Parallel Coordinate Descent Methods for Full Configuration Interaction

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The Many-Body Time-Independent Schrödinger Equation

- Ground-state wavefunction $|\boldsymbol{\Phi_0}\rangle$ and energy E_0 of a chemical system given by
$$\hat{H}|\boldsymbol{\Phi_0}\rangle = E_0|\boldsymbol{\Phi_0}\rangle,$$
where $\hat{H} = -\frac{1}{2}\sum_{i=1}^{n_{\mathrm{elec}}} \nabla_i^2 + V(x_1, x_2, \ldots, x_{n_{\mathrm{elec}}})$.
- Application: Molecular property calculations
- Computational Methods:
    - Density Functional Theory (DFT)
    - Many-Body Perturbation Theory (MBPT)
    - Coupled Cluster (CC)
    - **Full Configuration Interaction (FCI)**: for strongly-correlated system

Configuration Interaction Method

- Based on one-electron spin–orbitals $\{\chi_p\}_{p=1}^{n_{\text{orb}}}$ from Hartree–Fock procedure
- Wavefunction approximated as linear combination of anti-symmetrized tensor products (Slater determinants)

$$|\mathbf{\Phi_0}\rangle = \sum_{i=1}^{N_{\text{FCI}}} c_i |D_i\rangle = \sum_{i=1}^{N_{\text{FCI}}} c_i |\chi_{p_i}\chi_{p_j}\cdots\chi_{p_k}\rangle$$

- FCI variational space dimension: $N_{\text{FCI}} = \binom{n_{\text{orb}}}{n_{\text{elec}}}$
- Schrödinger equation transformed to eigenvalue problem

$$H\mathbf{c} = E_0\mathbf{c}, \quad H \in \mathbb{R}^{N_{\text{FCI}} \times N_{\text{FCI}}}, \quad \mathbf{c} \in \mathbb{R}^{N_{\text{FCI}}}$$

FCI Matrix Properties

Entry: $H_{ij} = \langle D_i | \hat{H} | D_j \rangle$, not guaranteed to be non-negative.

- Symmetric: $H_{ij} = H_{ji}$.
- Sparse: $H_{ij} = 0$ if $|D_i\rangle$ and $|D_j\rangle$ differ by more than two orbitals. H has $O(n_{\text{elec}}^2 n_{\text{orb}}^2)$ entries per row.
- Ground-state eigenvalue $E_0 < 0$, ground-state eigenvector $\mathbf{v_0}$ sparse in the sense of truncation.

Memory usage

Table: Different Molecule Systems and Storage cost

Molecule	Basis	Electrons	Spin–Orbitals	Dimension	Memory
H_2O	cc-pVDZ	10	48	$\sim 10^8$	$\sim$ 1 GB
N_2	cc-pVDZ	14	56	$\sim 10^{11}$	$\sim$ 1 TB
N_2	cc-pVTZ	14	120	$\sim 10^{16}$	$\sim$ 100 PB
Cr_2	Ahlrichs	48	84	$\sim 10^{22}$	-

Solution: Select a part of configurations!

Related Work and Our Contribution

- Related work:
  - Configuration Interaction by Perturbatively Selecting Iteration (CIPSI)
  - Adaptive Configuration Interaction (ACI)
  - Adaptive Sampling Configuration Interaction (ASCI)
  - Heat-Bath Configuration Interaction (HCI)
  - Semistochastic HCI (SHCI)
- Our contribution:
  - Performs configuration selection using coordinate descent
  - Visits important determinants efficiently
  - Captures the significant part of FCI space for ground state approximation

Coordinate Descent FCI (CDFCI)

FCI eigenvalue problem to unconstrained minimization problem

$$\min_{\mathbf{c}\in\mathbb{R}^{N_{\text{FCI}}}} f(\mathbf{c}) = \min_{\mathbf{c}} \|H + \mathbf{c}\mathbf{c}^{\mathsf{T}}\|_F^2$$

- The only two local minimizers are $\pm\sqrt{E_0}\mathbf{v_0}$.
- Ensures convergence to the ground state wavefunction, given a good starting point (Hartree–Fock ground state).

Coordinate gradient descent method

- Minimizes computational costs by avoiding operations with the entire Hamiltonian matrix.
- In each iteration, only *one coordinate* of the optimizing vector is updated.
- Computation for updating involves only *one column* of the Hamiltonian matrix.

CDFCI Framework

Store $\mathbf{c}$ and $\mathbf{b} = H\mathbf{c}$ in memory.
Initialize $\mathbf{c}^{(0)}$, $\mathbf{b}^{(0)} = H\mathbf{c}^{(0)}$.
For iteration $\ell = 1, 2, \ldots$

1. Select coordinate $i^{(\ell)} = \arg\max_i |\nabla_i f(\mathbf{c}^{(\ell-1)})|$.
2. Find stepsize by exact line search $\alpha^{(\ell)} = \arg\min_\alpha f(\mathbf{c}^{(\ell-1)} + \alpha \mathbf{e}_{i^{(\ell)}})$.
3. Update $\mathbf{c}^{(\ell)} = \mathbf{c}^{(\ell-1)} + \alpha^{(\ell)} \mathbf{e}_{i^{(\ell)}}$, $\mathbf{b}^{(\ell)} = \mathbf{b}^{(\ell-1)} + \alpha^{(\ell)} H_{:,i^{(\ell)}}$.

Remark: Gradient
$\nabla f(\mathbf{c}) = 4H\mathbf{c} + 4\mathbf{c}^\top \mathbf{c}\mathbf{c} = 4\mathbf{b} + 4\mathbf{c}^\top \mathbf{c}\mathbf{c}$.

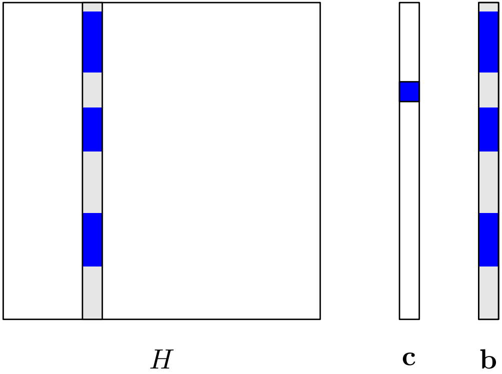


Figure: Update for one coordinate.

CDFCI Framework - for Two Coordinates?

Initialize $\mathbf{c}^{(0)}$, $\mathbf{b}^{(0)} = H\mathbf{c}^{(0)}$.

For iteration $\ell = 1, 2, \ldots$

1. Select coordinate
   $i^{(\ell)} = \arg\max_i |\nabla_i f(\mathbf{c}^{(\ell-1)})|$,
   $j^{(\ell)} = \arg\max_{j \neq i^{(\ell)}} |\nabla_j f(\mathbf{c}^{(\ell-1)})|$.
2. Find stepsize
   $\alpha^{(\ell)} = \arg\min_\alpha f(\mathbf{c}^{(\ell-1)} + \alpha \mathbf{e}_{i^{(\ell)}})$,
   $\beta^{(\ell)} = \arg\min_\beta f(\mathbf{c}^{(\ell-1)} + \beta \mathbf{e}_{j^{(\ell)}})$.
3. Update
   $\mathbf{c}^{(\ell)} = \mathbf{c}^{(\ell-1)} + \alpha^{(\ell)} \mathbf{e}_{i^{(\ell)}} + \beta^{(\ell)} \mathbf{e}_{j^{(\ell)}}$,
   $\mathbf{b}^{(\ell)} = \mathbf{b}^{(\ell-1)} + \alpha^{(\ell)} H_{:,i^{(\ell)}} + \beta^{(\ell)} H_{:,j^{(\ell)}}$.

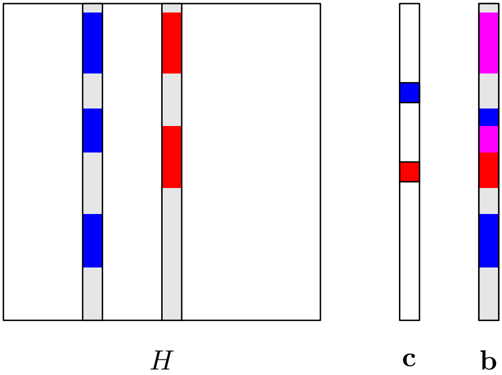


Figure: Update for two coordinates.

CDFCI Framework - Exact Line Search?

Initialize $\mathbf{c}^{(0)}$, $\mathbf{b}^{(0)} = H\mathbf{c}^{(0)}$.

For iteration $\ell = 1, 2, \ldots$

1. Select coordinate
   $i^{(\ell)} = \arg\max_i |\nabla_i f(\mathbf{c}^{(\ell-1)})|$,
   $j^{(\ell)} = \arg\max_{j \neq i^{(\ell)}} |\nabla_j f(\mathbf{c}^{(\ell-1)})|$.
2. Find stepsize $\alpha^{(\ell)}, \beta^{(\ell)} = \arg\min_{\alpha,\beta} f(\mathbf{c}^{(\ell-1)} + \alpha \mathbf{e}_{i^{(\ell)}} + \beta \mathbf{e}_{j^{(\ell)}})$.
3. Update
   $\mathbf{c}^{(\ell)} = \mathbf{c}^{(\ell-1)} + \alpha^{(\ell)} \mathbf{e}_{i^{(\ell)}} + \beta^{(\ell)} \mathbf{e}_{j^{(\ell)}}$,
   $\mathbf{b}^{(\ell)} = \mathbf{b}^{(\ell-1)} + \alpha^{(\ell)} H_{:,i^{(\ell)}} + \beta^{(\ell)} H_{:,j^{(\ell)}}$.

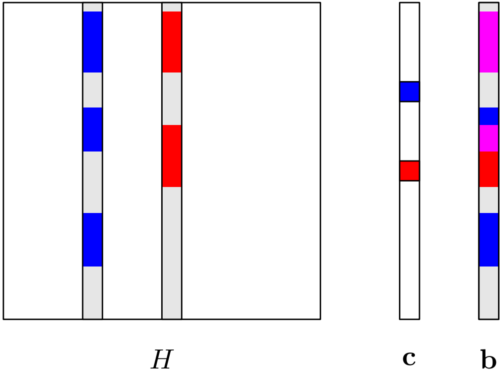


Figure: Update for two coordinates.

Add a Scalar γ for Exact Line Search

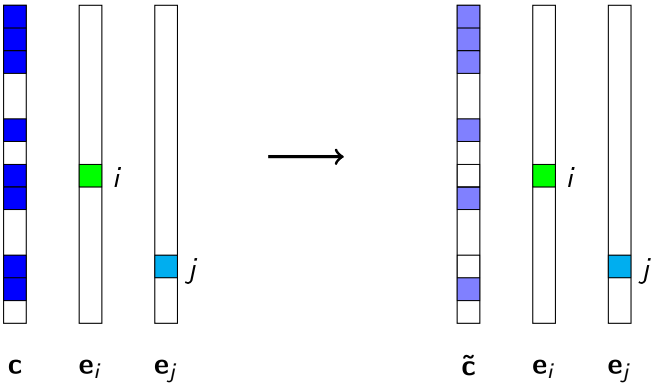
Modify the minimization problem from $\min_{\alpha,\beta} f(\mathbf{c} + \alpha \mathbf{e}_i + \beta \mathbf{e}_j)$ to

$$\min_{\gamma,\alpha,\beta} f(\gamma \mathbf{c} + \alpha \mathbf{e}_i + \beta \mathbf{e}_j) = f\left(\begin{bmatrix} \mathbf{c} & \mathbf{e}_i & \mathbf{e}_j \end{bmatrix} \begin{bmatrix} \gamma \\ \alpha \\ \beta \end{bmatrix}\right)$$

$$= \left\| H + \begin{bmatrix} \mathbf{c} & \mathbf{e}_i & \mathbf{e}_j \end{bmatrix} \begin{bmatrix} \gamma \\ \alpha \\ \beta \end{bmatrix} \begin{bmatrix} \gamma & \alpha & \beta \end{bmatrix} \begin{bmatrix} \mathbf{c}^T \\ \mathbf{e}_i^T \\ \mathbf{e}_j^T \end{bmatrix} \right\|_F^2 .$$

Matrix Orthogonalization

Construct $\begin{bmatrix} \tilde{\mathbf{c}} & \mathbf{e}_i & \mathbf{e}_j \end{bmatrix}$, where $\|\tilde{\mathbf{c}}\|_2 = 1$, $(\tilde{\mathbf{c}}, \mathbf{e}_i) = 0$, $(\tilde{\mathbf{c}}, \mathbf{e}_j) = 0$.



Add γ and $\tilde{\mathbf{c}}$ for Exact Line Search

Modify the minimization problem from $\min_{\alpha,\beta} f(\mathbf{c} + \alpha\mathbf{e}_i + \beta\mathbf{e}_j)$ to

$$\begin{aligned}
\min_{\gamma,\alpha,\beta} f(\gamma\tilde{\mathbf{c}} + \alpha\mathbf{e}_i + \beta\mathbf{e}_j) &= f\left(\begin{bmatrix}\tilde{\mathbf{c}} & \mathbf{e}_i & \mathbf{e}_j\end{bmatrix}\begin{bmatrix}\gamma\\ \alpha\\ \beta\end{bmatrix}\right)\\
&= \left\| H + \begin{bmatrix}\tilde{\mathbf{c}} & \mathbf{e}_i & \mathbf{e}_j\end{bmatrix}\begin{bmatrix}\gamma\\ \alpha\\ \beta\end{bmatrix}\begin{bmatrix}\gamma & \alpha & \beta\end{bmatrix}\begin{bmatrix}\tilde{\mathbf{c}}^{\mathsf{T}}\\ \mathbf{e}_i^{\mathsf{T}}\\ \mathbf{e}_j^{\mathsf{T}}\end{bmatrix}\right\|_F^2\\
&= \left\| \underbrace{\begin{bmatrix}\tilde{\mathbf{c}}^{\mathsf{T}}\\ \mathbf{e}_i^{\mathsf{T}}\\ \mathbf{e}_j^{\mathsf{T}}\end{bmatrix} H \begin{bmatrix}\tilde{\mathbf{c}} & \mathbf{e}_i & \mathbf{e}_j\end{bmatrix}}_{\in \mathbb{R}^{3\times 3}} + \begin{bmatrix}\gamma\\ \alpha\\ \beta\end{bmatrix}\begin{bmatrix}\gamma & \alpha & \beta\end{bmatrix}\right\|_F^2 .
\end{aligned}$$

Extension to Multi Coordinate Descent FCI

- Select a set of coordinates $I = \{i_1, \ldots, i_k\}, 1 \leq i_j \leq N_{\mathrm{FCI}}$ based on gradient $\nabla f(\mathbf{c}) = 4H\mathbf{c} + 4\mathbf{c}^{\mathrm{T}}\mathbf{c}\mathbf{c}$.
- Denote $\mathcal{E}_I = [e_{i_1}, \ldots, e_{i_k}] \in \mathbb{R}^{N_{\mathrm{FCI}} \times k}$.
- The update is given by

$$\mathbf{c} \leftarrow \gamma\mathbf{c} + \mathcal{E}_I\mathbf{a}.$$

- The values of γ and $\mathbf{a}$ are given by the eigenvector of

$$\begin{bmatrix} \tilde{\mathbf{c}}^{\mathrm{T}} \\ \mathcal{E}_I^{\mathrm{T}} \end{bmatrix} H \begin{bmatrix} \tilde{\mathbf{c}} & \mathcal{E}_I \end{bmatrix} \in \mathbb{R}^{(k+1)\times(k+1)}$$

  corresponding to the minimal eigenvalue $\lambda_{\min}$, which is the current energy estimate.

Implementation Details

- Each thread updates one coordinate: $\mathbf{\Delta b} = H_{:,j} a_j$.
- **c** and **b** are stored in a hash table that allows parallel read-write operations.
- The scaling factor γ is multiplied onto the scale of **c** at each iteration.

Scalability

Table: Speedup of mCDFCI for different number of coordinates (threads) of H_2O in cc-pVDZ basis

		Wall Time (s)	Speedup on k Cores					
Energy	Error	on Single Core	2	4	8	16	32	64
-76.2318601	10^{-2}	9.0	1.9×	3.7×	5.0×	7.0×	10.4×	13.1×
-76.2408601	10^{-3}	292.9	2.0×	3.7×	4.8×	8.2×	14.3×	20.3×
-76.2417601	10^{-4}	1837.6	2.0×	3.5×	6.1×	8.3×	16.2×	22.5×
-76.2418501	10^{-5}	9016.7	2.1×	4.1×	8.0×	12.3×	21.1×	29.0×
-76.2418591	10^{-6}	32931.7	2.1×	4.5×	9.3×	16.2×	29.1×	40.7×

Overall Speedup

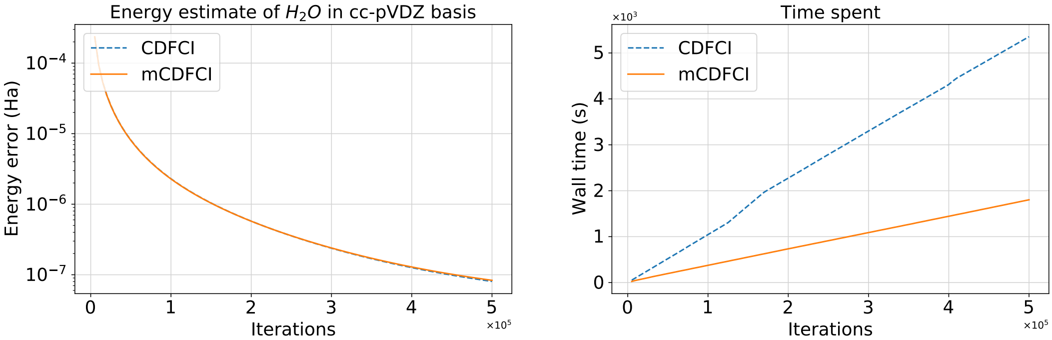


Figure: Speedup of mCDFCI compared with CDFCI(2019), both in 64 threads. We perform k coordinates ($k = 64$) descent per iteration for the original CDFCI.

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Thanks for Your Attention!