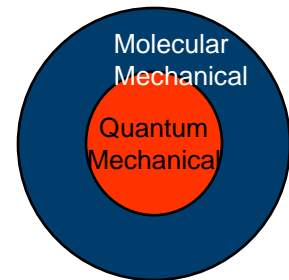
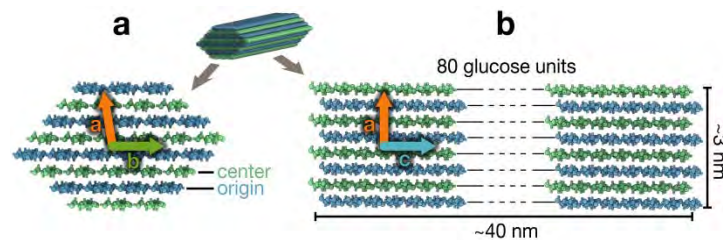
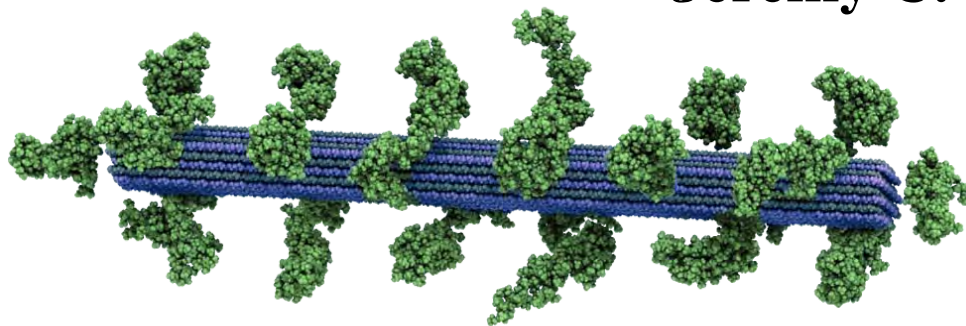


**Center for
Molecular
Biophysics**

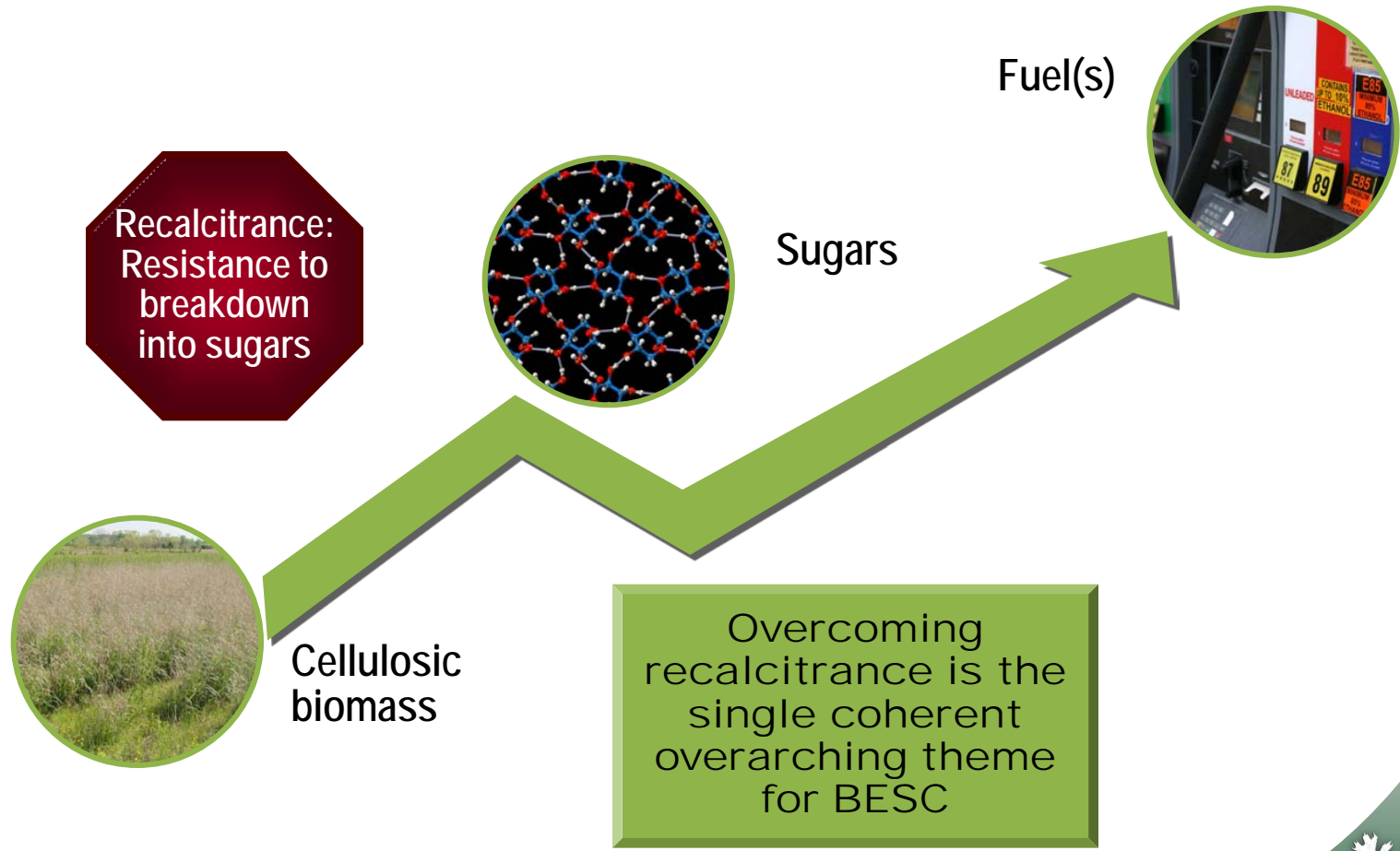


High-Performance Computer Simulation in the Energy Biosciences.

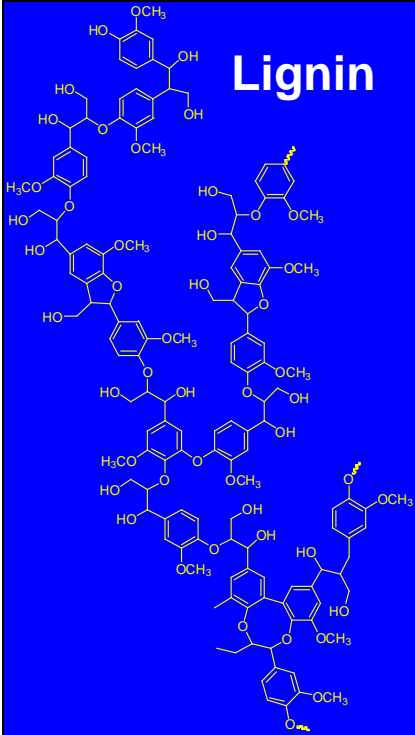
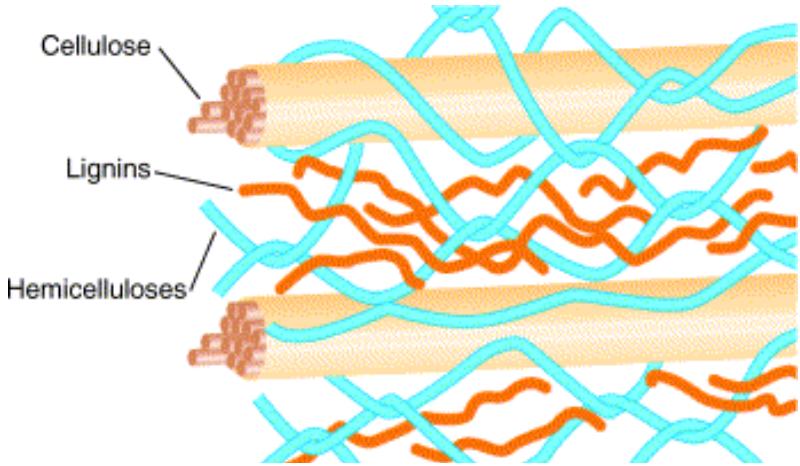
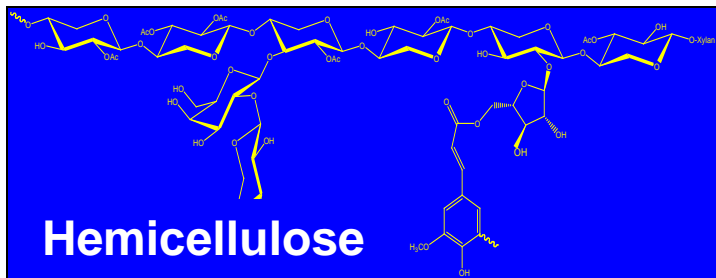
Jeremy C. Smith



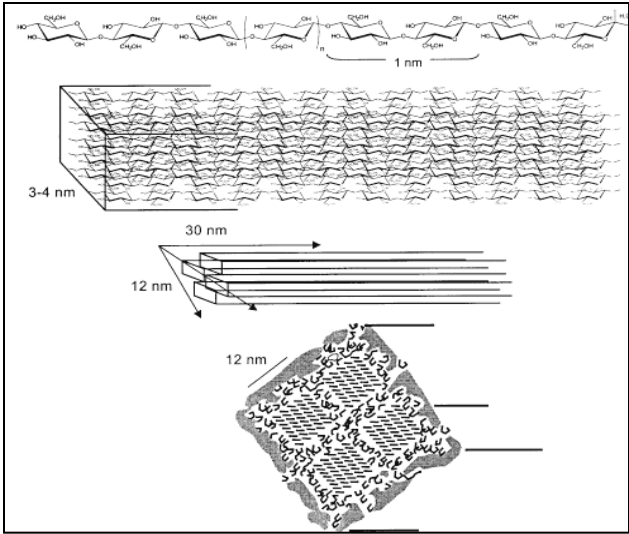
The fundamental science of biomass recalcitrance is poorly understood



Lignocellulosic Biomass Exhibits Structural Complexity

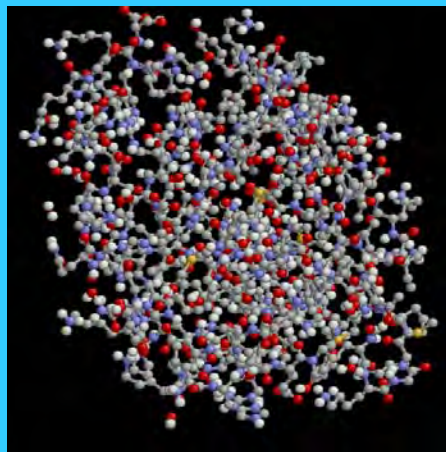


Cellulose

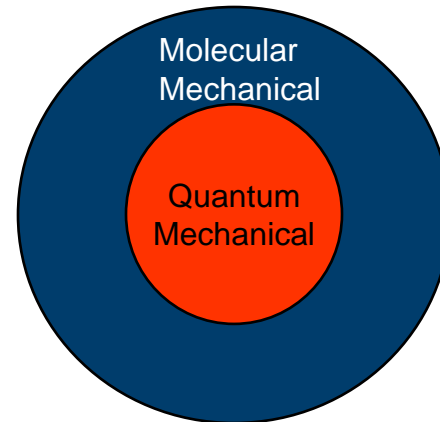


Atomic-Detail Computer Simulation - Basic Principles

Model System

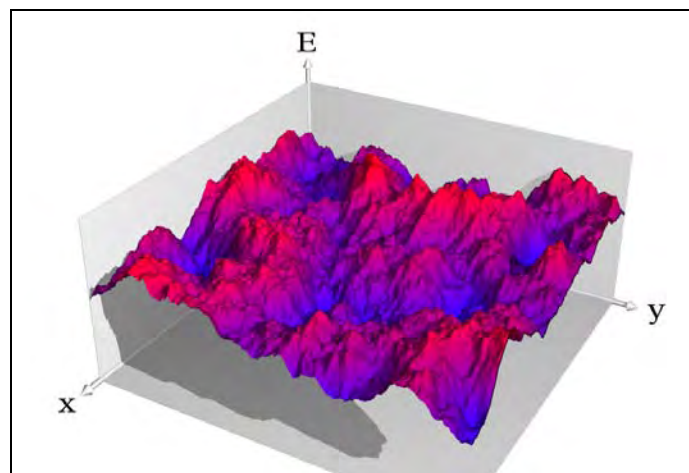


or QM/MM
Potential



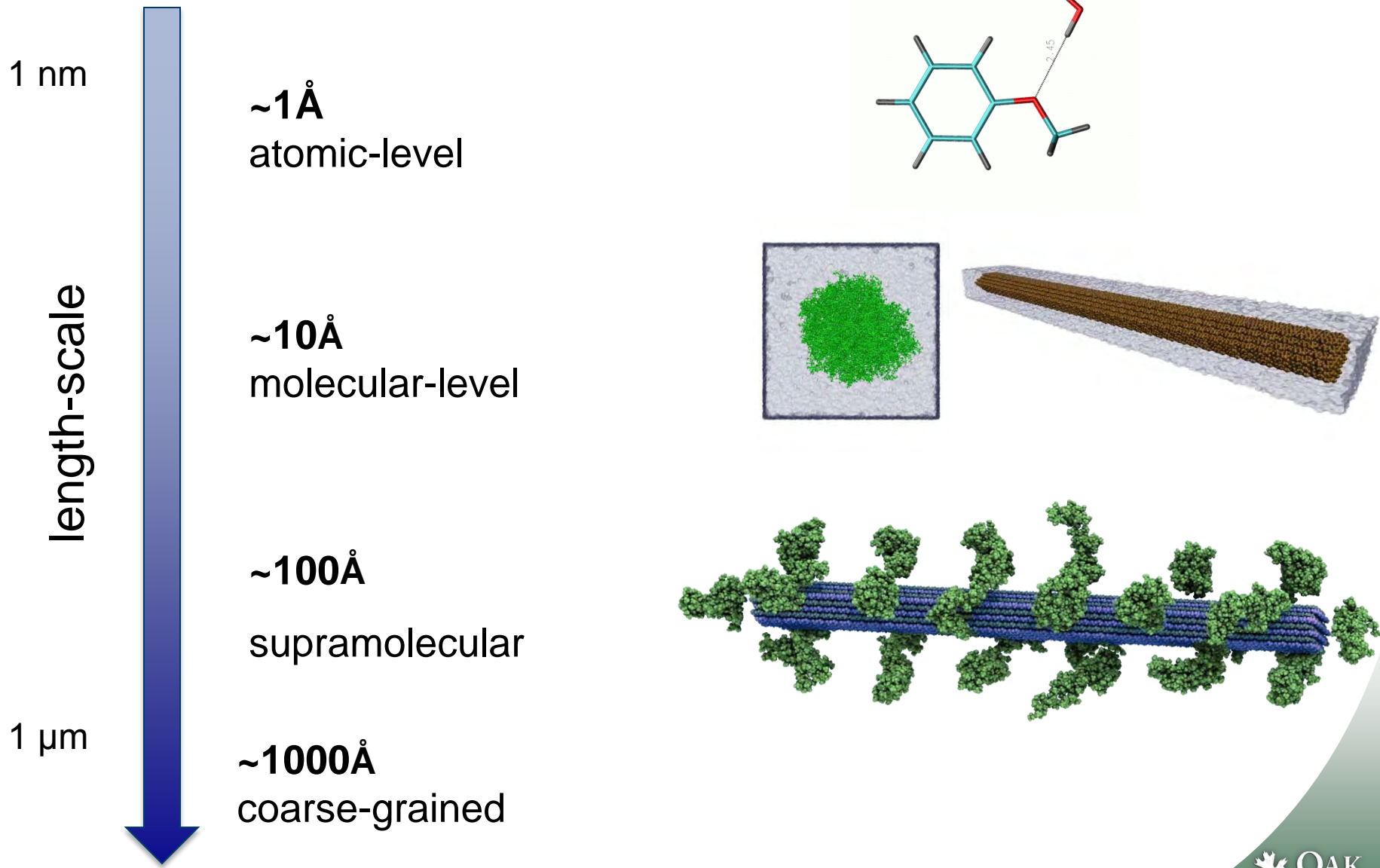
Molecular Mechanics Potential

$$V = \sum_{\text{bonds}} k_b (b - b_0)^2 + \sum_{\text{angles}} k_\theta (\theta - \theta_0)^2 + \sum_{\text{dihedrals}} \sum_{n=1}^N K_\phi^{(n)} [1 + \cos(n\phi - \delta)] + \sum_{i,j} 4\epsilon_{ij} \left[\left(\frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left(\frac{\sigma_{ij}}{r_{ij}} \right)^6 \right] + \sum_{i,j} \left(\frac{q_i q_j}{D r_{ij}} \right)$$



Simulation -
exploring the energy
landscape

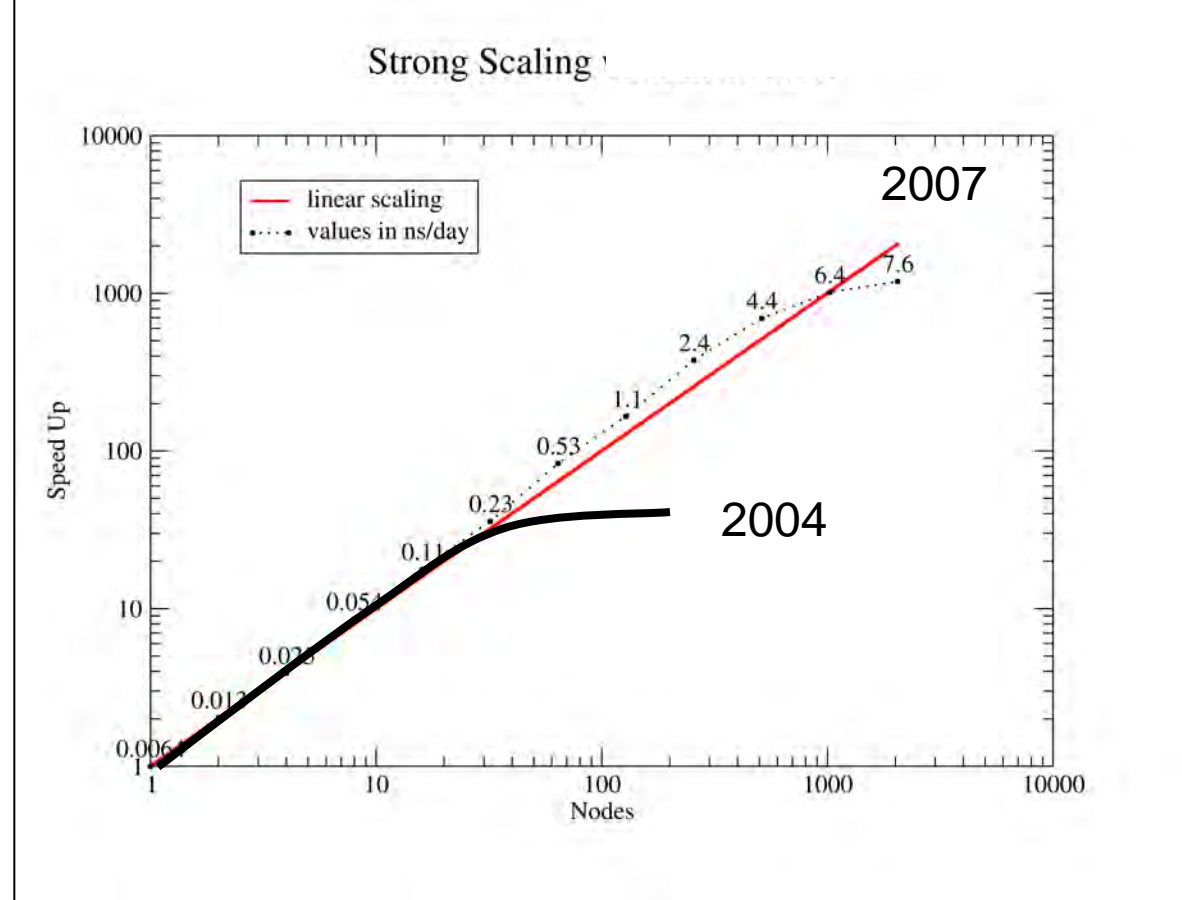
Simulation Model of Lignocellulose



One Million Atoms – Molecular Dynamics



**ROLAND
SCHULZ**

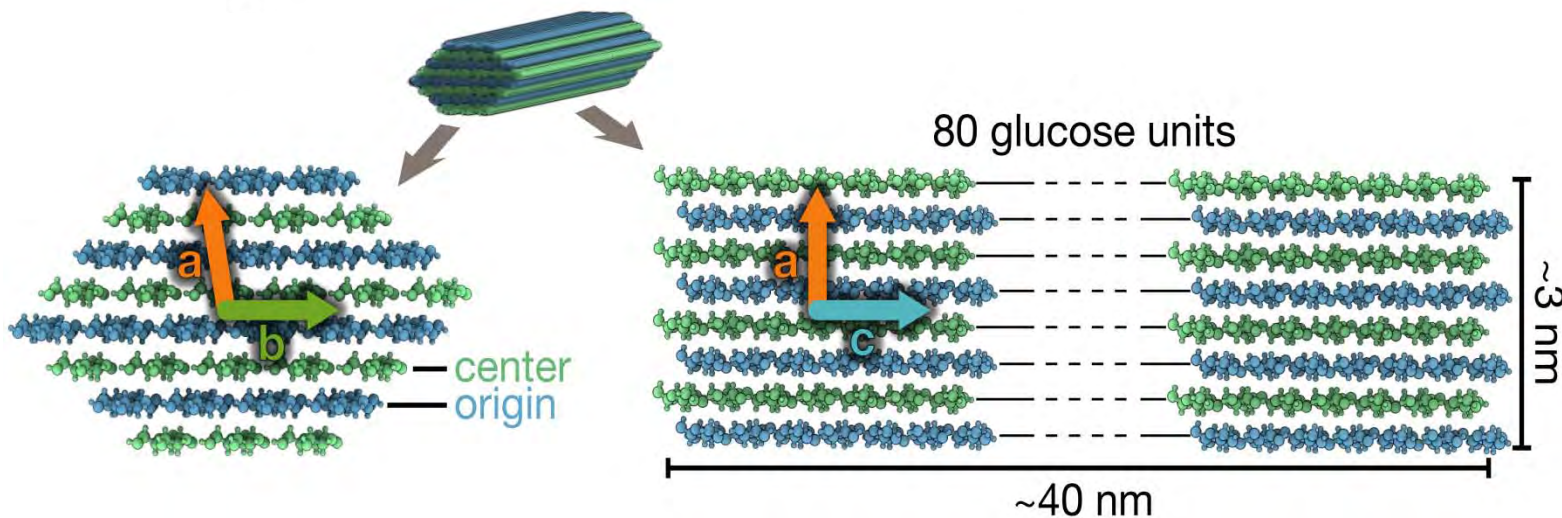
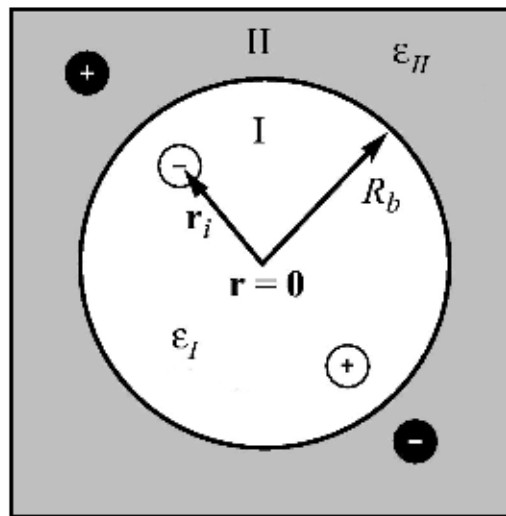


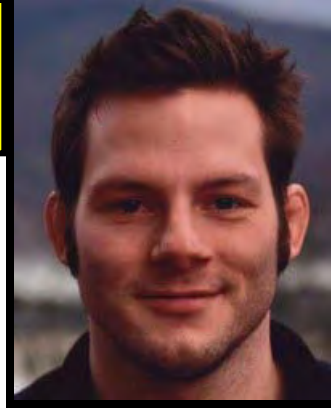
**Cray
XT4**

Electrostatics

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

- cuts simulation time to $\approx 1/3$ of PME (10k cores)
- improves scaling
- **accurate?**

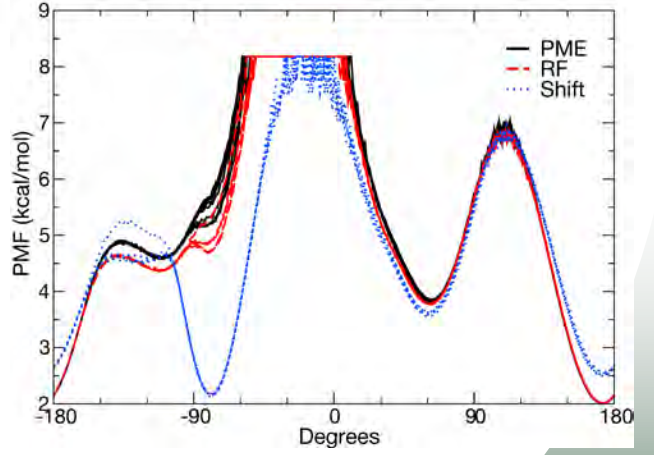
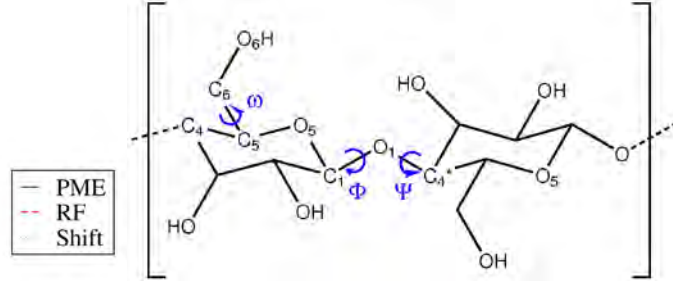
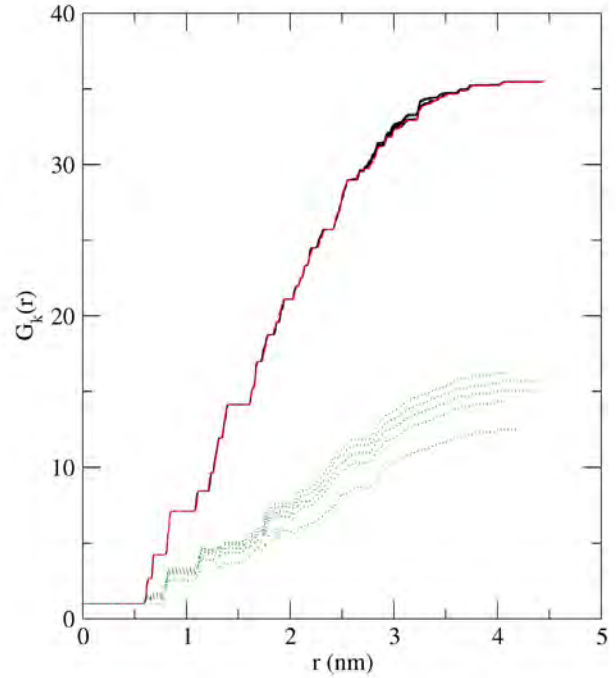
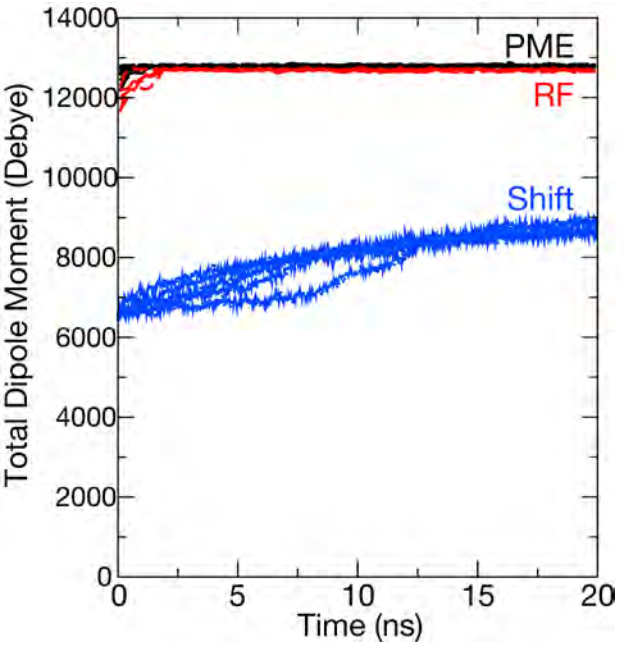




Electrostatics: Cellulose Fibril in Aqueous Solution

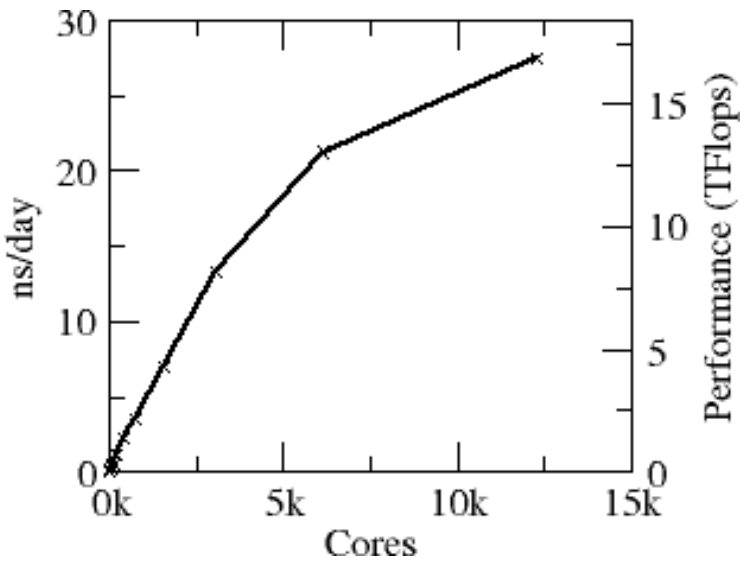
$$G_k(r) = \sum_{r_{ij} < r} \frac{\mu_i \cdot \mu_j}{\mu^2}$$

Kirkwood G Factor

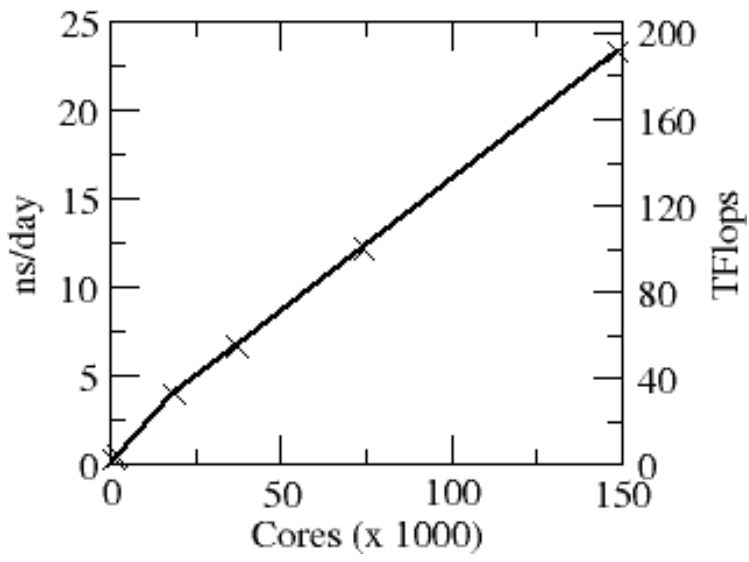




Molecular Dynamics Scaling



3.3M atom system



100M atom system

Spallation Neutron Source



Backscattering Spectrometer (BASIS) – BL 2 (2006)*
 Dynamics of macromolecules, constrained molecular systems, polymers, biology, chemistry, materials science
 Eugene Mamontov • 865.574.4108 • mamontov@ornl.gov

Nanoscale-Ordered Materials Diffractometer (NOMAD) – BL 1b (2010)
 Liquids, solutions, glasses, polymers, nanocrystalline and partially ordered complex materials
 Joerg Heidefeldt • 865.241.1825 • heidefeldt@ornl.gov

Wide Angular-Range Chopper Spectrometer (ARCS) – BL 18 (2007)
 Atomic-level dynamics in materials science, chemistry, condensed matter science
 Doug Abernathy • 865.576.5105 • abernathyd@ornl.gov

Fine-Resolution Fermi Chopper Spectrometer (SEQUOIA) – BL 17 (2008)
 Dynamics of complex fluids, quantum fluids, magnetism, condensed matter, materials science
 Garrett Granroth • 865.576.0900 • granrothg@ornl.gov

Ultra-small-Angle Neutron Scattering (USANS) Instrument – BL 1a (2012)
 Life sciences, polymers, materials science, and earth and environmental sciences
 Michael Agamian • 865.576.0903 • magamian@ornl.gov

Vibrational Spectrometer (VISION) – BL 16b (2011)
 Vibrational dynamics in molecular systems, chemistry
 Christoph Wildgruber • 865.574.5378 • wildgruber@ornl.gov

Spallation Neutrons and Pressure Diffractometer (SNAP) – BL 3 (2008)
 Materials science, geology, earth and environmental sciences
 Chris Tullis • 865.576.7038 • tullis@ornl.gov

Neutron Spin Echo Spectrometer (INSE) – BL 15 (2009)
 High-resolution dynamics of slow processes, polymers, and biological macromolecules
 Michael Ott • 865.574.8438 • ottm@ornl.gov

Hybrid Chopper Spectrometer (HYSPEC) – BL 14B (2011)
 Atomic-level dynamics in single crystals, magnetism, condensed matter sciences
 Mark Hagen • 865.241.9782 • hagenm@ornl.gov

Magnetism Reflectometer – BL 4a (2006)
 Chemistry, magnetism of layered systems and interfaces
 Halle Ambaye • 865.574.3096 • ambayeh@ornl.gov

BL 14a

Fundamental Physics Beam Line – BL 13 (2008)
 Fundamental properties of neutrons
 Geoffrey Greene • 865.574.8438 • greenge@ornl.gov

Liquids Reflectometer – BL 4b (2006)
 Interfaces in complex fluids, polymers, chemistry
 John Ankner • 865.576.5122 • anknerj@ornl.gov

Cold Neutron Chopper Spectrometer (CNCS) – BL 5 (2008)
 Condensed matter physics, materials science, chemistry, biology, environmental science
 Georg Eilers • 865.576.2911 • eilersg@ornl.gov

BL 8a

BL 8b

Elastic Diffuse Scattering Spectrometer (CORELLI) – BL 9 (2013)
 Detailed studies of disorder in crystalline materials
 TBO

Macromolecular Diffractometer (MANDI) – BL 11b (2012)
 Atomic-level structures of membrane proteins, drug complexes, and DNA
 Leighton Coates • 865.363.6180 • coatesl@ornl.gov

Single-Crystal Diffractometer (TOPAZ) – BL 12 (2009)
 Atomic-level structures in chemistry, biology, earth science, materials science, condensed matter physics
 Christina Hoffmann • 865.576.5127 • hoffmanncc@ornl.gov

Extended Q-Range Small-Angle Scattering Diffractometer (EQ-SANS) – BL 6 (2008)
 Life science, polymer and colloidal systems, materials science, earth and environmental sciences
 Jinkui Zhao • 864.574.0411 • zhaocj@ornl.gov

BL 10

Engineering Materials Diffractometer (VULCAN) – BL 7 (2008)
 Engineering, materials science, materials processing
 Xun-Li Wang • 865.574.9164 • wangxl@ornl.gov

Powder Diffractometer (POWGEN) – BL 11a (2008)
 Atomic-level structures in magnetism, chemistry, materials sciences
 Jason Hoopes • 865.576.7034 • hoopesj@ornl.gov

* Date shown is the scheduled commissioning date.

LEGEND		
	SNS TPC	
	DOE Grant	
	SING I	
	DOE NP	
		Non U.S.

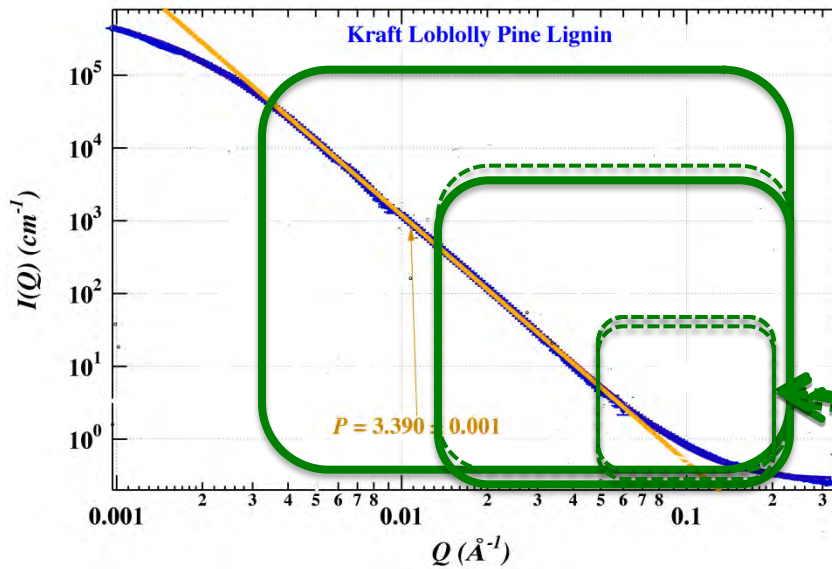


NEUTRON SCIENCES

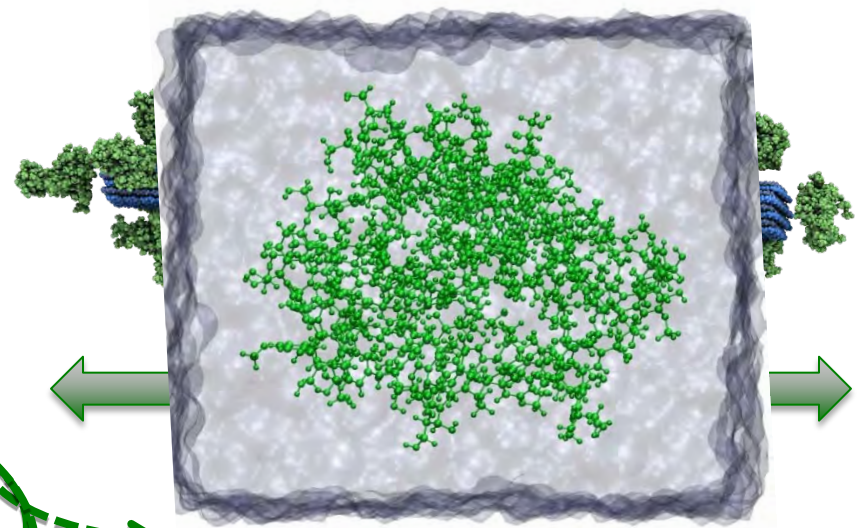
06-G00400H/am
National Laboratory

Length-Scale Correspondence: Simulation versus Experiment

Small-Angle Neutron Scattering



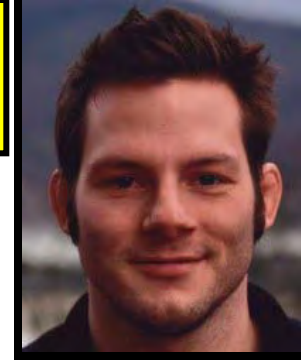
Simulation



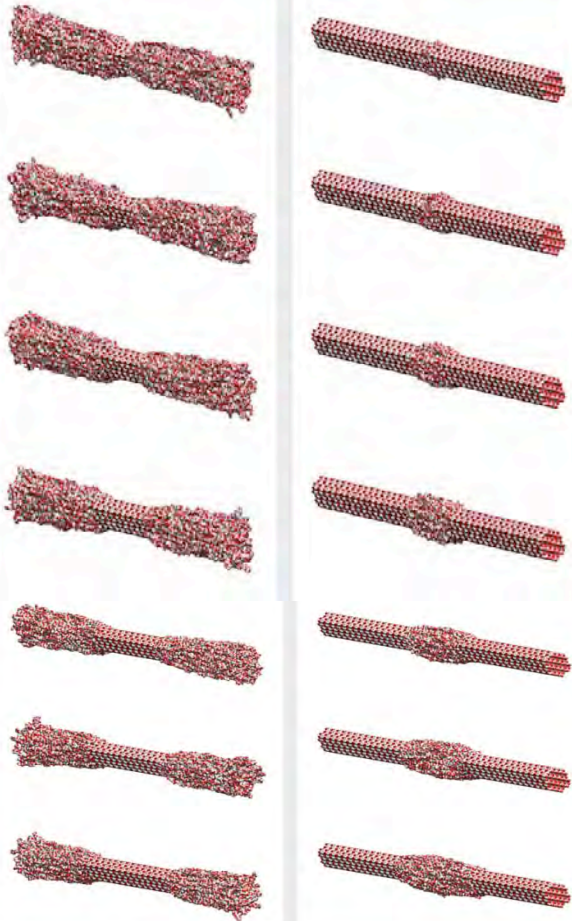
100 Å / ~500k atoms
 10 Å / ~50k atoms
 NSF Teragrid

Cellulose: Models of Pretreatment

**BENJAMIN
LINDNER**

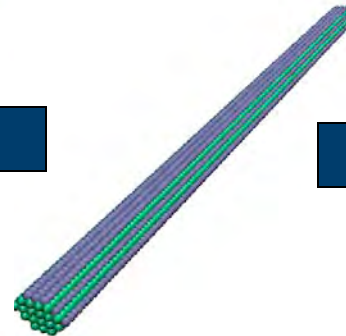
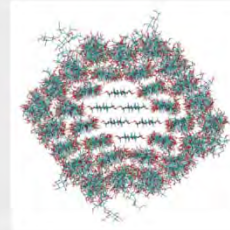
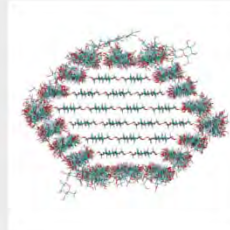


Crystalline/Amorphous



inner & outer core
crystalline

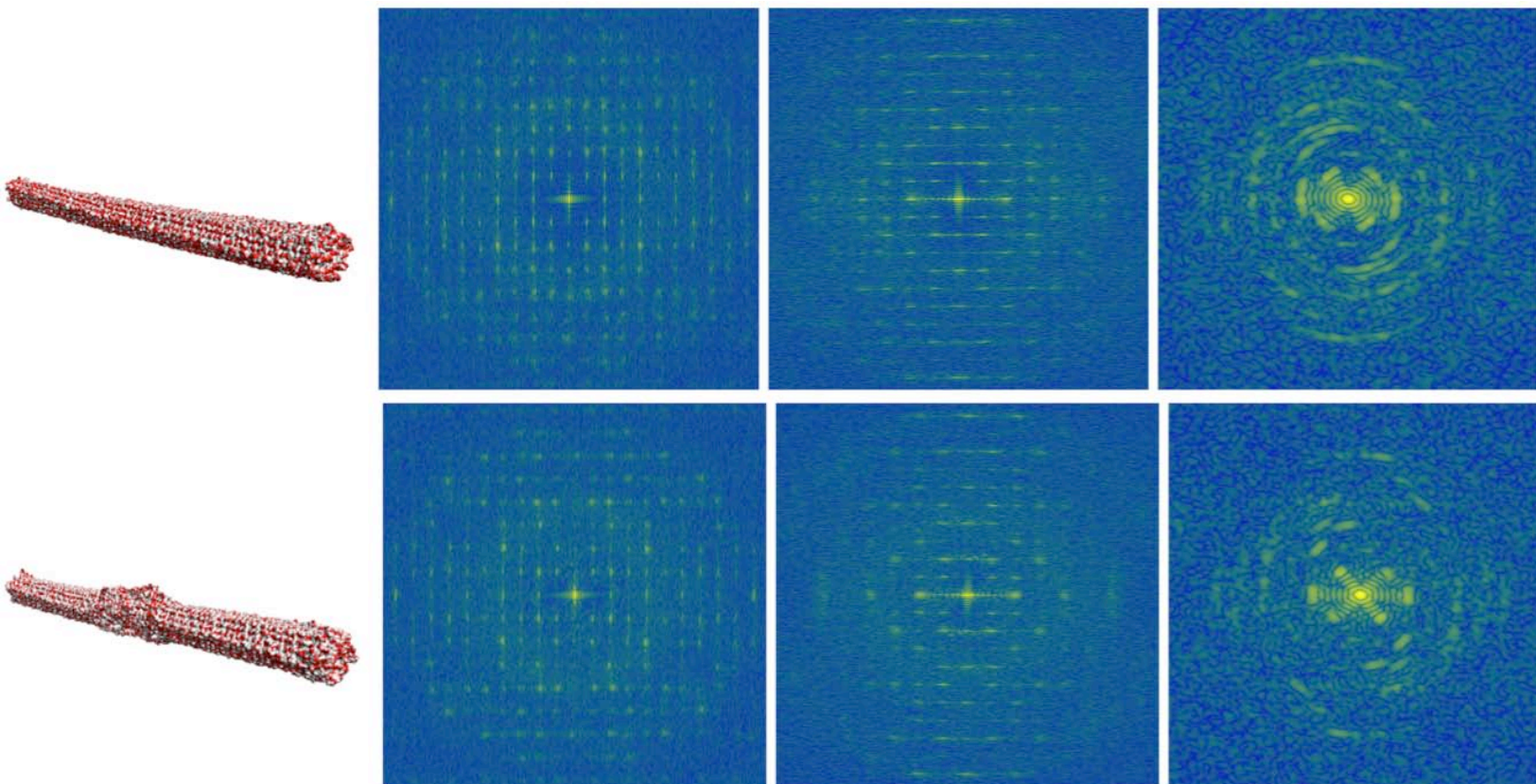
inner core
crystalline



Crystalline
(1ns@300K)

Amorphous
(1ns@650K)

Bragg Scattering from MD Simulation

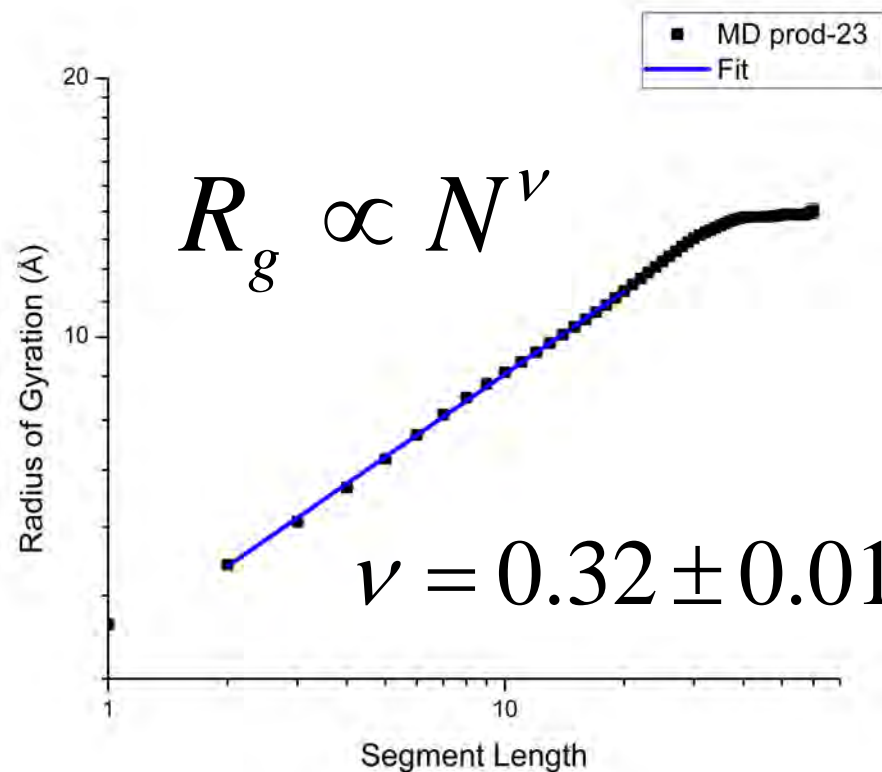
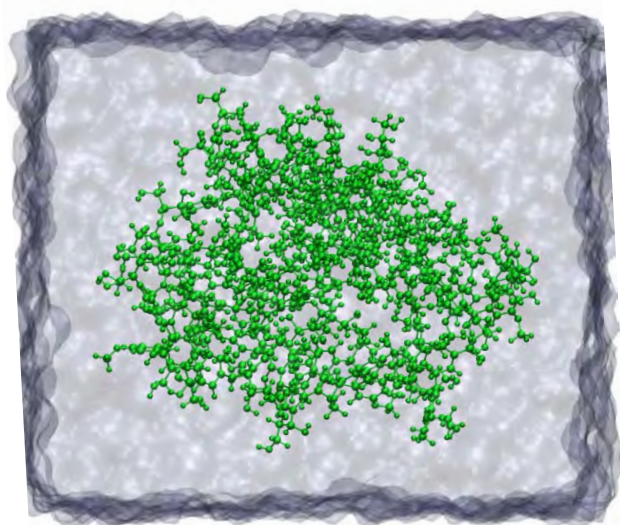
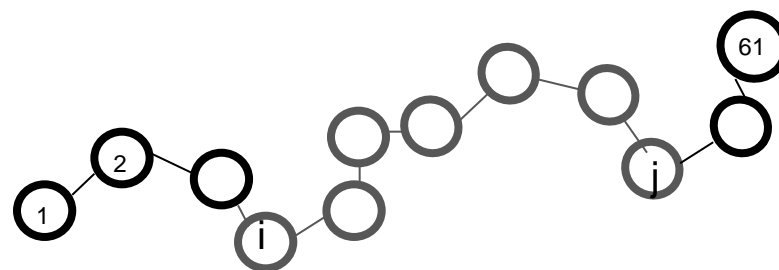




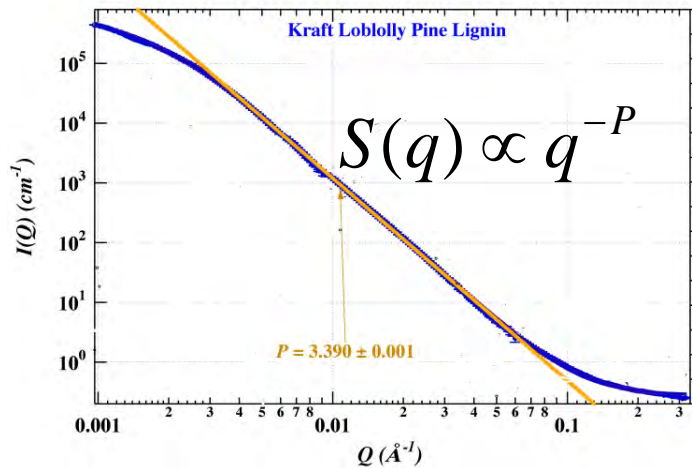
**LOUKAS
PETRIDIS**

Single Lignin Molecule

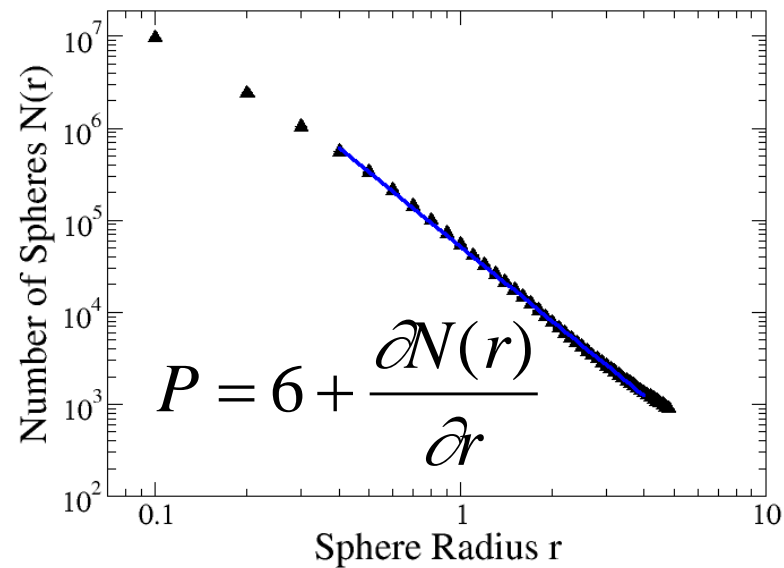
R_g = Segment Radius of Gyration



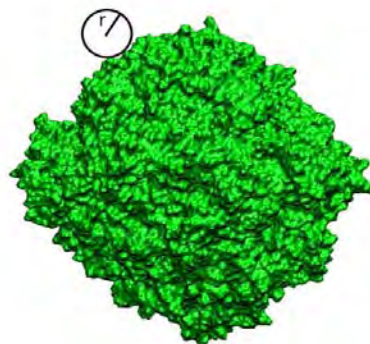
26-molecule lignin aggregate



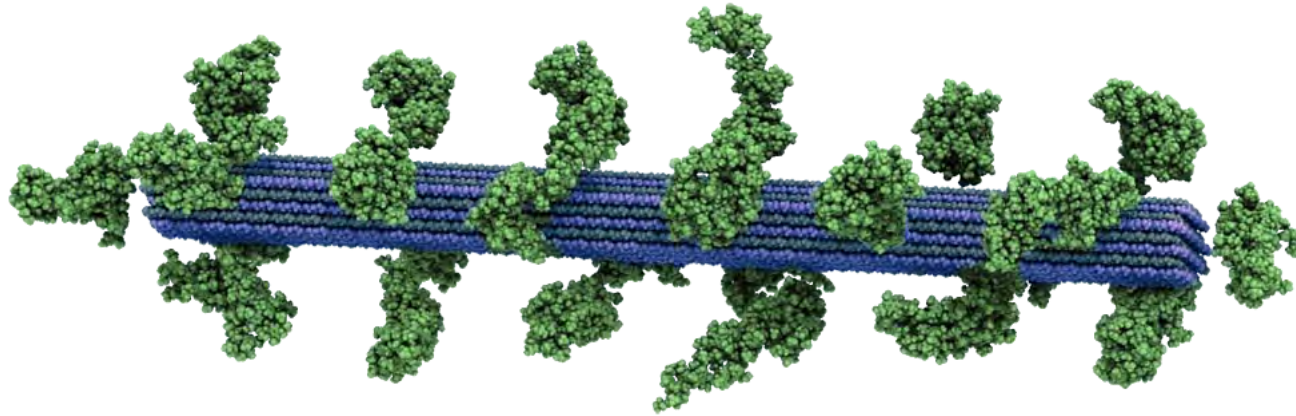
$$P_{\text{neutron}} = 3.39 \pm 0.001$$



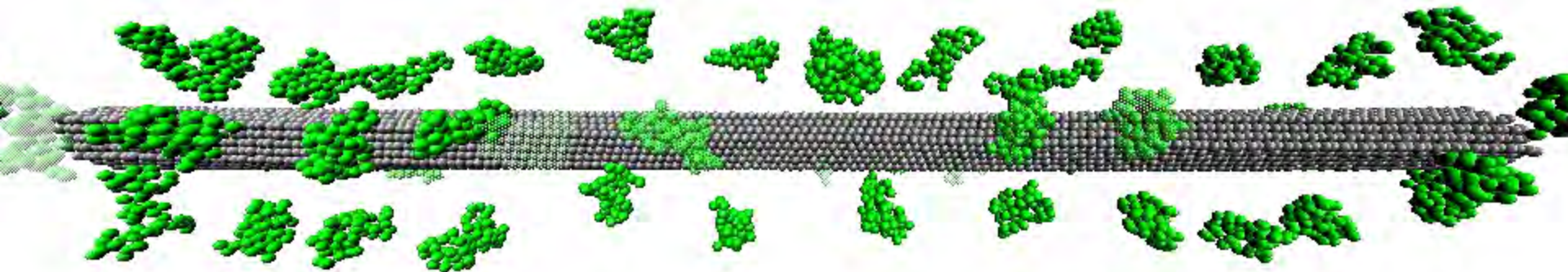
$$P_{\text{simulation}} = 3.31 \pm 0.001$$



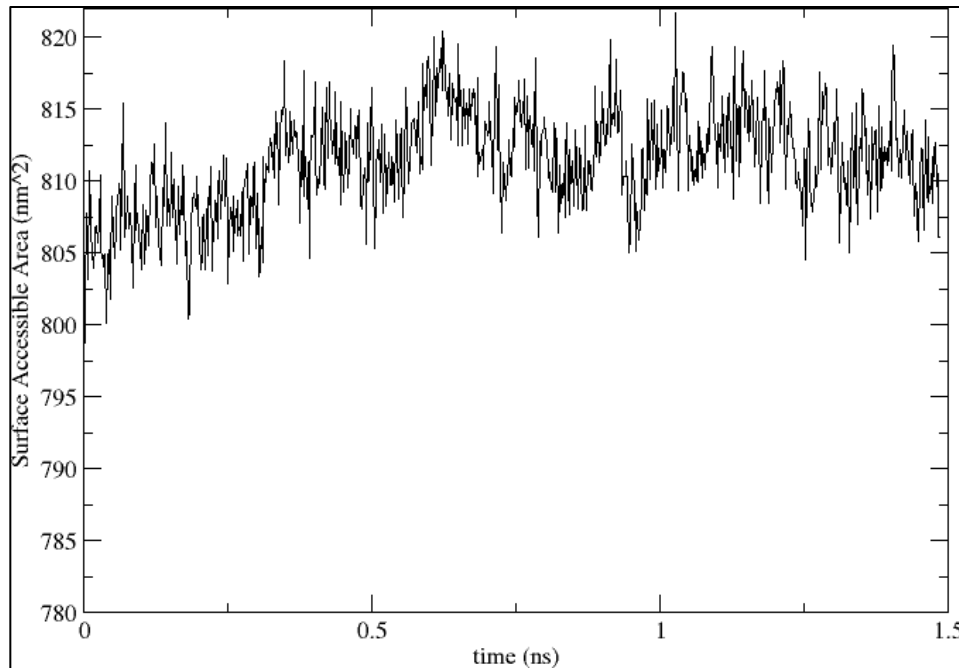
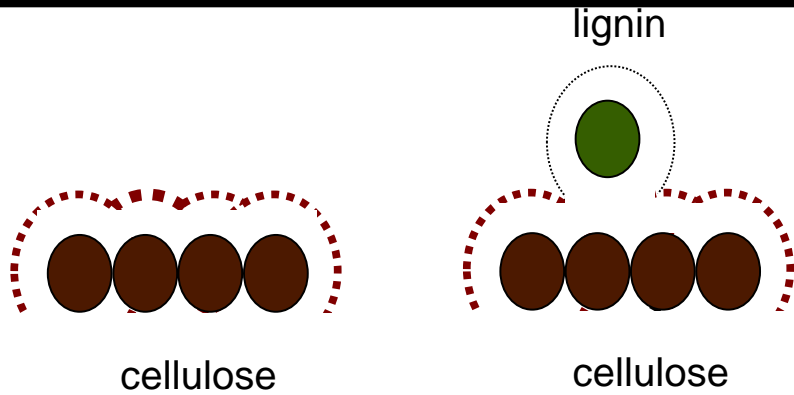
Simulation Model of Lignocellulose



Complex of cellulose and lignins
3.3 M atoms

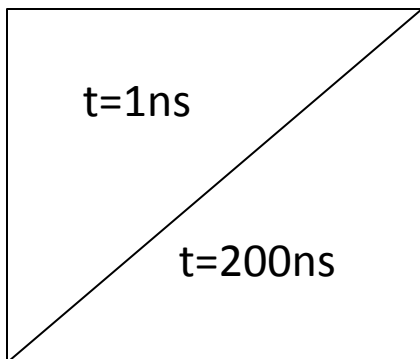


Cellulose Accessible Surface Area

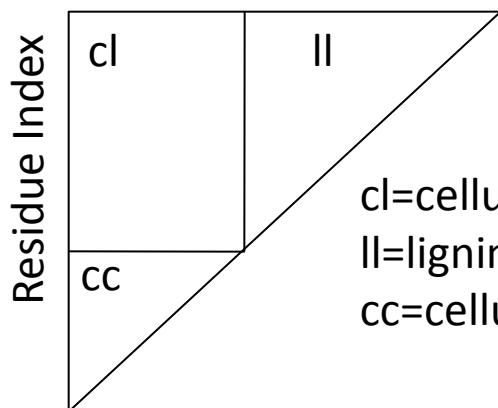
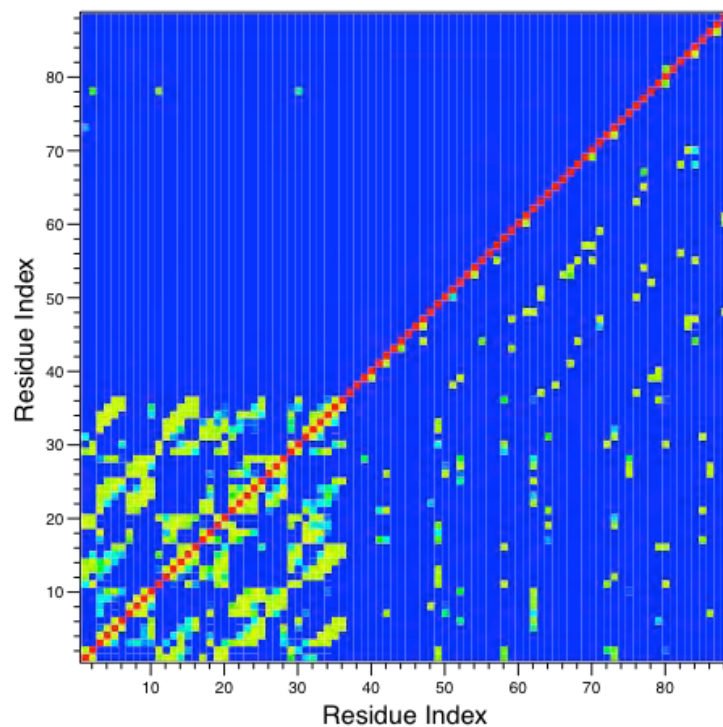


Topologic information via contact maps

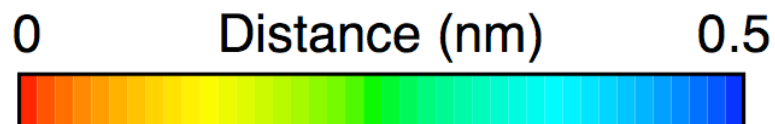
Combined Contact Map



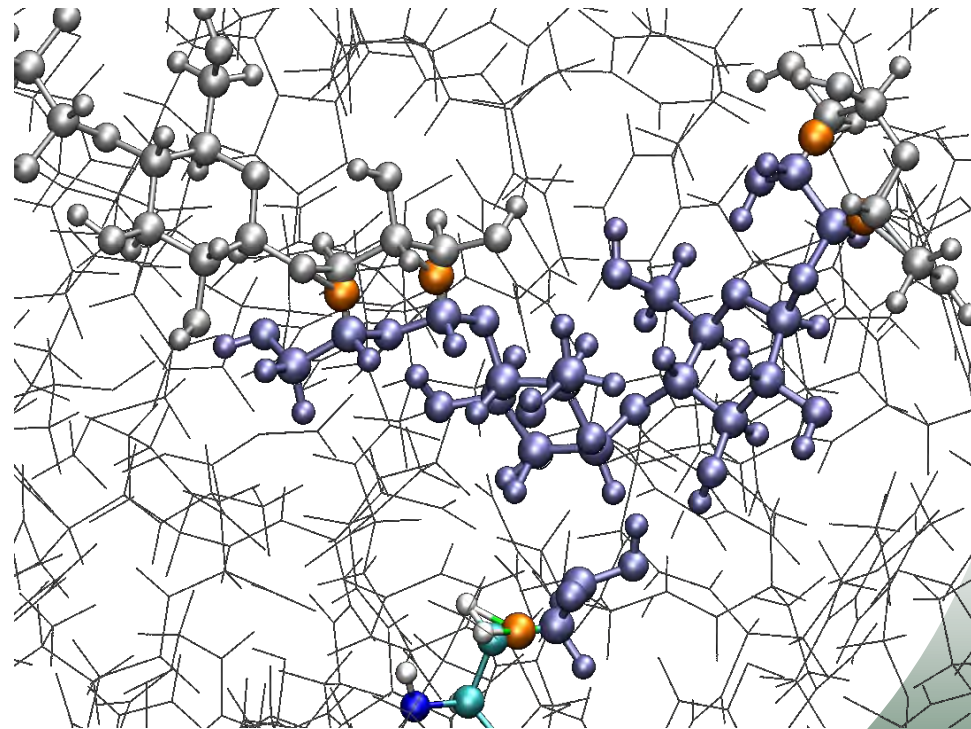
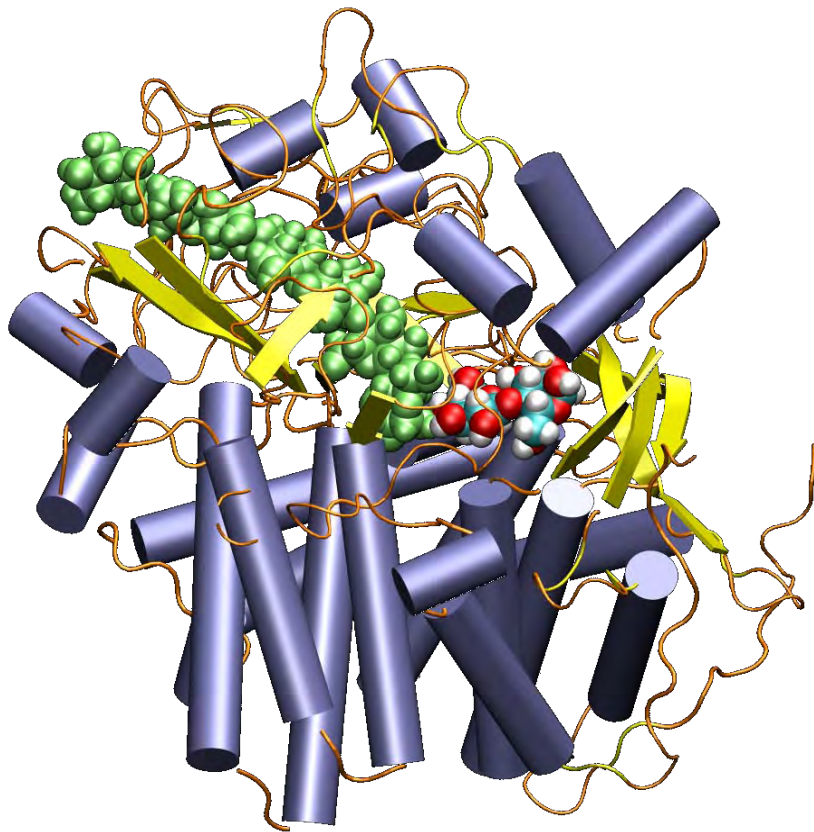
Amorph / far



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



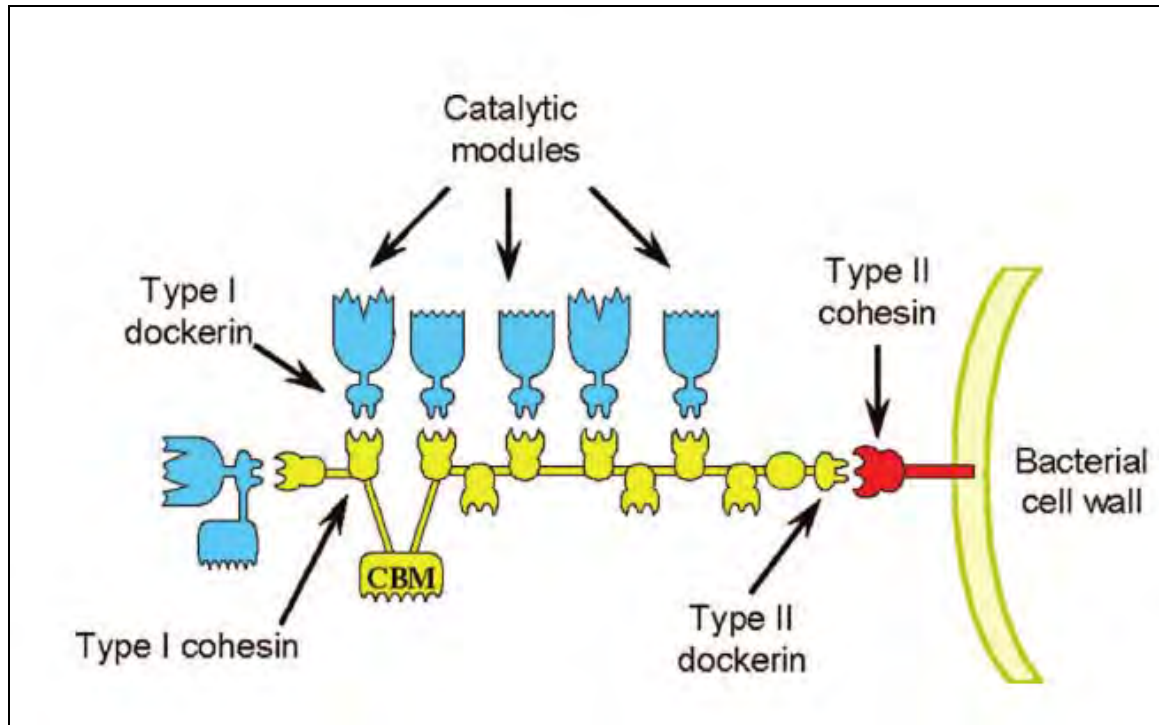
Catalytic Mechanism of Cellulase *CelS*



Catalytic Mechanism of Cellulase *CelS*



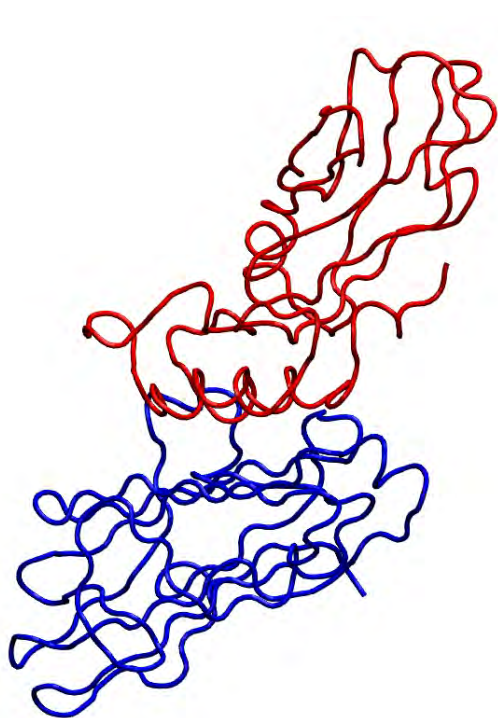
Cellulosome Organization



Cellulosome assembly is mediated by
cohesin-dockerin interaction.

Cohesin-Dockerin Principal Component Modes

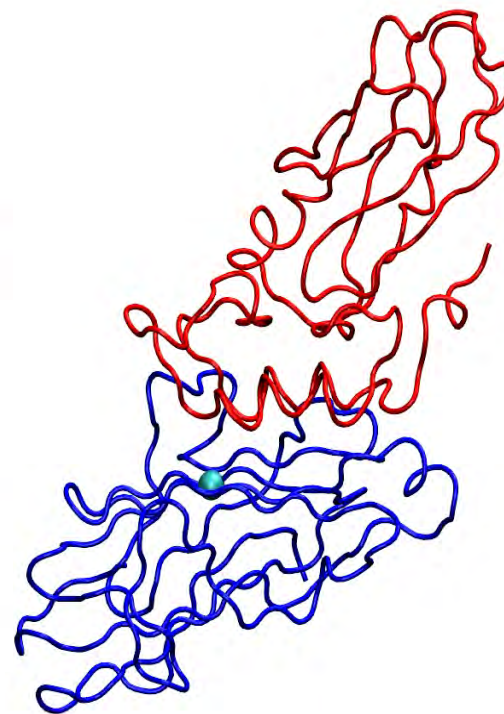
JIANCONG
XU



Wild Type

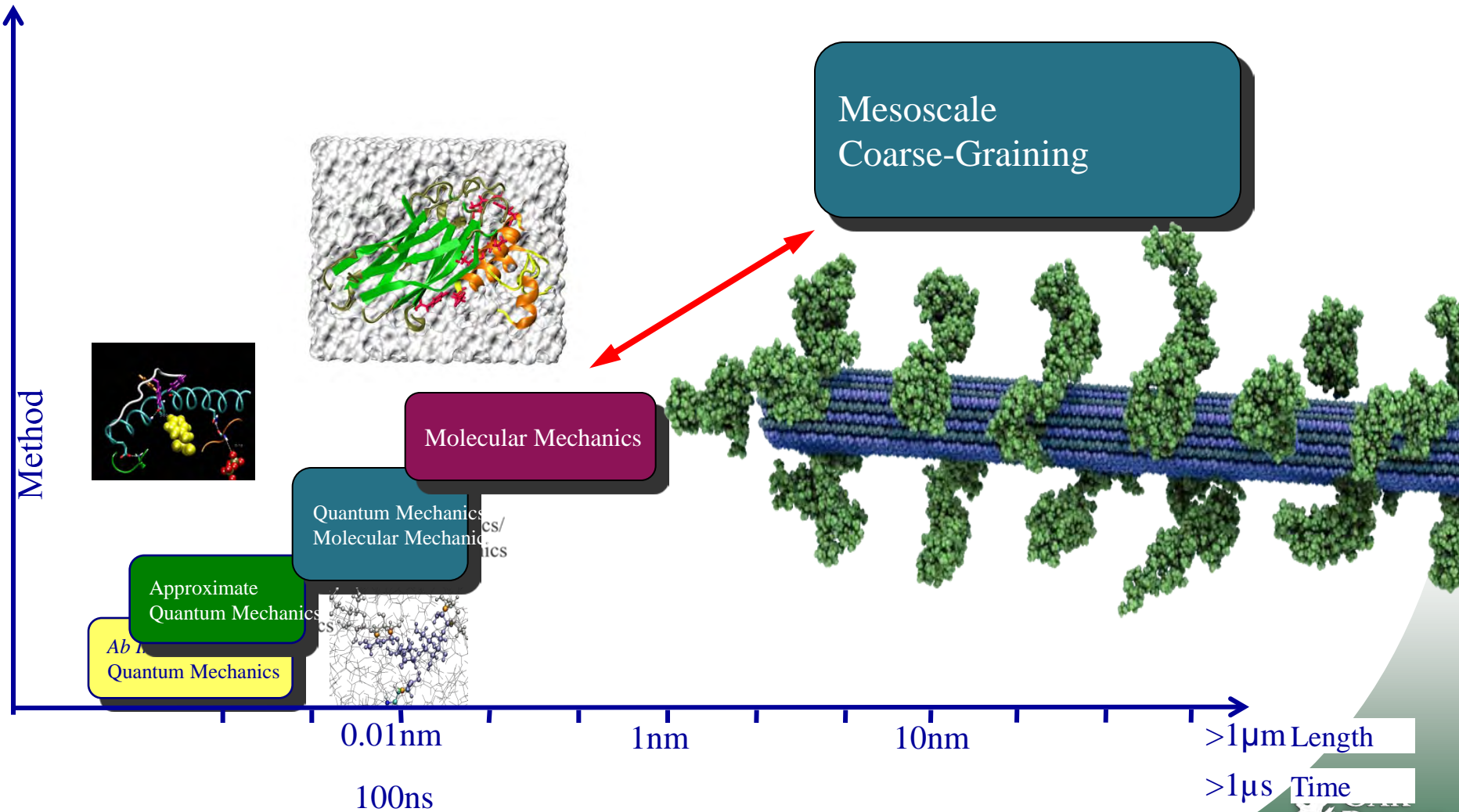


F162A



Q52A

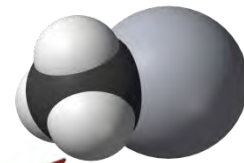
Multiscaling



Oak Ridge



Toxic
Methylmercury
 CH_3Hg^+



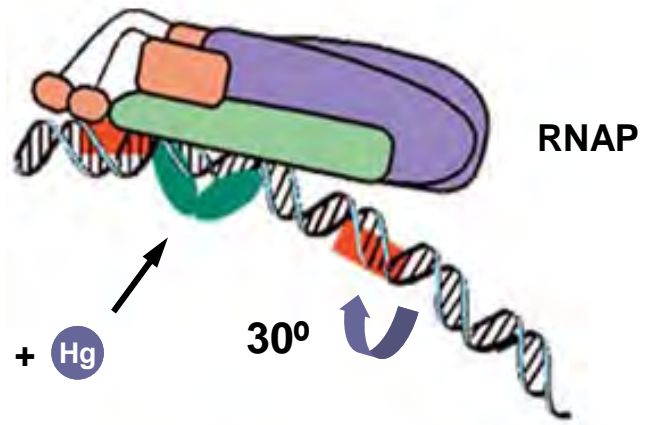
Inert
 $\text{Hg}(0)$



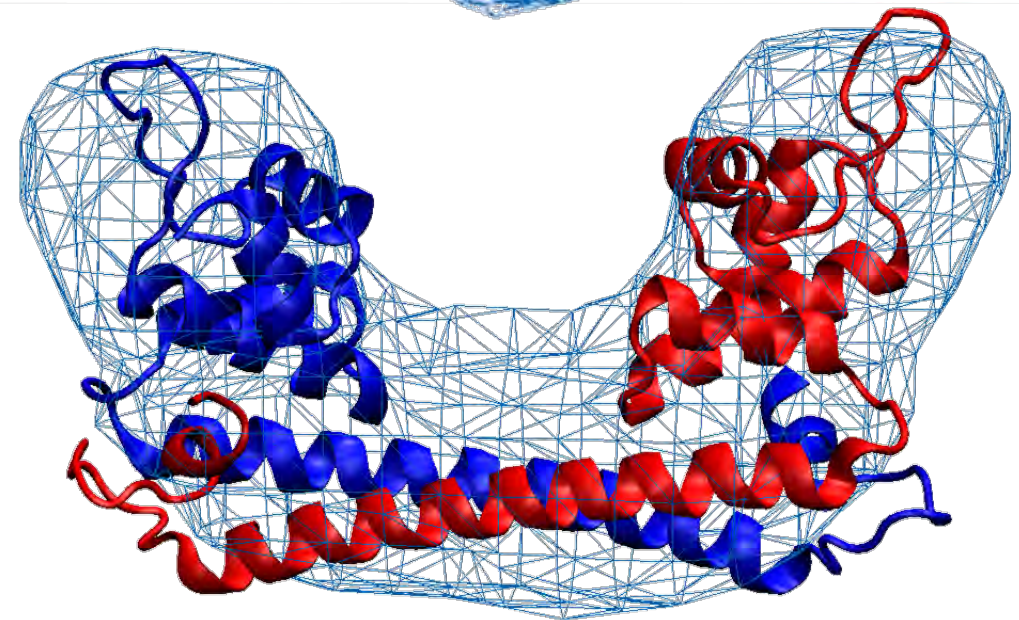
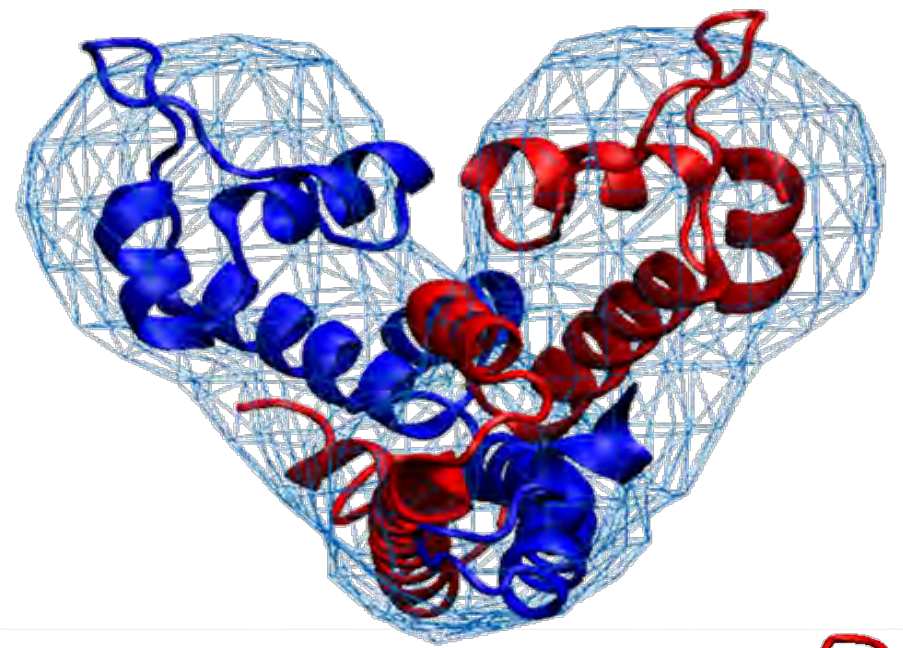
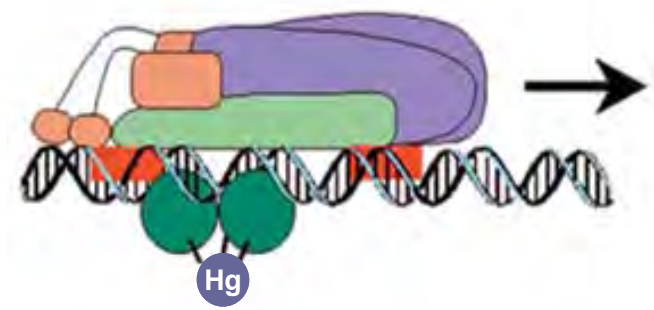
MerR



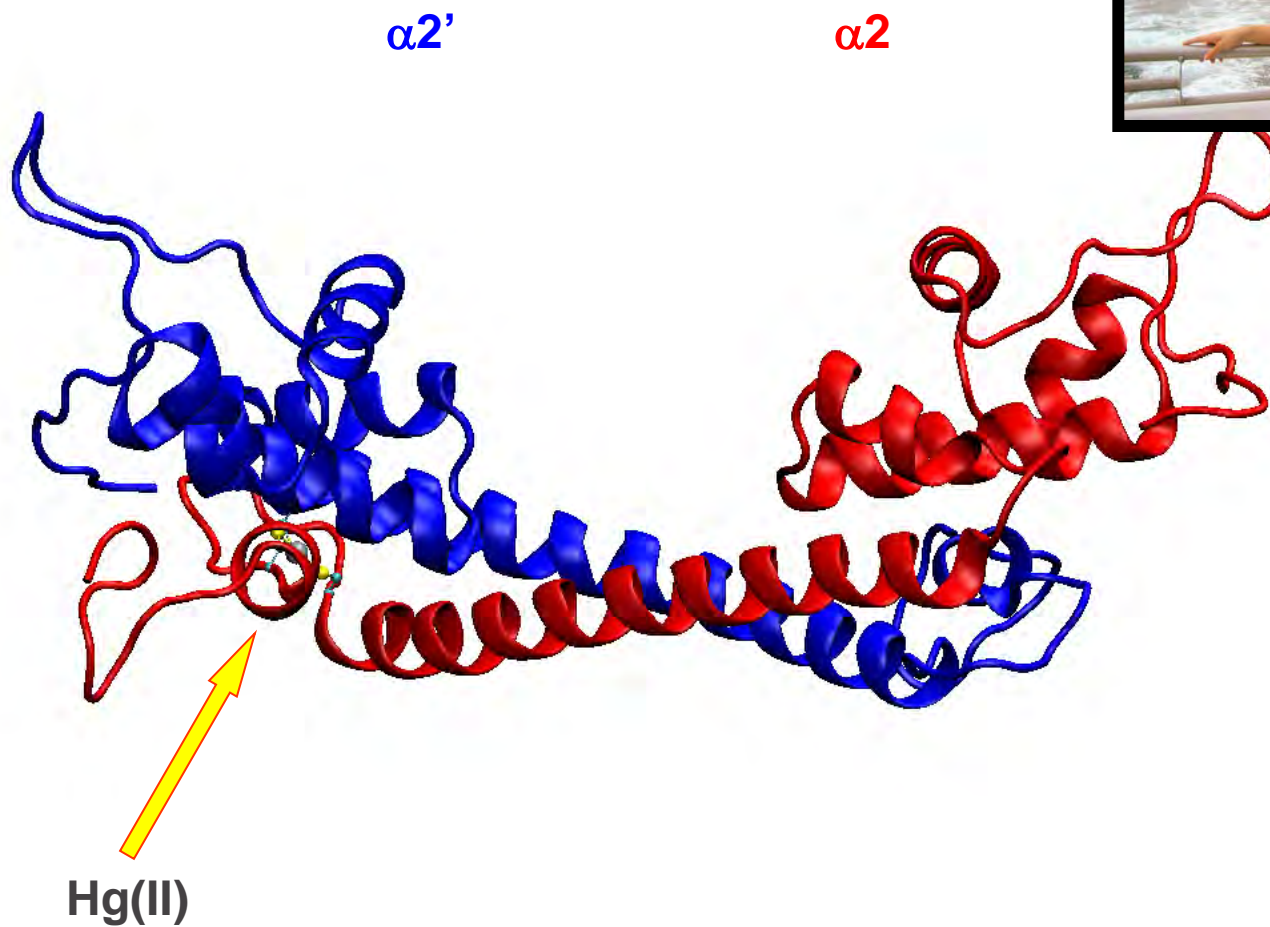
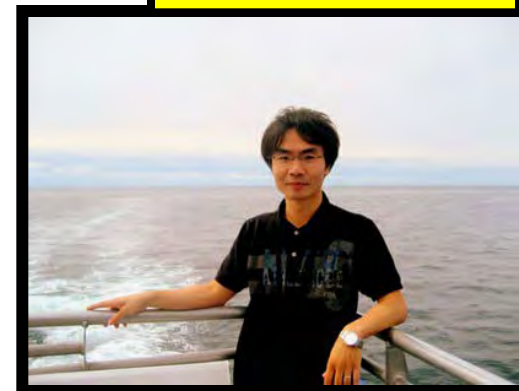
DNA



RNAP

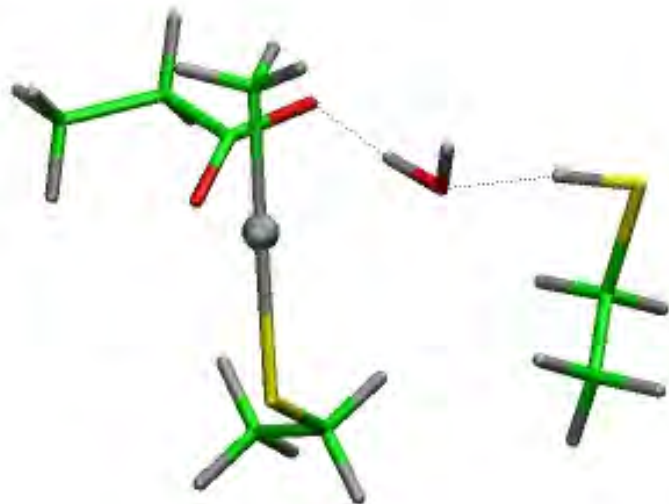
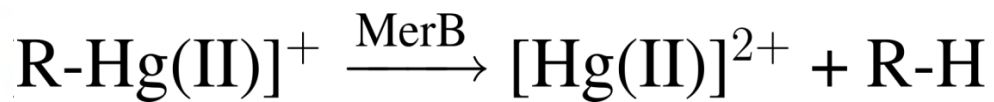
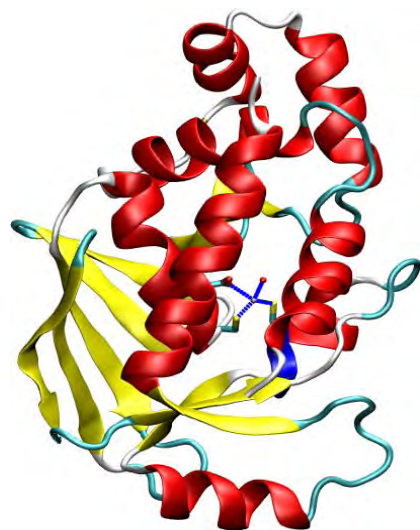


The MerR Machine

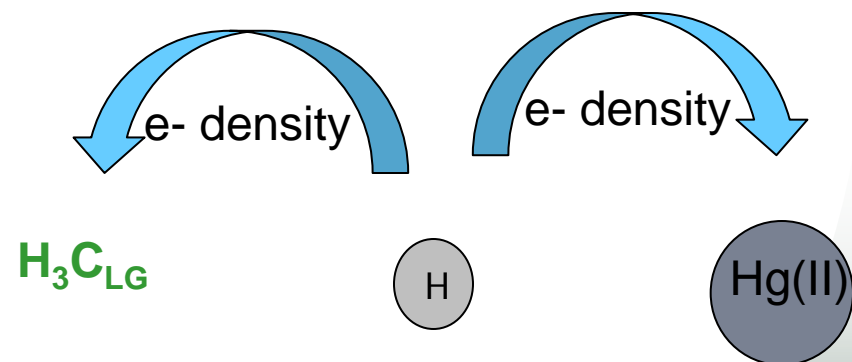


Catalytic Mechanism of MerB Hg-C Protonolysis

**JERRY
PARKS**



- 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**

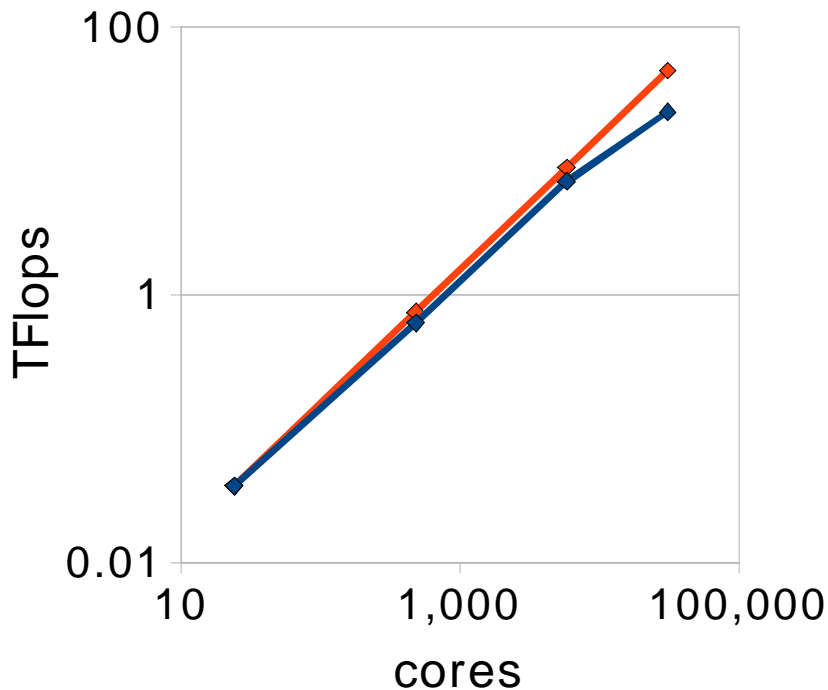
Molecular Dynamics Scaling

**ROLAND
SCHULZ**



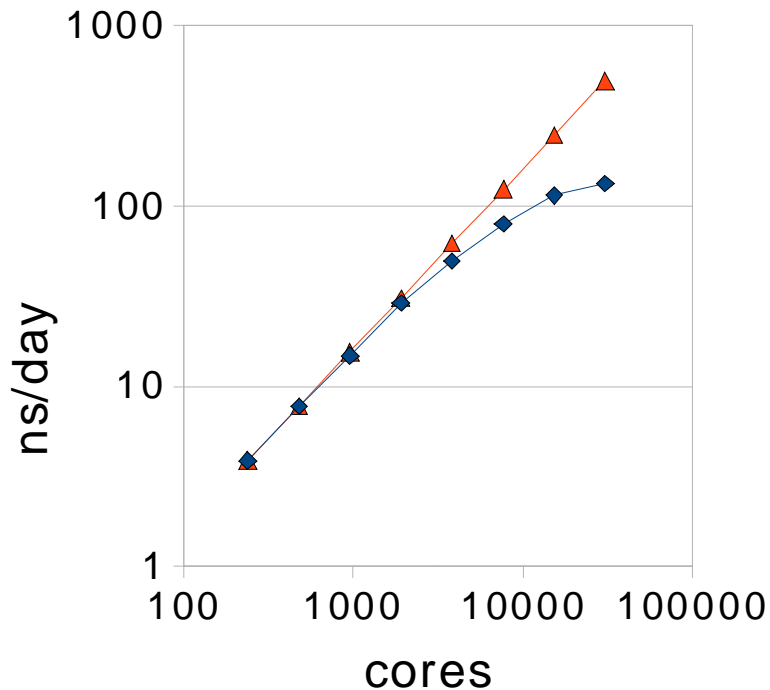
Weak Scaling

175 atoms/core

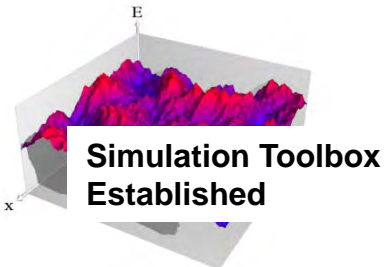


Strong Scaling

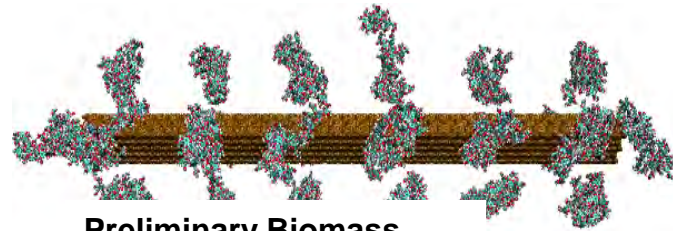
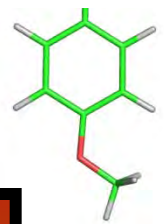
5.4 million atoms



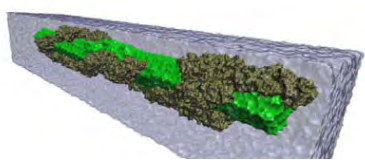
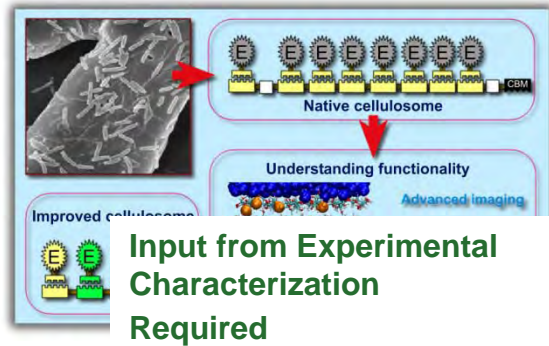
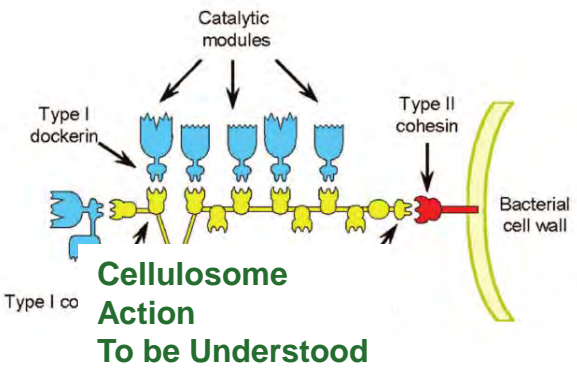
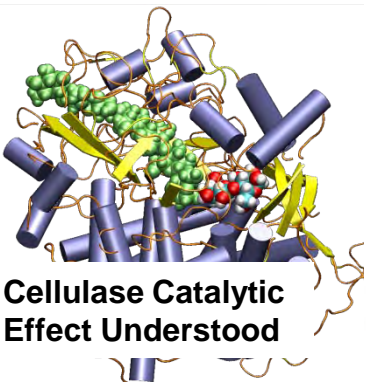
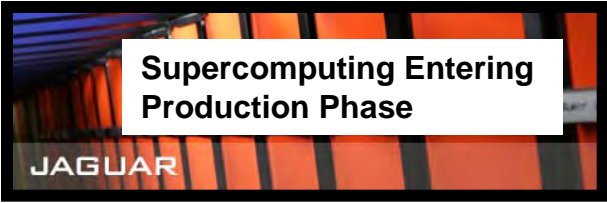
Future



Force Fields Established



Preliminary Biomass Models Built



Pretreatment Effects To be Examined