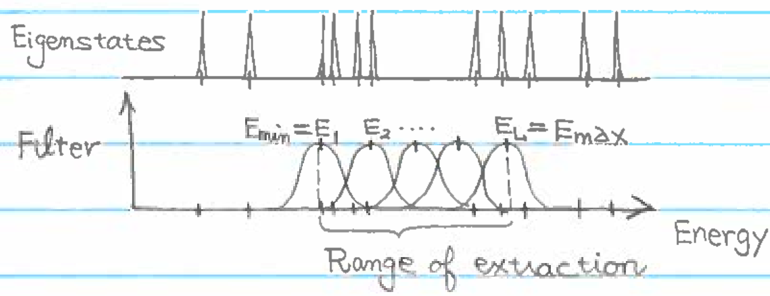
Filter Diagonalization

5/27/03

[M.R. Wall & D. Neuhauser, J. Chem. Phys. 102, 8011 (195)]

— O(N) extraction of eigenvalues

① Filter (overlapping energy windows)
② Disentangle (small diagonalization within each filter)



— Let $|\psi_0\rangle$ be a random initial wave function and $\hat{H}$ a single-electron Hamiltonian. We define the correlation function as

$$C(t) \equiv \langle \psi_0 | e^{-i\hat{H}t/\hbar} | \psi_0 \rangle \equiv \langle \psi_0 | \psi(t) \rangle \qquad (1)$$

$C(t)$ is calculated only once ($O(N) \propto$ # of mesh points) for $t \in [0, T_{max}]$ and extended to $t \in [-T_{max}, T_{max}]$ by the relation,

$$C(-t) = \langle \psi_0 | e^{i\hat{H}t/\hbar} | \psi_0 \rangle$$

$$= (\langle \psi_0 | e^{-i\hat{H}t/\hbar} | \psi_0 \rangle)^* = C^*(t) \qquad (2)$$

– Filter wave packets

$$\begin{cases} |\Phi(E_\ell)\rangle \equiv \displaystyle\int_{-\infty}^{\infty} dt\, w(t;E_\ell)\, |\psi(t)\rangle \qquad (\ell = 1, 2, \ldots, L) & (3) \\[2ex] w(t;E_\ell) = e^{-t^2/2T^2}\, e^{iE_\ell t/\hbar} & (4) \end{cases}$$

We define $E_{min} = E_1 < E_2 < \cdots < E_{L-1} < E_L = E_{max}$ and the range of extraction is $[E_{min}, E_{max}]$. The number of wave packets L must be chosen such that $L > M$, where M is the number of eigen states of $\hat{H}$ in $[E_{min}, E_{max}]$.
For a fixed $[E_{min}, E_{max}]$, M, and hence L, are $O(N)$; this will be partially remedied by the "overlapping extraction range" scheme later.

Note that

$$|\Phi(E_\ell)\rangle = \int_{-\infty}^{\infty} dt\, \exp\Big(\underbrace{-\frac{t^2}{2T^2} + i\frac{E_\ell t}{\hbar} - i\frac{\hat{H}t}{\hbar}}\Big)\, |\psi_0\rangle$$

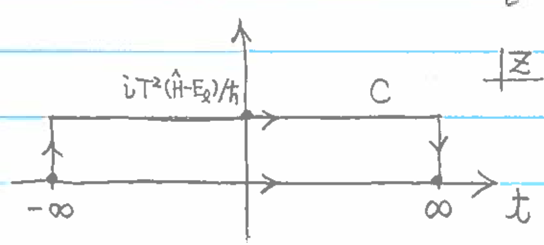
$$-\frac{t^2}{2T^2} - \frac{i}{\hbar}(\hat{H} - E_\ell)t$$

$$= -\frac{1}{2T^2}\left[t^2 + \frac{i2T^2}{\hbar}(\hat{H} - E_\ell)t\right]$$

$$= -\frac{1}{2T^2}\left\{\left[t + \frac{iT^2}{\hbar}(\hat{H} - E_\ell)\right]^2 + \frac{T^4}{\hbar^2}(\hat{H} - E_\ell)^2\right\}$$

$$= -\frac{1}{2T^2}\left[t + \frac{iT^2}{\hbar}(\hat{H} - E_\ell)\right]^2 - \frac{T^2}{2\hbar^2}(\hat{H} - E_\ell)^2$$

$$= \exp\left[-\frac{T^2}{2\hbar^2}(\hat{H} - E_\ell)^2\right] \int_{-\infty}^{\infty} dt\, \exp\Big\{-\frac{1}{2T^2}\Big[\underbrace{t + \frac{iT^2}{\hbar}(\hat{H} - E_\ell)}_{t'}\Big]^2\Big\}\, |\psi_0\rangle$$



By changing the integration path as shown above,

$$|\Psi(E_\ell)\rangle = \exp\left[-\frac{T^2}{2\hbar^2}(\hat{H}-E_\ell)^2\right] \underbrace{\int_{-\infty}^{\infty} dt' \exp\left(-\frac{t'^2}{2T^2}\right)}_{t'=\sqrt{2}Tx} |\psi_0\rangle$$

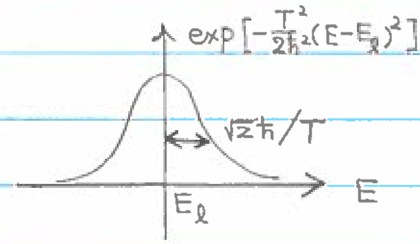
$$= \int_{-\infty}^{\infty} \sqrt{2}\, T dx\, e^{-x^2}$$

$$= \sqrt{2}\, T \underbrace{\int_{-\infty}^{\infty} dx\, e^{-x^2}}_{\sqrt{\pi}}$$

$$= \sqrt{2\pi}\, T$$

$$\therefore |\Psi(E_\ell)\rangle = \sqrt{2\pi}\, T \exp\left[-\frac{T^2}{2\hbar^2}(\hat{H}-E_\ell)^2\right] |\psi_0\rangle \qquad (5)$$

Thus $|\Psi(E_\ell)\rangle$ is a linear combination of eigen states in the range $E_\ell \pm \sqrt{2}\hbar/T$.



– Disentangle the wave packets

Let $|\phi_m\rangle$ be the eigen states of $\hat{H}$ with the eigenvalues ϵ_m:

$$\hat{H}|\phi_m\rangle = \epsilon_m|\phi_m\rangle \tag{6}$$

We wish to construct all $|\phi_m\rangle$ $(m=1,2,\ldots,M)$, such that $\epsilon_m \in [E_1, E_L]$, as a linear combination of $|\Psi(E_l)\rangle$ $(l=1,2,\ldots,L)$. It is necessary that $L \geq M$ for this to be possible.

(Generalized eigen value problem)

Let

$$|\phi_m\rangle = \sum_{l=1}^{L} b_l^{(m)} |\Psi(E_l)\rangle \quad (m=1,\ldots,M) \tag{7}$$

Substituting Eq. (7) in (6),

$$\hat{H}\sum_{l'=1}^{L} b_{l'}^{(m)} |\Psi(E_{l'})\rangle = \epsilon_m \sum_{l'=1}^{L} b_{l'}^{(m)} |\Psi(E_{l'})\rangle \tag{8}$$

$\langle\Psi(E_l)| \times$ Eq. (8)

$$\sum_{l'=1}^{L} \underbrace{\langle\Psi(E_l)|\hat{H}|\Psi(E_{l'})\rangle}_{H_{ll'}} \underbrace{b_{l'}^{(m)}}_{B_{l'm}} = \sum_{l'=1}^{L} \underbrace{\langle\Psi(E_l)|\Psi(E_{l'})\rangle}_{S_{ll'}} b_{l'}^{(m)} \epsilon_m$$

$$= \sum_{l'=1}^{L} \sum_{m'=1}^{M} S_{ll'} \underbrace{b_{l'}^{(m')}}_{B_{l'm'}} \underbrace{(\delta_{m'm}\epsilon_m)}_{\epsilon_{m'm}}$$

※ Note that $|\Psi(E_l)\rangle$ are non-orthogonal.

The generalized eigenvalue problem is thus.

$$\underset{L\times L}{\mathbb{H}}\ \underset{L\times M}{\mathbb{B}} = \underset{L\times L}{\mathbb{S}}\ \underset{L\times M}{\mathbb{B}}\ \underset{M\times M}{\mathbb{E}} \tag{9}$$

where

$$\begin{cases} H_{\ell\ell'} = \langle \Psi(E_\ell) | \hat{H} | \Psi(E_{\ell'}) \rangle & (\ell, \ell' = 1, \dots, L) \quad (10) \\ S_{\ell\ell'} = \langle \Psi(E_\ell) | \Psi(E_{\ell'}) \rangle & (\ell, \ell' = 1, \dots, L) \quad (11) \\ B_{\ell m} = b_\ell^{(m)} & (\ell = 1, \dots, L;\ m = 1, \dots, M) \quad (12) \\ E_{mm'} = \delta_{mm'} E_{m'} & (m, m' = 1, \dots, M) \quad (13) \end{cases}$$

The problem is :

① Find the number of eigen states, M, in the range $[E_1, E_L]$.
(This is also the number of linearly independent states in the space spanned by $|\Psi(E_\ell)\rangle$ $(\ell = 1, \dots, L)$, i.e., the rank of $S_{\ell\ell'}$.)

② Solve Eq. (9) to obtain $\mathbb{B}$ and $\mathbb{E}$.

These problems are solved by the singular value decomposition (SVD) of $S_{\ell\ell'}$.

— Singular value decomposition

(Prop) $S_{\ell\ell'}$ is Hermitian.

☺ $(S^{\dagger})_{\ell\ell'} = (S_{\ell'\ell})^{*} = (\langle \Psi(E_{\ell'}) | \Psi(E_{\ell}) \rangle)^{*} = \langle \Psi(E_{\ell}) | \Psi(E_{\ell'}) \rangle$
$= S_{\ell\ell'}$ //

Let $u_{\ell}^{(n)}$ be eigen states of $S_{\ell\ell'}$ with eigenvalues λ_m:

$$\sum_{\ell'=1}^{L} S_{\ell\ell'} u_{\ell'}^{(n)} = \lambda_n u_{\ell}^{(n)} \quad (n=1,\dots,L) \qquad (14)$$

(Prop) ① λ_m are real.
② $\lambda_m \geq \emptyset$.

☺① $\sum_{\ell=1}^{L} (u_{\ell}^{(n)})^{*} \times$ Eq. (14)

$$\underbrace{\sum_{\ell=1}^{L}\sum_{\ell'=1}^{L} (u_{\ell}^{(n)})^{*} S_{\ell\ell'} u_{\ell'}^{(n)}}_{a} = \lambda_n \underbrace{\sum_{\ell=1}^{L} |u_{\ell}^{(n)}|^2}_{real}$$

$$a^{*} = \sum_{\ell}\sum_{\ell'} u_{\ell}^{(n)} \underbrace{(S_{\ell\ell'})^{*}}_{(S^{\dagger})_{\ell'\ell} = S_{\ell'\ell} \ (☺ \text{ Hermitian})} (u_{\ell'}^{(n)})^{*} = \sum_{\ell'}\sum_{\ell} (u_{\ell'}^{(n)})^{*} S_{\ell'\ell} u_{\ell}^{(n)} = a$$

$\therefore$ a is real, and hence λ_n is real.

☺②

$$a = \sum_{\ell=1}^{L}\sum_{\ell'=1}^{L} (u_{\ell}^{(n)})^{*} \langle \Psi(E_{\ell}) | \Psi(E_{\ell'}) \rangle u_{\ell'}^{(n)}$$

$$= \underbrace{\left\{ \sum_{\ell=1}^{L} \langle \Psi(E_{\ell}) | (u_{\ell}^{(n)})^{*} \right\}}_{\langle \beta |} \underbrace{\left\{ \sum_{\ell'=1}^{L} u_{\ell'}^{(n)} | \Psi(E_{\ell'}) \rangle \right\}}_{| \beta \rangle}$$

$$= \langle \beta | \beta \rangle \geq \emptyset$$

Since $\sum_{\ell=1}^{L} |u_{\ell}^{(n)}|^2 \geq 0$, this implies that $\lambda_n \geq \emptyset$. //

(Prop) $u_\ell^{(n)}$ $(n=1,\dots,L)$ may be made orthonormal:

$$\sum_{\ell=1}^{L} (u_\ell^{(n)})^* u_\ell^{(n')} = \delta_{nn'} \quad (15)$$

∵ (non-degenerate case: $\lambda_n \neq \lambda_{n'}$)

$$\sum_{\ell'=1}^{L} S_{\ell\ell'} u_{\ell'}^{(n)} = \lambda_n u_\ell^{(n)} \quad (16)$$

$\sum_{\ell=1}^{L} (u_\ell^{(n')})^* \times (16)$

$$\sum_\ell \sum_{\ell'} (u_\ell^{(n')})^* S_{\ell\ell'} u_{\ell'}^{(n)} = \lambda_n \sum_\ell (u_\ell^{(n')})^* u_\ell^{(n)} \quad (17)$$

$$\sum_{\ell'=1}^{L} S_{\ell\ell'} u_{\ell'}^{(n')} = \lambda_{n'} u_\ell^{(n')} \quad (18)$$

$\sum_{\ell=1}^{L} (u_\ell^{(n)})^* \times (18)$

$$\sum_\ell \sum_{\ell'} (u_\ell^{(n)})^* S_{\ell\ell'} u_{\ell'}^{(n')} = \lambda_{n'} \sum_\ell (u_\ell^{(n)})^* u_\ell^{(n')}$$

$\downarrow$ c.c.

$$\sum_\ell \sum_{\ell'} (u_{\ell'}^{(n')})^* \underbrace{(S_{\ell\ell'})^*}_{S^\dagger_{\ell'\ell} = S_{\ell'\ell}\ (\because \text{Hermitian})} u_\ell^{(n)} = \overbrace{\lambda_{n'}^*}^{\lambda_{n'}} \sum_\ell (u_\ell^{(n')})^* u_\ell^{(n)}$$

$$\therefore \sum_\ell \sum_{\ell'} (u_\ell^{(n')})^* S_{\ell\ell'} u_{\ell'}^{(n)} = \lambda_{n'} \sum_\ell (u_\ell^{(n')})^* u_\ell^{(n)} \quad (19)$$

Eq. (17) − (19)

$$\emptyset = (\lambda_n - \lambda_{n'}) \sum_\ell (u_\ell^{(n')})^* u_\ell^{(n)}$$

Since $\lambda_n - \lambda_{n'} \neq 0$ by assumption, $\sum_\ell (u_\ell^{(n')})^* u_\ell^{(n)} = 0$

∵ (degenerate case: $\lambda_n = \lambda_{n'} = \lambda$)

$$\sum_{\ell'} S_{\ell\ell'} u_{\ell'}^{(n)} = \lambda u_\ell^{(n)} \quad \times\ a$$

$$+)\ \sum_{\ell'} S_{\ell\ell'} u_{\ell'}^{(n')} = \lambda u_\ell^{(n')} \quad \times\ b$$

$$\sum_{\ell'} S_{\ell\ell'} (a u_{\ell'}^{(n)} + b u_{\ell'}^{(n')}) = \lambda (a u_{\ell'}^{(n)} + b u_{\ell'}^{(n')})$$

Thus, any linear combination of $u_\ell^{(n)}$ and $u_\ell^{(n')}$ in the rank-2 space is an eigen state of $S_{\ell\ell'}$ with the eigenvalue λ.
We can orthonormalize the two states by the Gram-Schmidt procedure.

Example: $S_{\ell\ell'} = \begin{pmatrix} 2 & 0 \\ 0 & 2 \end{pmatrix}$.

$$\begin{pmatrix} 2 & 0 \\ 0 & 2 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \lambda \begin{pmatrix} x \\ y \end{pmatrix}$$

$$\begin{pmatrix} \lambda-2 & 0 \\ 0 & \lambda-2 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$$

$$\begin{vmatrix} \lambda-2 & 0 \\ 0 & \lambda-2 \end{vmatrix} = (\lambda-2)^2 = 0 \longrightarrow \lambda = 2 \quad \text{(degenerate)}$$

$\begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}$ can be satisfied by any $\begin{pmatrix} x \\ y \end{pmatrix}$.

Let $|u_1\rangle = \begin{pmatrix} \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} \end{pmatrix}$ $|u_2\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$. By the Gram-Schmidt procedure.

$$|u_2'\rangle \leftarrow |u_2\rangle - |u_1\rangle\langle u_1|u_2\rangle$$
$$= \begin{pmatrix} 1 \\ 0 \end{pmatrix} - \frac{1}{\sqrt{2}}\begin{pmatrix} \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} \end{pmatrix} = \begin{pmatrix} \frac{1}{2} \\ -\frac{1}{2} \end{pmatrix}$$

$$|u_2''\rangle \leftarrow \frac{|u_2'\rangle}{\sqrt{\langle u_2'|u_2'\rangle}} = \sqrt{2}\begin{pmatrix} \frac{1}{2} \\ -\frac{1}{2} \end{pmatrix} = \begin{pmatrix} \frac{1}{\sqrt{2}} \\ -\frac{1}{\sqrt{2}} \end{pmatrix}$$

//

Let

$$\mathcal{U}_\ell^{(n)} \equiv \mathbb{U}_{\ell n} \tag{20}$$

then Eq. (15) can be written as

$$\sum_{\ell=1}^{L} (\mathbb{U}_{\ell n})^* \mathbb{U}_{\ell n'} = \sum_{\ell=1}^{L} (\mathbb{U}^\dagger)_{n\ell} \mathbb{U}_{\ell n'} = \delta_{nn'}$$

Therefore, $\mathbb{U}$ is unitary:

$$\mathbb{U}^\dagger \mathbb{U} = \mathbb{I}_L \tag{21}$$

Substituting Eq. (20) in (14),

$$\sum_{\ell'=1}^{L} S_{\ell\ell'} \mathbb{U}_{\ell' n} = \underbrace{\mathbb{U}_{\ell n} \lambda_n}_{\sum_{\ell'=1}^{L} \mathbb{U}_{\ell\ell'} (\delta_{\ell' n} \lambda_n)}$$

$$\therefore \quad \mathbb{S}\,\mathbb{U} = \mathbb{U}\lambda \tag{22}$$

or

$$\mathbb{S} = \mathbb{U}\,\lambda\,\mathbb{U}^\dagger \tag{23}$$

where

$$\lambda_{nn'} = \delta_{nn'} \lambda_{n'} \tag{24}$$

Note that, if the matrix $S_{\ell\ell'}$ is rank deficient, some eigenvalues are $\emptyset$ (i.e., $S_{\ell\ell'}$ is singular), and $S_{\ell\ell'}$ is not invertable.

(Example)

$$S_{\ell\ell'} = \begin{pmatrix} 2 & 4 \\ 4 & 8 \end{pmatrix}$$

$$\begin{pmatrix} 2 & 4 \\ 4 & 8 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \lambda \begin{pmatrix} x \\ y \end{pmatrix}$$

$$\begin{pmatrix} \lambda-2 & -4 \\ -4 & \lambda-8 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = 0$$

For this to have a non-trivial solution,

$$\begin{vmatrix} \lambda-2 & -4 \\ -4 & \lambda-8 \end{vmatrix} = (\lambda-2)(\lambda-8)-16 = \lambda^2-10\lambda+\cancel{16}-\cancel{16} = \lambda(\lambda-10) = 0$$

$\therefore \lambda = 10, 0$

$\underline{\lambda_1 = 10}$

$$\begin{pmatrix} 8 & -4 \\ -4 & 2 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \quad \therefore \quad u_\ell^{(1)} = \frac{1}{\sqrt{5}}\begin{pmatrix} 1 \\ 2 \end{pmatrix}$$

$\underline{\lambda_2 = 0}$

$$\begin{pmatrix} -2 & -4 \\ -4 & -8 \end{pmatrix}\begin{pmatrix} x \\ y \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \quad \therefore \quad u_\ell^{(2)} = \frac{1}{\sqrt{5}}\begin{pmatrix} 2 \\ -1 \end{pmatrix}$$

$$\therefore \underbrace{\begin{pmatrix} 2 & 4 \\ 4 & 8 \end{pmatrix}}_{\mathbb{S}} \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} & \frac{2}{\sqrt{5}} \\ \frac{2}{\sqrt{5}} & \frac{-1}{\sqrt{5}} \end{pmatrix}}_{\mathbb{U}} = \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} & \frac{2}{\sqrt{5}} \\ \frac{2}{\sqrt{5}} & \frac{-1}{\sqrt{5}} \end{pmatrix}}_{\mathbb{U}} \underbrace{\begin{pmatrix} 10 & 0 \\ 0 & 0 \end{pmatrix}}_{\lambda} \tag{25}$$

If $\mathbb{S}$ is not singular (no $\lambda_n = 0$), then

$$\mathbb{S}^{-1} = \mathbb{U}\lambda^{-1}\mathbb{U}^\dagger \tag{26}$$

($\because$ $\mathbb{S} = \mathbb{U}\lambda\mathbb{U}^\dagger$ and thus $\mathbb{U}\lambda\underbrace{\mathbb{U}^\dagger(\mathbb{U}}_{\mathbb{I}}\lambda^{-1}\mathbb{U}^\dagger) = \mathbb{U}\underbrace{\lambda\lambda^{-1}}_{\mathbb{I}}\mathbb{U}^\dagger = \mathbb{U}\mathbb{U}^\dagger = \mathbb{I}$),

but this is not possible for singular $\mathbb{S}$.

The SVD recipe to solve the generalized eigenvalue problem, Eq. (9), with a rank-deficient overlap matrix, $\mathbb{S}$, is to introduce the low-rank, orthogonal representation of $\mathbb{S}$.

(Example of low-rank representation)

In Eq. (25), "contract" λ by throwing out all 0's:

$$\mathbb{S} = \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} & \frac{2}{\sqrt{5}} \\ \frac{2}{\sqrt{5}} & \frac{-1}{\sqrt{5}} \end{pmatrix}}_{\substack{\mathbb{U} \\ 2\times 2}} \underbrace{\begin{pmatrix} 10 & 0 \\ 0 & 0 \end{pmatrix}}_{\substack{\lambda \\ 2\times 2}} \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} & \frac{2}{\sqrt{5}} \\ \frac{2}{\sqrt{5}} & \frac{-1}{\sqrt{5}} \end{pmatrix}}_{\substack{\mathbb{U}^\dagger \\ 2\times 2}}$$

low-rank representation

$$\mathbb{S} = \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} \\ \frac{2}{\sqrt{5}} \end{pmatrix}}_{\substack{\tilde{\mathbb{U}} \\ 2\times 1}} \underbrace{(10)}_{\substack{\tilde{\lambda} \\ 1\times 1}} \underbrace{\begin{pmatrix} \frac{1}{\sqrt{5}} & \frac{2}{\sqrt{5}} \end{pmatrix}}_{\substack{\tilde{\mathbb{U}}^\dagger \\ 1\times 2}} = 10 \begin{pmatrix} \frac{1}{5} & \frac{2}{5} \\ \frac{2}{5} & \frac{4}{5} \end{pmatrix} = \begin{pmatrix} 2 & 4 \\ 4 & 8 \end{pmatrix}$$

Let's order the eigenstates of $\mathbb{S}$ such that

$$\lambda_1 \geq \lambda_2 \geq \cdots \geq \lambda_M (\neq 0) \geq \lambda_{M+1} = \cdots = \lambda_L = 0 \qquad (27)$$

and truncate Eq. (23) by throwing all L-M 0's:

$$S_{\ell\ell'} = \sum_{n=1}^{L} \sum_{n'=1}^{L} U_{\ell n} \underbrace{\lambda_{nn'}}_{\delta_{nn'}\lambda_{n'}} (U^\dagger)_{n'\ell'}$$

$$= \sum_{n=1}^{L} \sum_{n'=1}^{L} u_\ell^{(n)} (\delta_{nn'}\lambda_{n'}) u_{\ell'}^{*(n)}$$

$$= \sum_{n=1}^{L} u_\ell^{(n)} \lambda_n u_{\ell'}^{*(n)} \qquad (28)$$

low-rank representation

$$S_{\ell\ell'} = \sum_{n=1}^{M} u_\ell^{(n)} \lambda_n u_{\ell'}^{*(n)}$$

$$= \sum_{n=1}^{M} \sum_{n'=1}^{M} u_\ell^{(n)} (\lambda_n \delta_{nn'}) u_{\ell'}^{*(n')} \qquad (29)$$

Let

$$\begin{cases} \tilde{U}_{\ell n} = u_\ell^{(n)} & (\ell = 1,\dots,L;\ n = 1,\dots,M) \qquad (30) \\ \tilde{\lambda}_{nn'} = \delta_{nn'}\lambda_{n'} & (n,n' = 1,\dots,M) \qquad (31) \end{cases}$$

then

$$S_{\ell\ell'} = \sum_{n=1}^{M}\sum_{n'=1}^{M} \tilde{U}_{\ell n}\,\tilde{\lambda}_{nn'}\,\tilde{U}^{\dagger}_{n'\ell'} \qquad (32)$$

or

$$\underset{L\times L}{\mathbb{S}} = \underset{L\times M}{\tilde{\mathbb{U}}}\ \underset{M\times M}{\tilde{\lambda}}\ \underset{M\times L}{\tilde{\mathbb{U}}^{\dagger}} \qquad (33)$$

$$\begin{array}{c} \\ 1 \\ \\ L \end{array}\!\!\begin{array}{c} 1 \qquad\qquad L \\ \begin{bmatrix} S_{11} & \cdots & S_{1L} \\ & & \\ S_{L1} & \cdots & S_{LL} \end{bmatrix} \end{array} = \begin{array}{c} \\ 1 \\ \\ L \end{array}\!\!\begin{array}{c} 1 \qquad M \\ \begin{bmatrix} u_1^{(1)} \cdots u_1^{(M)} \\ |1\rangle \quad |M\rangle \\ u_L^{(1)} \cdots u_L^{(M)} \end{bmatrix} \end{array} \begin{array}{c} \\ 1 \\ M \end{array}\!\!\begin{array}{c} 1 \qquad M \\ \begin{bmatrix} \lambda_1 & & \\ & \ddots & \\ & & \lambda_M \end{bmatrix} \end{array} \begin{array}{c} \\ 1 \\ \\ M \end{array}\!\!\begin{array}{c} 1 \qquad\qquad L \\ \begin{bmatrix} u_1^{(1)*} & \langle 1| & u_L^{(1)*} \\ \vdots & & \vdots \\ u_1^{(M)*} & \langle M| & u_L^{(M)*} \end{bmatrix} \end{array}$$

$\tilde{U}_{\ell n} = u_\ell^{(n)}$
only M states

$\tilde{U}^{\dagger}_{n'\ell'} = u_{\ell'}^{(n')*}$
only M states

$$\sim \sum_{n=1}^{M} |n\rangle \lambda_n \langle n|$$

→ vector in $\mathbb{C}^L$

Note that the reduced $u_\ell^{(n)}$ are still orthonormal:

$$\sum_{\ell=1}^{L} \left(u_\ell^{(n)}\right)^{*} u_\ell^{(n')} = \delta_{nn'} \qquad (n,n' = 1,\dots,M) \qquad (34)$$

or

$$\sum_{\ell=1}^{L} \tilde{U}^{\dagger}_{n\ell}\,\tilde{U}_{\ell n'} = \delta_{nn'} \qquad (n, n' = 1,\dots,M) \qquad (35)$$

or

$$\underset{M\times L}{\tilde{\mathbb{U}}^{\dagger}}\ \underset{L\times M}{\tilde{\mathbb{U}}} = \underset{M\times M}{\mathbb{I}_M} \qquad (36)$$

— SVD-orthonormalized eigenvalue problem

From Eq. (33),

$$\underset{M\times L}{\tilde{\mathbb{U}}^{\dagger}}\,\underset{L\times L}{\mathbb{S}}\,\underset{L\times M}{\tilde{\mathbb{U}}} = \underset{M\times M}{\lambda} \tag{37}$$

$$\therefore \quad \underset{M\times M}{\tilde{\lambda}^{-1/2}}\,\underset{M\times L}{\tilde{\mathbb{U}}^{\dagger}}\,\underset{L\times L}{\mathbb{S}}\,\underset{L\times M}{\tilde{\mathbb{U}}}\,\underset{M\times M}{\tilde{\lambda}^{-1/2}} = \underset{M\times M}{\mathbb{I}_M} \tag{38}$$

or

$$\sum_{\ell=1}^{L}\sum_{\ell'=1}^{L} \lambda_n^{-1/2}\,\tilde{U}^{\dagger}_{n\ell}\,\underbrace{S_{\ell\ell'}}_{\langle\Psi(E_\ell)|\Psi(E_{\ell'})\rangle}\,\tilde{U}_{\ell'n'}\,\lambda_{n'}^{-1/2} = \delta_{nn'} \tag{39}$$

$$\therefore \left(\sum_{\ell=1}^{L}\lambda_n^{-1/2}\,\tilde{U}^{\dagger}_{n\ell}\,\langle\Psi(E_\ell)|\right)\sum_{\ell'=1}^{L}|\Psi(E_{\ell'})\rangle\,\tilde{U}_{\ell'n'}\,\lambda_{n'}^{-1/2} = \delta_{nn'}$$

Let's define a new basis,

$$|\chi_n\rangle \equiv \sum_{\ell=1}^{L}|\Psi(E_\ell)\rangle\,\tilde{U}_{\ell n}\,\lambda_n^{-1/2} \qquad (n=1,\ldots,M) \tag{40}$$

Then, this basis is orthonormal in the rank-M space:

$$\langle\chi_n|\chi_{n'}\rangle = \delta_{nn'} \tag{41}$$

The Hamiltonian $\hat{H}$ can be diagonalized in this orthonormal basis.

$$\begin{cases} \hat{H}|\phi_m\rangle = \epsilon_m|\phi_m\rangle \qquad (m=1,\ldots,M) & (42) \\[2ex] |\phi_m\rangle = \displaystyle\sum_{n=1}^{M} y_n^{(m)}\,|\chi_n\rangle & (43) \end{cases}$$

Substituting Eq. (43) in (42),

$$\sum_{n'=1}^{M} y_{n'}^{(m)} \hat{H} |\chi_{n'}\rangle = \epsilon_m \sum_{n'=1}^{M} y_{n'}^{(m)} |\chi_{n'}\rangle \tag{44}$$

$\langle \chi_n | \times$ Eq. (44)

$$\sum_{n'=1}^{M} \underbrace{\langle \chi_n | \hat{H} | \chi_{n'}\rangle}_{\tilde{H}_{nn'}} \underbrace{y_{n'}^{(m)}}_{Y_{n'm}} = \sum_{n'=1}^{M} \underbrace{\langle \chi_n | \chi_{n'}\rangle}_{\delta_{nn'}} y_{n'}^{(m)} \epsilon_m$$

$$= y_n^{(m)} \epsilon_m = \sum_{m'=1}^{M} \underbrace{y_n^{(m')} (\delta_{mm'} \epsilon_{m'})}_{Y_{nm'}\ \epsilon_{mm'}} \tag{45}$$

Let

$$\tilde{H}_{nn'} \equiv \langle \chi_n | \hat{H} | \chi_{n'}\rangle \tag{46}$$

$$= \sum_{l=1}^{L} \sum_{l'=1}^{L} \lambda_n^{-1/2} \tilde{U}_{nl}^{\dagger} \underbrace{\langle \Psi(E_l) | \hat{H} | \Psi(E_{l'})\rangle}_{H_{ll'}} \tilde{U}_{l'n} \lambda_n^{-1/2}$$

or

$$\underset{M\times M}{\tilde{\mathbb{H}}} \equiv \underset{M\times M}{\tilde{\lambda}^{-1/2}} \underset{M\times L}{\tilde{\mathbb{U}}^{\dagger}} \underset{L\times L}{\mathbb{H}} \underset{L\times M}{\tilde{\mathbb{U}}} \underset{M\times M}{\lambda^{-1/2}} \tag{47}$$

and

$$Y_{nm} = y_n^{(m)} \qquad (n, m = 1, \dots, M) \tag{48}$$

$$\epsilon_{mm'} = \delta_{mm'} \epsilon_{m'} \qquad (m, m' = 1, \dots, M) \tag{49}$$

Then Eq. (45) can be rewritten as

$$\underset{M\times M}{\tilde{\mathbb{H}}} = \underset{M\times M}{\mathbb{Y}^{\dagger}} \underset{M\times M}{\mathbb{E}} \underset{M\times M}{\mathbb{Y}} \tag{50}$$

Note that $|\Phi_m\rangle$ are orthonormal:

$$\langle \Phi_m | \Phi_{m'} \rangle = \delta_{mm'} \tag{51}$$

$$\therefore \sum_{n=1}^{M} \sum_{n'=1}^{M} \langle \chi_n | (y_n^{(m)})^* y_{n'}^{(m')} | \chi_{n'}\rangle = \sum_{n} \sum_{n'} (Y^{\dagger})_{mn} Y_{n'm'} \underbrace{\langle \chi_n | \chi_{n'}\rangle}_{\delta_{nn'}} = \delta_{mm'}$$

$$\sum_{n=1}^{M} (Y^{\dagger})_{mn} Y_{nm'} = \delta_{mm'}$$

Therefore, $\mathbb{Y}$ is unitary:

$$\mathbb{Y}^{\dagger} \mathbb{Y} = \mathbb{I}_M \tag{52}$$

(SVD algorithm)

1. Construct the overlap matrix

$$S_{\ell\ell'} = \langle \Psi(E_\ell) | \Psi(E_{\ell'}) \rangle \qquad (\ell, \ell' = 1, \ldots, L) \tag{11}$$

2. Perform SVD of the $L \times L$ $\mathbb{S}$

$$\underset{L\times L}{\mathbb{S}} = \underset{L\times L}{\mathbb{U}} \underset{L\times L}{\lambda} \underset{L\times L}{\mathbb{U}^\dagger} \tag{23}$$

where $\lambda_1 \geq \lambda_2 \geq \cdots \geq \lambda_M (\neq 0) \geq \lambda_{M+1} = \cdots = \lambda_L = 0$.

3. Contract Eq.(23) to derive rank-M representation of $\mathbb{S}$ by throwing out L−M Ø's in λ:

$$\begin{cases} \tilde{U}_{\ell n} = U_{\ell n} = u^n_{(\ell)} & (\ell = 1, \ldots, L;\ n = 1, \ldots, M) \\ \tilde{\lambda}_{nn'} = \delta_{nn'} \lambda_{n'} & (n, n' = 1, \ldots, M) \end{cases} \begin{matrix} (30) \\ (31) \end{matrix}$$

$$\underset{L\times L}{\mathbb{S}} = \underset{L\times M}{\tilde{\mathbb{U}}} \underset{M\times M}{\tilde{\lambda}} \underset{M\times L}{\tilde{\mathbb{U}}^\dagger} \tag{32}$$

4. Define the rank-M Hamiltonian,

$$\underset{M\times M}{\tilde{\mathbb{H}}} \equiv \underset{M\times M}{\tilde{\lambda}^{-1/2}} \underset{M\times L}{\tilde{\mathbb{U}}^\dagger} \underset{L\times L}{\mathbb{H}} \underset{L\times M}{\tilde{\mathbb{U}}} \underset{M\times M}{\tilde{\lambda}^{-1/2}} \tag{33}$$

where

$$H_{\ell\ell'} = \langle \Psi(E_\ell) | \hat{H} | \Psi(E_{\ell'}) \rangle \qquad (\ell, \ell' = 1, \ldots, L) \tag{34}$$

5. Diagonalize the (non-singular) Hamiltonian $\tilde{\mathbb{H}}$

$$\underset{M\times M}{\tilde{\mathbb{H}}} = \underset{M\times M}{\mathbb{Y}^\dagger} \underset{M\times M}{\mathbb{E}} \underset{M\times M}{\mathbb{Y}} \tag{50}$$

6. The eigen states of $\hat{H}$ are

$$|\Phi_m\rangle = \sum_{n=1}^{M} \underbrace{|\chi_n\rangle}_{\sum_{\ell=1}^{L} |\Psi(E_\ell)\rangle \tilde{U}_{\ell n} \tilde{\lambda}_n^{-1/2}} Y_{nm} \tag{43}$$

(∵ Eq.(40))

$$\therefore\ |\Phi_m\rangle = \sum_{\ell=1}^{L} \sum_{n=1}^{M} |\Psi(E_\ell)\rangle \tilde{U}_{\ell n} \tilde{\lambda}_n^{-1/2} Y_{nm} \qquad (m = 1, \ldots, M) \tag{44}$$

with the eigenvalues E_m.

– Construction of $\mathbb{S}$ and $\mathbb{H}$ via the correlation function

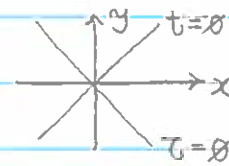
$$S_{\ell\ell'} = \langle \Psi(E_\ell) | \Psi(E_{\ell'}) \rangle \quad (\because \text{Eq.}(10))$$

$$= \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy \left(\underbrace{\langle \psi(x) |}_{\langle \psi_0 | e^{i\hat{H}x/\hbar}} e^{-iE_\ell x/\hbar} e^{-x^2/2T^2} \right) e^{-y^2/2T^2} e^{iE_{\ell'} y/\hbar} \underbrace{|\psi(y)\rangle}_{e^{-i\hat{H}y/\hbar}|\psi_0\rangle} \quad (\because \text{Eq.}(3))$$

$$= \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy\, e^{-(x^2+y^2)/2T^2} e^{-iE_\ell x/\hbar} e^{iE_{\ell'} y/\hbar} \underbrace{\langle \psi_0 | e^{-i\hat{H}(y-x)/\hbar} | \psi_0 \rangle}_{C(y-x)\ (\because \text{Eq.}(1))} \qquad (45)$$

We now change the variables to

$$y - x = t \qquad \text{or} \qquad y = \tau + \frac{t}{2}$$
$$\frac{y+x}{2} = \tau \qquad\qquad x = \tau - \frac{t}{2}$$



$$dx\,dy = \begin{vmatrix} \frac{\partial x}{\partial t} & \frac{\partial x}{\partial \tau} \\ \frac{\partial y}{\partial t} & \frac{\partial y}{\partial \tau} \end{vmatrix} dt\,d\tau = \begin{vmatrix} -\frac{1}{2} & 1 \\ \frac{1}{2} & 1 \end{vmatrix} dt\,d\tau = dt\,d\tau$$

$$x^2 + y^2 = \left(\tau + \frac{t}{2}\right)^2 + \left(\tau - \frac{t}{2}\right)^2 = 2\tau^2 + \frac{t^2}{2}$$

$$\therefore S_{\ell\ell'} = \int_{-\infty}^{\infty} d\tau \int_{-\infty}^{\infty} dt\, e^{-\tau^2/T^2} e^{-t^2/4T^2} e^{-iE_\ell(\tau - \frac{t}{2})/\hbar} e^{iE_{\ell'}(\tau + \frac{t}{2})/\hbar} C(t)$$

$$= \underbrace{\int_{-\infty}^{\infty} d\tau\, e^{-\tau^2/T^2} e^{-i(E_\ell - E_{\ell'})\tau/\hbar}}_{a} \int_{-\infty}^{\infty} dt\, e^{-t^2/4T^2} e^{i(E_\ell + E_{\ell'})t/2\hbar} C(t)$$

$$a = \int_{-\infty}^{\infty} d\tau \exp\left\{ \underbrace{-\frac{1}{T^2}\left[\tau^2 + i\frac{(E_\ell - E_{\ell'})\tau T^2}{\hbar}\right]}_{-\frac{1}{T^2}\left\{\left[\tau + \frac{i(E_\ell - E_{\ell'})T^2}{2\hbar}\right]^2 + \frac{(E_\ell - E_{\ell'})^2 T^4}{4\hbar^2}\right\}} \right\}$$

$$= \exp\left[-\frac{(E_\ell - E_{\ell'})^2 T^2}{4\hbar^2}\right] \underbrace{\int_{-\infty}^{\infty} d\tau \exp\left\{-\frac{1}{T^2}\left[\tau + \frac{i(E_\ell - E_{\ell'})T^2}{2\hbar}\right]^2\right\}}_{T\sqrt{\pi}}$$

$$\therefore S_{\ell\ell'} = \sqrt{\pi}\, T \exp\left[-\frac{(E_\ell - E_{\ell'})^2 T^2}{4\hbar^2}\right] \int_{-\infty}^{\infty} dt\, e^{-t^2/4T^2} e^{i(E_\ell + E_{\ell'})t/2\hbar} C(t) \qquad (46)$$

$$H_{\ell\ell'} = \langle \Phi(E_\ell)|\hat{H}|\Phi(E_{\ell'})\rangle \quad (\because \text{Eq.}(10))$$

$$= \underbrace{\sqrt{\pi}\,T \exp\left[-\frac{(E_\ell-E_{\ell'})^2T^2}{4\hbar^2}\right]}_{a} \int_{-\infty}^{\infty} dt\, e^{-t^2/4T^2} e^{i(E_\ell+E_{\ell'})t/2\hbar} \underbrace{\langle\psi_0|\hat{H} e^{-i\hat{H}t/\hbar}|\psi_0\rangle}_{i\hbar\frac{d}{dt}\underbrace{\langle\psi_0|e^{-i\hat{H}t/\hbar}|\psi_0\rangle}_{C(t)}}$$

$$= a\cdot i\hbar \int_{-\infty}^{\infty} dt\, e^{-t^2/4T^2} e^{i(E_\ell+E_{\ell'})t/2\hbar} \frac{d}{dt}C(t)$$

$$= -a\cdot i\hbar \int_{-\infty}^{\infty} dt \left[-\frac{t}{2T^2} + i\frac{(E_\ell+E_{\ell'})}{2\hbar}\right] \exp(\cdots)\, C(t)$$

$$= a\int_{-\infty}^{\infty} dt \left(\frac{E_\ell+E_{\ell'}}{2} + \frac{i\hbar t}{2T^2}\right) \exp(\cdots)\, C(t)$$

$$\therefore H_{\ell\ell'} = \frac{\sqrt{\pi}\,T}{2} \exp\left[-\frac{(E_\ell-E_{\ell'})^2T^2}{4\hbar^2}\right] \int_{-\infty}^{\infty} dt \left(E_\ell+E_{\ell'} + \frac{i\hbar t}{T^2}\right) e^{-t^2/4T^2} e^{i(E_\ell+E_{\ell'})t/2\hbar}\, C(t) \qquad (47)$$

(S and H construction algorithm)

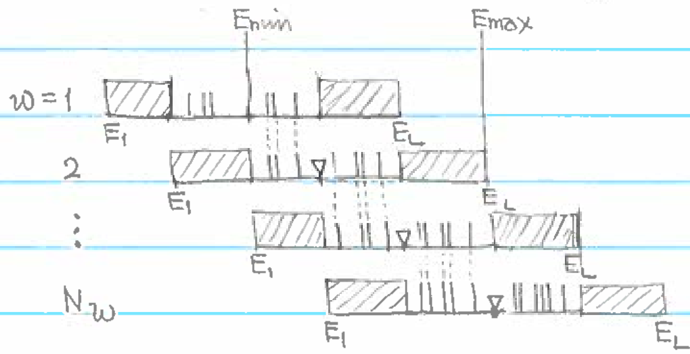
1. Prepare a random intial state $|\psi_0\rangle$
2. Propagate $|\psi(t)\rangle = e^{-i\hat{H}t/\hbar}|\psi_0\rangle$ for $[0, T_{max}]$ and record $C(t) = \langle\psi_0|\psi(t)\rangle$ for $t \in [0, T_{max}]$; extend $C(-t) = C^*(t)$ so that $C(t)$ is defined for $t \in [-T_{max}, T_{max}]$.
3. $$\begin{cases} S_{\ell\ell'} = \sqrt{\pi}\,T \exp\left[-\frac{(E_\ell-E_{\ell'})^2T^2}{4\hbar^2}\right] \int_{-\infty}^{\infty} dt\, e^{-t^2/4T^2} e^{i(E_\ell+E_{\ell'})t/2\hbar}\, C(t) & (46) \\ H_{\ell\ell'} = \frac{\sqrt{\pi}\,T}{2} \exp\left[-\frac{(E_\ell-E_{\ell'})^2T^2}{4\hbar^2}\right] \int_{-\infty}^{\infty} dt \left(E_\ell+E_{\ell'} + \frac{i\hbar t}{T^2}\right) e^{-t^2/4T^2} e^{i(E_\ell+E_{\ell'})t/2\hbar}\, C(t) & (47) \end{cases}$$

$(\ell, \ell' = 1, \ldots, L)$

— Low-rank "pipelining" algorithm

For a given energy window $[E_{min}, E_{max}]$, $L, M \propto N$, and thus $O(L^3)$ SVD and $O(M^3)$ eigenvalue problem requires $O(N^3)$ operation.

Instead, we divide $[E_{min}, E_{max}]$ into N_W overlapping subwindows each with L wavepackets; for each subwindow we throw out half the outerlying energy spectra.



(cf. density-matrix renormalization group)

Every (un-thrown) eigenstates are thus computed exactly twice, providing an error estimate.

$$E_{max} - E_{min} = (N_w - 1)\frac{E_L - E_1}{4} \quad (48)$$

We now have N_w $(= O(N))$ subproblems with each $O(L^3 + M^3) = O(1)$ operations.

※ Construction of $C(t)$ costs $O(N)$, but this is done only once for all subwindows and wavepackets.

(Algorithm)

1. Construct $C(t)$ for $t \in [-T_{max}, T_{max}]$
2. for $w = 1, N_w$

   construct $S_{\ell\ell'}$ & $H_{\ell\ell'}$ $(\ell, \ell' = 1, \ldots, L)$ in $[(w-\frac{1}{2})(E_L - E_1), (w+\frac{1}{2})(E_L - E_1)]$ $\qquad \frac{4(E_{max}-E_{min})}{N_w - 1}$

   SVD of $\mathbb{S}$; diagonalize $\tilde{\mathbb{H}}$

   retain inner half of the states.