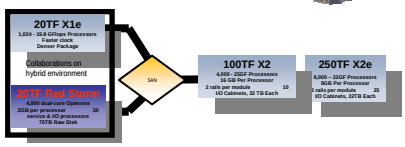
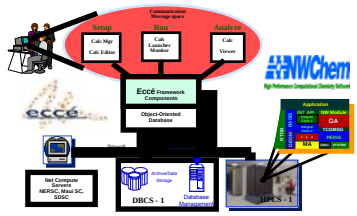
Chemical EndStation (ChemES): The Future of High Performance Computational Chemistry

National Leadership Computing Facility (NLCF)

"The focus of the proposed effort should be on capability computing in support of high-end science – rather than on enhanced computing capacity for general science users . . . ," Orbach

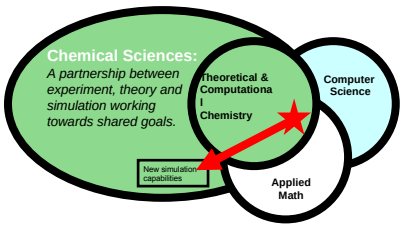


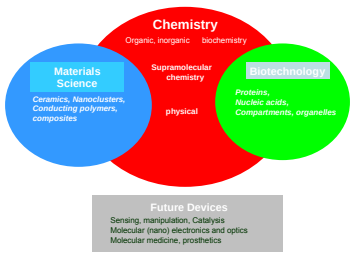
Chemical EndStation



NWChem & ECCE were conceived as an "endstation" for the molecular science computing facility at PNNL/EMSL. ChemES is conceived as being far larger, embracing a greater range of chemistry, and completely open. ChemES will be a community lead project, with NWChem & ECCE as a small subset.

Computational Chemical Sciences: A Multidisciplinary Approach





Vision: A comprehensive resource for chemical science centered in the core BES program, integrating experiment, theory and computation.

- Modern chemistry is inherently multidisciplinary
  - Chemistry must be understood and controlled wherever it occurs
    - e.g., nano, bio, interface, surface, ...
  - The theory, methods and tools are widely applicable
  - Advanced simulation demands novel math and computer science
- Advanced computing at the NLCF
- Close coordination with CNMS

High Performance Computing: Enabling Revolutionary Science

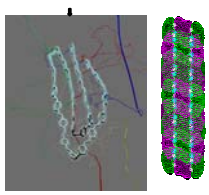
Designing New Materials

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

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

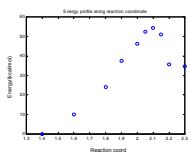
Issues:
- Control of chain organization and alignment
- Optimization of photophysical properties

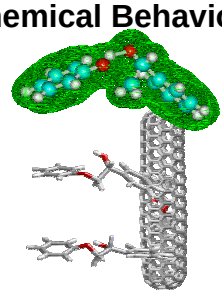
Key Breakthrough:
single molecule nanoparticles generated from dilute solutions of semiconducting polymers – Simulations show how 3-D confinement and solvent can lead to self-organization into the optimal structure!



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!**

Predicting Chemical Behavior





- OH substituent changes reaction rate by 20x in gas phase. Stabilization of the transition state confirmed by computation (MP2 6-31g**)
- Confinement causes large changes in electronic structure and reaction barrier heights (ΔE < 5Kcal/Mole)

Developing New Algorithms

Cray-X1 – definitive electronic structure calculations at 2.3 TFLOP/s on 256 MSP

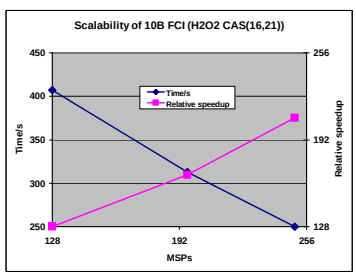
- **Full-configuration interaction *exactly* solves the N-electron problem in a given 1-electron basis**
- **It provides a critical benchmark for understanding and calibrating many-body calculations**
- **Several new parallel-vector algorithms have been developed each of which is optimal for certain parameter values – the new code automatically selects the optimal algorithm**
- **Optimal algorithms for the Cray differ sharply from those for IBM Power4**
- **The new code already enables computations 8x larger and 10x faster than previous work**

Benchmark calculations of Oxygen anion with different Full-CI algorithms

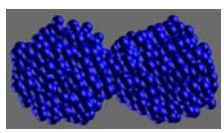
α–α Algorithm	**Time/s**
SAXPY Kernel-1	2240
SAXPY Kernel-2	355
Indexed SAXPY	485
DGEMM Kernel	55
α–β Algorithm	**Time/s**
SAXPY Kernel-2	1250
DGEMM Kernel	125

* FCI (7,79) calculation of Oxygen anion using aug-cc-pvdz basis. 1s electrons are frozen. The FCI involves 14,851,999,576 determinants in D2h symmetry. Timings are measured for one Davidson iteration on 256 MSPs, Cray-X1.

* The DGEMM-based algorithm runs at 9.5GFlops for alpha-beta piece and 8Gflops for alpha-alpha and beta-beta piece per MSP. It takes about 180 seconds (3 minutes) per iteration. The total calculation requires 18 iterations to converge to the required threshold of residual norm 1.D-04.

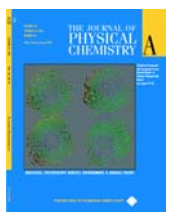


Studying Complex Materials



Recently, chemists at ORNL have successfully constructed well-defined structures from individually manipulated polymer particles.

Using molecular dynamics techniques, we have simulated the construction process and also investigated the physical properties of these systems.



Properties investigated include *normal modes*, *system structure*, and *thermal conductive properties*.