Supporting Information for "Dielectric matrix formulation of correlation energies in the Random Phase Approximation (RPA): inclusion of exchange effects"

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Starting from the general expression for the ground state correlation energy in the adiabatic connection fluctuation dissipation theorem (ACFDT) framework, it is shown that the dielectric matrix formulation, which is usually applied to calculate the direct random phase approximation (dRPA) correlation energy, can be used for alternative RPA expressions including exchange effects. Within this famework, the ACFDT analog of the second order screened exchange (SOSEX) approximation leads to a logarithmic formula for the correlation energy similar to the direct RPA expression. Alternatively, the contribution of the exchange can be included in the kernel used to evaluate the response functions. In this case the use of an approximate kernel is crucial to simplify the formalism and to obtain a correlation energy in logarithmic form. Technical details of the implementation of these methods are discussed and it is shown that one can take advantage of density fitting or Cholesky decomposition techniques to improve the computational efficiency; a discussion on the numerical quadrature made on the frequency variable is also provided. A series of test calculations on atomic correlation energies and molecular reaction energies shows that exchange effects are instrumental to improve over direct RPA results.
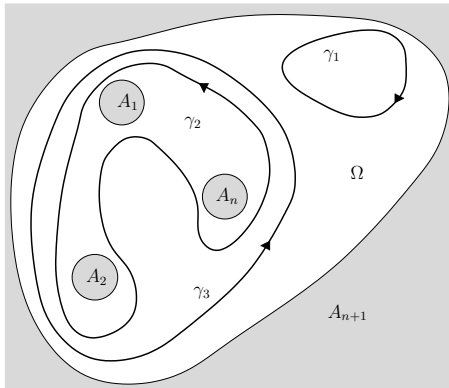
FIG. 1. Ω is a region with holes $A_1, A_2, \ldots, A_n, A_{n+1}$ (A_{n+1} is the "exterior" of Ω). We have : $\gamma_1 \sim 0[\Omega]$, *i.e.* the closed curve does not wind around any point that is outside of Ω. And : $\gamma_2 \sim \gamma_3[\Omega]$, *i.e.* the two closed curves wind around all the points outside of Ω in the same manner.

I. BACKGROUND OF COMPLEX INTEGRATION

Using the method followed by McLachlan *et al.* [1] the frequency integral in Eq. (56) of the main paper can be solved via an integration by parts of the logarithm and by invoking the *argument principle*, leading to the result of Eq. (57) of the main paper after contour integration. To explain these steps, a review of some complex integration notions is presented. A comprehensive account on the subject can be found in Ref. 2.

An important notion to consider when dealing with complex integration is the *index of a point a with respect to a closed curve γ*, denoted $n(\gamma, a)$. In layman's terms, it indicates how many times a closed curve winds around a point not on the curve, and is defined as:

$$n(\gamma, a) = \frac{1}{2\pi i} \int_\gamma \frac{dz}{z - a}. \tag{1}$$

Having defined the notion of index, a property of the closed curves emerges : the *homology*. In simple words, considering a region Ω with holes $A_1, A_2, \ldots, A_n, A_{n+1}$, such as in Fig. (1), we say that closed curves γ_i and γ_j are homologous with respect to Ω when they wind around every point outside of Ω the same number of times (this is written $\gamma_i \sim \gamma_j[\Omega]$). In Fig. (1), the closed curved γ_2 and γ_3 are homologous with respect to Ω.

For any region Ω like seen in Fig. (1), we are able to construct an homology basis as a set of closed curves $\{\gamma_i\}$ around

the holes A_i of Ω, such that for any closed curve γ:

$$\gamma \sim (c_1\gamma_1 + c_2\gamma_2 + \cdots + c_n\gamma_n)[\Omega], \qquad (2)$$

where the c_i are the (constant) indexes $n(\gamma, a_i)$ of the points a_i in a hole A_i.

In particular, considering a function f analytic in the whole region except for isolated poles $a_1, a_2, \ldots, a_n$ that lay inside the holes $A_1, A_2, \ldots, A_n$ (f is said to be meromorphic), Cauchy's theorem yields that:

$$\int_\gamma f dz = c_1 \int_{\gamma_1} f dz + c_2 \int_{\gamma_2} f dz + \cdots + c_n \int_{\gamma_n} f dz$$
$$= 2\pi i(c_1 R_1 + c_2 R_2 + \cdots + c_n R_n), \qquad (3)$$

which defines the R_i, the *residues of f at point a_i*. Equation (3) is called the residue theorem.

An application of the residue theorem, called the *argument principle* is of particular interest for us. For any meromorphic function f with zeros a_i of order h_i and poles b_j of order h_j, the function f'/f has simple poles a_i and b_j of residues h_i and $-h_j$. Application of the residue theorem to f'/f gives:

$$\frac{1}{2\pi i} \int_\gamma \frac{f'(z)}{f(z)} dz = \sum_i n(\gamma, a_i)h_i - \sum_j n(\gamma, b_j)h_j. \qquad (4)$$

This is the argument principle, which can be generalized to a function $g \times (f'/f)$:

$$\frac{1}{2\pi i} \int_\gamma g(z)\frac{f'(z)}{f(z)} dz = \sum_i n(\gamma, a_i)g(a_i)h_i$$
$$- \sum_j n(\gamma, b_j)g(b_j)h_j. \qquad (5)$$

Note that one will often find the argument principle written with a convention where the summation over the zeros and poles are repeated as many times as their order indicates, which allows a more compact expression:

$$\frac{1}{2\pi i} \int_\gamma g(z)\frac{f'(z)}{f(z)} dz = \sum_{\tilde{i}} n(\gamma, a_{\tilde{i}})g(a_{\tilde{i}})$$
$$- \sum_{\tilde{j}} n(\gamma, b_{\tilde{j}})g(b_{\tilde{j}}). \qquad (6)$$

Having this in mind, let us introduce a change of variable $\omega = i\xi$ in Eq. (56) of the main paper :

$$E_c^{\text{dRPA-I}} = \frac{1}{2} \int_{-\infty}^{\infty} \frac{-d\xi}{2\pi i} \sum_{ia} \left\{ \log\left(1 + \frac{\Omega_{ia}^2 - \epsilon_{ia}^2}{\epsilon_{ia}^2 - \xi^2}\right) - \frac{M_{1,ia,ia} - \epsilon_{ia}^2}{\epsilon_{ia}^2 - \xi^2}\right\}. \qquad (7)$$

The second term integrates to $-K_{ia,ia}$, and the logarithm can be integrated by parts:

$$-\frac{1}{2\pi i} \int \log(f(z)) dz = -\frac{1}{2\pi i} \left[\log(f(z)) z\right] + \frac{1}{2\pi i} \int \frac{f'(z)}{f(z)} z$$
$$= \sum_i a_i - \sum_j b_j. \qquad (8)$$

The first term vanishes since $\lim_{z\to\pm\infty} \log(f(z)) z = 0$ and we have applied the argument principle to the second term, with a contour chosen so that all indexes are equal to one. The zero of our function f is Ω_{ia}, its pole is ϵ_{ia}, hence the final expression :

$$E_c^{\text{dRPA-I}} = \frac{1}{2} \left\{\sum_{ia} \Omega_{ia} - \sum_{ia}(\epsilon_{ia} + K_{ia,ia})\right\}. \qquad (9)$$

(This is because in the particular case of simples poles in a_i, the residues take the value of $(z - a_i)f(z)$ for $z = a_i$).

II. PROOF OF VANISHING SECOND-ORDER CONTRIBUTIONS IN THE DERIVATION OF DRPA-IIA FROM DRPA-II

The minor correction to the integrand of Eq. (17) of the main paper directly reads as (without the unitary transformation used for the major contribution):

$$\alpha \, \text{tr}\left\{(\mathbf{I} - \alpha\mathbf{\Pi}_0\mathbf{K})^{-1}(\mathbf{\Pi}_0^+\mathbf{K}\,\mathbf{\Pi}_0^+ + \mathbf{\Pi}_0^-\mathbf{K}\,\mathbf{\Pi}_0^-)(\mathbf{A}' - \mathbf{B})\right\}. \qquad (10)$$

We consider the matrix identity

$$(\mathbf{I} - \alpha\mathbf{\Pi}_0\mathbf{K})^{-1} = \mathbf{I} + \alpha(\mathbf{I} - \alpha\mathbf{\Pi}_0\mathbf{K})^{-1}\mathbf{\Pi}_0\mathbf{K}$$
$$= \mathbf{Q}\left[\mathbf{I} + \alpha(\mathbf{I} - \alpha\mathbf{D})^{-1}\mathbf{D}\right]\mathbf{Q}^{-1}, \qquad (11)$$

where the diagonal matrix $\mathbf{D}(i\omega)$ contains the eigenvalues of $\mathbf{\Pi}_0(i\omega)\mathbf{K}$, and the matrix $\mathbf{Q}(i\omega)$ it's eigenvectors. The contribution coming from the identity term in the second member of Eq. (11) dominates over the contribution coming from the second term. Indeed, since $-\alpha d_{ia} > 0$ (d_{ia} being the diagonal elements of $\mathbf{D}$), we have that $|\alpha d_{ia}/(1 - \alpha d_{ia})| < 1$. Furthermore, since d_{ia} decreases rapidly to small values as a function of the index ia and of $i\omega$ [3], we can assume that $|\alpha d_{ia}/(1 - \alpha d_{ia})| \ll 1$ for most ia's.

By virtue of this, we include only the first contribution to Eq. (11), $\mathbf{I}$, in Eq. (10), and we are left with the task of proving that:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \left(\mathbf{\Pi}_0^+(i\omega)\mathbf{K}\mathbf{\Pi}_0^+(i\omega) + \mathbf{\Pi}_0^-(i\omega)\mathbf{K}\mathbf{\Pi}_0^-(i\omega)\right) = 0. \qquad (12)$$

We first note that:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \, \text{tr}\{\mathbf{\Pi}_0^-(i\omega)\mathbf{K}\mathbf{\Pi}_0^-(i\omega)\} = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \, \text{tr}\{\mathbf{\Pi}_0^+(i\omega)\mathbf{K}\mathbf{\Pi}_0^+(i\omega)\}, \qquad (13)$$

which may be seen by replacing ω with $-\omega$, adapting the integration limits and noting that $\mathbf{\Pi}_0^-(-i\omega) = \mathbf{\Pi}_0^+(i\omega)$. Now with

$$[\mathbf{\Pi}_0^+(i\omega)\mathbf{K}\mathbf{\Pi}_0^+(i\omega)]_{ia,jb} = \frac{K_{ia,jb}}{(i\epsilon_{ia} + \omega)(i\epsilon_{jb} + \omega)}, \quad (14)$$

one can see that the denominator of each of these integrals has simple poles at $\omega = -i\epsilon_{ia}$ and $\omega = -i\epsilon_{jb}$ if $\epsilon_{ia} \neq \epsilon_{jb}$ and a twofold pole otherwise. In any case it has no poles on the positive imaginary axis. Replacing ω by a complex number z and choosing an integration contour Γ which passes along the real axis from $-\infty$ to ∞ and encircles the complex half-plane with positive imaginary values in a counter-clockwise way and finally noting that the integrand vanishes quadratically for large $|z|$, it follows from the residue theorem that:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{1}{(i\epsilon_{ia} + \omega)(i\epsilon_{jb} + \omega)} = \int_{\Gamma} \frac{dz}{2\pi} \frac{1}{(i\epsilon_{ia} + z)(i\epsilon_{jb} + z)} = 0. \tag{15}$$

For the case $\epsilon_{ia} = \epsilon_{jb}$ the integral trivially vanishes since the integrand then corresponds to a member of order -2 in a Laurent series.

III.   DENSITY FITTING

Let us start from the expression for the ACFDT dRPA-IIa correlation energy of Eq. (21) of the main paper for a closed shell system, using spatial orbitals

$$E_c^{\text{dRPA-IIa}} = \frac{1}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \text{tr}\Big\{(\log(\mathbf{I} - \mathbf{\Pi}_0(i\omega)\mathbf{K}) + \mathbf{\Pi}_0(i\omega)\mathbf{K}) \times$$
$$\Big(\mathbf{1} - (\mathbf{\Pi}_0(i\omega)\mathbf{K})^{-1}\mathbf{\Pi}_0(i\omega)\tilde{\mathbf{K}}\Big)\Big\}, \quad (16)$$

where (in chemist's notation) $K_{ia,jb} = 2(ia|jb)$ and $\tilde{K}_{ia,jb} = (ib|ja)$. The non-interacting response function explicitly reads as

$$[\mathbf{\Pi}_0(i\omega)]_{ij,ab} = \frac{-2\epsilon_{ia}\delta_{ij}\delta_{ab}}{\epsilon_{ia}^2 + \omega^2}, \tag{17}$$

while the density fitting expression for the two types of two-electron integrals may be written as

$$K_{ia,jb} = \left[\mathbf{MM}^{\text{T}}\right]_{ia,jb} = \sum_G M_{ia,G}M_{G,jb} = 2\sum_G L_{ia,G}L_{G,jb} \tag{18}$$

$$\tilde{K}_{ia,jb} = \left[\mathbf{NN}^{\text{T}}\right]_{ia,jb} = \sum_G N_{ia,G}N_{G,jb} = \sum_G L_{ib,G}L_{G,ja}. \tag{19}$$

With these expressions, the first contribution to Eq. (16) corresponding to $E_c^{\text{dRPA-I}}$ can be written as

$$\text{tr}\{\log(\mathbf{I} - \mathbf{\Pi}_0(i\omega)\mathbf{K}) + \mathbf{\Pi}_0(i\omega)\mathbf{K}\}$$
$$= \text{tr}\Big\{\log\Big(\mathbf{I} - \mathbf{\Pi}_0(i\omega)\mathbf{MM}^{\text{T}}\Big) + \mathbf{\Pi}_0(i\omega)\mathbf{MM}^{\text{T}}\Big\}$$
$$= \text{tr}\Big\{\log\Big(\mathbf{I} - \mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}\Big) + \mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}\Big\}$$
$$= \text{tr}\{\log(\mathbf{I} - \mathbf{C}(i\omega)) + \mathbf{C}(i\omega)\}, \tag{20}$$

where we used the cyclic invariance of the trace and the fact that the non-zero eigenvalues of $\mathbf{\Pi}_0(i\omega)\mathbf{MM}^{\text{T}}$ and $\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}$ are identical. We defined:

$$C_{PQ}(i\omega) = [\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}]_{PQ} = \sum_{ia} L_{P,ia} \frac{-4\epsilon_{ia}}{\epsilon_{ia}^2 + \omega^2} L_{ia,Q}. \tag{21}$$

The exchange ("SOSEX-like") contribution to Eq. (16) is more complicated. In order to keep the notation as simple as possible we define the function g of a generic matrix $\mathbf{A}$ as

$$g(\mathbf{A}) = [\log(\mathbf{I} - \mathbf{A}) + \mathbf{A}]\mathbf{A}^{-1}. \tag{22}$$

In this definition we intend $\mathbf{A}^{-1}$ as a pseudoinverse that does not include the contribution from (almost) zero eigenvalues, as discussed in Sec. 2.2 of the main paper ; by passing to the density fitting or Cholesky representation a possible linear dependence of the matrix $\mathbf{A}$ is removed and the inverse of $\mathbf{A}$ is well defined and coincides with its pseudoinverse.

We can express the exchange contribution as

$$\text{tr}\Big\{(\log(\mathbf{I} - \mathbf{\Pi}_0(i\omega)\mathbf{K}) + \mathbf{\Pi}_0(i\omega)\mathbf{K})(\mathbf{\Pi}_0(i\omega)\mathbf{K})^{-1}\mathbf{\Pi}_0(i\omega)\tilde{\mathbf{K}}\Big\}$$
$$= \text{tr}\Big\{g(\mathbf{\Pi}_0(i\omega)\mathbf{K})\mathbf{\Pi}_0(i\omega)\tilde{\mathbf{K}}\Big\}$$
$$= \text{tr}\Big\{\mathbf{M}^{\text{T}} g(\mathbf{\Pi}_0(i\omega)\mathbf{MM}^{\text{T}})\mathbf{\Pi}_0(i\omega)\mathbf{NN}^{\text{T}}(\mathbf{MM}^{\text{T}})^{-1}\mathbf{M}\Big\}, \quad (23)$$

where we multiplied by $\mathbf{1} = (\mathbf{MM}^{\text{T}})^{-1}\mathbf{MM}^{\text{T}}$. The following identity (demonstrated in Appendix IV):

$$\text{tr}\Big\{\mathbf{M}^{\text{T}} g(\mathbf{\Pi}_0(i\omega)\mathbf{MM}^{\text{T}})\mathbf{B}\Big\} = \text{tr}\Big\{g(\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M})\mathbf{M}^{\text{T}}\mathbf{B}\Big\}, \tag{24}$$

allows us to further derive:

$$\text{tr}\Big\{(\log(\mathbf{I} - \mathbf{\Pi}_0(i\omega)\mathbf{K}) + \mathbf{\Pi}_0(i\omega)\mathbf{K})(\mathbf{\Pi}_0(i\omega)\mathbf{K})^{-1}\mathbf{\Pi}_0(i\omega)\tilde{\mathbf{K}}\Big\}$$
$$= \text{tr}\Big\{g(\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M})\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{NN}^{\text{T}}(\mathbf{MM}^{\text{T}})^{-1}\mathbf{M}\Big\}$$
$$= \text{tr}\Big\{(\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M})^{-1} g(\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}) \times$$
$$\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{NN}^{\text{T}}(\mathbf{MM}^{\text{T}})^{-1}\mathbf{MM}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}\Big\}$$
$$= \text{tr}\Big\{\mathbf{C}(i\omega)^{-1} g(\mathbf{C}(i\omega))\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{NN}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}\Big\}, \quad (25)$$

where we multiplied by $\mathbf{1} = \mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M}(\mathbf{M}^{\text{T}}\mathbf{\Pi}_0(i\omega)\mathbf{M})^{-1}$, applied the definition of $\mathbf{C}(i\omega)$ and resolved the inverse. The last matrix products under the trace read:

$$\left[\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0(\mathrm{i}\omega)\mathbf{N}\mathbf{N}^{\mathrm{T}}\mathbf{\Pi}_0(\mathrm{i}\omega)\mathbf{M}\right]_{QP} =$$

$$= \sum_{ia,jb}\sum_G \left(M_{Q,ia}\frac{-2\epsilon_{ia}}{\epsilon_{ia}^2+\omega^2}N_{ia,G}\right)\left(N_{G,jb}\frac{-2\epsilon_{jb}}{\epsilon_{jb}^2+\omega^2}M_{jb,P}\right)$$

$$= 2\sum_{ia,jb}\sum_G \left(L_{Q,ia}\frac{-2\epsilon_{ia}}{\epsilon_{ia}^2+\omega^2}L_{ib,G}\right)\left(L_{G,ja}\frac{-2\epsilon_{jb}}{\epsilon_{jb}^2+\omega^2}L_{jb,P}\right)$$

$$= 2\sum_{ij}\sum_G \left(\sum_a L_{Q,ia}\frac{-2\epsilon_{ia}}{\epsilon_{ia}^2+\omega^2}L_{ja,G}\right)\left(\sum_b L_{G,ib}\frac{-2\epsilon_{jb}}{\epsilon_{jb}^2+\omega^2}L_{jb,P}\right). \tag{26}$$

By introducing the quantities

$$[\mathbf{X}^{ij}(\mathrm{i}\omega)]_{QG} = \sum_a L_{Q,ia}\frac{-2\epsilon_{ia}}{\epsilon_{ia}^2+\omega^2}L_{ja,G}, \tag{27}$$

and

$$\mathbf{Y}(\mathrm{i}\omega) = 2\sum_{ij}\mathbf{X}^{ij}(\mathrm{i}\omega)\mathbf{X}^{ij}(\mathrm{i}\omega), \tag{28}$$

and remembering the definition of the matrix function g in Eq. (22), the general exchange term in Eq. (25) can be written as

$$\mathrm{tr}\left\{\left(\log\left(\mathbf{I}-\mathbf{C}(\mathrm{i}\omega)\right)+\mathbf{C}(\mathrm{i}\omega)\right)\mathbf{C}^{-1}(\mathrm{i}\omega)\mathbf{Y}(\mathrm{i}\omega)\mathbf{C}^{-1}(\mathrm{i}\omega)\right\}. \tag{29}$$

We note that $\mathbf{C}(\mathrm{i}\omega)$ can be defined from the "diagonal elements" of the $\mathbf{X}^{ij}(\mathrm{i}\omega)$ super-matrices:

$$C_{PQ}(\mathrm{i}\omega) = \sum_{ia} L_{P,ia}\frac{-4\epsilon_{ia}}{\epsilon_{ia}^2+\omega^2}L_{ia,Q} = 2\sum_i X_{PQ}^{ii}(\mathrm{i}\omega). \tag{30}$$

Combining everything together the density-fitting dRPA-IIa correlation energy in Eq. (16) finally becomes:

$$E_{\mathrm{c}}^{\mathrm{dRPA\text{-}IIa}} = \int_0^\infty \frac{d\omega}{2\pi}\,\mathrm{tr}\Big\{\left(\log(\mathbf{1}-\mathbf{C}(\mathrm{i}\omega))+\mathbf{C}(\mathrm{i}\omega)\right)\times$$

$$\left(\mathbf{1}-\mathbf{C}^{-1}(\mathrm{i}\omega)\mathbf{Y}(\mathrm{i}\omega)\mathbf{C}^{-1}(\mathrm{i}\omega)\right)\Big\}. \tag{31}$$

IV. PROOF OF EQ. (24)

In order to derive the final result of Appendix III it is necessary to demonstrate the validity of the equation (in order to keep the notation simple, the explicit dependence of $\mathbf{\Pi}_0$ on iω has been dropped):

$$\mathrm{tr}\left\{\mathbf{M}^{\mathrm{T}}g(\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}})\mathbf{B}\right\} = \mathrm{tr}\left\{g(\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0\mathbf{M})\mathbf{M}^{\mathrm{T}}\mathbf{B}\right\}, \tag{32}$$

where $\mathbf{B}=\mathbf{\Pi}_0\mathbf{N}\mathbf{N}^{\mathrm{T}}(\mathbf{M}\mathbf{M}^{\mathrm{T}})^{-1}\mathbf{M}$ is the ($N_{\mathrm{exc}}\times N_{\mathrm{aux}}$) matrix seen in Eq. (23) and $\mathbf{M}\mathbf{M}^{\mathrm{T}}$ denotes the Cholesky decomposition in Eq. (18). By remembering that a generic function of a matrix is defined as $f(\mathbf{A})=\mathbf{Q}f(\mathbf{D})\mathbf{Q}^{-1}$, we have

$$\mathrm{tr}\left\{\mathbf{M}^{\mathrm{T}}g(\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}})\mathbf{B}\right\} =$$

$$\sum_G^{N_{\mathrm{aux}}}\sum_{ia,jb,kc}^{N_{\mathrm{exc}}} M_{G,ia}\,Q_{ia,kc}\,g(d_{kc})\,Q_{kc,jb}^{-1}\,B_{jb,G}. \tag{33}$$

From the definition of the function g in Eq. (22) we have that $g(0)=0$, namely the zero (or below a certain threshold) eigenvalues d_{kc} do not contribute to Eq. (33), that can be rewritten as

$$\mathrm{tr}\left\{\mathbf{M}^{\mathrm{T}}g(\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}})\mathbf{B}\right\} =$$

$$\sum_G^{N_{\mathrm{aux}}}\sum_{ia,jb}^{N_{\mathrm{exc}}}\sum_{kc}^{N_{\mathrm{eig}}} M_{G,ia}\,Q_{ia,kc}\,g(d_{kc})\,Q_{kc,jb}^{-1}\,B_{jb,G}, \tag{34}$$

where N_{eig} indicate that the sum is performed only for $d_{kc}\neq 0$. In Eqs. (33-34) the index G runs over the N_{aux} auxiliary vectors generated by the Cholesky decomposition. These auxiliary vectors span a basis set that "removes" a possible linear dependence in the two-electron integrals $\mathbf{K}$ and $\tilde{\mathbf{K}}$. Since this is equivalent to removing the contribution of the 0 eigenvalues from the matrix $\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}$, we can assume that $N_{\mathrm{eig}}=N_{\mathrm{aux}}$. In compact matrix notation we can write Eq. (34) as

$$\mathrm{tr}\left\{\mathbf{M}^{\mathrm{T}}g(\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}})\mathbf{B}\right\} = \mathrm{tr}\left\{\mathbf{M}^{\mathrm{T}}\bar{\mathbf{Q}}\,g(\bar{\mathbf{D}})\,\bar{\mathbf{Q}}^-\mathbf{B}\right\}, \tag{35}$$

where we have defined the ($N_{\mathrm{exc}}\times N_{\mathrm{aux}}$) rectangular matrix $\bar{\mathbf{Q}}$, the ($N_{\mathrm{aux}}\times N_{\mathrm{exc}}$) rectangular matrix $\bar{\mathbf{Q}}^-$, and the $N_{\mathrm{aux}}\times N_{\mathrm{aux}}$ diagonal matrix $\bar{\mathbf{D}}$ that contains the non-zero eigenvalues of the non-Hermitian matrix $\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}$. The matrix $\bar{\mathbf{Q}}$ ($\bar{\mathbf{Q}}^-$) contains the right (left) eigenvectors of $\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}$ corresponding to non-zero eigenvalues. One should note that by definition $\bar{\mathbf{Q}}^-\bar{\mathbf{Q}}=\mathbf{1}$ (the ($N_{\mathrm{aux}}\times N_{\mathrm{aux}}$) identity matrix) while $\bar{\mathbf{Q}}\bar{\mathbf{Q}}^-\neq\mathbf{I}$ (the ($N_{\mathrm{exc}}\times N_{\mathrm{exc}}$) identity matrix).

The eigenvalue problem corresponding to the non-zero eigenvalues of the $\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}$ can be written in matrix form as

$$\left(\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}\right)\bar{\mathbf{Q}} = \bar{\mathbf{Q}}\bar{\mathbf{D}}. \tag{36}$$

which we can multiply by $\mathbf{M}^{\mathrm{T}}$ to obtain:

$$\left(\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0\mathbf{M}\right)\mathbf{M}^{\mathrm{T}}\bar{\mathbf{Q}} = \mathbf{M}^{\mathrm{T}}\bar{\mathbf{Q}}\bar{\mathbf{D}}, \tag{37}$$

and to find that the columns of the ($N_{\mathrm{aux}}\times N_{\mathrm{aux}}$) nonsingular square matrix $\mathbf{M}^{\mathrm{T}}\bar{\mathbf{Q}}$ are the eigenvectors of the Hermitian matrix $\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0\mathbf{M}$. The comparison of Eq. (37) and Eq. (36) implies that the non-zero eigenvalues of the matrices $\mathbf{\Pi}_0\mathbf{M}\mathbf{M}^{\mathrm{T}}$ and $\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0\mathbf{M}$ are identical. Eq. (37) can be used to define the function g on the matrix $\mathbf{M}^{\mathrm{T}}\mathbf{\Pi}_0\mathbf{M}$:

$$g(\mathbf{M}^T \mathbf{\Pi}_0 \mathbf{M}) = (\mathbf{M}^T \bar{\mathbf{Q}}) \, g(\bar{\mathbf{D}}) \, (\mathbf{M}^T \bar{\mathbf{Q}})^{-1}. \qquad (38)$$

Hence, we have:

$$
\begin{aligned}
g(\mathbf{M}^T \mathbf{\Pi}_0 \mathbf{M}) \, \mathbf{M}^T \mathbf{B} &= \mathbf{M}^T \bar{\mathbf{Q}} \, g(\bar{\mathbf{D}}) \, (\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T \mathbf{B} \\
&= \mathbf{M}^T \bar{\mathbf{Q}} \, g(\bar{\mathbf{D}}) \, \bar{\mathbf{Q}}^- \mathbf{B}, \qquad (39)
\end{aligned}
$$

where in the last step we assumed that

$$(\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T = \bar{\mathbf{Q}}^-. \qquad (40)$$

This identity can be inferred from the fact that by definition the rows of the matrix $\bar{\mathbf{Q}}^-$ contain the left eigenvectors of the non-Hermitian matrix $\mathbf{\Pi}_0 \mathbf{M} \mathbf{M}^T$ corresponding to non-zero eigenvalues:

$$\bar{\mathbf{Q}}^- \mathbf{\Pi}_0 \mathbf{M} \mathbf{M}^T = \bar{\mathbf{D}} \bar{\mathbf{Q}}^-, \qquad (41)$$

and so do the rows of the matrix $(\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T$ since

$$
\begin{aligned}
(\mathbf{M}^T & \bar{\mathbf{Q}})^{-1} \mathbf{M}^T \mathbf{\Pi}_0 \mathbf{M} \mathbf{M}^T \\
&= (\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T \mathbf{\Pi}_0 \mathbf{M} (\mathbf{M}^T \bar{\mathbf{Q}})(\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T \\
&= \bar{\mathbf{D}} (\mathbf{M}^T \bar{\mathbf{Q}})^{-1} \mathbf{M}^T, \qquad (42)
\end{aligned}
$$

where the result in Eq. (37) has been used.

By using the results in Eq. (35) and in Eq. (39) we have

$$
\begin{aligned}
\text{tr} \left\{ \mathbf{M}^T g(\mathbf{\Pi}_0 \mathbf{M} \mathbf{M}^T) \mathbf{B} \right\} &= \text{tr} \left\{ \mathbf{M}^T \bar{\mathbf{Q}} g(\bar{\mathbf{D}}) \bar{\mathbf{Q}}^- \mathbf{B} \right\} \\
&= \text{tr} \left\{ g(\mathbf{M}^T \mathbf{\Pi}_0 \mathbf{M}) \mathbf{M}^T \mathbf{B} \right\}, \qquad (43)
\end{aligned}
$$

that demonstrates the identity in Eq. (24).

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