Implementation of Density Functional Theory based Electronic Structure Codes on Advanced Computing Architectures

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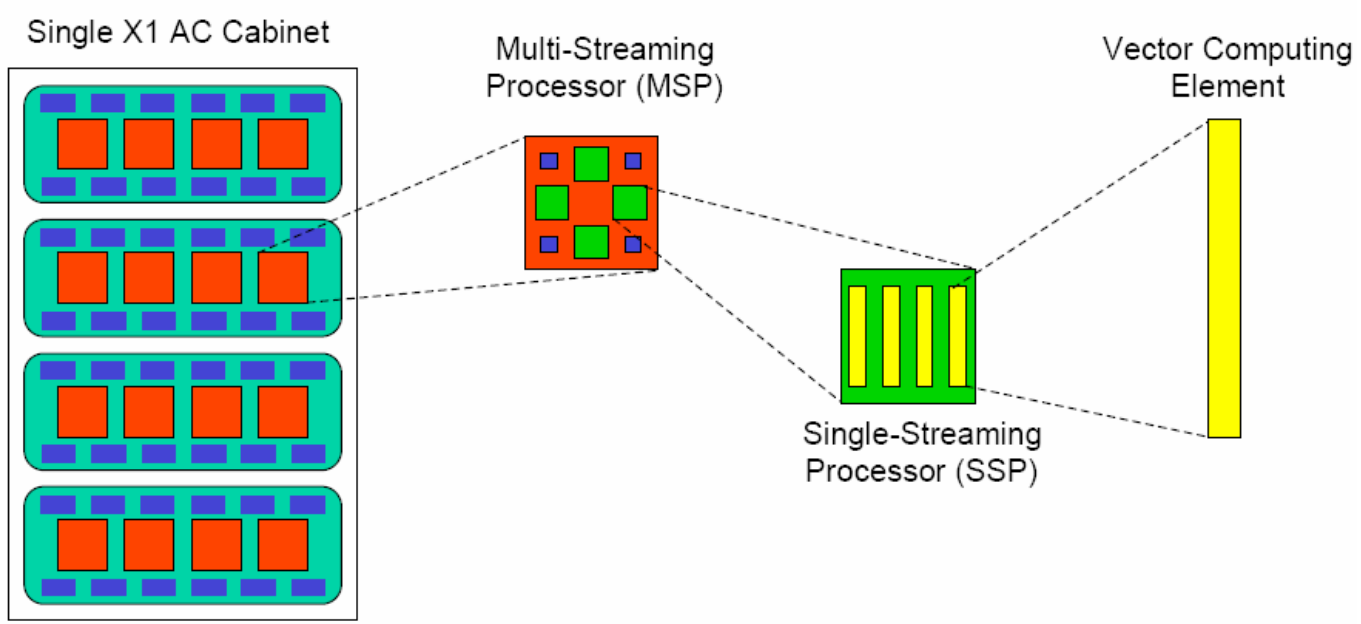
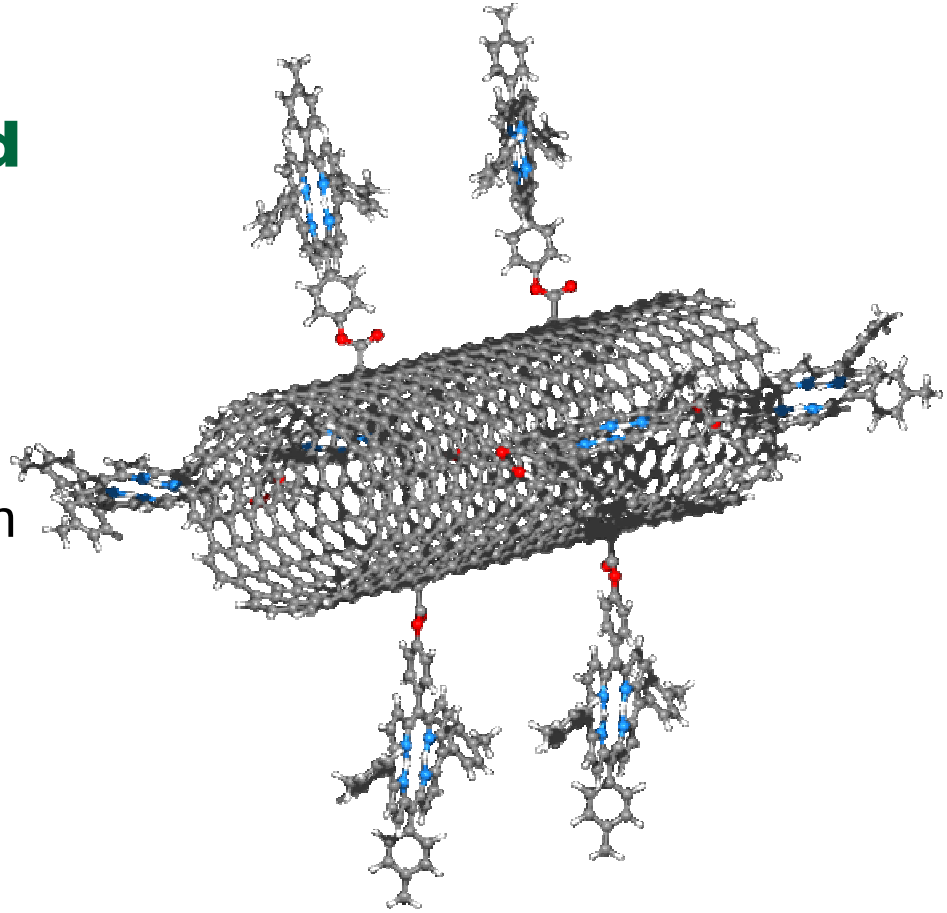
Cray-X1E

- 1024 Multi-streaming vector processor (MSP)
  - Each MSP has 2 MB of cache and a peak computation rate of 12.8 GF
  - 4 single-streaming processors (SSPs) form a node with 16 Gbytes of shared memory
  - Memory is physically distributed on individual modules
  - all memory is directly addressable to and accessible by any MSP in the system through the use of load and store instructions

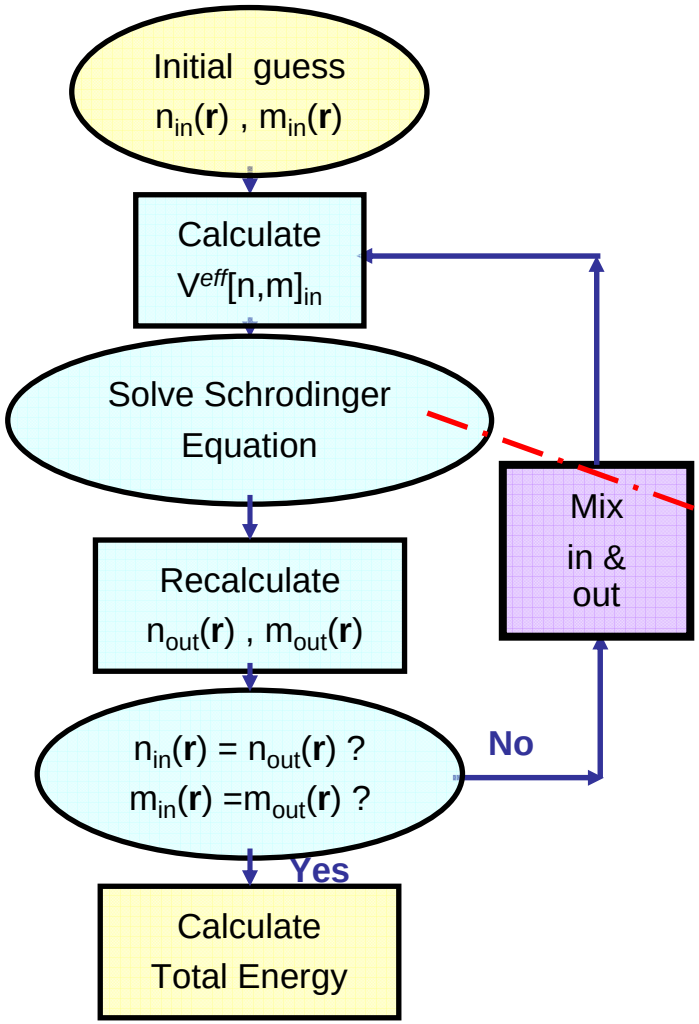


Applications

- **Porphyrin Functionalized Nanotube**
- 1532 atoms
- STO-3G basis set = 6380 basis functions
- Do the covalently attached porphyrins undergo facile absorption of visible light and transfer electrons to the nanotube
- What type of efficiencies does one obtain
- Time for LDA energy + gradient using 300 processors = less than 1 hour



LSDA &Multiple Scattering Theory (MST)



- Multiple Scattering Theory (MST)
  J. Korringa, Physica 13, 392, (1947)
  W. Kohn, N. Rostoker, PR, 94, 1111,(1954)
- MST Green function methods
  B. Gyorffy, and M. J. Stott, "Band Structure Spectroscopy of Metals and Alloys", Ed. D.J. Fabian and L. M. Watson (Academic 1972)
  S.J. Faulkner and G.M. Stocks, PR B 21, 3222, (1980)

$$\{(\varepsilon + \frac{\hbar}{2m}\nabla^2)\hat{1} - \hat{V}^{eff}\}\hat{G}(\mathbf{r},\mathbf{r}';\varepsilon) = \hat{1}\delta(\mathbf{r}-\mathbf{r}')$$

$$n(\mathbf{r}) = \mathrm{Im}\frac{1}{\pi}\int d\varepsilon\, f(\varepsilon-\mu)\ \mathrm{Tr}\hat{G}(\mathbf{r},\mathbf{r}';\varepsilon)$$

$$\mathbf{m}(\mathbf{r}) = \mathrm{Im}\frac{1}{\pi}\int d\varepsilon\, f(\varepsilon-\mu)\ \tilde{\sigma}\mathrm{Tr}\hat{G}(\mathbf{r},\mathbf{r}';\varepsilon)$$

Algorithm Design for future generation architectures

- More accurate
  - Spectral or pseudo-spectral accuracy
- Wider range of applicability
- Sparse representation
  - Memory requirements grow linearly
  - Each processor can treat thousands of atoms
- Make use of large number of processors
- Message-Passing
  - Each atom/node local message-passing is independent of the size of the system
- Time consuming step of model
  - Sparse linear solver
  - Direct or preconditioned iterative approach

Multiple Scattering Theory

- Multiple scattering theory
  - Green function

$$G^{nm}(\vec{r},\vec{r}';\varepsilon)=\sum_{LL'}Z_L^n(\vec{r}_n;\varepsilon)\tau_{LL'}^{nm}(\varepsilon)Z_{L'}^m(\vec{r}_m';\varepsilon)-Z_L^n(\vec{r}_<;\varepsilon)J_{L'}^m(\vec{r}_>';\varepsilon)\delta_{LL'}$$

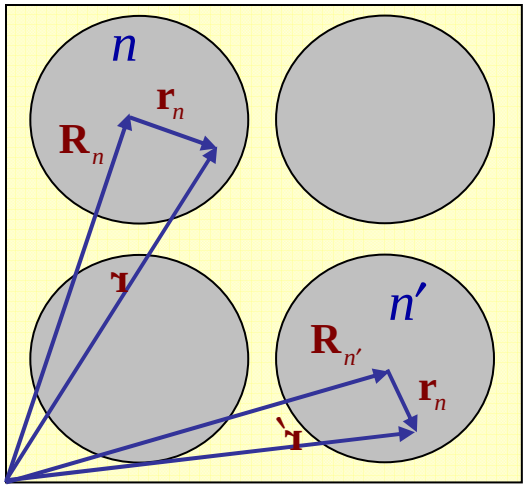
  - Scattering path matrix

$$\tau^{nm}(\varepsilon)=t_n(\varepsilon)\delta_{nm}+\sum_{n\neq m}t_n(\varepsilon)G(\vec{R}_n-\vec{R}_m)\tau^{nm}(\varepsilon)$$

$$\tau^{nm}(\varepsilon)=(M^{nm}(\varepsilon))^{-1}$$

$$M_{LL'}^{nm}(\varepsilon)=t_{LL'}^{-1}\delta_{LL'}\delta_{nm}-G_{LL'}(\vec{R}_n-\vec{R}_m)$$

Generalization of t-matrix. Converts incoming wave at site n into outgoing wave at site m in the presence of all the other sites

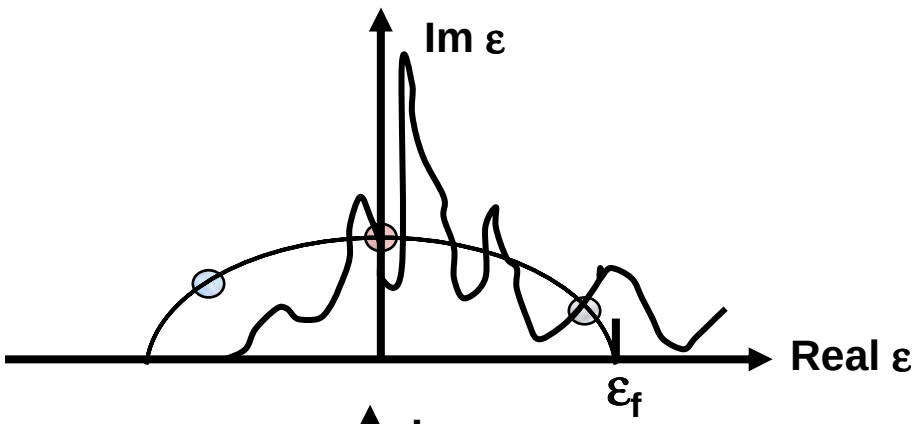


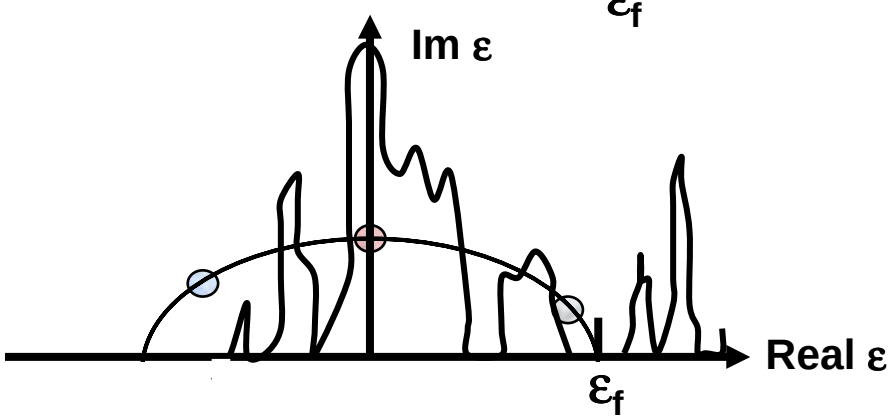
$$M(\varepsilon)=\begin{bmatrix} t_0^{-1} & -G^{01}(\varepsilon) & -G^{02}(\varepsilon) & -G^{03}(\varepsilon) \\ -G^{10}(\varepsilon) & t_1^{-1} & -G^{12}(\varepsilon) & -G^{1m}(\varepsilon) \\ -G^{20}(\varepsilon) & -G^{21}(\varepsilon) & t_2^{-1} & -G^{2m}(\varepsilon) \\ -G^{30}(\varepsilon) & -G^{31}(\varepsilon) & -G^{32}(\varepsilon) & t_m^{-1} \end{bmatrix}$$

$$G^{nm}(\varepsilon)=-\frac{1}{4\pi}\frac{e^{i\varepsilon^{1/2}\left|\vec{R}_n-\vec{R}_m\right|}}{\left|\vec{R}_n-\vec{R}_m\right|}$$

decay slowly with increasing distance
contain free-electron singularities

Complex Energy Plane





ε_f is the highest occupied electronic state in energy

- Scattering is local since there are no states near the bottom of the energy contour
- Scattering is local since a large Im ε is equivalent to rising temperature which smears out the states
- Near ε_f scattering is non-local (metal)

Semi-conductors and insulators could work well since they have no states at ε_f

The scattering properties at complex energy can be used to develop highly efficient real-space and k-space methods

Multiple Scattering Theory

$$\tau^{-1}{}_{LL'}(\varepsilon) = t^{-1}_{L,L',0}(\varepsilon) \; - \; G_{LL'}(R_{ij},\varepsilon)$$

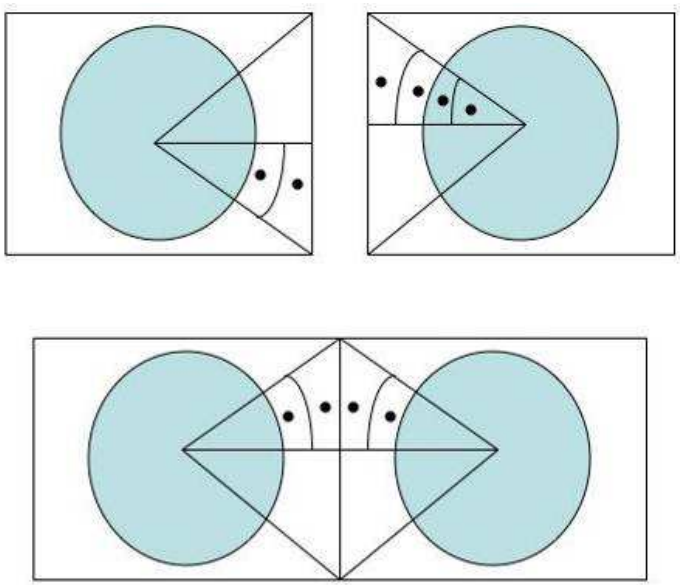
Depends on constituent V(r)
Independent of lattice

Depends on ε and
Underlying lattice structure

Representation is ideal for disorder systems since t-matrix is site diagonal !!
Coherent Potential Approximation (CPA)
Non-local CPA (based Dynamical Cluster Approximation to Dynamical mean-field theory

t-matrix

- Solve for t-matrix inside Voroni polyhedron
  - Using Calergo method



$$R_\ell(r) = j_\ell(\kappa r) - \kappa \int s^2 V(s) R_\ell(s) \left[n_\ell(\kappa s) j_\ell(\kappa r) - j_\ell(\kappa s) n_\ell(\kappa r) \right] ds$$

$$R_\ell(r) = c_\ell(r) j_\ell(\kappa r) - s_\ell n_\ell(\kappa r)$$

$$\frac{dc_\ell(r)}{dr} = -\kappa r^2 V(r) n_\ell(\kappa r) \left[c_\ell(r) j_\ell(\kappa r) - s_\ell n_\ell(\kappa r) \right]$$

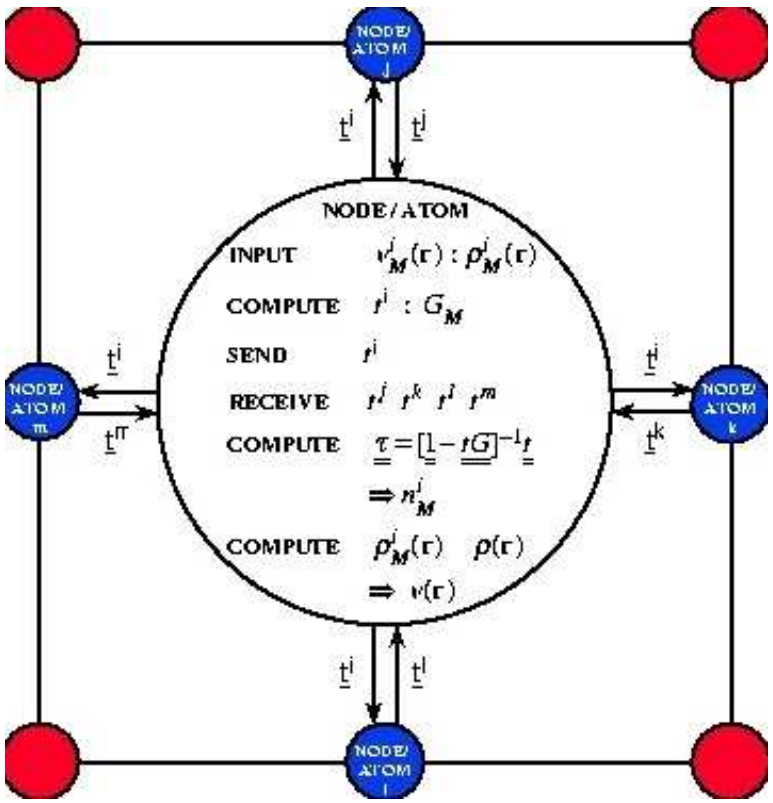
$$\frac{ds_\ell(r)}{dr} = -\kappa r^2 V(r) j_\ell(\kappa r) \left[c_\ell(r) j_\ell(\kappa r) - s_\ell n_\ell(\kappa r) \right]$$

Equations solved for both regular and irregular solutions
Since each atom and ℓ is independent can solve in shared memory. Similar for the structure constants.

Parallel Implementation

- **Green's function**

$$G^{nm}(\vec{r},\vec{r}\,';\varepsilon)=\sum_{LL'}Z_L^n(\vec{r}_n;\varepsilon)\tau_{LL'}^{nm}(\varepsilon)Z_{L'}^m(\vec{r}_m\,';\varepsilon)-Z_L^n(\vec{r}_<;\varepsilon)J_{L'}^m(\vec{r}_>\,';\varepsilon)\delta_{LL'}$$



- Scattering path matrix: real space

$$\underline{\tau}=\underline{M}^{-1}$$

$$\underline{M}=[\underline{t}^{-1}(\varepsilon)-\underline{G}(R_{nm},\varepsilon)]$$

t : scattering from single site

G: structure constant matrix

- Once M is fixed increasing N does not affect the local calculation of $\underline{M}^{-1}$

Tight-Binding MST Representation

Tight Binding Multiple Scattering Theory

Embed a constant repulsive potential

Shifts the energy zero allowing for calculations at negative energy

Rapidly decaying interactions

Free electron singularities are not a problem

Sparse representation

$$G^{nm,s}(\varepsilon) \propto e^{-(V^s-\varepsilon)^{1/2}|\vec{R}_n-\vec{R}_m|}$$

$$G^{nm}(\varepsilon) \propto \frac{e^{-\varepsilon^{1/2}\left|\vec{R}_n-\vec{R}_m\right|}}{\left|\vec{R}_n-\vec{R}_m\right|}$$

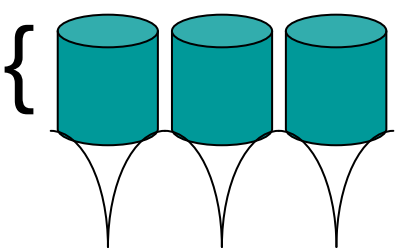
$$G = G_0 + G_0 t G_0 + G_0 t G_0 t G_0 = G_0 (I - t G_0)^{-1}$$

$$G^{-1} = G_0^{-1} - t$$

$$G^r = G_0 + G_0 t^r G_0 + G_0 t^r G_0 t^r G_0 = G_0 (I - t^r G_0)^{-1}$$

$$(G^r)^{-1} = G_0^{-1} - t^r \rightarrow G_0^{-1} = (G^r)^{-1} + t^r$$

$$G^{-1} = (G^r)^{-1} + t^r - t \rightarrow (G^r)^{-1} - \Delta t$$

2 Ryd. 

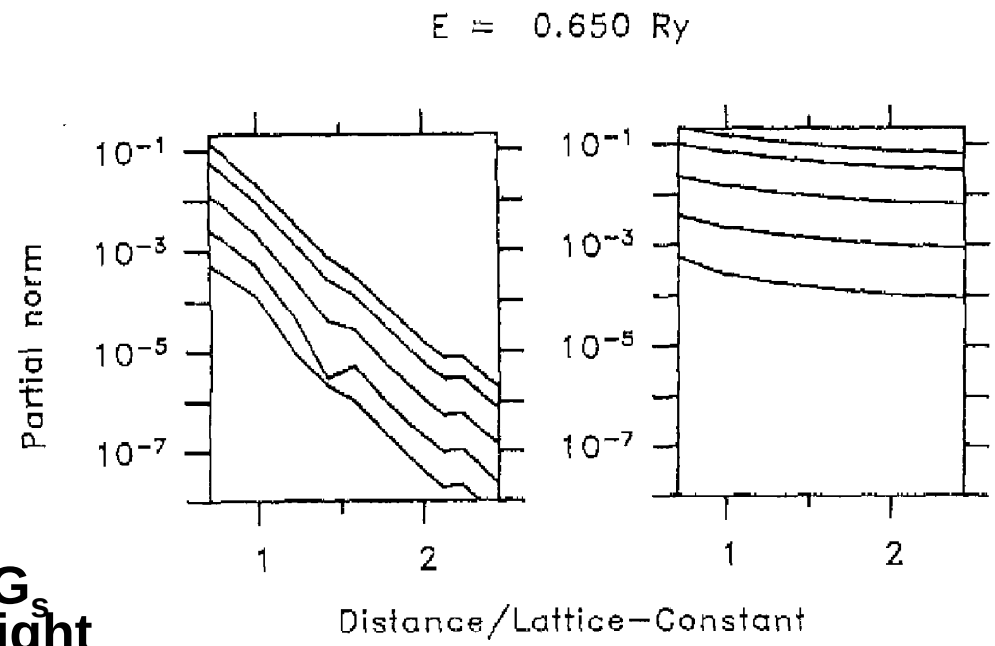
$$V^r \begin{cases} \text{Constant inside a sphere} & \vec{R} \le \vec{R}_{mt} \\ 0 & \vec{R} > \vec{R}_{mt} \end{cases}$$

Screened Structure Constants

- **Linear solve using m atom cluster that is less than the n atom system**
- **Easy to perform Fourier transform**
  - **K-space method**

$$G^s(\varepsilon) = [I - t^s G^{free}(\varepsilon)]^{-1}$$

$$G(\vec{k},\varepsilon) = \sum_m G^{m,s}(\varepsilon) e^{i\vec{k}\bullet\vec{R}_m}$$

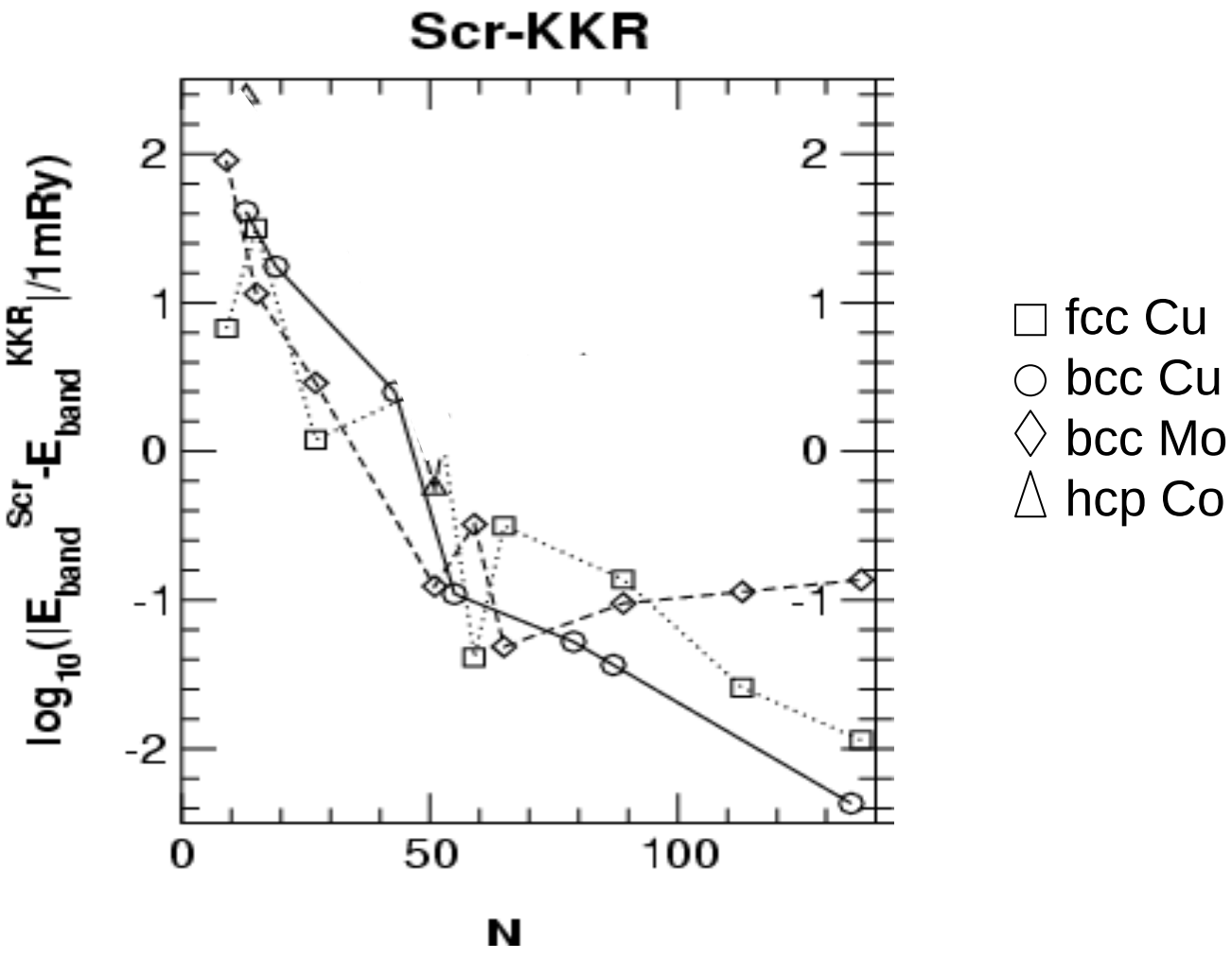


- **Screened Structure Constants G_s on the left unscreened on the right**
  - **Screened structure constants rapidly go to zero, whereas the free space structure constants have hardly changed**

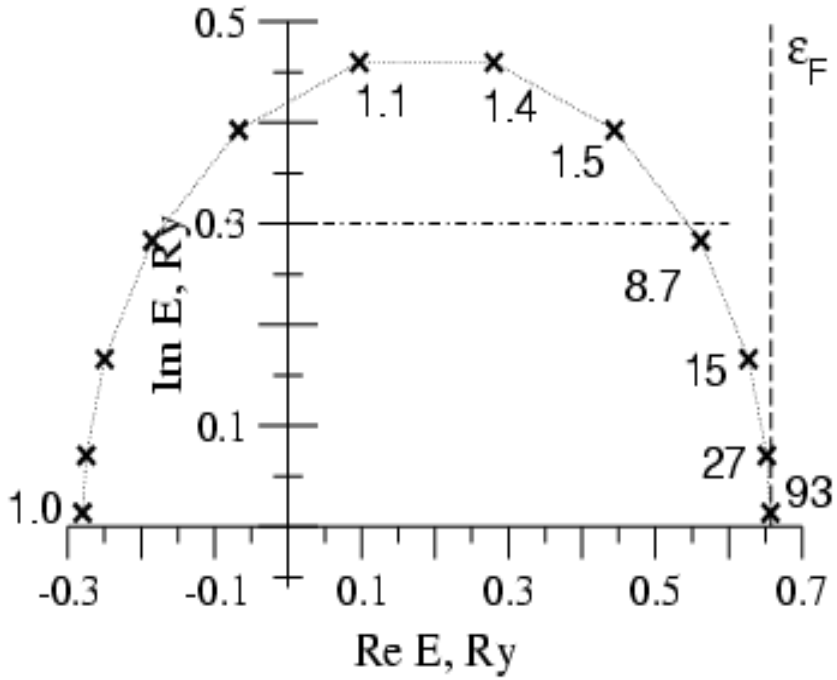
Screened MST Methods

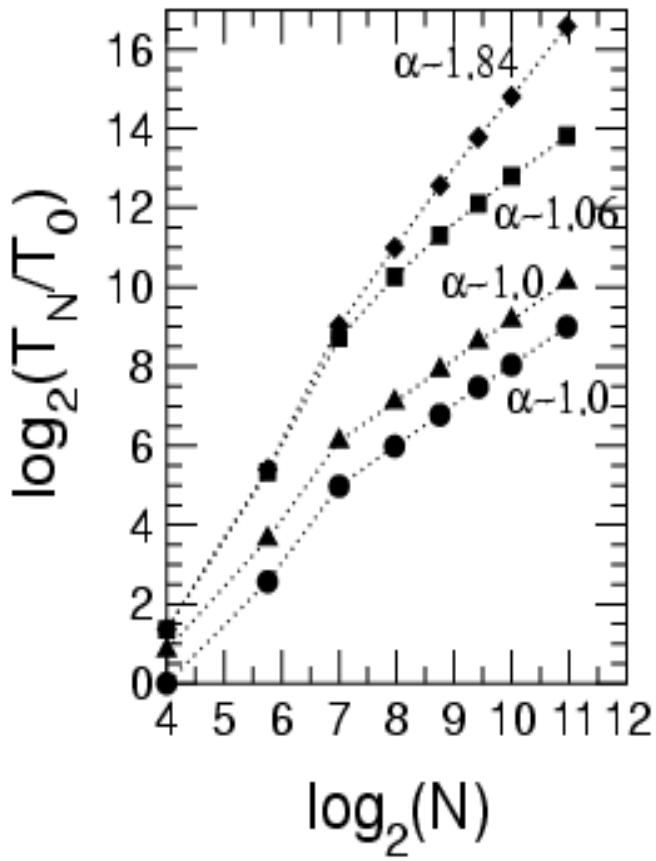
- Formulation produces a sparse matrix representation
  - 2-D case has tridiagonal structure with a few distant elements due to periodicity
  - 3-D case has scattered elements
    - Mainly due to mapping 3-D structure to a matrix (2-D)
    - A few elements due to periodic boundary conditions
- Require block diagonals of the inverse of $\tau(\varepsilon)$ matrix
  - Block diagonals represent the site $\tau(\varepsilon)$ matrix and are needed to calculate the Green's function for each atomic site
- Sparse direct and preconditioned iterative methods are used to calculate $\tau^{ii}(\varepsilon)$
  - SuperLU
  - Transpose free Quasi-Minimal Residual Method (TFQMR)

Screened KKR Accuracy



Timing and Scaling of Scr-K KKR-CPA





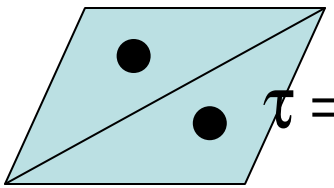
Conclusion

- **Initial benchmarking of the Screened KKR method**
  - **SuperLU $N^{1.8}$ for finding the inverse of the upper left block of τ**
  - **TFQMR with block Jacobi preconditioner $N^{1.06}$ for finding the inverse of the upper left block of τ**
- **Extremely high sparsity (97%-99% zeros increases with increasing system size)**
- **Large number of atoms on a single processor**
- **Real-space/Scr-KKR hybrid may provide the most efficient parallel approach for new generation architectres**
- **Single code contains**
  - **LSMS, KKR-CPA, Scr-LSMS and Scr-KKR-CPA**

Local Poisson Equation

Discontinuous Galerkin approach

$$-\Delta u = f \text{ in } \Omega$$



$$\nabla u = \sigma$$

τ = tensor basis in d-dimension

$$\nabla \cdot \sigma = f$$

ν = basis

u_h = is the representation of u on an element,

same for σ_h

$$\int_K \sigma_h \cdot \tau \, dx = -\int_K u_h \nabla \cdot \tau \, dx + \int_K \hat{u}_K \cdot n_K \tau \, ds, \ \forall \tau \in \sum(K)$$

$$\int_K \sigma_h \cdot \nabla \nu \, dx = -\int_K f \nu \, dx + \int_K \hat{\sigma}_K \cdot n_K \nu \, ds, \ \forall \nu \in P(K)$$