An introduction to FCIQMC, the NECI codebase, and more

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Lodz
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Overview of the three lectures and tutorials

- Introduction, Full CI Quantum Monte Carlo and NECI (AA)
- Transcorrelation: combining real-space methods such as VMC with quantum chemistry. Integral calculation with TCHint (PLR)
- Transcorrelated Coupled Cluster (DK): solving the TC Hamiltonian with CC (e-co.jl)
- Practical calculations (Philip Haupt, Johannes Hauskrecht)

Many-Electron Schrödinger equation

$$H = -\frac{1}{2}\sum_{i=1}^{N}\nabla_i^2 + \sum_{i<j}\frac{1}{r_{ij}} + \sum_i v(\mathbf{r}_i)$$

$$\begin{aligned} H\Psi_0 &= E_0\Psi_0 \\ \Psi_0 &= \Psi_0(\mathbf{x}_1, ..., \mathbf{x}_N) \qquad\qquad \mathbf{x} = (\mathbf{r}, \sigma) \end{aligned}$$

Electrons are Fermions:

$$\Psi_0(....\mathbf{x}_i, ..., \mathbf{x}_j ...) = -\Psi_0(....\mathbf{x}_j, ..., \mathbf{x}_i ...)$$

Atomic units $\hbar = m_e = |e| = 1$ E_h = 27.211 eV

The "standard" Quantum Chemical Hamiltonian

2nd quantisation in finite basis sets

Introduce $M \gg N$ spatial orbitals,
together with their **fermionic creation and annihilation operators**

$$\hat{H} = \sum_{pq\sigma} h_q^p c_{p\sigma}^\dagger c_{q\sigma} + \sum_{pqrs\sigma\tau} v_{rs}^{pq} c_{p\sigma}^\dagger c_{q\tau}^\dagger c_{s\tau} c_{r\sigma}$$

$$h_q^p = \langle \phi_p | \hat{h} | \phi_q \rangle \qquad \hat{h} = -\frac{1}{2}\nabla^2 + v_{ext}(\mathbf{r})$$

$$v_{rs}^{pq} = \langle \phi_p \phi_q | \hat{U} | \phi_r \phi_s \rangle \qquad \hat{U} = r_{12}^{-1}$$

Orbitals usually given by Restricted Hartree-Fock for weak-correlation and CAS-SCF for strong correlation

Major capabilities of NECI

- FCIQMC: exact stochastic FCI method which can overcome the fermion sign problem
- High parallelisability (upto ~20000 cores) allowing > 10^{10} walker simulations
- Initiator and adaptive-shift approximation for large systems
- Transcorrelated Hamiltonians
- Calculation of 1- and 2-body RDMs and TDMs
- Excited states
- Spin-adapted methodology (GUGA) for large open-shell systems (more than 20 OS orbitals)
- Real-time propagation, and spectral functions

Imaginary time Schrödinger Equation and determinant based expansions

$$-\partial_\tau \Psi = (\hat{H} - E)\Psi = 0$$

$$\Psi(\tau) = \sum_i C_i(\tau)\,|D_i\rangle, \qquad |D_i\rangle = \frac{1}{\sqrt{N!}}\,|\phi_{a\sigma}\phi_{b\tau}\ldots.|$$

$$N_{FCI} = \binom{M}{N_\alpha}\binom{M}{N_\beta}, \quad N_\alpha + N_\beta = N$$

M=number of spatial orbitals
N=number of electrons

$$\Psi_0 \propto \lim_{\tau\to\infty} \Psi(\tau) = \lim_{\tau\to\infty} e^{-\tau\hat{H}}\Psi(0)$$

We will solve this problem via a stochastic propagation of signed walkers with explicit annihilation (FCIQMC)

FCIQMC: population dynamics of a set of walkers

$$\Psi = \sum_i \left(\sum_\gamma^{N_w} s_\gamma \delta(i - i_\gamma) \right) | D_i \rangle \qquad N_w = \sum_\gamma | s_\gamma |$$

Booth, Thom and Alavi, J Chem Phys, **131**, 054106, (2009)

Initiator Method: controlling the sign problem

$$-\partial_\tau \Psi = (H[\Psi] - E)\Psi = 0$$

Cleland, Booth, Alavi, J Chem Phys, **132**, 041103, (2010)

Adaptive Shift: unbiasing initiator bias in large systems

$$-\partial_\tau \Psi = (\hat{H}[\Psi] - E[\Psi])\Psi = 0$$

Ghanem, Lozovoi, Alavi, J Chem Phys, **151**, 224108 (2019)
Ghanem, Guther, Alavi, J Chem Phys, **153**, 224115 (2020)

Schrödinger Equation

$$-\partial_\tau \Psi = (\hat{H} - E)\Psi = 0$$

FCIQMC: population dynamics of a set of walkers

$$\Psi = \sum_i \left(\sum_\gamma^{N_w} s_\gamma \delta(i - i_\gamma) \right) |D_i\rangle \qquad N_w = \sum_\gamma |s_\gamma|$$

Master equation for the first-order kinetics of the walkers:

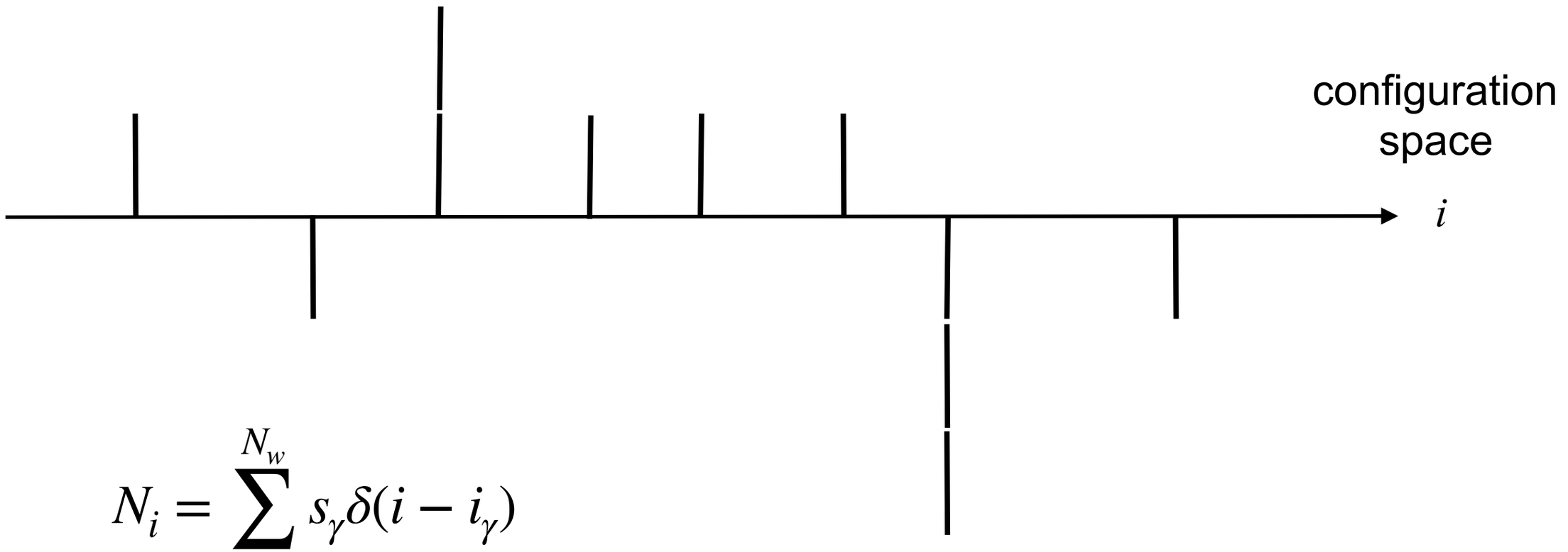
$$N_i = \sum_\gamma^{N_w} s_\gamma \delta(i - i_\gamma)$$

$$-\frac{dN_i}{d\tau} = (H_{ii} - S)N_i + \sum_{j \neq i} H_{ij} N_j$$

Shift

Pictorial example

$$N_w = \sum_\gamma |s_\gamma| = 11$$



$$N_i = \sum_\gamma^{N_w} s_\gamma \delta(i - i_\gamma)$$

$$\text{L}_2 \text{ norm} \quad = \frac{1}{\sqrt{\sum_i N_i^2}} = \frac{1}{\sqrt{19}}$$

Overview of FCIQMC algorithm:

a random Game of Life, death and annihilation

Start with N (positive) walkers on D_0, an initial value of S, and time-step τ

↓

Spawning (birth) processes

(Diagonal) death/cloning processes

Annihilation step

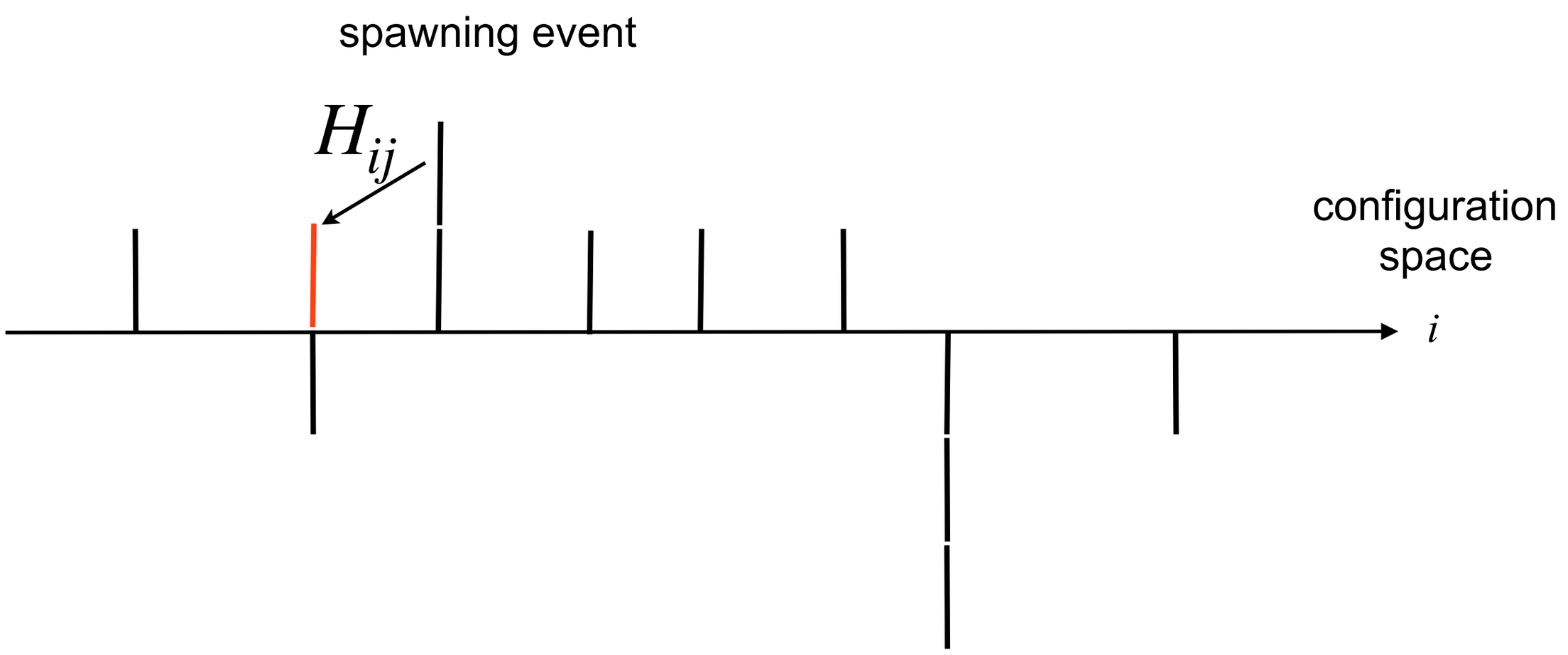
Pairwise removal of walkers of opposite sign on the same det

Adjust Shift (in constant N mode)

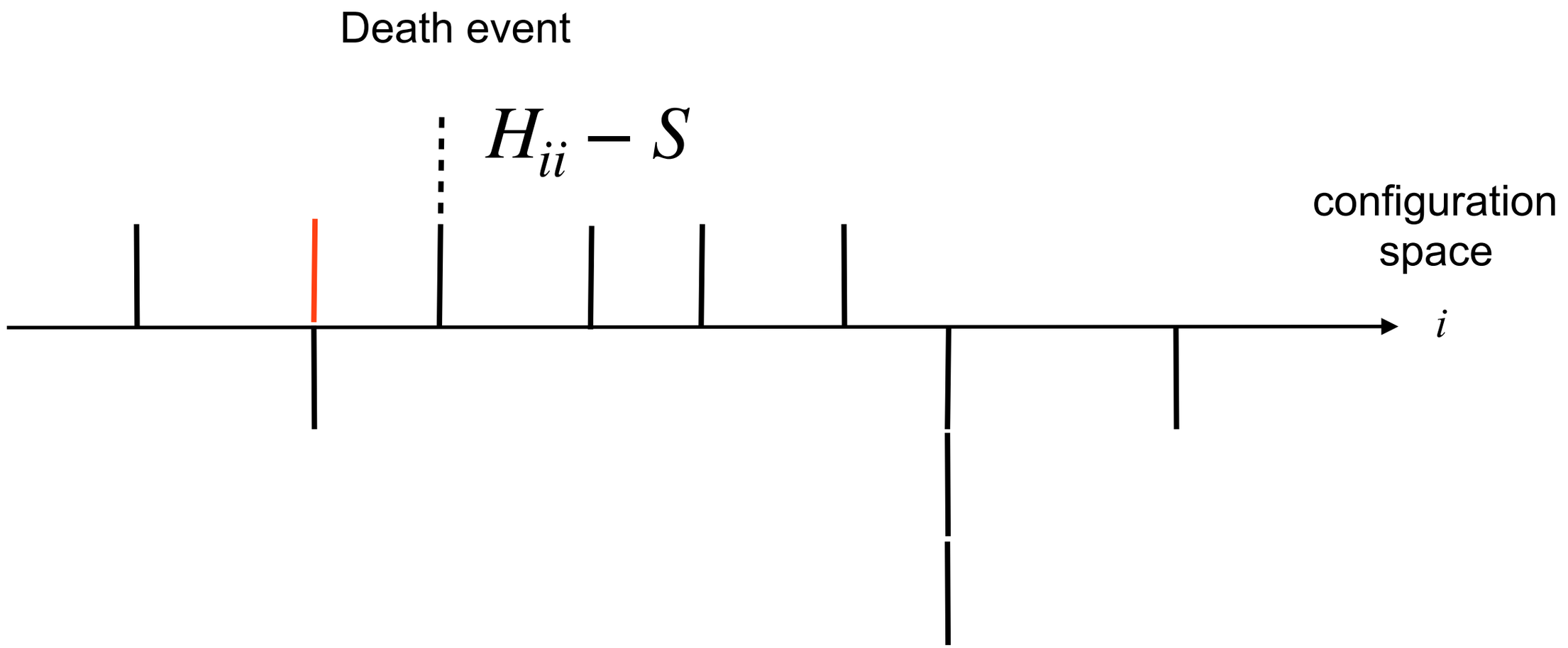
$$S \rightarrow S - \frac{1}{A\tau} \ln \frac{N_{current}}{N_{old}}$$

Booth, Thom and Alavi, J Chem Phys, 131, 054106, (2009)

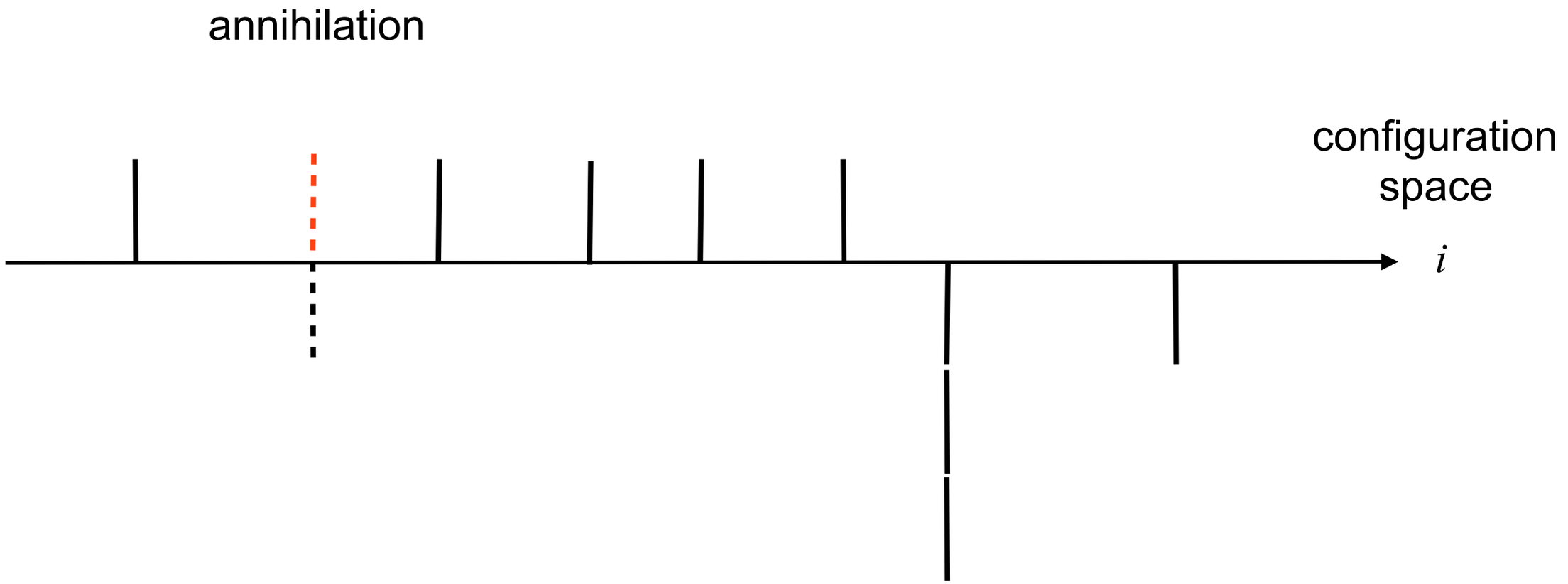
Spawning



Death



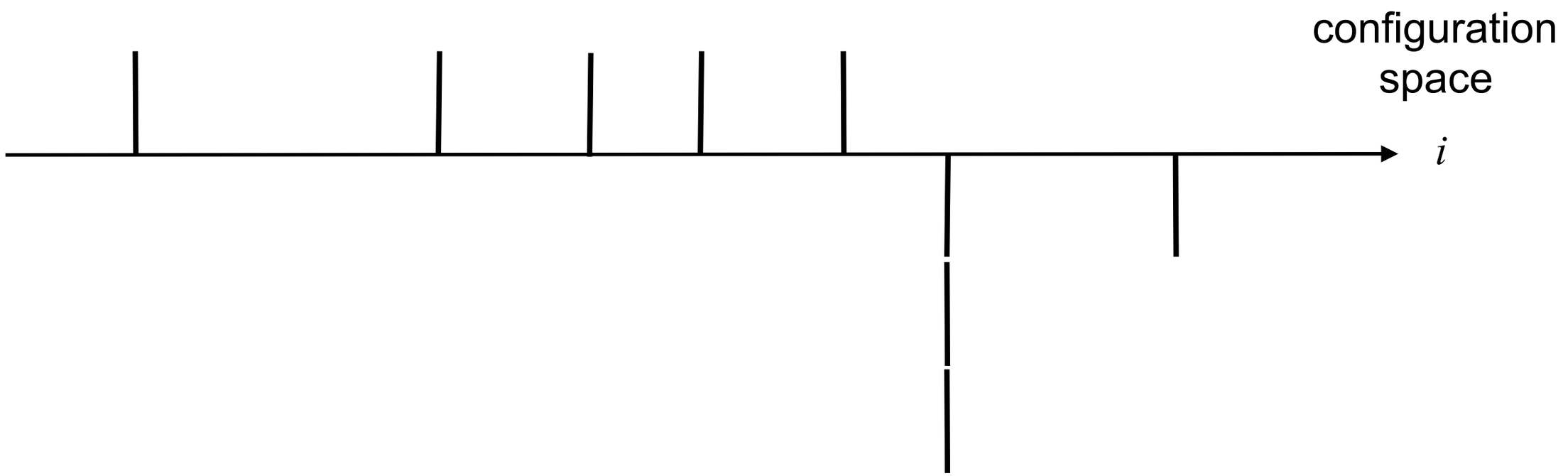
Annihilation



Hash algorithm: $O(N_w)$

Booth, Smart, Alavi, Mol. Phys., **112** (14), (2014),1855-1869

Annihilation

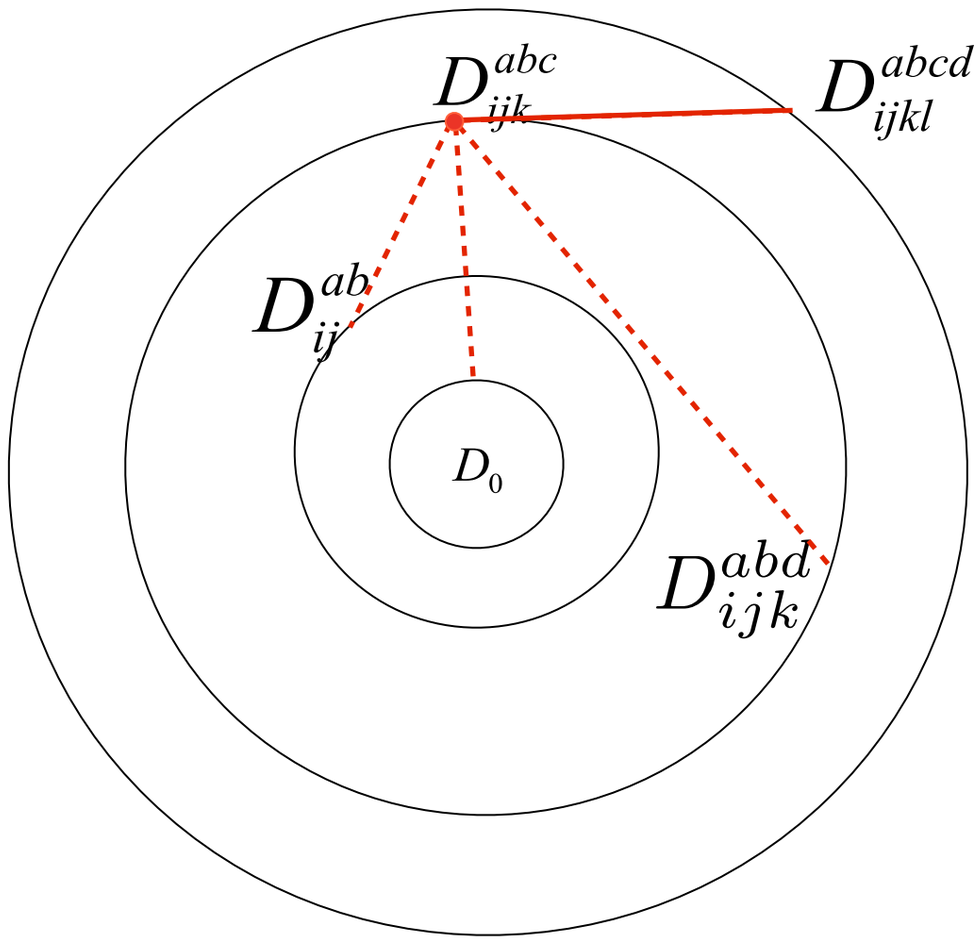


The rules of FCIQMC

(derived from the underlying imaginary-time S.E.)

Probability of death

$$p_d = \tau |H_{\mathbf{ii}} - E_{HF} - S|$$



Probability to spawn new walker

$$p_s = \tau \frac{|H_{\mathbf{ij}}|}{p_{gen}(\mathbf{j}|\mathbf{i})}$$

$$\sum_{\mathbf{j}} p_{gen}(\mathbf{j}|\mathbf{i}) = 1$$

$$p_{gen}(\mathbf{j}|\mathbf{i}) \sim (N^2M^2 + NM)^{-1}$$

If Hij < 0 , child has same sign as parent.
If Hij > 0 child has opposite sign of parent

The projected energy

(non-variational)

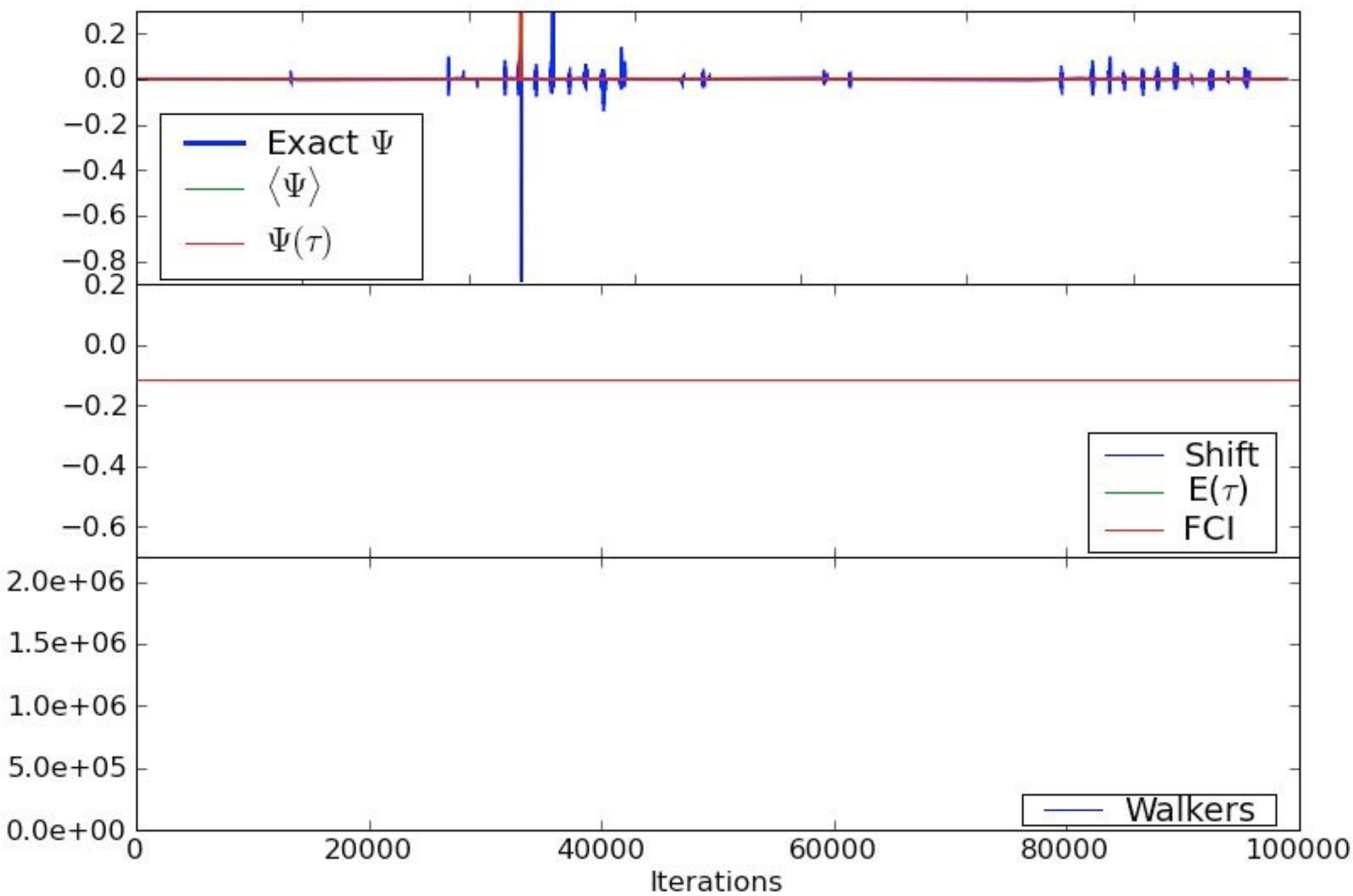
$$E_{proj} = \frac{\langle \Psi_T | H | \Psi \rangle}{\langle \Psi_T | \Psi \rangle}$$

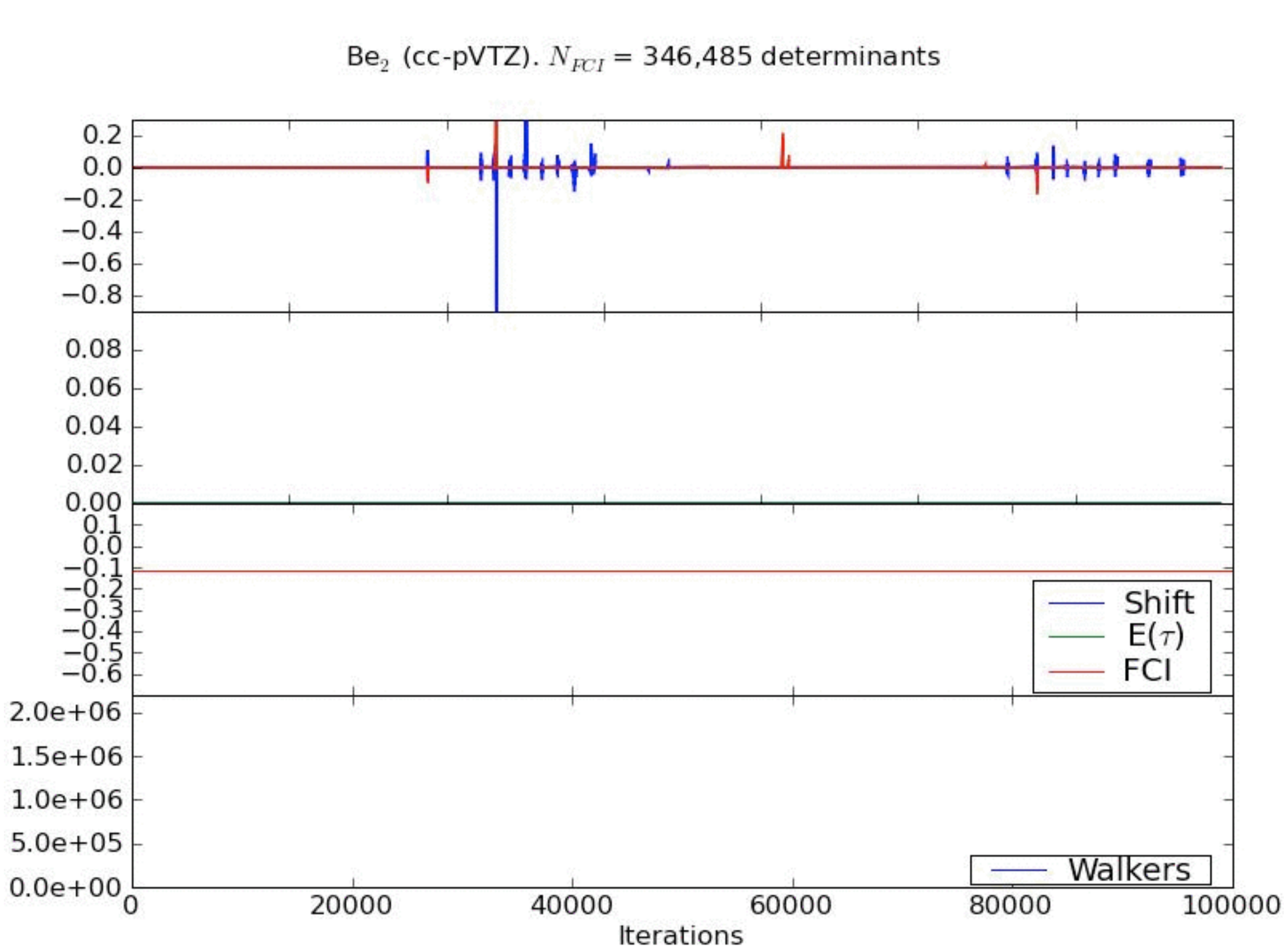
For single reference problems $\Psi_T = D_{HF}$

For multi-reference problems $\Psi_T = \sum_{i \in \mathcal{T}} c_i D_i$

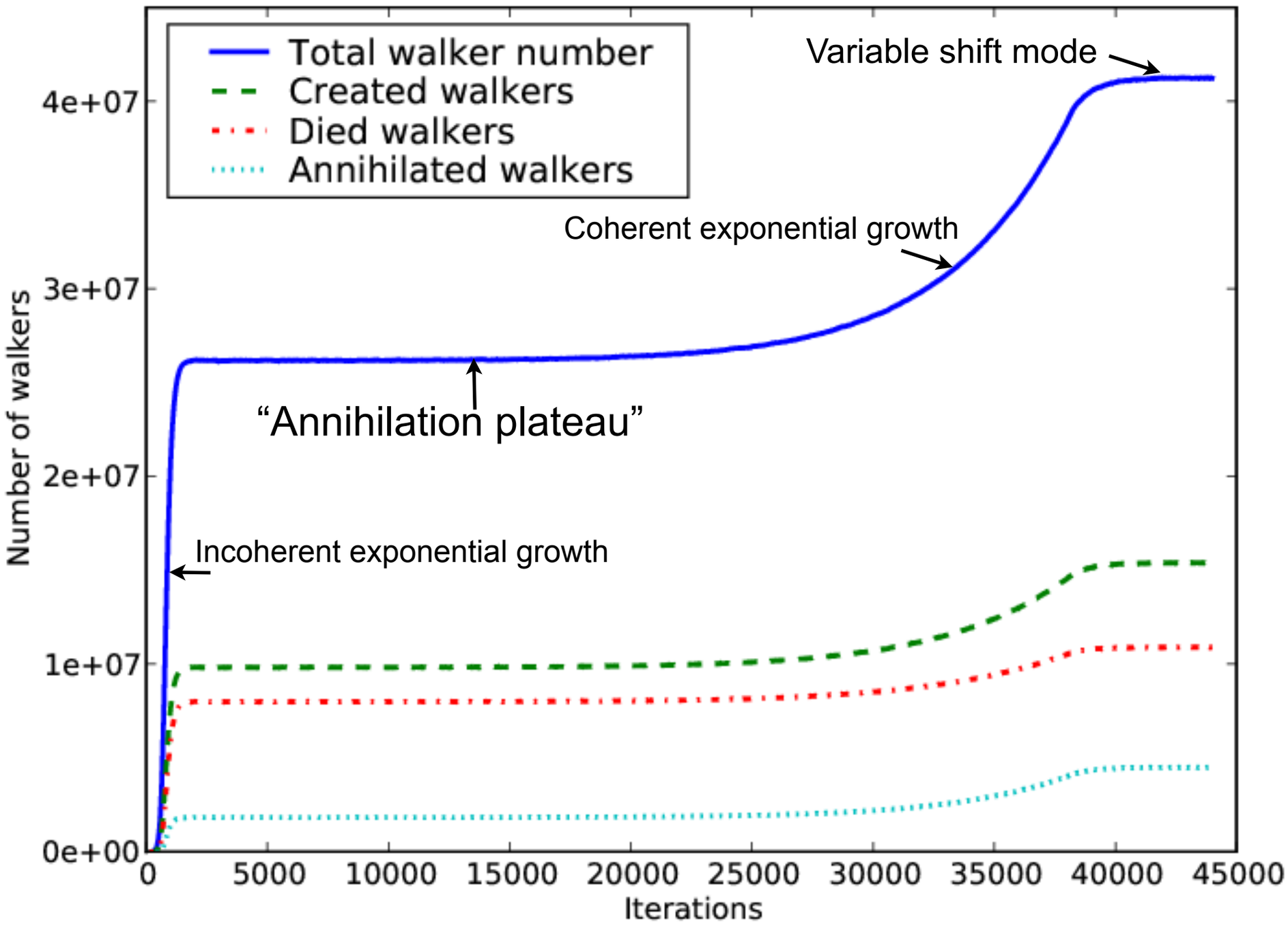
$$E_{var} = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle}$$

Can be estimated via reduced density matrices


Be$_2$ (cc-pVTZ). N_{FCI} = 346,485 determinants
0.2
0.0
−0.2
−0.4
−0.6
−0.8
Exact Ψ
⟨Ψ⟩
Ψ(τ)
0.2
0.0
−0.2
−0.4
−0.6
Shift
E(τ)
FCI
2.0e+06
1.5e+06
1.0e+06
5.0e+05
0.0e+00
Walkers
0
20000
40000
60000
80000
100000
Iterations


Be$_2$ (cc-pVTZ). N_{FCI} = 346,485 determinants
0.2
0.0
−0.2
−0.4
−0.6
−0.8
0.08
0.06
0.04
0.02
0.00
0.1
0.0
−0.1
−0.2
−0.3
−0.4
−0.5
−0.6
Shift
E(τ)
FCI
2.0e+06
1.5e+06
1.0e+06
5.0e+05
0.0e+00
Walkers
0
20000
40000
60000
80000
100000
Iterations

H2O (all electron, cc-pVDZ, 452×10^6 determinants)

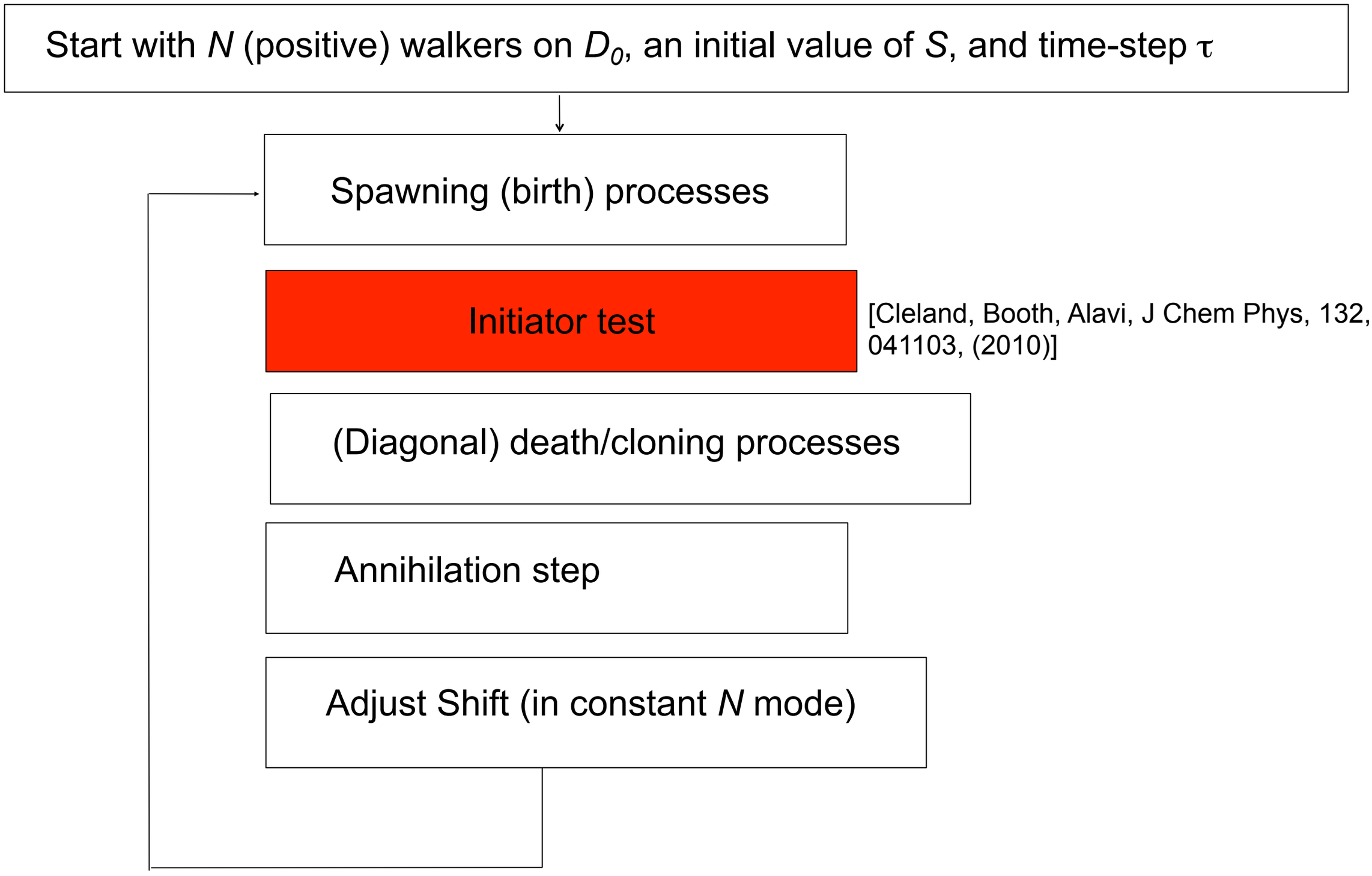


Predicted FCI results from the 2009 paper

TABLE II. Predicted FCI results. The geometries of the molecules were (in Å): CN (1.1941), HF (0.91622), CH_4(r_{CH}=1.087 728), CO (1.1448), H_2O(r_{OH}=0.975 512, θ=110.565°) (Ref. 35), O_2 (1.2074), and NaH (1.885 977). CN and O_2 orbitals were constructed from a restricted open-shell HF calculation with a spin multiplicity of two and three, respectively. CN, CH_4, CO, and O_2 had frozen core electrons. The number in brackets represents the error in the previous digit, obtained through a Flyvbjerg–Petersen blocking analysis (Ref. 37) of $E(\tau)$.

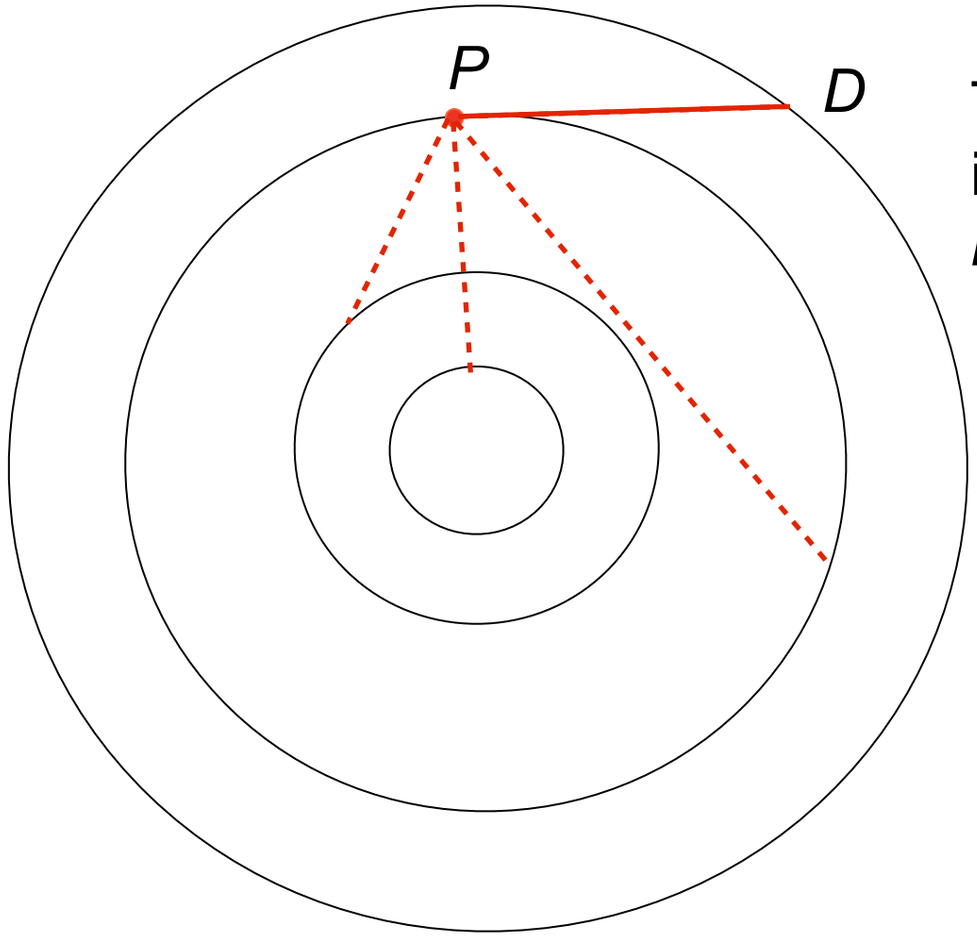
System	(N,M)	$N_{FCI}/10^6$	$N_c/10^6$	f_c	E_{total}	$E_{CCSD(T)}$
Be: cc-V5Z	(4,91)	2.11	0	0	−14.646 38(2)	−14.646 29
CN: cc-pVDZ	(9,26)	246	173	0.704	−92.493 8(3)	−92.491 64
HF: cc-pCVDZ	(10,23)	283	0.998	0.0035	−100.270 98(3)	−100.270 44
CH_4: cc-pVDZ	(8,33)	419	377	0.898	−40.387 52(1)	−40.389 74
CO: cc-pVDZ	(10,26)	1080	777	0.719	−113.056 44(4)	−113.054 97
H_2O: cc-pCVDZ	(10,28)	2410	47.4	0.0196	−76.280 91(3)	−76.280 28
O_2: cc-pVDZ	(12,26)	5409	2651	0.490	−149.987 5(2)	−149.985 62
NaH: cc-pCVDZ	(12,32)	205 300	63.8	0.000 31	−162.609 0(1)	−162.609 01

Overview of initiator-FCIQMC



The initiator test: should the newly spawned walker survive?

If D is empty,
child of P spawned onto D survives
only if P is an initiator ($N_P > n_{add}$)



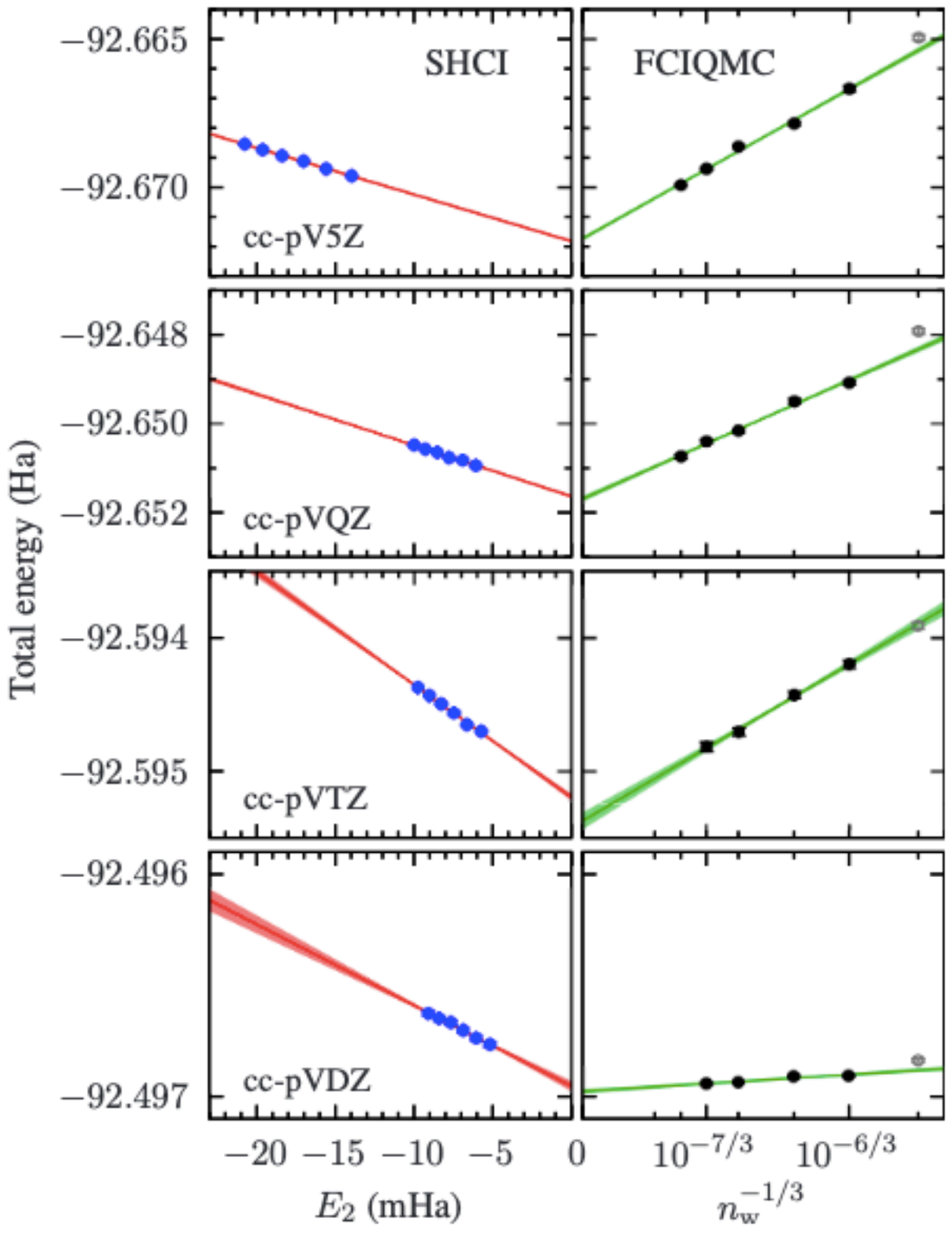
The value of n_{add} is not crucial, as long as it is sensibly chosen. We typically use n_{add} =2 or 3.

Initiators can bring to life new determinants

Demonstration

Extrapolation to the infinite-walker limit using an $N_w^{-1/3}$ law

Haupt et al, arXiv: 2302.13683



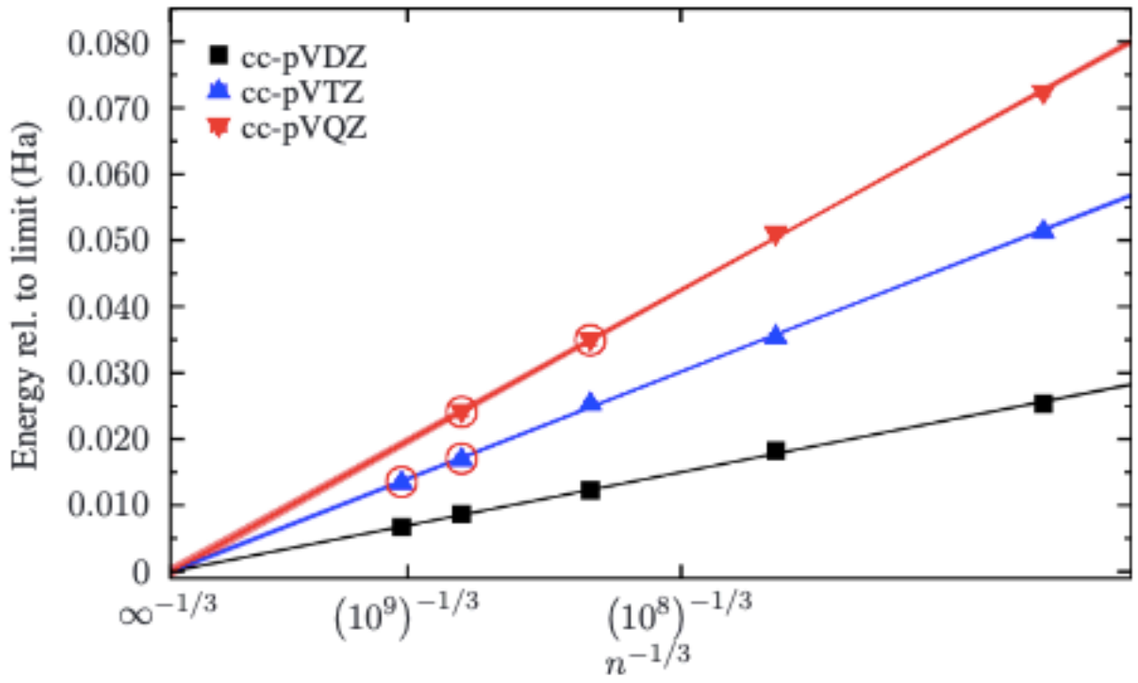
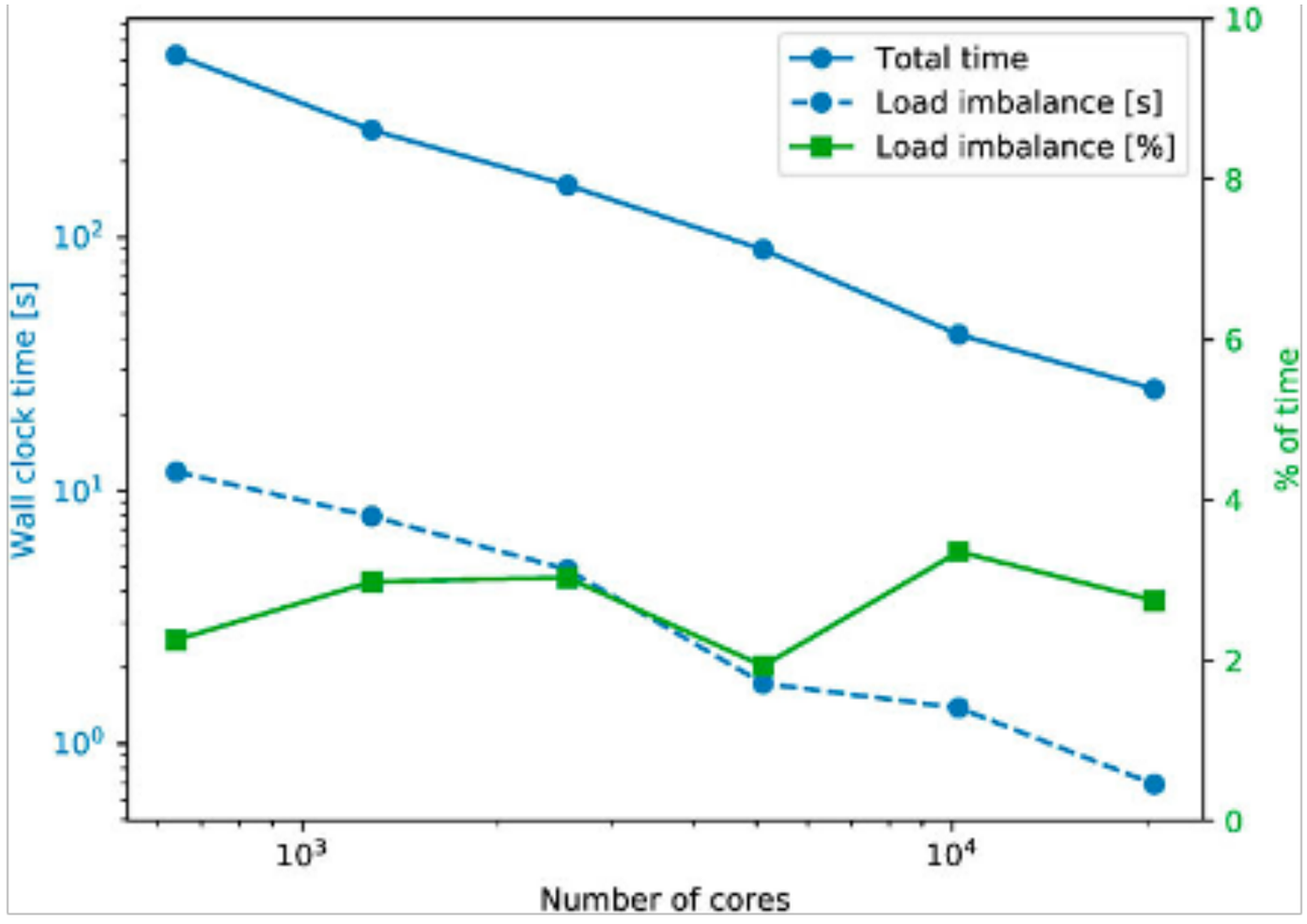


FIG. 14. FCIQMC for Cr_2.

Parallelisability of NECI



Total time and time lost due to load imbalance for running 100 iterations with 1.6×10^9 walkers for the Cr_2/cc-pVDZ (28e in 76o) on 640–20 480 cores (not counting initialization). The calculations were run on Intel Xeon Gold 6148 Skylake processors with a 100 Gb/s OmniPath node interconnect.

Guther et al ; *J. Chem. Phys.* **153,** 034107 (2020) DOI: 10.1063/5.0005754

Transcorrelated Hamiltonians

non-unitary similarity transformations

$$\Psi = e^{\hat{J}}\Phi \qquad \hat{J} = \hat{J}^{\dagger}$$

$$\hat{H}\Psi = E\Psi$$

Only if $\hat{J}^{\dagger} = -\hat{J}$ is the transformation unitary

$$\Rightarrow (e^{-\hat{J}}\hat{H}e^{\hat{J}})\Phi = E\Phi$$

Baker-Campbell-Hausdorff expansion of the similarity-transformed Hamiltonian:

$$e^{-\hat{J}}\hat{H}e^{\hat{J}} \equiv \tilde{H} = \hat{H} + [\hat{H},\hat{J}] + \frac{1}{2!}[[\hat{H},\hat{J}],\hat{J}] + \ldots$$

Two forms of the correlators lead to analytically evaluable (exact) BCH expansion

(1) **Jastrow** correlator, useful for ab initio Hamiltonians, starts in 1st quantisation,

$$J(\mathbf{R}) = \sum_{i<j} u(\mathbf{r}_i, \mathbf{r}_j), \qquad \mathbf{R} = \{\mathbf{r}_1, \ldots, \mathbf{r}_N\}$$

u is real symmetric $[u(\mathbf{r}_i, \mathbf{r}_j) = u(\mathbf{r}_j, \mathbf{r}_i)]$ but not necessarily merely a function of $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$

Sophisticated *u's* suitable for the TC method can be obtained from Variational Monte Carlo (PLR)

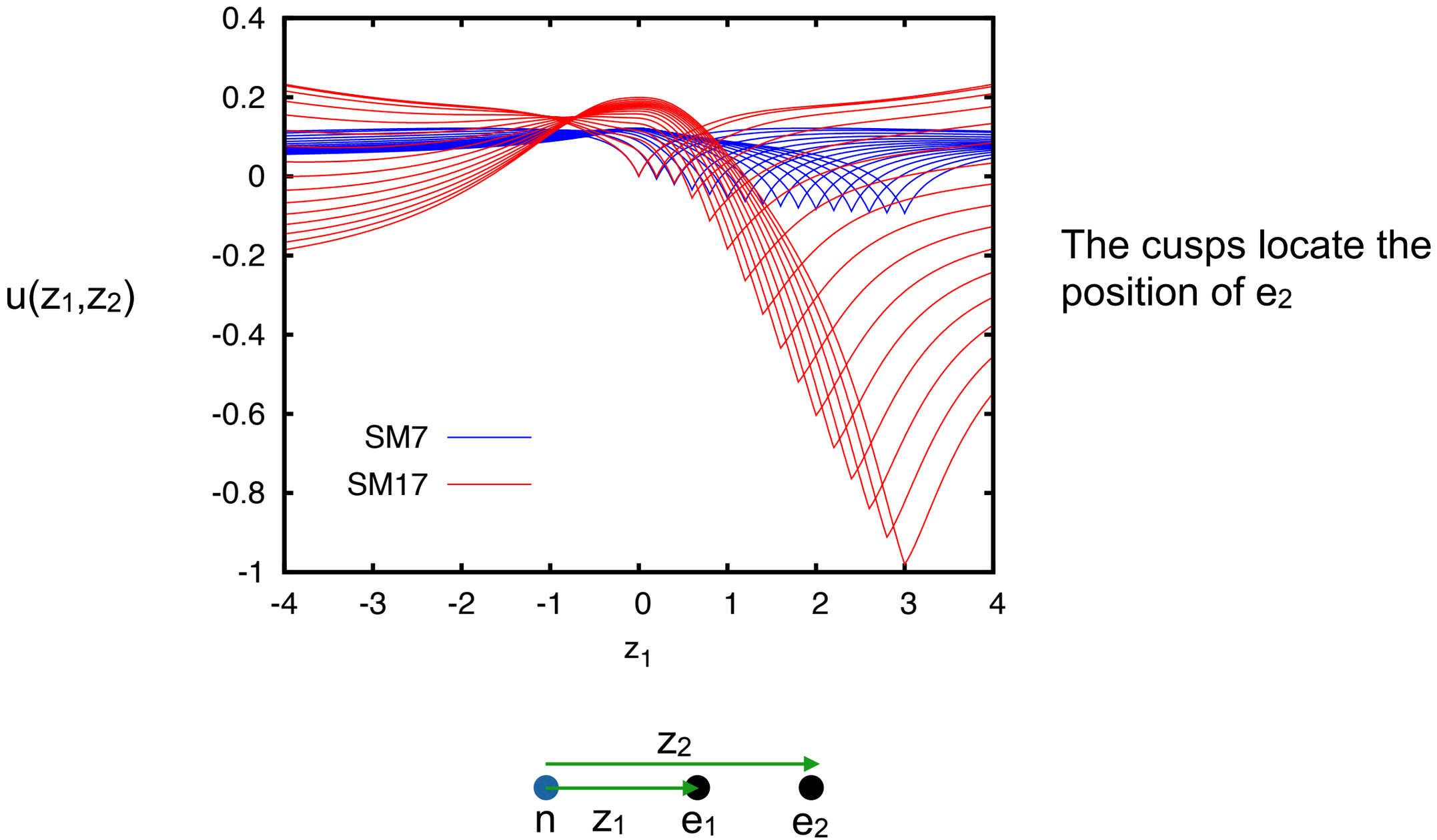
(2) **Gutzwiller** correlator, useful in the Hubbard model, starts in 2nd quantisation

$$\hat{J} = J \sum_i \hat{n}_{i\uparrow}\hat{n}_{i\downarrow}$$

Hubbard model is a good `toy' model to study strong correlation

Dobrautz, Luo, Alavi, PRB **99**, 075119 (2019)

Correlation factor (Ne) with and without e-e-n term



The cusps locate the position of e_2

Jastrow Factorised Similarity Transformation of the S.E.

(Hirschfelder 1963, Boys and Handy 1969)

$$J(\mathbf{R}) = \sum_{i<j} u(\mathbf{r}_i, \mathbf{r}_j), \qquad \mathbf{R} = \{\mathbf{r}_1, \ldots, \mathbf{r}_N\}$$

BCH expansion terminates at second order (kinetic energy is 2nd order one-body differential operator):

$$\tilde{H} = \hat{H} - \sum_i \left(\frac{1}{2} \nabla_i^2 J + (\nabla_i J) \nabla_i + \frac{1}{2} (\nabla_i J)^2 \right)$$

$$= \hat{H} - \sum_{i<j} \hat{K}(\mathbf{r}_i, \mathbf{r}_j) - \sum_{i<j<k} L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k)$$

Jastrow Factorised Similarity Transformation of the S.E.

$$\tilde{H} = \hat{H} - \sum_{i<j} \hat{K}(\mathbf{r}_i, \mathbf{r}_j) - \sum_{i<j<k} L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k)$$

$$\hat{K}(\mathbf{r}_i, \mathbf{r}_j) = \frac{1}{2}\left(\underbrace{\nabla_i^2 u + \nabla_j^2 u} + (\nabla_i u)^2 + (\nabla_j u)^2 \right)$$

$$+\left(\nabla_i u \cdot \nabla_i + \nabla_j u \cdot \nabla_j \right)$$

$$L(\mathbf{r}_i, \mathbf{r}_j, \mathbf{r}_k) = \nabla_i u_{ij} \cdot \nabla_i u_{ik} + \nabla_j u_{ji} \cdot \nabla_j u_{jk} + \nabla_k u_{ki} \cdot \nabla_k u_{kj}$$

The TC Hamiltonian in 2nd quantised form

$$\tilde{H} = \sum_{pq\sigma} h_q^p a_{p\sigma}^\dagger a_{q\sigma} + \frac{1}{2}\sum_{pqrs}(V_{rs}^{pq} - K_{rs}^{pq})\sum_{\sigma\tau} a_{p\sigma}^\dagger a_{q\tau}^\dagger a_{s\tau} a_{r\sigma}$$

$$-\frac{1}{6}\sum_{pqrstu} L_{stu}^{pqr}\sum_{\sigma\tau\lambda} a_{p\sigma}^\dagger a_{q\tau}^\dagger a_{r\lambda}^\dagger a_{u\lambda} a_{t\tau} a_{s\sigma}$$

$$K_{rs}^{pq} = \langle \phi_p \phi_q | \hat{K} | \phi_r \phi_s \rangle \qquad L_{stu}^{pqr} = \langle \phi_p \phi_q \phi_r | L | \phi_s \phi_t \phi_u \rangle$$

The TC integrals are computed using **TCHint**

Excellent two-body-only approximation to the TC Hamiltonian (xTC) exists, and will be covered by Daniel Kats

$\tilde{H}$ is a "pseudo-Hermitian" operator

- The non-Hermitian nature of $\tilde{H}$ has been considered a great source of difficulty in the past, which has prevented the wide-spread adoption of the TC method
- As a non-unitary similarity transformation,

  $\tilde{H}$ is **iso-spectral** with $\hat{H}$
- The eigenvalues of $\tilde{H}$ are real
- However $\tilde{H}$ has distinct **left** and **right**-eigenvectors

$$\tilde{H}\,|\,\Phi^{(R)}\rangle = E\,|\,\Phi^{(R)}\rangle$$

$$\langle\Phi^{(L)}\,|\,\tilde{H} = \langle\Phi^{(L)}\,|\,E$$

- Note that in $\tilde{H}^\dagger$, τ acts in the **wrong** way

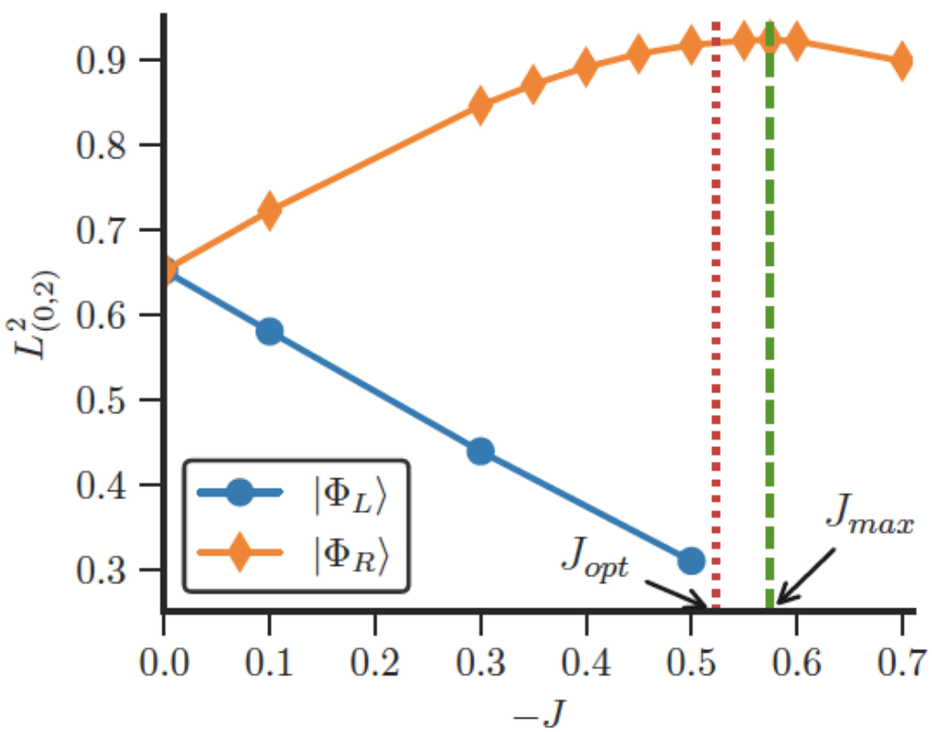
$$\tilde{H}^\dagger \left| \Phi^{(L)} \right\rangle = E \left| \Phi^{(L)} \right\rangle$$

$$\tilde{H}^\dagger = e^J \hat{H} e^{-J}, \text{ i.e. } \quad J \to -J$$

- The left-eigenvector is less compact.
- Therefore approaches which must compute $\left\langle \Phi^{(L)} \right|$ are not ideal (eg bi-variational)
- Need methods which need **only** compute the **right-eigenvector** of $\tilde{H}$

Dobrautz, Luo, Alavi, PRB **99**, 075119 (2019)

18-site 2D Hubbard model, U/t=4



Imaginary-times methods such as FCIQMC can be used as the projective diagonaliser of the similarity transformed Hamiltonian

$$\Psi(\beta) = e^{-\beta(\hat{H}-E_0)}\Psi(0)$$

$$\Psi_0 = \lim_{\beta\to\infty} e^{-\beta(\hat{H}-E_0)}\Psi(0)$$

$$\Psi(\beta) = e^{\hat{J}}\Phi(\beta)$$

$$\Phi(\beta) = e^{-\beta(\tilde{H}-E_0)}\Phi(0)$$

$$\Phi_0 = \lim_{\beta\to\infty} e^{-\beta(\tilde{H}-E_0)}\Phi(0)$$

Strategies and approximations solve transcorrelated Hamiltonians

- Transcorrelated FCIQMC

$$\Phi^{(R)} = \sum_I C_I \,|\, D_I\rangle$$

Main use is in **multi-reference problems**

- Transcorrelated CC $\quad \Phi^{(R)} = e^{\hat{T}} \,|\, D_0\rangle, \quad \hat{T} \approx \hat{T}_1 + \hat{T}_2$

  Main use is for **weak/medium correlation**

- Transcorrelated Perturbation theory: test of behaviour for metals

Non-hermitian character TC Hamiltonian does not cause a problem for the above methodologies.

Proof

[Luo, Alavi, JCTC, **14**, 1403, (2018)]

$$e^{\hat{J}}\Phi(\beta) = \Psi(\beta) = e^{-\beta(\hat{H}-E_0)}\Psi(0) = e^{-\beta(\hat{H}-E_0)}e^{\hat{J}}\Phi(0)$$

$$\Rightarrow \Phi(\beta) = e^{-\hat{J}}e^{-\beta(\hat{H}-E_0)}e^{\hat{J}}\Phi(0)$$

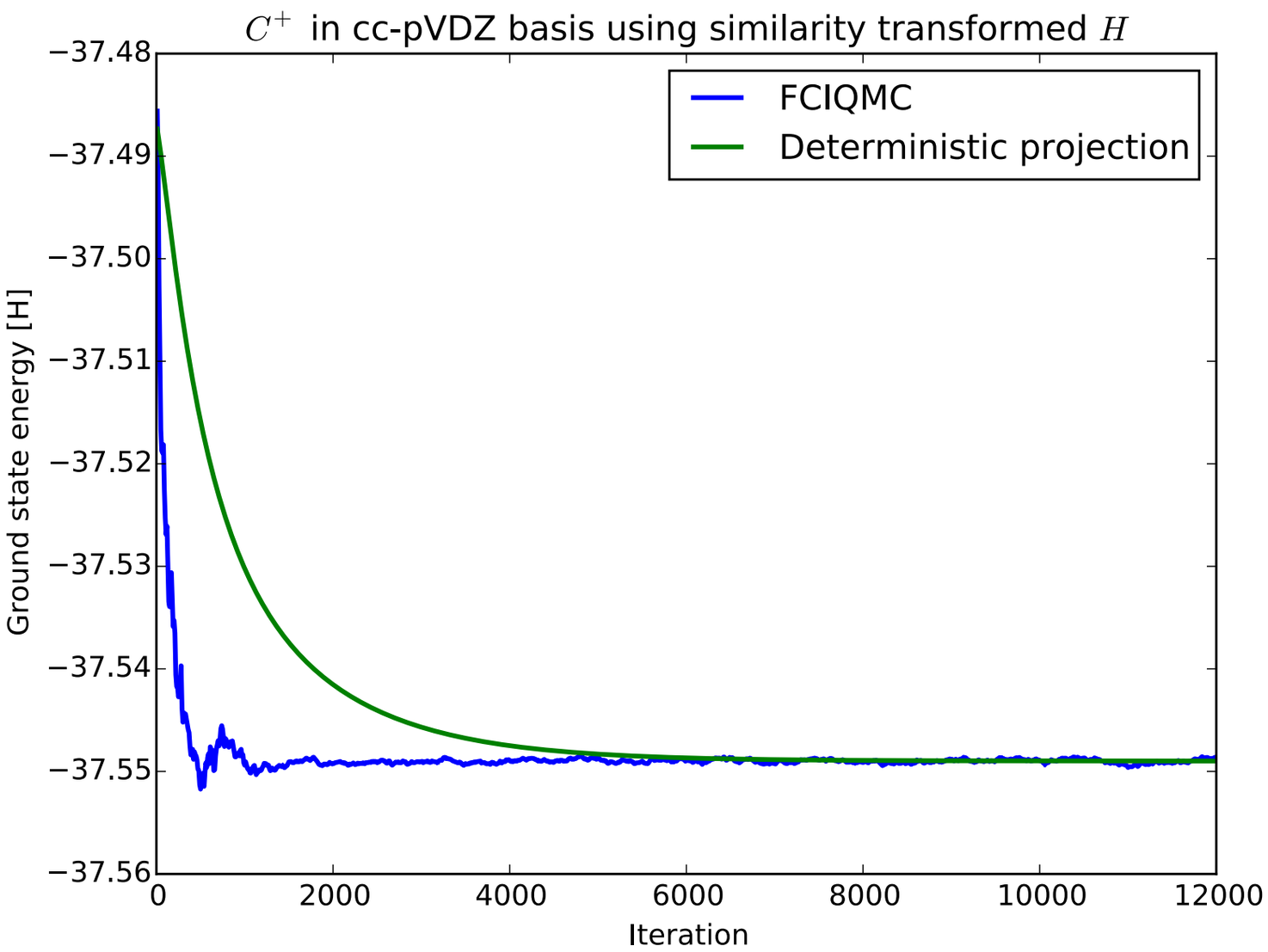
$$= \lim_{M\to\infty} e^{-\hat{J}}\left(1-\frac{\beta}{M}\left(\hat{H}-E_0\right)\right)^M e^{\hat{J}}\Phi(0)$$

$$= \lim_{M\to\infty} e^{-\hat{J}}\left(1-\frac{\beta}{M}\left(\hat{H}-E_0\right)\right)e^{\hat{J}}e^{-\hat{J}}\left(1-\frac{\beta}{M}\left(\hat{H}-E_0\right)\right)\ldots e^{\hat{J}}\Phi(0)$$

$$= \lim_{M\to\infty}\left(1-\frac{\beta}{M}\left(e^{-\hat{J}}\hat{H}e^{\hat{J}}-E_0\right)\right)^M\Phi(0)$$

$$= e^{-\beta(\tilde{H}-E_0)}\Phi(0)$$

Imaginary time propagation with $\tilde{H}$ leads to the ground state!



Boys-Handy form for *u*

$$u(\mathbf{r}_i, \mathbf{r}_j) = \sum_{\substack{mno \\ m+n+o \le 6}} c_{nml}(\bar{r}_i^m \bar{r}_j^n + \bar{r}_j^m \bar{r}_i^n)\bar{r}_{ij}^o$$

Includes e-e, e-n, and e-e-n terms

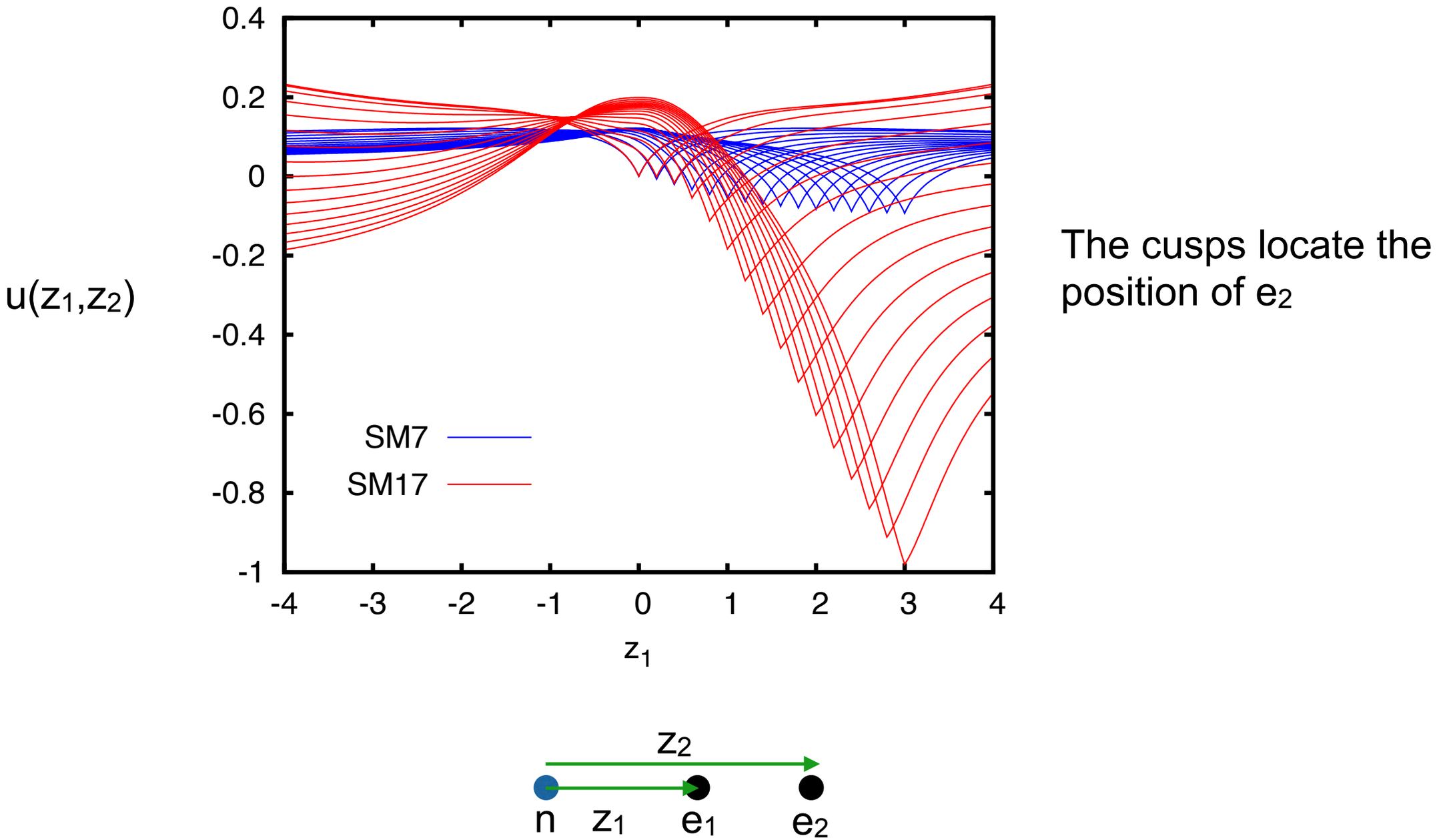
$$\bar{r} = \frac{r}{1+r}$$

$\bar{r} \approx r - r^2$ for small r

$\bar{r} \approx 1 - 1/r \to 1$ for large r

For the first-row atoms, the 17 parameters have been obtained by a **Variance minimisation** VMC by Schmidt and Moskowitz, JCP, **93**, 4172 (1990)

Correlation factor (Ne) with and without e-e-n term (SM17 vs SM7)



The cusps locate the position of e_2

First application of TC-FCIQMC: First row atoms

Cohen, Luo, Guther, Dobrautz, Tew, Alavi, JCP **151**, 0161101 (2019)

TABLE I. Total atomic energies (Hartrees), for CCSD(T), UCSSD(T)-F12, and the ST Hamiltonian, using the SM7, and SM17 correlation factors. The Mean Absolute Error (MAE) for each method across the series is also shown.

basis	Li	Be	B	C	N	O	F	Ne	MAE/H
CCSD(T)									
cc-pVDZ	-7.43264	-14.61741	-24.59026	-37.76156	-54.47994	-74.911155	-99.52932	-128.68069	0.121
cc-pVTZ	-7.44606	-14.62379	-24.60538	-37.78953	-54.52487	-74.98494	-99.63219	-128.81513	0.069
cc-pVQZ	-7.44983	-14.64008	-24.62350	-37.81209	-54.55309	-75.02319	-99.68158	-128.87676	0.039
F12									
cc-pVDZ	-7.47458	-14.65400	-24.63121	-37.80901	-54.53707	-74.99208	-99.63623	-128.81125	0.053
cc-pVTZ	-7.47267	-14.65653	-24.63626	-37.81883	-54.55293	-75.01752	-99.66994	-128.85890	0.036
cc-pVQZ	-7.47370	-14.65933	-24.64187	-37.82884	-54.56916	-75.04056	-99.70070	-128.89816	0.020
SM7									
cc-pVDZ	-7.46726	-14.65517	-24.63279	-37.81469	-54.53448	-74.97785	-99.60602	-128.78385	0.063
cc-pVTZ	-7.47627	-14.65943	-24.64458	-37.83703	-54.57236	-75.04055	-99.69421	-128.89389	0.019
cc-pVQZ	-7.47785	-14.66791	-24.65417	-37.84791	-54.58778	-75.06296	-99.72507	-128.92967	0.003
SM17									
cc-pVDZ	-7.47707	-14.66793	-24.64521	-37.82772	-54.55719	-75.01639	-99.65834	-128.83682	0.036
cc-pVTZ	-7.47804	-14.66789	-24.65003	-37.83928	-54.57989	-75.05303	-99.71377	-128.90944	0.010
cc-pVQZ	-7.47845	-14.66749	-24.65287	-37.84461	-54.58844	-75.06609	-99.73283	-128.93542	0.001
Expt	-7.47806	-14.66736	-24.65391	-37.84500	-54.58920	-75.06730	-99.73390	-128.93760	

The role of the *K* and *L* terms

Ne atom cc-pVQZ with SM17 correlation factor

$$\langle D_{HF} | \hat{K} | D_{HF} \rangle = -382 \text{ mH}$$

$$\langle D_{HF} | L | D_{HF} \rangle = +108 \text{ mH}$$

The effect of the three-body (*L*) terms is to raise the energy, countering the large negative (non-Hermitian) contributions coming from the two-body (*K*) terms.

It is necessary to have an accurate treatment of the 3-body terms

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