Wave Function Optimization Algorithms in QMCPACK

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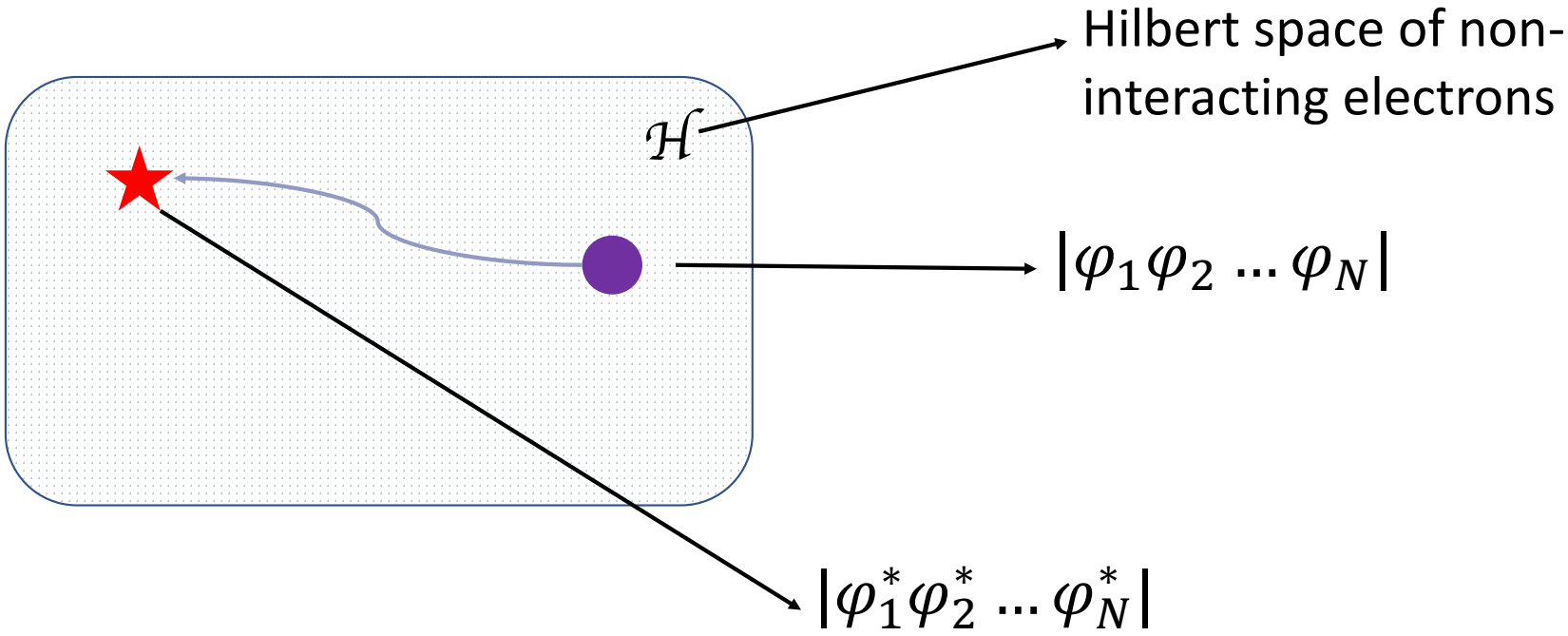
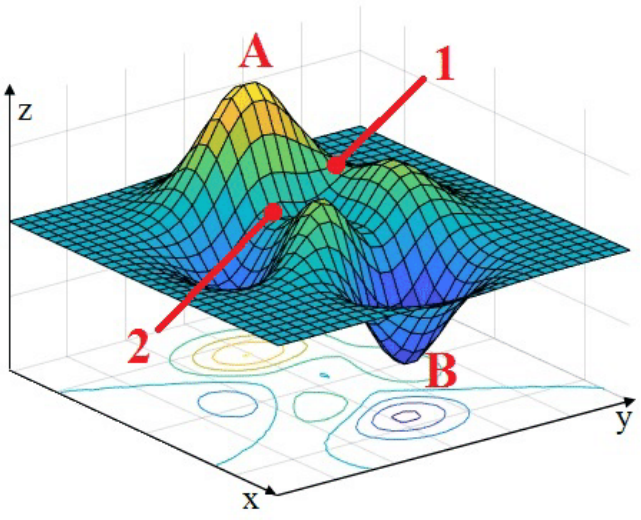
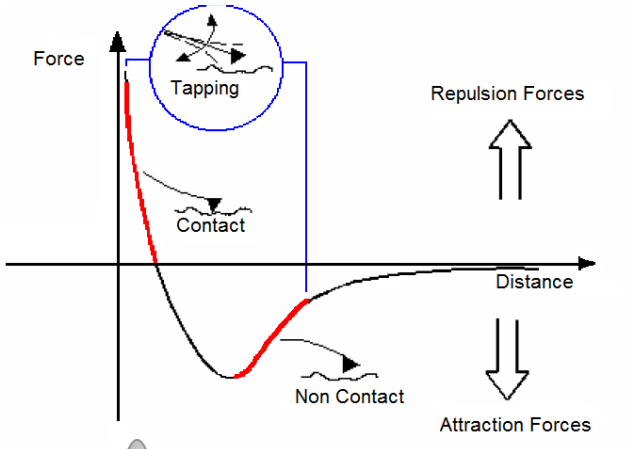
The Motivation of Wave Function Optimization



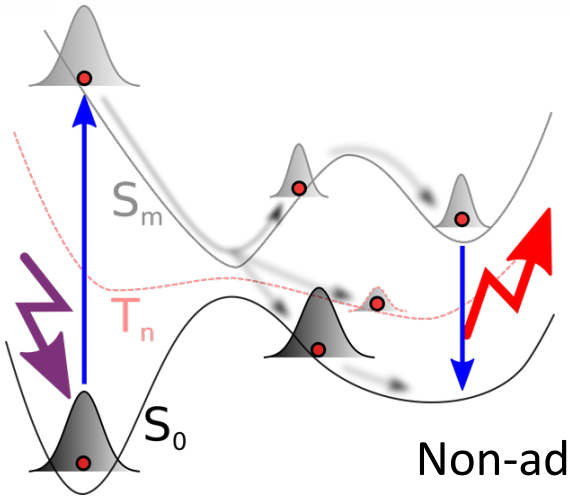
What can we compute with better wave functions?



Potential Energy Surfaces: Barrier Height, Excitation Energy, Reaction Rate



Forces

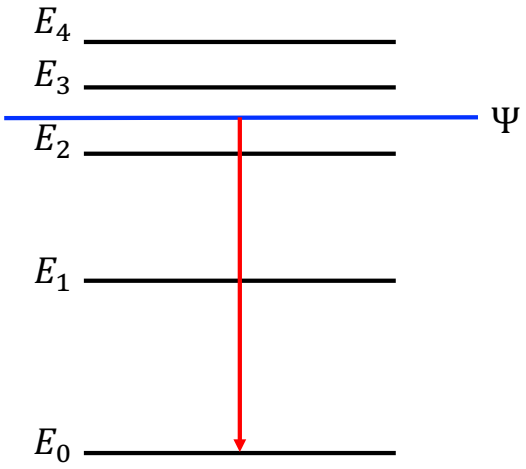


Non-adiabatic Dynamics

Variational Principle For Ground and Excited State

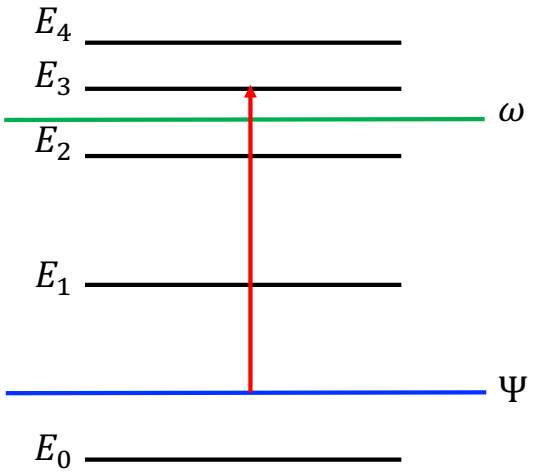
Ground State

$$E[\Psi] = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle} \geq E_0$$



Excited State

$$\Omega[\Psi] = \frac{\langle \Psi | \omega - H | \Psi \rangle}{\langle \Psi | (\omega - H)^2 | \Psi \rangle}$$



Optimization Method in QMCPACK: The Linear Method

Wave Function with Parameters $\vec{p}$: $\Psi(\vec{p})$

First Order Taylor Expansion around $\vec{p}$:

$$\Psi(\vec{p}+\Delta\vec{p}) = \Psi(\vec{p}) + \sum_{i=1} \frac{\partial \Psi}{\partial p_i} \Delta p_i = \sum_{i=0} \frac{\partial \Psi}{\partial p_i} \Delta p_i$$

Plug $\Psi(\vec{p}+\Delta\vec{p})$ in $E[\Psi]$ or $\Omega[\Psi]$:

$$E[\Psi(\vec{p}+\Delta\vec{p})] = \frac{\sum_{ij} \left\langle \frac{\partial \Psi}{\partial p_i} \middle| H \middle| \frac{\partial \Psi}{\partial p_j} \right\rangle}{\sum_{ij} \left\langle \frac{\partial \Psi}{\partial p_i} \middle| \frac{\partial \Psi}{\partial p_j} \right\rangle} \qquad \Omega[\Psi(\vec{p}+\Delta\vec{p})] = \frac{\sum_{ij} \left\langle \frac{\partial \Psi}{\partial p_i} \middle| \omega - H \middle| \frac{\partial \Psi}{\partial p_j} \right\rangle}{\sum_{ij} \left\langle \frac{\partial \Psi}{\partial p_i} \middle| (\omega - H)^2 \middle| \frac{\partial \Psi}{\partial p_j} \right\rangle}$$

Optimization Method in QMCPACK: The Linear Method

Taking derivatives w.r.t. $\Delta\vec{p}$ and set the derivatives to 0:

$$\sum_j \left\langle \frac{\partial\Psi}{\partial p_i} \middle| H \middle| \frac{\partial\Psi}{\partial p_j} \right\rangle \Delta p_j = E \sum_j \left\langle \frac{\partial\Psi}{\partial p_i} \middle| \frac{\partial\Psi}{\partial p_j} \right\rangle \Delta p_j$$

$$\sum_j \left\langle \frac{\partial\Psi}{\partial p_i} \middle| \omega - H \middle| \frac{\partial\Psi}{\partial p_j} \right\rangle \Delta p_j = \Omega \sum_j \left\langle \frac{\partial\Psi}{\partial p_i} \middle| (\omega - H)^2 \middle| \frac{\partial\Psi}{\partial p_j} \right\rangle \Delta p_j$$

Both equations are in the form of a generalized eigenvalue problem:

$$\tilde{H}\Delta\vec{p} = \lambda\tilde{S}\Delta\vec{p}$$

Optimization Method in QMCPACK: The Linear Method

Perform Inverse Taylor expansion:

$$p_i = p_i + \Delta p_i$$

1. Build $\tilde{H}$ and $\tilde{S}$ matrices.
2. Solve the Generalized Eigenvalue Problem.
3. Update Wave Function Parameters.
4. If not converged, go to step 1.

One Step in LM

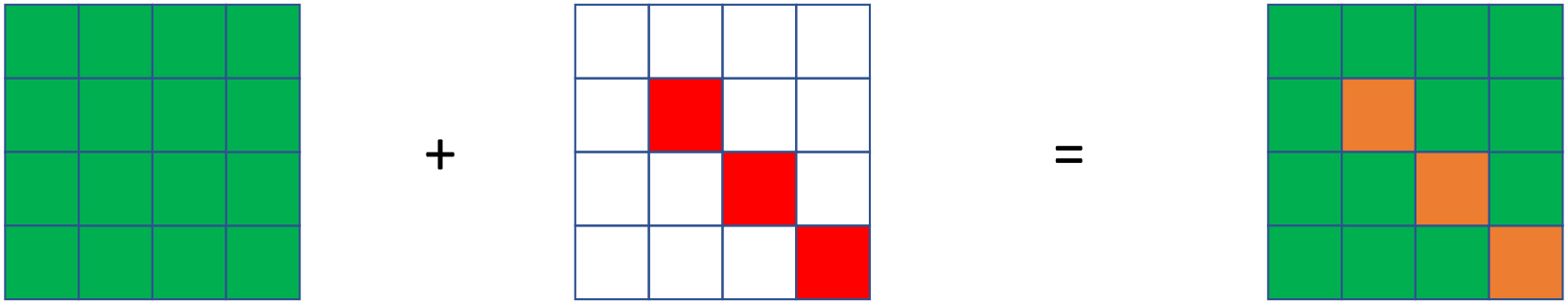
Stabilizing Linear Method: Step Size Control

Modify the Hamiltonian:

$$\widetilde{H} = \widetilde{H} + \alpha A + \beta B$$

Diagonal Shift Matrix A:

$$A_{xy} = \delta_{xy}(1 - \delta_{x0})$$



Stabilizing Linear Method: Step Size Control

Projection Matrix Q: Project the current wave function out

$$Q_{xy} = \delta_{xy} - \delta_{x0}(1 - \delta_{y0})S_{0y}$$

Shift Matrix T: Penalize directions with larger norms

$$T_{xy} = (1 - \delta_{x0}\delta_{y0})[Q^{\dagger}SQ]_{xy}$$

Shift Matrix B: Transform back to the original basis

$$B = \left(Q^{\dagger}\right)^{-1} T Q^{-1}$$

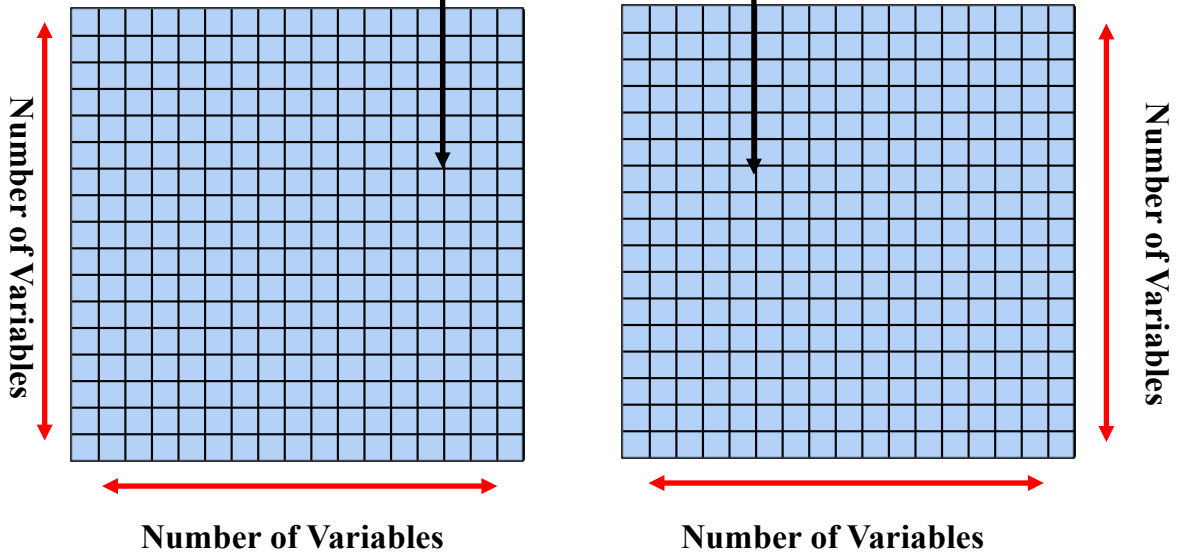
How to Choose the Shift Value?

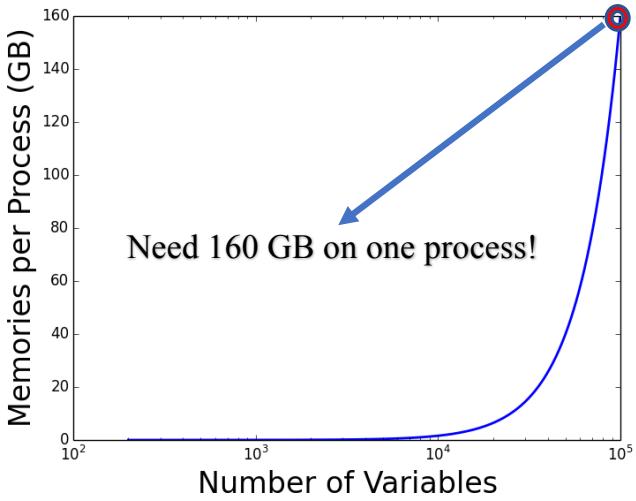
adaptive:

1. Set initial α and β
2. Small shifts: $\alpha/4$, $\beta/4$, Medium Shifts: $\alpha/4$, $\beta/4$, Large Shifts: 4α, 4β
3. Solve the generalized eigenvalue problem for these three sets of shifts.
4. Use correlated sampling to compare the updated wave function based on three shifts
5. Small shifts wins: lower the shifts Medium shifts wins: no change to the shifts Large shifts wins: increase the shifts

Memory Bottleneck of LM

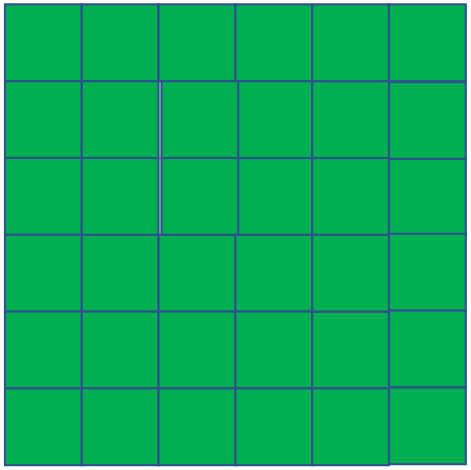
$$\sum_{j=1}^{Nv} \tilde{H}_{ij}\Delta p_j = E \sum_{j=1}^{Nv} \tilde{S}_{ij}\Delta p_j$$

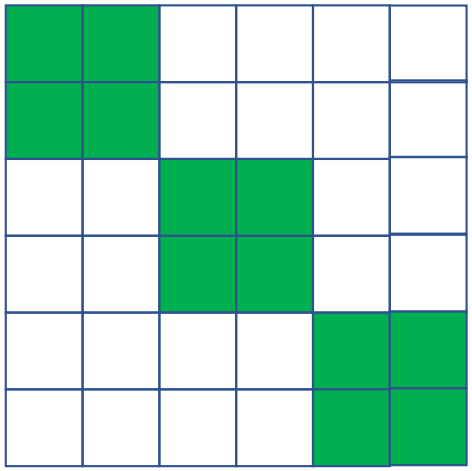




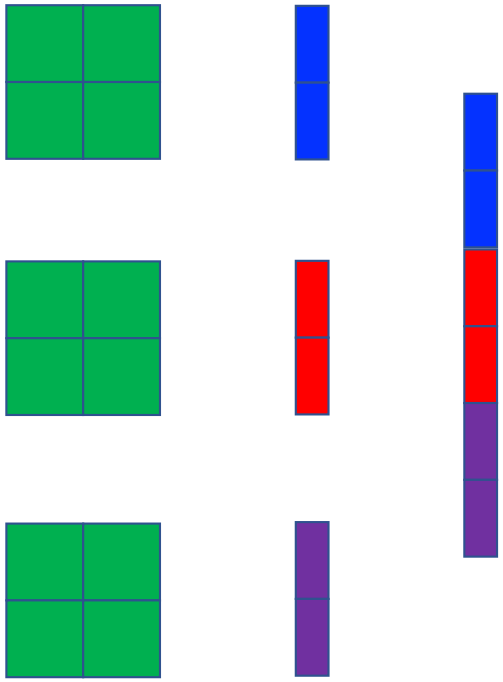
A few GB of memory

Blocked LM: A Crude Idea

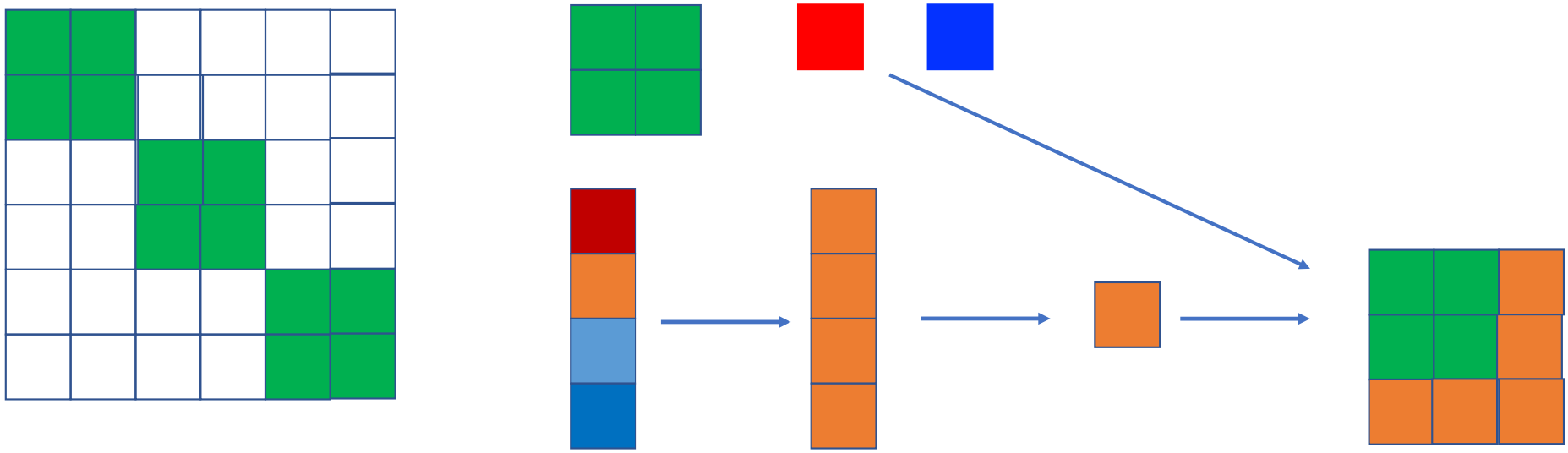




Divide Variables into Blocks

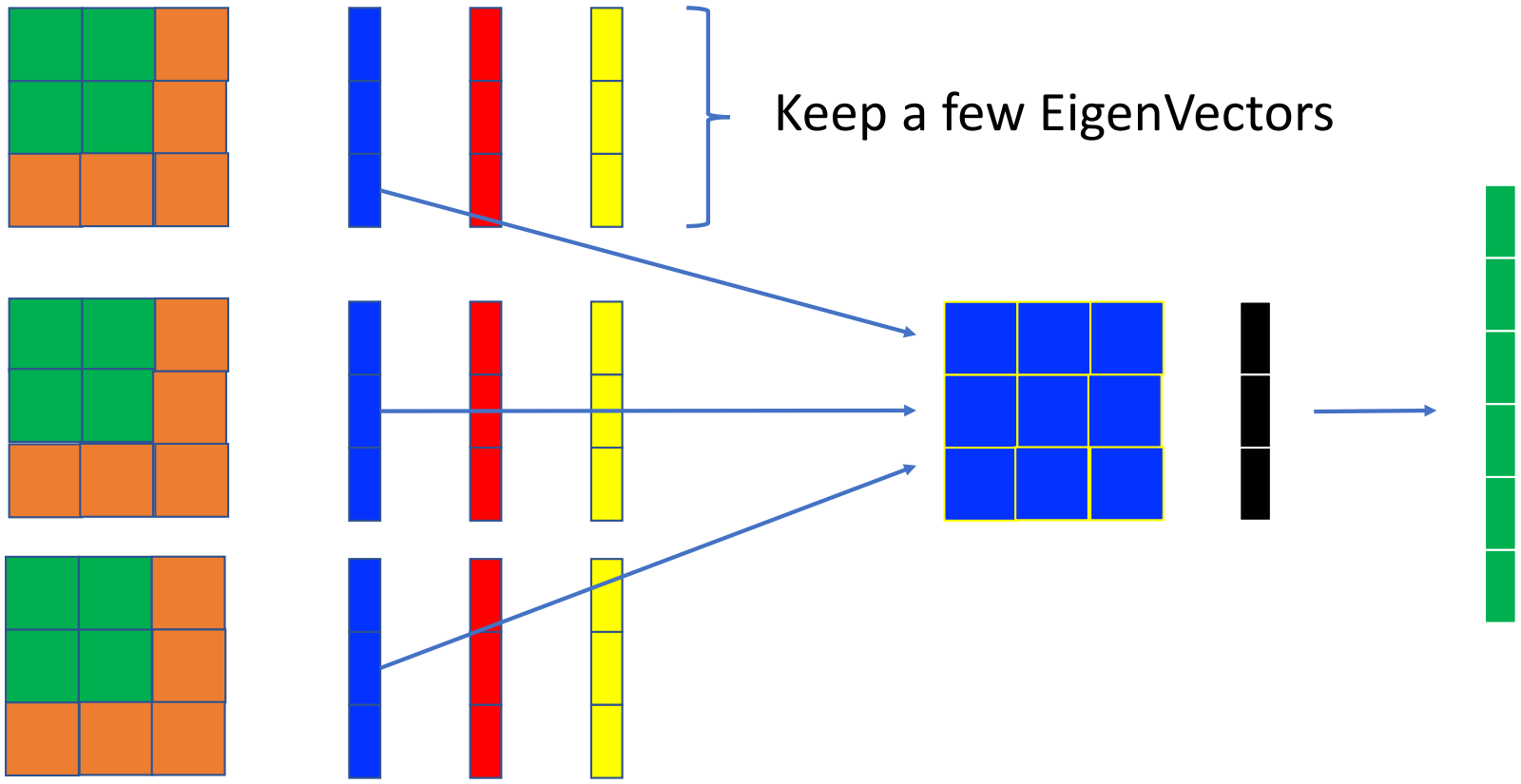


Blocked LM: Account for Inter-Block Coupling



Old Update Directions

Blocked LM: Account for Inter-Block Coupling



How to use LM/BLM in QMCPACK

```
<loop max="10">
    <qmc method="linear" move="pbyp" checkpoint="-1" gpu="no">
      <parameter name="MinMethod"> adaptive </parameter>
      <parameter name="targetExcited"> yes </parameter>
      <parameter name="omega"> -17.779  </parameter>
       …….
    </qmc>
</loop>
```

How many steps you want to run

Specify linear method

Parameters of LM in QMCPACK

1. targetExcited: yes/no: whether to do target function/energy minimization
2. omega: real number: Energy shift used to target different excited states
3. shift_i: 1.0: value of the identity shift
4. shift_s: 1.0: value of the overlap shift
5. max_param_change: 0.3: the maximum allowed parameter update
6. chase_lowest: yes/no: whether to chase the lowest eigenvector in iterative solver
7. chase_closest: yes/no: whether to chase the eigenvector that is closest to the initial guess

Parameters of BLM in QMCPACK

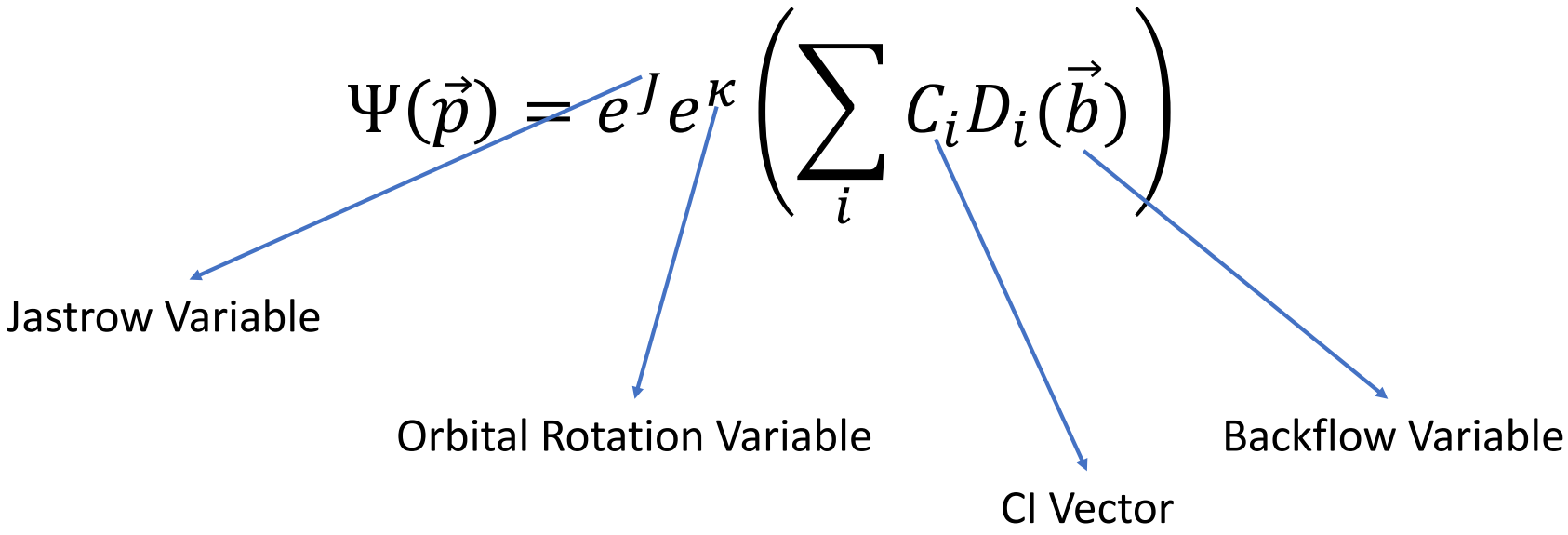
1. block_lm: yes/no: whether to switch to BLM
2. nblocks: integer: number of blocks in BLM
3. nkept: integer: number of eigenvectors to keep per block in BLM
4. nolds: integer: number of old update vectors used in BLM

Rule of Thumb: BLM should be turned on when the number of parameters > 5000.

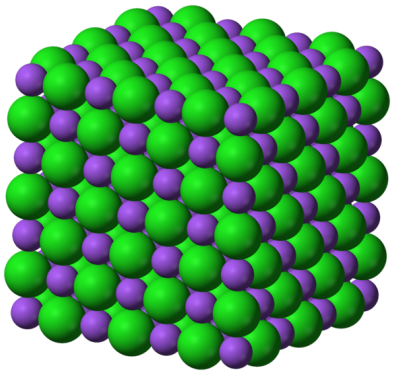
Usually a few hundred blocks and a handful of nkept and nolds are sufficient to balance memory and accuracy.

Type of Parameters that LM/BLM can optimize

Everything!



Upcoming Optimization Features in QMCPACK



Complex Wave Function Parameters: Solid State

$$\Psi_G^2(R) = |\Psi(R)|^2 + \varepsilon \sum_i \Psi_i^2(R)$$

$$\Psi_G^2(R) = |\Psi(R)|^2 + \varepsilon |\nabla^2 \Psi(R)|$$

Guiding Function other than $\Psi(R)$

Examples in the Hands on Session

1. Energy Optimization of all-electron H_4 with Jastrow Multi-Slater Wave Function and Orbital Optimization.
2. Excited State Optimization of H_2O with Jastrow Multi-Slater Wave Function and Orbital Optimization.
3. Blocked LM Optimization of C_2.

Acknowledgement

- Eric Neuscamman
- Graduate students and postdocs in Neuscamman group
- Collaborators in CPSFM

QMCPACK

CPSFM