Coupling-constant-dependent ERPA matrices

For a given reference wavefunction $\Psi^{\text{ref}}$, introduce spin-free 1- and 2-RDMs, $\gamma$ and $\Gamma$, respectively, as

$$
\gamma_{pq} = \langle \Psi^{\text{ref}} | \alpha_{q\alpha}^\dagger \alpha_{p\alpha} | \Psi^{\text{ref}} \rangle + \langle \Psi^{\text{ref}} | \alpha_{q\beta}^\dagger \alpha_{p\beta} | \Psi^{\text{ref}} \rangle
$$

(1)
and

\[ 2\Gamma_{pqrs} = \langle \Psi^\text{ref} | \hat{a}_{r\alpha}^\dagger \hat{a}_{s\beta}^\dagger \hat{a}_{q\alpha} \hat{a}_{p\beta} | \Psi^\text{ref} \rangle + \langle \Psi^\text{ref} | \hat{a}_{r\alpha}^\dagger \hat{a}_{s\beta}^\dagger \hat{a}_{q\beta} \hat{a}_{p\alpha} | \Psi^\text{ref} \rangle \\
+ \langle \Psi^\text{ref} | \hat{a}_{r\beta}^\dagger \hat{a}_{s\alpha}^\dagger \hat{a}_{q\alpha} \hat{a}_{p\beta} | \Psi^\text{ref} \rangle + \langle \Psi^\text{ref} | \hat{a}_{r\beta}^\dagger \hat{a}_{s\alpha}^\dagger \hat{a}_{q\beta} \hat{a}_{p\alpha} | \Psi^\text{ref} \rangle . \]  

(2)

Using the representation of the natural orbitals

\[ \gamma_{pq} = 2\delta_{pq}n_p \]  

(3)

(notice that \( \forall p \ 0 \leq n_p \leq 1 \)) the spin-free ERPA matrices

\[ \mathcal{A}_+^\alpha = N^{-1/2} (\mathcal{A}^\alpha + \mathcal{B}^\alpha) N^{-1/2} \]  

(4)

\[ \mathcal{A}_-^\alpha = N^{-1/2} (\mathcal{A}^\alpha - \mathcal{B}^\alpha) N^{-1/2} \]  

(5)

\[ \forall p > q \quad \forall r > s \quad N_{pq,rs} = \delta_{pr}\delta_{qs}(n_p - n_q) \]  

(6)

are given explicitly in terms of 1- and 2-RDMs and the pertinent expression reads

\[
\forall_{pqrs} \quad [\mathcal{A}^\alpha]_{pq,rs} = [\mathcal{B}^\alpha]_{pq,rs} \\
= h_{sq}^\alpha \delta_{pr}(n_p - n_s) + h_{rp}^\alpha \delta_{sq}(n_q - n_r) \\
+ \sum_{tu} \langle st|qu \rangle^\alpha \Gamma_{putr} + \sum_{tu} \langle st|uq \rangle^\alpha \Gamma_{putr} \\
+ \sum_{tu} \langle up|tr \rangle^\alpha \Gamma_{stqu} + \sum_{tu} \langle up|rt \rangle^\alpha \Gamma_{stuq} \\
- \sum_{tu} \langle ps|tu \rangle^\alpha \Gamma_{tuqr} - \sum_{tu} \langle tu|qr \rangle^\alpha \Gamma_{sput} \\
- \frac{1}{2} \delta_{sq} \sum_{tuv} \langle tp|wv \rangle^\alpha \Gamma_{wutr} - \frac{1}{2} \delta_{sq} \sum_{tuv} \langle tp|uw \rangle^\alpha \Gamma_{wurt} \\
- \frac{1}{2} \delta_{pr} \sum_{tuv} \langle tu|wq \rangle^\alpha \Gamma_{swut} - \frac{1}{2} \delta_{pr} \sum_{tuv} \langle tu|qw \rangle^\alpha \Gamma_{satu} .
\]  

(7)
The coupling-constant-dependent modified one- and two-electron integrals are defined as

\[ h_{pq}^\alpha = \alpha h_{pq} + \delta_{I_pI_q} (1 - \alpha) h_{pq}^{\text{eff}} \]  
(8)

\[ h_{pq}^{\text{eff}} = h_{pq} + \sum_{J \neq I} \sum_{r \in J} n_r [2 \langle pr | qr \rangle - \langle pr | rq \rangle] \]  
(9)

\[ \langle pq | rs \rangle_\alpha = [\alpha + \delta_{I_pI_s} \delta_{I_qI_r} (1 - \alpha)] \langle pq | rs \rangle \]  
(10)

the symbol \( \delta_{I_pI_q} \) stands for 1 if orbitals \( p \) and \( q \) are from the same orbital group (both are inactive, occupied or virtual) and 0 otherwise.

The matrices \( \mathcal{A}_\pm^{(0)} \) and \( \mathcal{A}_\pm^{(1)} \) needed to find the AC correlation energy using the iterative scheme and the Cholesky decomposition, see Eqs. (35)-(39) in the main text, follow immediately from Eqs. (4), (5), namely

\[ \mathcal{A}_\pm^{(0)} = \mathcal{A}^{(0)} \pm B^{(0)} \]  
(11)

\[ \mathcal{A}_\pm^{(1)} = \mathcal{A}^{(1)} \pm B^{(1)} \]  
(12)

where the matrices \( \mathcal{A}^{(0)}, B^{(0)} \) are obtained from Eq. (7) by replacing \( h_{pq}^\alpha \) and \( \langle pq | rs \rangle_\alpha \) integrals with their zero-order counterparts \( h_{pq}^{(0)}, \langle pq | rs \rangle^{(0)} \) reading

\[ h_{pq}^{(0)} = \delta_{I_pI_q} h_{pq}^{\text{eff}} \]  
(13)

\[ \langle pq | rs \rangle^{(0)} = \delta_{I_pI_s} \delta_{I_qI_r} \langle pq | rs \rangle \]  
(14)

Similarly, first-order matrices \( \mathcal{A}^{(1)}, B^{(1)} \) follow from Eq. (7) if the elements

\[ h_{pq}^{(1)} = \alpha h_{pq} - \delta_{I_pI_q} h_{pq}^{\text{eff}} \]  
(15)

\[ \langle pq | rs \rangle^{(1)} = (1 - \delta_{I_pI_s} \delta_{I_qI_r}) \langle pq | rs \rangle \]  
(16)

replace \( h_{pq}^\alpha \) and \( \langle pq | rs \rangle_\alpha \), respectively.
Frequency-dependent density-density linear response function in the ERPA approximation

Begin with the coupling-constant dependent ERPA problem

$$
\begin{pmatrix}
A_\alpha - & 0 \\
0 & A_\alpha +
\end{pmatrix}
\begin{pmatrix}
\tilde{Y}_\nu^\alpha \\
\tilde{X}_\nu^\alpha
\end{pmatrix}
= \omega_\nu^\alpha
\begin{pmatrix}
0 & 1 \\
1 & 0
\end{pmatrix}
\begin{pmatrix}
\tilde{Y}_\nu^\alpha \\
\tilde{X}_\nu^\alpha
\end{pmatrix},
$$

(17)

where the eigenvectors are orthonormal in the following sense

$$
\forall_{\mu,\nu} \quad 2\tilde{Y}_\mu^\alpha \tilde{X}_\nu^\alpha = \delta_{\mu\nu}
$$

(18)

The matrices $A_{\pm}^\alpha$, defined in Eqs. (4)-(7), are assumed to be positive definite. This condition is met if the reference wavefunction satisfies the Hellman-Feynman theorem, since in such a case the EPRA matrices are equivalent to the Hessian matrix describing variations of energy with respect to orbital rotations. It can be shown [see theorem A.5 in Furche, JCP 114, 5982 (2001)] that spectral decomposition of the matrices $A_{\pm}^\alpha$ is given by eigenvalues and eigenvectors

$$
\tilde{X}_\nu^\alpha = \sqrt{2}\tilde{X}_\nu^\alpha
$$

(19)

$$
\tilde{Y}_\nu^\alpha = \sqrt{2}\tilde{Y}_\nu^\alpha
$$

(20)

orthonormal in the following sense

$$
\forall_{\mu,\nu} \quad \tilde{Y}_\mu^\alpha \tilde{X}_\nu^\alpha = \delta_{\mu\nu}
$$

(21)

as
\[ \mathcal{A}_+^\alpha = \left( \begin{array}{ccccc} \ddot{Y}_1^\alpha & \ddot{Y}_2^\alpha & \cdots \\ \end{array} \right) \left( \begin{array}{cccc} \omega_1^\alpha & 0 & \cdots \\ 0 & \omega_2^\alpha & \cdots \\ \cdots & \cdots & \cdots \\ \end{array} \right) \left( \begin{array}{c} [\dot{Y}_1^\alpha]^T \\ [\dot{Y}_2^\alpha]^T \\ \vdots \\ \end{array} \right) \]  

\hspace{1cm} (22) 

and

\[ \mathcal{A}_-^\alpha = \left( \begin{array}{ccccc} \ddot{X}_1^\alpha & \ddot{X}_2^\alpha & \cdots \\ \end{array} \right) \left( \begin{array}{cccc} \omega_1^\alpha & 0 & \cdots \\ 0 & \omega_2^\alpha & \cdots \\ \cdots & \cdots & \cdots \\ \end{array} \right) \left( \begin{array}{c} [\dot{X}_1^\alpha]^T \\ [\dot{X}_2^\alpha]^T \\ \vdots \\ \end{array} \right). \]  

\hspace{1cm} (23) 

Using the orthonormality condition, Eq. (21), and Eqs. (19)-(20), it can be checked by inspection that a matrix \( C^\alpha(\omega) \) satisfying the following equation

\[ \left[ \mathcal{A}_+^\alpha \mathcal{A}_-^\alpha + \omega^2 \mathbf{1} \right] C^\alpha(\omega) = \mathcal{A}_+^\alpha \]  

\hspace{1cm} (24)

is the dynamic linear response function, see Eq. (21) in the main text,

\[ \forall_{p>q} \quad C^\alpha_{pq,rs}(\omega) = 2 \sum_{\nu} \left[ \ddot{Y}_\nu^\alpha \right]_{pq} \frac{\omega_\nu}{(\omega_\nu)^2 + \omega^2} \left[ \ddot{Y}_\nu^\alpha \right]_{rs}. \]  

\hspace{1cm} (25)
Table 1: Convergence check for biradicals molecules. Energies in [Ha], ST gaps (differences between triplet and singlet energies) in [eV]. Systems (Sys.): 1 – C₄H₄, 2 – C₅H₅⁺, 3 – C₄H₃NH₂, 4 – C₄H₅CHO, 5 – C₄H₂NH₂(CHO), 6 – C₄H₂-1,2-(CH₂)₂, 7 – C₄H₂-1,3-(CH₂)₂, 8 – divergent case of C₅H₅⁺ and CAS(2,4).

| Sys. | n  | ACₙ(S) | ACₙ(T) | ACₙ(S)-AC(S) | ACₙ(T)-AC(T) | ACₙ(ST) | ACₙ(ST)-AC(ST) |
|------|----|--------|--------|--------------|--------------|---------|----------------|
| 1    | 1  | -154.38675 | -154.38681 | -0.01993 | -0.02478 | -0.00144 | -0.13201 |
|      | 2  | -154.35211 | -154.36192 | 0.01471 | 0.00011 | -0.26684 | -0.39741 |
|      | 3  | -154.37064 | -154.36790 | -0.00381 | -0.00587 | 0.07450 | -0.05607 |
|      | 4  | -154.36520 | -154.35905 | 0.00162 | 0.00297 | 0.16744 | 0.03687 |
|      | 5  | -154.36751 | -154.36354 | -0.00069 | -0.00151 | 0.10811 | -0.02246 |
|      | 7  | -154.36697 | -154.36251 | -0.00015 | -0.00049 | 0.12129 | -0.00928 |
|      | 10 | -154.36681 | -154.36192 | 0.00001 | 0.00011 | 0.13320 | 0.00263 |
|      | 12 | -154.36682 | -154.36198 | 0.00000 | 0.00004 | 0.13174 | 0.00117 |
|      | 15 | -154.36683 | -154.36204 | 0.00000 | -0.00002 | 0.13021 | -0.00036 |
|      | AC | -154.36682 | -154.36202 | - | - | 0.13057 | - |
| 2    | 1  | -192.78598 | -192.81767 | -0.02010 | -0.02386 | -0.86239 | -0.10249 |
|      | 2  | -192.74597 | -192.77359 | 0.01992 | 0.02022 | -0.75165 | 0.00824 |
|      | 3  | -192.77033 | -192.79950 | -0.00444 | -0.00569 | -0.79389 | -0.03400 |
|      | 4  | -192.76255 | -192.79086 | 0.00333 | 0.00295 | -0.77025 | -0.01035 |
|      | 5  | -192.76682 | -192.79519 | -0.00094 | -0.00139 | -0.77207 | -0.01217 |
|      | 7  | -192.76610 | -192.79422 | -0.00022 | -0.00041 | -0.76512 | -0.00523 |
|      | 10 | -192.76553 | -192.79372 | 0.00035 | 0.00009 | -0.76691 | -0.00702 |
|      | 12 | -192.76565 | -192.79377 | 0.00023 | 0.00004 | -0.76510 | -0.00521 |
|      | 15 | -192.76586 | -192.79381 | 0.00003 | 0.00000 | -0.76059 | -0.00069 |
|      | AC | -192.76588 | -192.79381 | - | - | -0.75989 | - |
| 3    | 1  | -209.68148 | -209.68454 | -0.03926 | -0.04462 | -0.08319 | -0.14584 |
|      | 2  | -209.61974 | -209.61310 | 0.02248 | 0.02682 | 0.18065 | 0.11800 |
| Sys. | n  | $AC_n(S)$ | $AC_n(T)$ | $AC_n(S)$-$AC(S)$ | $AC_n(T)$-$AC(T)$ | $AC_n(ST)$ | $AC_n(ST)$-$AC(ST)$ |
|------|----|----------|----------|-----------------|-----------------|-----------|-------------------|
| 3    | -209.64932 | -209.64934 | -0.00709 | -0.00942 | -0.00061 | -0.06326 |
| 4    | -209.63933 | -209.63531 | 0.00289 | 0.00461 | 0.10950 | 0.04685 |
| 5    | -209.64365 | -209.64235 | -0.00142 | -0.00243 | 0.03523 | -0.02742 |
| 7    | -209.64256 | -209.64071 | -0.0033 | -0.0079 | 0.05023 | -0.01242 |
| 10   | -209.64216 | -209.63970 | 0.00006 | 0.0022 | 0.06702 | 0.00437 |
| 12   | -209.64219 | -209.63981 | 0.00003 | 0.0011 | 0.06479 | 0.00214 |
| 15   | -209.64220 | -209.63993 | 0.00002 | 0.0000 | 0.06193 | -0.00072 |
| AC   | -209.64222 | -209.63992 | - | - | 0.06265 | - |

| 1    | -267.57835 | -267.57912 | -0.06117 | -0.06606 | -0.02111 | -0.13292 |
| 2    | -267.48330 | -267.47536 | 0.03387 | 0.03771 | 0.21615 | 0.10434 |
| 3    | -267.53097 | -267.52893 | -0.01379 | -0.01587 | 0.05536 | -0.05646 |
| 4    | -267.51019 | -267.50456 | 0.00699 | 0.00851 | 0.15309 | 0.04128 |
| 5    | -267.52136 | -267.51816 | -0.00418 | -0.00509 | 0.08705 | -0.02476 |
| 7    | -267.51880 | -267.51512 | -0.00162 | -0.00205 | 0.09996 | -0.01185 |
| 10   | -267.51662 | -267.51233 | 0.00056 | 0.00074 | 0.11661 | 0.00480 |
| 12   | -267.51686 | -267.51265 | 0.00032 | 0.00042 | 0.11454 | 0.00273 |
| 15   | -267.51724 | -267.51318 | -0.00006 | -0.00011 | 0.11055 | -0.00126 |
| AC   | -267.51718 | -267.51307 | - | - | 0.11181 | - |

| 1    | -322.88405 | -322.87894 | -0.07863 | -0.08765 | 0.13894 | -0.24570 |
| 2    | -322.75998 | -322.74413 | 0.04545 | 0.04716 | 0.43122 | 0.04658 |
| 3    | -322.82401 | -322.81120 | -0.01858 | -0.01991 | 0.34854 | -0.03610 |
| 4    | -322.79486 | -322.78070 | 0.01057 | 0.01059 | 0.38516 | 0.00052 |
| 5    | -322.81221 | -322.79752 | -0.00679 | -0.00624 | 0.39960 | 0.01496 |
| 7    | -322.80884 | -322.79377 | -0.00341 | -0.00248 | 0.41001 | 0.02537 |
| 10   | -322.80384 | -322.79036 | 0.00159 | 0.00093 | 0.36678 | -0.01786 |
| 12   | -322.80431 | -322.79075 | 0.00111 | 0.00054 | 0.36910 | -0.01553 |

S7
| Sys. | n  | $AC_n(S)$ | $AC_n(T)$ | $(S) - AC(S)$ | $(T) - AC(T)$ | $(ST) - AC(ST)$ | $AC_n(ST)$ | $AC_n(ST) - AC(ST)$ |
|------|----|-----------|-----------|--------------|--------------|----------------|-----------|-------------------|
| 15   |    | -322.80606 | -322.79141 | -0.00064    | -0.00012     | 0.39881        | 0.01417   |                   |
| AC   |    | -322.80542 | -322.79129 | -0.00065    | -0.00012     | 0.38464        | -         |                   |
| 1    |    | -231.74407 | -231.61704 | -0.03381    | -0.03307     | 3.45658        | 0.02014   |                   |
| 2    |    | -231.68434 | -231.56013 | 0.02591     | 0.02384      | 3.38011        | -0.05632  |                   |
| 3    |    | -231.71815 | -231.59113 | -0.00790    | -0.00716     | 3.45641        | 0.01998   |                   |
| 4    |    | -231.70636 | -231.58071 | 0.00389     | 0.00326      | 3.41918        | -0.01725  |                   |
| 5    |    | -231.71231 | -231.58559 | -0.00206    | -0.00163     | 3.44816        | 0.01173   |                   |
| 6    |    | -231.71092 | -231.58440 | -0.00067    | -0.00043     | 3.44276        | 0.00632   |                   |
| 7    |    | -231.71012 | -231.58386 | 0.00014     | 0.00011      | 3.43570        | -0.00074  |                   |
| 10   |    | -231.71018 | -231.58390 | 0.00007     | 0.00007      | 3.43621        | -0.00022  |                   |
| 12   |    | -231.71024 | -231.58394 | 0.00001     | 0.00003      | 3.43697        | 0.00054   |                   |
| AC   |    | -231.71026 | -231.58397 | -0.00008    | -0.00003     | 3.43643        | -         |                   |
| 1    |    | -231.64530 | -231.67913 | -0.03032    | -0.03372     | -0.92055       | -0.01902  |                   |
| 2    |    | -231.58760 | -231.62092 | 0.02469     | 0.02450      | -0.90673       | -0.00520  |                   |
| 3    |    | -231.61972 | -231.65290 | -0.00744    | -0.00749     | -0.90286       | -0.00133  |                   |
| 4    |    | -231.60880 | -231.64197 | 0.00348     | 0.00344      | -0.90265       | -0.00112  |                   |
| 5    |    | -231.61416 | -231.64717 | -0.00187    | -0.00176     | -0.89839       | 0.00314   |                   |
| 7    |    | -231.61288 | -231.64591 | -0.00060    | -0.00049     | -0.89866       | 0.00288   |                   |
| 10   |    | -231.61219 | -231.64531 | 0.00010     | 0.00011      | -0.90132       | 0.00021   |                   |
| 12   |    | -231.61224 | -231.64536 | 0.00005     | 0.00006      | -0.90128       | 0.00025   |                   |
| 15   |    | -231.61228 | -231.64540 | 0.00000     | 0.00002      | -0.90104       | 0.00050   |                   |
| AC   |    | -231.61228 | -231.64541 | -0.00000    | -0.00000     | -0.90153       | -         |                   |
| 1    |    | -192.79829 | -192.82560 | -0.04506    | -0.03862     | -0.74314       | 0.17552   |                   |
| 2    |    | -192.72167 | -192.75934 | 0.03156     | 0.02764      | -1.02505       | -0.10639  |                   |
| 3    |    | -192.77238 | -192.79638 | -0.01916    | -0.00940     | -0.65307       | 0.26559   |                   |
| Sys. | n   | AC_n(S) | AC_n(T) | AC_n(S)-AC(S) | AC_n(T)-AC(T) | AC_n(ST) | AC_n(ST)-AC(ST) |
|------|-----|---------|---------|--------------|--------------|----------|-----------------|
| 4    | -192.74718 | -192.78110 | 0.00605 | 0.00589 | -0.92301 | -0.00435 |
| 5    | -192.76827 | -192.78992 | -0.01505 | -0.00294 | -0.58913 | 0.32953 |
| 6    | -192.75531 | -192.78477 | -0.00208 | 0.00222 | -0.80165 | 0.11701 |
| 7    | -192.77100 | -192.78820 | -0.01778 | -0.00121 | -0.46776 | 0.45090 |
| 8    | -192.76309 | -192.78593 | -0.00986 | 0.00106 | -0.62151 | 0.29715 |
| 9    | -192.77912 | -192.78757 | -0.02589 | -0.00058 | -0.23021 | 0.68845 |
| 10   | -192.77532 | -192.78642 | -0.02209 | 0.00057 | -0.30205 | 0.61661 |
| AC   | -192.75323 | -192.78699 | - | - | -0.91866 | - |
Timings

Table 2 shows timings for AC, AC\(_n\) and AC0 correlation energy calculations for singlet energy of C\(_4\)H\(_2\)-1,3-(CH\(_2\))\(_2\) biradical in two basis sets: aug-cc-pVTZ (aTZ) and aug-cc-pVQZ (aQZ).

Table 2: CPU times of computing correlation energy for C\(_4\)H\(_2\)-1,3-(CH\(_2\))\(_2\) in two basis sets: aug-cc-pVTZ (aTZ) and aug-cc-pVQZ (aQZ). AC\(_n\) corresponds to n=10. The timings were measured on a 4 core Intel(R) Xeon(R) Gold 6240 CPU machine clocked at 2.60 GHz using a threaded MKL BLAS library.

| active space | AC aTZ | AC aQZ | AC\(_n\) aTZ | AC\(_n\) aQZ | AC0 aTZ | AC0 aQZ |
|--------------|--------|--------|---------------|---------------|--------|--------|
| (4,4)        | 3h 14m 3s | 19h 55m 18s | 42m 9s | 4h 1m 17s | 22s | 1m 26s |
| (6,6)        | 3h 54m 36s | 21h 38m 33s | 46m 46s | 4h 9m 57s | 28s | 1m 36s |
| (8,8)        | 3h 56m 39s | 24h 52m 45s | 50m 15s | 4h 28m 1s | 37s | 2m 22s |
DMRG single-orbital entropies and mutual information for selected biradicals

For all studied biradicals we have used geometries from Ref. [1] and aug-cc-pVTZ basis set. [2] For the representative examples of C_4H_4 and C_5H_5^+ we have performed the CASSCF orbital optimizations in the medium size active spaces of Ref. [3] (mCPO). Moreover, for C_4H_4 we have also performed DMRG-SCF optimizations in eCPO with fixed bond dimensions $M = 2000$, i.e. CAS(20,22), to verify that there is essentially no difference between correlations in mCPO and eCPO. In other words, the additional orbitals not included in mCPO are only weakly correlated. Finally, we have computed the DMRG ($M_{\text{max}} = 4000$) single-orbital entropies ($s_i$), as well as mutual information ($I_{ij}$), [4] which provide additional information on the correlation structure.

The DMRG single-orbital entropies for C_4H_4 (eCPO, mCPO) and C_5H_5^+ (mCPO) are shown in Figures 1 - 3.

![Figure 1: The DMRG single-orbital entropies for C_4H_4 and eCPO, i.e. CAS(20,22). The orbital indices correspond to the ordering from Figures 4 and 5.](image-url)
CASSCF natural orbitals for selected biradicals

The DMRG-SCF (C₄H₄, eCPO) natural orbitals with their occupation numbers are depicted in Figures 4 and 5. CASSCF (C₄H₄, C₅H₅⁺, mCPO) natural orbitals and the corresponding occupation numbers are depicted in Figures 6, 7, 8, and 9.
Figure 4: C₄H₄, singlet state, DMRG-SCF(20, 22)
Figure 5: C₄H₄, triplet state, DMRG-SCF(20, 22)
Figure 6: C\textsubscript{4}H\textsubscript{4}, singlet state, CASSCF(12, 14)
Figure 7: C$_{4}$H$_{4}$, triplet state, CASSCF(12, 14)
Figure 8: C₅H₅, singlet state, CASSCF(14, 16)
Figure 9: C$_5$H$_5$, triplet state, CASSCF(14, 16)
References

(1) Saito, T.; Nishihara, S.; Yamanaka, S.; Kitagawa, Y.; Kawakami, T.; Yamada, S.; Isobe, H.; Okumura, M.; Yamaguchi, K. Symmetry and broken symmetry in molecular orbital description of unstable molecules IV: comparison between single-and multi-reference computational results for antiaromatic molecules. *Theor. Chem. Acc.* **2011**, *130*, 749–763.

(2) Kendall, R. A.; Dunning Jr, T. H.; Harrison, R. J. Electron affinities of the first-row atoms revisited. Systematic basis sets and wave functions. *J. Chem. Phys.* **1992**, *96*, 6796–6806.

(3) Stoneburner, S. J.; Shen, J.; Ajala, A. O.; Piecuch, P.; Truhlar, D. G.; Gagliardi, L. Systematic design of active spaces for multi-reference calculations of singlet–triplet gaps of organic diradicals, with benchmarks against doubly electron-attached coupled-cluster data. *J. Chem. Phys.* **2017**, *147*, 164120.

(4) Szalay, S.; Pfeffer, M.; Murg, V.; Barcza, G.; Verstraete, F.; Schneider, R.; Örs Legeza, Tensor product methods and entanglement optimization for ab initio quantum chemistry. *Int. J. Quant. Chem.* **2015**, *115*, 1342–1391.