

Extensions of the traditional linear scaling concepts

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Outline

- The locality properties of interacting Fermion systems
- Hypersparsity of the independent particle density matrix
- Linear scaling geometry optimization

Basic strategy for achieving linear scaling

S. Goedecker: Rev. of Mod. Phys. 71, (1999)

- Formulate the electronic structure problem in terms of localized quantities
 - Wannier functions $W_i(\mathbf{r})$ centered at point \mathbf{R}_i

$$W_i(\mathbf{r}) \longrightarrow 0 \\ |\mathbf{R}_i - \mathbf{r}| \rightarrow \infty$$

- Density matrix $F(\mathbf{r}, \mathbf{r}')$

$$F(\mathbf{r}, \mathbf{r}') \longrightarrow 0 \\ |\mathbf{r} - \mathbf{r}'| \rightarrow \infty$$

Both decay exponentially in insulator

- Do not calculate exponentially small parts of the Wannier functions and density matrix. Calculate them only in the "localization region" where they are big enough.

Definition of Wannier functions

Periodic solid with Bloch functions $\Psi_{n,\mathbf{k}}(\mathbf{r})$

$$W_i(\mathbf{r}) = \frac{V}{(2\pi)^3} \int_{BZ} d\mathbf{k} e^{-i\mathbf{k}\mathbf{R}} \Psi_{n,\mathbf{k}}(\mathbf{r})$$

$$i = \mathbf{R}_i, n$$

Definition of Density matrix

At zero temperature ($2N$ electrons)

$$\begin{aligned} F(\mathbf{r}, \mathbf{r}') &= 2 \sum_{i=1}^N \Psi_i^*(\mathbf{r}) \Psi_i(\mathbf{r}') \\ &= 2 \sum_{i=1}^N W_i^*(\mathbf{r}) W_i(\mathbf{r}') \end{aligned}$$

$$H\Psi_i = \epsilon_i\Psi_i$$

Localization of the density matrix and Wannier functions in the many body context

E. Koch and S. Goedecker, Sol. State Comm., 119 105 (2001)

Questions:

- Does the one-particle reduced density matrix $D(\mathbf{r}, \mathbf{r}')$

$$D(\mathbf{x}'_1, \mathbf{x}_1) = \int \dots \int d\mathbf{x}_2 \dots d\mathbf{x}_N \Psi(\mathbf{x}'_1, \mathbf{x}_2, \dots, \mathbf{x}_N) \Psi(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N)$$

decay in the same way as the independent particle density matrix $F(\mathbf{x}, \mathbf{x}')$

- Can one construct one-particle Wannier functions from a many body wave function $\Psi(\mathbf{x}_1, \mathbf{x}_2, \dots, \mathbf{x}_N)$

Analogies

In a periodic solid $D(\mathbf{x}, \mathbf{x}')$ has the same symmetry properties as an independent particle Hamiltonian H ; Bloch's theorem applies

\mathbf{k} dependent (Kohn Sham) Hamiltonian

$$H_{\mathbf{k}}$$

(Kohn Sham) orbitals $\Psi_{n,\mathbf{k}}^H$ with band energies $\epsilon_{n,\mathbf{k}}$

$$H_{\mathbf{k}} \Psi_{n,\mathbf{k}}^H = \epsilon_{n,\mathbf{k}} \Psi_{n,\mathbf{k}}^H$$

$$H(\mathbf{r}, \mathbf{r}') \propto$$

$$\int_{BZ} H_{\mathbf{k}}(\mathbf{r}', \mathbf{r}) e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')} d\mathbf{k}$$

Independent particle Wannier functions

$$W_n^H(\mathbf{r} - \mathbf{R}) \propto$$

$$\int_{BZ} d\mathbf{k} e^{-i\mathbf{k}\mathbf{R}} \Psi_{n,\mathbf{k}}^H(\mathbf{r})$$

\mathbf{k} dependent one particle density matrix

$$D_{\mathbf{k}}$$

Natural orbitals $\Psi_{n,\mathbf{k}}^D$ with occupation number bands $n_{n,\mathbf{k}}$

$$D_{\mathbf{k}} \Psi_{n,\mathbf{k}}^D = n_{n,\mathbf{k}} \Psi_{n,\mathbf{k}}^D$$

$$D(\mathbf{r}, \mathbf{r}') \propto$$

$$\int_{BZ} D_{\mathbf{k}}(\mathbf{r}', \mathbf{r}) e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')} d\mathbf{k}$$

"Natural" Wannier functions

$$W_n^D(\mathbf{r} - \mathbf{R}) \propto$$

$$\int_{BZ} d\mathbf{k} e^{-i\mathbf{k}\mathbf{R}} \Psi_{n,\mathbf{k}}^D(\mathbf{r})$$

Decay Properties of D

Postulate:

In an insulator all \mathbf{k} dependent quantities are analytic with respect to \mathbf{k} , in a metal they are discontinuous at the Fermi level.

Paley-Wiener theorem:

$D(\mathbf{r}, \mathbf{r}')$ decays exponentially

Numerical results for decay constant in the Hubbard model

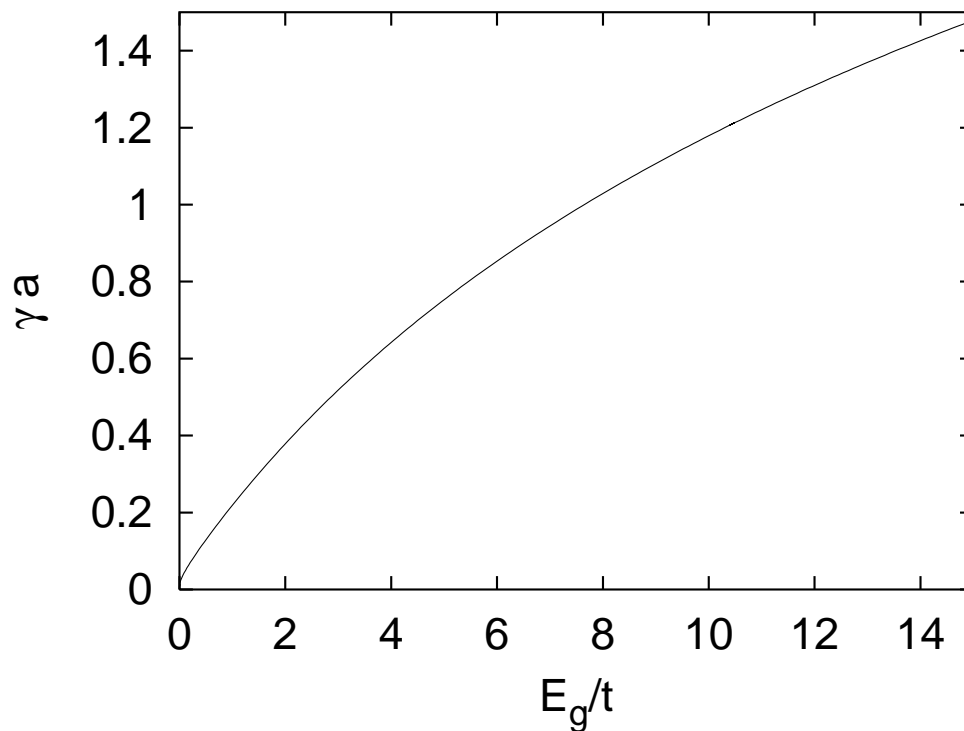


Figure 1: Decay constant γa of the density matrix for the Hubbard chain as function of the gap E_g .

Natural Wannier function

- Decay exponentially
- Virtual natural Wannier functions are presumably better localized than their independent particle counterparts
- Virtual natural Wannier functions have never been calculated for a realistic system

One can distinguish an insulator from a metal by looking only at ground state properties

Sparsity of W and F for independent particle schemes

How sparse are W and F ?

- The number of significant elements of F and W is small enough to be stored if small basis sets such as Gaussian type orbitals (GTO's) are used
- Problems arise if large high quality basis sets such as finite elements are used

Example: A system near the crossover point:

100 atoms each contributing 2 electrons: $N = 100$

1000 basis functions per atom: $M = 100\ 000$

Localization region contains all the 100 atoms

- W : $N \times M$, i.e. 10 million elements, feasible
- F : $M \times M$, i.e. 10 billion elements, unfeasible

Hyper-sparsity of the density matrix

S. Goedecker and O. Ivanov, PRB 59 7270 (1999)

Initial observation:

To get the best possible sparsity for the density matrix of the homogeneous electron gas we have to use plane waves

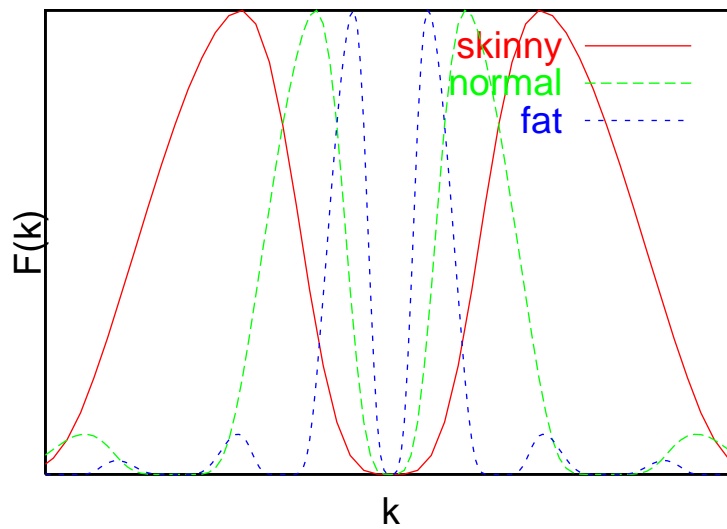
Hope:

- By using a basis set that is localized both in real and in Fourier space we can obtain a better sparsity for F for any system. A basis set with this property are wavelets.
- Obtain linear or close to linear scaling for metals at zero temperature

Frequency localization properties

Daubechies Degree 8 wavelets

Fourier power spectrum of 3 wavelets on neighboring resolution levels



Results: One dimensional insulator:

Scaling function basis: $F_{i,j} = \int \int \phi_i(r) F(r, r') \phi_j(r')$

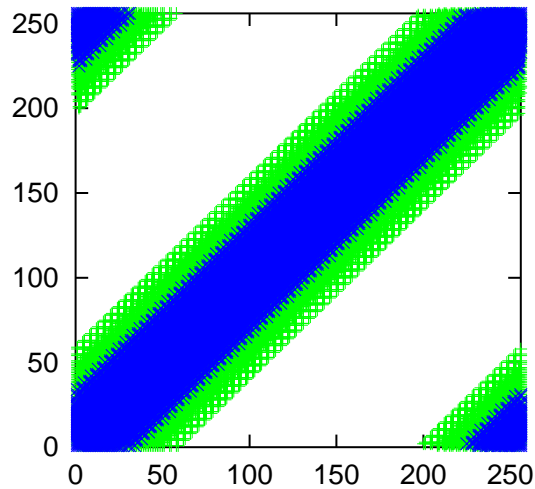


Figure 2: *Structure of the density matrix in a scaling function representation. Elements larger than 1.e-3 are denoted by green areas elements larger than 1.e-2 by blue areas.*

Wavelet basis: $F_{i,j} = \int \int \psi_i(r) F(r, r') \psi_j(r')$

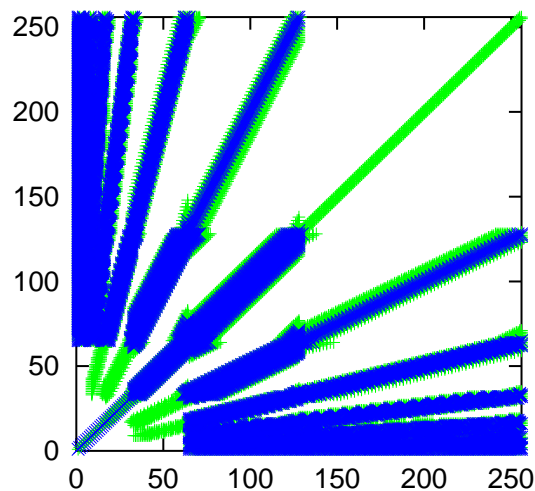
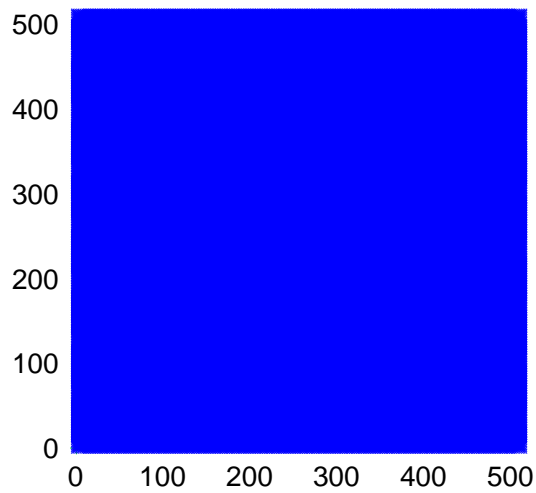


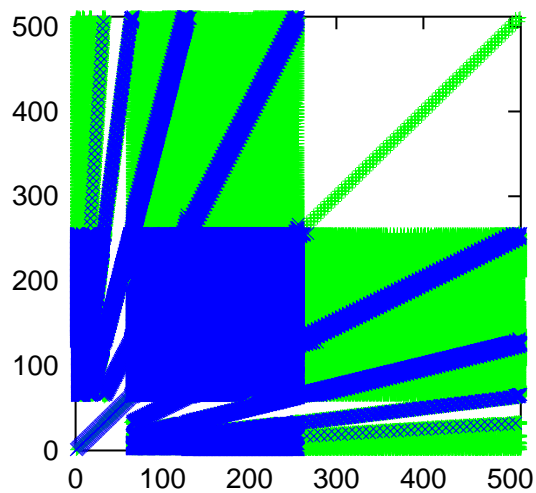
Figure 3: *Structure of the density matrix in a wavelet representation*

Results: One dimensional metal:

Scaling function basis: $elements \propto atoms^2$



Wavelet basis: $elements \propto atoms^{1.66}$



Compression rates

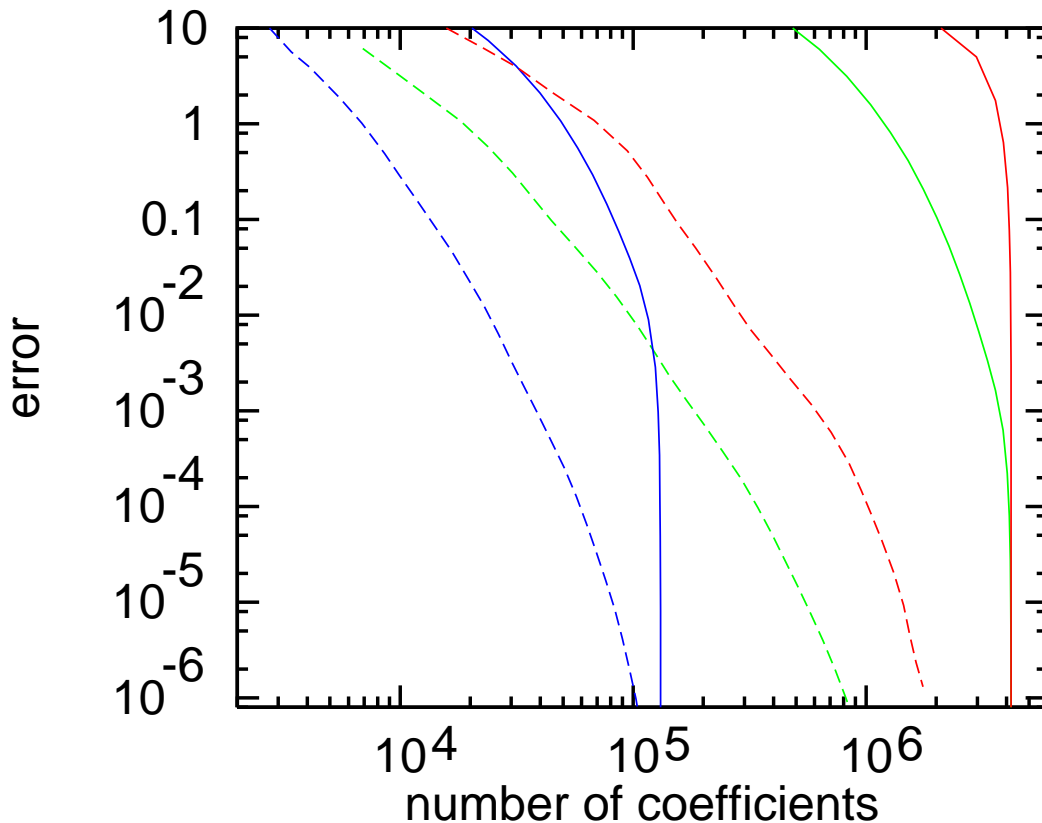


Figure 4: *The error in the density matrix versus the size of the data set necessary for its representation. Solid lines correspond to a scaling function representation, dashed lines to a wavelet representation. The two red curves are for a metallic system, the two green curves for an insulator and the two blue curves for an insulator where the density matrix was constructed indirectly via the Wannier functions.*

Geometry optimization

Basic steps of iteration it :

- Calculate forces on all the atoms \mathbf{f}_{it}
We assume linear scaling for this part
- Calculate displacements \mathbf{d}_{it} from the forces \mathbf{f}_{it}
 - Steepest descent:

$$\mathbf{d}_{it} = \alpha \mathbf{f}_{it}$$

- Conjugate gradient:

$$\mathbf{d}_{it} = \text{linear combination of } \{\mathbf{f}_1, \mathbf{f}_2, \dots, \mathbf{f}_{it}\}$$

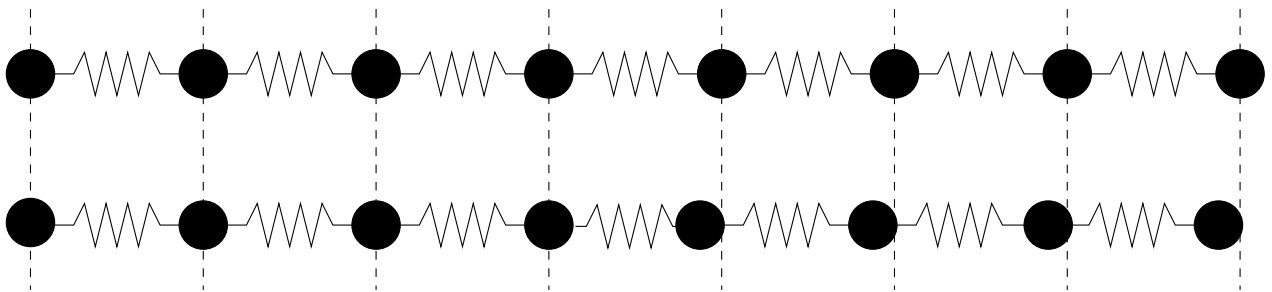
- Update atomic positions: $\mathbf{r}_{it+1} = \mathbf{r}_{it} + \mathbf{d}_{it}$

$O(N)$ geometry optimization

S. Goedecker et al., Rap. Comm. in Phys. Rev. B **64**, 161102-1 (2001)

Problem: number of iterations n_{it} increases with system size

Pictorial derivation:



For a chain of N_{at} atoms at least N_{at} iteration steps are required

For faster relaxation long wavelengths have to be excited

Mathematical analysis of n_{it} :

- Conjugate gradient:

$$n_{it} \propto \frac{\omega_{max}}{\omega_{min}} \log(\epsilon) \propto N_{at}$$

- Steepest descent:

$$n_{it} \propto \left(\frac{\omega_{max}}{\omega_{min}} \right)^2 \log(\epsilon) \propto N_{at}^2$$

ω_{max} highest phonon frequency

ω_{min} lowest phonon frequency

ϵ convergence threshold

N Number of atoms in chain

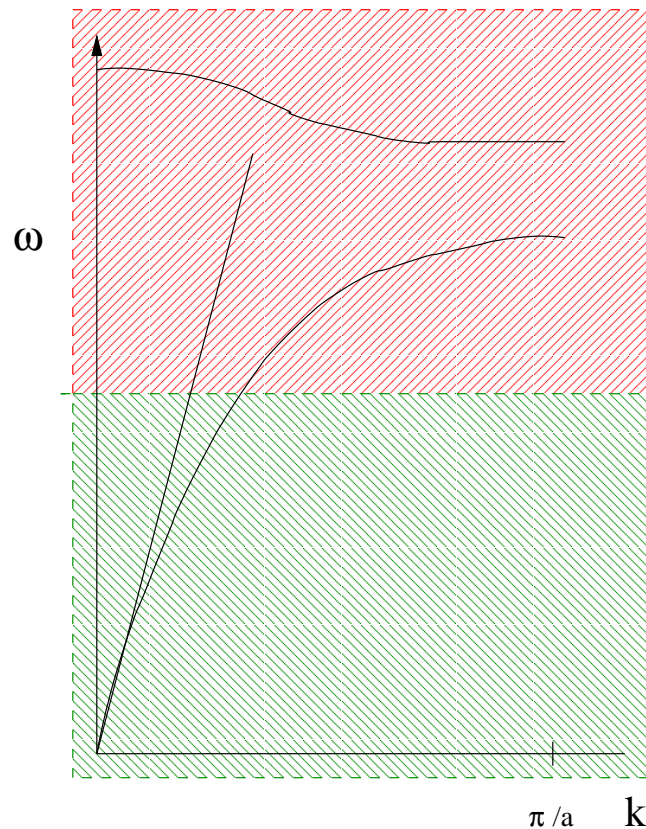
Three-dimensional bulk materials:

Conjugate gradient: $n_{it} \propto L = N_{at}^{1/3}$

Basic idea

To keep the number of iterations constant, treat high and low frequencies differently

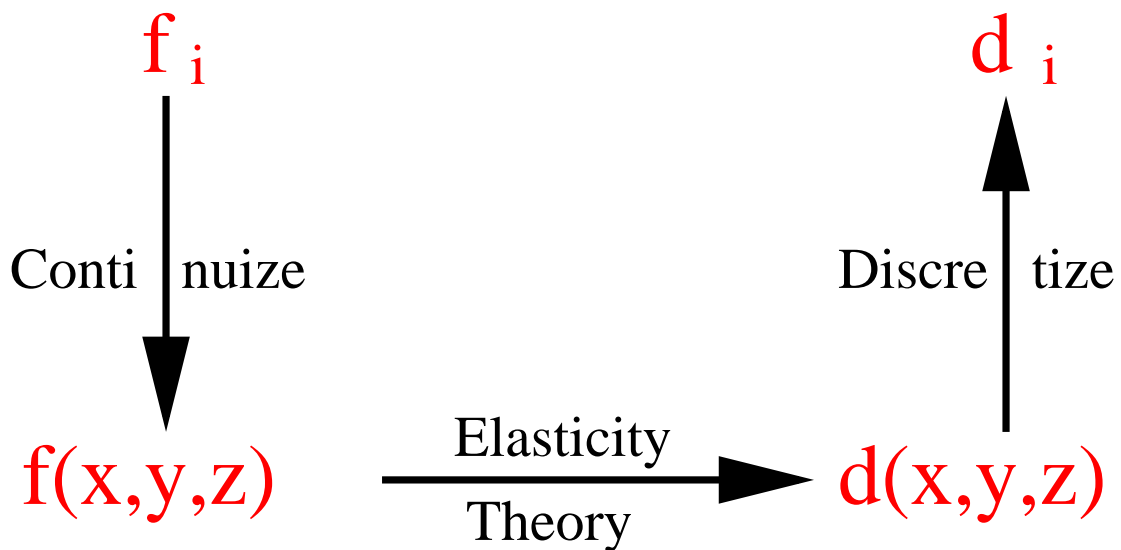
- high frequencies: ordinary steepest descent
- low frequencies: Elasticity theory



Elasticity theory is a good approximation on a length scale of a few inter-atomic distances

Algorithm

- high frequencies: ordinary steepest descent
- low frequencies: Elasticity theory



Elasticity equations are solved by multi-grid or Fourier techniques

Results for periodic systems

Table 1: Number of force evaluations n_f and CPU time T in seconds for the conjugate gradient (CG) and the linear scaling (SC) method for a divacancy in silicon.

number of atoms	n_f (CG)	n_f (LS)	T(CG)	T(LS)
510	102	106	.41	.50
998	124	106	.90	.93
1726	146	109	1.7	1.6
4094	184	115	5.1	4.2
13822	260	115	24.	14.
110592	502	115	373.	135.
884734	934	117	5586.	1147.

Results for non-periodic systems

Table 2: Number of force evaluations n_f for the conjugate gradient (CG) and the linear scaling (SC) for the relaxation of a Si quantum dot.

number of atoms	n_f (CG)	n_f (LS)
172	132	156
992	198	162
4158	294	210

Method also applicable to non-uniform systems

Comparison of 3 systems:

A: 1dim crystal with acoustic modes only

B: 1dim crystal with acoustic and optic modes

C: 1dim quasi crystal

Table 3: Number of iterations with the linear scaling method

number of atoms	64/55	128/144	256/233	512/610
System A	7	7	7	7
System B	11	11	11	11
System C	12	13	12	13

Conclusions

- Single particle Wannier functions can also be constructed in the many body context. These natural Wannier functions and the one-particle reduced density matrix decay exponentially in an insulator
- By exploiting the Fourier space localization properties additional sparsity (hypersparsity) of the density matrix can be obtained.
- Linear scaling geometry optimization allows for the efficient relaxation of large systems