GF2 and GW with decomposed interactions

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I. DECOMPOSED INTERACTION

The electronic Hamiltonian in a molecule with N orbitals takes the form

$$H = \sum_{ij}\sum_{\sigma} h_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \frac{1}{2}\sum_{ijkl} U_{ijkl} \sum_{\sigma\sigma'} c^{\dagger}_{i\sigma} c^{\dagger}_{k\sigma'} c_{l\sigma'} c_{j\sigma}. \tag{1}$$

The interaction tensor $U_{ijkl} = [ij|kl]$ usually has the following symmetry:

$$[ij|kl] = [ji|kl] \tag{2}$$
$$[ij|kl] = [ij|lk] \tag{3}$$
$$[ij|kl] = [kl|ij]. \tag{4}$$

Making use of one of the symmetry (4), we can view U_{ijkl} as a symmetric matrix with dimension $N^2 \times N^2$. One can perform a eigenvalue decomposition such that

$$U_{ijkl} = \sum_{\alpha=1}^{N^2} d^{\alpha} V^{\alpha}_{ij} V^{\alpha}_{kl}, \tag{5}$$

where $\{d^{\alpha}\}$ are eigenvalues and $\{V^{\alpha}\}$ are "eigenvectors". The decomposition step scales as $\mathcal{O}(N^6)$. The decomposed interaction tensor V^{α}_{ij} usually retains the symmetry in the remaining two orbital indices, i.e. $V^{\alpha}_{ij} = V^{\alpha}_{ji}$. Note that one can alternatively perform a Cholesky decomposition (U is usually positive (semi-)definite) or directly use the density fitting integrals. The eigenvalues $\{d^{\alpha}\}$, when ordered in descending order, usually decay very rapidly, and we can truncate the values at certain precision tolerance and throw away small eigenvalues. After the truncation, the number of eigenvalues kept N_{α} is usually much smaller than N^2 and scales as $\mathcal{O}(N)$.

II. HARTREE-FOCK

The Hartree-Fock contribution with the full interaction tensor is

$$\Sigma^{\mathrm{HF}}_{ij,\sigma} = \sum_{kl} U_{ijkl} \sum_{\sigma'} \rho_{kl,\sigma'} - \sum_{kl} U_{ilkj} \rho_{kl,\sigma}, \tag{6}$$

which is a $\mathcal{O}(N^4)$ procedure. With the decomposed version, it becomes

$$\Sigma^{\mathrm{HF}}_{ij,\sigma} = \sum_{kl}\sum_{\alpha} d^{\alpha} V^{\alpha}_{ij} V^{\alpha}_{kl} \sum_{\sigma'} \rho_{kl,\sigma'} - \sum_{kl}\sum_{\alpha} d^{\alpha} V^{\alpha}_{il} V^{\alpha}_{kj} \rho_{kl,\sigma}. \tag{7}$$

The Hartree term can be constructed as

$$X^{\alpha} := \sum_{kl} d^{\alpha} V^{\alpha}_{kl} \sum_{\sigma'} \rho_{kl,\sigma'}, \quad \mathcal{O}(N^2 N_{\alpha}) \tag{8a}$$
$$\Sigma^{\mathrm{Hartree}}_{ij,\sigma} = \sum_{\alpha} V^{\alpha}_{ij} X^{\alpha}. \quad \mathcal{O}(N^2 N_{\alpha}) \tag{8b}$$

For the Fock term, we can do

$$X^{\alpha}_{lj,\sigma} := \sum_{k} d^{\alpha} V^{\alpha}_{kj} \rho_{kl,\sigma}, \quad \mathcal{O}(N^3 N_{\alpha}) \tag{9a}$$
$$\Sigma^{\mathrm{Fock}}_{ij,\sigma} = \sum_{l}\sum_{\alpha} V^{\alpha}_{il} X^{\alpha}_{lj,\sigma}. \quad \mathcal{O}(N^3 N_{\alpha}) \tag{9b}$$

Overall this would be faster than reassembling the full tensor and use Eq. (6), because the reassemble step is $\mathcal{O}(N^4 N_{\alpha})$.

III. GF2 WITHOUT DECOMPOSED INTERACTION

The GF2 self-energy has two terms. The "bubble" term is

$$\Sigma^{\mathrm{bubble}}_{ij,\sigma}(\tau) = -\sum_{lnp}\sum_{kqm} U_{ilnp} U_{kjqm} \times G_{lk,\sigma}(\tau) \sum_{\sigma'} G_{mn,\sigma'}(-\tau) G_{pq,\sigma'}(\tau), \tag{10}$$

and the "exchange" term is

$$\Sigma^{\mathrm{exchange}}_{ij,\sigma}(\tau) = \sum_{lnp}\sum_{qkm} U_{ilnp} U_{qjkm} \times G_{lk,\sigma}(\tau) G_{mn,\sigma}(-\tau) G_{pq,\sigma}(\tau). \tag{11}$$

The contraction can be performed as follows. For the bubble term,

$$X_{ilnq,\sigma\sigma'}(\tau) \leftarrow \sum_{p} U_{ilnp,\sigma} G_{pq,\sigma'}(\tau), \quad \mathcal{O}(N^5) \tag{12a}$$
$$X_{ilmq,\sigma}(\tau) \leftarrow \sum_{n}\sum_{\sigma'} X_{ilnq,\sigma\sigma'}(\tau) G_{mn,\sigma'}(-\tau), \quad \mathcal{O}(N^5) \tag{12b}$$
$$X_{ikmq,\sigma}(\tau) \leftarrow \sum_{l} X_{ilmq,\sigma}(\tau) G_{lk,\sigma}(\tau), \quad \mathcal{O}(N^5) \tag{12c}$$
$$\Sigma^{\mathrm{bubble}}_{ij,\sigma}(\tau) = -\sum_{kmq} X_{ikmq,\sigma}(\tau) U_{kjqm}. \quad \mathcal{O}(N^5) \tag{12d}$$

Naively, the memory scaling of this procedure is $\mathcal{O}(N^4)$. One can improve this to $\mathcal{O}(N^3)$ by doing an outer loop of the external index i.

Similarly for the exchange term,

$$X_{ilnq,\sigma}(\tau) \leftarrow \sum_q U_{ilnp,\sigma} G_{pq,\sigma}(\tau), \quad \mathcal{O}(N^5) \tag{13a}$$

$$X_{ilmq,\sigma}(\tau) \leftarrow \sum_n X_{ilnq,\sigma}(\tau) G_{mn,\sigma}(-\tau), \quad \mathcal{O}(N^5) \tag{13b}$$

$$X_{ikmq,\sigma}(\tau) \leftarrow \sum_l X_{ilmq,\sigma}(\tau) G_{lk,\sigma}(\tau), \quad \mathcal{O}(N^5) \tag{13c}$$

$$\Sigma^{\text{bubble}}_{ij,\sigma}(\tau) = \sum_{kmq} X_{ikmq,\sigma}(\tau) U_{qjkm}. \quad \mathcal{O}(N^5) \tag{13d}$$

The first tree steps are almost the same as in the bubble term except for the absence of spin summation. In the spin restricted (paramagnetic) case, the two terms can be combined into the same procedure.

IV. GF2 WITH DECOMPOSED INTERACTION

With the decomposed interaction, the "bubble" terms becomes

$$\Sigma^{\text{bubble}}_{ij,\sigma}(\tau) = \sum_{lnp}\sum_{kqm}\sum_{\alpha\beta} d^\alpha V^\alpha_{il} V^\alpha_{np} d^\beta V^\beta_{kj} V^\beta_{qm} \times G_{lk,\sigma}(\tau) \sum_{\sigma'} G_{mn,\sigma'}(-\tau) G_{pq,\sigma'}(\tau). \tag{14}$$

We can build this up by doing the following contractions (scaling for each τ point.)

$$X^\beta_{qn,\sigma}(\tau) := \sum_m V^\beta_{qm} G_{mn,\sigma}(-\tau), \qquad \mathcal{O}(N^3 N_\alpha) \tag{15a}$$

$$Y^\alpha_{nq,\sigma}(\tau) := \sum_l V^\alpha_{np} G_{pq,\sigma}(\tau), \qquad \mathcal{O}(N^3 N_\alpha) \tag{15b}$$

$$\Pi^{\alpha\beta}(\tau) := \sum_{nq}\sum_\sigma d^\beta d^\alpha X^\beta_{qn,\sigma}(\tau) Y^\alpha_{nq,\sigma}(\tau), \qquad \mathcal{O}(N^2 N_\alpha^2) \tag{15c}$$

$$Z^\beta_{kj}(\tau) := \sum_\alpha \Pi^{\alpha\beta}(\tau) V^\beta_{kj}, \qquad \mathcal{O}(N^2 N_\alpha^2) \tag{15d}$$

$$\Sigma^{\text{bubble}}_{ij,\sigma}(\tau) = \sum_k \sum_\beta Z^\beta_{kj}(\tau) Y^\beta_{ik}(\tau). \qquad \mathcal{O}(N^3 N_\alpha) \tag{15e}$$

The "exchange" term is

$$\Sigma^{\text{exchange}}_{ij,\sigma}(\tau) = \sum_{lnp}\sum_{qkm}\sum_{\alpha\beta} d^\alpha V^\alpha_{il} V^\alpha_{np} d^\beta V^\beta_{qj} V^\beta_{km} \times G_{lk,\sigma}(\tau) G_{mn,\sigma}(-\tau) G_{pq,\sigma}(\tau). \tag{16}$$

And the construction steps are the following.

$$X^\beta_{kn,\sigma}(\tau) := \sum_m d^\beta V^\beta_{km} G_{mn,\sigma}(-\tau), \qquad \mathcal{O}(N^3 N_\alpha) \tag{17a}$$

$$\tilde{X}^\beta_{ln,\sigma}(\tau) := \sum_k X^\beta_{kn,\sigma}(\tau) G_{lk,\sigma}(\tau), \qquad \mathcal{O}(N^3 N_\alpha) \tag{17b}$$

$$Z_{qjln,\sigma}(\tau) := \sum_\beta \tilde{X}^\beta_{ln,\sigma} V^\beta_{qj}, \qquad \mathcal{O}(N^4 N_\alpha) \tag{17c}$$

$$Y^\alpha_{nq,\sigma}(\tau) := \sum_l d^\alpha V^\alpha_{np} G_{pq,\sigma}(\tau), \qquad \mathcal{O}(N^3 N_\alpha) \tag{17d}$$

$$\tilde{Y}^\alpha_{jl,\sigma}(\tau) := \sum_{nq} Y^\alpha_{nq,\sigma}(\tau) Z_{qjln,\sigma}(\tau), \qquad \mathcal{O}(N^4 N_\alpha) \tag{17e}$$

$$\Sigma^{\text{exchange}}_{ij,\sigma}(\tau) = \sum_l \sum_\alpha \tilde{Y}^\alpha_{jl,\sigma}(\tau) V^\alpha_{il}. \qquad \mathcal{O}(N^3 N_\alpha) \tag{17f}$$

We don't really benefit much from the decomposition here, since the overall scaling $\mathcal{O}(N^4 N_\alpha)$ is usually not as good as the $\mathcal{O}(N^5)$ scaling without the decomposition.

V. *GW* WITH DECOMPOSED INTERACTION

GW is not practical without the decomposed interaction, as the scaling would be $\mathcal{O}(N^6)$ for each frequency in each iteration. The *GW* self-energy takes the form

$$\Sigma^{GW}_{ij,\sigma}(\tau) = -\sum_{kl} G_{lk,\sigma}(\tau) W_{ilkj}(\tau), \tag{18}$$

in which the screened interaction derived from a Bethe-Salpeter equation

$$W_{ijkl}(\mathrm{i}\omega_n) = U_{ijkl} + \sum_{pqmn} W_{ijpq}(\mathrm{i}\omega_n) \Pi_{qpnm}(\mathrm{i}\omega_n) U_{mnkl}, \tag{19}$$

and the "polarization" object is the bare "bubble"

$$\Pi_{qpnm}(\tau) = \sum_\sigma G_{qm,\sigma}(\tau) G_{np,\sigma}(-\tau). \tag{20}$$

The screened interaction contains a frequency-independent contribution from the bare interaction U, which actually gives the Fock diagram. The actual "beyond-HF" contribution comes from the frequency-dependent part

$$\tilde{W}_{ijkl}(\mathrm{i}\omega_n) = W_{ijkl}(\mathrm{i}\omega_n) - U_{ijkl}, \tag{21}$$

and Eq. (19) becomes

$$\tilde{W}_{ijkl}(\mathrm{i}\omega_n) = \sum_{pqmn} (\tilde{W}_{ijpq}(\mathrm{i}\omega_n) + U_{ijpq}) \Pi_{qpnm}(\mathrm{i}\omega_n) U_{mnkl}. \tag{22}$$

With the decomposed interaction, the equation for the screened interaction becomes

$$\tilde{W}_{ijkl}(\mathrm{i}\omega_n) = \sum_{pqmn} (\tilde{W}_{ijpq}(\mathrm{i}\omega_n) + \sum_{\alpha} d^{\alpha} V^{\alpha}_{ij} V^{\alpha}_{pq}) \times \Pi_{qpnm}(\mathrm{i}\omega_n) \sum_{\beta} d^{\beta} V^{\beta}_{mn} V^{\beta}_{kl}. \tag{23}$$

If we define the quantities $\tilde{\Pi}$ and X such that

$$\tilde{\Pi}^{\alpha\beta} = \sum_{pqnm} V^{\alpha}_{pq} \Pi_{qpnm} V^{\beta}_{mn}, \tag{24}$$

$$\tilde{W}_{ijkl} = \sum_{\alpha\beta} V^{\alpha}_{ij} F^{\alpha\beta} V^{\beta}_{kl}, \tag{25}$$

we can rewrite (23) as

$$\begin{aligned} F^{\alpha\beta}(\mathrm{i}\omega_n) &= \sum_{\alpha'} (F^{\alpha\alpha'}(\mathrm{i}\omega_n) + d^{\alpha}\delta^{\alpha\alpha'}) \tilde{\Pi}^{\alpha'\beta}(\mathrm{i}\omega_n) d^{\beta} \\ &= \sum_{\alpha'} F^{\alpha\alpha'}(\mathrm{i}\omega_n) \tilde{\Pi}^{\alpha'\beta}(\mathrm{i}\omega_n) d^{\beta} + d^{\alpha} \tilde{\Pi}^{\alpha\beta}(\mathrm{i}\omega_n) d^{\beta}. \end{aligned} \tag{26}$$

In matrix form, this can be written as

$$F(\mathrm{i}\omega_n) = F(\mathrm{i}\omega_n)\tilde{\Pi}(\mathrm{i}\omega_n) D + D\tilde{\Pi}(\mathrm{i}\omega_n) D, \tag{27}$$

where D is the diagonal matrix with elements d^{α}. F can now be solved from a $N_\alpha \times N_\alpha$ linear equation:

$$F(\mathrm{i}\omega_n)(\mathbf{1} - \tilde{\Pi}(\mathrm{i}\omega_n) D) = D\tilde{\Pi}(\mathrm{i}\omega_n) D, \tag{28}$$

which scales as $\mathcal{O}(N_\alpha^3)$. The construction of $\tilde{\Pi}^{\alpha\beta}(\tau) = \sum_{pqnm} V^{\alpha}_{pq} \sum_{\sigma} G_{qm,\sigma}(\tau) G_{np,\sigma}(-\tau) V^{\beta}_{mn}$ can be performed as follows:

$$X^{\alpha}_{pm,\sigma}(\tau) \leftarrow \sum_{q} V^{\alpha}_{pq} G_{qm,\sigma}(\tau), \quad \mathcal{O}(N^3 N_\alpha) \tag{29a}$$

$$X^{\alpha}_{nm,\sigma}(\tau) \leftarrow \sum_{p} X^{\alpha}_{pm,\sigma}(\tau) G_{np,\sigma}(-\tau), \quad \mathcal{O}(N^3 N_\alpha) \tag{29b}$$

$$\tilde{\Pi}^{\alpha\beta}(\tau) = \sum_{\sigma}\sum_{mn} X^{\alpha}_{nm,\sigma}(\tau) V^{\beta}_{mn}. \quad \mathcal{O}(N^2 N_\alpha^2) \tag{29c}$$

Finally, the GW contribution to the self-energy takes the form

$$\begin{aligned} \tilde{\Sigma}^{GW}_{ij}(\tau) &= -\sum_{kl} G_{lk,\sigma}(\tau) \tilde{W}_{ilkj}(\tau) \\ &= -\sum_{kl} G_{lk,\sigma}(\tau) \sum_{\alpha\beta} V^{\alpha}_{il} F^{\alpha\beta}(\tau) V^{\beta}_{kj}, \end{aligned} \tag{30}$$

which can be constructed as

$$X^{\alpha}_{ik,\sigma}(\tau) \leftarrow \sum_{l} G_{lk,\sigma}(\tau) V^{\alpha}_{il}, \quad \mathcal{O}(N^3 N_\alpha) \tag{31a}$$

$$X^{\beta}_{ik,\sigma}(\tau) \leftarrow \sum_{\alpha} X^{\alpha}_{ik,\sigma} F^{\alpha\beta}(\tau), \quad \mathcal{O}(N^2 N_\alpha^2) \tag{31b}$$

$$\tilde{\Sigma}^{GW}_{ij}(\tau) = -\sum_{k}\sum_{\beta} X^{\beta}_{ik,\sigma} V^{\beta}_{kj}. \quad \mathcal{O}(N^3 N_\alpha) \tag{31c}$$

The total complexity is dominated by $\mathcal{O}(N_\alpha^3 N_\omega)$ from Eq. (28) or $\mathcal{O}(N^2 N_\alpha^2 N_\tau)$ form Eq. (29) or (31).