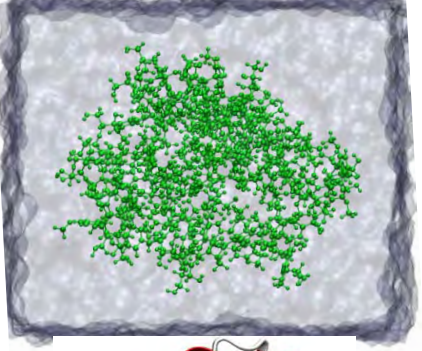
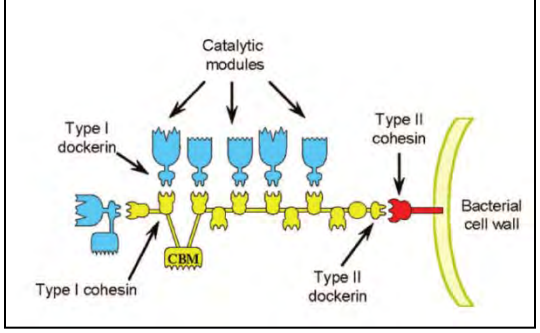
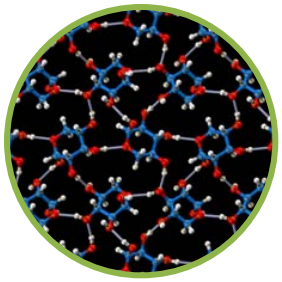
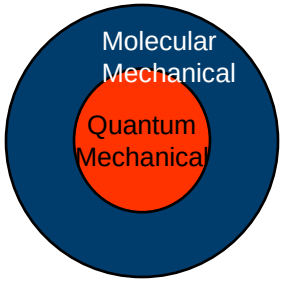
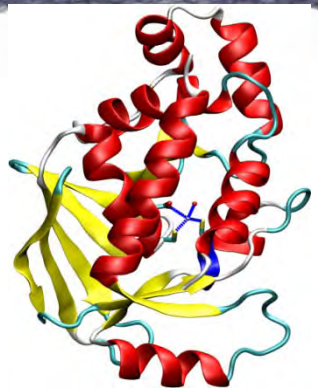








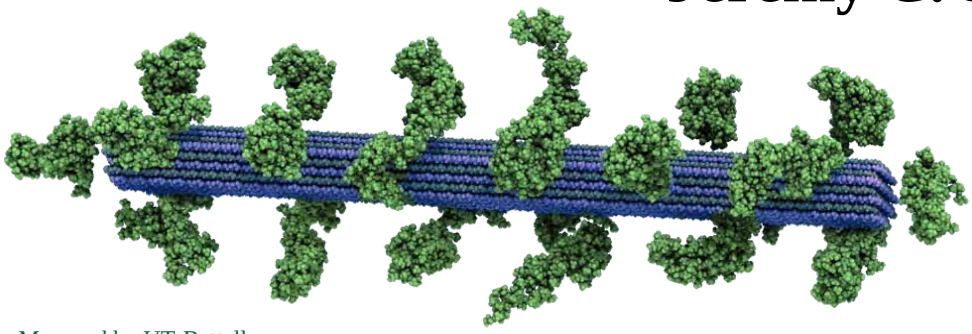


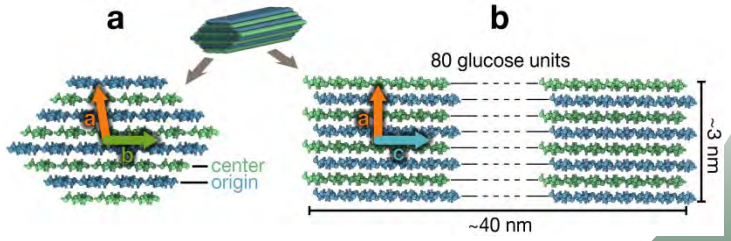
Center for Molecular Biophysics

ORNL

High-Performance Computer Simulation in the Energy Biosciences.

Jeremy C. Smith

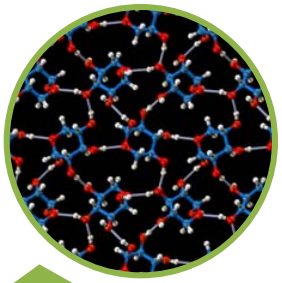




OAK RIDGE National Laboratory

The fundamental science of biomass recalcitrance is poorly understood

Recalcitrance: Resistance to breakdown into sugars



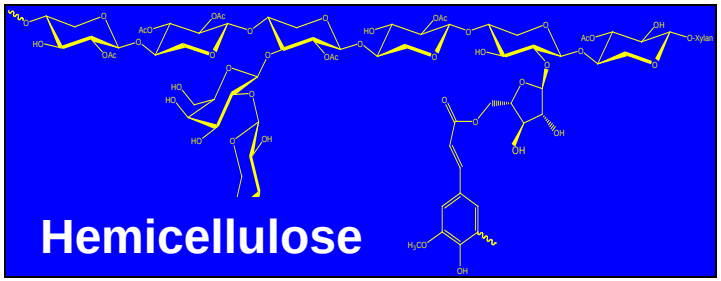
Sugars

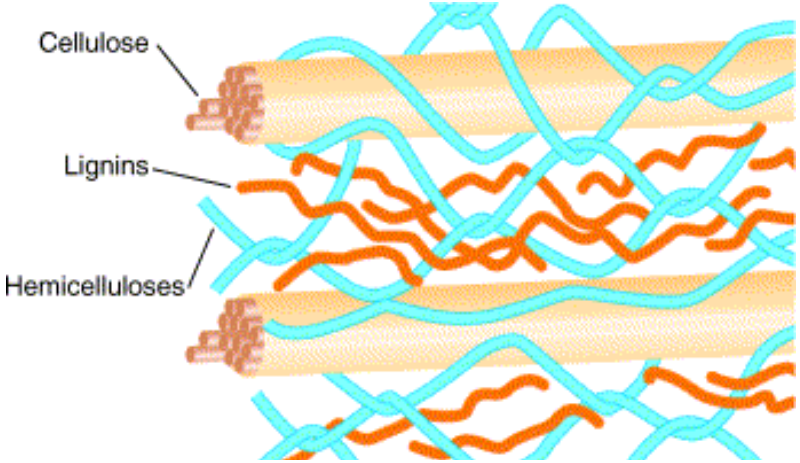
Fuel(s)

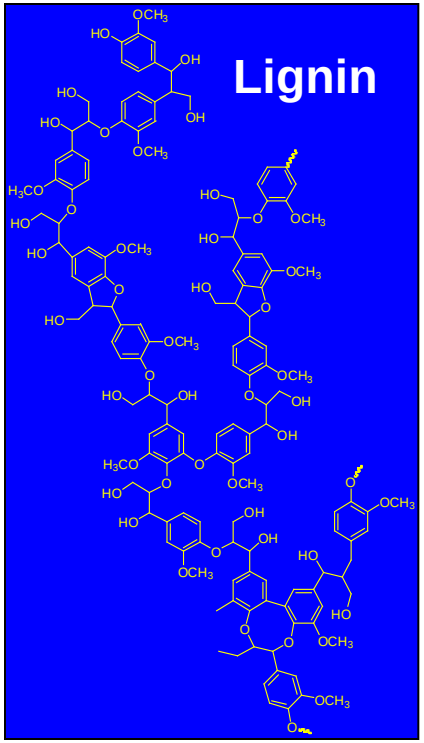
Cellulosic biomass

Overcoming recalcitrance is the single coherent overarching theme for BESC

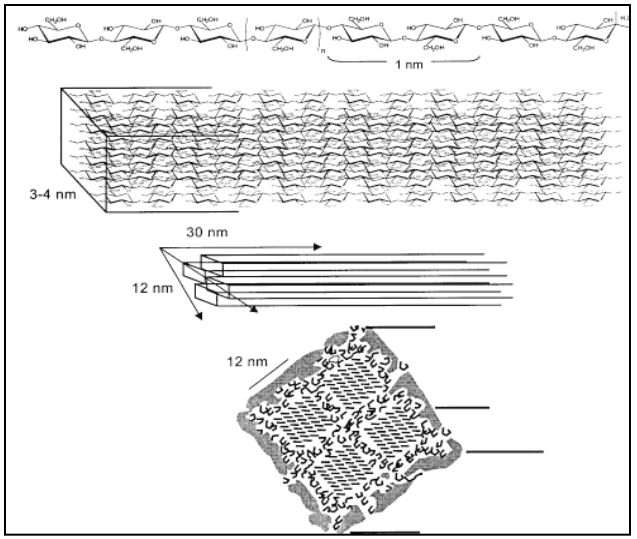
Lignocellulosic Biomass Exhibits Structural Complexity





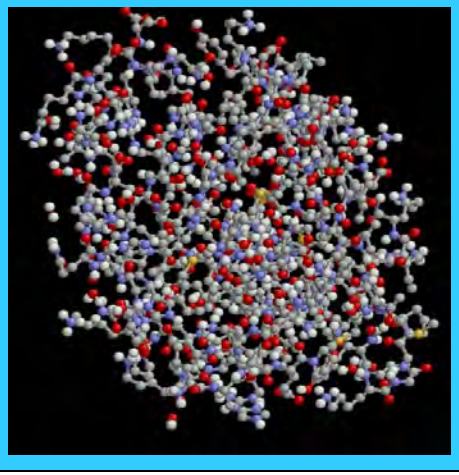


Cellulose



Atomic-Detail Computer Simulation - Basic Principles

Model System



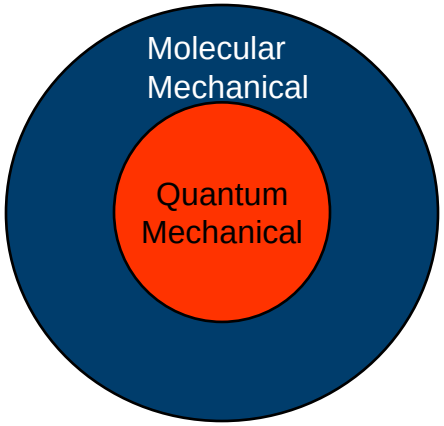
Molecular Mechanics Potential

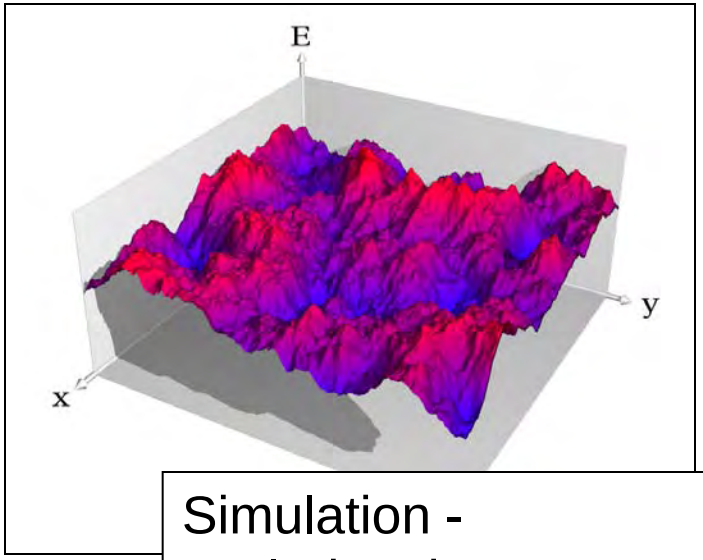
$$V = \sum_{bonds} k_b (b - b_0)^2 + \sum_{angles} k_\theta (\theta - \theta_0)^2 +$$

$$+ \sum_{dihedrals} \sum_{n=1}^{N} K_\phi^{(n)} [1 + \cos(n\phi - \delta)]$$

$$+ \sum_{i,j} 4\varepsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^{6} \right] + \sum_{i,j} \left(\frac{q_i q_j}{D r_{ij}} \right)$$

or QM/MM Potential





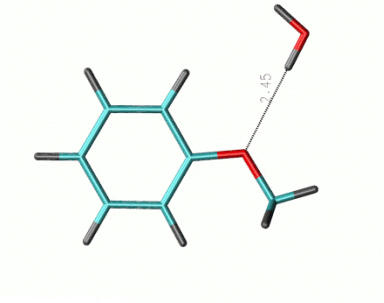
Simulation - exploring the energy landscape

Simulation Model of Lignocellulose

1 nm

length-scale

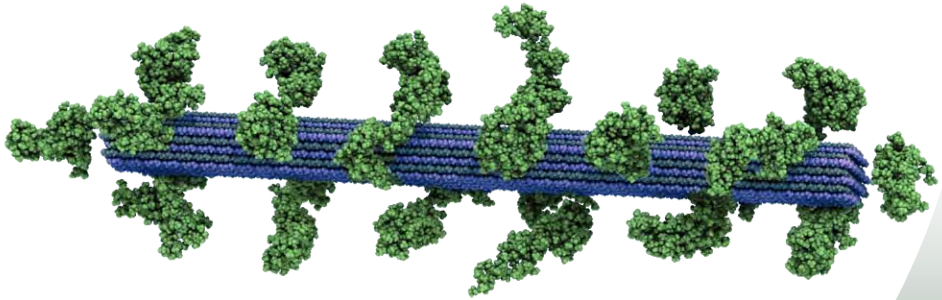
~1Å
atomic-level



~10Å
molecular-level

~100Å

supramolecular

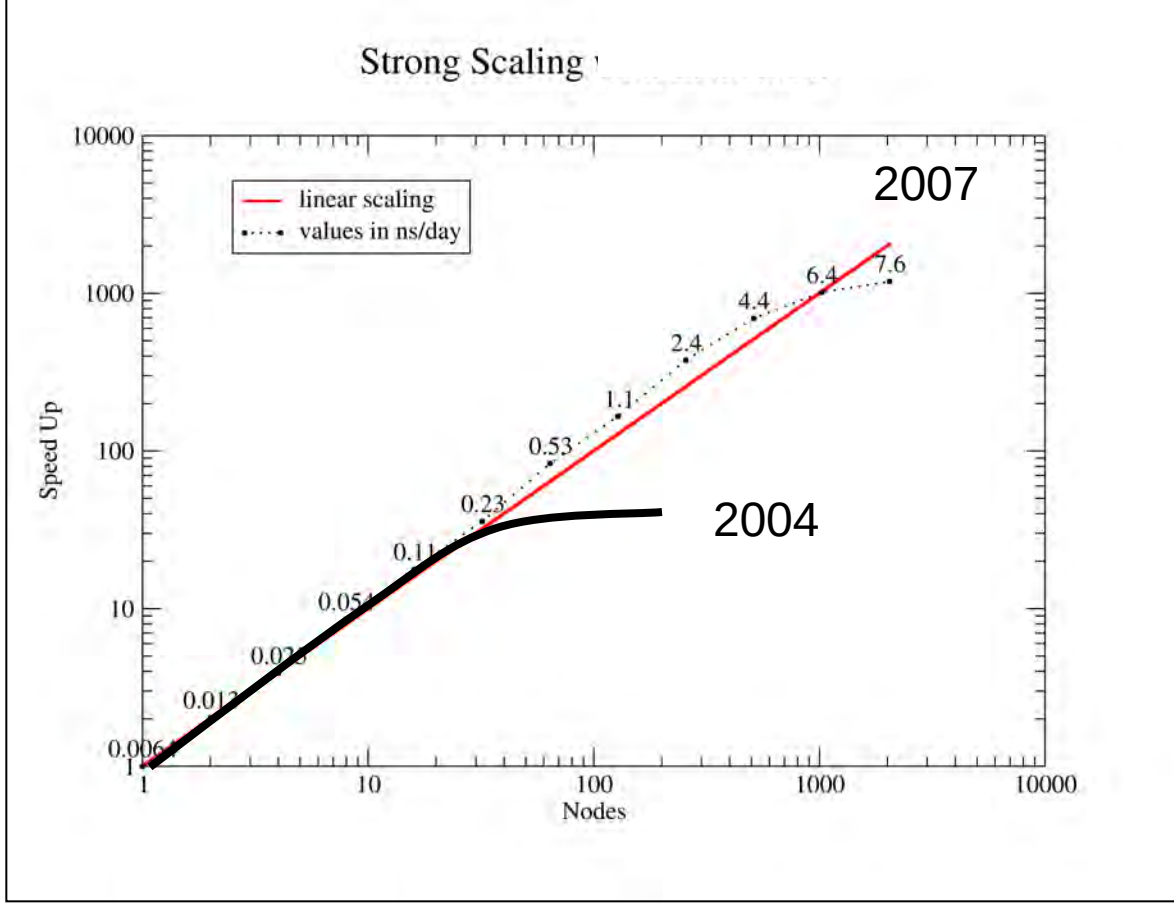


1 μm

~1000Å
coarse-grained

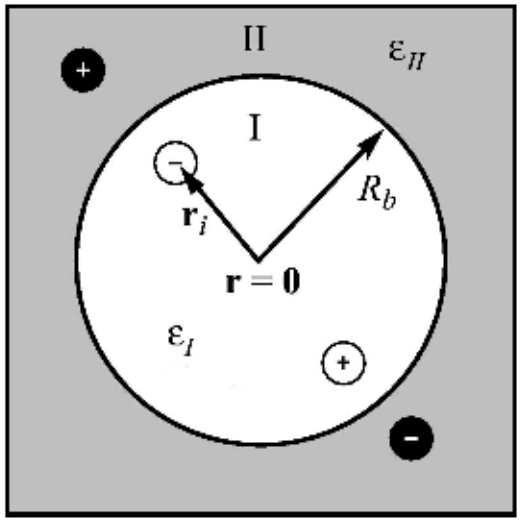
One Million Atoms – Molecular Dynamics

ROLAND SCHULZ

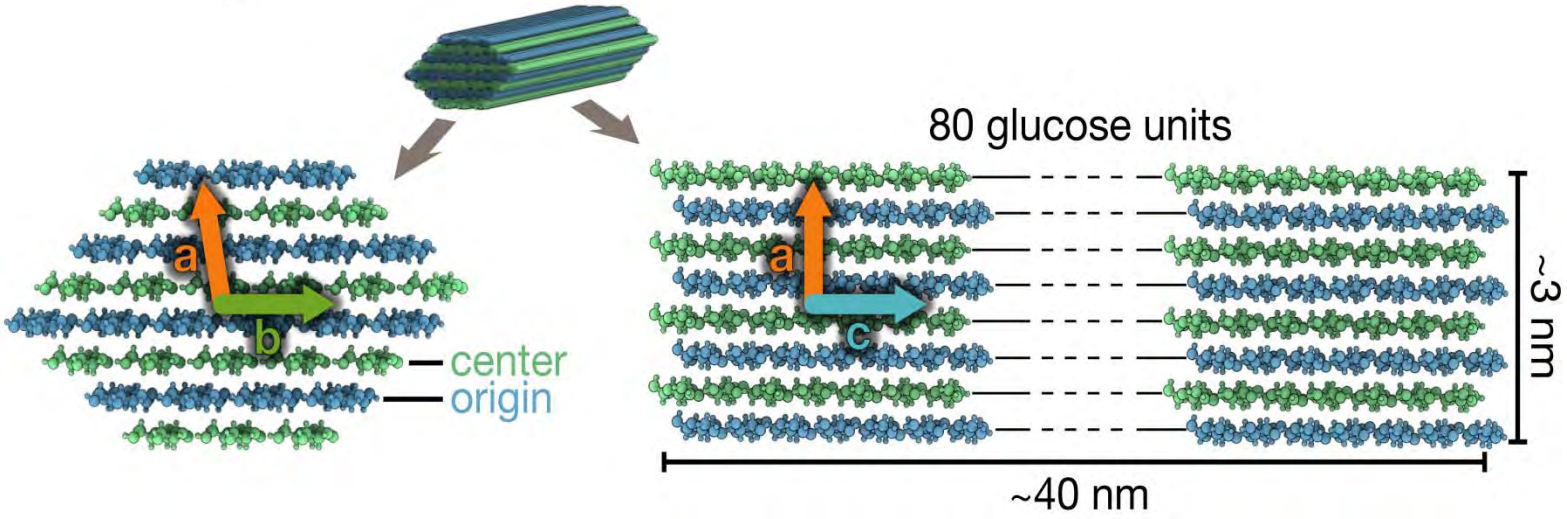


Electrostatics

1. Shift Truncation (fast, not accurate)
2. Particle Mesh Ewald (slow, accurate)
3. **Reaction Field**

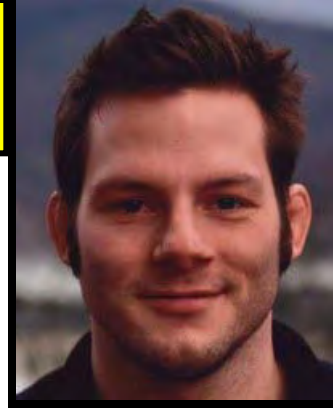


- cuts simulation time to ≈1/3 cf PME (10k cores)
- improves scaling
- **accurate?**



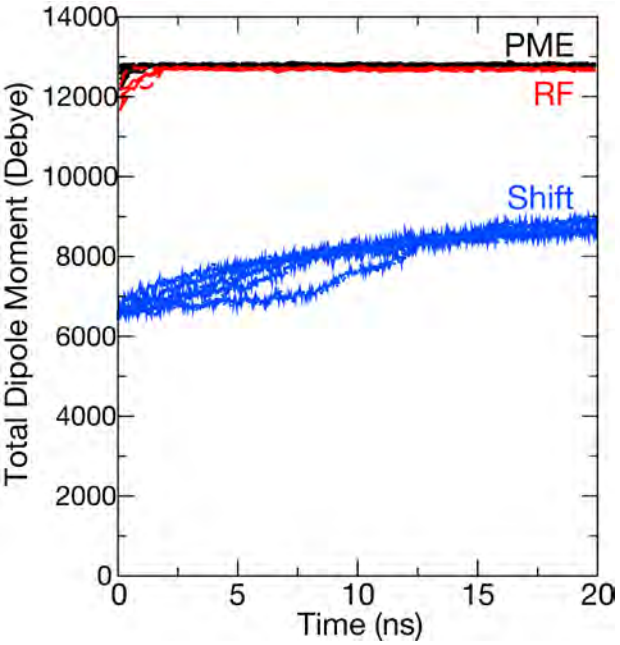
Electrostatics: Cellulose Fibril in Aqueous Solution

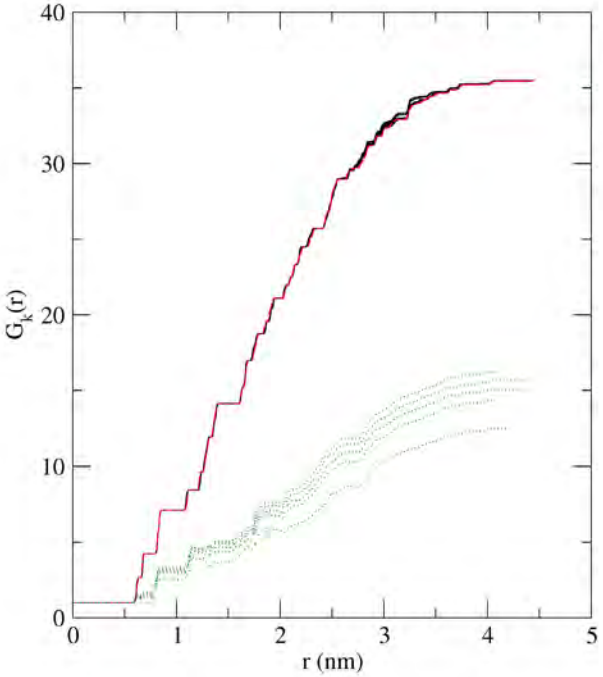
BENJAMIN LINDNER

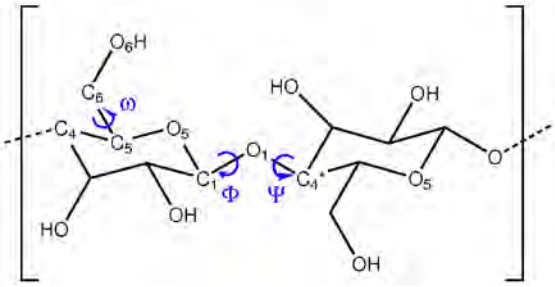


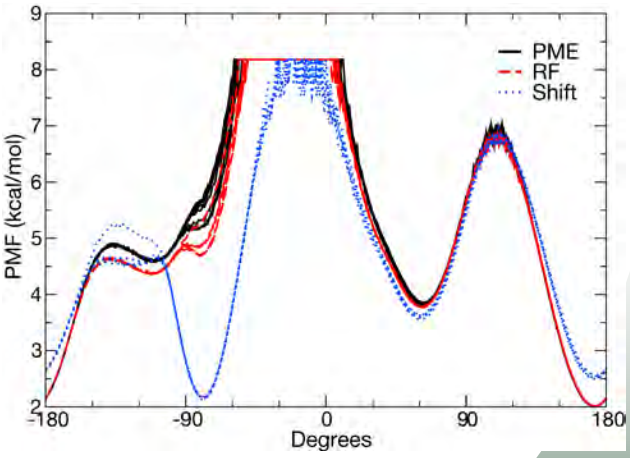
$$G_k(r) = \sum_{r_{ij}<r} \frac{\boldsymbol{\mu}_i \cdot \boldsymbol{\mu}_j}{\mu^2}$$

Kirkwood G Factor



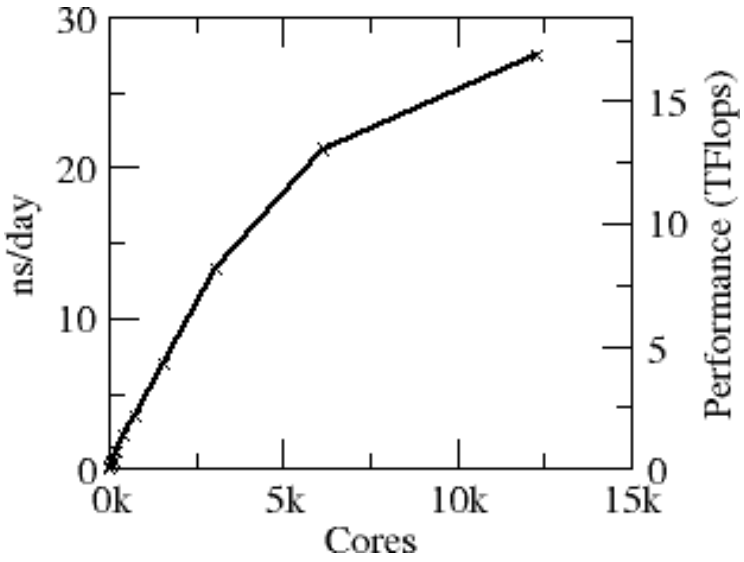




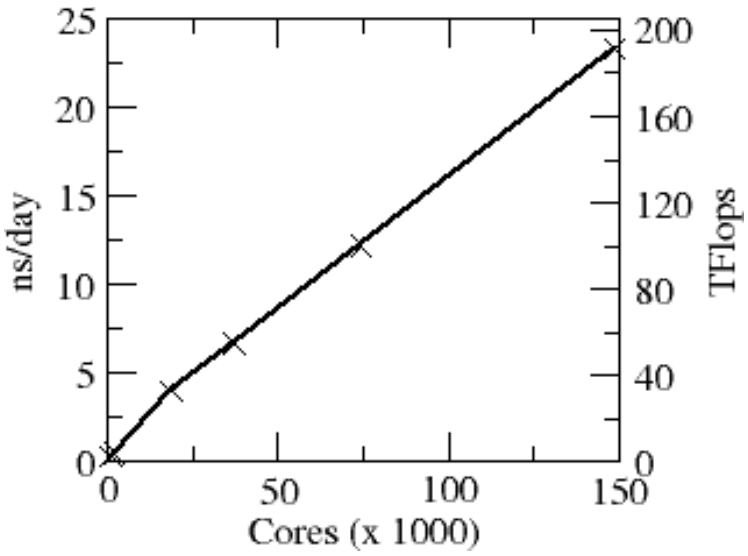


Molecular Dynamics Scaling

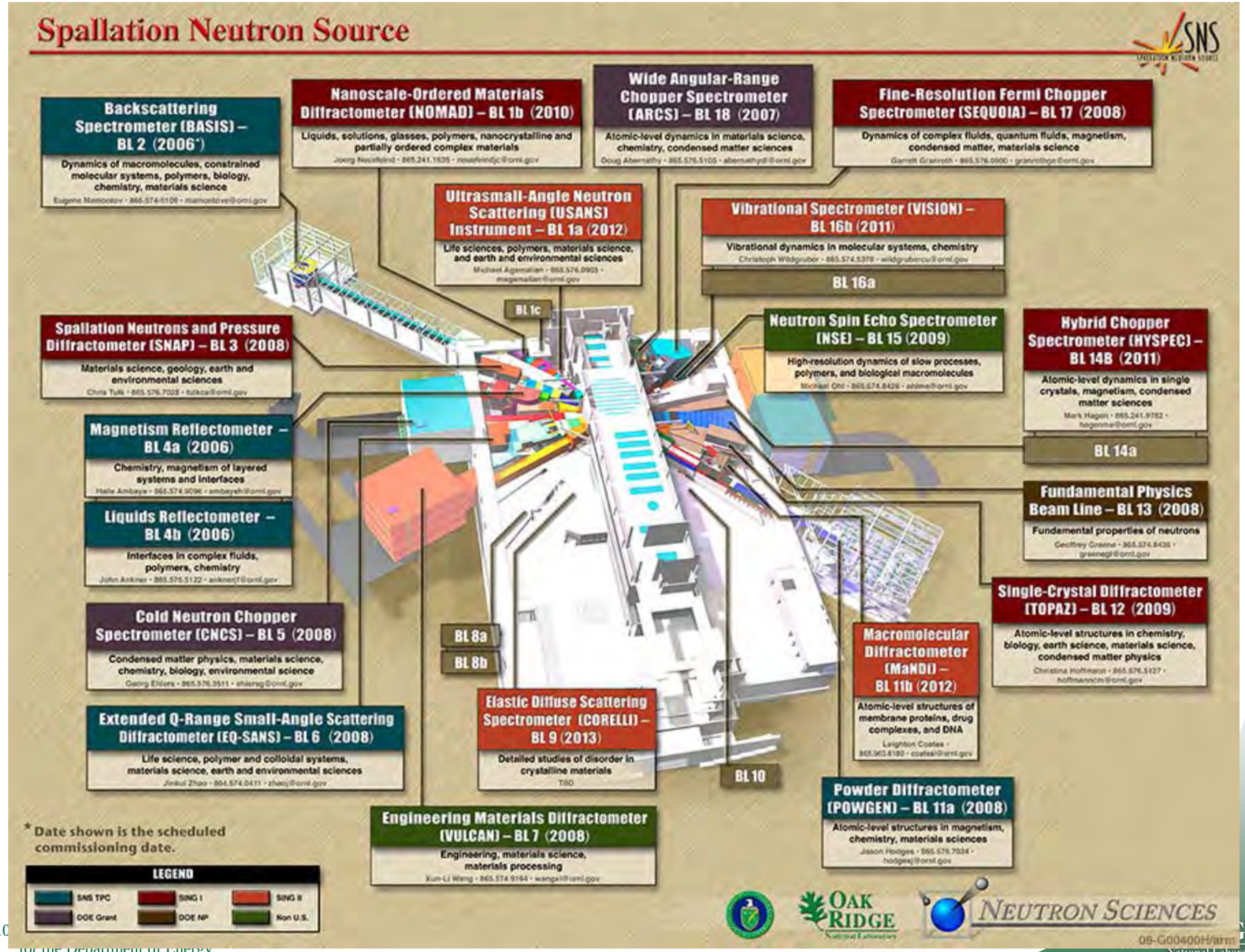
ROLAND SCHULZ



3.3M atom system



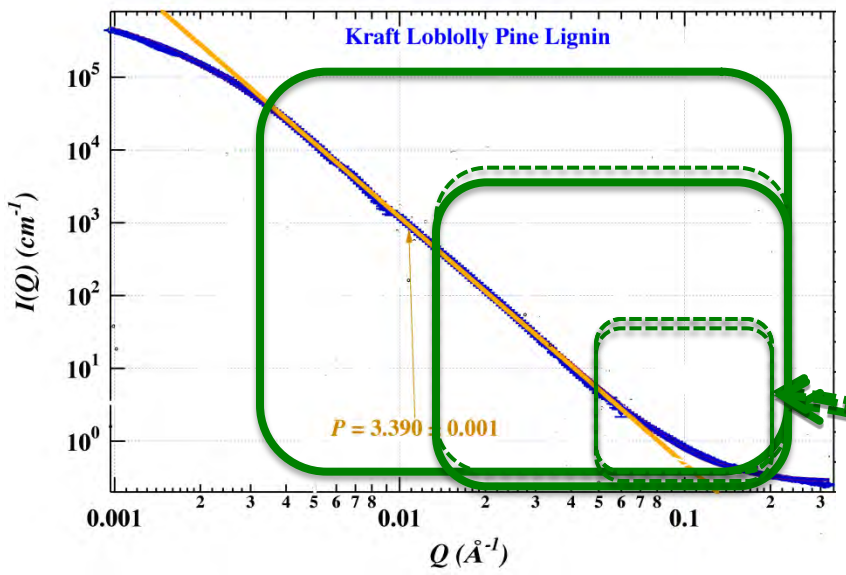
100M atom system



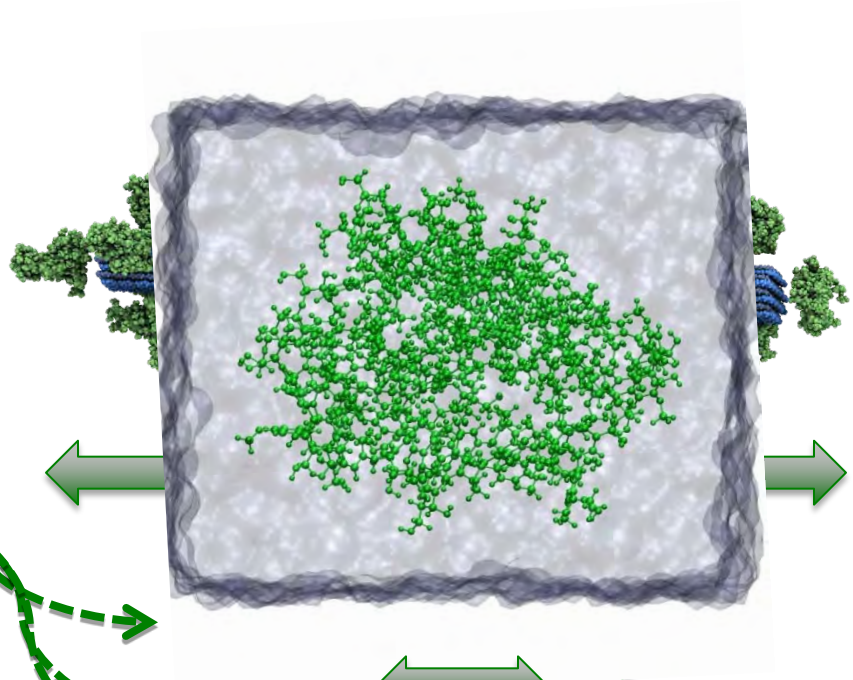
for the Department of Energy
National Laboratory

Length-Scale Correspondence: Simulation versus Experiment

Small-Angle Neutron Scattering



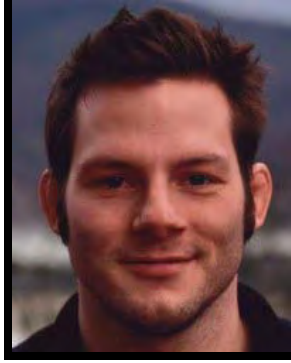
Simulation

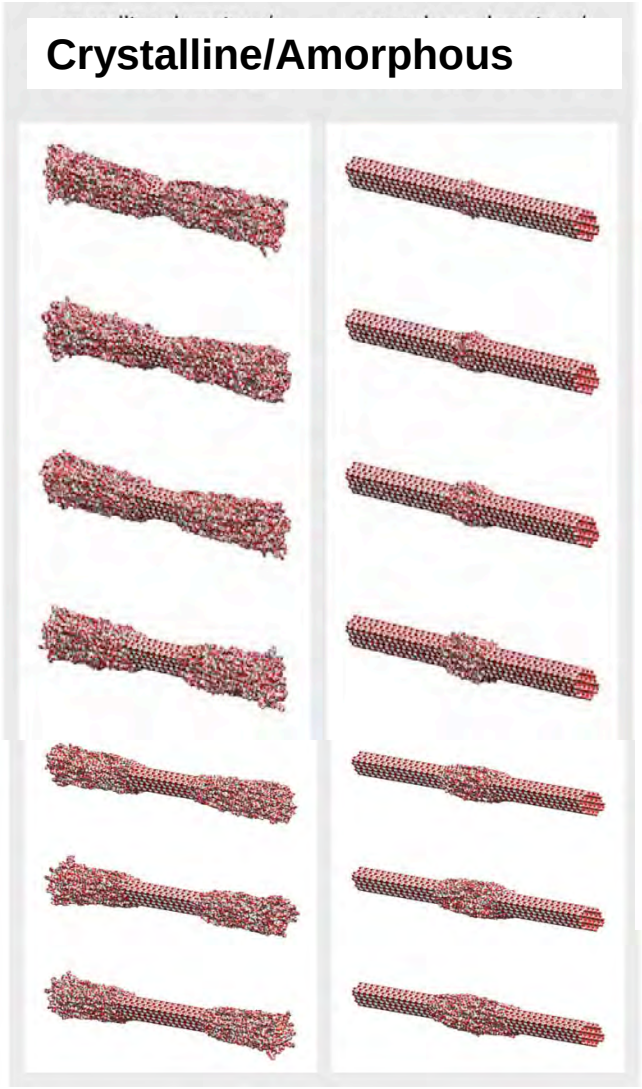


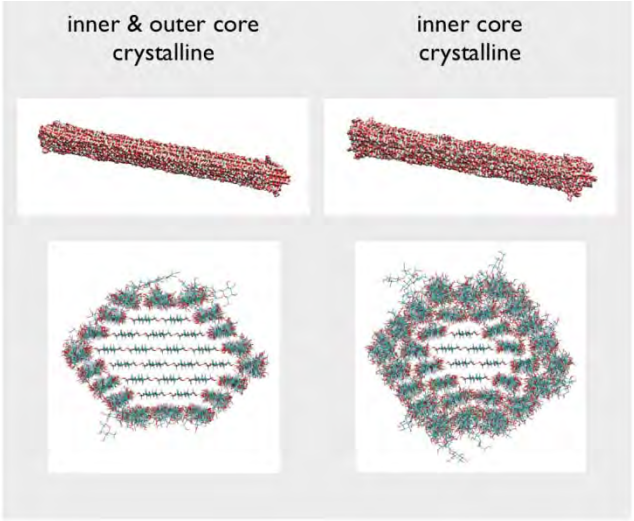
100 Å / ~500k atoms
10 Å / ~50k atoms
INCITE XT4
NSF Teragrid

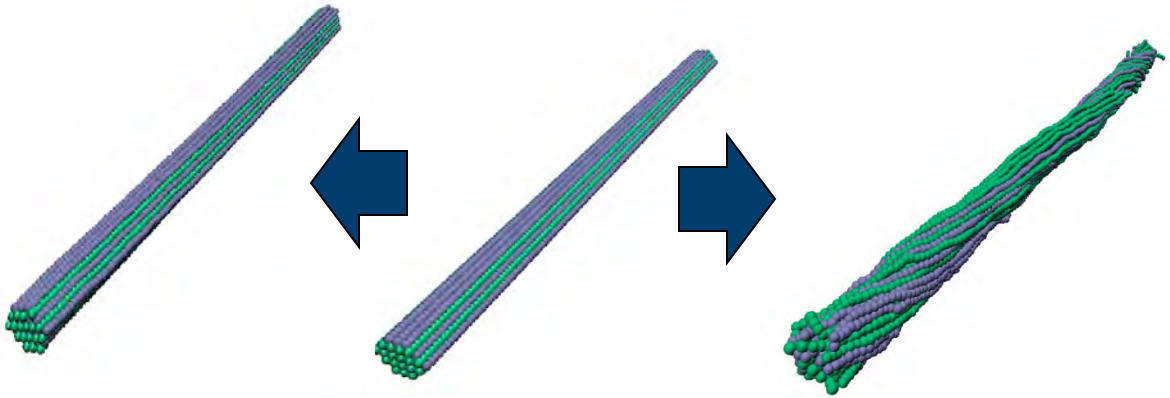
Cellulose: Models of Pretreatment

BENJAMIN LINDNER





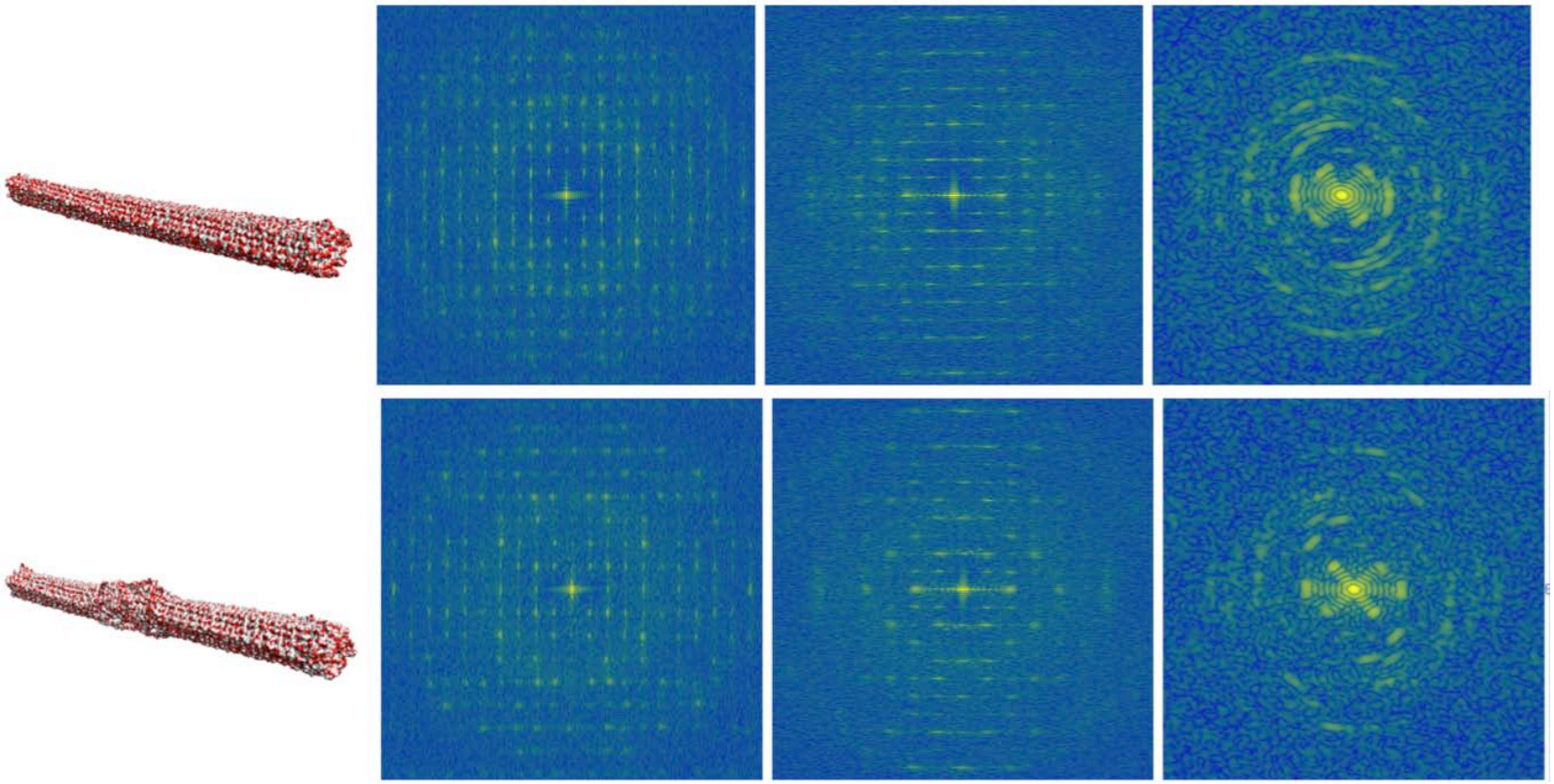




Crystalline (1ns@300K)

Amorphous (1ns@650K)

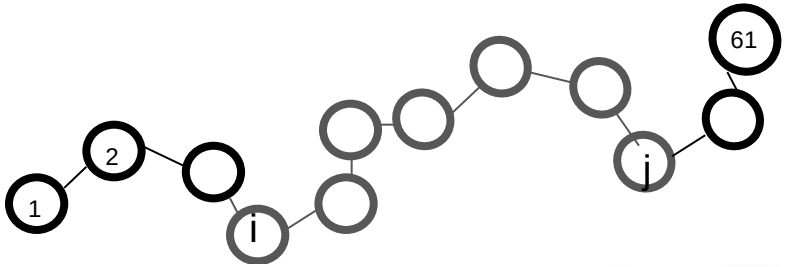
Bragg Scattering from MD Simulation

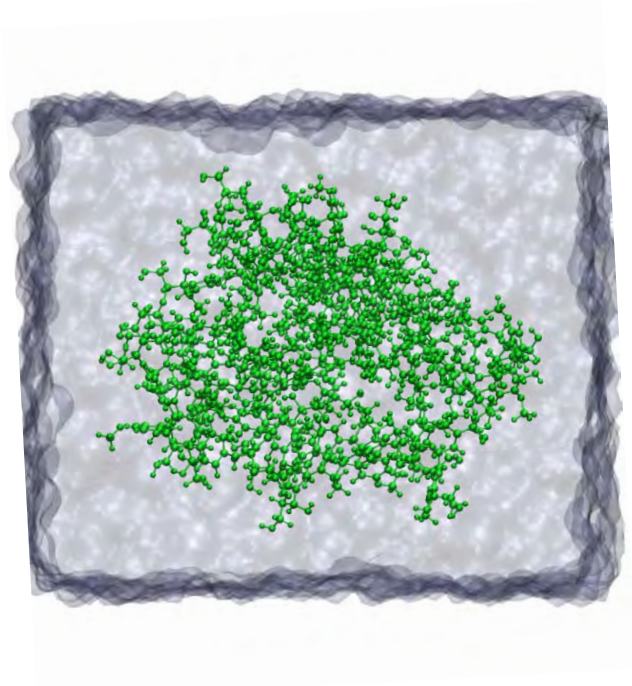


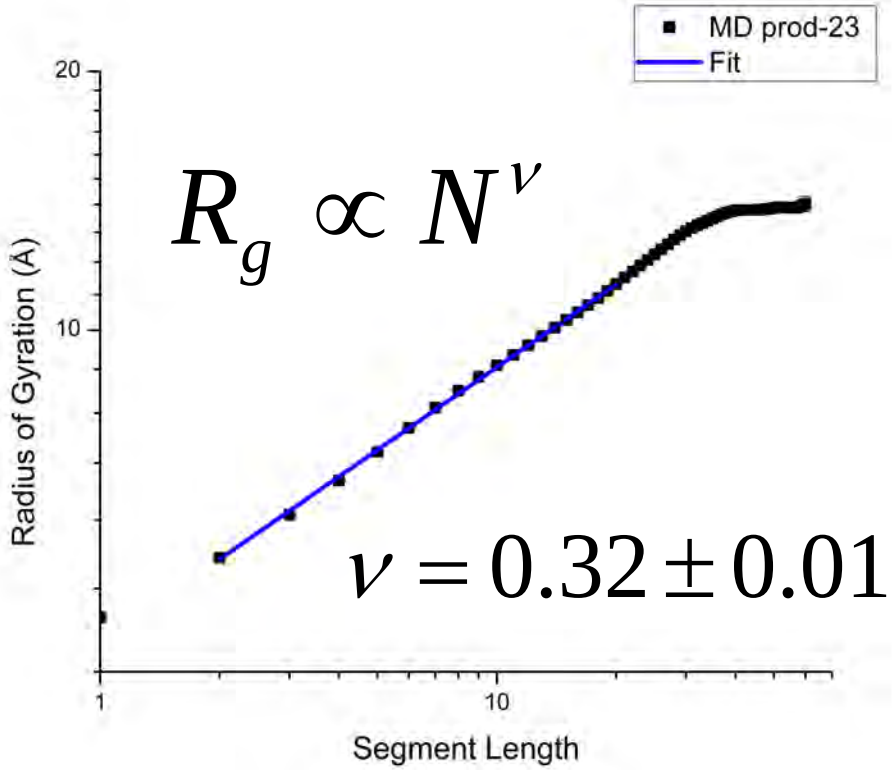
LOUKAS PETRIDIS

Single Lignin Molecule

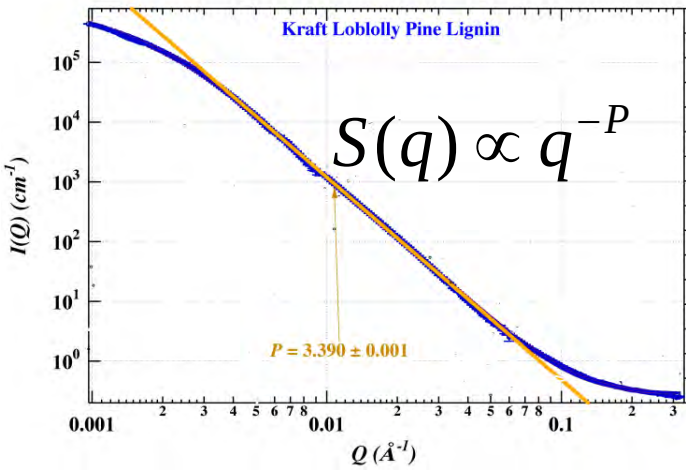
R_g=Segment Radius of Gyration



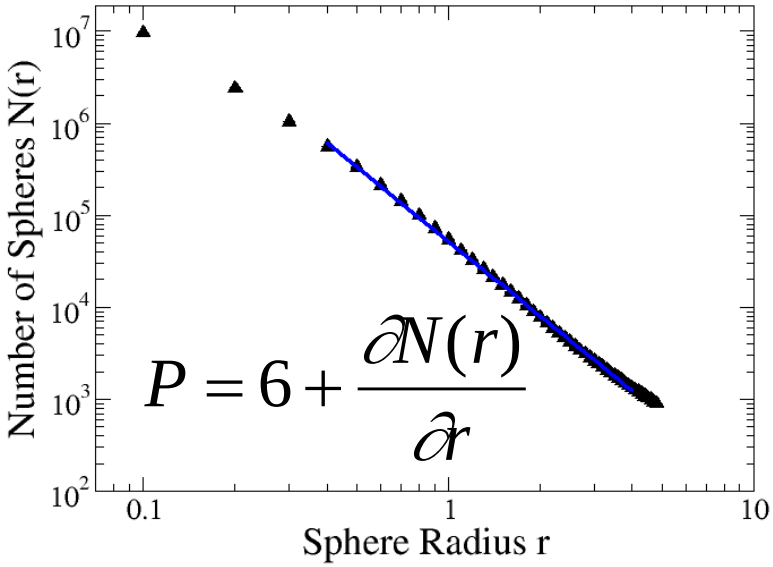




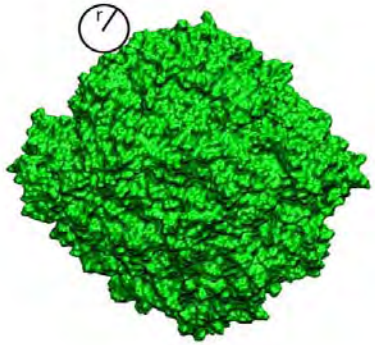
26-molecule lignin aggregate



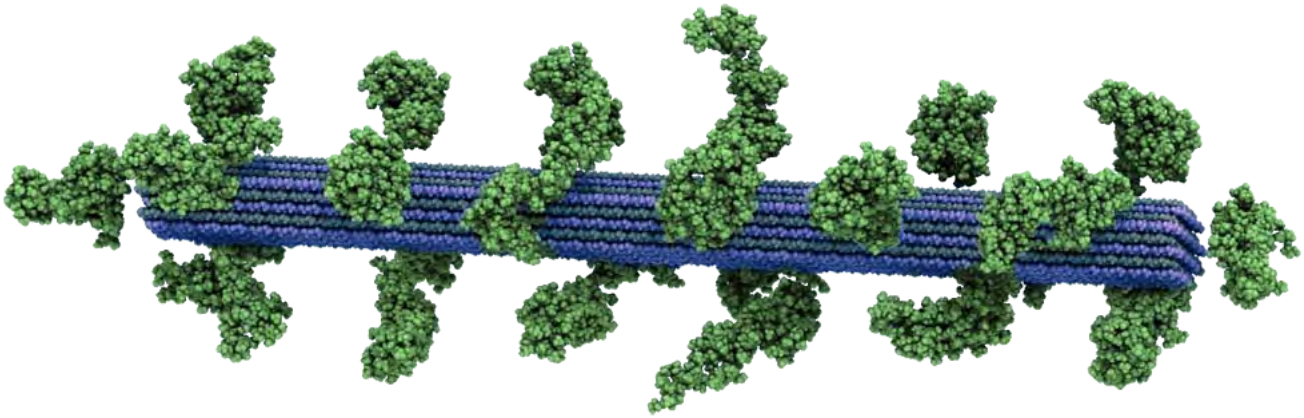
$P_{neutron}$=3.39+/-0.001



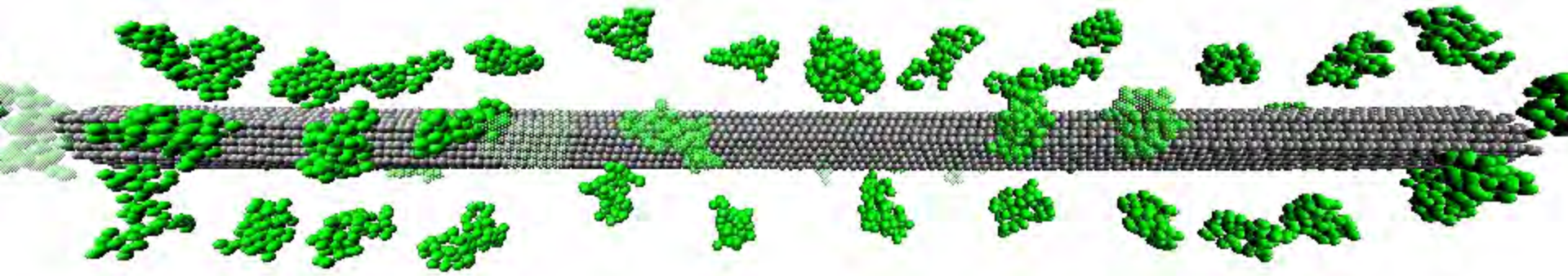
$P_{simulation}$=3.31+/-0.001



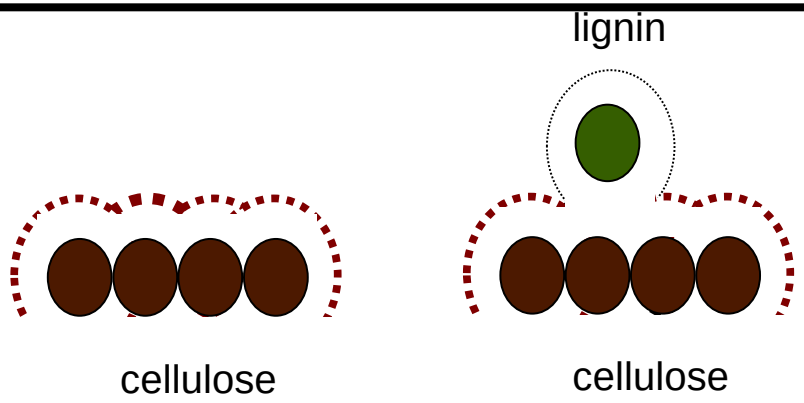
Simulation Model of Lignocellulose

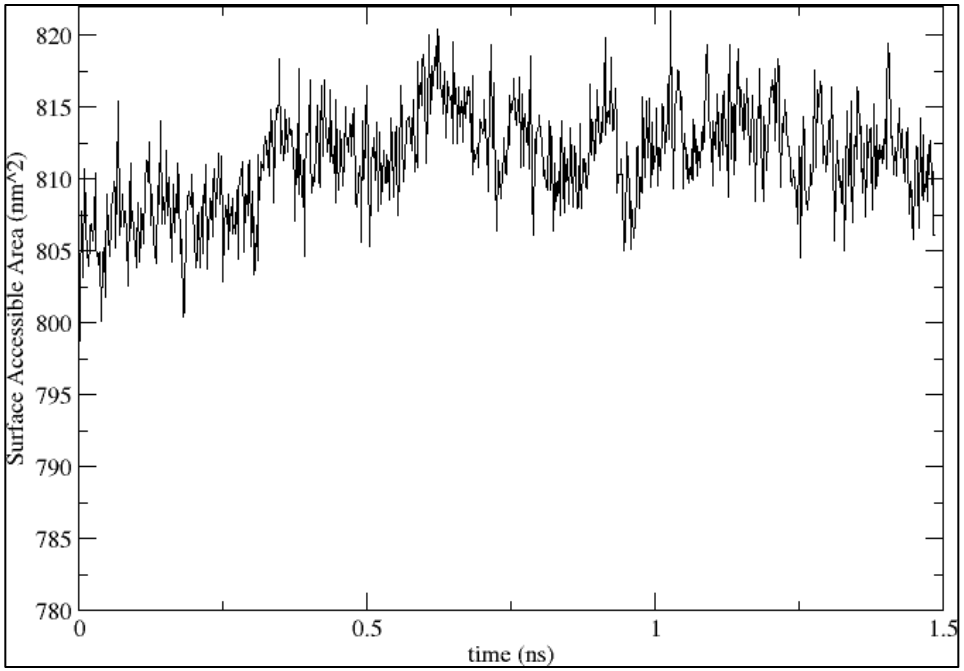


Complex of cellulose and lignins
3.3 M atoms



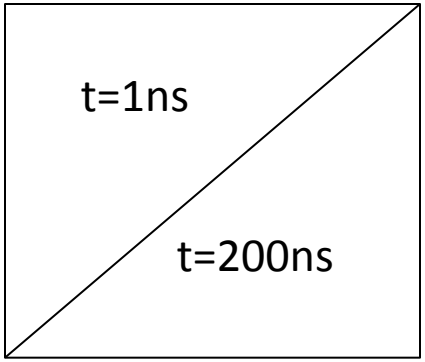
Cellulose
Accessible Surface Area

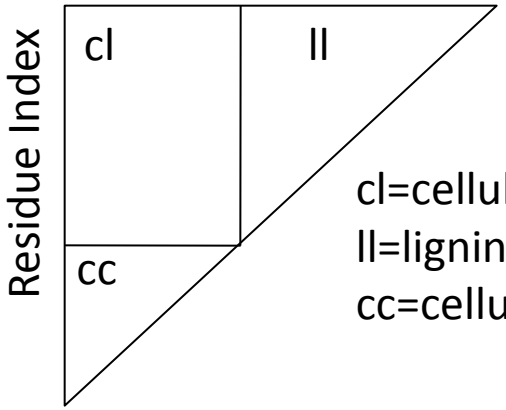




Topologic information via contact maps

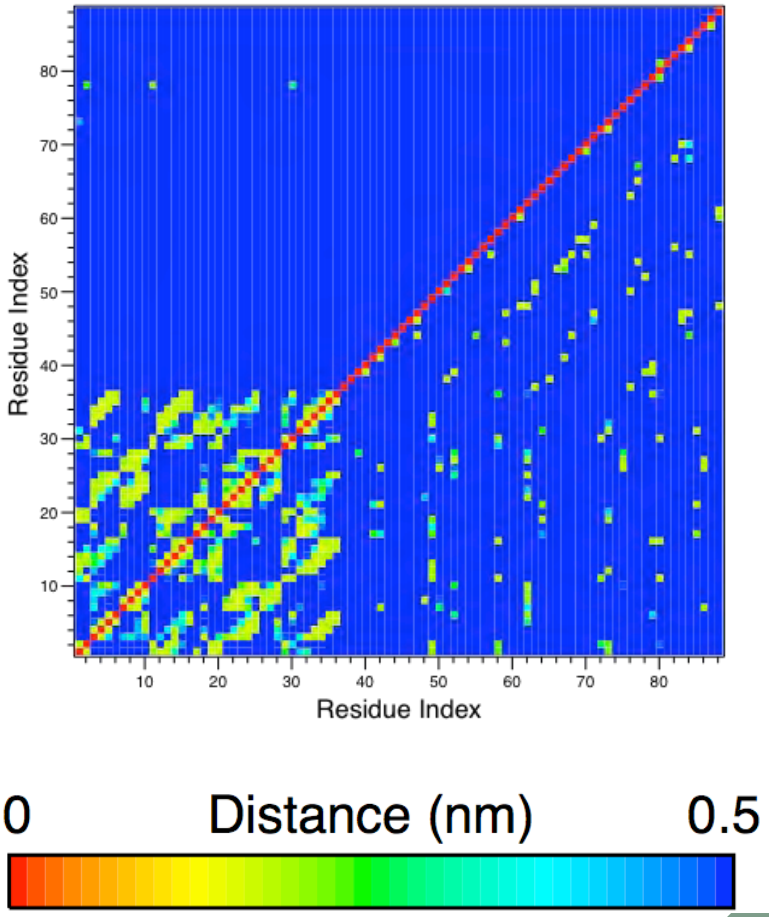
Combined Contact Map





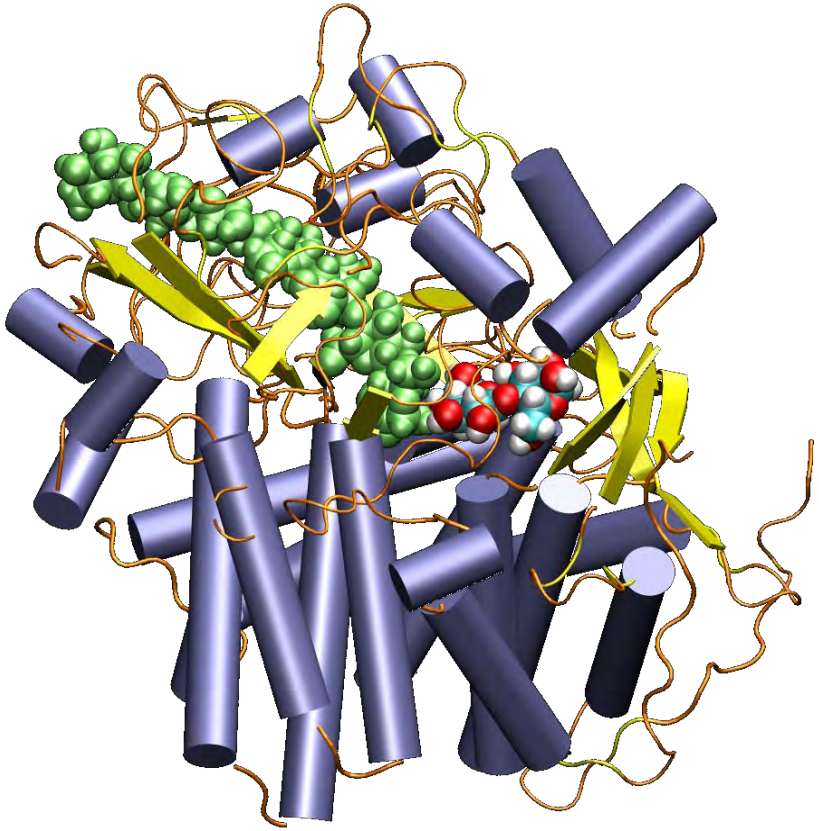
cl=cellulose-lignin
ll=lignin-lignin
cc=cellulose-cellulose

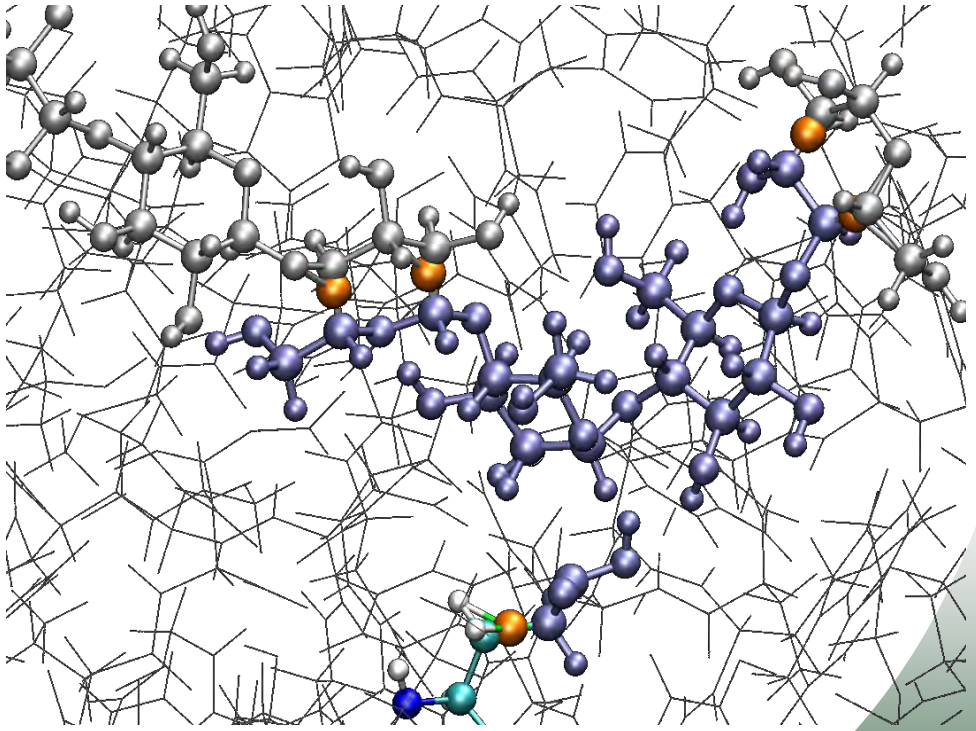
Amorph / far



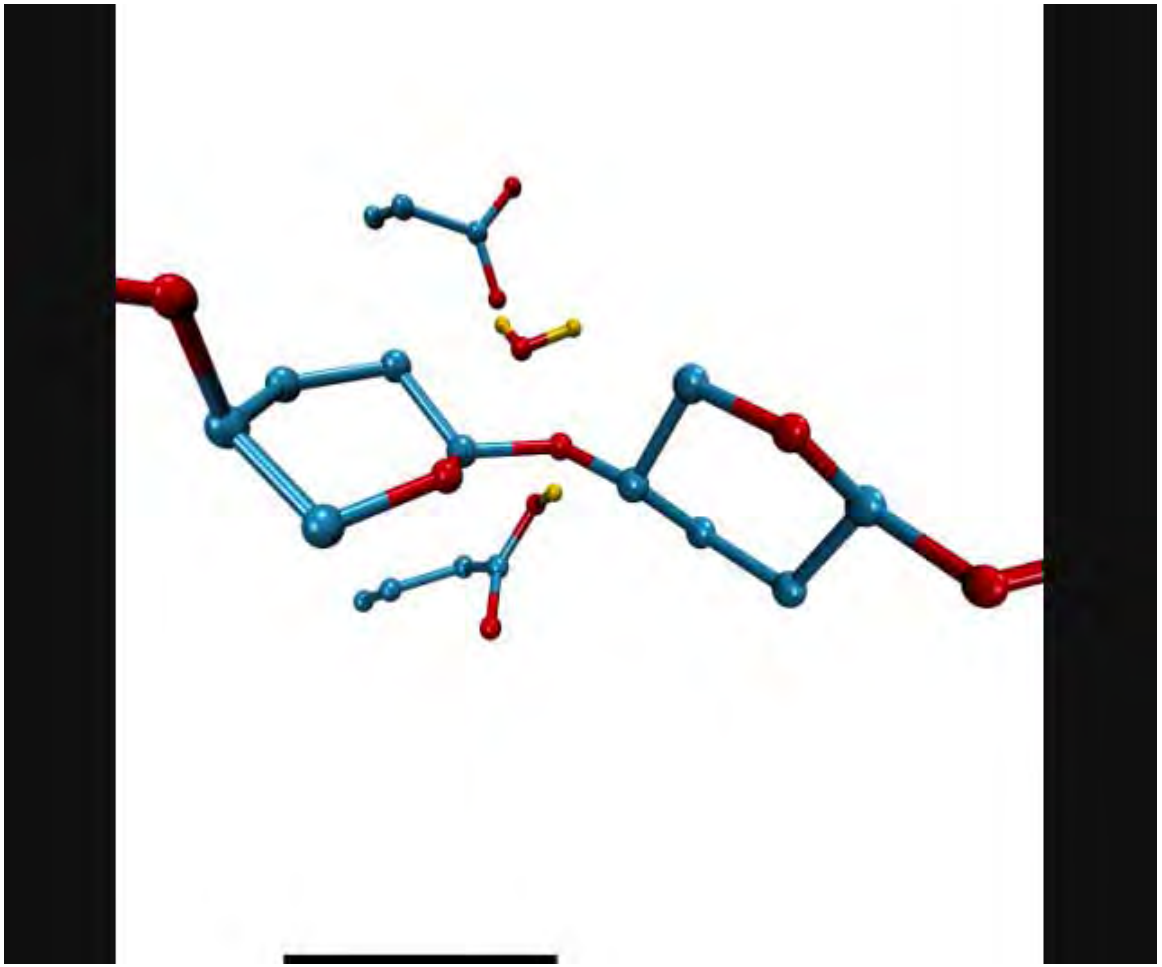
MOUMITA SAHARAY

Catalytic Mechanism of Cellulase *CelS*

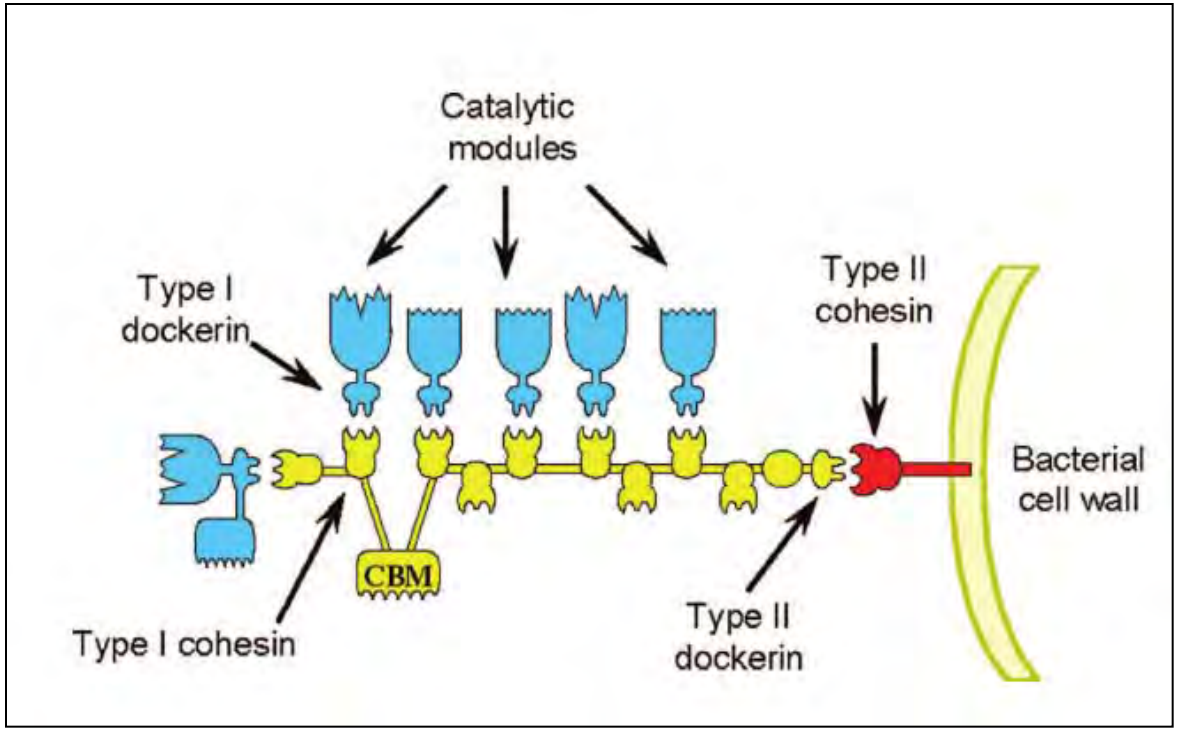




Catalytic Mechanism of Cellulase *CelS*



Cellulosome Organization

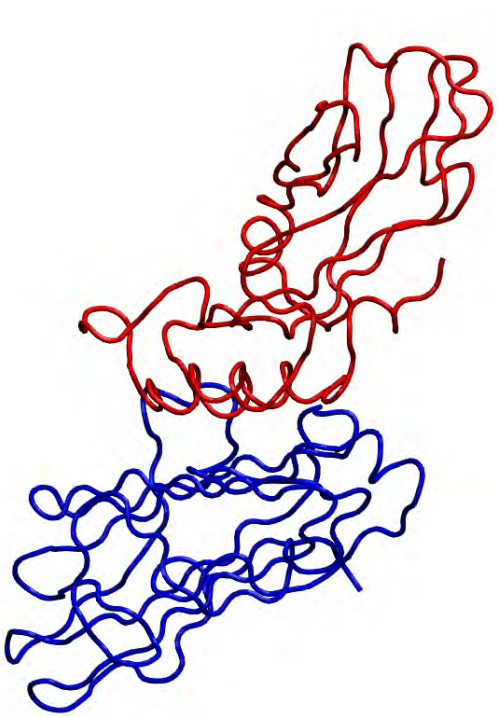


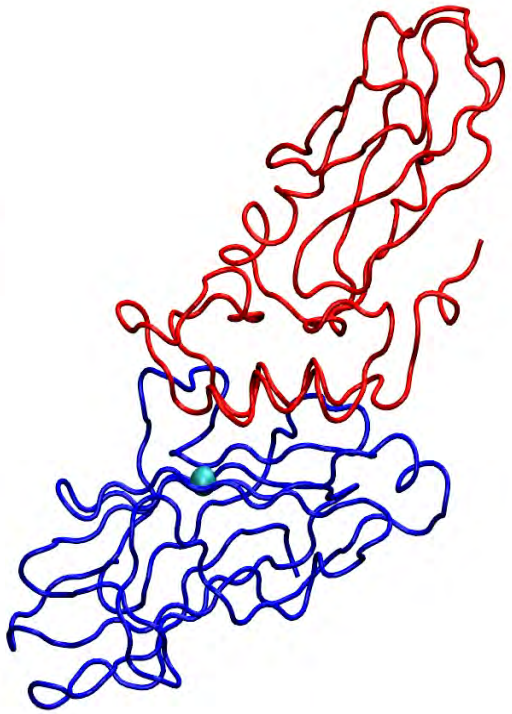
Cellulosome assembly is mediated by

cohesin-dockerin interaction.

Cohesin-Dockerin Principal Component Modes

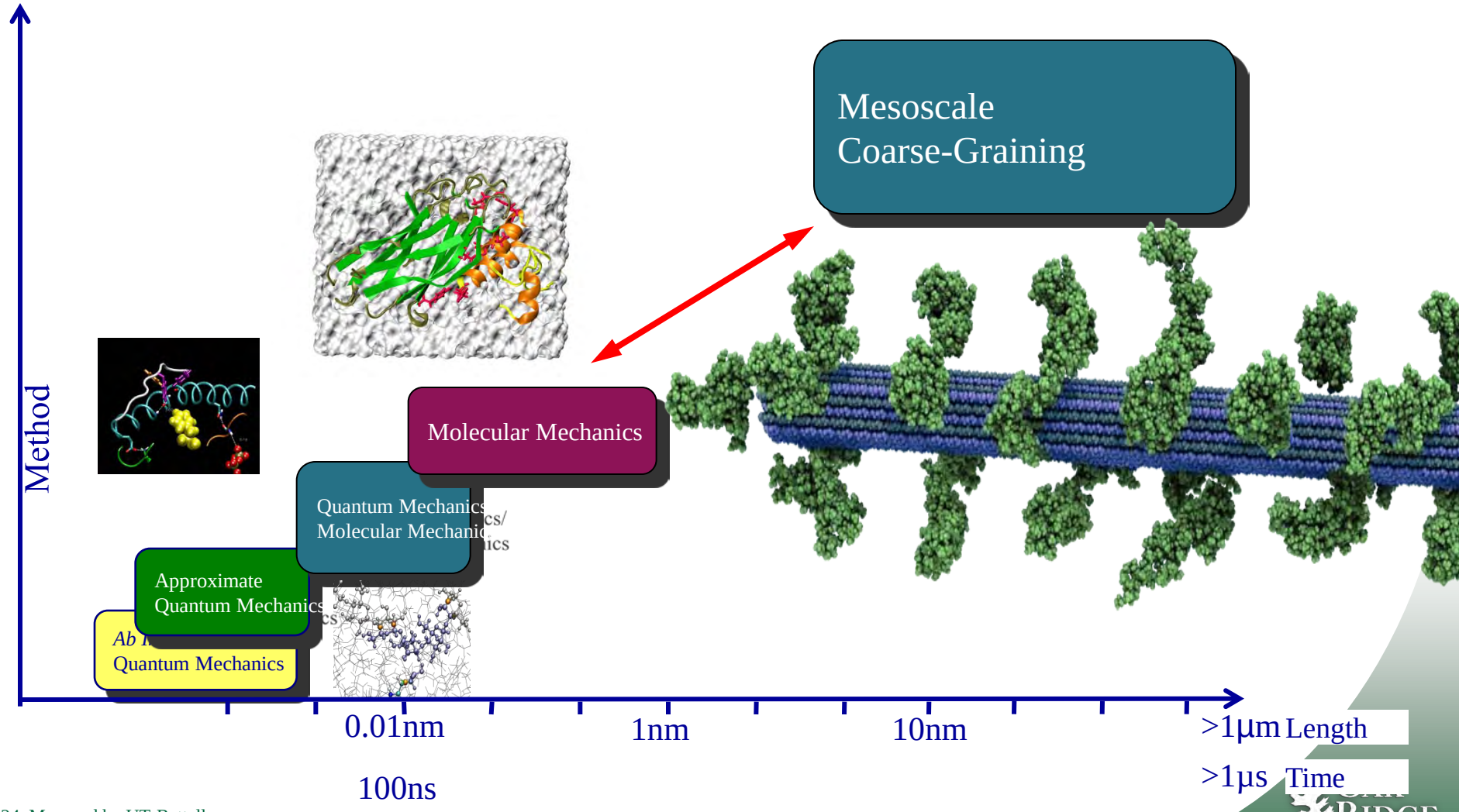
JIANCONG XU





Wild Type **F162A** **Q52A**

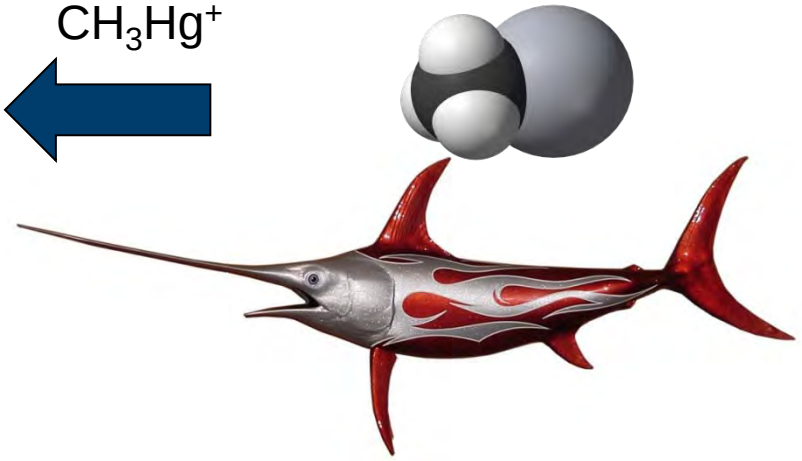
Multiscaling



Oak Ridge

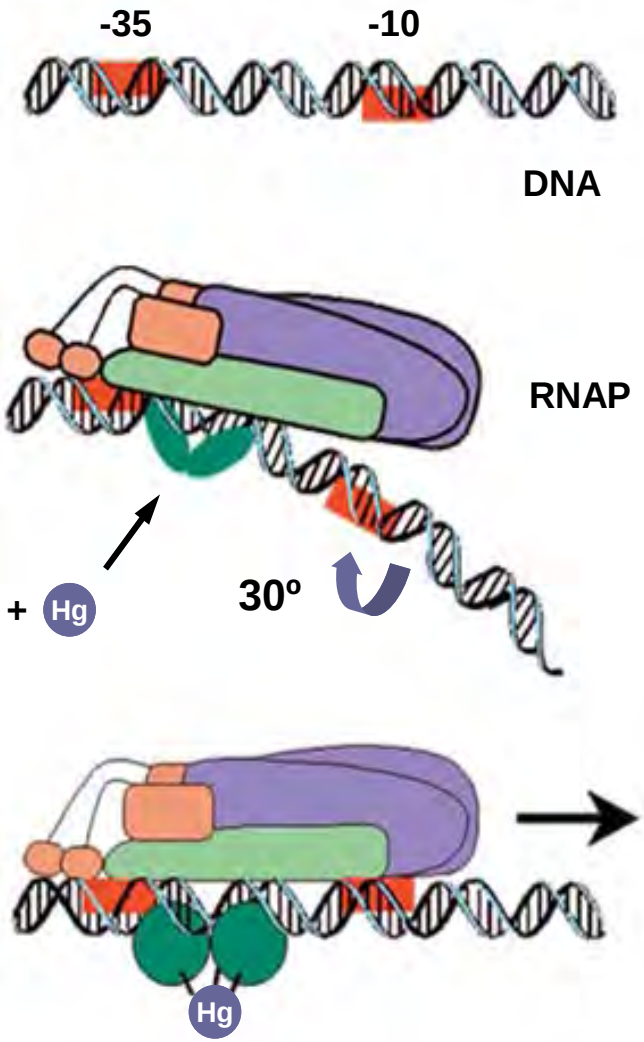
Emptying Hg flasks at the dumping shed (1955)

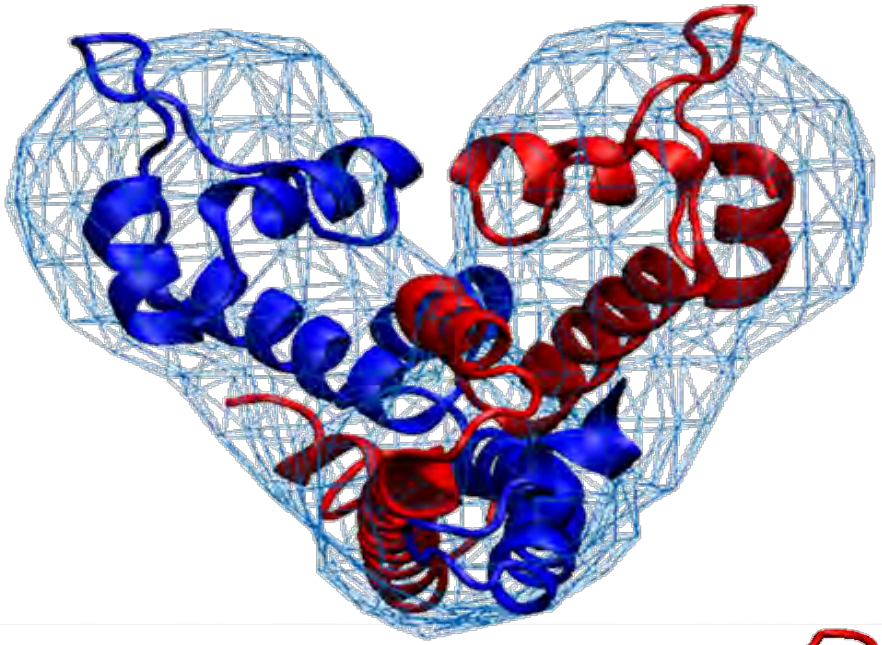
Toxic Methylmercury CH_3Hg^+

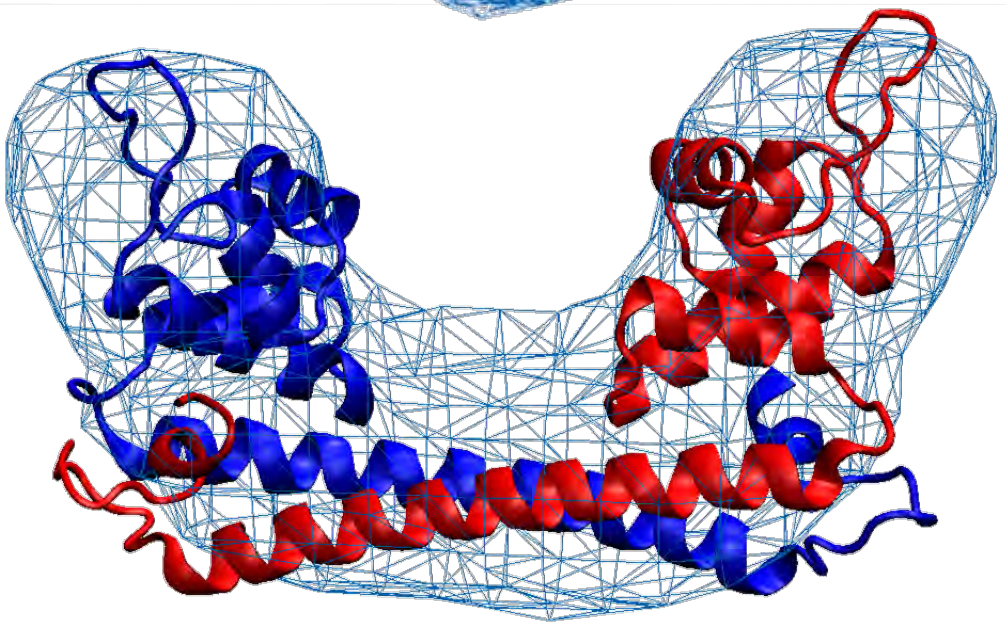


Inert Hg(0)

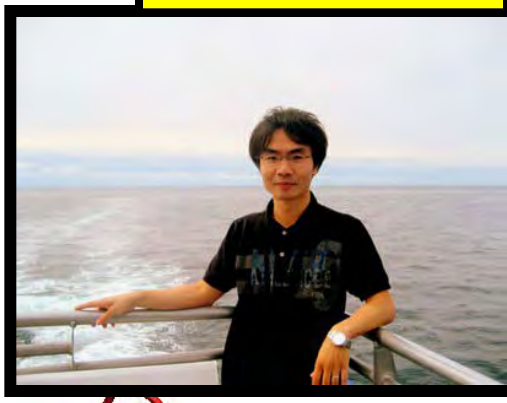
MerR



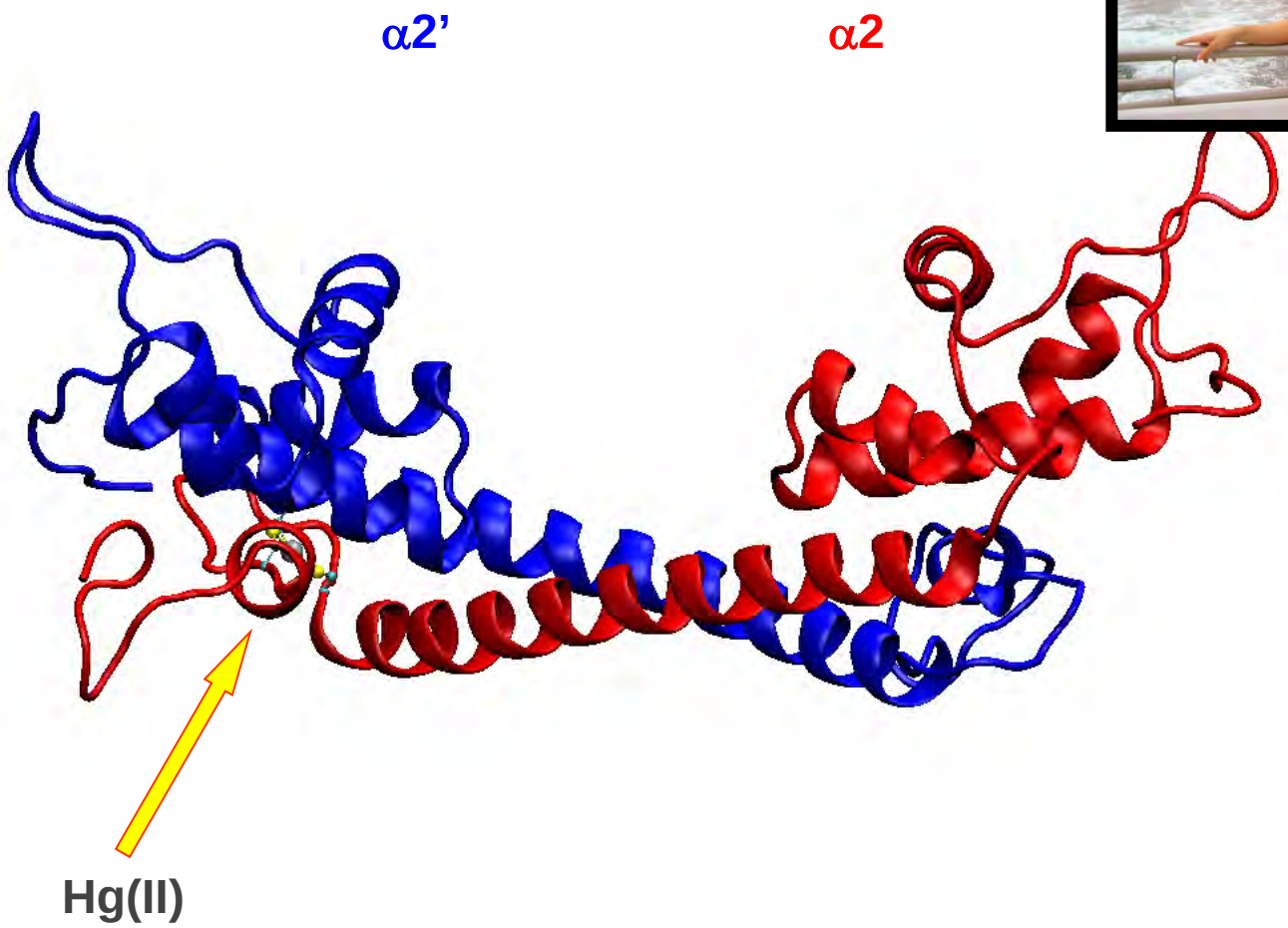




HAO-BO GUO

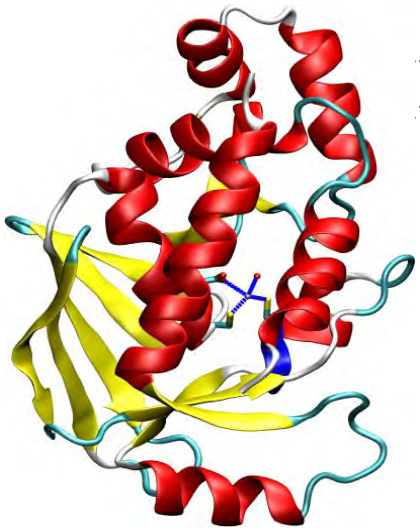


The MerR Machine



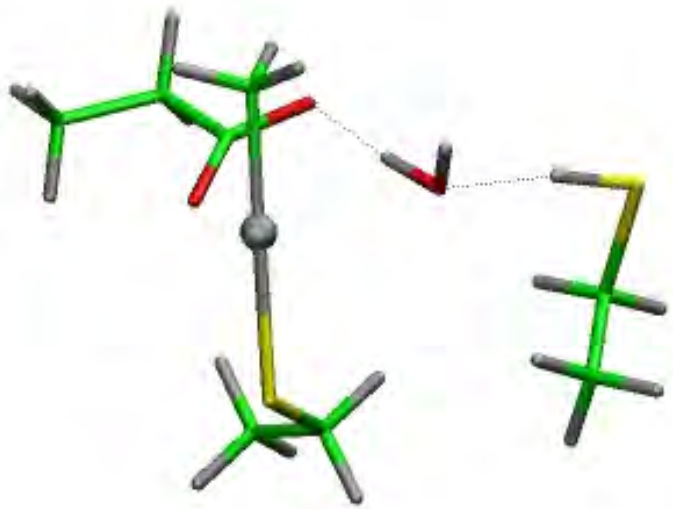
Catalytic Mechanism of MerB Hg-C Protonolysis

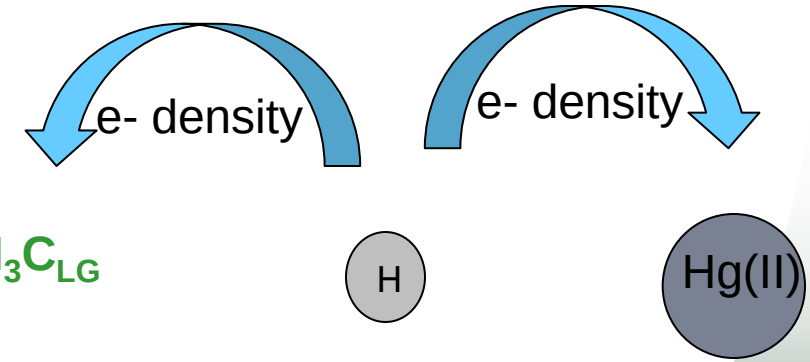
JERRY PARKS



$$R\text{-}Hg(II)]^{+} \xrightarrow{MerB} [Hg(II)]^{2+} + R\text{-}H$$

- **Thiol *bis*-coordination of Hg at Transition State Polarizes Attacking Proton.**





Funding

US Department of Energy

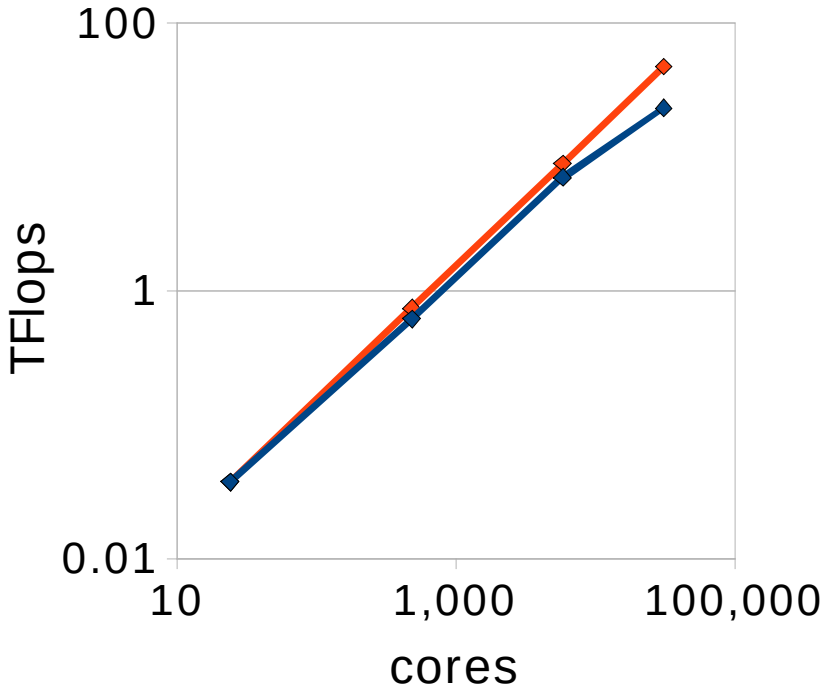
- **OBER Bioenergy Science Center.**
- **OBER ERSP SFA Biogeochemical and Molecular Mechanisms Controlling Contaminant Transformation in the Environment.**
- **OBER FWP Integration of Neutron Scattering and Computer Simulation in the Imaging of Lignocellulosic Biomass**

Molecular Dynamics Scaling

ROLAND SCHULZ

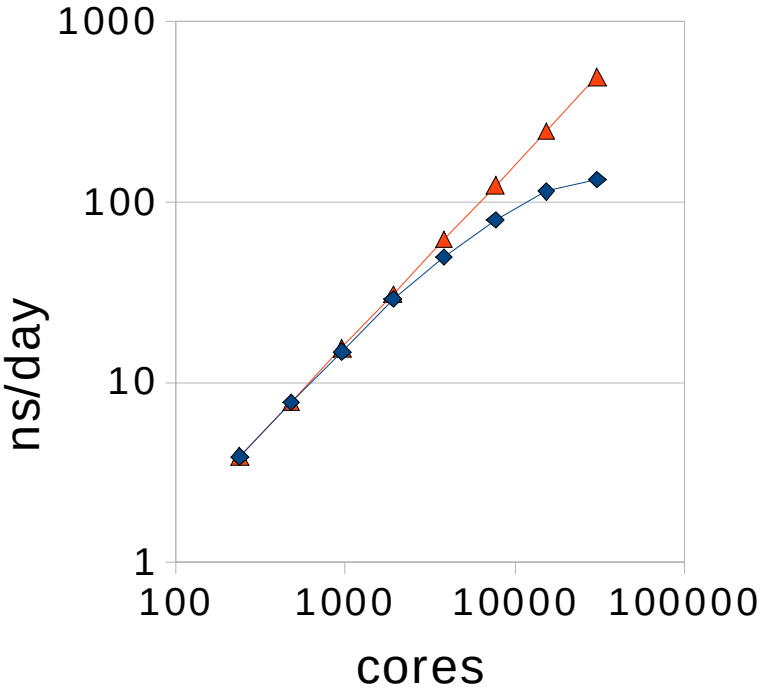
Weak Scaling

175 atoms/core

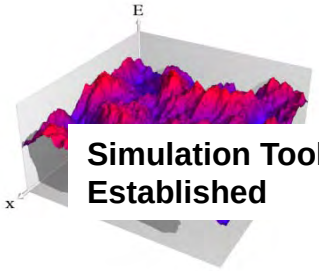


Strong Scaling

5.4 million atoms

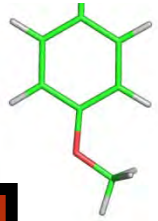


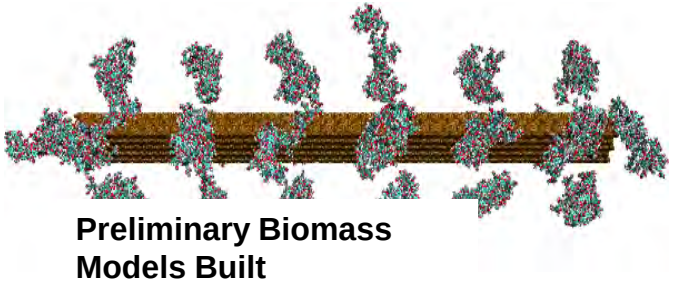
Future



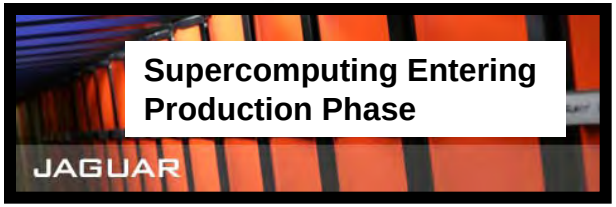
Simulation Toolbox Established

Force Fields Established

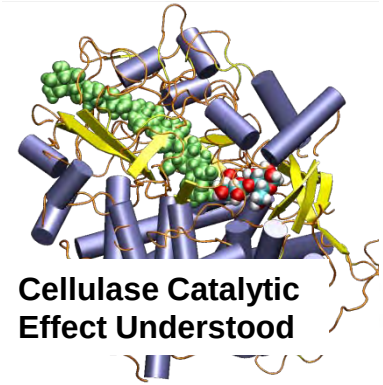




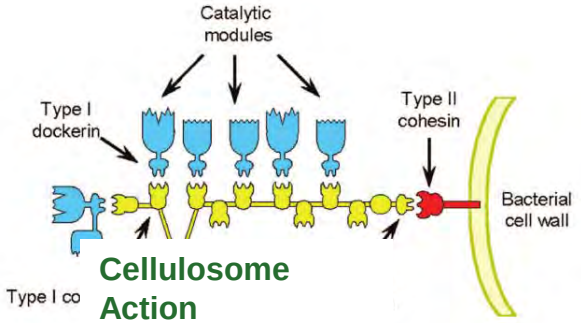
Preliminary Biomass Models Built



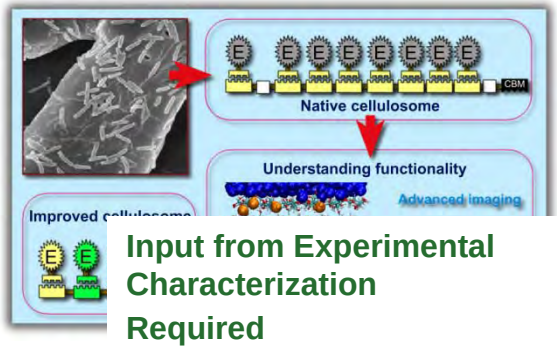
Supercomputing Entering Production Phase



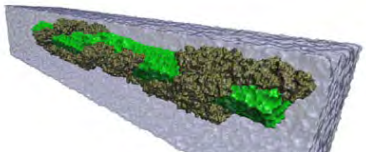
Cellulase Catalytic Effect Understood



Cellulosome Action To be Understood



Input from Experimental Characterization Required



Pretreatment Effects To be Examined