A Parallel Universal Configuration Interaction Engine

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1 Notations

- μ , ν , λ , σ: atomic orbitals (AOs)
- p, q, r, s: general MOs
- t, u, v, w: model space MOs
- *I, J, K, L*: Slater determinants
- b_p: the occupancy status of MO p, which can be either 0 or 1, representing unoccupied and occupied orbital, respectively.
- $\boldsymbol{b}$ in bold font: orbitals associated with excitation operator
- $|...b_p\boldsymbol{b_q}b_rb_s...\rangle$: binary representations of determinants, where the order of orbitals is from right to left.

2 Review on Current Efficient Implementations on $\boldsymbol{\sigma}$ = HC

2.1 CI Electronic Hamiltonian and $\boldsymbol{\sigma}$ Vector

The electronic Hamiltonian for the correlated space has the form,

$$\hat{H} = \sum_{pq} h_{pq}^{\mathrm{FC}} \hat{E}_{pq} + \frac{1}{2} \sum_{pqrs} (pq|rs) \hat{e}_{pqrs} \tag{1}$$

$$h_{pq}^{\mathrm{FC}} = h_{pq} + \sum_{r'} [(pq|r'r') - (pr'|r'q)] \tag{2}$$

where r' belong to the frozen core space, and $\{p, q, r, s\}$ are in the correlated space. Equation (2) describes the interaction between electrons in the frozen core space and those in the correlated space. The total MRCI energy is then defined as

$$E = \langle \Psi_{\mathrm{MRCI}} | \hat{H} | \Psi_{\mathrm{MRCI}} \rangle + E^{\mathrm{FC}} \tag{3}$$

$$E^{\mathrm{FC}} = \sum_{p'} h_{p'p'} + \sum_{p'>q'} [(p'p'|q'q') - (p'q'|q'p')] \tag{4}$$

where E^{FC}, computed using reference (HF or CASSCF) orbitals, is the energetic contribution from the frozen core space.

The rapid scaling in CI expansion usually requires a careful selection of correlated space and the use of iterative diagonalization approaches. In this work the matrix-vector product,

$$\sigma_K = \sum_L H_{KL} C_L \tag{5}$$

is computed *on-the-fly*.

Plugging the electronic Hamiltonian, Eq. (1), into Eq. (5), we can obtain the $\boldsymbol{\sigma}$ vectors

in the form of

$$\sigma_K = \sum_{pq} \sum_{L} h_{pq}^{\mathrm{FC}} \langle K| \hat{E}_{pq} |L\rangle C_L + \frac{1}{2} \sum_{pqrs} \sum_{L} (pq|rs) \langle K| \hat{e}_{pqrs} |L\rangle C_L \tag{6}$$

2.2 The Factorized Methods and the Sparsity of the Electron Interactions

In the computation of $\boldsymbol{\sigma}$, the sparsity of the electron interactions is utilized as:

$$\begin{array}{c} \langle K| \hat{E}_{pq} |L\rangle \\ (\text{or } \langle K| \hat{e}_{pqrs} |L\rangle) \end{array} = \begin{cases} \pm 1, & \text{when } |K\rangle \text{ and } |L\rangle \text{ only differ on } b_p, b_q, (b_r, b_s) \\ 0, & \text{otherwise} \end{cases} \tag{7}$$

In Eq. (6), the outer summation over the configurations yields the configuration-driven algorithm, while over MO indices yields the integral-driven algorithm. In either cases, one-electron excitation lists containing the information of non-zero elements of $\langle K| \hat{E}_{tu} |L\rangle$, *i.e.*, $\{K, L, t, u, \langle K| \hat{E}_{tu} |L\rangle\}$, is pre-computed and stored in core. To avoid the additional storage for $\langle K| \hat{e}_{tuwv} |L\rangle$, the two-electron excitation operator can be factorized into multiplications of one-electron operators *via*

$$\hat{e}_{pqrs} = \hat{E}_{pq}\hat{E}_{rs} - \delta_{qr}\hat{E}_{ps} \tag{8}$$

As the result, the $\boldsymbol{\sigma}$ vector now has the form of:

$$\begin{aligned} \sigma_K =& \sum_{pq} \sum_{L} h_{pq}^{\mathrm{FC}} \langle K| \hat{E}_{pq} |L\rangle C_L + \frac{1}{2} \sum_{pqrs} (pq|rs)\delta_{qr} \langle K| \hat{E}_{ps} |L\rangle C_L \\ &+ \frac{1}{2} \sum_{pqrs} \sum_{L} (pq|rs) \langle K| \hat{E}_{pq}\hat{E}_{rs} |L\rangle C_L \qquad (9) \\ =& \sum_{pq} \sum_{L} h'_{pq} \langle K| \hat{E}_{pq} |L\rangle C_L + \frac{1}{2} \sum_{pqrs} \sum_{L} \sum_{J} (pq|rs) \langle K| \hat{E}_{pq} |J\rangle \langle J| \hat{E}_{rs} |L\rangle C_L \qquad (10) \end{aligned}$$

where the h'_{pq} is defined as

$$h'_{pq} = h_{pq}^{\mathrm{FC}} - \frac{1}{2} \sum_{r} (pr|rq) \tag{11}$$

2.3 Matrix Form of $\boldsymbol{\sigma} = \mathbf{HC}$

In order to take advantages of the existing highly optimized linear algebra libraries, such as basic linear algebra subprograms (BLAS) and linear algebra packages (LAPACK), efficient algorithms for updating $\boldsymbol{\sigma}$ vector were implemented in the form of matrix-vector or matrix-matrix multiplications. Equation (10) could be separated as one-electron and two-electron contributions:

$$ {}^{1e}\sigma_K = \sum_{pq} \sum_{L} h'_{pq} \langle K| \hat{E}_{pq} |L\rangle C_L \tag{12} $$

$$ {}^{2e}\sigma_K = \frac{1}{2} \sum_{pqrs} \sum_{L} \sum_{J} (pq|rs) \langle K| \hat{E}_{pq} |J\rangle \langle J| \hat{E}_{rs} |L\rangle C_L \tag{13} $$

The one-electron symbolic matrix elements is defined as:

$$ A^{pq}_{KL} = \langle K| \hat{E}_{pq} |L\rangle \tag{14} $$

Then, two intermediate matrices could be defined:

$$ \Omega_{KL} = \sum_{pq} h'_{pq} A^{pq}_{KL} \tag{15} $$

$$ \Lambda^{pq}_{JL} = \sum_{rs} (pq|rs) A^{rs}_{JL} \tag{16} $$

Thus, in the matrix form, ${}^{1e}\boldsymbol{\sigma}$ and ${}^{2e}\boldsymbol{\sigma}$ are computed as:

$$ {}^{1e}\boldsymbol{\sigma} = \boldsymbol{\Omega} \cdot \mathbf{C} \tag{17} $$

$$ {}^{2e}\boldsymbol{\sigma} = \frac{1}{2} \sum_{pq} \boldsymbol{A}^{pq} \cdot \boldsymbol{\Lambda}^{pq} \cdot \mathbf{C} = \frac{1}{2} \sum_{pq} \boldsymbol{A}^{pq} \cdot \ {}^{2e}\boldsymbol{\sigma}^{pq} \tag{18} $$

2.4 Current Efficient Implementations for real-RHF CASCI

Practically, the major concern in terms of implementations of above matrix-vector product is the storage of the one-electron symbolic matrix and those intermediate matrices. For example, in RHF framework, a CAS(10,10) calculation yields the number of configurations as $N = 65536$, and the dimension of each matrix become 65536×65536 ($\sim$34 GBytes Hang ▶*from shaopeng's thesis*◀). Besides, in this form, the sparsity of the symbolic matrix element (Eq. (7)) is not used.

In RHF framework, the major feature of the electronic configuration is that electron occupancy could be separated based on their spins and no spin-flip excitation is allowed. Thus, each electron configuration could be decomposed as a combination of a alpha electron configuration and a beta one, and so does the excitation operators.

$$|K\rangle = |K_\alpha K_\beta\rangle \tag{19}$$

$$\hat{E}_{pq} = \hat{E}^\alpha_{pq} + \hat{E}^\beta_{pq} \tag{20}$$

In a CASCI calculations, it also very convenient to write $\boldsymbol{\sigma}$ and $\mathbf{C}$ in the matrix form with dimension $N_\alpha \times N_\beta$. Then, Eqs. (12) and (13) could be re-written as:

$$\begin{aligned} {}^{1e}\sigma(K_\alpha, K_\beta) = &\sum_{pq}\sum_{L_\alpha} h'_{pq} \langle K_\alpha| \hat{E}^\alpha_{pq} |L_\alpha\rangle C(L_\alpha, K_\beta) \\ &+ \sum_{pq}\sum_{L_\beta} h'_{pq} \langle K_\beta| \hat{E}^\beta_{pq} |L_\beta\rangle C(K_\alpha, L_\beta) \end{aligned} \tag{21}$$

$$
\begin{aligned}
{}^{2e}\sigma(K_\alpha, K_\beta) =& \frac{1}{2}\sum_{pqrs}\sum_{L_\alpha}\sum_{J_\alpha}(pq|rs)\left\langle K_\alpha\right|\hat{E}^\alpha_{pq}\left|J_\alpha\right\rangle\left\langle J_\alpha\right|\hat{E}^\alpha_{rs}\left|L_\alpha\right\rangle C(L_\alpha, K_\beta) \\
&+ \frac{1}{2}\sum_{pqrs}\sum_{L_\beta}\sum_{J_\beta}(pq|rs)\left\langle K_\beta\right|\hat{E}^\beta_{pq}\left|J_\beta\right\rangle\left\langle J_\beta\right|\hat{E}^\beta_{rs}\left|L_\beta\right\rangle C(K_\alpha, L_\beta) \\
&+ \frac{1}{2}\sum_{pqrs}\sum_{L_\alpha}\sum_{L_\beta}(pq|rs)\left\langle K_\alpha\right|\hat{E}^\alpha_{pq}\left|L_\alpha\right\rangle\left\langle K_\beta\right|\hat{E}^\beta_{rs}\left|L_\beta\right\rangle C(L_\alpha, L_\beta) \\
&+ \frac{1}{2}\sum_{pqrs}\sum_{L_\alpha}\sum_{L_\beta}(pq|rs)\left\langle K_\beta\right|\hat{E}^\beta_{pq}\left|L_\beta\right\rangle\left\langle K_\alpha\right|\hat{E}^\alpha_{rs}\left|L_\alpha\right\rangle C(L_\alpha, L_\beta) \qquad (22)
\end{aligned}
$$

The one-electron symbolic matrix elements are now defined in spin form as:

$$
\boldsymbol{A}^{pq}_{\gamma\gamma} : A^{pq}_{K_\gamma L_\gamma} = \left\langle K_\gamma\right|\hat{E}^\gamma_{pq}\left|L_\gamma\right\rangle \qquad (23)
$$

Then the two intermediate matrices are now:

$$
\boldsymbol{\Omega}_{\gamma\gamma} = \sum_{p_\gamma q_\gamma} h'_{p_\gamma q_\gamma}\boldsymbol{A}^{pq}_{\gamma\gamma} \qquad (24)
$$

$$
\boldsymbol{\Lambda}^{p_\gamma q_\gamma}_{\gamma'\gamma'} = \sum_{r_{\gamma'}s_{\gamma'}}(p_\gamma q_\gamma|r_{\gamma'}s_{\gamma'})\boldsymbol{A}^{rs}_{\gamma'\gamma'} \qquad (25)
$$

And in matrix form, the $\boldsymbol{\sigma}$ becomes:

$$
{}^{1e}\boldsymbol{\sigma}_{\alpha\beta} = \boldsymbol{\Omega}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \left[\boldsymbol{\Omega}_{\beta\beta}\cdot\mathbf{C}^{\mathrm{T}}_{\alpha\beta}\right]^{\mathrm{T}} = \boldsymbol{\Omega}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \cdot\mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Omega}^{\mathrm{T}}_{\beta\beta} \qquad (26)
$$

$$
\begin{aligned}
{}^{2e}\boldsymbol{\sigma}_{\alpha\beta} =& \frac{1}{2}\sum_{p_\alpha q_\alpha}\boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \frac{1}{2}\left[\sum_{p_\beta q_\beta}\boldsymbol{A}^{pq}_{\beta\beta}\cdot\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\beta\beta}\cdot\mathbf{C}^{\mathrm{T}}_{\alpha\beta}\right]^{\mathrm{T}} \\
&+ \frac{1}{2}\sum_{p_\alpha q_\alpha}\boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\left[\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\beta\beta}\cdot\mathbf{C}^{\mathrm{T}}_{\alpha\beta}\right]^{\mathrm{T}} + \frac{1}{2}\left[\sum_{p_\beta q_\beta}\boldsymbol{A}^{pq}_{\beta\beta}\cdot\left[\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta}\right]^{\mathrm{T}}\right]^{\mathrm{T}} \\
=& \frac{1}{2}\sum_{p_\alpha q_\alpha}\boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \frac{1}{2}\sum_{p_\beta q_\beta}\mathbf{C}_{\alpha\beta}\cdot\left[\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\beta\beta}\right]^{\mathrm{T}}\cdot\left[\boldsymbol{A}^{pq}_{\beta\beta}\right]^{\mathrm{T}} \\
&+ \frac{1}{2}\sum_{p_\alpha q_\alpha}\boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta}\cdot\left[\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\beta\beta}\right]^{\mathrm{T}} + \frac{1}{2}\sum_{p_\beta q_\beta}\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta}\cdot\left[\boldsymbol{A}^{pq}_{\beta\beta}\right]^{\mathrm{T}} \qquad (27)
\end{aligned}
$$

In the real RHF framework the intermediate matrice have properties as:

$$\left[\boldsymbol{A}^{pq}_{\gamma\gamma}\right]^{\mathrm{T}} = \boldsymbol{A}^{qp}_{\gamma\gamma} \tag{28}$$

$$\boldsymbol{\Omega}^{\mathrm{T}}_{\gamma\gamma} = \boldsymbol{\Omega}_{\gamma\gamma} \tag{29}$$

$$\left[\boldsymbol{\Lambda}^{p_\gamma q_\gamma}_{\gamma'\gamma'}\right]^{\mathrm{T}} = \boldsymbol{\Lambda}^{q_\gamma p_\gamma}_{\gamma'\gamma'} = \boldsymbol{\Lambda}^{p_\gamma q_\gamma}_{\gamma'\gamma'} \tag{30}$$

Then, Eqs. (26) and (27) could be further simplified as:

$$^{1e}\boldsymbol{\sigma}_{\alpha\beta} = \boldsymbol{\Omega}_{\alpha\alpha}\mathbf{C}_{\alpha\beta} + \cdot\mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Omega}_{\beta\beta} \tag{31}$$

$$\begin{aligned}
^{2e}\boldsymbol{\sigma}_{\alpha\beta} =& \frac{1}{2}\sum_{p_\alpha q_\alpha} \boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \frac{1}{2}\sum_{p_\beta q_\beta}\mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\beta\beta}\cdot\boldsymbol{A}^{pq}_{\beta\beta} \\
&+ \frac{1}{2}\sum_{p_\alpha q_\alpha} \boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Lambda}^{q_\alpha p_\alpha}_{\beta\beta} + \frac{1}{2}\sum_{p_\beta q_\beta}\boldsymbol{\Lambda}^{p_\beta q_\beta}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta}\cdot\boldsymbol{A}^{qp}_{\beta\beta} \\
=& \frac{1}{2}\sum_{p_\alpha q_\alpha} \boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\left[\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Lambda}^{p_\alpha q_\alpha}_{\beta\beta}\right] + \frac{1}{2}\sum_{p_\beta q_\beta}\left[\boldsymbol{\Lambda}^{q_\beta p_\beta}_{\alpha\alpha}\cdot\mathbf{C}_{\alpha\beta} + \mathbf{C}_{\alpha\beta}\cdot\boldsymbol{\Lambda}^{q_\beta p_\beta}_{\beta\beta}\right]\cdot\boldsymbol{A}^{pq}_{\beta\beta} \\
=& \frac{1}{2}\sum_{p_\alpha q_\alpha} \boldsymbol{A}^{pq}_{\alpha\alpha}\cdot\ {}^{2e}\boldsymbol{\sigma}^{p_\alpha q_\alpha}_{\alpha\beta} + \frac{1}{2}\sum_{p_\beta q_\beta}\ {}^{2e}\boldsymbol{\sigma}^{p_\beta q_\beta}_{\alpha\beta}\cdot\boldsymbol{A}^{pq}_{\beta\beta}
\end{aligned} \tag{32}$$

In this way, the dimension of those intermediate matrices are now only $N_\alpha \times N_\alpha$ or $N_\beta \times N_\beta$, which can be easily stored up to CAS(18,18). With the scaling still being large for CASCI, it would still be very difficult to tackle extremely-large CASCI problem. And for non-CASCI, writing $\boldsymbol{\sigma}$ and $\mathbf{C}$ in the matrix form of alpha and beta spin would create vast number of zero matrix elements, resulting in severe compromise of efficiency. For GHF calculations, it is impossible to decompose the electron configuration by their spins, because of the use of spinor orbitals.

3 A Parallel Universal CI Engine

3.1 Excess Information in Electron Symbolic Matrix Elements

The non-zero one-electron symbolic matrix elements is normally computed as:

$$\forall p < q : A_{KL}^{pq} = \langle K | \, a_p^\dagger a_q \, | L \rangle = \begin{cases} +1, \text{ if } \sum_{i=p+1}^{q-1} b_i \text{ is even} \\ -1, \text{ if } \sum_{i=p+1}^{q-1} b_i \text{ is odd} \end{cases} \tag{33}$$

Obviously, a set of pairs of K and L will share same A_{KL}^{pq} value, if they meet conditions that 1) K and L only differ on electron occupancy b_p and b_q; 2) the number of electrons between p and q are same. By group those pair of K and L, it offers the potential to build intermediate matrices with dimensions that are only subset of the total space, like the separation of alpha and beta spins in Sec. 2.4. To generalize this idea for arbitrary separation of electronic configurations, the concepts in generalized active space (GAS) methods are adapted.

3.2 String Methods in the context of GAS

Similar to RAS, in GAS calculations, the total correlation space can now be partitioned into any arbitrary number of active spaces, with occupation restrictions within each GAS, and/or excitation restrictions between any 2 GAS imposed. The basic concepts and notations are defined as:

- GAS X: denoted as $\mathbb{G}_X$ and indexed by X, is a set of molecular orbitals or spinor orbitals.
- Category Y: denoted as $\mathbb{D}_Y$ and indexed by Y, is the set of electron determinants with fixed electron occupancy in each GAS. $\mathbb{D}_Y^X$ is the set of electron configurations in GAS X of $\mathbb{D}_Y$.
- Categorical excitation set and operator: denoted as $\mathbb{E}_{XX'}$ and $\hat{\mathbb{E}}_{XX'}$, respectively. $\mathbb{E}_{XX'}$ is a set of all possible excitation operators with one electron excited from GASX' to

GASX and $\hat{\mathbb{E}}^{XX'}$ is the sum of all the elements in $\mathbb{E}_{XX'}$:

$$\mathbb{E}_{XX'} = \left\{ \hat{E}_{pq} \middle| p \in \mathbb{G}_X \text{ and } q \in \mathbb{G}_{X'} \right\} \tag{34}$$

- $K_Y^{X\gamma}$: Sub-string with γ spin in GAS X of category Y;
- n, n_Y^X: number of electrons in full space or in GAS X of category Y;
- $n_{\min}^X$, $n_{\max}^X$: minimal and maximum electrons allowed in GAS X
- m, m^X: number of orbitals in full space or GAS X;
- M: number of GAS in full space;
- $A(K)$, $A^{\mathbb{D}}(K), A^{\mathbb{G}}(K_Y^X)$: global address of electronic configuration $|K\rangle$, address of $|K\rangle$ its category or local address of $\left|K_Y^X\right\rangle$ in its GAS X and category Y.

To build up the full space, a reference category ($\mathbb{D}_0$) is constructed by applying all the intra-GAS excitation on the reference HF or CASSCF configuration $|0\rangle$:

$$\mathbb{D}_0 = \left\{ \hat{E}_{pq} |0\rangle \middle| \hat{E}_{pq} \in \bigcup_X \mathbb{E}_{XX} \right\} \tag{35}$$

Then, all other categories are built by applying inter-GAS excitation operators. Note if occupation restrictions are imposed, they are also re-interpreted as excitation restrictions. Based on excitation levels, each singly ($\mathbb{D}(X_1X_1')$), doubly ($\mathbb{D}(X_1X_1', X_2X_2')$),..., excited categories are built as:

$$\forall(X_1 \neq X_1') : \quad \mathbb{D}(X_1X_1') = \left\{ \hat{E}_{pq} |I\rangle \middle| |I\rangle \in \mathbb{D}_0 \text{ and } \hat{E}_{pq} \in \mathbb{E}_{X_1X_1'} \right\} \tag{36}$$

$$\begin{aligned} \forall(X_2 \neq X_2' \quad & \wedge X_2 \geq X_1 \wedge X_2' \geq X_1' \wedge X_2 \neq X_1' \wedge X_2' \neq X_1) : \\ & \mathbb{D}(X_1X_1', X_2X_2') = \left\{ \hat{E}_{rs} |K\rangle \middle| |K\rangle \in \mathbb{D}(X_1X_1') \text{ and } \hat{E}_{rs} \in \mathbb{E}_{X_2X_2'} \right\} \end{aligned} \tag{37}$$

And the total singly($\mathbb{D}_S$), doubly ($\mathbb{D}_D$), ..., excited categorical spaces are:

$$\mathbb{D}_S = \bigcup_{\text{valid } X_1, X_1'} \mathbb{D}(X_1 X_1') \tag{38}$$

$$\mathbb{D}_D = \bigcup_{\text{valid } X_1, X_1', X_2, X_2'} \mathbb{D}(X_1 X_1', X_2 X_2') \tag{39}$$

In order to organize the total space, a three-level addressing scheme is proposed based on the Kozlowski and Pulay's RAS addressing scheme. In first level, the total space is divided into segments that associated with each category $\mathbb{D}_Y$, then the global address of determinant $|K\rangle$ is defined as:

$$A(K) = A^{\mathbb{D}}(K) + \sum_{Y'=0}^{Y-1} N_{Y'} \tag{40}$$

where the length of the category is computed as the products of the length of each GAS in it:

$$N_Y = \prod_X N_Y^X \tag{41}$$

$$N_Y^X = \binom{m_Y^X}{n_Y^X} \tag{42}$$

In the second level, the address of determinant $|K\rangle$ is resolved in its category:

$$A^{\mathbb{D}}(K) = 1 + \sum_X \left[\left(A^{\mathbb{G}}(K_Y^X) - 1 \right) \times \prod_{X'=1}^{X-1} N_Y^{X'} \right] \tag{43}$$

In the third level, each individual GAS string segments $\left|K_Y^X\right\rangle$ is addressed using Knowles and Handy's[?] addressing methods:

$$A^{\mathbb{G}}(K_Y^X) = 1 + \sum_{n_i=1}^{n_Y^X} Z(n_i, m_j) \tag{44}$$

where, n_i and m_j are the i^{th} electron and the j^{th} orbital in $\left|K_Y^X\right\rangle$ and $Z(n_i, m_j)$ are matrix

elements of the addressing matrix defined as:

$$Z(n_i, m_j) = \begin{cases} \sum_{k=m^X-m_j+1}^{m^X-n_i} \left[\binom{k}{n_Y^X-n_i} - \binom{k-1}{n_Y^X-n_i-1} \right], & \text{if } m_Y^X - n_Y^X + n_i \geq m_j \geq n_i \\ & \text{and } n_i < m^X \\ m_j - n_Y^X, & \text{if } m_X \geq m_j \geq n_Y^X \\ 0, & \text{otherwise} \end{cases} \tag{45}$$

At this point, we can decompose the determinant $|K\rangle$ as its individual components in each GAS, and denoted as:

$$|K\rangle = \left|K_Y^M ... K_Y^X ... K_Y^1\right\rangle = \left|K_Y^{M...X...1}\right\rangle \tag{46}$$

Then, all non-zeros one-electron symbolic matrix element are now separated into two cases: 1) intra-XY excitation, or 2) inter-XY excitation. They are computed as:

$$\forall \begin{pmatrix} |K\rangle \in \mathbb{D}_Y, |L\rangle \in \mathbb{D}_{Y'} \\ p \in \mathbb{G}_X, q \in \mathbb{G}_{X'} \end{pmatrix} \wedge \begin{pmatrix} \forall X'' \neq X \wedge X'' \neq X', \\ \left|K_Y^{X''}\right\rangle = \left|L_Y^{X''}\right\rangle \end{pmatrix} :$$

$$A_{KL}^{pq} = \begin{cases} A_{KL}^{pq}(Y, X) = \left\langle K_Y^X \right| \hat{E}_{pq} \left| L_Y^X \right\rangle, & \text{if } Y = Y' \wedge X = X' \\ A_{KL}^{pq}(Y, X; Y', X') = \left\langle K_Y^{XX'} \right| \hat{E}_{pq} \left| L_{Y'}^{XX'} \right\rangle \times \text{sgn}_Y^{\{XX'\}}, & \text{if } Y \neq Y' \wedge X \neq X' \end{cases} \tag{47}$$

where in inter-XY excitation, the contribution between GAS X and GAS X' are denoted as a offset sign change:

$$\forall (X > X') : \text{sgn}_Y^{\{X'X\}} = sgn_Y^{\{XX'\}} = \begin{cases} +1, \text{ if } \sum_{X''=X'+1}^{X-1} n_Y^{X''} \text{ is even} \\ -1, \text{ if } \sum_{X''=X'+1}^{X-1} n_Y^{X''} \text{ is odd} \end{cases} \tag{48}$$

Then the Eqs. (12) and (13) could be re-written as:

$$\forall(|K\rangle \in \mathbb{D}_Y \wedge |L\rangle \in \mathbb{D}_{Y'} \wedge |J\rangle \in \mathbb{D}_{Y''}) :$$

$$^{1e}\sigma_K = \sum_X \sum_{pq\in\mathbb{G}_X} \sum_{L_Y^X} h'_{pq} A^{pq}_{KL}(Y, X) C_L + \sum_X \sum_{\substack{Y'\neq Y \\ X'\neq X}} \sum_{\substack{p\in\mathbb{G}_X \\ q\in\mathbb{G}_{X'}}} \sum_{L_{Y'}^{XX'}} h'_{pq} A^{pq}_{KL}(Y, X; Y', X') C_L \tag{49}$$

$$\begin{aligned} ^{2e}\sigma_K =& \frac{1}{2} \sum_{X_1} \sum_{pq\in\mathbb{G}_{X_1}} \sum_{J_Y^{X_1}} A^{pq}_{KJ}(Y, X_1) \sum_{X_2} \Bigg[\sum_{rs\in\mathbb{G}_{X_2}} \sum_{L_Y^{X_2}} (pq|rs) A^{pq}_{JL}(Y, X_2) C_L \\ &+ \sum_{\substack{Y''\neq Y \\ X_2'\neq X_2}} \sum_{\substack{r\in\mathbb{G}_{X_2} \\ s\in\mathbb{G}_{X_2'}}} \sum_{L_{Y'}^{X_2X_2'}} (pq|rs) A^{pq}_{JL}(Y, X_2; Y', X_2') C_L \Bigg] \\ &+ \frac{1}{2} \sum_{X_1} \sum_{\substack{Y''\neq Y \\ X_1'\neq X_1}} \sum_{\substack{p\in\mathbb{G}_{X_1} \\ q\in\mathbb{G}_{X_1'}}} \sum_{J_{Y''}^{X_1X_1'}} A^{pq}_{KJ}(Y, X_1; Y'', X_1') \sum_{X_2} \Bigg[\sum_{rs\in\mathbb{G}_{X_2}} \sum_{L_{Y''}^{X_2}} (pq|rs) A^{pq}_{JL}(Y'', X_2) C_L \\ &+ \sum_{\substack{Y''\neq Y' \\ X_2'\neq X_2}} \sum_{\substack{r\in\mathbb{G}_{X_2} \\ s\in\mathbb{G}_{X_2'}}} \sum_{L_{Y'}^{X_2X_2'}} (pq|rs) A^{pq}_{JL}(Y'', X_2; Y', X_2') C_L \Bigg] \end{aligned} \tag{50}$$

3.3 Matrix/Tensor Formalism of GASCI

An important observation is that each categorical segments of total space can be view as the Cartesian products of its sub-GAS strings:

$$\mathbb{D}_Y = \mathbb{D}_Y^1 \times ... \times \mathbb{D}_Y^M = \mathbb{D}_Y^{1\times...\times M} \tag{51}$$

In another word, the vector $\boldsymbol{\sigma}_Y$ and $\boldsymbol{C}_Y$ could be viewed as rank-M tensors. Here we consider they are complex-value vectors, then:

$$\underline{\boldsymbol{\sigma}}_Y, \underline{\boldsymbol{C}}_Y \in \mathbb{C}^{N_Y^1\times...\times N_Y^M} \tag{52}$$

Now, the intra- and inter-XY one-electron symbolic matrix are now defined as matrices and rank-4 tensors, respectively:

$$\boldsymbol{A}^{pq}_{\{Y,X\}} \in \mathbb{C}^{N_Y^X \times N_Y^X} : \quad A^{pq}_{KL}(Y,X) = \left\langle K_Y^X \right| \hat{E}_{pq} \left| L_Y^X \right\rangle \tag{53}$$

$$\underline{\boldsymbol{A}}^{pq}_{\{Y,X;Y',X'\}} \in \mathbb{C}^{N_Y^X \times N_Y^{X'} \times N_{Y'}^X \times N_{Y'}^{X'}} :$$

$$A^{pq}_{KL}(Y,X;Y',X') = \left\langle K_Y^{XX'} \right| \hat{E}_{pq} \left| L_{Y'}^{XX'} \right\rangle \times \mathrm{sgn}_Y^{\{XX'\}} \tag{54}$$

Then, the intermediate matrices and tensors are defined accordingly as:

$$\boldsymbol{\Omega}_{\{Y,X\}} = \sum_{pq \in \mathbb{G}_X} h'_{pq} \boldsymbol{A}^{pq}_{\{Y,X\}} \tag{55}$$

$$\underline{\boldsymbol{\Omega}}_{\{Y,X;Y',X'\}} = \sum_{p \in \mathbb{G}_X, q \in \mathbb{G}_{X'}} h'_{pq} \underline{\boldsymbol{A}}^{pq}_{\{Y,X;Y',X'\}} \tag{56}$$

$$\boldsymbol{\Lambda}^{pq}_{\{Y,X\}} = \sum_{rs \in \mathbb{G}_X} (pq|rs) \boldsymbol{A}^{pq}_{\{Y,X\}} \tag{57}$$

$$\underline{\boldsymbol{\Lambda}}^{pq}_{\{Y,X;Y',X'\}} = \sum_{r \in \mathbb{G}_X, s \in \mathbb{G}_{X'}} (pq|rs) \underline{\boldsymbol{A}}^{rs}_{\{Y,X;Y',X'\}} \tag{58}$$

The dimension of those matrices are $\mathbb{C}^{N_Y^X \times N_Y^X}$, and of those tensors are $\mathbb{C}^{N_Y^X \times N_Y^{X'} \times N_{Y'}^X \times N_{Y'}^{X'}}$, which could be arbitrarily small under appropriate GAS partitions and would not cause any problems in terms of storage.

To simplify the discussion, notations related to tensor operations are introduced. The mode-(a,b) tensor contraction between two tensors, $\underline{\boldsymbol{Z}}_i \in \mathbb{C}^{I_1 \times \ldots \times I_A}$ and $\underline{\boldsymbol{Z}}_j \in \mathbb{C}^{J_1 \times \ldots \times J_B}$ is denoted as:

$$\forall I_a = J_b, \quad \underline{\boldsymbol{Z}}_k = \underline{\boldsymbol{Z}}_i \times_a^b \underline{\boldsymbol{Z}}_j :$$

$$Z_k(i_1, .., i_{a-1}, i_{a+1}, ...i_A, j_1, ..., j_{b-1}, j_{b+1}..., j_B) =$$

$$\sum_{i_a} Z_i(i_1, .., i_{a-1}, i_a, i_{a+1}, ...i_A) \times Z_j(j_1, .., j_{b-1}, i_a, j_{b+1}, ...j_B) \tag{59}$$

If $a = A$ or $b = 1$, those index could be omitted, yielding the simplified notations:

$$\underline{\boldsymbol{Z}}_i \times_A^1 \underline{\boldsymbol{Z}}_j = \underline{\boldsymbol{Z}}_i \times^1 \underline{\boldsymbol{Z}}_j = \underline{\boldsymbol{Z}}_i \times_N \underline{\boldsymbol{Z}}_j = \underline{\boldsymbol{Z}}_i \cdot \underline{\boldsymbol{Z}}_j \tag{60}$$

Tensor contractions over multiple dimensions are denoted as $\times_{a,c,...}^{b,d,...}$.

Then, a tensor transpose is defined as permutations among its indices of different dimensions:

$$\begin{aligned} \forall \quad \boldsymbol{\pi} = (\pi_1, ..., \pi_A) \quad \wedge \quad \{\pi_1, ..., \pi_A\} = \{i_1, ..., i_A\} : \\ [\underline{\boldsymbol{Z}}_i(i_1, ..., i_A)]^{T_{\boldsymbol{\pi}}} = \underline{\boldsymbol{Z}}_i(\pi_1, ..., \pi_A) \end{aligned} \tag{61}$$

Two simplified tensor transpose are defined as:

$$T_a = T_{\boldsymbol{\pi}_a}, \text{ where } \boldsymbol{\pi}_a = (a, 1, 2, ..., a-1, a+1, ..., A) \tag{62}$$

$$T_a^\dagger = T_{\boldsymbol{\pi}'_a}, \text{ where } \boldsymbol{\pi}'_a = (2, ..., a-1, a, 1, a+1, ..., A) \tag{63}$$

It is obviously that:

$$\left[\underline{\boldsymbol{Z}}^{T_a}\right]^{T_a^\dagger} = \underline{\boldsymbol{Z}} \tag{64}$$

At this point the tensor formalism of $\underline{\boldsymbol{\sigma}}_Y$ can be expressed as:

$$\begin{aligned} &\forall \quad X >= X' : \\ {}^{1e}\underline{\boldsymbol{\sigma}}_Y = &\sum_X \left[\boldsymbol{\Omega}_{\{Y,X\}} \times^X \underline{\boldsymbol{C}}_Y\right]^{T_X^\dagger} + \sum_{\substack{Y' \neq Y \\ X' \neq X}} \left[\sum_X \left(\boldsymbol{\Omega}_{\{Y,X;Y',X'\}} \times^{X,X'} \underline{\boldsymbol{C}}_{Y'}\right)^{T_X^\dagger}\right]^{T_{X'}^\dagger} \end{aligned} \tag{65}$$

$\forall \quad X_1 >= X_1' \wedge X_2 >= X_2' :$

$$ {}^{2e}\underline{\boldsymbol{\sigma}}^{pq}_{Y''} = \sum_{X_2} \left[\boldsymbol{\Lambda}^{pq}_{\{Y'',X_2\}} \times^{X_2} \underline{\boldsymbol{C}}_{Y''} \right]^{T^{\dagger}_{X_2}} + \sum_{\substack{Y'\neq Y'' \\ X_2'\neq X_2}} \left[\sum_{X_2} \left(\boldsymbol{\Lambda}^{pq}_{\{Y'',X_2;Y',X_2'\}} \times^{X_2,X_2'} \underline{\boldsymbol{C}}_{Y'} \right)^{T^{\dagger}_{X_2}} \right]^{T^{\dagger}_{X_2'}} \tag{66} $$

$$ \begin{aligned} {}^{2e}\underline{\boldsymbol{\sigma}}_{Y} =& \frac{1}{2} \sum_{X_1} \left[\sum_{pq\in\mathbb{G}_{X_1}} \boldsymbol{A}^{pq}_{\{Y,X_1\}} \times^{X_1} {}^{2e}\underline{\boldsymbol{\sigma}}^{pq}_{Y} \right]^{T^{\dagger}_{X_1}} \\ &+ \frac{1}{2} \sum_{\substack{Y''\neq Y \\ X_1'\neq X_1}} \left[\sum_{X_1} \left(\sum_{\substack{p\in\mathbb{G}_{X_1} \\ q\in\mathbb{G}_{X_1'}}} \boldsymbol{A}^{pq}_{\{Y,X_1;Y'',X_1'\}} \times^{X_1,X_1'} {}^{2e}\underline{\boldsymbol{\sigma}}^{pq}_{Y''} \right)^{T^{\dagger}_{X_1}} \right]^{T^{\dagger}_{X_1'}} \end{aligned} \tag{67} $$

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