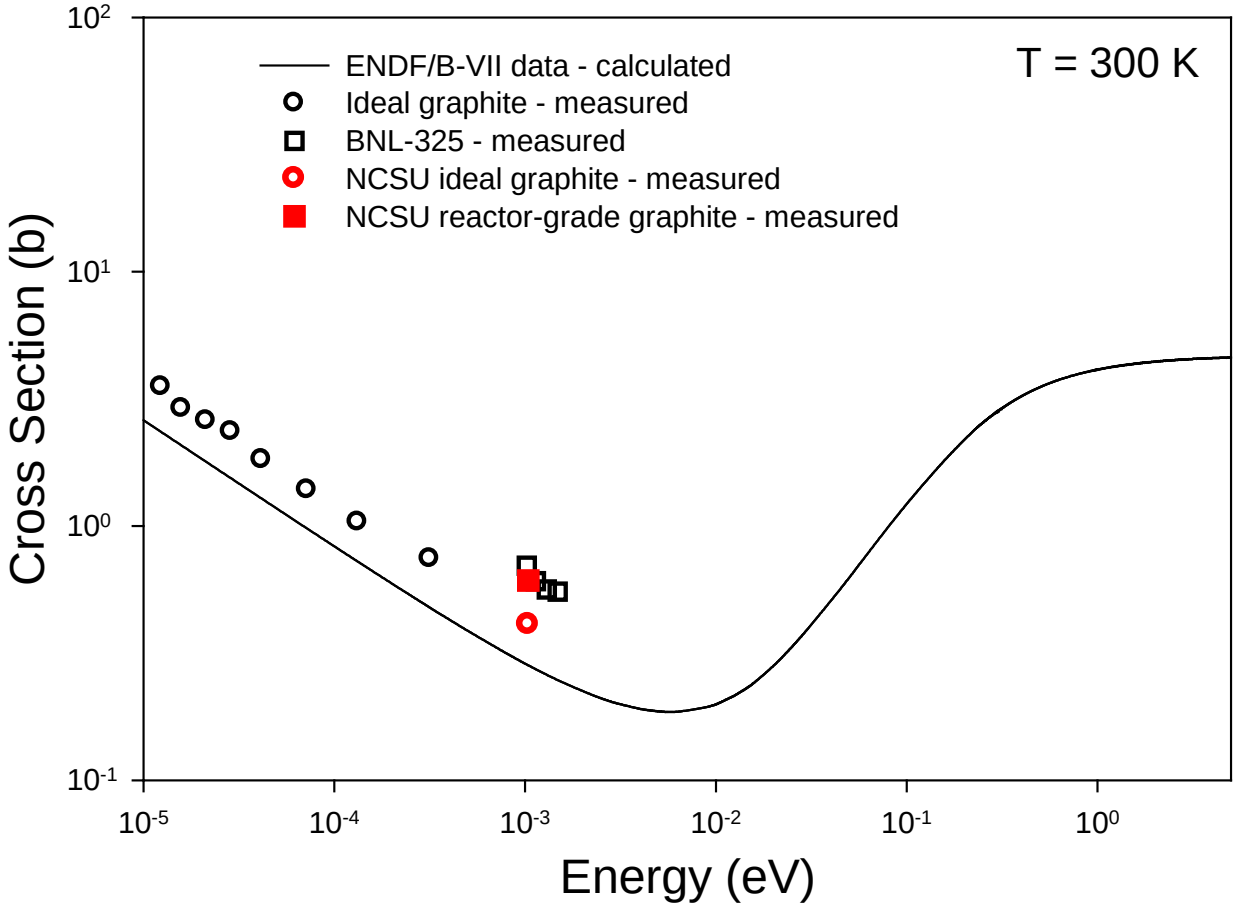


- Hexagonal Structure
- 4 atoms per unit cell
- a=b=2.46 Å
- c=6.7 Å

Graphite – 1


ENDF/B-VII data - calculated
T = 300 K
Cross Section (b)
10^{2}
10^{1}
10^{0}
10^{-1}
10^{-5}
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
Energy (eV)

Graphite – 2


ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

$S(\kappa,\omega)$ - particle density autocorrelation

Scattering law is computed directly from the atomic positions as:

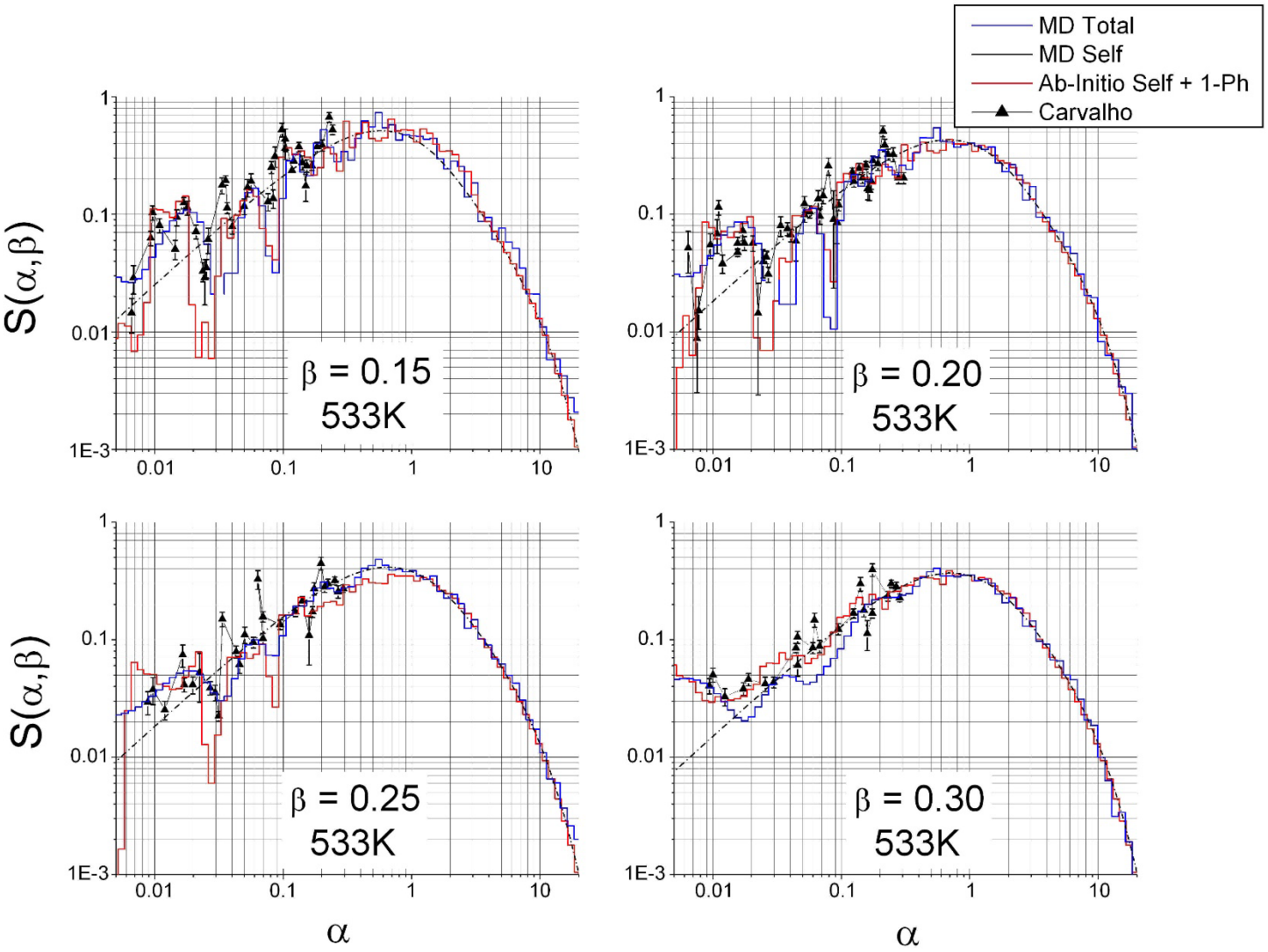
$$I^{cl}(\kappa,t)=\frac{1}{N}\sum_{j,j'}\left\langle \exp\left[-i\vec{\kappa}\cdot\vec{R}_{j'}(0)\right]\exp\left[i\vec{\kappa}\cdot\vec{R}_{j}(t)\right]\right\rangle$$

$$S^{cl}(\kappa,\omega)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} I(\kappa,t)\exp(-i\omega t)\,dt$$

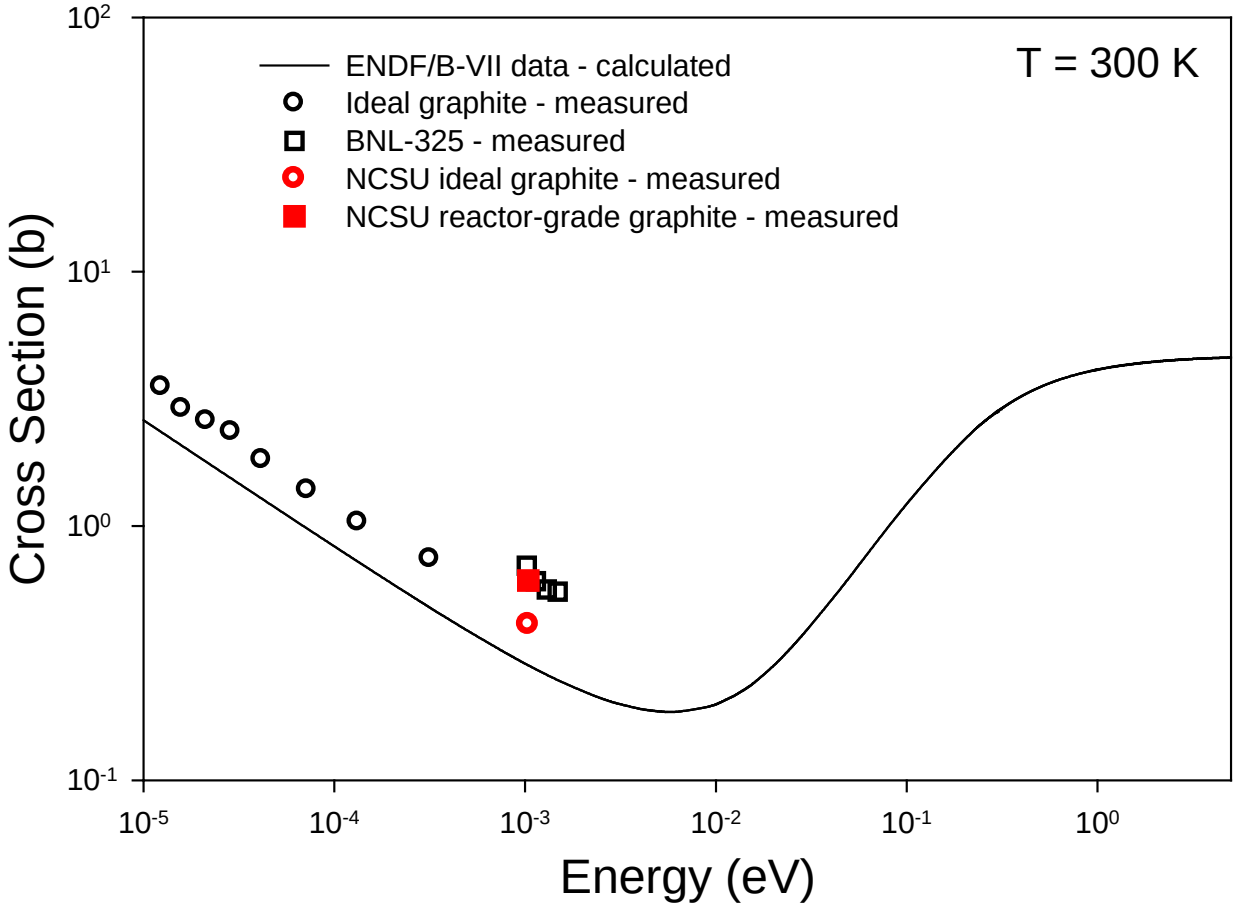
where the positions are known at discrete time steps and the Fourier transforms are taken in discrete form.

- Detailed balance
- Response of scattering system to neutron interaction

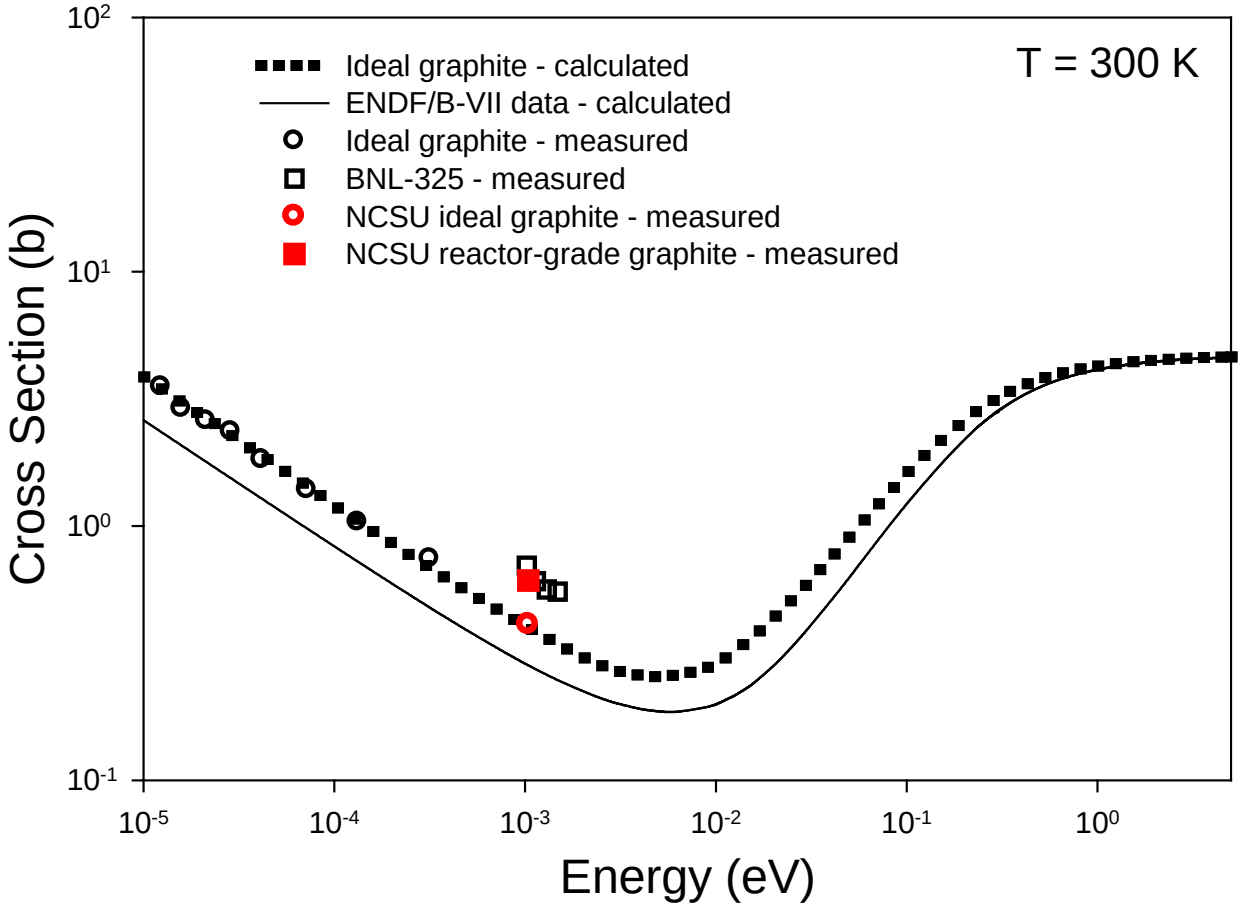
Graphite Scattering Law



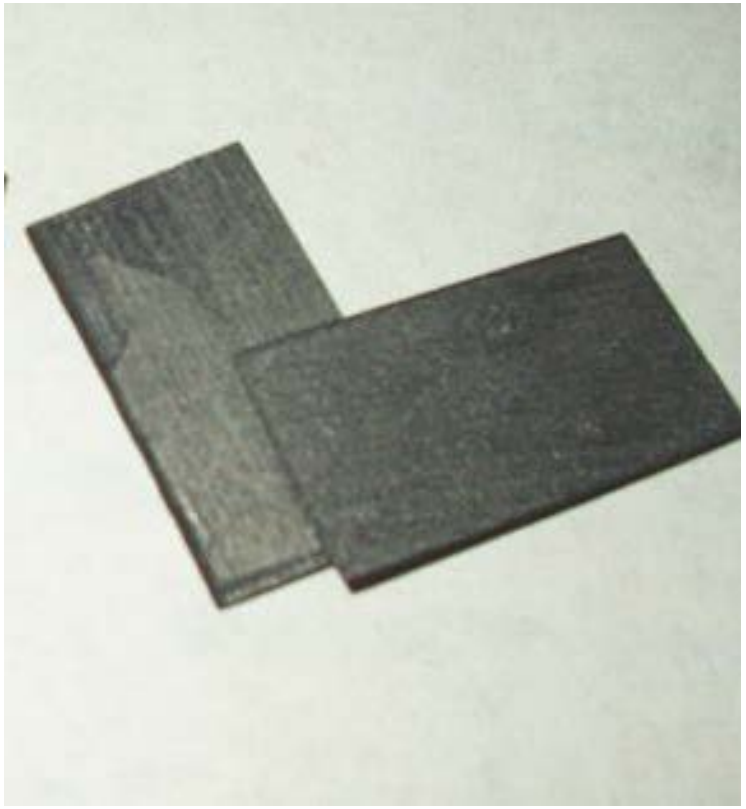
Graphite – 2


ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

Graphite – 3

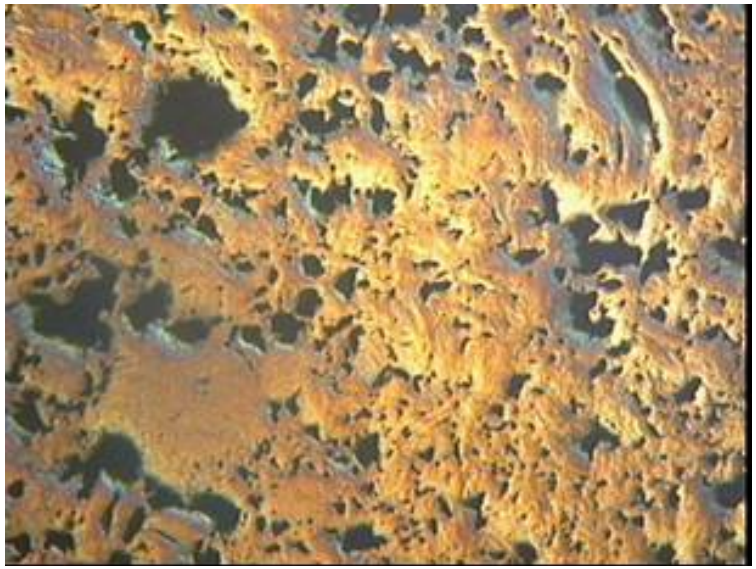

Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

Graphite Types



Ideal Graphite

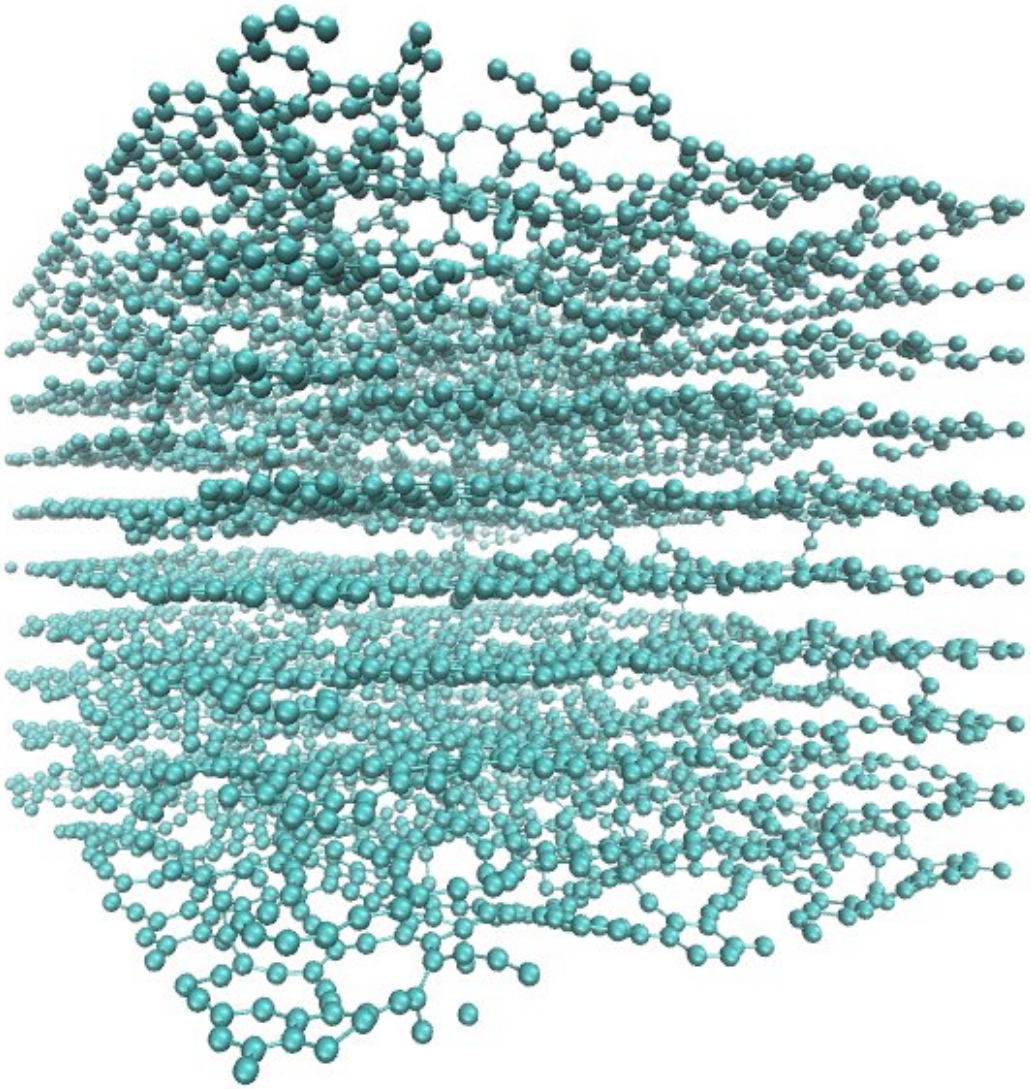
Density = 2.25 g/cm^3



Nuclear Graphite

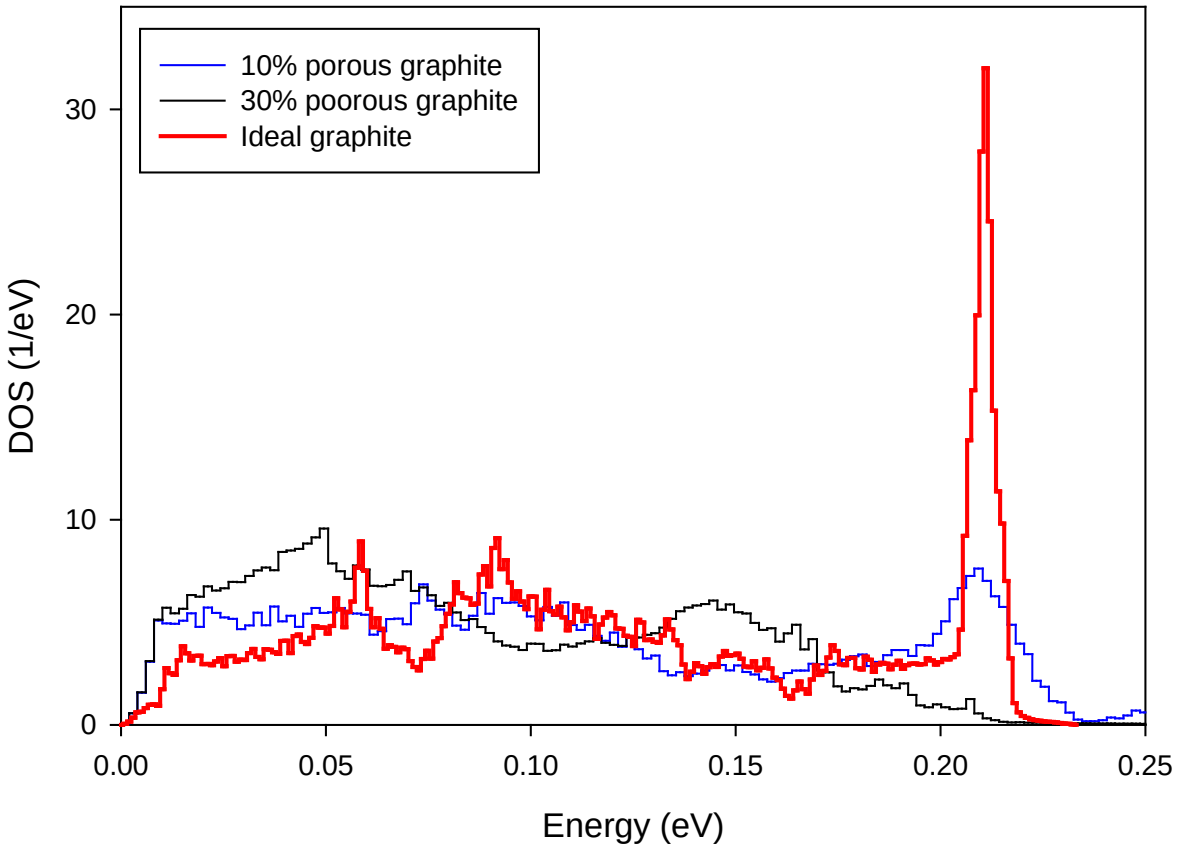
Density = 1.5 – 1.8 g/cm^3

MD Models

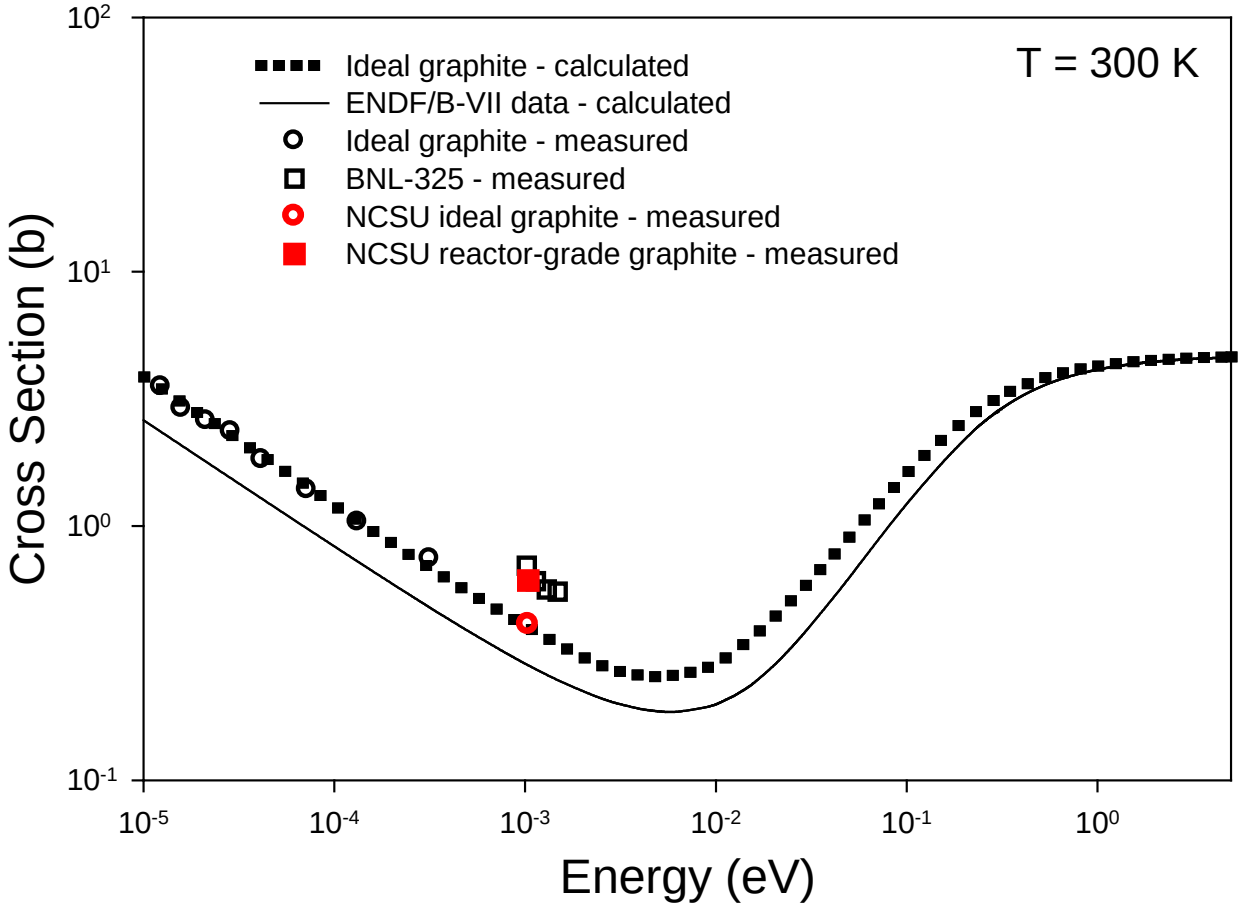


8000 atoms
30% porosity
NVT ensemble
T = 300 K

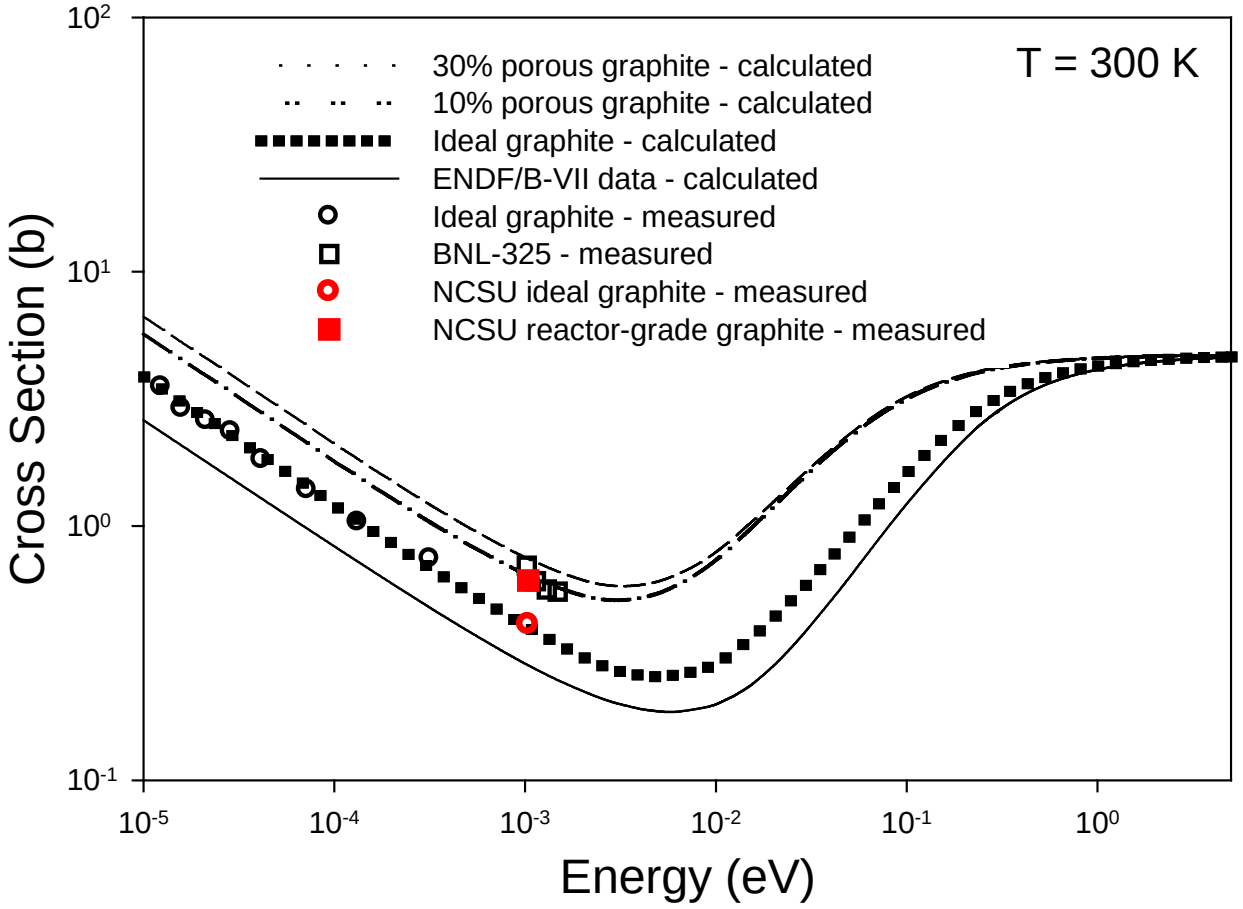
MD Models


10% porous graphite
30% poorous graphite
Ideal graphite
DOS (1/eV)
0
10
20
30
0.00
0.05
0.10
0.15
0.20
0.25
Energy (eV)

Graphite – 3

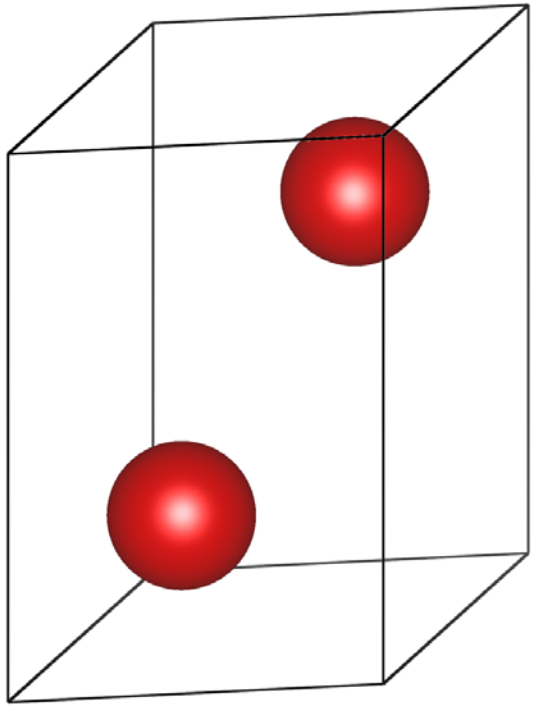

Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
T = 300 K
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

Graphite – 4


T = 300 K
30% porous graphite - calculated
10% porous graphite - calculated
Ideal graphite - calculated
ENDF/B-VII data - calculated
Ideal graphite - measured
BNL-325 - measured
NCSU ideal graphite - measured
NCSU reactor-grade graphite - measured
Cross Section (b)
Energy (eV)
10^2
10^1
10^0
10^-1
10^-5
10^-4
10^-3
10^-2
10^-1
10^0

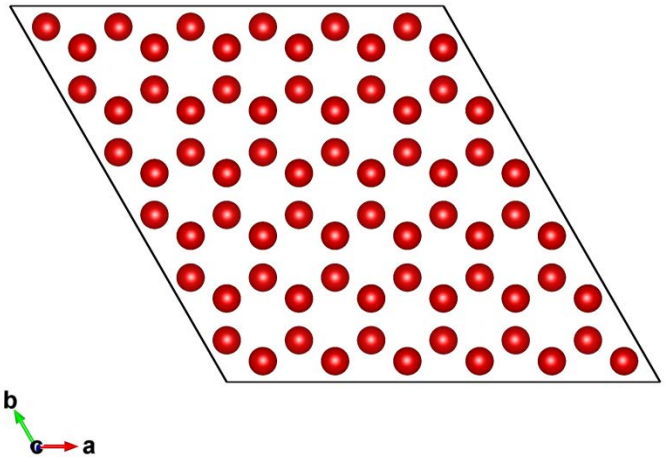
AIMD Model of Be

- HCP ($P6_3/mmc$)
  - a=2.2856 (2.27 AIMD)
  - c=3.5832 (3.55 AIMD)
- VASP
  - GGA-PAW
  - 3x3x3 k-mesh
  - 350eV Plane-wave cut-off

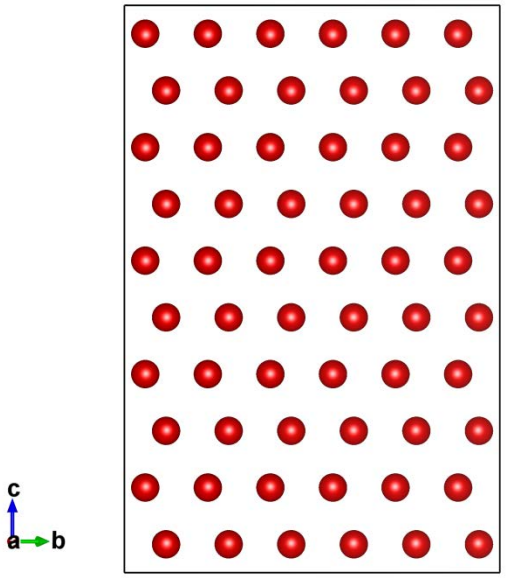


AIMD Model of Be

- VASP 5x5x5 super-cell (250 atoms)
- Temperature of 300K under NVE
- Equilibrated with velocity scaling (1.5ps)
- 10ps simulation with 1fs time steps



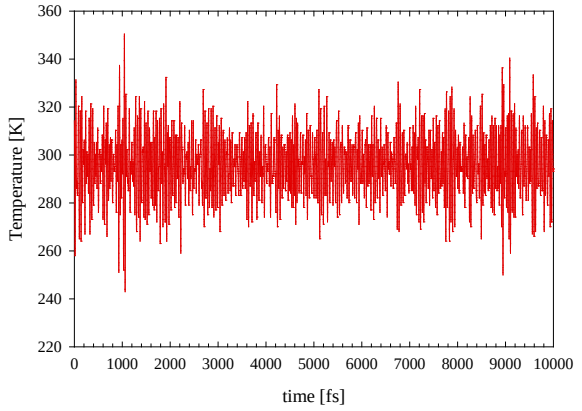
Supercell: c-axis

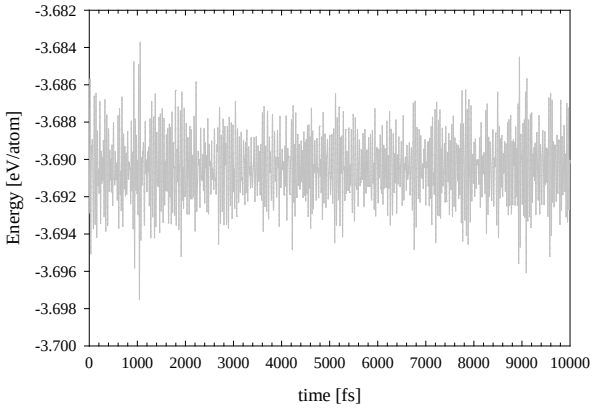


Supercell: a-axis

Verification of Model Behavior

- Atoms well-behaved
  - No-Diffusion
  - MSD
  - Velocity Distribution
- Temperature Fluctuations
  - Reasonable standard deviation
- Energy Fluctuations
  - Relative fluctuation must be small





Computational Approach

$$C(t) = \left\langle \overrightarrow{v_j(0)} \cdot \overrightarrow{v_j(t)} \right\rangle = \frac{1}{N} \sum_{j-1}^{N} \overrightarrow{v_j(0)} \cdot \overrightarrow{v_j(t)}$$

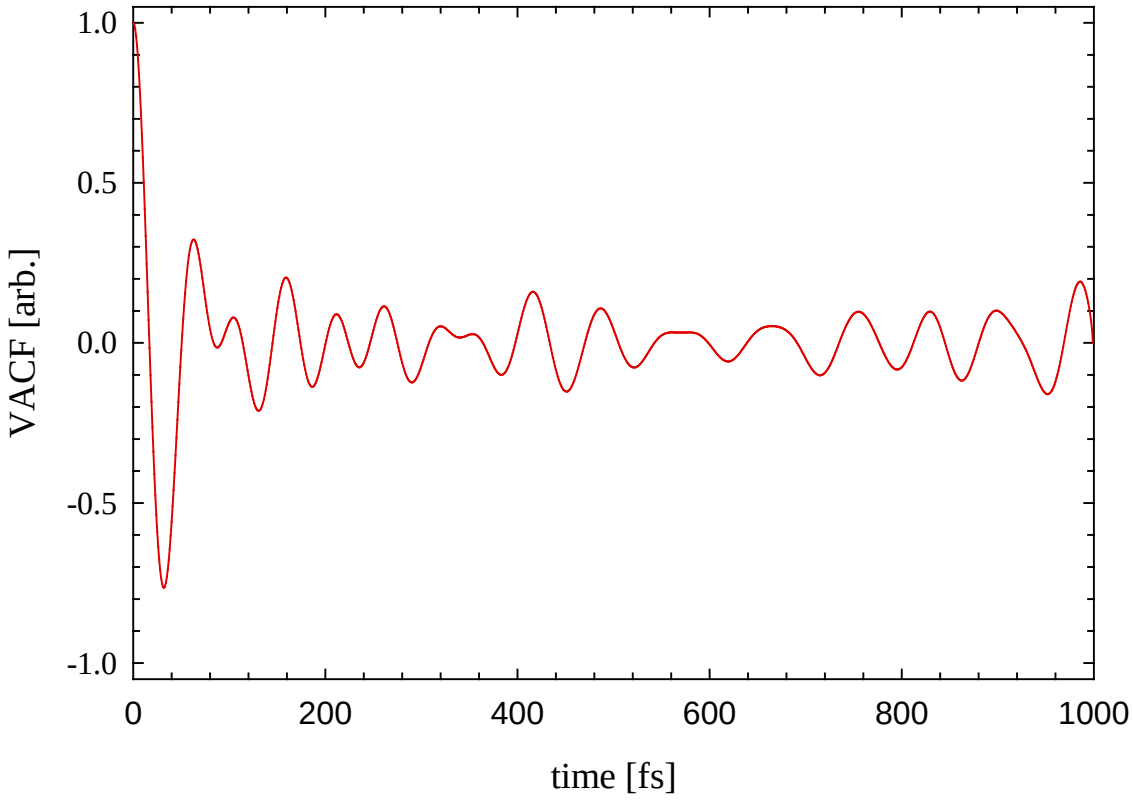
$$\rho(\omega) = \frac{M}{3\pi k_\beta T} \int_{-\infty}^{\infty} C(t) e^{-i\omega t} dt$$

Use NJOY/LEAPR

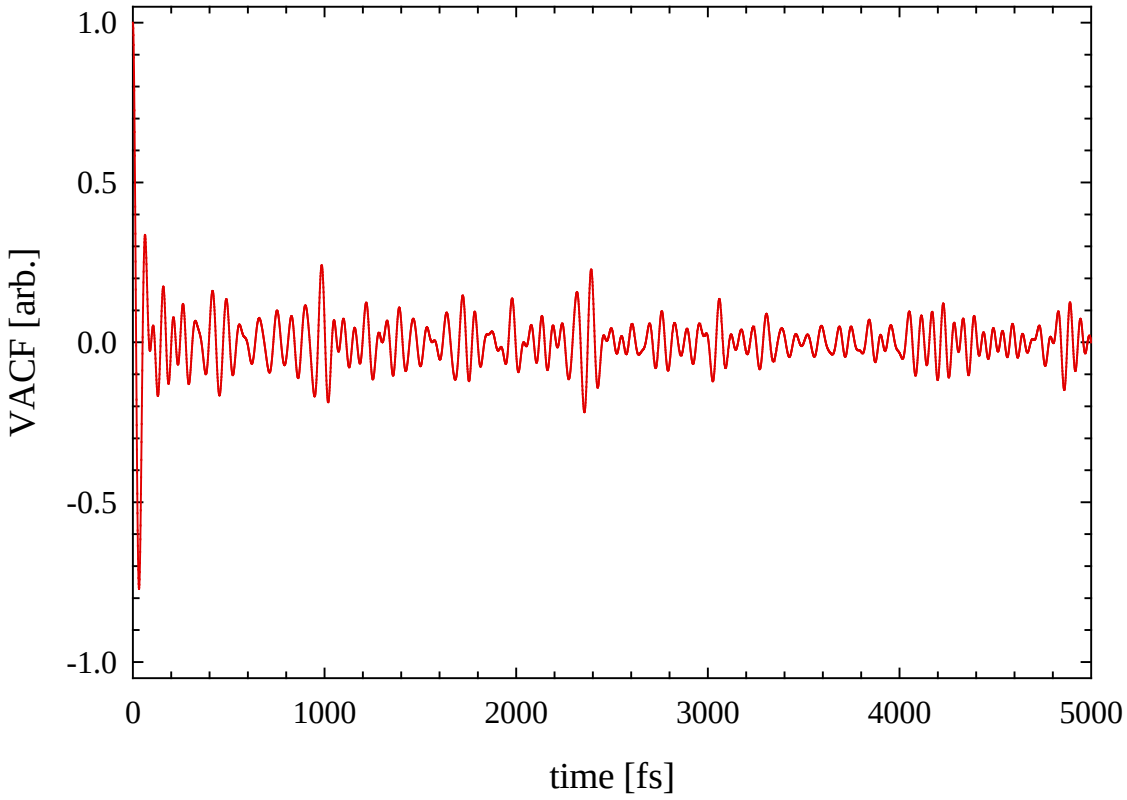
$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\beta t} e^{-\gamma(t, \rho(\omega))} dt$$

$$\left. \frac{d^2\sigma}{d\Omega dE} \right|_{\text{inelastic}} = \frac{\sigma}{2k_B T} \sqrt{\frac{E'}{E}} e^{-\frac{\beta}{2}} S(\alpha, \beta)$$

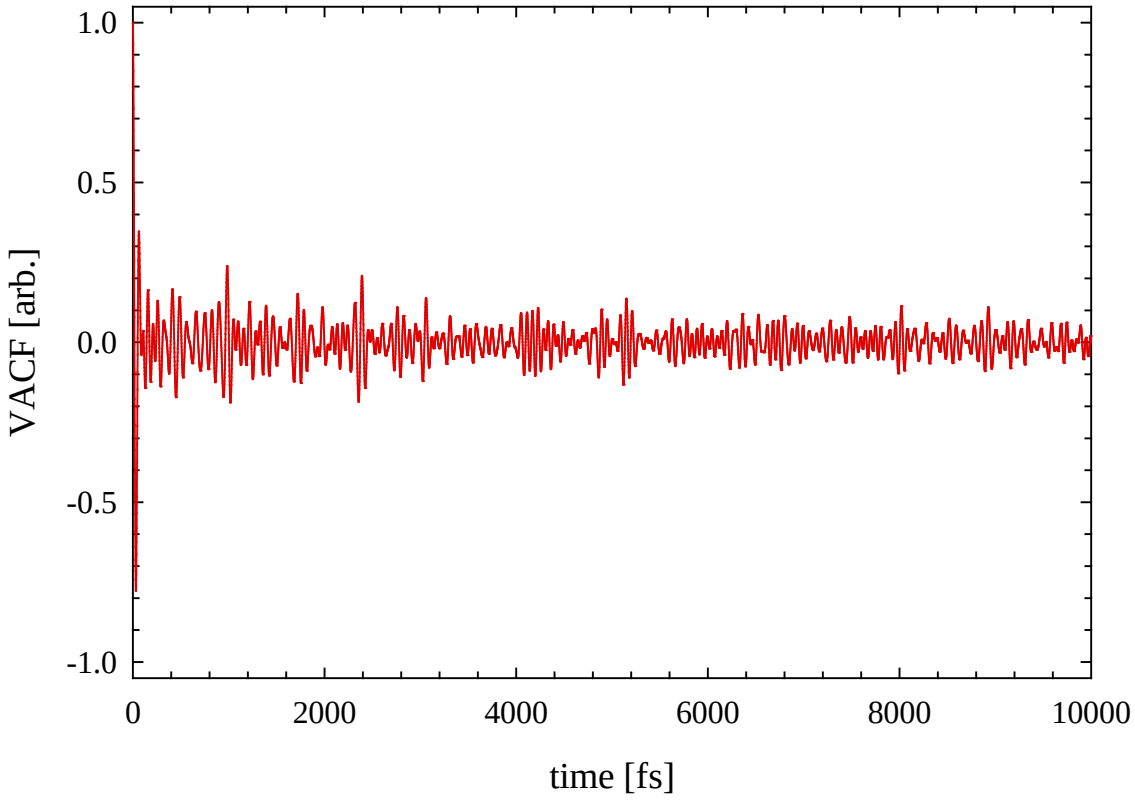
Velocity Auto-Correlation Function



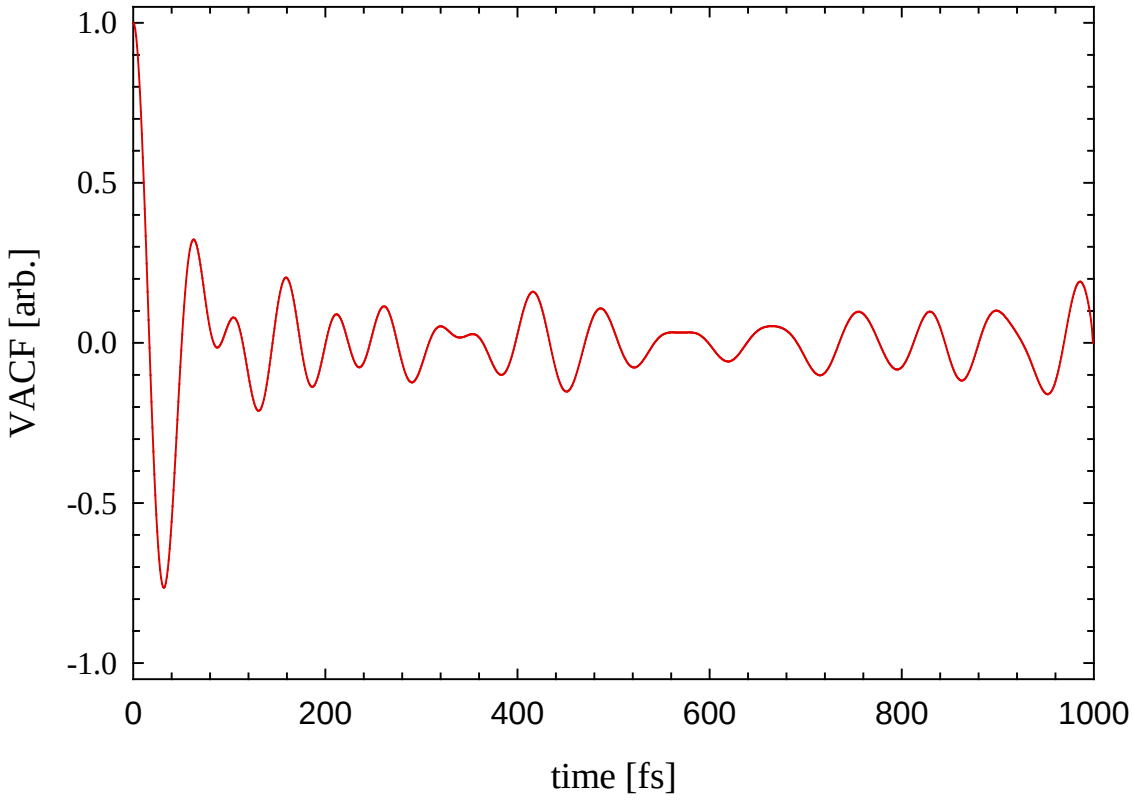
Velocity Auto-Correlation Function



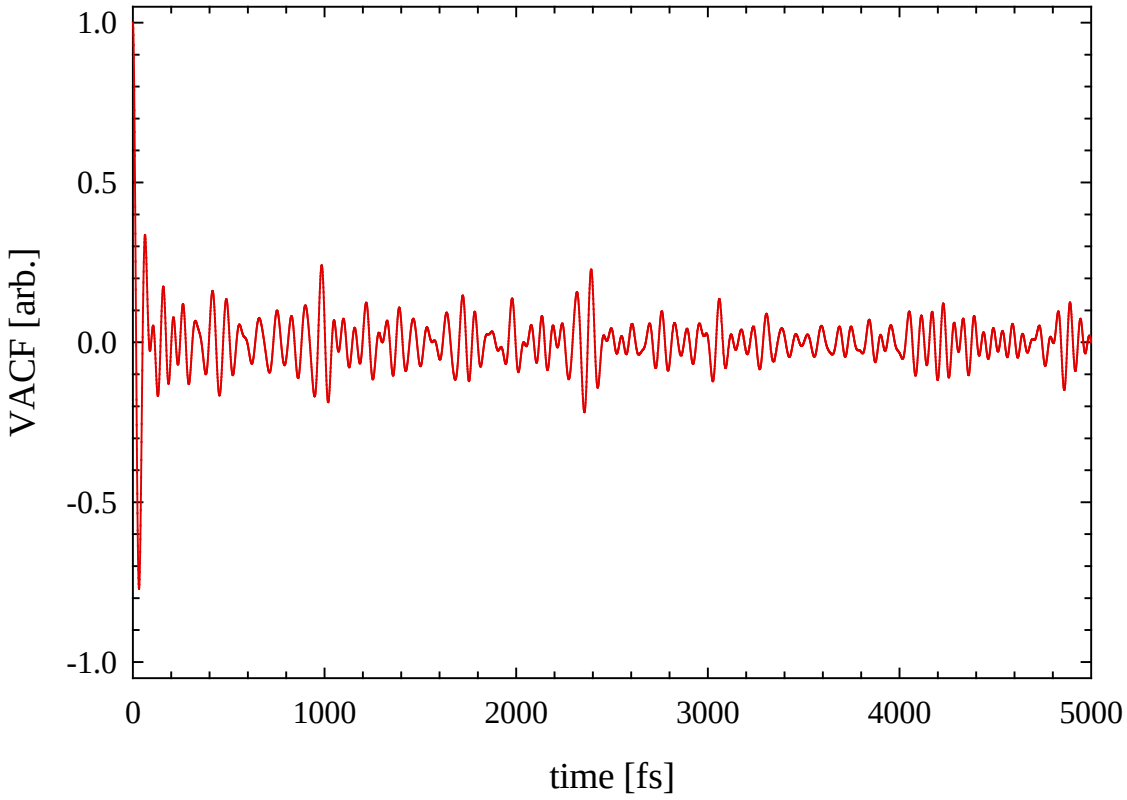
Velocity Auto-Correlation Function



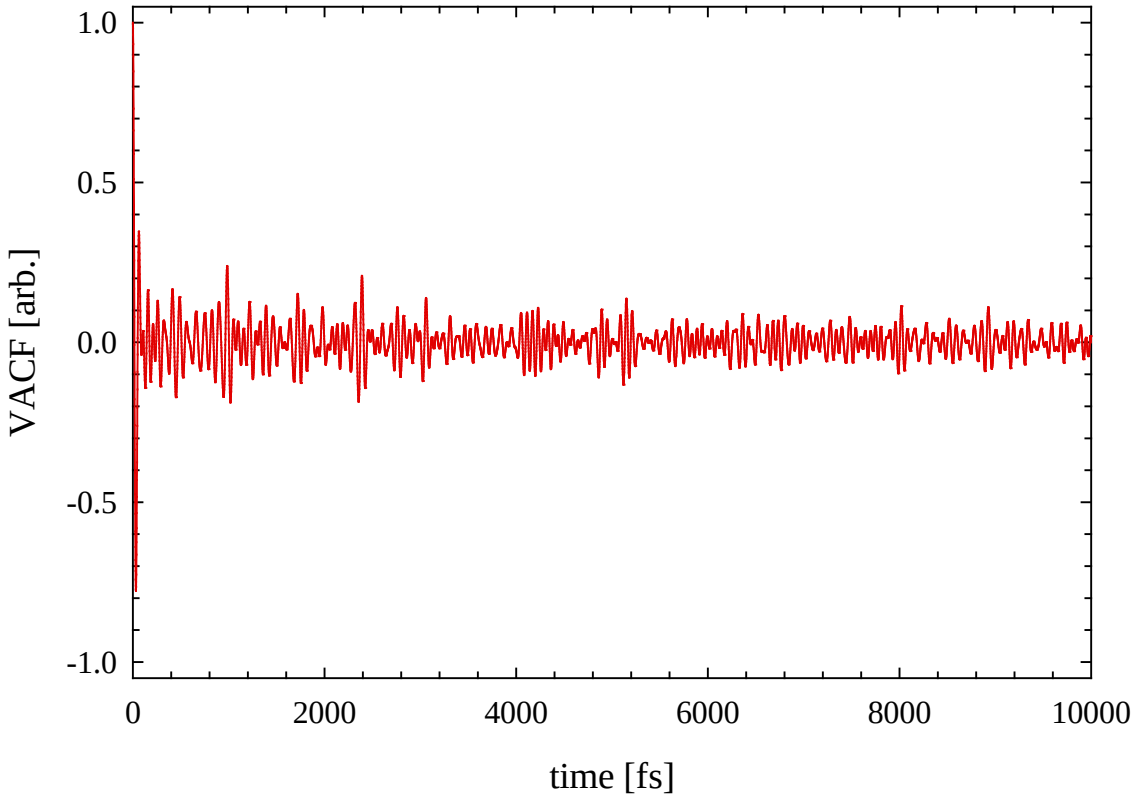
Velocity Auto-Correlation Function



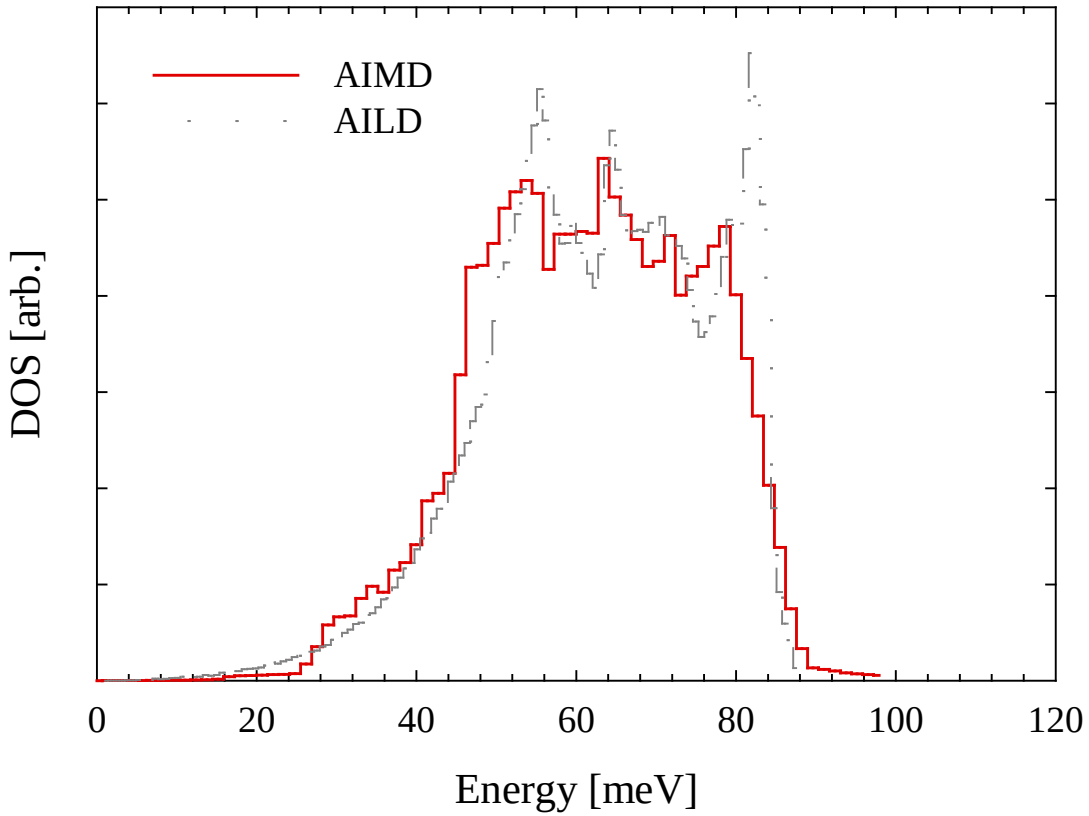
Velocity Auto-Correlation Function



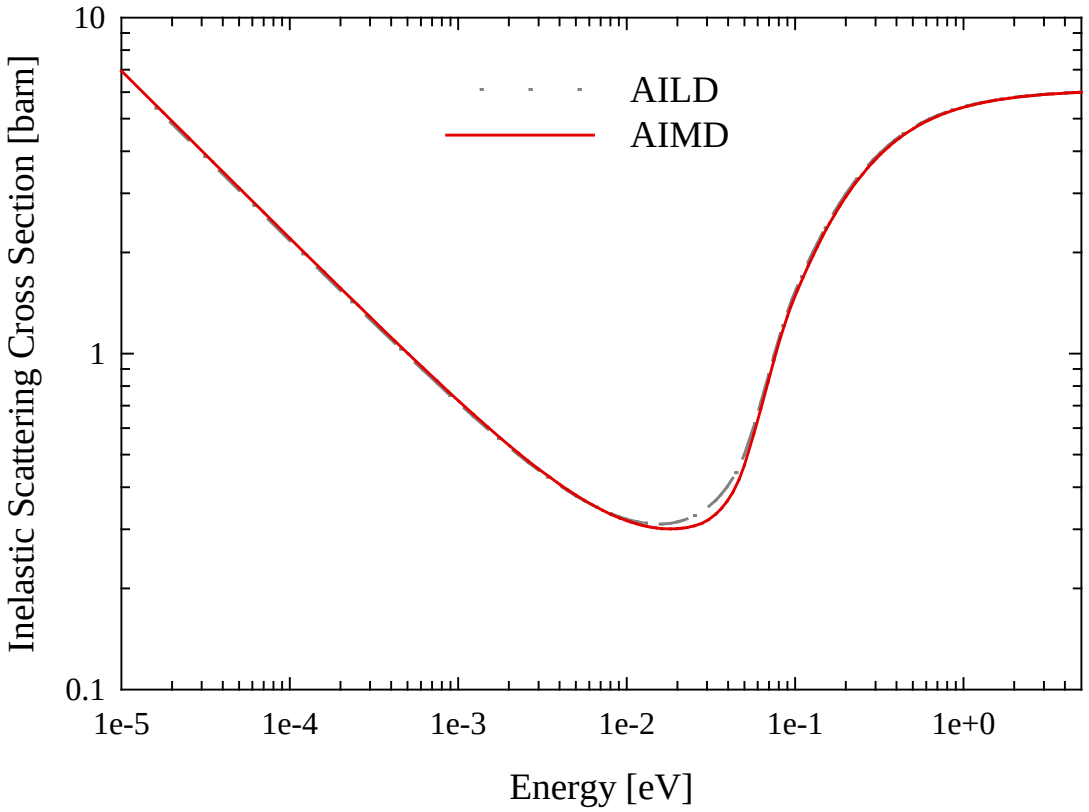
Velocity Auto-Correlation Function



Phonon Density of States

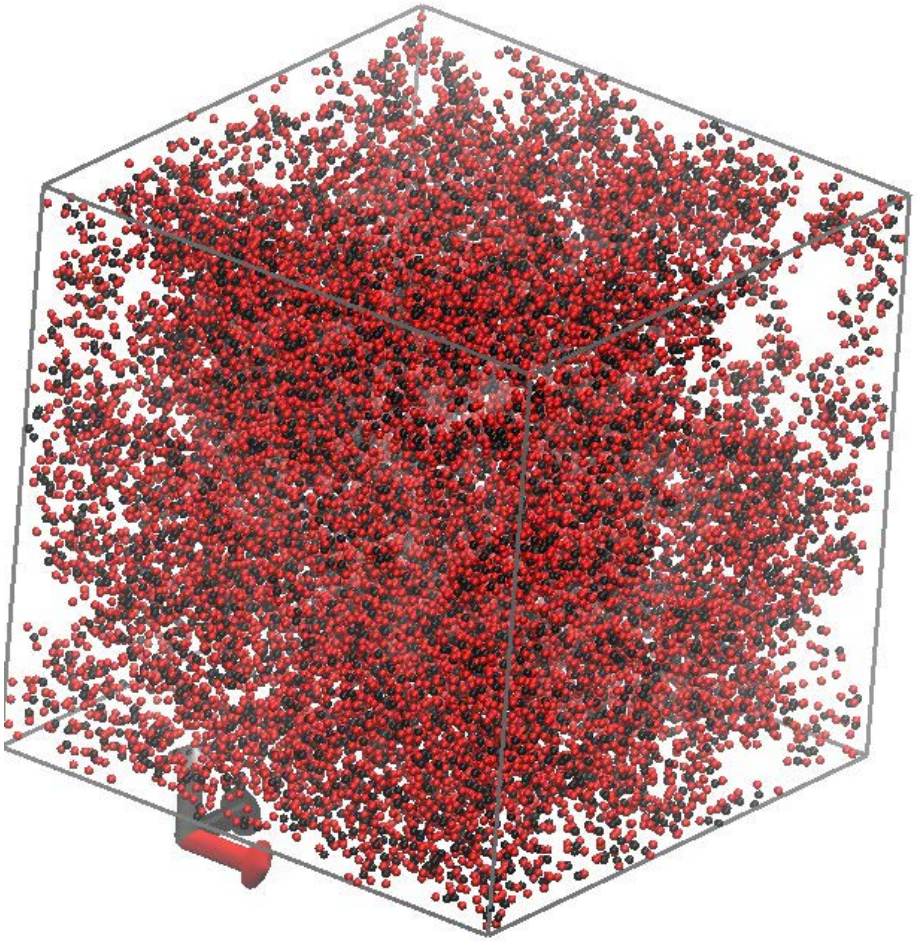


Inelastic Thermal Neutron Scattering Cross Section

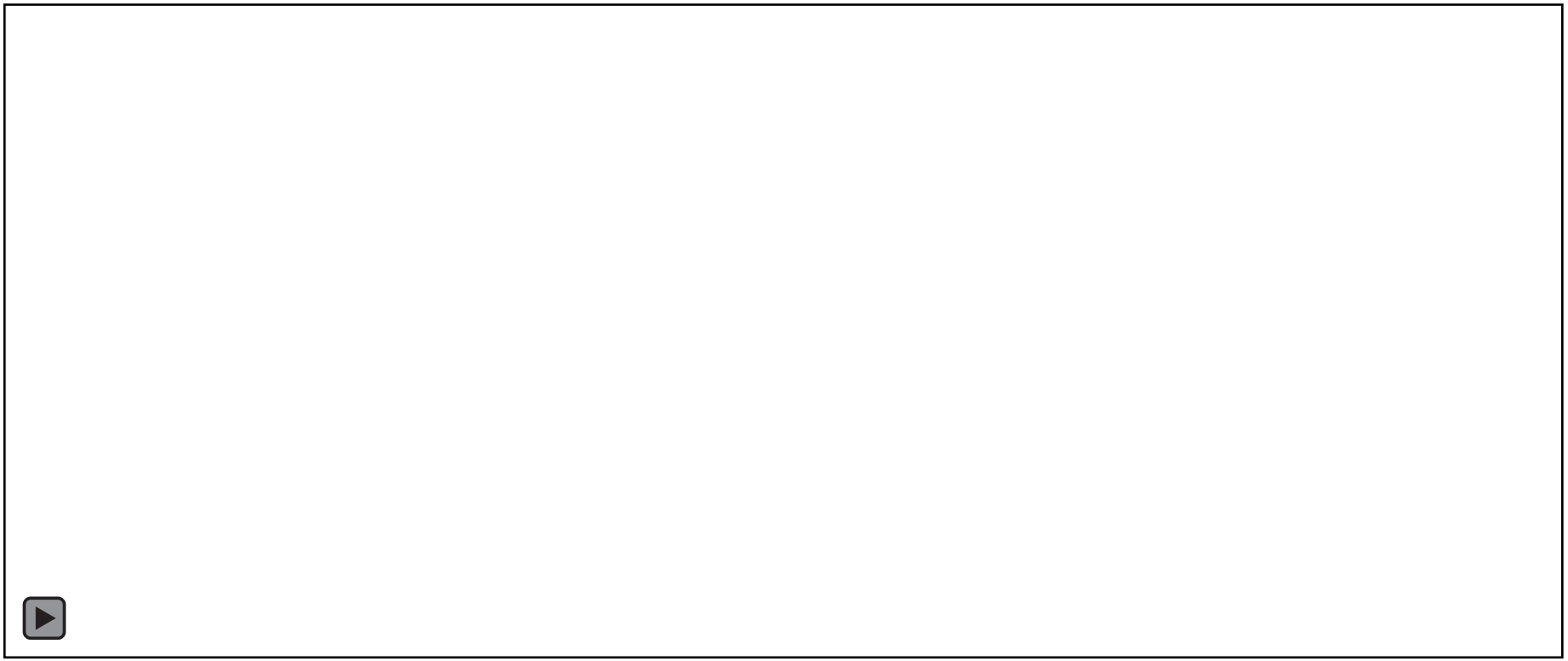


Polyethylene System

- Systems of multiple polymer chains are needed to account for inter-chain interactions.
- Initial system, 20 polymer chains, each 200 monomers long, 24,000 atoms.



MD Simulation



Summary

- Developed a modern approach for thermal neutron cross section calculations based on the use of atomistic simulations
  - Ab initio lattice dynamics
  - Molecular dynamics
- The approach is predictive
  - New materials
  - All states of matter (solid, liquid, gas)
  - Imperfect structure