Macromolecular-Based Multicomponent Materials

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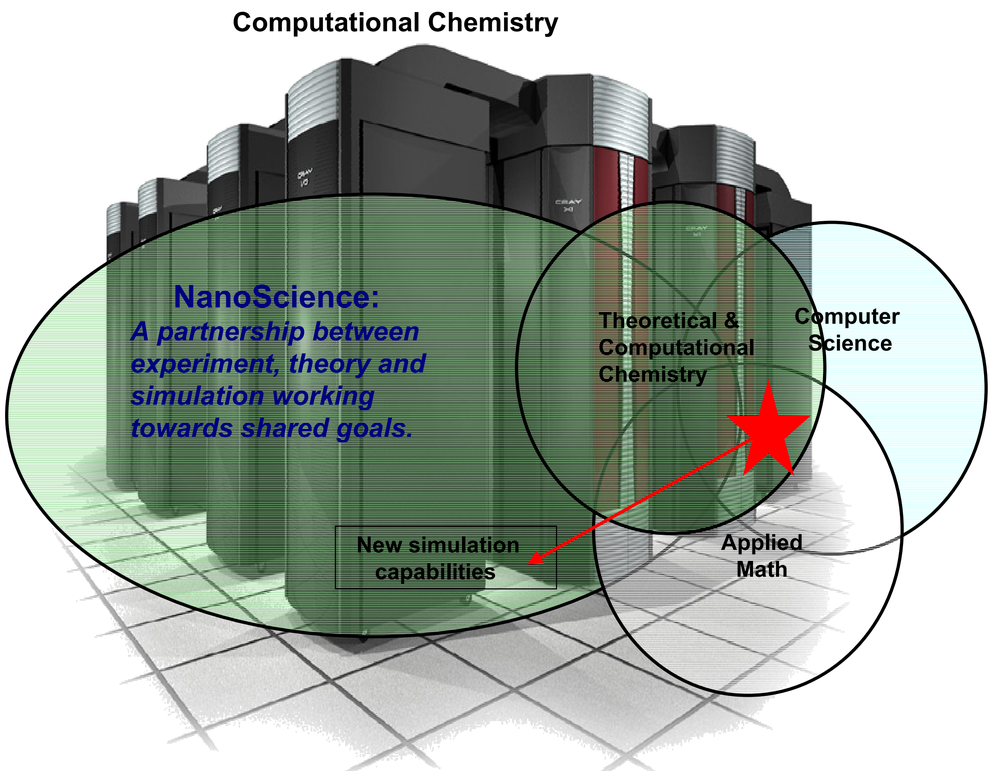
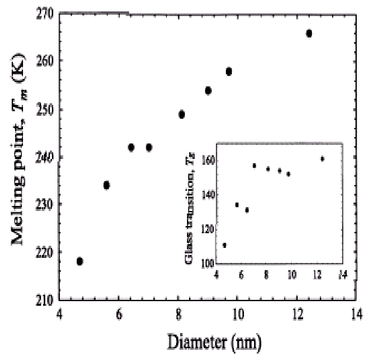


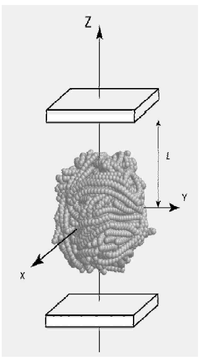
Atomic-scale methods provide information on the structure, dynamics, and thermodynamics of the system.
Molecular dynamics (MD), mechanics (MM), Monte Carlo
Large-scale normal-mode analysis (NMA).

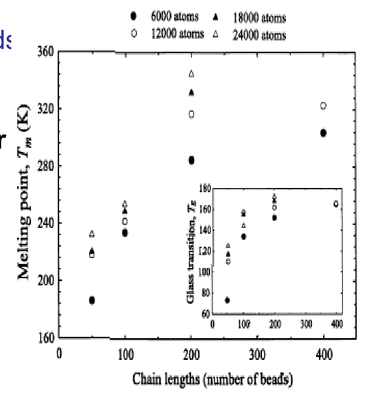
Ab Initio (DFT-MD) methods used to compute electronic and molecular structure, and to obtain interaction potentials, activation energies for transitions, and electronic spectra as input into MD, kinetic MC, course grain approaches.
NWChem, CPMD, CASTEP, VASP, new Wavelet based methods

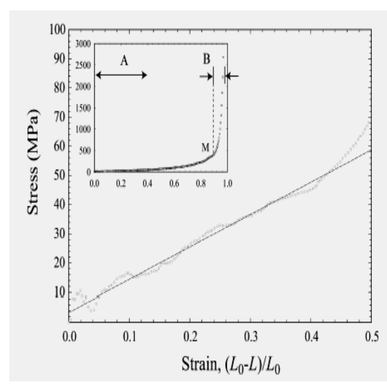
Heuristic methods provide complementary ways to perform efficient optimization and modeling based on experimental or simulation data
Computational neural networks, evolutionary algorithms

All codes are implemented in parallel and run on ORNL/CCS resources







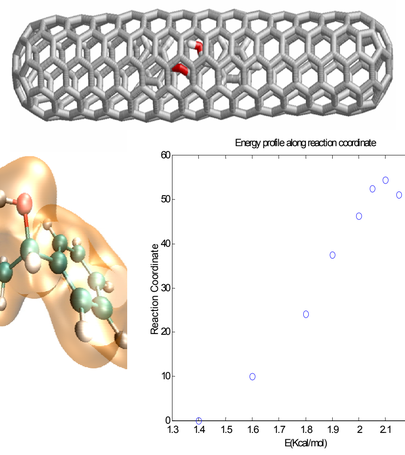


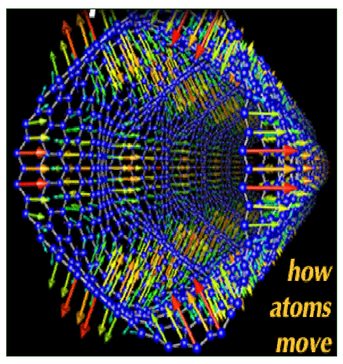
Effects of Nano-Confinement

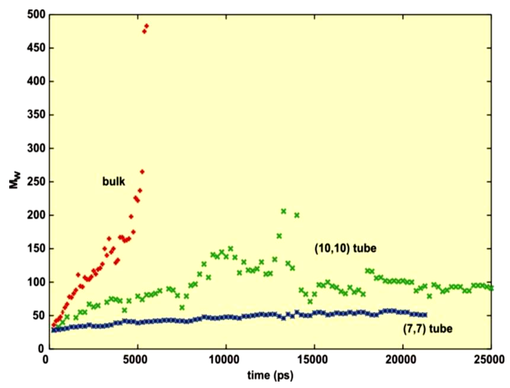
Reactions involving: dissociation, association, re-arrangements

- *Thermal decomposition, polymerization, isomerization, etc*
- *Classically the reaction kinetics are inhibited by confinement; quantum predicts enhanced rates*

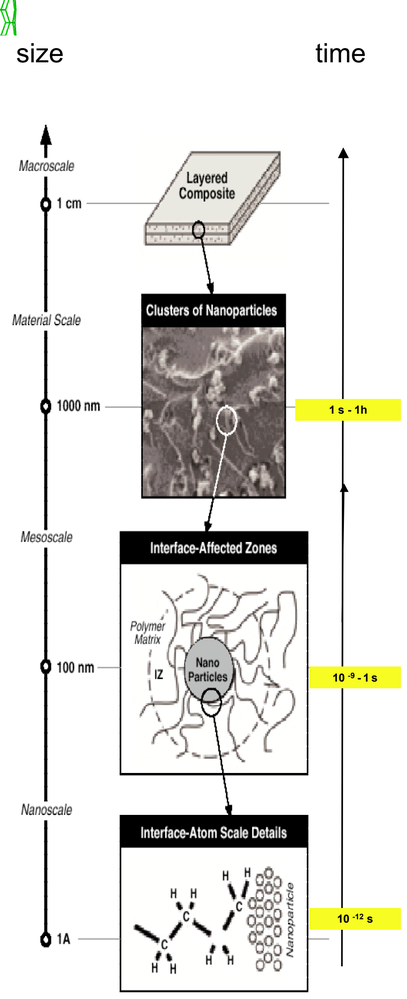
examine coupling to vibrational modes ➔ mode specificity

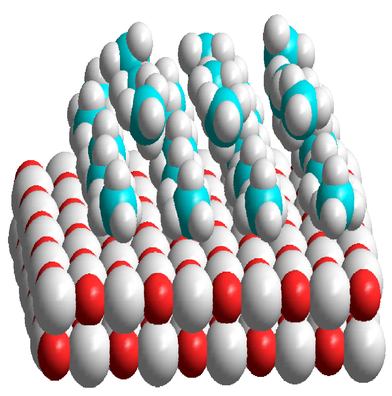


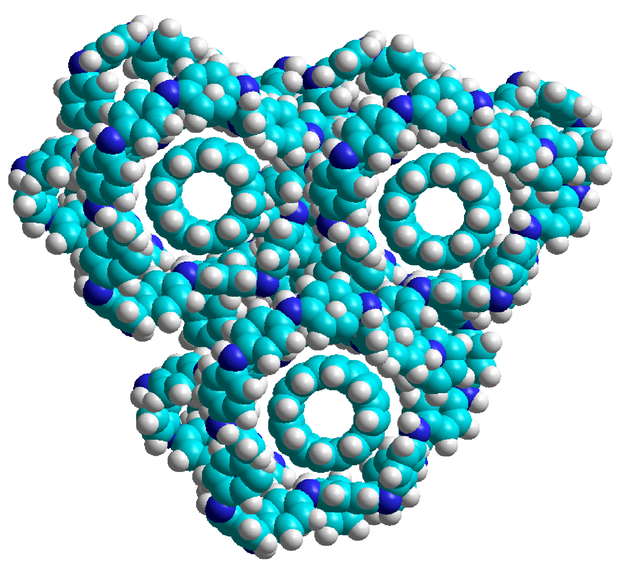




size time







Using Nano-Confinement to Tailor Semiconducting Polymers: New Generation Optoelectronics

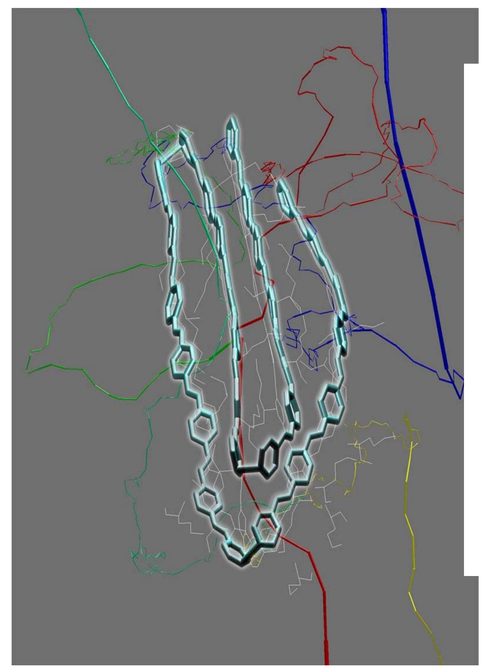
Goal: To make polymer analogues of inorganic semiconductor quantum dots *without* specialized synthetic chemistry

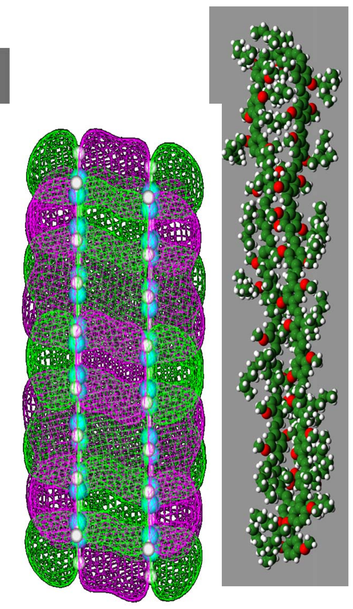
Issues:
- Control of chain organization and alignment
- Optimization of photophysical properties

Applications: Electronic paper, luminescent clothing, display technologies, photovoltaic devices, light emitting diodes, field-effect transistors, solid state lasers, biomedical imaging!
"…brighter, thinner, lighter, faster"

Key Breakthrough: single molecule nanoparticles generated from dilute solutions of semiconducting polymers – Simulations show how 3-D confinement and solvent can lead to self-organization into the optimal structure!

The nanoparticles act like single (z-oriented) atoms - signature of quantum dot behavior

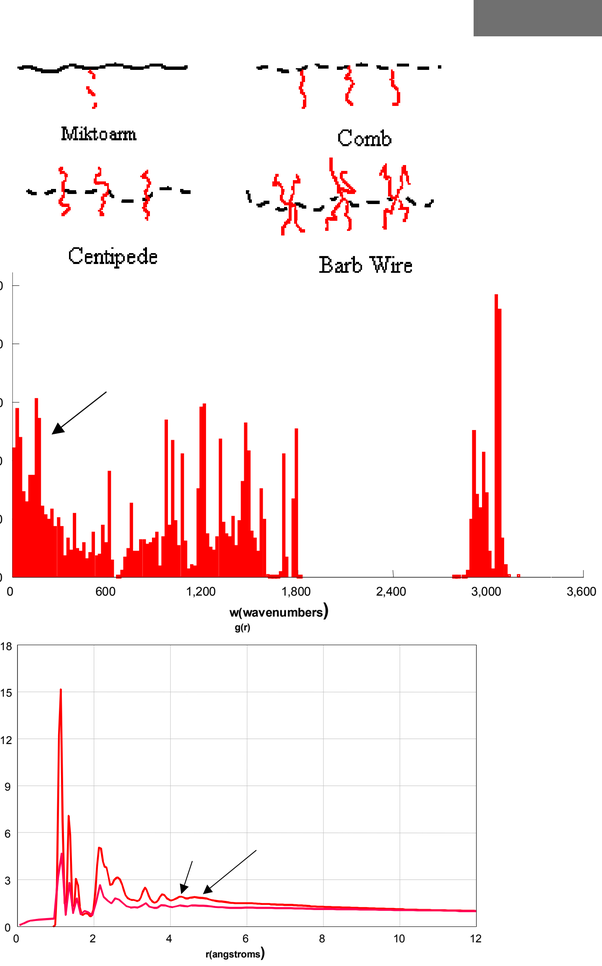


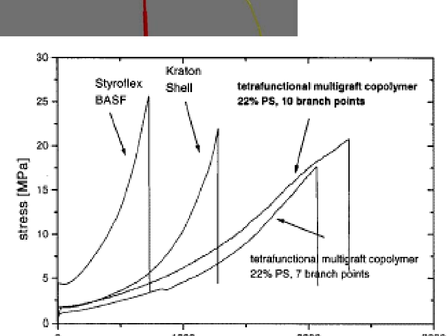


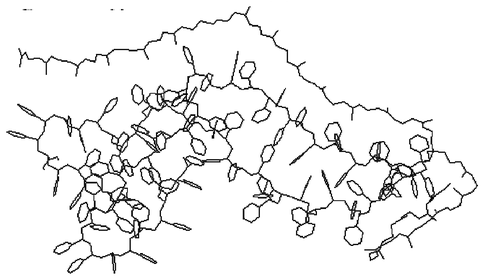
Results

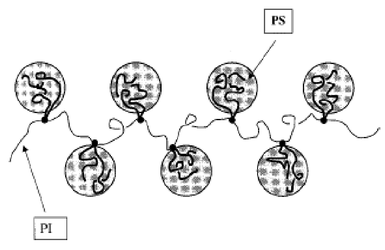
•Optimized photophysical properties by using dilute solution to generate single molecule nanoparticles

•Photostability and spectral bandwidth superior to inorganic quantum dots under ambient conditions!









References: Sumpter, et. al. J. Phys. Chem. B 109, 7671 (2005); Polymer News 29, 302 (2004); Macromolecules 37, 6132 (2004); Nanolett 3, 603 (2003); J. Am. Chem. Soc. 126, 3376 (2004); Polymer 44, 4389 (2002); Macro. Theory Simul. 11, 711(2002); Polymer 44, 3761 (2003); Polymer Sci. B. (in press 2005)