Institute for the Theory of Advanced Materials in Information Technology

Computational Challenges and Solution Methods in DFT and TDDFT

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Current and recent members of the team: (CS side)

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Main issues of interest:

- Solve the eigenvalue problem efficiently [specificity: large number of eigenvalues]
- Find alternatives [avoid eigenvectors, eigenvalues]
- Solve various related computational problems [TDDFT, computation of dielectric matrix, ...]

Challenges in DFT

- DFT leads to a one-electron Shrödinger equation of the form

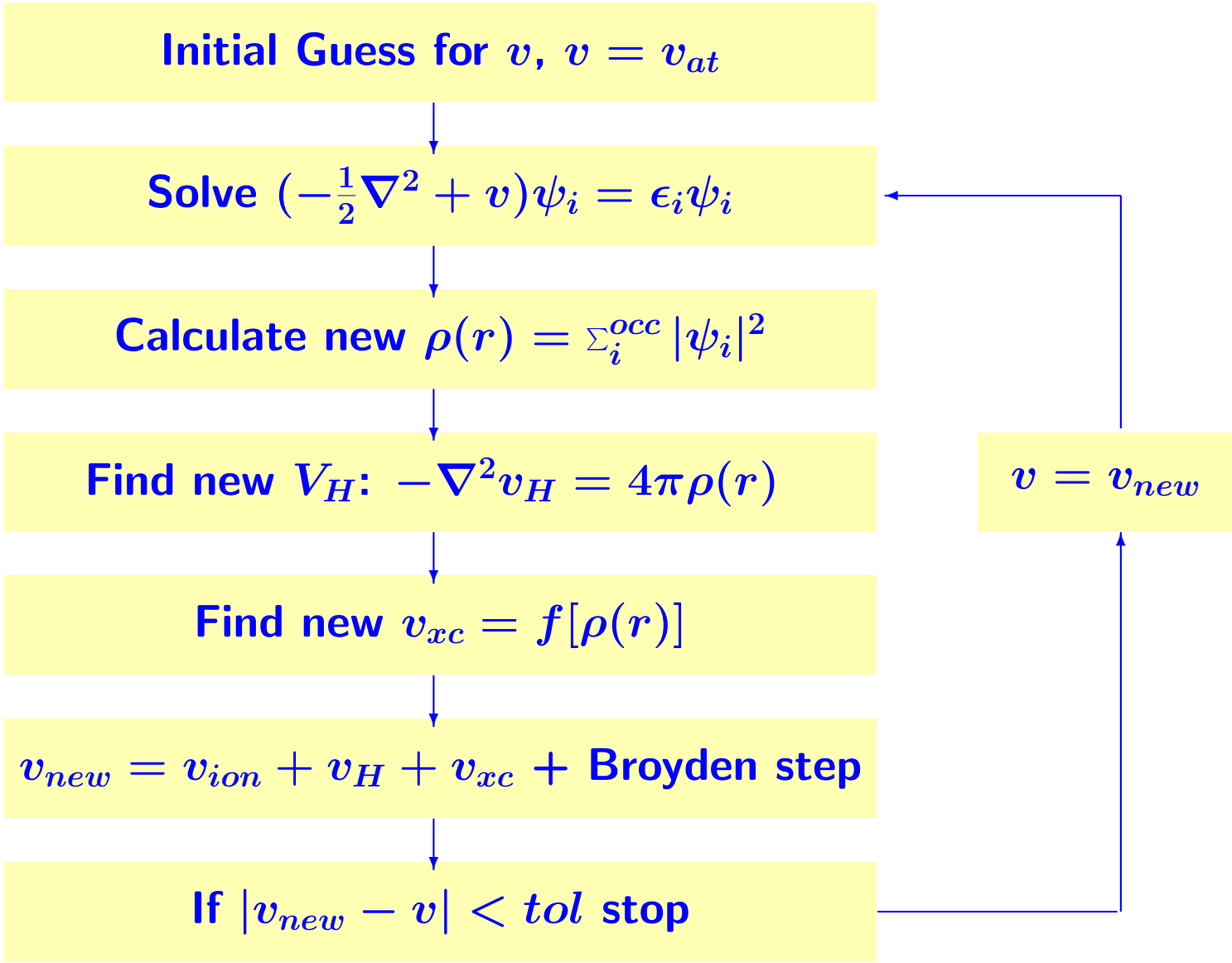
$$\left[-\frac{h^2}{2m}\nabla^2 + \underbrace{V_{ion} + V_H + V_{xc}}_{V_{tot}}\right]\Psi(r) = E\Psi(r)$$

- V_H = Hartree potential — local
- V_{xc} = Exchange & Correlation potential — local (LDA)
- V_{ion} = Ionic potential — Non-local

- Electron Density:

$$\rho(r) = \Sigma_i^{occup} |\Psi_i(r)|^2$$

- Above problem can be viewed as a nonlinear eigenvalue problem.


Initial Guess for v, $v = v_{at}$
Solve $(-\frac{1}{2}\nabla^2 + v)\psi_i = \epsilon_i \psi_i$
Calculate new $\rho(r) = \Sigma_i^{occ} |\psi_i|^2$
Find new V_H: $-\nabla^2 v_H = 4\pi\rho(r)$
Find new $v_{xc} = f[\rho(r)]$
$v_{new} = v_{ion} + v_H + v_{xc}$ + Broyden step
If $|v_{new} - v| < tol$ stop
$v = v_{new}$

- Most time-consuming part = computing eigenvalues / eigenvectors.

- Difficulty: large number of eigenvalues/eigenvectors to compute [number of occupied states]. For example, in real space, matrix size can be $N = 1,000,000$ and the number of eigenvectors could be 2,000.

- Self-consistent loop takes a few iterations (typically 4 to 12)

Standard methods ('The old')

- Lanczos method. Basis generated from 3-term recurrence:

$$\beta_{j+1} v_{j+1} = A v_j - \alpha_j v_j - \beta_j v_{j-1}$$

- Add preconditioning: Davidson.

$$v_{j+1} = M_j^{-1}(A u_j - \theta_j u_j); \quad [u_j, \theta_j] = \text{current eigenpair}$$

- Add implicit restart – clever way of restarting Lanczos process.

- Software. Best-known code = ARPACK

Issues

Preconditioning

- No clear-cut convincing approach for large numbers of eigenvalues.
- Simplest method: filtering by averaging..

Orthogonalization

- Becomes problematic for large number of eigenvalues -
- Idea of spectrum sclicing may be attractive but hard to implement

The outer loop

- Each self consistent loop should utilize previous information when starting – not easy to do with standard packages such as ARPACK

Current work on eigenvalue algorithms ('The new')

Focus:

(1) AMLS and related methods

(2) Block versions of restarted Lanczos

Motivation:

(1) Excellent success of AMLS in structural engineering. Similarity: large number of eigenvectors to compute

(2) Standard packages (ARPACK) do not easily take advantage of self-consistent loop. Also: not specialized for large number of eigenvalues.

Block-Lanczos – advantages

- Basic principle of the Block Lanczos algorithm: operate on block of b columns instead of only one column as in standard Lanczos.

Advantages:

- Can exploit a block of several initial guesses of eigenvectors
- Deals well with clustered or multiple ('degenerate') eigenvalues
- Can yield better cache performance (BLAS 3 instead of BLAS 2)

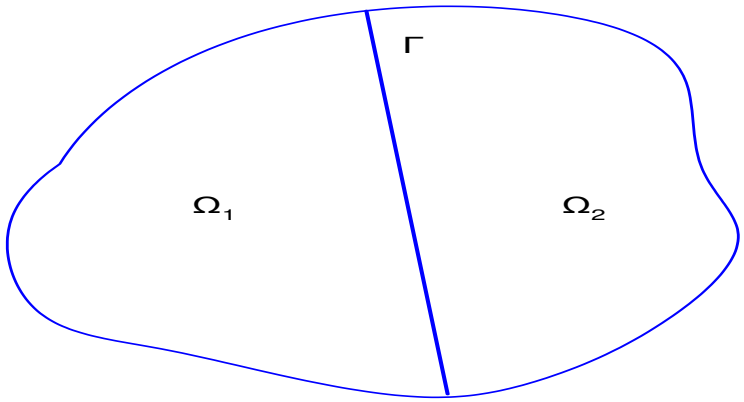
Issues:

- How to implement implicit restarts?
- Important to dynamically adapt block size

Automatic Multi-Level Substructuring

Origin: Extention of substructuring for eigenvalue problems.

Background: Domain decomposition. Let $A \in \mathbb{C}^{n\times n}$, Hermitian



$$\rightarrow A = \begin{pmatrix} B & E \\ E^* & C \end{pmatrix} \quad B \in \mathbb{C}^{(n-p)\times(n-p)}$$

Note: B is block-diagonal

Main Reference:

J. K. Bennighof and R. B. Lehoucq, **An automated multilevel substructuring method for eigenspace computation in linear elastodynamics, To appear in SIAM. J. Sci. Comput.**

Basic idea of the method for two levels

First step: eliminate the blocks E, E^*.

$$U = \begin{pmatrix} I & -B^{-1}E \\ 0 & I \end{pmatrix} \rightarrow U^*AU = \begin{pmatrix} B & 0 \\ 0 & S \end{pmatrix} ; \quad S = C - E^*B^{-1}E.$$

Original problem is equivalent to $U^*AUu = \lambda U^*Uu \rightarrow$

$$\begin{pmatrix} B & 0 \\ 0 & S \end{pmatrix} u = \lambda \begin{pmatrix} I & -B^{-1}E \\ -E^*B^{-1} & M_S \end{pmatrix} u \; ; M_S = I + E^*B^{-2}E$$

Second step: neglect the coupling in right-hand side matrix:

$$Bv = \mu \, v$$

$$Sw = \eta \, M_S w.$$

- Compute a few of the smallest engenvalues of the above problem.

Third step: Build a 'good' subspace to approximate to eigenfunctions of original problem. The basis used for this projection is of the form

$$\left\{ \hat{v}_i = \begin{pmatrix} v_i \\ 0 \end{pmatrix} \quad i = 1, \ldots, m_B; \qquad \hat{w}_j = \begin{pmatrix} 0 \\ w_j \end{pmatrix} \quad j = 1, \ldots, m_S \right\},$$

where $m_B < (n-p)$ and $m_S < p$.

Then use this subspace for a Rayleigh-Ritz projection applied to

$$\begin{pmatrix} B & 0 \\ 0 & S \end{pmatrix} \begin{pmatrix} u^B \\ u^S \end{pmatrix} = \lambda \begin{pmatrix} I & -B^{-1}E \\ -E^* B^{-1} & M_S \end{pmatrix} \begin{pmatrix} u^B \\ u^S \end{pmatrix}$$

(Note: not the original problem.)

Final step: exploit recursion –

NOTE: algorithm does only one shot of descent - ascent (no iterative improvement).

Issues investigated

- AMLS is a one shot algorithm - accuracy unlikely to be sufficient for electronic structure calculations. Problem: develop an improved version.

- AMLS can be viewed as Domain-Decomposition applied to shift-and-invert – with one shift at zero. Problem: develop a multi-shift version.

- Example: use AMLS with shift σ – obtain approximate eigenvectors (one shot 'descend and ascend'). Keep in basis. Change σ, add new eigenvectors to basis, etc..

Avoiding the eigenvalue problem

Recall: Density matrix,

$$\rho(r,r') = \sum_j f(E_j)\Psi_j(r)\overline{\Psi_j(r')} \quad \text{with} f(\lambda) = \text{occupancy factor}$$
$$\equiv f(H)$$

Main observation: Charge density ρ can be viewed as the diagonal of the density matrix: When $\psi_j(r)$ is discretized w.r.t. r then $\rho(r_i,r_i) \equiv \rho_{ii}$, the diagonal entry of $\rho(r,r')$, is the charge density at location r_i. Standard methods compute this by computing explicitly the eigenfunctions. ➨ Any orthogonal basis of the same space can be used. Is the eigenbasis an overkill?

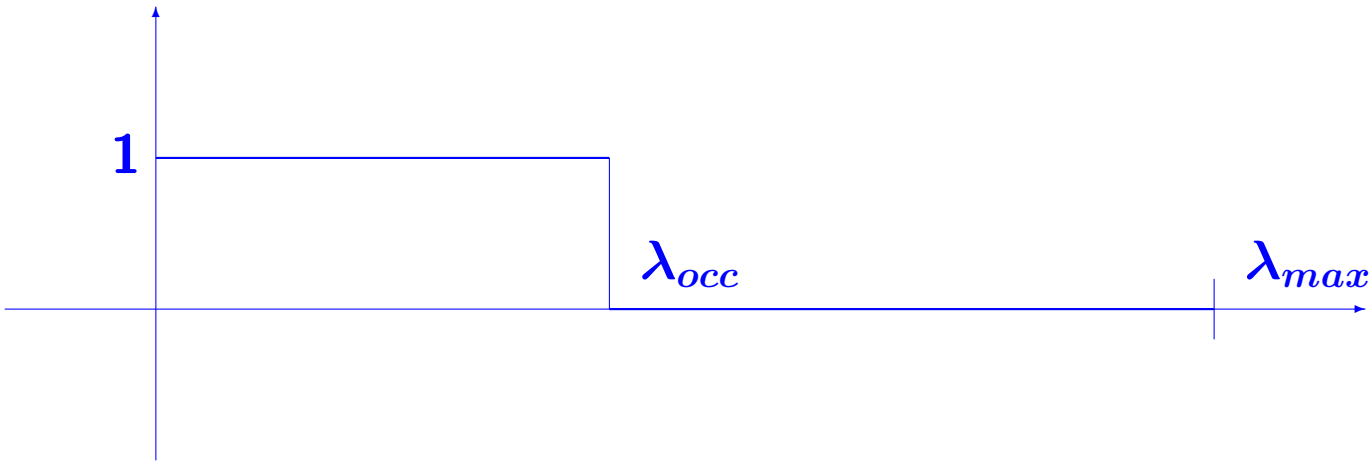
➨ 'Order n methods' find approximations to $\rho(r,r')$ by exploiting its decay properties

Previous work - Use of Chebychev polynomials ('the old')

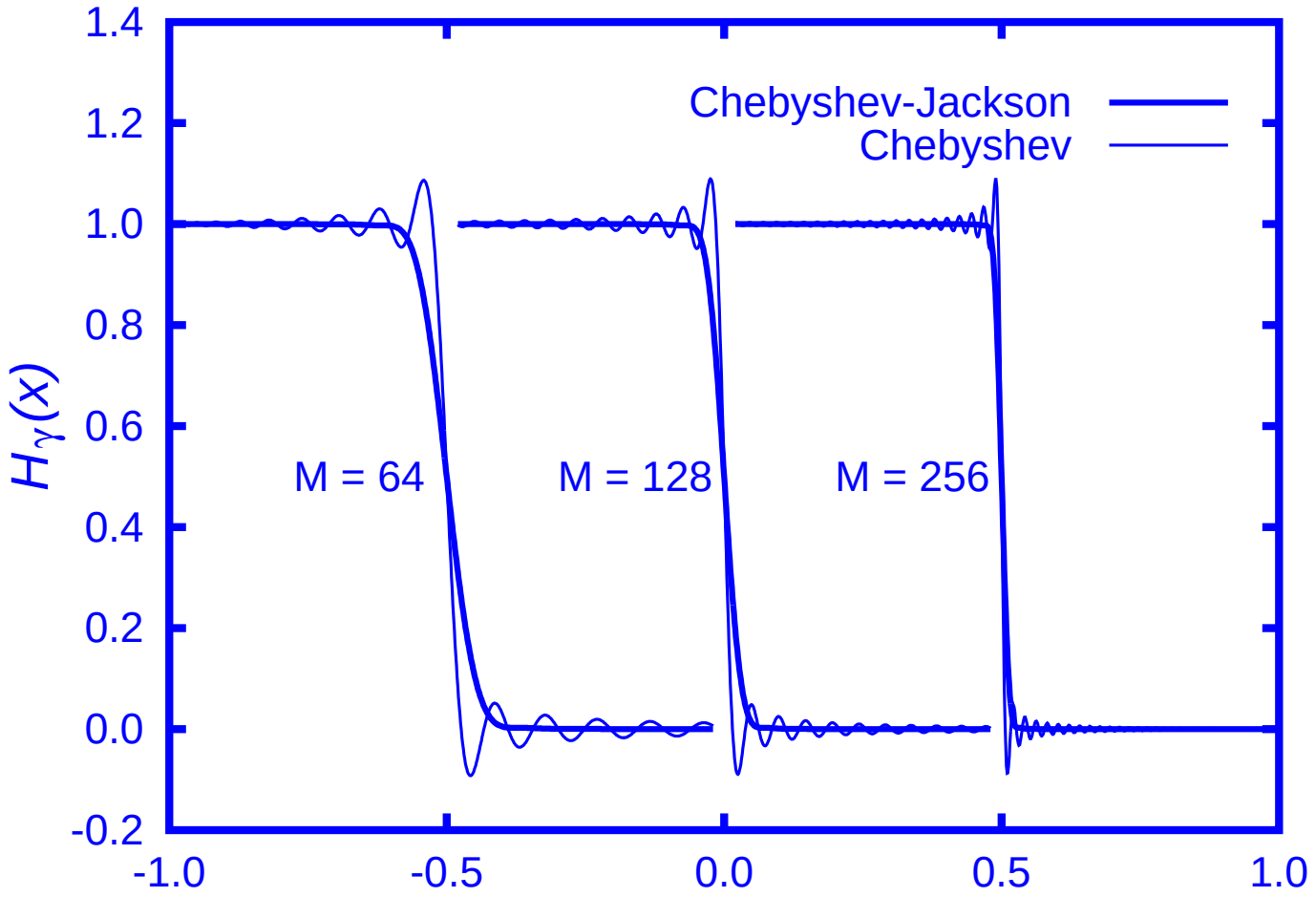
Main idea: Call H the original Hamiltonian matrix. Then

$$P = f(H)$$

where $f(t)$ is the Heaviside function:



- Replace f by a polynomial using Chebyshev expansions.
- This is now done in the planewave space not real space.



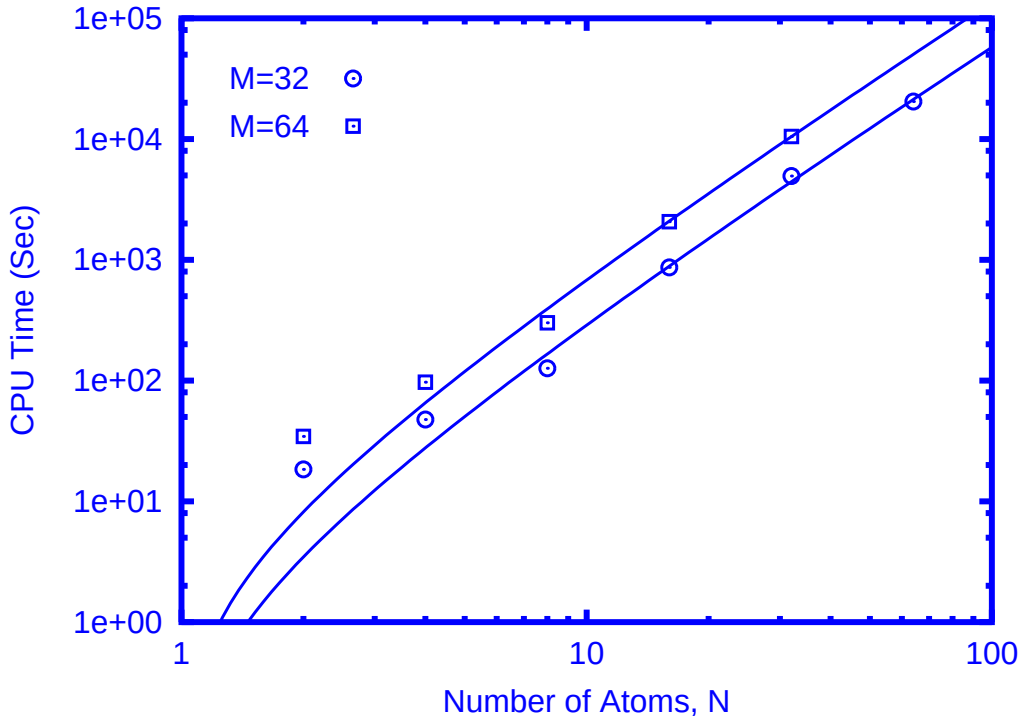
Chebyshev and Jackson polynomials of degree 64, 128, and 258

[Jackson expansions are modifications of the least-squares Chebyshev expansions that avoid Gibbs oscillations.]

- P is approximated by $p_m(H)X$ where X is a matrix of size $n \times p$ [e.g., 1st p columns of identity]

- Exploit recurrence relations of Chebyshev polynomials and near bandedness of P in planewave basis

- Test : with (Crystalline silicon) [See L. Jay et al. 1998]
- Times scale like $n^2 \log n$
- Cost is still high but : (1) can avoid eigenvectors completely, and (2) can still iterate to self-consistency



Current Approaches ('the new')

$$P = f(H)$$

where f is a step function. Approximate f by, e.g., a polynomial

- Result: can obtain columns of P inexpensively via:

$$Pe_j \approx p_k(H)e_j$$

- Exploit sparsity of P (especially in planewave basis)- ideas of "probing" allow to compute several columns of P at once.

- Statistical approach: work of Hutchinson for estimating trace of a matrix [used in image processing] adapted to estimating diagonals.

- Many variants currently being investigated

Example 1 : Statistical approach

- Let a sequence of random vectors $v^1, \ldots, v^s$ with entries satisfying a normal distribution. Diagonal of a matrix B can be approximated by

$$D^s = \left[\sum_{k=1}^{s} v^k \odot Bv^k\right] \oslash \left[\sum_{k=1}^{s} v^k \odot v^k\right]$$

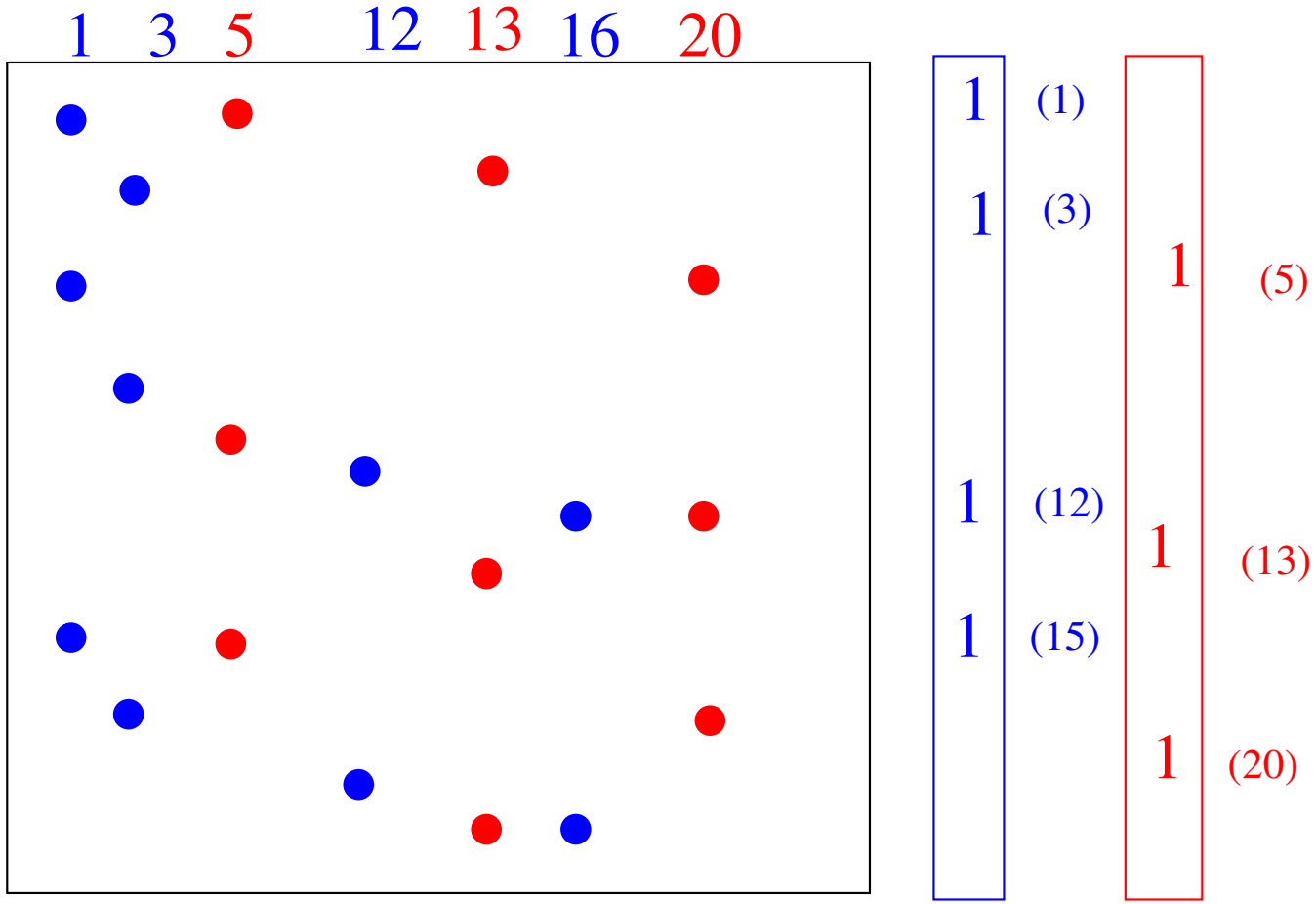
in which $\odot$ is a componentwise product of vectors, and similarly $\oslash$ represents a componentwise division of vectors.

- Deterministic approach: For a banded matrix (bandwidth p), there exists p vectors such the above formula yields the exact diagonal.

- These methods would require computing $p_k(H)v$ for several v's. Generally: method is expensive unless bandwith is small.

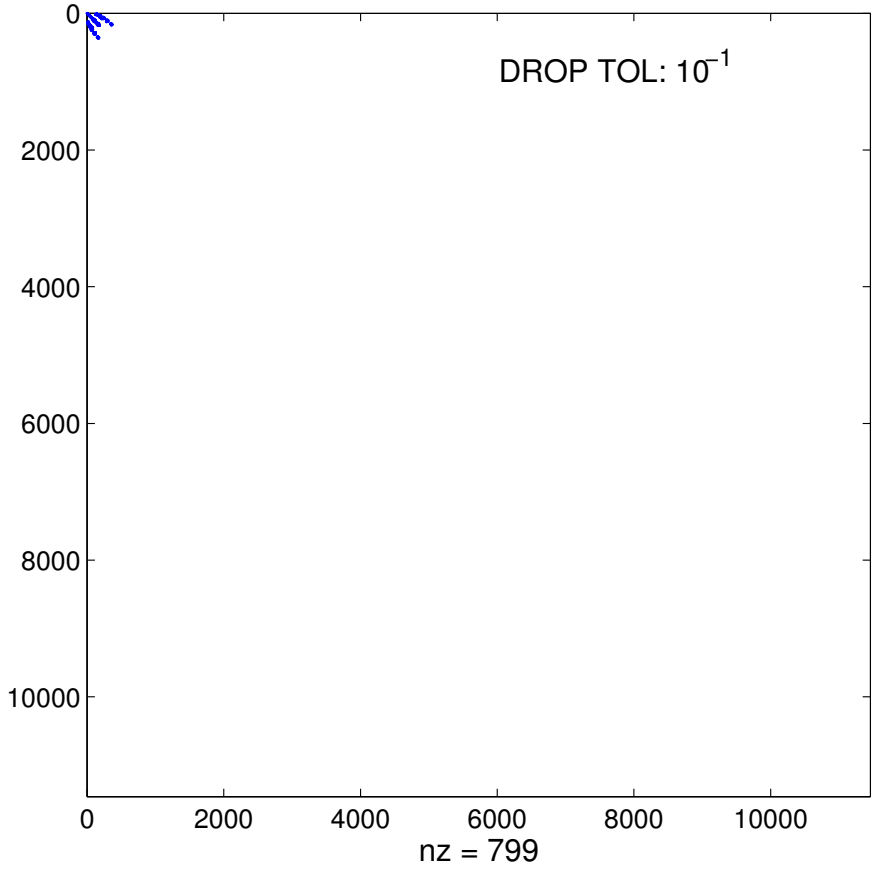
Example 2 : Sparsifying the density matrix in PW basis

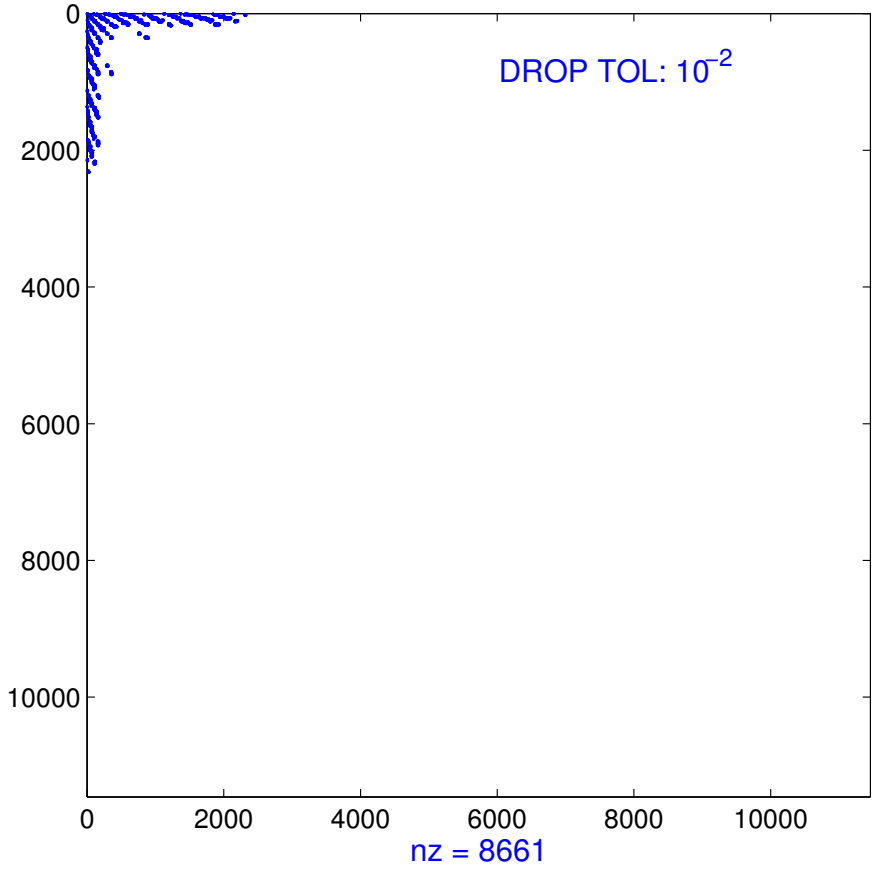
- Recall $P = f(H) \;=\; \{\rho(r, r')\}$
- Consider the expression in the G-basis
- Most entries are small –
- Idea: use technique of "probing" or "CPR" or "Sparse Jacobian" estimators ....

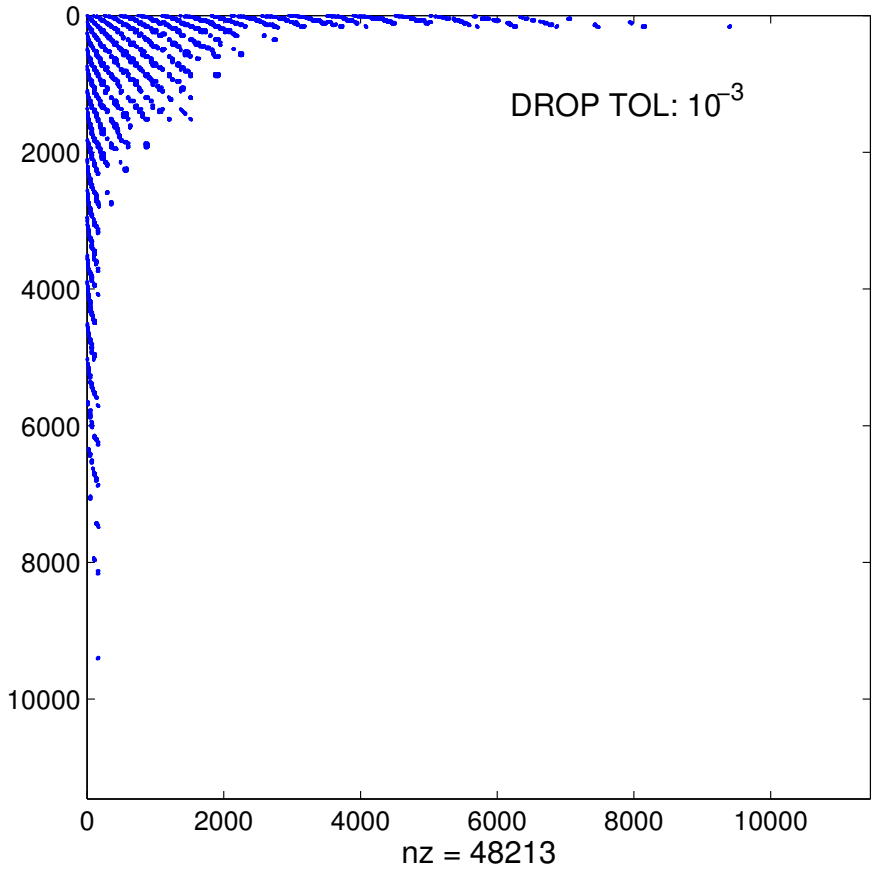


Probing in action: blue columns can be computed at once by one matrix-vector product. Then red columns can be coumputed the same way

Density matrix for Si64




DROP TOL: 10^-2
0
2000
4000
6000
8000
10000
nz = 8661


DROP TOL: 10^{-3}
0
2000
4000
6000
8000
10000
nz = 48213

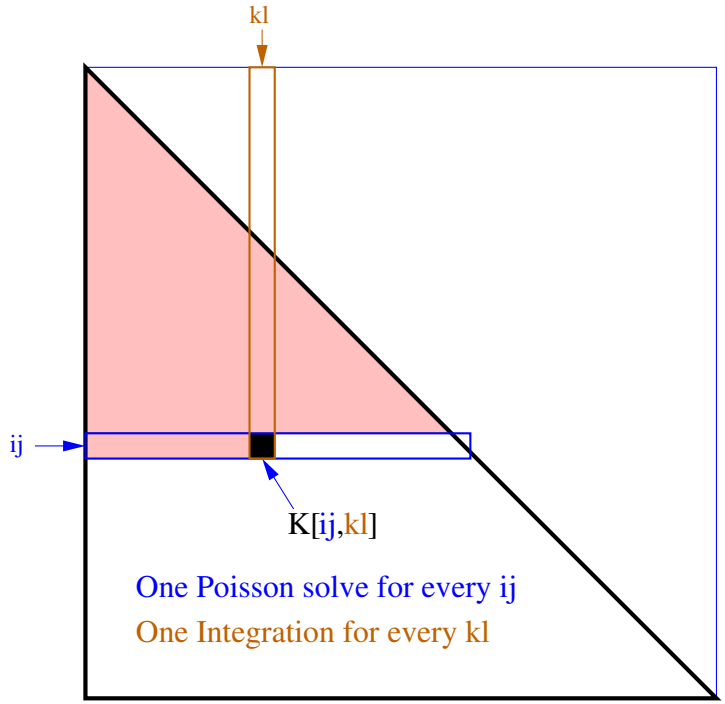
TIME DEPENDENT DFT

TDLDA: Use of planewave bases and FFT

Recall :

$$K_{ij,kl} = \int_\Omega \left(\Psi_i(\mathrm{r}) \bar{\Psi}_j(\mathrm{r}) \frac{dV_{\mathbf{xc}}(\mathrm{r})}{d\rho(\mathrm{r})} + \Phi_{ij}(\mathrm{r}) \right) \Psi_k(\mathrm{r}) \bar{\Psi}_l(\mathrm{r}) d\mathrm{r}.$$

With $\triangle \Phi_{ij}(\mathrm{r}) = -4\pi \Psi_i \bar{\Psi}_j(\mathrm{r}).$



Coupling Matrix K

- Previous work [our group] : work in real space + use CG to solve Poisson's equation.

- Real space approach does not exploit specific features of the physics when solving Poisson's equation.

- Idea is to use FFTs: (In essence: Use "fast Poisson solvers")

- Expand each wavefunction in planewave basis:

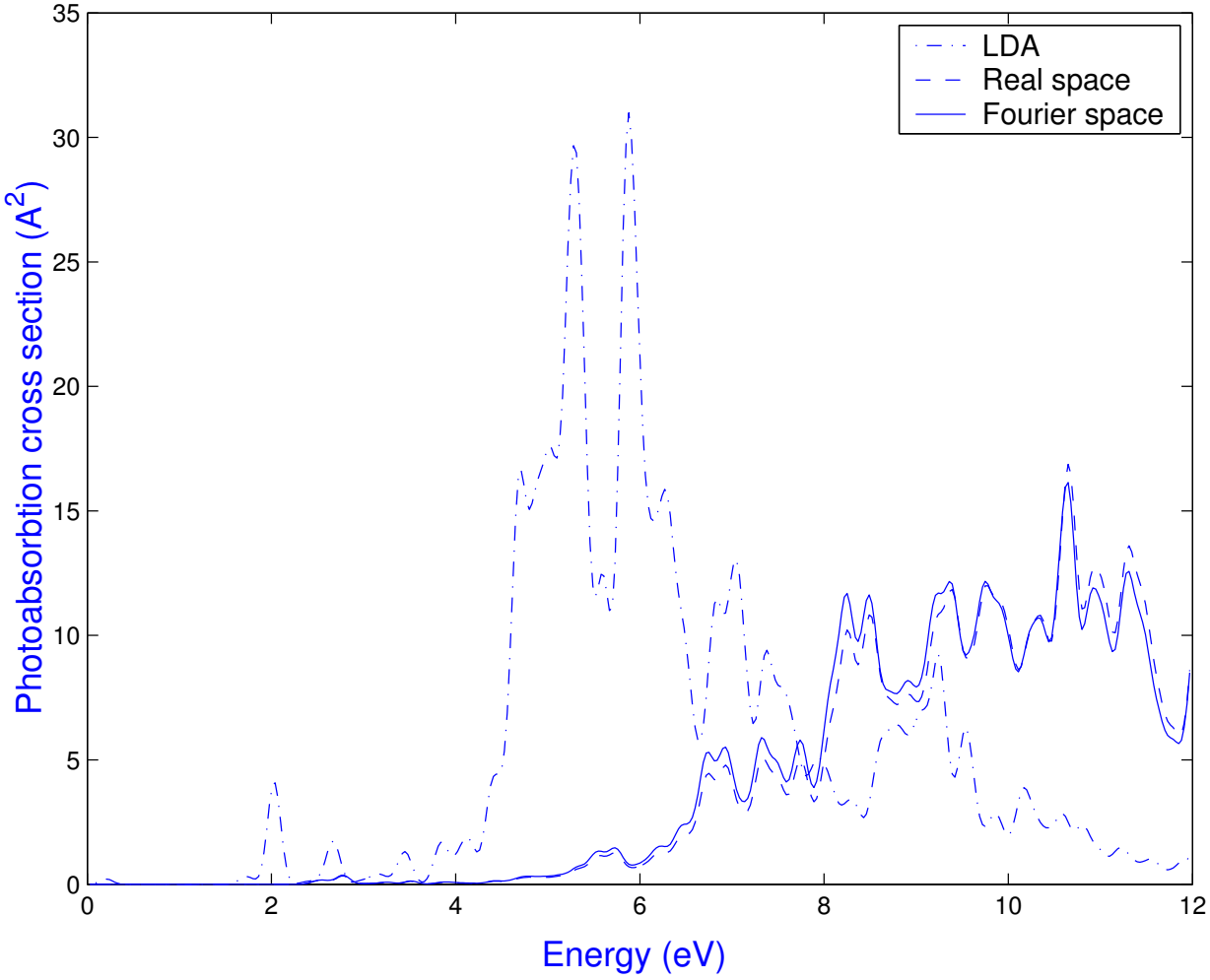
$$\Psi_j(\mathrm{r}) = \sum_{\mathrm{l}} \psi_{\mathrm{l}}^j \exp i(\mathrm{l}.\mathrm{r}) \longrightarrow \Phi_{ij}(\mathrm{r}) = 4\pi \sum_{(\mathrm{l},\mathrm{l}')\mathrm{l}\neq\mathrm{l}'} \frac{\psi_{\mathrm{l}}^i \bar{\psi}_{\mathrm{l}'}^j}{\|\mathrm{l}-\mathrm{l}'\|^2} e^{i(\mathrm{l}-\mathrm{l}').\mathrm{r}}.$$

- Many improvements can now be made. For example, in practice meaningful 'support' of $\psi_i\psi_j$ is small

$$\mathcal{F}(\Psi_i\bar{\Psi}_j)(\mathrm{k}) = \sum_{\mathrm{r}} e^{i\mathrm{k}.\mathrm{r}}(\Psi_i\bar{\Psi}_j)(\mathrm{r}) = \sum_{\mathrm{r}\,\in\,\mathbf{Supp}(\Psi_i\bar{\Psi}_j)} e^{i\mathrm{k}.\mathrm{r}}(\Psi_i\bar{\Psi}_j)(\mathrm{r}).$$

Results

- **Compare Real space code with planewave code for $Si34H36$**



- Compare times for Real space code and planewave code [for $Si34H36$]

Method	Wall-Clock Time (hours)
Real Space Code	15:30
PW: Initial Implementation	3:30
PW: Optimized load balancing	2:30

Wall-clock time of the parallel TDLDA code using Fourier space and Real Space for the Si34H36 test case running on 8 processors

Note: Gain a factor of 5-6 wrt to optimized version of TDLDA code. Compound with another factor of 3-4 from original to optimized real-space code $\longrightarrow$ 15 to 24 faster than [Vasiliev et al. 2000]

- More to come!

Specific Plans (TDDFT)

- 1. Optimize PW-based code – more opportunities for improvements;
- 2. Develop an efficient library for TDDFT
- 3. Do a large challenging new calculation;
- 4. In TDLDA, is it possible to better exploit the special nature of right-hand sides in Poisson's equation:

$$\nabla^2 \Phi_{ij\sigma}(\mathrm{r}) = -4\pi \psi_{i\sigma}(\mathrm{r}) \psi_{j\sigma}(\mathrm{r}).$$

More general plans

- 1. Most costly computation currently is still: the eigenvalue problem. Q: Can the success of AMLS [Automatic Multi-Level Substrututing] be extended from structural engineering to electronic structures calculations..

- 2. Challenge number 1 is: Can we avoid eigenvalue calculations altogether and still get good accuracy? [at lower cost?]