A Short Overview of Computational Nanoscience

Juan Meza
High Performance Computing Research
Lawrence Berkeley National Laboratory

ALS Seminar

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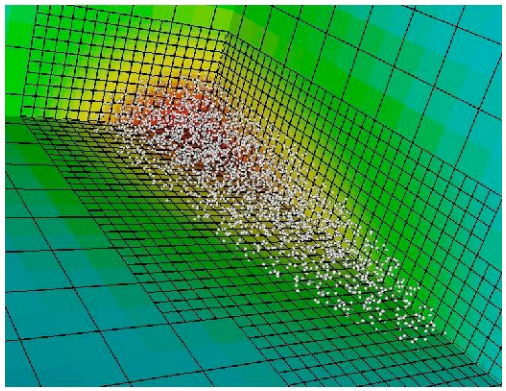
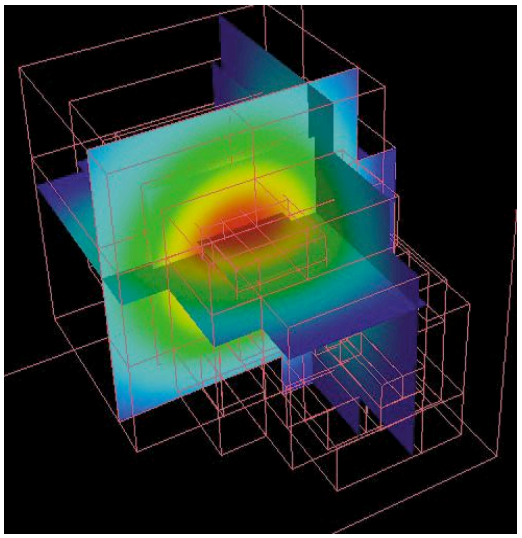
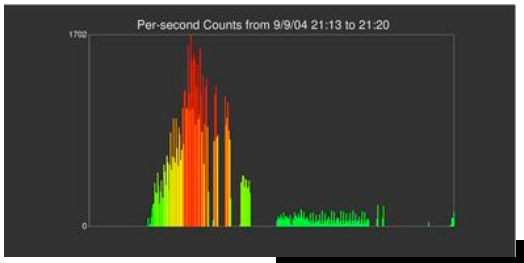
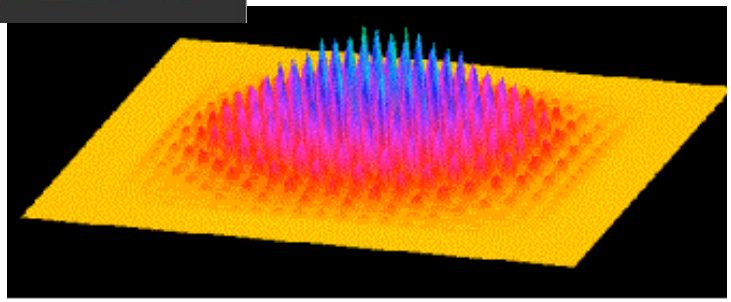
High Performance Computing Research Department









Juan Meza, Department Head

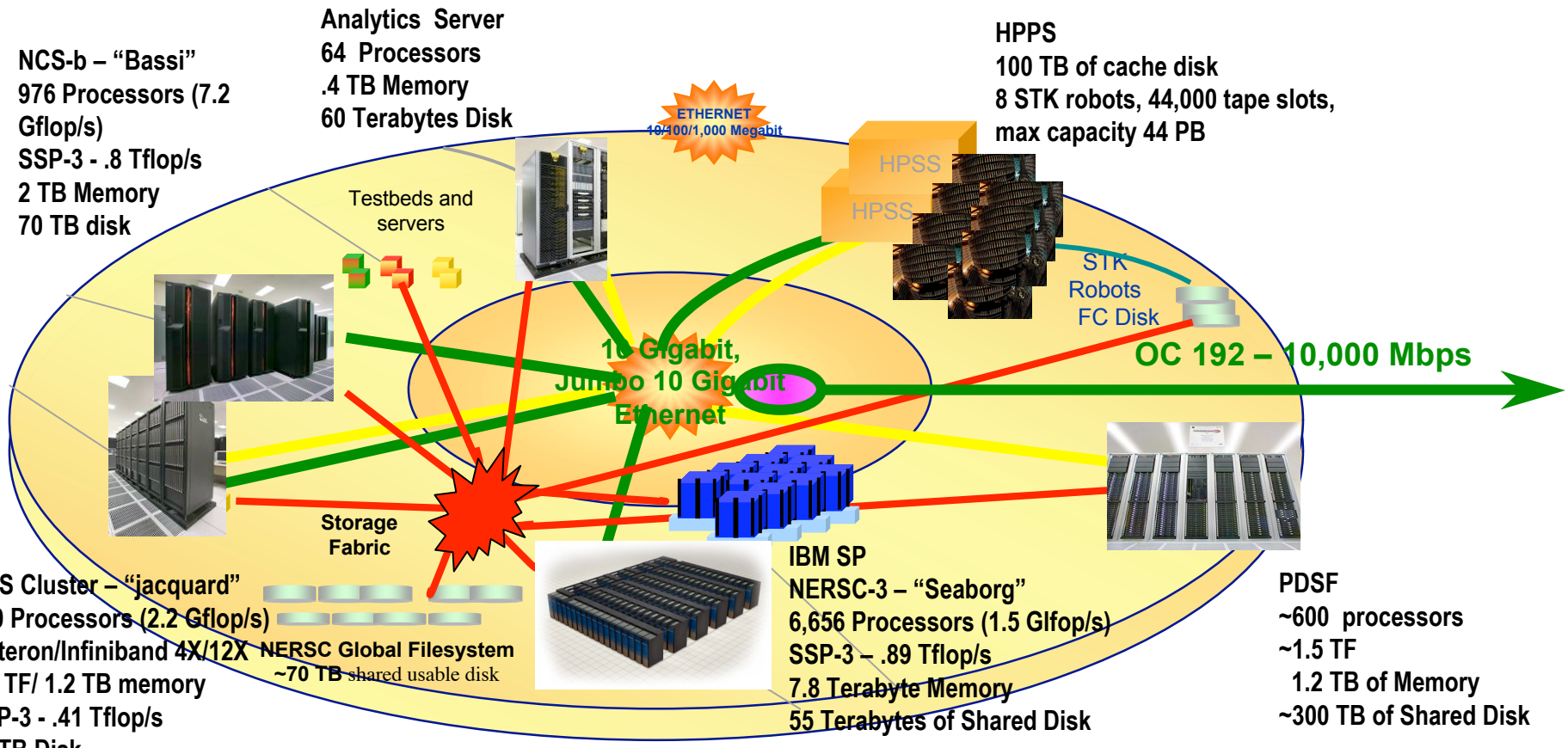
7 Groups:

- Applied Numerical Algorithms
- Center for Computational Sciences and Engineering
- Future Technologies
- Mathematics
- Scientific Computing
- Scientific Data Management
- Visualization

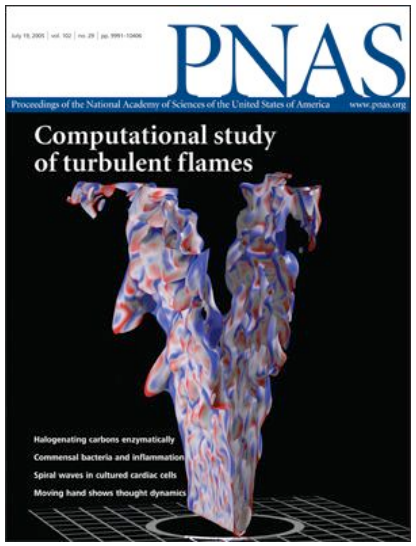
Total Staff: ~150

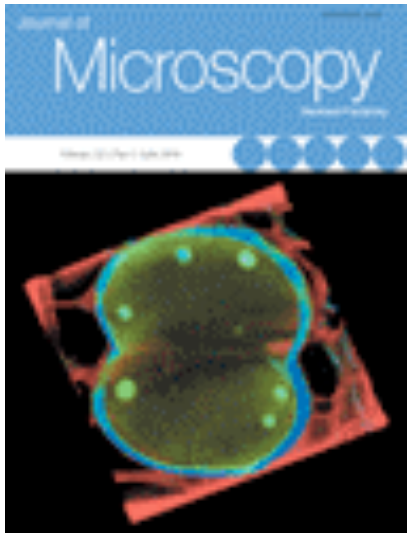
The High Performance Computing Research Department conducts research and development in mathematical modeling, algorithm design, software implementation, and system architectures, and evaluates new and promising computer technologies.

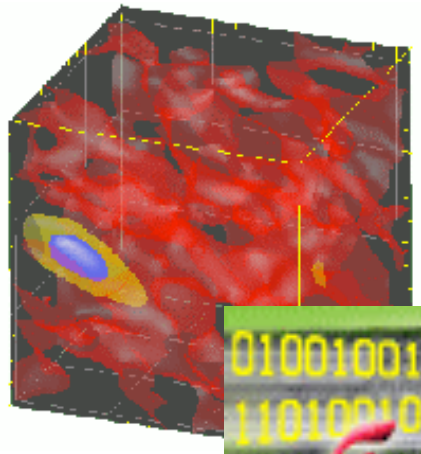
NERSC Systems 2007



Selected Scientific Highlights







- Combustion simulations lead to a better understanding of turbulent flames
- Level-set methods used to improve industrial and medical processes
- New data management techniques improve the efficiency of searching databases
- Improved computational methods for understanding material science properties of nanostructures
- Computer architecture evaluations show promise of new processors for scientific computing

Computational Nanoscience Projects

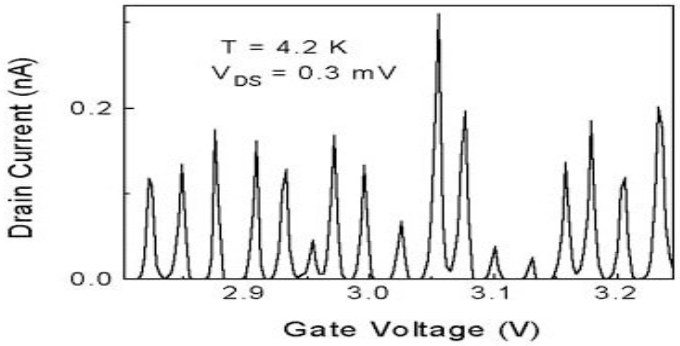
- Materials Science Support for NERSC: PEtot, PARATEC, Escan (ASCR)
- Inorganic-Organic Nanocomposites (BES)
- Computational Materials Science Network (BES)
- Computational Methods for Electronic Structure Codes in Materials Science (LDRD)
- Bonding in Low-Dimensional Structure: Theory and Computation (LDRD)
- Two nanoscience projects jointly funded by ASCR and BES in October 2003
  - Predicting the Electronic Properties of 3D, Million-Atom Semiconductor Nanostructure Architectures
  - Scalable Methods for Electronic Excitations and Optical Responses of Nanostructures: Mathematics to Algorithms to Observables

We're interested in many aspects

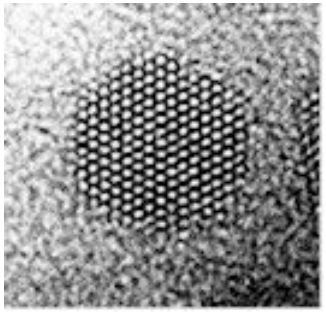
- Band gap increase

CdSe quantum dot (size)

- Single electron effects on transport (Coulomb blockade).

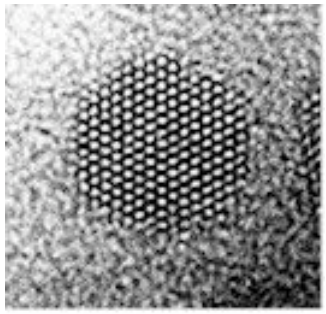


- Mechanical properties, surface effects and no dislocations



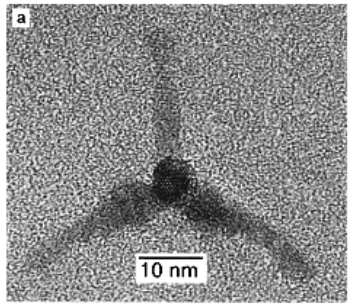
Why are large scale electronic structure calculations required?

- Nanosystems often involve 1,000 to 1,000,000 atoms



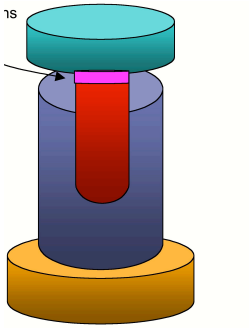
CdSe QD

~1000-5000



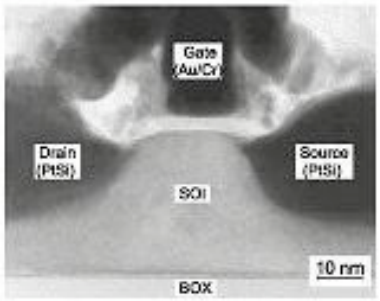
Tetrapod

>10,000



A Solar Cell

>50,000

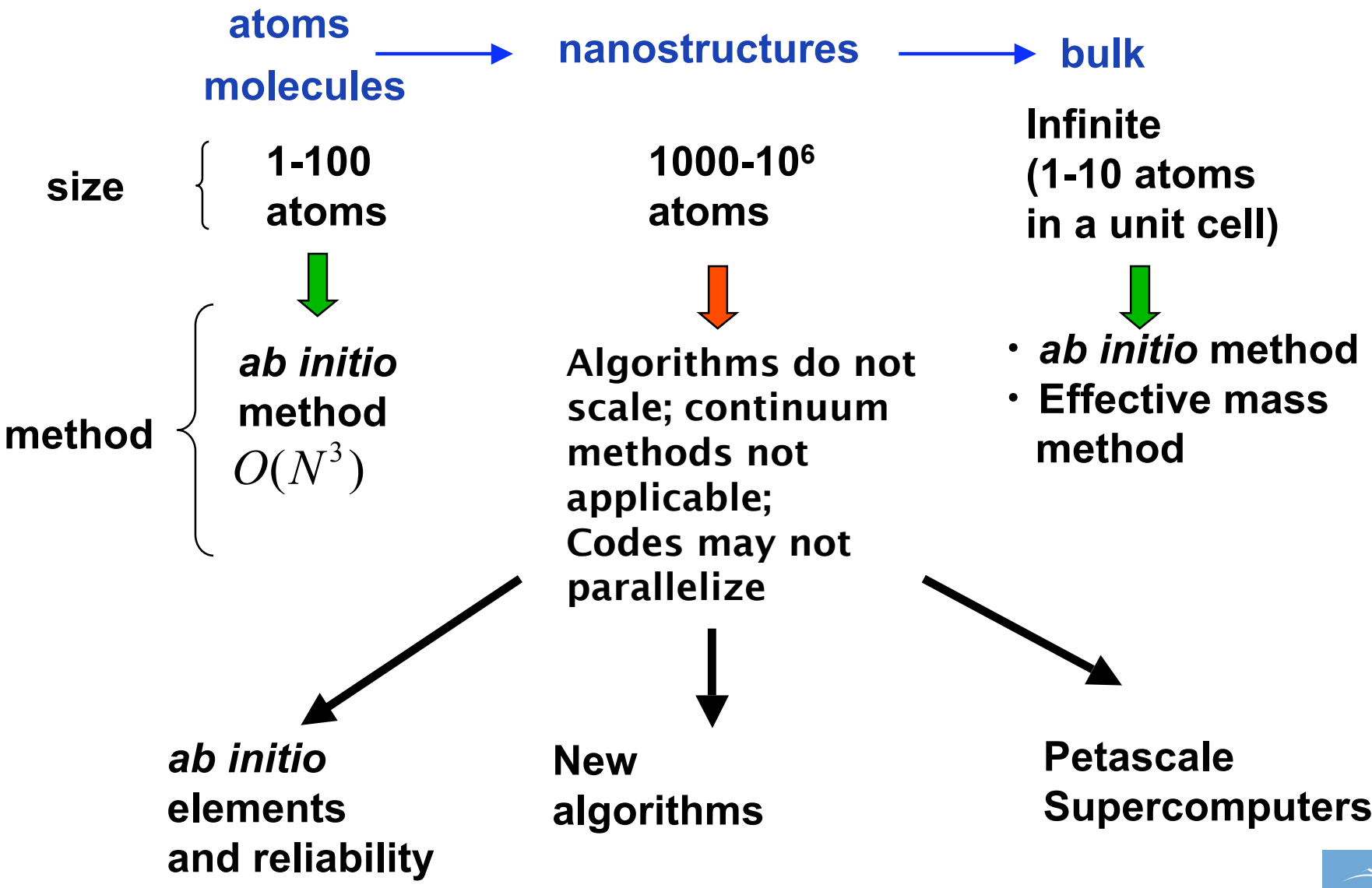


CMOS

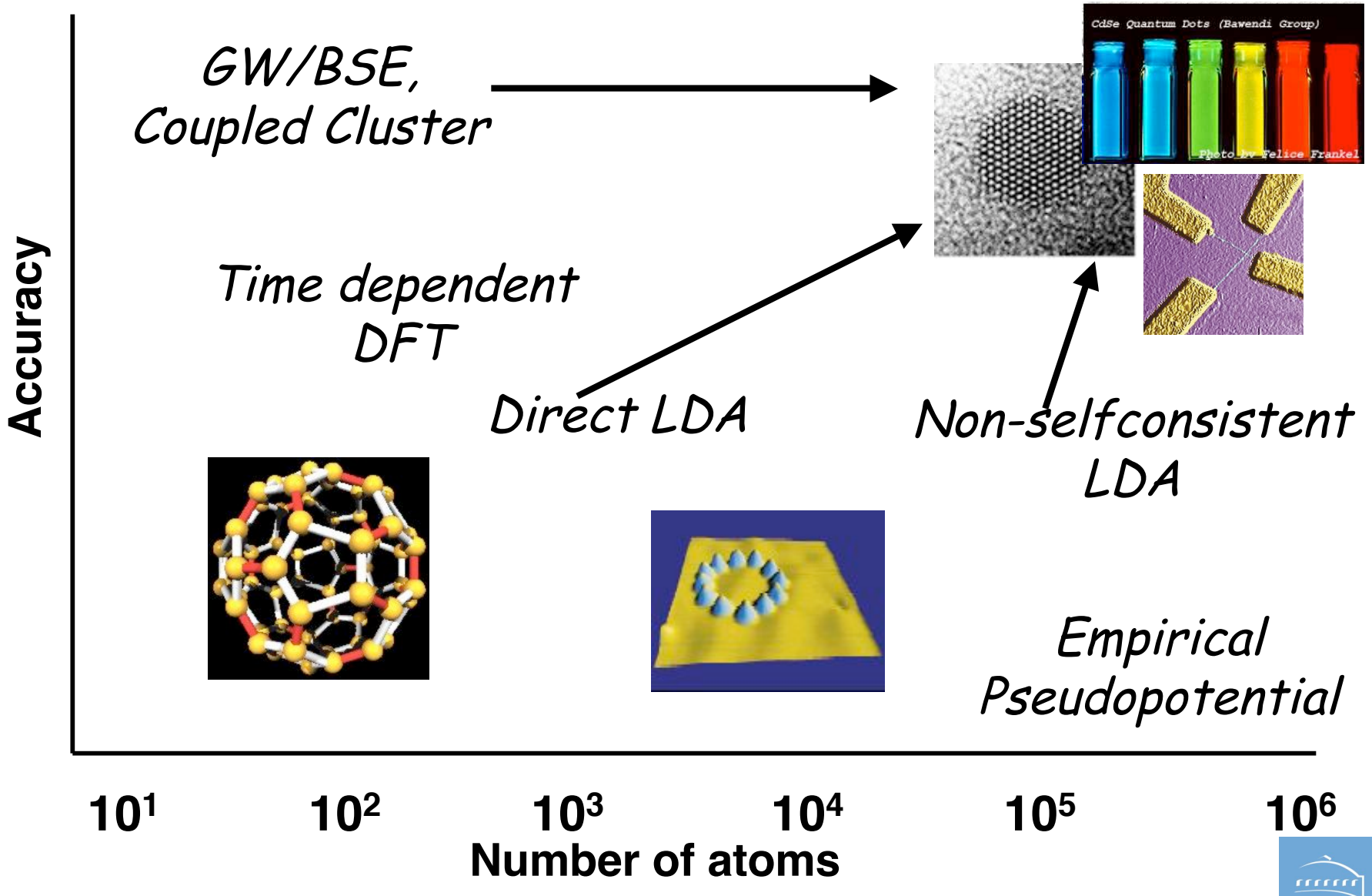
~1,000,000 atoms

- Atomic details are still important:
  - surface charge, impurity, dopant, symmetry, passivation
- Thus, *ab initio* total energy electronic structure calculations are needed.

Challenge for computational nanoscience is three-fold



Applicability versus Accuracy of Current Methods



Brief Overview of Fundamental Equations

Many-body Schrodinger's Equation

$$H\psi\left(r_1, r_2 \ldots r_n\right) = E\psi\left(r_1, r_2 \ldots r_n\right)$$

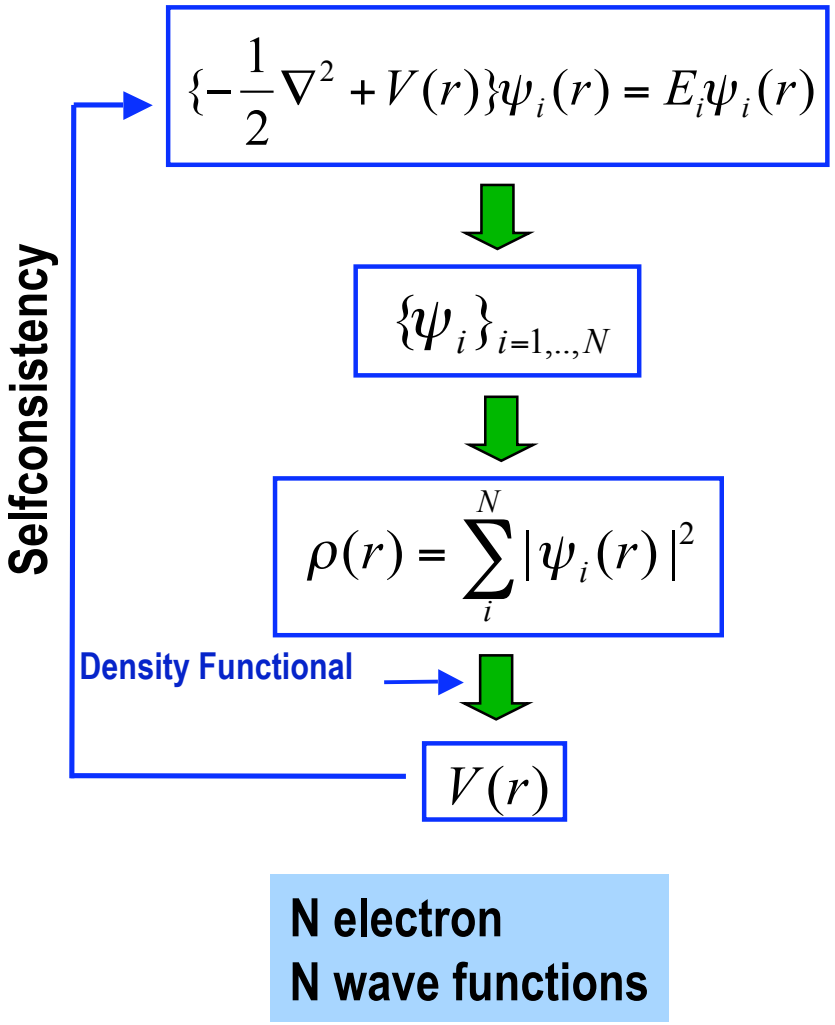
ψ Contains all the information of the system. Satisfies special symmetry of particles.

$|\psi|^2$ Probability density of finding the system at a certain state.

E Quantized energy of the system.

Density functional theory and local density approximation (LDA)

Solve Kohn-Sham equation:



$$E_{tot} = \sum_{i=1,M}\int \psi_i^*(r)[-\frac{1}{2}\nabla^2]\psi_i(r)dr + \int V_{ion}(r)\rho_{tot}(r)dr +$$

$$\frac{1}{2}\int \frac{\rho_{tot}(r)\rho_{tot}(r')}{|r-r'|}drdr' + \int \varepsilon_{xc}(\rho_{tot}(r))dr,$$

$$\rho_{tot}(r) = \sum_{i=1,M}|\psi_i(r)|^2 .$$

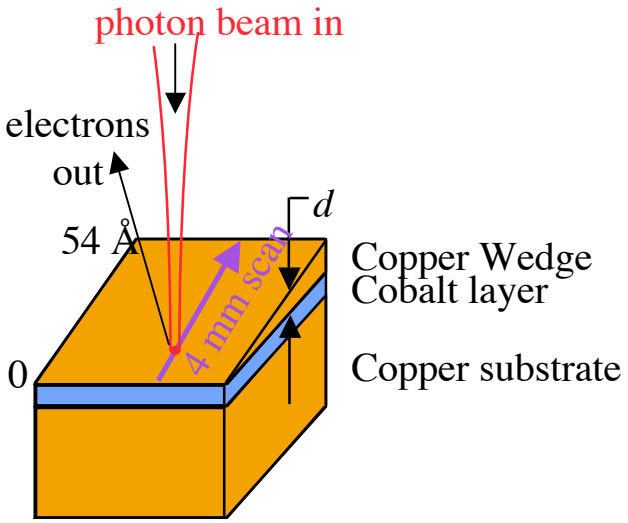
Where $\int \psi_i^*(r)\psi_j(r)d^3r = \delta_{i,j}$

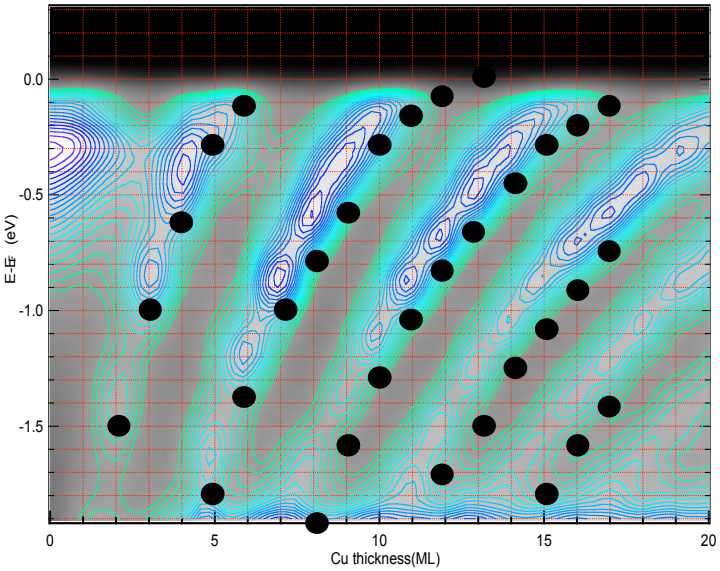
- **Overall Complexity *O(N³)***
- **Major computational work (for plane wave codes):**
  - **3D FFT**
  - **Orthogonalization**
  - **Nonlocal potential**

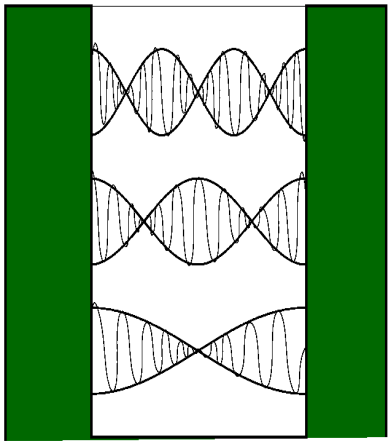
Project Highlights

Low Dimensional Structure Case Study:The Quantization Condition of Quantum-well States in Cu/Co(100)

- Theoretical investigation of Quantum Well states in Cu films using our codes (PARATEC, PEtot) to compare with experiments at the ALS (E. Rotenberg, Y.Z. Wu, Z.Q. Qiu)
- New computational methods for metallic systems used in the calculations.
- Led to an understanding of surface effects on the Quantum Well States. Improves on simple Phase Accumulation Model used previously







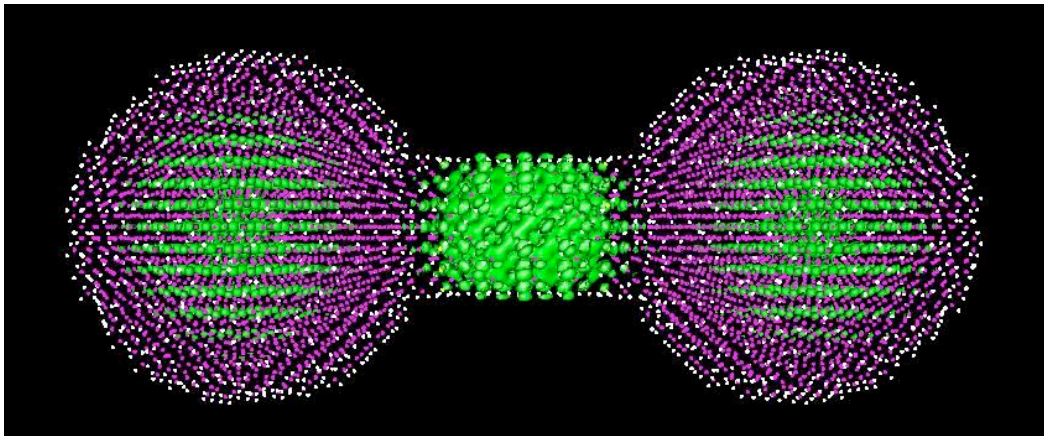
QW states in Copper Wedge

Difference between theory and experiment improved by taking surface effects into account

Predicting the Electronic Properties of 3D, Million-Atom Semiconductor Nanostructure Architectures

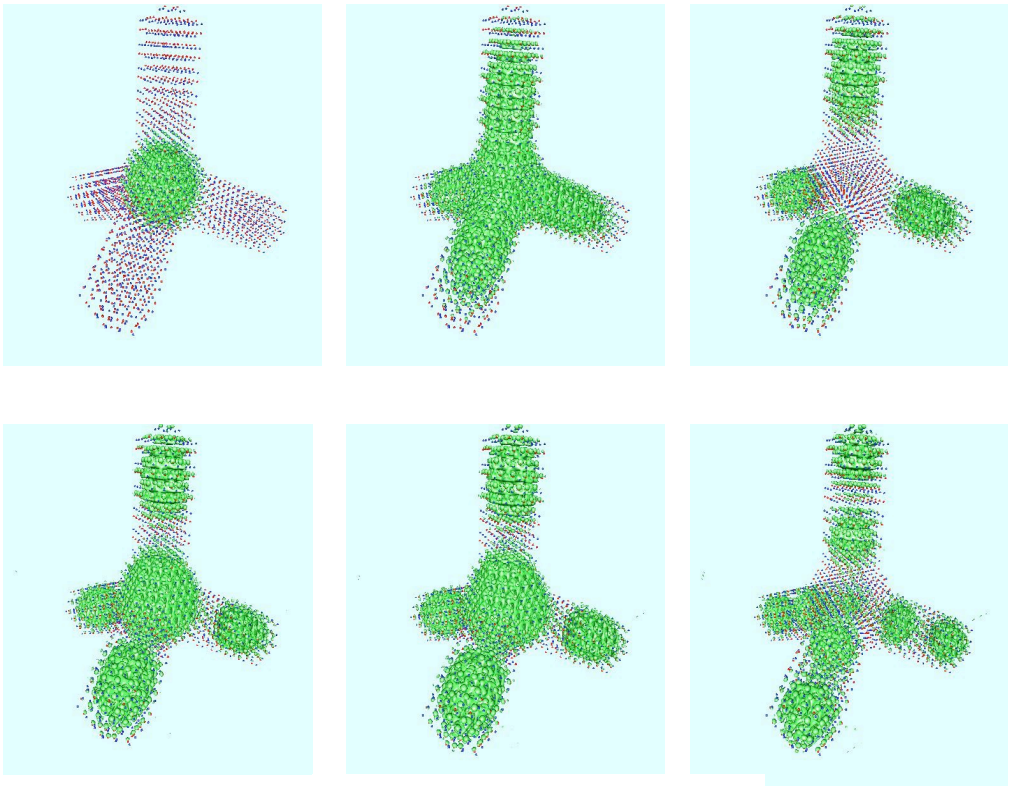
LBNL investigators: L.W. Wang (CRD), A. Canning (CRD), O. Marques (CRD)

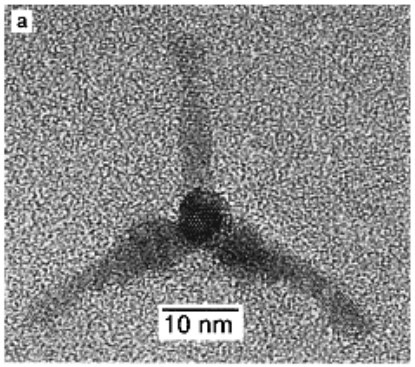
- Developed the charge patching method, which makes thousand atom ab-initio calculations possible.
- Developed a way to treat the polarizability of the charge motif in the charge patching method, thereby allowing a selfconsistent treatment for long range electric fields and their charge response.
- Developed and tested new mathematical algorithms to improve the efficiency of the current codes (e.g, Escan) used in 1000 atom electronic structure calculations.
- Developed a new algorithm for electronic quantum transport, which can be used to study larger systems (a few hundred atoms) than previously capable.



The electronic state of a 6000 atom dumbbell consisting of CdTe/CdSe/CdTe. This state was calculated with the charge patching method and the Escan code, both developed at LBNL.

CdSe tetrapod conduction band wavefunctions





- First 6 electron wavefunctions of a CdSe tetrapod quantum structure
- System contains ~1000 atoms
- Solution of large eigenvalue problem is required

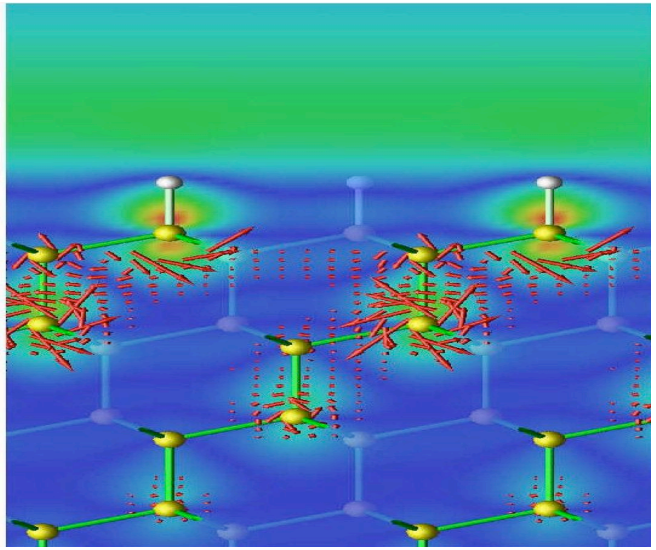
Andrew Canning, Lin-Wang Wang,
Scientific Computing, LBNL

Computational Methods for Electronic Structure Codes in Materials Science

Investigators: A.Canning (CRD), S.Louie (MSD), L-W Wang (Collaborator) + 2 postdocs

- New eigensolver based on Grassman conjugate gradient approach incorporated into PARATEC DFT-based electronic structure code (more stable and faster then previous solver).
- New charge mixing scheme (Thomas-Fermi) programmed into PARATEC giving large improvement for inhomogeneous systems.
- Calculation of forces for excitons (electron-hole states) in the context of the GW+BS approach used by Louie for excited states.
- Codes used by other groups at LBNL eg. to study Quantum Well states in layered Cu/Co/Cu systems with Prof Qiu's group (MSD) (Thomas-Fermi mixing approach vital to converge these difficult systems)
- **Implementation of above approaches in efficient parallel codes used by many different groups, code PARATEC distributed on web site.**

PARATEC (PARAllel Total Energy Code)



- PARATEC performs first-principles quantum mechanical total energy calculation using pseudopotentials & plane wave basis set
- Written in F90 and MPI
- Designed to run on large parallel machines IBM SP etc. but also runs on PCs

- PARATEC uses all-band CG approach to obtain wavefunctions of electrons
- Generally obtains high percentage of peak on different platforms
- Developed with Louie and Cohen's groups (UCB, LBNL), Raczkowski (Multiple 3d FFTs Peter Haynes and Michel Cote)

PARATEC: Performance

Problem	P	NERSC (Power3)		Jacquard (Opteron)		Thunder (Itanium2)		Phoenix (X1)		NEC ES (SX6*)		NEC SX8	
		Gflops/P	%peak	Gflops/P	%peak	Gflops/P	%peak	Gflops/P	%peak	Gflops/P	%peak	Gflops/P	%peak
488 Atom CdSe Quantum Dot	128	0.93	62%			2.8	51%	3.2	25%	5.1	64%	7.5	64%
	256	0.85	67%	1.98	45%	2.6	47%	3.0	24%	5.0	62%	6.8	62%
	512	0.73	49%	0.95	21%	2.4	44%			4.4	55%		
	1024	0.60	40%			1.8	32%			3.6	46%		

- All architectures generally achieve high performance due to computational intensity of code (BLAS3, FFT)
- ES achieves highest overall performance to date: 5.5Tflop/s on 2048 procs
  - Main ES advantage for this code is fast interconnect
- SX8 achieves highest per-processor performance
- X1 shows lowest % of peak
  - Non-vectorizable code much more expensive on X1

Developed with Louie and Cohen's groups (UCB, LBNL), also work with L. Oliker, J Carter

Pattern Search Methods for Structure Determination

(joint work with Michel van Hove (ALS))

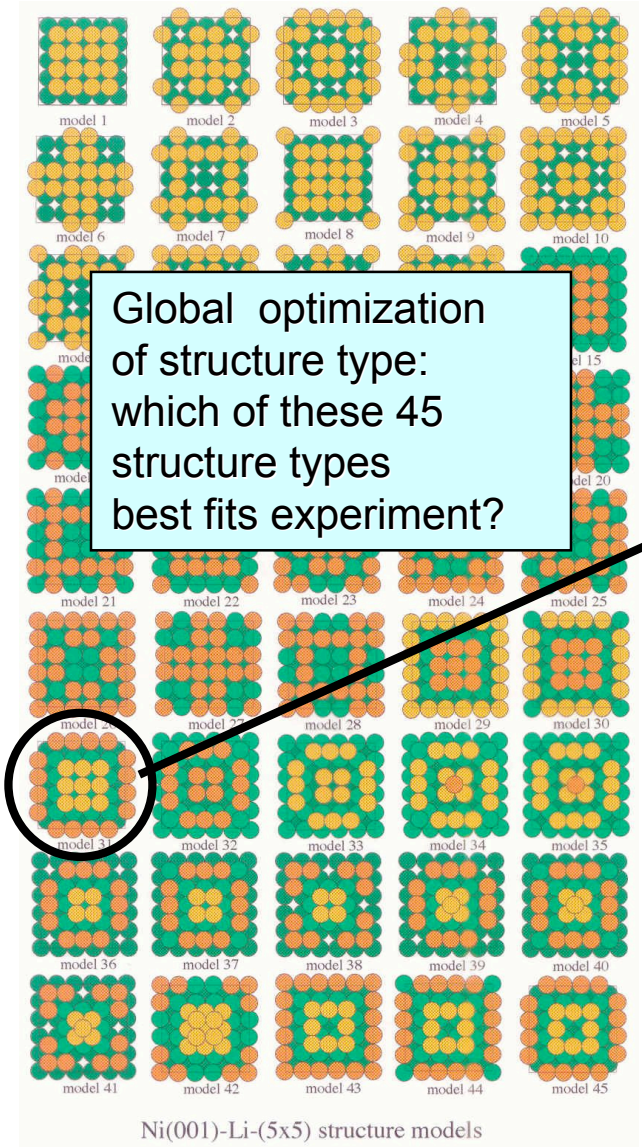
Scalable Methods for Electronic Excitations and Optical Responses of Nanostructures: Mathematics to Algorithms to Observables

- Initiate a program on the theory and modeling of the electronic excited-state and optical properties of various nanoscience structure
- Address existing bottlenecks in simulating excitations and optical responses of nanostructure
- Seek novel reformulations of the underlying physical theories by exploring new ideas in applied mathematics
- Apply the methodology to targeted problems in nanosciences

Participants:

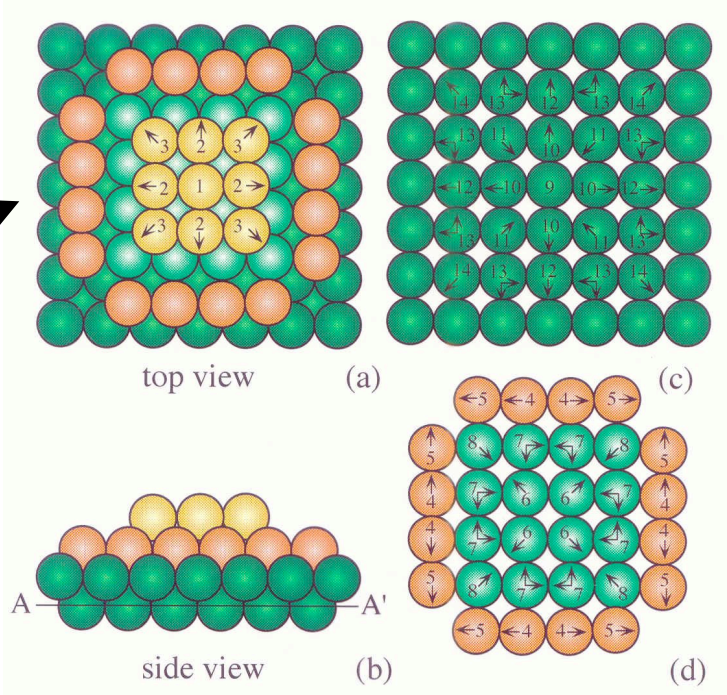
BES (Lab):	Martin Head-Gordon, Steven Louie, Michel van Hove, Lin-Wang Wang
BES (University):	Emily Carter(Princeton), James Chelikowsky (UMN)
ASCR (Lab):	John Bell, Andrew Canning, Byounghak Lee, Juan Meza, Chuck Rendleman, Chao Yang, Zhengji Zhao
ASCR (University):	John Dennis (Rice University), Yousef Saad (UMN)

Surface structure determination from experiment



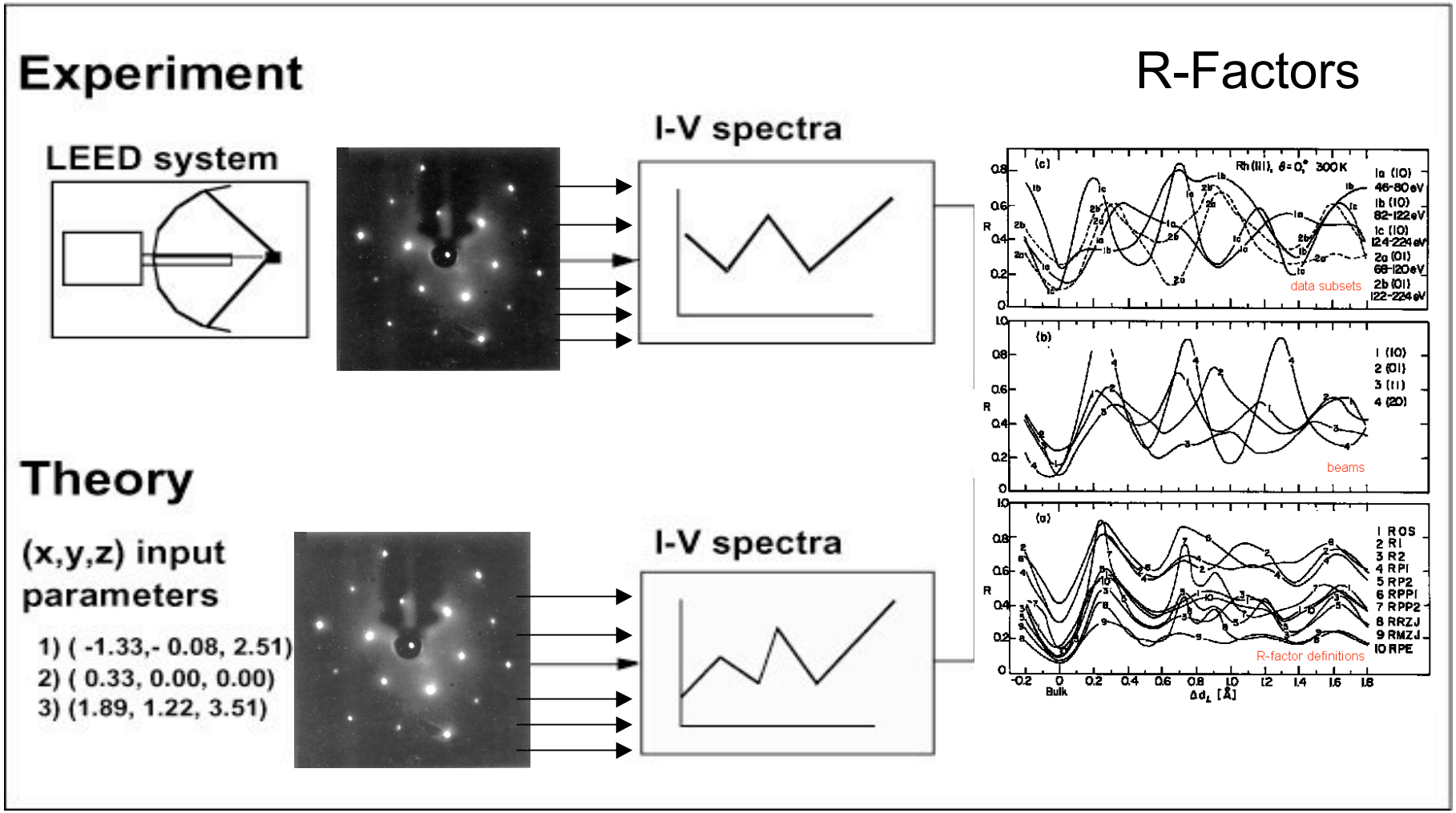
Global optimization of structure type: which of these 45 structure types best fits experiment?

- Electron diffraction determination of atomic positions in a surface:
  - Li atoms on a Ni surface

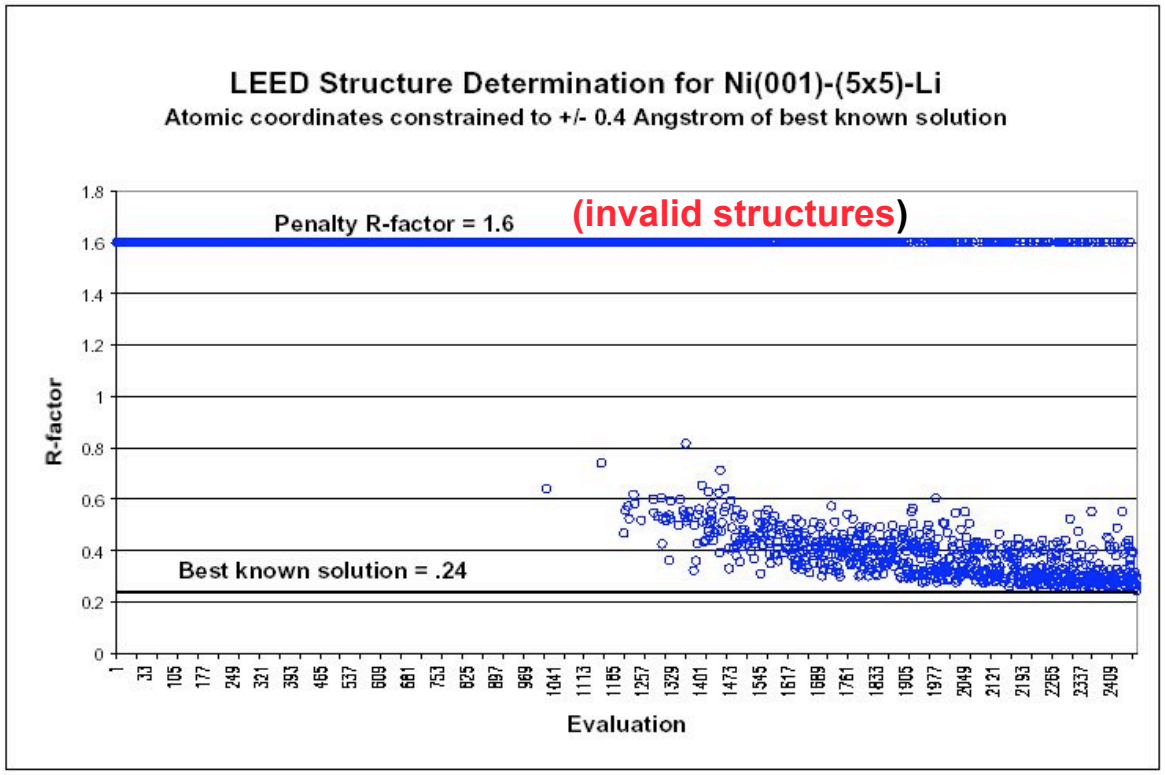


Local optimization of structure parameters: which are the best interatomic distances and angles?

Low Energy Electron Diffraction



Previous Work



- Previous work used genetic algorithms to solve the optimization method.
- Large number of invalid structures generated (more on this later).
- Overall, a solution was found - *after adding sufficient constraints*.

1. *Global Optimization in LEED Structure Determination Using Genetic Algorithms,* **R. Döll and M.A. Van Hove**, Surf. Sci. **355**, L393-8 (1996).
2. *A Scalable Genetic Algorithm Package for Global Optimization Problems with Expensive Objective Functions,* **G. S. Stone**, M.S. dissertation, Computer Science Dept., San Francisco State University, 1998.

Generalized Pattern Search Framework

1. Initialization: Given Δ_0 , x_0 , M_0, P_0
2. For k = 0, 1, …

   a) SEARCH: Evaluate f on a finite subset of trial points on the mesh M_k

   b) POLL: Evaluate f on the frame P_k

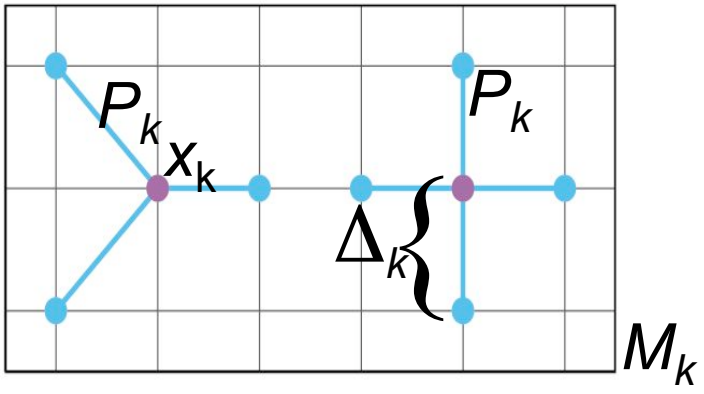
   Global phase can include user heuristics or surrogate functions

   Local phase more rigid, but necessary to ensure convergence

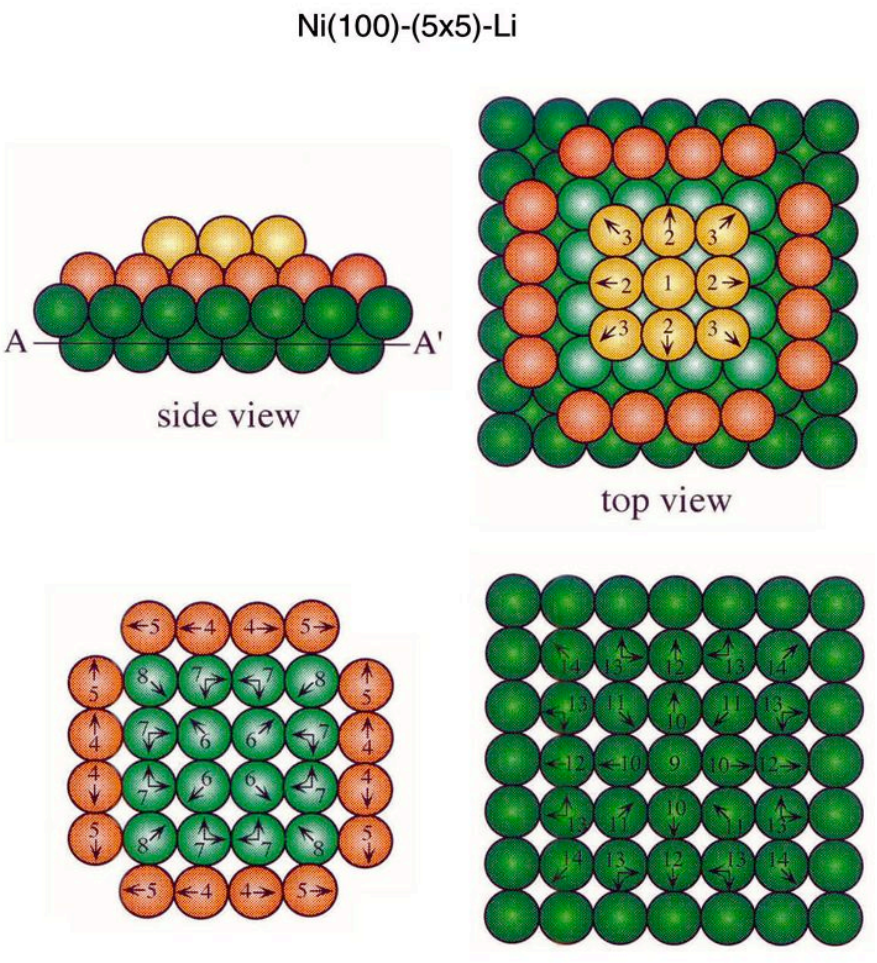
3. If successful - mesh expansion:

   a) $x_{k+1} = x_k + \Delta_k d_k$

4. Otherwise contract mesh



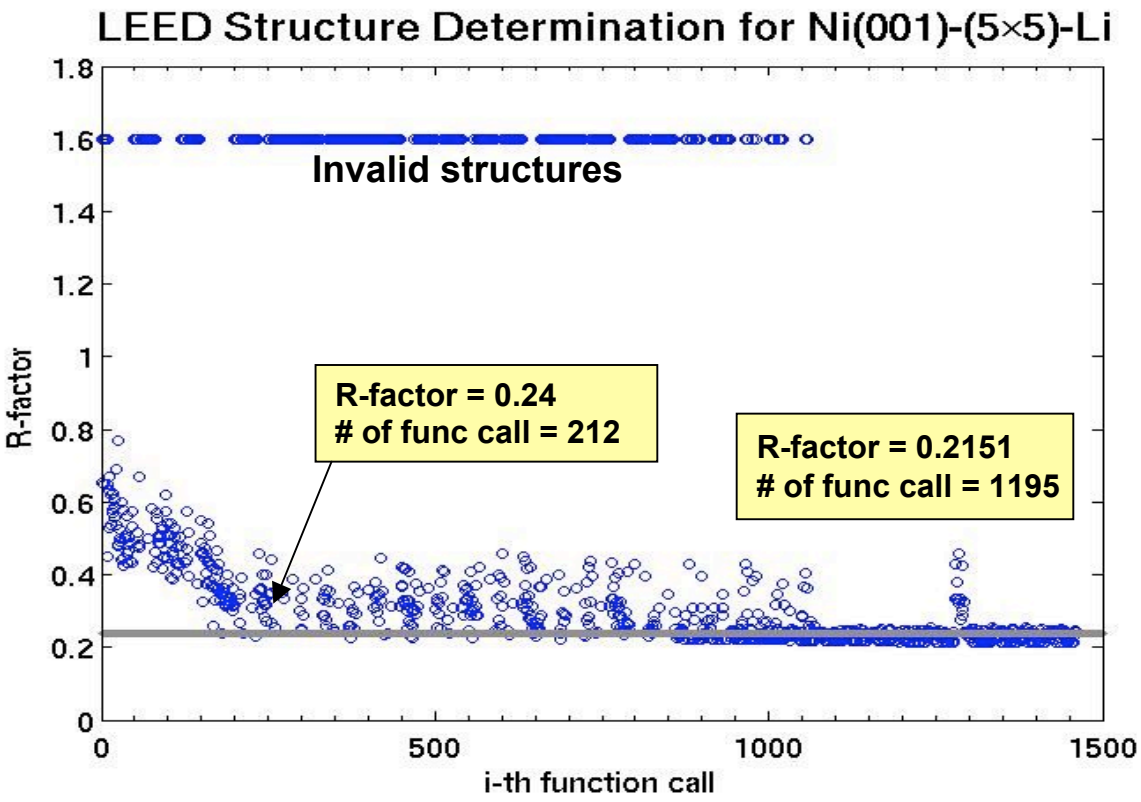
Test problem

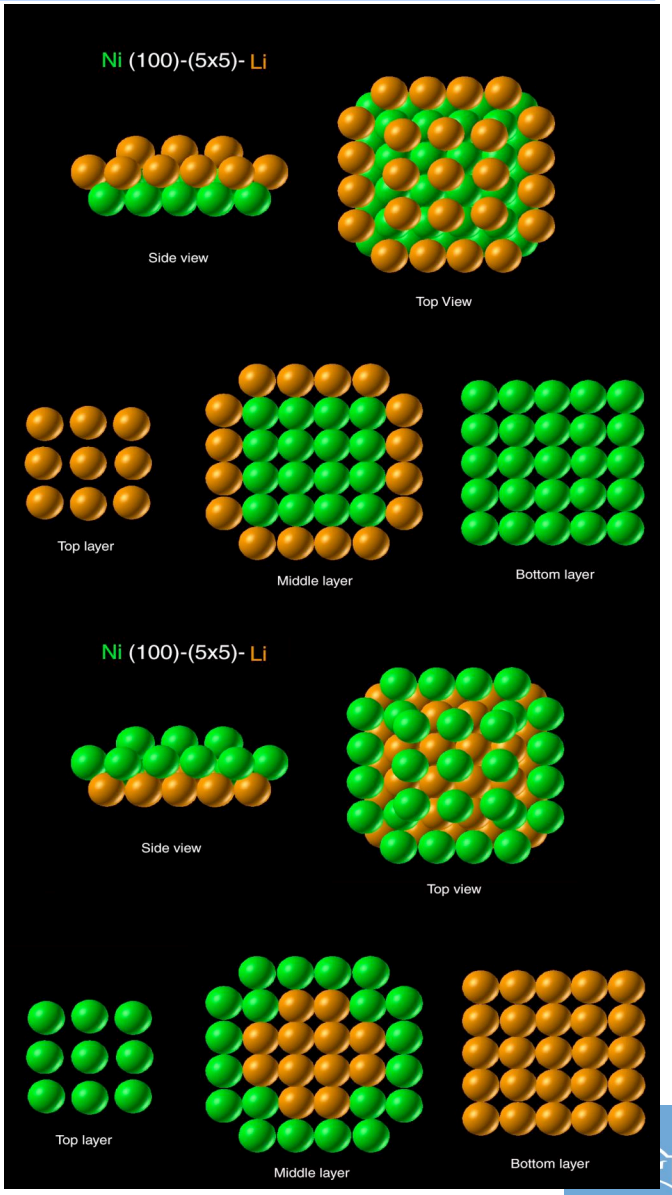


Model 31 from set of TLEED model problems

- Model contains three layers of atoms
- Using symmetry considerations we can reduce the problem to 14 atoms
  - 14 categorical variables
  - 42 continuous variables
- Positions of atoms constrained to lie within a box
- Best known previous solution had R-factor = .24

Minimization with respect to both types of variables removes coordinate constraints



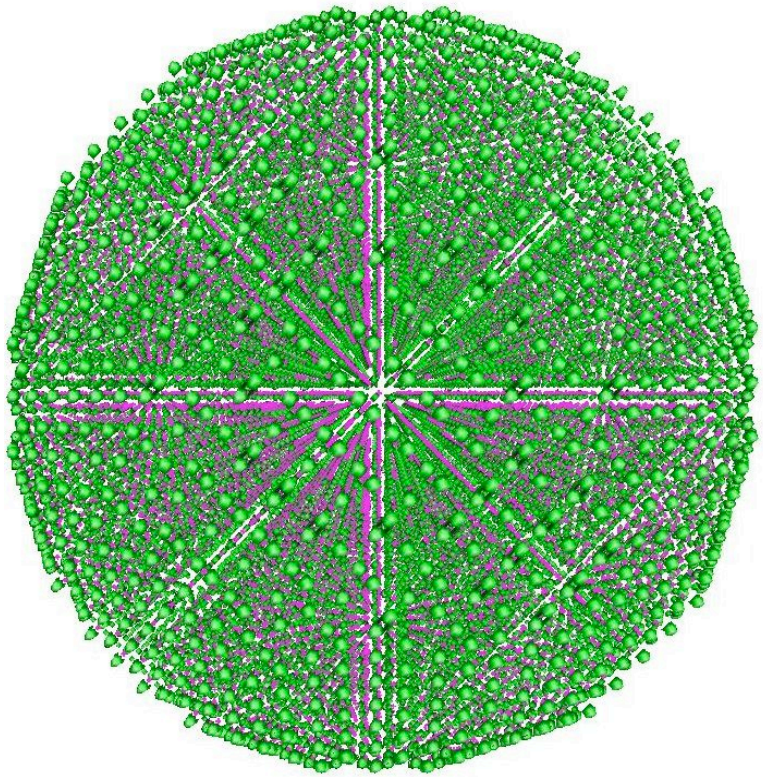


Previous best known solution R-factor = 0.24
New solution found with R-factor = .2151
Final (global) solution with R-factor = .1184

New Linear Scaling Methods

Beyond 10000 atoms

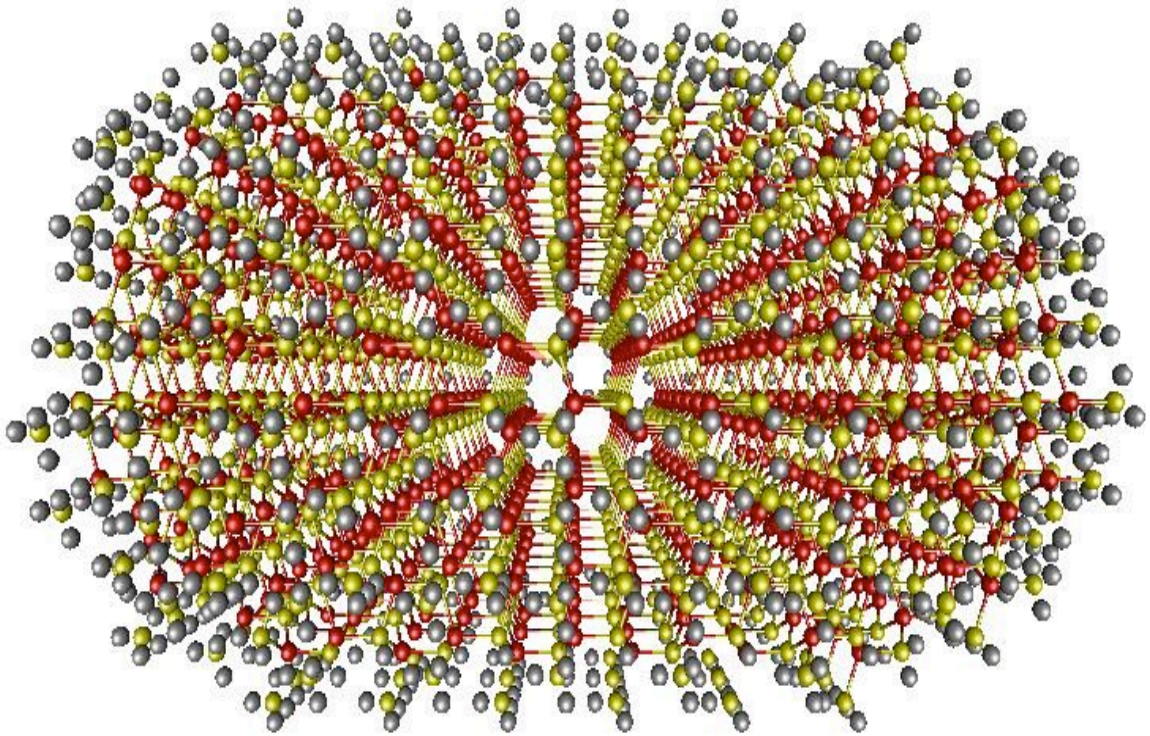
Linear Scaling 3D Fragment (LS3DF) method



- Uses a novel divide and conquer approach to solve DFT
- Scales linearly with the number of atoms and has excellent parallel scaling
- Numerically equivalent to LDA
  - The total energy difference is 3meV/atom ~ 0.1 kcal/mol
  - Charge density difference: 0.2%
  - Atomic force difference: 10^{-5} a.u

The charge density of a 15,000 atom quantum dot, $Si_{13607}H_{2236}$. Using 2048 processors at NERSC the calculation took about 5 hours, while a direct LDA calculation would have taken a few months.

Dipole Moment calculation



- The calculated dipole moment of a 2633 atom CdSe quantum rod, $Cd_{961}Se_{724}H_{948}$.
- Using 2560 processors at NERSC the calculation took about 30 hours.

Summary

- Several new algorithms and approaches have been developed to address some common problems in computational nanoscience.
- New charge patching method developed which makes thousand-atom ab-initio calculations possible.
- Development of a linearly scalable DFT code for many thousands of atoms calculations.
- New robust optimization methods for determination of atomic-scale structure of surfaces from experiments.
- Several codes available for general use, all supported on NERSC machines

Future plans

- Continue improvements to all of the codes, PeTOT, PARATEC, …
- Investigate new algorithms using new optimization methods and adaptive mesh refinement techniques
- Expand applicability of methods to new systems, perhaps biological?
- Interested in new collaborations, especially with experimentalists

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Questions

The New York Times

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The First Nanotechnologists

Ancient stained-glass makers knew that by putting varying, tiny amounts of gold and silver in the glass, they could produce the red and yellow found in stained-glass windows. Similarly, today's scientists and engineers have found that it takes only small amounts of a nanoparticle, precisely placed, to change a material's physical properties.

Gold particles in glass

Size*: 25 nm
Shape: sphere
Color reflected:

100 nanometers =
0.0001 millimeter

Silver particles in glass

Size*: 100 nm
Shape: sphere
Color reflected:

200 nm

Had medieval artists been able to control the size and shape of the nanoparticles, they would have been able to use the two metals to produce other colors. Examples:

Size*: 50 nm
Shape: sphere
Color reflected:

Size*: 40 nm
Shape: sphere
Color reflected:

Size*: 100 nm
Shape: sphere
Color reflected:

Size*: 100 nm
Shape: prism
Color reflected:

Source: Dr. Chad A. Mirkin, Institute of Nanotechnology, Northwestern University

*Approximate