Supporting Information for: "Description of Sudden Polarization in the Excited Electronic States with an Ensemble Density Functional Theory Method"

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Contents

1 The SSR(2,2) method

The SSR(2,2) energies are

$$E_0^{SSR} = a_{00}^2 E_0^{PPS} + a_{10}^2 E_1^{OSS} + 2a_{00}a_{10}\Delta_{01}^{SA} \tag{1a}$$
$$E_1^{SSR} = a_{01}^2 E_0^{PPS} + a_{11}^2 E_1^{OSS} + 2a_{01}a_{11}\Delta_{01}^{SA} \tag{1b}$$

where the SA-REKS energies and orbitals are obtained from minimizing the functional

$$\begin{aligned} E^{SA-REKS} &= w_{PPS}E_0^{PPS} + w_{OSS}E_1^{OSS} \\ w_{PPS} &+ w_{OSS} = 1 \end{aligned} \tag{2}$$

with respect to the orbitals and the occupation numbers in the PPS state, and the coefficients a_{kl} are obtained from solving a 2×2 secular equation

$$\begin{pmatrix} E_0^{PPS} & \Delta_{01}^{SA} \\ \Delta_{01}^{SA} & E_1^{OSS} \end{pmatrix} \begin{pmatrix} a_{00} & a_{01} \\ a_{10} & a_{11} \end{pmatrix} = \begin{pmatrix} E_0^{SSR} & 0 \\ 0 & E_1^{SSR} \end{pmatrix} \begin{pmatrix} a_{00} & a_{01} \\ a_{10} & a_{11} \end{pmatrix} \tag{3}$$

with the interstate coupling element Δ_{01}^{SA} defined as

$$\Delta_{01}^{SA} = \left(\sqrt{n_r} - \sqrt{n_s}\right) W_{rs} \tag{4}$$

2 The SSR(2,2) gradient

The derivatives of the SSR(2,2) energies are

$$\frac{\partial E_0^{SSR}}{\partial \lambda} = a_{00}^2 \frac{\partial E_0^{PPS}}{\partial \lambda} + a_{10}^2 \frac{\partial E_1^{OSS}}{\partial \lambda} + 2a_{00}a_{10}\frac{\partial \Delta_{01}^{SA}}{\partial \lambda} \tag{5a}$$
$$\frac{\partial E_1^{SSR}}{\partial \lambda} = a_{01}^2 \frac{\partial E_0^{PPS}}{\partial \lambda} + a_{11}^2 \frac{\partial E_1^{OSS}}{\partial \lambda} + 2a_{01}a_{11}\frac{\partial \Delta_{01}^{SA}}{\partial \lambda} \tag{5b}$$

The derivatives of the individual energies E_Y^X ($\{X, Y\}$ = {PPS,0}, {OSS,1}) are obtained as

$$\begin{aligned} \frac{\partial E_Y^X}{\partial \lambda} = \sum_{L=L_{min}}^{L_{max}} C_L^X \frac{\partial' E_L}{\partial \lambda} - \frac{1}{2}\sum_{i,j}^{all}\left({}^i\epsilon_{ij}^X + {}^j\epsilon_{ij}^X\right) S_{ji}^{\lambda} \\ + \sum_{i,j}^{all}\left({}^i\epsilon_{ij}^X - {}^j\epsilon_{ij}^X\right) U_{ji}^{\lambda} \end{aligned} \tag{6}$$

where the λ superscript denotes derivative with respect to the external parameter λ and prime at the differentiation symbol means that only the molecular integrals are to be differentiated. The Lagrange multipliers ${}^{i}\epsilon_{pq}^{X}$ are

$$ {}^{i}\epsilon_{pq}^{X} = \sum_{L=L_{min}}^{L_{max}} C_L^X \sum_{\sigma} n_{i,L}^{\sigma} \langle \varphi_p | \hat{F}_L^{\sigma} | \varphi_q \rangle = \langle \varphi_p | n_i \hat{F}_i | \varphi_q \rangle \tag{7} $$

and the orbital derivatives are defined as

$$ \frac{\partial \mathbf{C}}{\partial \lambda} = \mathbf{C}\, \mathbf{U}^{\lambda} \tag{8} $$

where $\mathbf{C}$ are the coefficients used in the MO expansion $\boldsymbol{\varphi} = \boldsymbol{\chi}\, \mathbf{C}$.

3 The CP-REKS equations

The orbital derivatives are obtained from the equation

$$ \mathbf{A}\, \mathbf{U}^{\lambda} = \mathbf{B}^{\lambda} \tag{9} $$

where the matrix $\mathbf{B}^{\lambda}$ on the right hand side of the equation collects all terms pertaining to the specific perturbation λ. The elements of the orbital rotation Hessian matrix $\mathbf{A}$ are independent of the perturbation and are given by

$$ \begin{aligned} A_{ij,pq} = {} & \frac{\delta_{ip}}{2} \left({}^{i}\epsilon_{jq}^{SA} - {}^{j}\epsilon_{jq}^{SA} \right) - \frac{\delta_{jq}}{2} \left({}^{i}\epsilon_{ip}^{SA} - {}^{j}\epsilon_{ip}^{SA} \right) - \frac{\delta_{iq}}{2} \left({}^{i}\epsilon_{jp}^{SA} - {}^{j}\epsilon_{jp}^{SA} \right) + \frac{\delta_{jp}}{2} \left({}^{i}\epsilon_{iq}^{SA} - {}^{j}\epsilon_{iq}^{SA} \right) \\ & + (ij|\, \tilde{f}_{Hxc}^{(i-j)(p-q)}\, |qp) - w_{PPS}\, G^{(1)}\, \Omega_{ij}\, \Omega_{pq} \end{aligned} \tag{10} $$

where a convention $p < q$ ($i < j$) is used and the elements of the symmetrized Hxc kernel are given by

$$ (ij|\, \tilde{f}_{Hxc}^{(i-j)(p-q)}\, |qp) = \sum_{L=L_{min}}^{L_{max}} C_L^{SA} \sum_{\sigma,\sigma'} (n_{i,L}^{\sigma} - n_{j,L}^{\sigma})\, (n_{p,L}^{\sigma'} - n_{q,L}^{\sigma'})\, (ij|\tilde{f}_{Hxc,L}^{\sigma\sigma'}|qp) \tag{11} $$

The symmetrized Hxc kernel $\tilde{f}_{Hxc,L}^{\sigma\sigma'}$ is defined as

$$ (pq|\tilde{f}_{Hxc,L}^{\sigma\sigma'}|ij) = \frac{1}{2} \left((pq|f_{Hxc,L}^{\sigma\sigma'}|ij) + (pq|f_{Hxc,L}^{\sigma\sigma'}|ji) \right) \tag{12} $$

where $f_{Hxc,L}^{\sigma\sigma'}$ is the usual Hxc kernel

$$ (pq|f_{Hxc,L}^{\sigma\sigma'}|ij) = (pq|ij) - \delta_{\sigma\sigma'} (pj| \frac{g_{HF}}{r_{12}} |iq) + (pq|g_{DFT} f_{x,L}^{\sigma\sigma'}|ij) + (pq|f_{c,L}^{\sigma\sigma'}|ij) \tag{13} $$

In eq. (13), g_{HF} and g_{DFT} are the parameters (or functions) defining the splitting between the exact exchange and the DFT exchange energy, respectively.

In Eq. (10), the antisymmetric matrices $\mathbf{\Omega}$ represent coupling between the FONs (in the PPS state) and orbital variations and are given by

$$\Omega_{pq} = \sum_{L=1}^{6} I_L \sum_{\sigma} (n_{p,L}^{\sigma} - n_{q,L}^{\sigma}) \epsilon_{L,pq}^{\sigma} \tag{14}$$

where the coefficients I_L are

$$\begin{aligned} I_1 &= -I_2 = 1 \\ I_3 &= I_4 = -I_5 = -I_6 = \frac{(E_1 - E_2)}{E_5 + E_6 - E_3 - E_4} \end{aligned} \tag{15}$$

$G^{(1)}$ is the energy weighted derivative of the interpolating function $f(x)$

$$G^{(1)} = 2 \left(\frac{\partial^2 f(x)}{\partial x^2} \right)^{-1} \frac{1}{E_5 + E_6 - E_3 - E_4} \tag{16}$$

where E_L are the energies of the respective microstates and $x = n_r/2$.

The $\mathbf{B}^{\lambda}$ vector on the right hand side of eq. (9) is the part of the orbital gradient independent of the orbital displacement given by

$$\begin{aligned} B_{pq}^{\lambda} = &\frac{1}{2} \sum_{L} C_L^{SA} \sum_{\sigma} (n_{p,L}^{\sigma} - n_{q,L}^{\sigma}) \frac{\partial' \langle p | \hat{F}_L^{\sigma} | q \rangle}{\partial \lambda} - \frac{1}{4} \sum_{t}^{all} \left({}^{p}\epsilon_{tq}^{SA} - {}^{q}\epsilon_{tq}^{SA} \right) S_{tp}^{\lambda} - \frac{1}{4} \sum_{t}^{all} \left({}^{p}\epsilon_{pt}^{SA} - {}^{q}\epsilon_{pt}^{SA} \right) S_{tq}^{\lambda} \\ &- \frac{1}{2} \sum_{u,v}^{all} \left((pq | \tilde{f}_{Hxc}^{pu} | uv) - (pq | \tilde{f}_{Hxc}^{qu} | uv) \right) S_{vu}^{\lambda} \\ &- \frac{1}{2} w_{PPS} G^{(1)} \Omega_{pq} \sum_{L} I_L \left(\frac{\partial' E_L}{\partial \lambda} + \frac{1}{2} \sum_{i,j}^{all} S_{ji}^{\lambda} \left({}^{i}\epsilon_{ij}^{L} + {}^{j}\epsilon_{ij}^{L} \right) \right) \end{aligned} \tag{17}$$

where the multipliers ${}^{i}\epsilon_{ij}^{L}$ are

$${}^{q}\epsilon_{ij}^{L} = \sum_{\sigma} n_{q,L}^{\sigma} \langle \varphi_i | \hat{F}_L^{\sigma} | \varphi_j \rangle \,. \tag{18}$$

As the orbital gradient term in Eq. (9) is always of the form $\mathbf{X}_X^{\mathsf{T}} \cdot \mathbf{U}^{\lambda}$ (T stands for matrix transposition), where $\mathbf{X}_X^{\mathsf{T}}$ ($X =$ REKS or OSS) is

$$\left(\mathbf{X}_X^{\mathsf{T}} \right)_{ij} = {}^{i}\epsilon_{ij}^{X} - {}^{j}\epsilon_{ij}^{X} \,, \tag{19}$$

a replacement

$$\mathbf{Z}^{\mathsf{T}} \, \mathbf{A} = \mathbf{X}^{\mathsf{T}} \tag{20}$$

can be introduced to simplify Eq. (9) to

$$\frac{\partial E_Y^X}{\partial \lambda} = \sum_{L=L_{min}}^{L_{max}} C_L^X \frac{\partial' E_L}{\partial \lambda} - \frac{1}{2} \sum_{i,j}^{all} \left({}^{i}\epsilon_{ij}^{X} + {}^{j}\epsilon_{ij}^{X} \right) S_{ji}^{\lambda} - 2 \sum_{i<j}^{all} Z_{ij}^{X} B_{ij}^{\lambda} \tag{21}$$

4 Solving the CP-REKS equations

The Z-vector equation (20) is typically solved by the conjugate gradient (CG) method. The most time consuming part of eq. (20) is the computation of the inner product of the Hessian $\mathbf{A}$ with a trial vector $\mathbf{Y}$; the latter vector converges to $\mathbf{Z}^{\intercal}$. In terms of the basis functions, the latter product becomes

$$\sum_{i<j} Y_{ij}\, A^{2e}_{ij,pq} = \sum_{L} C^{SA}_{L} \sum_{\sigma'} \sum_{\gamma,\tau} \mathcal{Z}^{\sigma'}_{L,\gamma\tau}\, (n^{\sigma'}_{p,L} - n^{\sigma'}_{q,L}) C^{\dagger}_{q\gamma} C_{\tau p} \tag{22}$$

where

$$\mathcal{Z}^{\sigma'}_{L,\gamma\tau} = \sum_{\sigma} \sum_{\mu,\nu} R^{\sigma}_{L,\mu\nu}\, (\mu\nu|\tilde{f}^{\sigma\sigma'}_{Hxc,L}|\gamma\tau) \tag{23}$$

and

$$R^{\sigma}_{L,\nu\mu} = \sum_{i<j} Y_{ij}\, (n^{\sigma}_{i,L} - n^{\sigma}_{j,L}) C^{\dagger}_{i\mu} C_{\nu j} \tag{24}$$

5 PPS and OSS relaxed density matrix

When the perturbation λ is an external electric field $\mathbf{F}$, then the derivative of the PPS or OSS energy becomes the negative of its dipole moment $\boldsymbol{\mu}$

$$\frac{\partial E^{X}_{Y}}{\partial \mathbf{F}} = \sum_{L=1}^{6} C_L \sum_{\mu,\nu} P^{(L)}_{\nu\mu} \mathbf{d}_{\mu\nu} - 2 \sum_{i<j} \left({}^{i}\epsilon^{X}_{ij} - {}^{j}\epsilon^{X}_{ij} \right) U^{\mathbf{F}}_{ij} \tag{25a}$$

$$= \sum_{\mu,\nu} P^{(o)}_{\nu\mu} \mathbf{d}_{\mu\nu} - 2\ \mathrm{tr}\, \mathbf{Z}^{\intercal}\, \mathbf{B}^{\mathbf{F}} \tag{25b}$$

$$= \sum_{\mu,\nu} \left(P^{(o)}_{\nu\mu} + P^{(r)}_{\nu\mu} \right) \mathbf{d}_{\mu\nu} \tag{25c}$$

$$= -\boldsymbol{\mu} \tag{25d}$$

where the orbital part $\mathbf{P}^{(o)}$ of the total PPS or OSS density matrix is obtained using the SA-REKS orbital coefficients and the response part $\mathbf{P}^{(r)}$ is

$$P^{(r)}_{\nu\mu} = -2 \left(\sum_{p<q} Z_{pq} (f_p - f_q) C_{\nu q} C^{\dagger}_{p\mu} - w_{PPS} G^{(1)} (\sum_{p<q} Z_{pq} \Omega_{pq}) (C_{\nu r} C^{\dagger}_{r\mu} - C_{\nu s} C^{\dagger}_{s\mu}) \right) \tag{26}$$

In Eq. (25c), $\mathbf{d}_{\mu\nu}$ are the dipole moment integrals

$$\mathbf{d}_{\mu\nu} = \frac{\partial h_{\mu\nu}}{\partial \mathbf{F}} = \bar{e} \langle \chi_\mu | \mathbf{r} | \chi_\nu \rangle \tag{27}$$

where $\mathbf{r}$ is the electron coordinate vector and $\bar{e} = -1$ is the electron charge.

6 PPS and OSS gradient

When λ is, *e.g.*, a nuclear coordinate, Eq. (9) becomes

$$\frac{\partial E_Y^X}{\partial \lambda} = \sum_L \Big(C_L^X + w_{PPS} G^{(1)} I_L \sum_{p<q} Z_{pq}^X \Omega_{pq} \Big) \Big(\frac{\partial' E_L}{\partial \lambda} - \frac{1}{2} \sum_{i,j}^{all} ({}^{i}\epsilon_{ij}^{L} + {}^{j}\epsilon_{ij}^{L}) S_{ji}^{\lambda} \Big) \tag{28}$$

$$- \sum_L C_L^{SA} \sum_\sigma \mathrm{tr}\, {}^X\mathbf{R}_L^\sigma \mathbf{T}_L^{\sigma,\lambda} + \mathrm{tr} \left({}^X\mathbf{Q}^{(1)} + {}^X\mathbf{Q}^{(2)} \right) \mathbf{S}^\lambda$$

where ${}^X\mathbf{R}_L^\sigma$ is

$${}^X R_{L,\nu\mu}^{\sigma} = \sum_{i<j} Z_{ji}^X \, (n_{i,L}^\sigma - n_{j,L}^\sigma) C_{i\mu}^\dagger C_{\nu j} \, , \tag{29}$$

the matrix $\mathbf{T}_L^{\sigma,\lambda}$ is the derivative $\partial' \langle p | \hat{F}_L^\sigma | q \rangle / \partial \lambda$ transformed to the atomic orbital basis set representation

$$T_{L,\mu\nu}^{\sigma,\lambda} = h_{\mu\nu}^{\lambda} + \sum_{\kappa,\tau} \sum_{\sigma'} P_{L,\tau\kappa}^{\sigma'} \big((\mu\nu|\kappa\tau)^\lambda - \delta_{\sigma\sigma'} (\mu\tau | \frac{g_{HF}}{r_{12}} | \kappa\nu)^\lambda \big)$$

$$+ \frac{\partial' (\mu\nu | g_{DFT} \, V_{xc,L}^\sigma)}{\partial \lambda} \, , \tag{30}$$

(the λ superscript denotes differentiation), $V_{xc,L}^\sigma(\mathbf{r}) = \delta E_{xc,L} / \delta \rho_L^\sigma(\mathbf{r})$ is the XC potential of the L-th microstate, $P_{L,\nu\mu}^\sigma = \sum_i^{occ} n_{i,L}^\sigma C_{\nu i} C_{i\mu}^\dagger$ are the elements of the density matrix of the L-th microstate, and the matrices ${}^X\mathbf{Q}^{(1)}$ and ${}^X\mathbf{Q}^{(2)}$ are

$${}^X Q_{pt}^{(1)} = \frac{1}{2} \sum_{q(>p)}^{all} Z_{pq}^X \left({}^{p}\epsilon_{qt}^{SA} - {}^{q}\epsilon_{qt}^{SA} \right)$$

$$- \frac{1}{2} \sum_{q(<p)}^{all} Z_{qp}^X \left({}^{p}\epsilon_{qt}^{SA} - {}^{q}\epsilon_{qt}^{SA} \right) \tag{31}$$

$${}^X Q_{vu}^{(2)} = \frac{1}{2} \sum_{p<q}^{all} Z_{pq}^X (pq | \tilde{f}_{Hxc}^{(p-q)(u+v)} | uv) \, . \tag{32}$$

In terms of the basis functions, Eq. (32) becomes

$${}^X Q_{vu}^{(2)} = \frac{1}{2} \sum_L C_L^{SA} \sum_{\sigma'} \sum_{\gamma,\tau} {}^X \mathcal{Z}_{\gamma\tau}^{\sigma',L} (n_{u,L}^{\sigma'} + n_{v,L}^{\sigma'}) C_{u\gamma}^\dagger C_{\tau v} \tag{33}$$

where

$${}^X \mathcal{Z}_{\gamma\tau}^{\sigma',L} = \sum_\sigma \sum_{\mu,\nu} {}^X R_{L,\nu\mu}^{\sigma} (\mu\nu | \tilde{f}_{Hxc,L}^{\sigma\sigma'} | \gamma\tau) \, . \tag{34}$$

7 Derivatives of the SSR(2,2) energies

For the SSR method, the derivatives take a somewhat more complicated form. A new term, $\frac{\partial \Delta_{01}^{SA}}{\partial \lambda}$, occurs in the expression for the SSR gradient Eq. (5). This term is given by

$$\frac{\partial \Delta_{01}^{SA}}{\partial \lambda} = -G^{(1)} R_{sr} \sum_L I_L \Big(\frac{\partial' E_L}{\partial \lambda} - \frac{1}{2} \sum_{i,j}^{all} \big({}^{i}\epsilon_{ij}^{L} + {}^{j}\epsilon_{ij}^{L} \big) S_{ji}^{\lambda} \Big) + \sum_L C_L^{SA} \sum_{\sigma=\alpha,\beta} \operatorname{tr} {}^{\Delta}\mathbf{R}_L^{\sigma} \mathbf{T}_L^{\sigma,\lambda} \\ - \operatorname{tr} \left({}^{\Delta}\mathbf{Q}^{(1)} + {}^{\Delta}\mathbf{Q}^{(2)} \right) \mathbf{S}^{\lambda} + 2 \operatorname{tr} \mathbf{X}_{\Delta}^{\intercal} \mathbf{U}^{\lambda} \tag{35}$$

where

$${}^{\Delta}R_{L,\nu\mu}^{\sigma} = \left(\sqrt{n_r} n_{r,L}^{\sigma} - \sqrt{n_s} n_{s,L}^{\sigma} \right) C_{\nu s} C_{r\mu}^{\dagger} \tag{36}$$

$$R_{sr} = \left(\frac{1}{\sqrt{n_r}} + \frac{1}{\sqrt{n_s}} \right) {}^{r}\epsilon_{sr}^{SA} + w_{PPS} \sum_L I_L \sum_{\sigma} \left(\sqrt{n_r} n_{r,L}^{\sigma} \epsilon_{L,sr}^{\sigma} - \sqrt{n_s} n_{s,L}^{\sigma} \epsilon_{L,rs}^{\sigma} \right) , \tag{37}$$

the elements of the vector $\mathbf{X}_{\Delta}^{\intercal}$ (with $p < q$) are

$$\begin{aligned} \left(\mathbf{X}_{\Delta}^{\intercal} \right)_{pq} = & \frac{\delta_{qs}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{pr}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{pr}^{SA} \right) + \frac{\delta_{qr}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{ps}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{ps}^{SA} \right) \\ & - \frac{\delta_{ps}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{qr}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{qr}^{SA} \right) - \frac{\delta_{pr}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{qs}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{qs}^{SA} \right) \\ & + G^{(1)} R_{sr} \Omega_{pq} - \sqrt{n_r} (rs | \tilde{f}_{Hxc}^{r(p-q)} | pq) + \sqrt{n_s} (rs | \tilde{f}_{Hxc}^{s(p-q)} | pq) , \end{aligned} \tag{38}$$

and the matrices ${}^{\Delta}\mathbf{Q}^{(1)}$ and ${}^{\Delta}\mathbf{Q}^{(2)}$ are given by

$$\begin{aligned} {}^{\Delta}Q_{pq}^{(1)} = & \frac{\delta_{qs}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{pr}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{pr}^{SA} \right) + \frac{\delta_{qr}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{ps}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{ps}^{SA} \right) \\ & + \frac{\delta_{ps}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{qr}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{qr}^{SA} \right) + \frac{\delta_{pr}}{2} \left(\sqrt{n_r}\, {}^{r}\epsilon_{qs}^{SA} - \sqrt{n_s}\, {}^{s}\epsilon_{qs}^{SA} \right) \end{aligned} \tag{39}$$

$${}^{\Delta}Q_{pq}^{(2)} = \sqrt{n_r} (sr | \tilde{f}_{Hxc}^{r(p+q)} | pq) - \sqrt{n_s} (rs | \tilde{f}_{Hxc}^{s(p+q)} | pq) \tag{40}$$

Collecting all the terms in Eq. (5) yields

$$\frac{\partial E_m^{SSR}}{\partial \lambda} = \sum_L \left(\left(C_L^{SSR_m} + w_{PPS} G^{(1)} I_L \sum_{p<q} Z_{pq}^{SSR_m} \Omega_{pq} \right) \Big(\frac{\partial' E_L}{\partial \lambda} - \frac{1}{2} \sum_{i,j}^{all} \big({}^{i}\epsilon_{ij}^{L} + {}^{j}\epsilon_{ij}^{L} \big) S_{ji}^{\lambda} \Big) \right) \\ - \sum_L C_L^{SA} \sum_{\sigma} \operatorname{tr} \left({}^{SSR_m}\mathbf{R}_L^{\sigma} \mathbf{T}_L^{\sigma,\lambda} \right) + \operatorname{tr} \left({}^{SSR_m}\mathbf{Q}^{(1)} + {}^{SSR_m}\mathbf{Q}^{(2)} \right) \mathbf{S}^{\lambda} , \tag{41}$$

where the $\mathbf{Z}^{SSR_m}$ vector is obtained from the Z-vector equation (20) with the right hand side replaced by

$$\mathbf{X}_{SSR_m}^{\intercal} = a_{0m}^2 \mathbf{X}_{PPS}^{\intercal} + a_{1m}^2 \mathbf{X}_{OSS}^{\intercal} - 2 a_{0m} a_{1m} \mathbf{X}_{\Delta}^{\intercal} , \tag{42}$$

the $C_L^{SSR_m}$ weighting factors are

$$C_L^{SSR_m} = a_{0m}^2 C_L^{PPS} + a_{1m}^2 C_L^{OSS} - 2 a_{0m} a_{1m} G^{(1)} R_{sr} I_L \tag{43}$$

and the matrices ${}^{SSR_m}\mathbf{R}_L^{\sigma}$, ${}^{SSR_m}\mathbf{Q}^{(1)}$, and ${}^{SSR_m}\mathbf{Q}^{(2)}$ are given by

$$ {}^{SSR_m}\mathbf{R}_L^{\sigma} = {}^{(m)}\mathbf{R}_L^{\sigma} - 2a_{0m}a_{1m}{}^{\Delta}\mathbf{R}_L^{\sigma} \tag{44} $$

$$ {}^{SSR_m}\mathbf{Q}^{(1)} = {}^{(m)}\mathbf{Q}^{(1)} - 2a_{0m}a_{1m}{}^{\Delta}\mathbf{Q}^{(1)} \tag{45} $$

$$ {}^{SSR_m}\mathbf{Q}^{(2)} = {}^{(m)}\mathbf{Q}^{(2)} - 2a_{0m}a_{1m}{}^{\Delta}\mathbf{Q}^{(2)} \tag{46} $$

where ${}^{(m)}\mathbf{R}_L^{\sigma}$, ${}^{(m)}\mathbf{Q}^{(1)}$, and ${}^{(m)}\mathbf{Q}^{(2)}$ are obtained from eqs. (29), (31), and (32), respectively, by replacing $\mathbf{Z}^X$ with $\mathbf{Z}^{SSR_m}$.

8 SSR(2,2) relaxed density matrix

When the external perturbation λ is an electric field $\mathbf{F}$, Eq. (41) becomes

$$ \frac{\partial E_m^{SSR}}{\partial \mathbf{F}} = \sum_{\mu,\nu} \left({}^{SSR_m}P_{\nu\mu}^{(o)} + 2a_{0m}a_{1m}{}^{\Delta}P_{\nu\mu}^{(n)} \right) \mathbf{d}_{\mu\nu} - 2\,\mathrm{tr}(\mathbf{Z}^{SSR_m})^{\intercal}\mathbf{B}^{\mathbf{F}}\,, \tag{47} $$

where

$$ {}^{SSR_m}P_{\nu\mu}^{(o)} = a_{0m}^2\,{}^{PPS}P_{\nu\mu}^{(o)} + a_{1m}^2\,{}^{OSS}P_{\nu\mu}^{(o)} + 2a_{0m}a_{1m}{}^{\Delta}P_{\nu\mu}^{(o)} \tag{48} $$

$$ {}^{X}P_{\nu\mu}^{(o)} = \sum_{L=1}^{L_{max}} C_L^X \sum_q^{occ} n_{q,L}^{\sigma} C_{\nu q} C_{q\mu}^{\dagger}\,; \quad X = PPS,\, OSS \tag{49} $$

$$ {}^{\Delta}P_{\nu\mu}^{(o)} = \frac{1}{2}\left(C_{\nu s}C_{\mu r} + C_{\nu r}C_{\mu s}\right)\left(\sqrt{n_r} - \sqrt{n_s}\right) \tag{50} $$

$$ {}^{\Delta}P_{\nu\mu}^{(n)} = \frac{1}{2}w_{PPS}\left((n_r-1)\sqrt{n_r} - (n_s-1)\sqrt{n_s}\right)\left(C_{\nu s}C_{\mu r} + C_{\nu r}C_{\mu s}\right) - 2G^{(1)}R_{sr}\left(C_{\nu r}C_{\mu r} - C_{\nu s}C_{\mu s}\right)\,, \tag{51} $$

and the last term is

$$ \mathrm{tr}(\mathbf{Z}^{SSR_m})^{\intercal}\mathbf{B}^{\mathbf{F}} = \sum_{\mu,\nu}\mathbf{d}_{\mu\nu}\left(\sum_{p<q} Z_{pq}^{SSR_m}(f_p - f_q)C_{\nu q}C_{p\mu}^{\dagger} - w_{PPS}G^{(1)}\left(\sum_{p<q} Z_{pq}^{SSR_m}\Omega_{pq}\right)\left(C_{\nu r}C_{r\mu}^{\dagger} - C_{\nu s}C_{s\mu}^{\dagger}\right)\right) \tag{52} $$

Hence, the full relaxed density matrix is

$$ {}^{X}P_{\nu\mu}^{r} = {}^{SSR_m}P_{\nu\mu}^{(o)} + 2a_{0m}a_{1m}{}^{\Delta}P_{\nu\mu}^{(n)} - 2\left(\sum_{p<q} Z_{pq}^{SSR_m}(f_p - f_q)C_{\nu q}C_{p\mu}^{\dagger} - w_{PPS}G^{(1)}\left(\sum_{p<q} Z_{pq}^{SSR_m}\Omega_{pq}\right)\left(C_{\nu r}C_{r\mu}^{\dagger} - C_{\nu s}C_{s\mu}^{\dagger}\right)\right) \tag{53} $$

9 Alternative expression for SA/SSR relaxed density matrix

From the 1e part of the SA/SSR gradient, Eqs. (28) and (41), it follows that the relaxed density matrix can be written as

$$ {}^{X}P^{r}_{\nu\mu} = \sum_{L}\sum_{\sigma}\left(\tilde{C}^{X}_{L}\, P^{\sigma}_{L,\nu\mu} - C^{SA}_{L}\,{}^{X}R^{\sigma}_{L,\mu\nu}\right) , \tag{54} $$

where

$$ \tilde{C}^{X}_{L} = C^{X}_{L} + w_{PPS} G^{(1)} I_{L} \sum_{p<q} Z^{X}_{pq}\Omega_{pq} \tag{55} $$

and $X = PPS, OSS, SSR_m$, $m = 0, 1$ as defined in the previous equations.

In the MO representation, after some algebra, and using definition of the occupation numbers f^{X}_{p}

$$ f^{X}_{p} = \frac{1}{2}\sum_{L}\sum_{\sigma} C^{X}_{L} n^{\sigma}_{p,L} \tag{56} $$

one obtains for $p < q$

$$ {}^{X}P^{r}_{qp} = \delta_{qp}\sum_{L}\tilde{C}^{X}_{L}\,(n^{\alpha}_{p,L} + n^{\beta}_{p,L}) - 2\,Z^{X}_{qp}\,(f^{SA}_{p} - f^{SA}_{q}) \tag{57a} $$

$$ - 4\,\delta_{qs}\,\delta_{pr}\,a_{0m}a_{1m}(\sqrt{n_r}f^{SA}_{r} - \sqrt{n_s}f^{SA}_{s}) , \tag{57b} $$

where the term in the second line applies only to $X = SSR_m$, $m = 0, 1$ and not to $X = PPS$, OSS. After computing P_{qp}, it needs to be symmetrized, *i.e.*,

$$ {}^{X}P^{r}_{pq} = \frac{1}{2}\left({}^{X}P^{r}_{qp} + {}^{X}P^{r}_{pq}\right) . \tag{58} $$

The relaxed density matrix obtained in this way is fully equivalent to the one defined previously, as was checked by implementing both expressions in the program and comparing the results.

10 Alternative expression for SA/SSR gradient

Using the new expression for the relaxed density matrix Eq. (57) the analytic gradient expressions Eqs. (28) and (41) can be written as

$$ \frac{\partial E^{X}}{\partial\lambda} = \mathrm{tr}\,{}^{X}\mathbf{P}^{r}\,\mathbf{h}^{\lambda} - \frac{1}{2}\,\mathrm{tr}\,\tilde{\mathbf{W}}^{X}\,\mathbf{S}^{\lambda} \tag{59a} $$

$$ + \sum_{L}\tilde{C}^{X}_{L}\,\frac{\partial' E^{2e}_{L}}{\partial\lambda} - \sum_{L} C^{SA}_{L}\sum_{\sigma}\mathrm{tr}\,{}^{X}\mathbf{R}^{\sigma}_{L}\mathbf{T}^{\sigma,\lambda(2e)}_{L} , \tag{59b} $$

where $X = PPS, OSS, SSR_m$, $m = 0, 1$, ${}^X\mathbf{P}^r$ is defined in Eqs. (57) and (58), $\tilde{\mathbf{W}}$ is the new Lagrangian matrix

$$\tilde{W}^X_{pq} = \sum_L \tilde{C}^X_L \left({}^p\epsilon^L_{pq} + {}^q\epsilon^L_{pq}\right) - 2\left({}^XQ^{(1)}_{pq} + {}^XQ^{(2)}_{pq}\right) \tag{60}$$

and E^{2e}_L and $\mathbf{T}^{\sigma,\lambda(2e)}_L$ are the two-electron parts of the respective quantities in Eqs. (28) and (41)

11 The SSR(3,2) method

The two lowest SSR(3,2) energies are

$$E^{SSR(3,2)}_0 = a^2_{00}E^{PPS}_0 + a^2_{10}E^{OSS}_1 + a^2_{20}E^{DES}_2 + 2a_{00}a_{10}\Delta^{SA}_{01} + 2a_{10}a_{20}\Delta^{SA}_{12} \tag{61a}$$

$$E^{SSR(3,2)}_1 = a^2_{01}E^{PPS}_0 + a^2_{11}E^{OSS}_1 + a^2_{21}E^{DES}_2 + 2a_{01}a_{11}\Delta^{SA}_{01} + 2a_{11}a_{21}\Delta^{SA}_{12} \tag{61b}$$

where the SA-REKS energies and orbitals are obtained from minimizing the functional in Eq. (2). The coefficients a_{kl} are obtained from solving a 3×3 secular equation

$$\begin{pmatrix} E^{PPS}_0 & \Delta^{SA}_{01} & 0 \\ \Delta^{SA}_{01} & E^{OSS}_1 & \Delta^{SA}_{12} \\ 0 & \Delta^{SA}_{12} & E^{DES}_2 \end{pmatrix}\begin{pmatrix} a_{00} & a_{01} & a_{02} \\ a_{10} & a_{11} & a_{12} \\ a_{20} & a_{21} & a_{22} \end{pmatrix} = \begin{pmatrix} E_0 & 0 & 0 \\ 0 & E_1 & 0 \\ 0 & 0 & E_2 \end{pmatrix}\begin{pmatrix} a_{00} & a_{01} & a_{02} \\ a_{10} & a_{11} & a_{12} \\ a_{20} & a_{21} & a_{22} \end{pmatrix} \tag{62}$$

where the interstate coupling element Δ^{SA}_{01} is defined in Eq. (4) and the new coupling element Δ^{SA}_{12} is

$$\Delta^{SA}_{12} = \left(\sqrt{n_r} + \sqrt{n_s}\right) W_{rs} \tag{63}$$

12 Derivatives of the SSR(3,2) energies

The derivatives of the SSR(3,2) energies in Eq. (61) are

$$\frac{\partial E^{SSR(3,2)}_k}{\partial\lambda} = a^2_{0k}\frac{\partial E^{PPS}_0}{\partial\lambda} + a^2_{1k}\frac{\partial E^{OSS}_1}{\partial\lambda} + a^2_{2k}\frac{\partial E^{DES}_2}{\partial\lambda} + 2a_{0k}a_{1k}\frac{\partial\Delta^{SA}_{01}}{\partial\lambda} + 2a_{1k}a_{2k}\frac{\partial\Delta^{SA}_{12}}{\partial\lambda} \tag{64}$$
$$k = 1, 2$$

Compared to the SSR derivatives in Eq. (5) two new contributions occur in the SSR(3,2) derivatives, *i.e.*, the derivative of the E^{DES}_2 energy and the derivative of the Δ^{SA}_{12} coupling element. As the formulae for the new derivatives can be easily obtained from $\partial E^{PPS}_0/\partial\lambda$ and $\partial\Delta^{SA}_{12}/\partial\lambda$, the new derivatives become

$$\frac{\partial E^{SSR(3,2)}_k}{\partial\lambda} = \sum_L \left(\left(C^{SSR(3,2)_k}_L + w_{GSS}G^{(1)}I_L\sum_{p<q} Z^{SSR(3,2)_k}_{pq}\Omega_{pq}\right)\left(\frac{\partial' E_L}{\partial\lambda} - \frac{1}{2}\sum^{all}_{i,j}\left({}^i\epsilon^L_{ij} + {}^j\epsilon^L_{ij}\right)S^\lambda_{ji}\right)\right)$$

$$- \sum_L C_L^{SA} \sum_{\sigma=\alpha,\beta} \mathrm{tr}\left({}^{(k)}\mathbf{R}_L^{\sigma} \mathbf{T}_L^{\sigma,\lambda} \right) + \mathrm{tr}\left({}^{(k)}\mathbf{Q}^{(1)} + {}^{(k)}\mathbf{Q}^{(2)} \right) \mathbf{S}^{\lambda} \tag{65}$$
$$+ 2a_{0k}a_{1k} \sum_L C_L^{SA} \sum_{\sigma=\alpha,\beta} \mathrm{tr}\left({}^{\Delta}\mathbf{R}_L^{\sigma} \mathbf{T}_L^{\sigma,\lambda} \right) - 2a_{0k}a_{1k}\, \mathrm{tr}\left({}^{\Delta}\mathbf{Q}^{(1)} + {}^{\Delta}\mathbf{Q}^{(2)} \right) \mathbf{S}^{\lambda}$$
$$+ 2a_{1k}a_{2k} \sum_L C_L^{SA} \sum_{\sigma=\alpha,\beta} \mathrm{tr}\left({}^{\Delta_{12}}\mathbf{R}_L^{\sigma} \mathbf{T}_L^{\sigma,\lambda} \right) - 2a_{1k}a_{2k}\, \mathrm{tr}\left({}^{\Delta_{12}}\mathbf{Q}^{(1)} + {}^{\Delta_{12}}\mathbf{Q}^{(2)} \right) \mathbf{S}^{\lambda}$$

where

$$C_L^{SSR(3,2)_k} = a_{0k}^2 C_L^{PPS} + a_{1k}^2 C_L^{OSS} + a_{2k}^2 C_L^{DES} - 2a_{0k}a_{1k} G^{(1)} R_{sr} I_L - 2a_{1k}a_{2k} G^{(1)} R_{sr}^{(12)} I_L \tag{66}$$
$$R_{sr}^{(12)} = \left(\frac{1}{\sqrt{n_r}} - \frac{1}{\sqrt{n_s}} \right) {}^{r}\epsilon_{sr}^{SA} + w_{PPS} \sum_L I_L \sum_{\sigma} \left(\sqrt{n_r} n_{r,L}^{\sigma} \epsilon_{L,sr}^{\sigma} + \sqrt{n_s} n_{s,L}^{\sigma} \epsilon_{L,rs}^{\sigma} \right) \tag{67}$$
$${}^{\Delta_{12}}R_{L,\nu\mu}^{\sigma} = \left(\sqrt{n_r} n_{r,L}^{\sigma} + \sqrt{n_s} n_{s,L}^{\sigma} \right) C_{\nu s} C_{r\mu}^{\dagger} \tag{68}$$
$$\begin{aligned} {}^{\Delta_{12}}Q_{pq}^{(1)} &= \frac{\delta_{qs}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{pr}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{pr}^{SA}) + \frac{\delta_{qr}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{ps}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{ps}^{SA}) \\ &+ \frac{\delta_{ps}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{qr}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{qr}^{SA}) + \frac{\delta_{pr}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{qs}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{qs}^{SA}) \end{aligned} \tag{69}$$
$${}^{\Delta_{12}}Q_{pq}^{(2)} = \sqrt{n_r} (sr|\tilde{f}_{Hxc}^{r(p+q)}|pq) + \sqrt{n_s} (rs|\tilde{f}_{Hxc}^{s(p+q)}|pq) \tag{70}$$

and the Z-vector $\mathbf{Z}^{SSR(3,2)_k}$ is obtained from solving CP-REKS Eq. (20) with the right hand side given by

$$\mathbf{X}_{SSR(3,2)_k}^{\intercal} = a_{0k}^2 \mathbf{X}_{PPS}^{\intercal} + a_{1k}^2 \mathbf{X}_{OSS}^{\intercal} + a_{2k}^2 \mathbf{X}_{DES}^{\intercal} - 2a_{0k}a_{1k} \mathbf{X}_{\Delta}^{\intercal} - 2a_{1k}a_{2k} \mathbf{X}_{\Delta_{12}}^{\intercal} \tag{71}$$

where

$$\begin{aligned} \left(\mathbf{X}_{\Delta_{12}}^{\intercal} \right)_{pq} &= \frac{\delta_{qs}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{pr}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{pr}^{SA}) + \frac{\delta_{qr}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{ps}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{ps}^{SA}) \\ &- \frac{\delta_{ps}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{qr}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{qr}^{SA}) - \frac{\delta_{pr}}{2} (\sqrt{n_r}\, {}^{r}\epsilon_{qs}^{SA} + \sqrt{n_s}\, {}^{s}\epsilon_{qs}^{SA}) \\ &+ G^{(1)} R_{sr}^{(12)} \Omega_{pq} - \sqrt{n_r} (rs|\tilde{f}_{Hxc}^{r(p-q)}|pq) - \sqrt{n_s} (rs|\tilde{f}_{Hxc}^{s(p-q)}|pq)\,. \end{aligned} \tag{72}$$

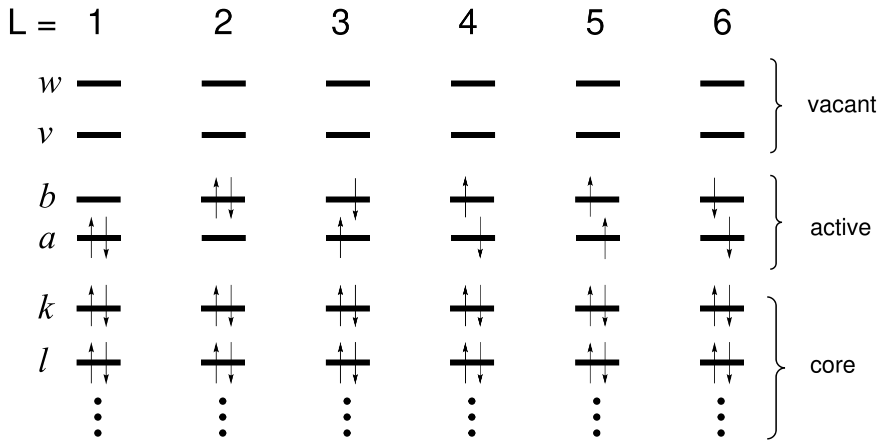
Finally, the SSR(3,2) gradient is given by Eqs. (59), (54), and (60), where

$$\tilde{C}_L^{SSR(3,2)_k} = C_L^{SSR(3,2)_k} + w_{PPS} G^{(1)} I_L \sum_{p<q} Z_{pq}^{SSR(3,2)_k} \Omega_{pq} \tag{73}$$
$${}^{SSR(3,2)_k}\mathbf{R}_L^{\sigma} = {}^{(k)}\mathbf{R}_L^{\sigma} - 2a_{0k}a_{1k}{}^{\Delta}\mathbf{R}_L^{\sigma} - 2a_{1k}a_{2k}{}^{\Delta_{12}}\mathbf{R}_L^{\sigma} \tag{74}$$
$${}^{SSR(3,2)_k}\mathbf{Q}^{(1)} = {}^{(k)}\mathbf{Q}^{(1)} - 2a_{0k}a_{1k}{}^{\Delta}\mathbf{Q}^{(1)} - 2a_{1k}a_{2k}{}^{\Delta_{12}}\mathbf{Q}^{(1)} \tag{75}$$
$${}^{SSR(3,2)_k}\mathbf{Q}^{(2)} = {}^{(k)}\mathbf{Q}^{(2)} - 2a_{0k}a_{1k}{}^{\Delta}\mathbf{Q}^{(2)} - 2a_{1k}a_{2k}{}^{\Delta_{12}}\mathbf{Q}^{(2)} \tag{76}$$

The other contributions in Eqs. (59) and (54) remain the same as for the SSR method.

Appendix A REKS microstates

The microstates used in the (SI-SA-)REKS(2,2) method are shown in Scheme 1 and the weighting factors are collected in Table 1.



Scheme 1: Microstates of the REKS(2,2) method.

Table 1: Weighting factors C_L of REKS microstates

state	L					
	1	2	3	4	5	6
PPS	$\frac{n_r}{2}$	$\frac{n_s}{2}$	$-\frac{1}{2}\,f(\frac{n_r}{2})$	$-\frac{1}{2}\,f(\frac{n_r}{2})$	$\frac{1}{2}\,f(\frac{n_r}{2})$	$\frac{1}{2}\,f(\frac{n_r}{2})$
OSS	–	–	1	1	$-\frac{1}{2}$	$-\frac{1}{2}$
DES	$\frac{n_s}{2}$	$\frac{n_r}{2}$	$\frac{1}{2}\,f(\frac{n_r}{2})$	$\frac{1}{2}\,f(\frac{n_r}{2})$	$-\frac{1}{2}\,f(\frac{n_r}{2})$	$-\frac{1}{2}\,f(\frac{n_r}{2})$
SA	$\frac{w_{PPS}\,n_r}{2}$	$\frac{w_{PPS}\,n_s}{2}$	$\frac{2\,w_{OSS}-w_{PPS}\,f(\frac{n_r}{2})}{2}$	$\frac{2\,w_{OSS}-w_{PPS}\,f(\frac{n_r}{2})}{2}$	$\frac{w_{PPS}\,f(\frac{n_r}{2})-w_{OSS}}{2}$	$\frac{w_{PPS}\,f(\frac{n_r}{2})-w_{OSS}}{2}$

In Table 1, function f is

$$f(x) = (4\,x\,(1-x))^{1-\frac{1}{2}\left(\frac{4\,x\,(1-x)+\delta}{1+\delta}\right)}\,; \qquad 1 \leq x \leq \frac{1}{2} \tag{77}$$

where $\delta = 0.4$.

The the fixed and integer occupation numbers $n^{\sigma}_{q,L}$ in the microstates are given in Table 2.

Table 2: Occupation numbers of orbitals $n^{\sigma}_{q,L}$ in REKS microstates

shell, p	σ	L					
		1	2	3	4	5	6
core, k	α	1	1	1	1	1	1
	β	1	1	1	1	1	1
active, a	α	1	0	1	0	1	0
	β	1	0	0	1	0	1
active, b	α	0	1	0	1	1	0
	β	0	1	1	0	0	1
vacant, v	α	0	0	0	0	0	0
	β	0	0	0	0	0	0

Appendix B RT term

$$-\sum_L C_L^{SA} \sum_\sigma \sum_{\mu,\nu} R^\sigma_{L,\nu\mu} T^{\sigma,\lambda(2e)}_{L,\mu\nu} \tag{78a}$$

$$= -\sum_L C_L^{SA} \sum_\sigma \sum_{\mu,\nu} R^\sigma_{L,\nu\mu} \left(\sum_{\kappa\tau} \sum_{\sigma'} P^{\sigma'}_{L,\tau\kappa} \left[(\mu\nu|\kappa\tau)^\lambda - \delta_{\sigma\sigma'} g_{HF} (\mu\kappa|\nu\tau)^\lambda \right] \right) \tag{78b}$$

$$= -\sum_{\mu,\nu,\sigma} \sum_{\kappa,\tau,\sigma'} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^\sigma_{L,\nu\mu} P^{\sigma'}_{L,\tau\kappa} \right) + \sum_{\mu,\nu,\sigma} \sum_{\kappa,\tau,\sigma'} (\mu\kappa|\nu\tau)^\lambda \left(\delta_{\sigma\sigma'} g_{HF} \sum_L C_L^{SA} R^\sigma_{L,\nu\mu} P^{\sigma'}_{L,\tau\kappa} \right) \tag{78c}$$

$$= -\sum_{\mu,\nu,\sigma} \sum_{\kappa,\tau,\sigma'} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^\sigma_{L,\nu\mu} P^{\sigma'}_{L,\tau\kappa} \right) + \sum_{\mu,\nu,\sigma} \sum_{\kappa,\tau,\sigma'} (\mu\nu|\kappa\tau)^\lambda \left(\delta_{\sigma\sigma'} g_{HF} \sum_L C_L^{SA} R^\sigma_{L,\kappa\mu} P^{\sigma'}_{L,\tau\nu} \right) \tag{78d}$$

Let $R^{s,\sigma}_{L,\nu\mu} = (R^\sigma_{L,\nu\mu} + R^\sigma_{L,\mu\nu})/2$ and $R^{a,\sigma}_{L,\nu\mu} = (R^\sigma_{L,\nu\mu} - R^\sigma_{L,\mu\nu})/2$. Then, $R^\sigma_{L,\nu\mu} = R^{s,\sigma}_{L,\nu\mu} + R^{a,\sigma}_{L,\nu\mu}$. The same for the P matrix; however, the P^a part vanishes, as the P matrix is symmetric. Further, let $R^{s,t}_{L,\nu\mu} = R^{s,\alpha}_{L,\nu\mu} + R^{s,\beta}_{L,\nu\mu}$ and $R^{s,m}_{L,\nu\mu} = R^{s,\alpha}_{L,\nu\mu} - R^{s,\beta}_{L,\nu\mu}$. Then, $R^{s,\alpha}_{L,\nu\mu} = (R^{s,t}_{L,\nu\mu} + R^{s,m}_{L,\nu\mu})/2$ and $R^{s,\beta}_{L,\nu\mu} = (R^{s,t}_{L,\nu\mu} - R^{s,m}_{L,\nu\mu})/2$. The same relations hold for the R^a matrices and for the P matrices. Then,

$$= -\sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^{s,t}_{L,\nu\mu} P^t_{L,\tau\kappa} \right) - \sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^{a,t}_{L,\nu\mu} P^t_{L,\tau\kappa} \right) \tag{78e}$$

$$+ \sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(g_{HF} \sum_L C_L^{SA} \left[R^{s,\alpha}_{L,\kappa\mu} P^\alpha_{L,\tau\nu} + R^{s,\beta}_{L,\kappa\mu} P^\beta_{L,\tau\nu} + R^{a,\alpha}_{L,\kappa\mu} P^\alpha_{L,\tau\nu} + R^{a,\beta}_{L,\kappa\mu} P^\beta_{L,\tau\nu} \right] \right) \tag{78f}$$

Substituting $R^{s,\alpha}_{L,\kappa\mu} = (R^{s,t}_{L,\kappa\mu} + R^{s,m}_{L,\kappa\mu})/2$, $R^{s,\beta}_{L,\kappa\mu} = (R^{s,t}_{L,\kappa\mu} - R^{s,m}_{L,\kappa\mu})/2$, $R^{a,\alpha}_{L,\kappa\mu} = (R^{a,t}_{L,\kappa\mu} + R^{a,m}_{L,\kappa\mu})/2$, and $R^{a,\beta}_{L,\kappa\mu} = (R^{a,t}_{L,\kappa\mu} - R^{a,m}_{L,\kappa\mu})/2$, and the expressions for the P^α and P^β matrices yields, after some manipulation

$$= -\sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^{s,t}_{L,\nu\mu} P^t_{L,\tau\kappa} \right) - \sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(\sum_L C_L^{SA} R^{a,t}_{L,\nu\mu} P^t_{L,\tau\kappa} \right) \tag{78g}$$

$$+ \sum_{\mu,\nu} \sum_{\kappa,\tau} (\mu\nu|\kappa\tau)^\lambda \left(\frac{1}{2} g_{HF} \sum_L C_L^{SA} \left[R^{s,t}_{L,\kappa\mu} P^t_{L,\tau\nu} + R^{s,m}_{L,\kappa\mu} P^m_{L,\tau\nu} + R^{a,t}_{L,\kappa\mu} P^t_{L,\tau\nu} + R^{a,m}_{L,\kappa\mu} P^m_{L,\tau\nu} \right] \right) \tag{78h}$$

The second term in Eq. (78g) vanishes, as it implies summation with respect to the indices μ and ν over the whole range of the product of a symmetric and an anti-symmetric matrices. Then, the RT term becomes

$$= -\sum_{\mu,\nu}\sum_{\kappa,\tau}(\mu\nu|\kappa\tau)^{\lambda}\left(\sum_{L} C_L^{SA} R_{L,\nu\mu}^{s,t} P_{L,\tau\kappa}^{t}\right) \tag{78i}$$

$$+\sum_{\mu,\nu}\sum_{\kappa,\tau}(\mu\nu|\kappa\tau)^{\lambda}\left(\frac{1}{2}g_{HF}\sum_{L} C_L^{SA}\left[R_{L,\kappa\mu}^{s,t}P_{L,\tau\nu}^{t} + R_{L,\kappa\mu}^{s,m}P_{L,\tau\nu}^{m} + R_{L,\kappa\mu}^{a,t}P_{L,\tau\nu}^{t} + R_{L,\kappa\mu}^{a,m}P_{L,\tau\nu}^{m}\right]\right) \tag{78j}$$

As a matter of fact, the antisymmetric part of R does not make any contribution to the RT term either. So, it can be removed yielding

$$= -\sum_{\mu,\nu}\sum_{\kappa,\tau}(\mu\nu|\kappa\tau)^{\lambda}\left(\sum_{L} C_L^{SA} R_{L,\nu\mu}^{s,t} P_{L,\tau\kappa}^{t}\right) \tag{78k}$$

$$+\sum_{\mu,\nu}\sum_{\kappa,\tau}(\mu\nu|\kappa\tau)^{\lambda}\left(\frac{1}{2}g_{HF}\sum_{L} C_L^{SA}\left[R_{L,\kappa\mu}^{s,t}P_{L,\tau\nu}^{t} + R_{L,\kappa\mu}^{s,m}P_{L,\tau\nu}^{m}\right]\right) \tag{78l}$$

13 Coordinates of all species

13.1 C_2H_4 MEP from FC to MECI$_{tw-pyr}$

6
E = -78.256093917
C 0.0000000 0.0000000 -0.6596543
C 0.0000000 0.0000000 0.6596543
H 0.0000000 0.9172556 -1.2241830
H 0.0000000 -0.9172556 -1.2241830
H 0.0000000 -0.9172556 1.2241830
H 0.0000000 0.9172556 1.2241830
6
E = -78.277046438
C 0.0002117 -0.0000860 -0.7157623
C 0.0002331 0.0000068 0.7157272
H -0.0009942 0.9245042 -1.2383944
H 0.0007787 -0.9244458 -1.2385904
H -0.0010010 -0.9244330 1.2386022
H 0.0007717 0.9244538 1.2384178
6
E = -78.280832587
C -0.0056042 0.0002289 -0.7315354
C -0.0052927 -0.0004088 0.7316919
H -0.0437991 0.9270422 -1.2615479
H 0.0492576 -0.9264256 -1.2609961
H -0.0438418 -0.9271362 1.2619036
H 0.0492801 0.9266995 1.2604838
6
E = -78.284404832
C -0.0074259 0.0005219 -0.7294689
C -0.0069547 -0.0004955 0.7297858
H -0.0819882 0.9237793 -1.2693454
H 0.0890905 -0.9226890 -1.2671926
H -0.0819000 -0.9241347 1.2690077
H 0.0891779 0.9230181 1.2672134
6
E = -78.287007905
C -0.0072330 0.0006794 -0.7271423
C -0.0061797 -0.0007031 0.7277949
H -0.1034309 0.9174985 -1.2737832

H 0.1100178 -0.9167821 -1.2725276
H -0.1033646 -0.9182285 1.2738682
H 0.1101904 0.9175360 1.2717901
6
E = -78.290194961
C -0.0100253 0.0005009 -0.7233305
C -0.0095939 -0.0005754 0.7235644
H -0.1252546 0.9150777 -1.2776560
H 0.1349525 -0.9129746 -1.2743531
H -0.1251209 -0.9153026 1.2775422
H 0.1350416 0.9132742 1.2742330
6
E = -78.296837819
C -0.0075453 -0.0004435 -0.7159446
C -0.0049026 -0.0002033 0.7170716
H -0.1693542 0.9025301 -1.2837710
H 0.1749240 -0.9010693 -1.2824558
H -0.1687015 -0.9034808 1.2838771
H 0.1755794 0.9026662 1.2812230
6
E = -78.303918592
C -0.0065059 -0.0021640 -0.7088704
C -0.0006384 -0.0003798 0.7107807
H -0.2108958 0.8879129 -1.2880936
H 0.2131704 -0.8867276 -1.2919632
H -0.2101831 -0.8887001 1.2909537
H 0.2150529 0.8900584 1.2871928
6
E = -78.307925515
C -0.0058968 -0.0033916 -0.7051028
C 0.0021354 -0.0007156 0.7074915
H -0.2334818 0.8791335 -1.2896107
H 0.2336313 -0.8780832 -1.2975864
H -0.2328731 -0.8795526 1.2948121
H 0.2364853 0.8826097 1.2899961
6
E = -78.311993418
C -0.0053359 -0.0049716 -0.7016263
C 0.0052612 -0.0013434 0.7044833
H -0.2560552 0.8698099 -1.2900311
H 0.2538460 -0.8687602 -1.3034868

H -0.2557755 -0.8694299 1.2985901
H 0.2580593 0.8746953 1.2920707
6
E = -78.319354026
C -0.0040713 -0.0083667 -0.6958158
C 0.0113206 -0.0031942 0.6992672
H -0.2967988 0.8530287 -1.2872495
H 0.2900512 -0.8516283 -1.3132565
H -0.2982358 -0.8498482 1.3038475
H 0.2977343 0.8600092 1.2932072
6
E = -78.332100139
C 0.0017552 -0.0116185 -0.6886316
C 0.0205240 -0.0095631 0.6909766
H -0.3712109 0.8248214 -1.2762912
H 0.3588532 -0.8218648 -1.3215992
H -0.3797186 -0.8130033 1.3069655
H 0.3697973 0.8312286 1.2885797
6
E = -78.338136336
C 0.0051102 -0.0105847 -0.6859877
C 0.0219689 -0.0126827 0.6870481
H -0.4100175 0.8083809 -1.2729591
H 0.3977888 -0.8042271 -1.3198563
H -0.4220997 -0.7930077 1.3059995
H 0.4072496 0.8121216 1.2857559
6
E = -78.344125782
C -0.0011863 -0.0135234 -0.6791163
C 0.0175273 -0.0183092 0.6780915
H -0.4632970 0.7933448 -1.2750381
H 0.4617140 -0.7807303 -1.3234519
H -0.4769697 -0.7762533 1.3146558
H 0.4622115 0.7954716 1.2848593
6
E = -78.347685558
C 0.0012577 -0.0069739 -0.6718940
C 0.0162232 -0.0130366 0.6700714
H -0.4936161 0.7597091 -1.2782886
H 0.4920581 -0.7473630 -1.3047206
H -0.5042333 -0.7467448 1.3051271

H 0.4883103 0.7544094 1.2797052
6
E = -78.349195322
C 0.0008739 -0.0054398 -0.6774415
C 0.0149012 -0.0176876 0.6766520
H -0.5108103 0.7444694 -1.2807173
H 0.5136144 -0.7327288 -1.3103235
H -0.5210930 -0.7297532 1.3074521
H 0.5025139 0.7411401 1.2843780
6
E = -78.349874900
C 0.0048316 -0.0051198 -0.6792122
C 0.0151519 -0.0098415 0.6799897
H -0.5219412 0.7251017 -1.2887292
H 0.5249732 -0.7181483 -1.3150345
H -0.5310831 -0.7156259 1.3071405
H 0.5080672 0.7236339 1.2958462
6
E = -78.350510385
C 0.0097974 -0.0041438 -0.6730614
C 0.0140247 -0.0140508 0.6707644
H -0.5353488 0.7222488 -1.2839655
H 0.5353567 -0.7178591 -1.3094413
H -0.5430422 -0.7139878 1.2996769
H 0.5192121 0.7277926 1.2960270
6
E = -78.351293103
C -0.0034756 -0.0026233 -0.6773932
C 0.0049216 -0.0197055 0.6733208
H -0.5498450 0.7243557 -1.2934703
H 0.5631284 -0.7065837 -1.2988923
H -0.5612330 -0.7120514 1.3070031
H 0.5465039 0.7166081 1.2894319
6
E = -78.351944805
C -0.0008558 -0.0004897 -0.6764679
C 0.0021159 -0.0137840 0.6731143
H -0.5581971 0.7034179 -1.3008197
H 0.5734724 -0.6888749 -1.3013863
H -0.5689947 -0.6980836 1.3064140
H 0.5524596 0.6978148 1.2991459

6
E = -78.352262309
C 0.0052140 -0.0008200 -0.6753297
C 0.0039918 -0.0082224 0.6736398
H -0.5693527 0.6871993 -1.3015198
H 0.5819382 -0.6790026 -1.3054715
H -0.5775715 -0.6874871 1.3005722
H 0.5557804 0.6883331 1.3081093
6
E = -78.352544254
C -0.0004535 -0.0037557 -0.6764534
C -0.0009959 -0.0092885 0.6756313
H -0.5821368 0.6790133 -1.3056861
H 0.6012082 -0.6617910 -1.3114925
H -0.5885727 -0.6788936 1.3107942
H 0.5709509 0.6747156 1.3072065
6
E = -78.352671257
C 0.0047707 0.0011699 -0.6764979
C 0.0026905 -0.0055705 0.6752494
H -0.5963422 0.6628807 -1.3119292
H 0.6109907 -0.6556208 -1.3084525
H -0.6004093 -0.6677097 1.3027813
H 0.5782999 0.6648507 1.3188490
6
E = -78.352804654
C 0.0049482 0.0035988 -0.6759609
C -0.0004202 -0.0032843 0.6752180
H -0.5986685 0.6608462 -1.3100643
H 0.6184539 -0.6535704 -1.2976355
H -0.6056893 -0.6689596 1.2927849
H 0.5813761 0.6613699 1.3156575
6
E = -78.355304534
C -0.0059034 0.0130859 -0.6541966
C -0.1089868 0.0914918 0.6959304
H -0.6032784 0.6376686 -1.3350589
H 0.6457625 -0.6388795 -1.2657082
H -0.5674216 -0.6947715 1.2769096
H 0.6398278 0.5914051 1.2821233
6

E = -78.356833336
C -0.0105686 0.0165640 -0.6535804
C -0.1286826 0.1090210 0.6952058
H -0.6033913 0.6357050 -1.3398447
H 0.6508445 -0.6361778 -1.2494350
H -0.5548452 -0.6993727 1.2715506
H 0.6466430 0.5742607 1.2761035
6
E = -78.363028942
C -0.0182203 0.0222314 -0.6420023
C -0.1923780 0.1626868 0.7058905
H -0.6078387 0.6218989 -1.3549310
H 0.6696700 -0.6246938 -1.2286190
H -0.5330163 -0.7133556 1.2636438
H 0.6817830 0.5312324 1.2560178
6
E = -78.367365315
C -0.0244338 0.0297555 -0.6356718
C -0.2295092 0.2068448 0.7100343
H -0.6051109 0.6170373 -1.3621304
H 0.6663522 -0.6319221 -1.1927895
H -0.4981150 -0.7224248 1.2361157
H 0.6908169 0.5007093 1.2444414
6
E = -78.363414880
C -0.0418042 0.0602463 -0.6165287
C -0.2637051 0.2854024 0.7157149
H -0.6014700 0.5952094 -1.3905946
H 0.6608650 -0.6457601 -1.0794556
H -0.4475276 -0.7186897 1.1405689
H 0.6936422 0.4235917 1.2302955
6
E = -78.365254990
C -0.0439370 0.0754023 -0.6078028
C -0.2625136 0.3239900 0.7140794
H -0.6047091 0.5804387 -1.4050089
H 0.6553683 -0.6586505 -1.0393435
H -0.4408378 -0.7140064 1.0860230
H 0.6966296 0.3928259 1.2520530
6
E = -78.366878054

C -0.0460699 0.0905583 -0.5990769
C -0.2613221 0.3625776 0.7124439
H -0.6079483 0.5656680 -1.4194231
H 0.6498716 -0.6715408 -0.9992315
H -0.4341480 -0.7093232 1.0314770
H 0.6996169 0.3620600 1.2738105
6
E = -78.367840955
C -0.0364335 0.0763895 -0.5987467
C -0.2406120 0.3895329 0.7137685
H -0.6090139 0.5564317 -1.4114618
H 0.6472314 -0.6781197 -1.0160161
H -0.4529330 -0.7028252 1.0060137
H 0.6917609 0.3585904 1.3064430
6
E = -78.368755600
C 0.0370320 0.0845240 -0.6020582
C -0.1342463 0.4351000 0.7120406
H -0.5158826 0.5561831 -1.4073822
H 0.5825652 -0.8054956 -0.9513306
H -0.6157076 -0.6015916 0.7891220
H 0.6462394 0.3312800 1.4596087
6
E = -78.368714757
C 0.0422364 0.0721814 -0.6010374
C -0.1391317 0.4381717 0.7169812
H -0.4809687 0.5633952 -1.4110600
H 0.5906112 -0.8157266 -0.9506764
H -0.6409598 -0.6049662 0.7762760
H 0.6282128 0.3469449 1.4695165
6
E = -78.369270901
C -0.0100635 0.0828344 -0.6008909
C -0.1928723 0.4227540 0.7182734
H -0.5743813 0.5495075 -1.4101205
H 0.6265957 -0.7317121 -0.9757187
H -0.5251278 -0.6689393 0.8890089
H 0.6758492 0.3455555 1.3794478

13.2 TME S_1 MEP: from $S_{1,min}$ to $S_{1,TS}$

14
E = -233.049629
C -0.0000000 -0.0000000 0.8343992
C -0.0000000 -0.0000000 -0.6552107
C -0.3245031 1.2254564 1.3873061
C 0.3245031 -1.2254564 1.3873061
C 0.4178713 1.0797604 -1.3988424
C -0.4178713 -1.0797604 -1.3988424
H -0.3592604 1.3239830 2.4596013
H -0.9246457 1.9346586 0.8378908
H -0.4055216 -1.0642196 -2.4775489
H -0.7701179 -1.9630053 -0.8980014
H 0.4055216 1.0642196 -2.4775489
H 0.7701179 1.9630053 -0.8980014
H 0.3592604 -1.3239830 2.4596013
H 0.9246457 -1.9346586 0.8378908
14
E = -233.049615
C 0.0002554 -0.0001498 0.8324948
C 0.0008346 -0.0002610 -0.6569744
C -0.3281374 1.2233004 1.3872157
C 0.3279056 -1.2235767 1.3877938
C 0.4192435 1.0808345 -1.3984383
C -0.4170721 -1.0813536 -1.3986666
H -0.3641684 1.3207432 2.4595810
H -0.9233165 1.9352271 0.8361253
H -0.4078838 -1.0667251 -2.4773852
H -0.7736610 -1.9619829 -0.8962393
H 0.4044925 1.0683863 -2.4771140
H 0.7750613 1.9616395 -0.8957834
H 0.3637538 -1.3198810 2.4602748
H 0.9226924 -1.9362009 0.8371158
14
E = -233.049569
C -0.0016687 0.0001260 0.8308472
C -0.0018197 0.0003328 -0.6585163
C -0.3301487 1.2220971 1.3885757
C 0.3313107 -1.2217390 1.3875651
C 0.4185713 1.0820170 -1.3982344

C -0.4216565 -1.0814671 -1.3982305
H -0.3739338 1.3150914 2.4612025
H -0.9162660 1.9403156 0.8356209
H -0.3980418 -1.0743910 -2.4768566
H -0.7790578 -1.9607859 -0.8941194
H 0.4017057 1.0720068 -2.4768460
H 0.7795921 1.9599449 -0.8940725
H 0.3736324 -1.3149299 2.4603558
H 0.9177809 -1.9386188 0.8327085
14
E = -233.049526
C -0.0012585 -0.0000728 0.8288573
C -0.0010621 0.0000739 -0.6603195
C -0.3334776 1.2198682 1.3883838
C 0.3345538 -1.2196965 1.3880594
C 0.4203809 1.0829341 -1.3977431
C -0.4202591 -1.0833165 -1.3981436
H -0.3773101 1.3123839 2.4609853
H -0.9142990 1.9410257 0.8339919
H -0.4006962 -1.0768572 -2.4767859
H -0.7809606 -1.9606093 -0.8926657
H 0.3990296 1.0768756 -2.4763399
H 0.7838524 1.9588808 -0.8919171
H 0.3755604 -1.3114014 2.4609900
H 0.9159461 -1.9400882 0.8326471
14
E = -233.049471
C -0.0002174 0.0000207 0.8269775
C -0.0006815 0.0001291 -0.6621061
C -0.3365655 1.2178022 1.3885437
C 0.3389027 -1.2174334 1.3880665
C 0.4204157 1.0843817 -1.3975285
C -0.4197933 -1.0847805 -1.3978042
H -0.3801058 1.3095119 2.4612591
H -0.9131878 1.9413078 0.8331550
H -0.4046011 -1.0789125 -2.4764898
H -0.7833883 -1.9599374 -0.8906144
H 0.4024864 1.0788376 -2.4761873
H 0.7851597 1.9590569 -0.8904664
H 0.3766125 -1.3088670 2.4610715
H 0.9149636 -1.9411172 0.8321235

14
E = -233.049404
C 0.0000415 0.0001437 0.8250787
C -0.0015732 0.0004454 -0.6638652
C -0.3387253 1.2161311 1.3890055
C 0.3417581 -1.2154893 1.3879458
C 0.4201198 1.0858240 -1.3973947
C -0.4207266 -1.0858188 -1.3975187
H -0.3816748 1.3069354 2.4618226
H -0.9111726 1.9423552 0.8328775
H -0.4023467 -1.0833206 -2.4761130
H -0.7860232 -1.9595455 -0.8890791
H 0.4022651 1.0820852 -2.4760082
H 0.7870802 1.9588346 -0.8889653
H 0.3781100 -1.3063154 2.4609439
H 0.9128679 -1.9422651 0.8312701
14
E = -233.049327
C -0.0003004 0.0001429 0.8232503
C -0.0027324 0.0006758 -0.6655898
C -0.3413727 1.2142715 1.3893734
C 0.3442759 -1.2137208 1.3877941
C 0.4197062 1.0872144 -1.3970698
C -0.4210748 -1.0871609 -1.3974012
H -0.3828208 1.3048079 2.4621753
H -0.9092167 1.9434377 0.8327296
H -0.4031596 -1.0863649 -2.4759355
H -0.7875425 -1.9597895 -0.8877949
H 0.4043612 1.0843207 -2.4756955
H 0.7885097 1.9586569 -0.8872808
H 0.3794600 -1.3038617 2.4609043
H 0.9119068 -1.9426300 0.8305406
14
E = -233.049239
C -0.0010516 0.0002582 0.8213950
C -0.0048571 0.0012207 -0.6673407
C -0.3434189 1.2127316 1.3901095
C 0.3467014 -1.2119127 1.3872671
C 0.4181512 1.0888447 -1.3968610
C -0.4228747 -1.0880524 -1.3971717
H -0.3826775 1.3026364 2.4630530

H -0.9070694 1.9449516 0.8333464
H -0.4001775 -1.0911629 -2.4755650
H -0.7899639 -1.9597569 -0.8864328
H 0.4070289 1.0860438 -2.4754523
H 0.7889273 1.9587993 -0.8858971
H 0.3802609 -1.3019589 2.4603995
H 0.9110209 -1.9426427 0.8291501
14
E = -233.049147
C -0.0019324 0.0001679 0.8195546
C -0.0061716 0.0013932 -0.6690804
C -0.3454698 1.2110627 1.3906087
C 0.3489894 -1.2102039 1.3869998
C 0.4177742 1.0900262 -1.3965352
C -0.4232623 -1.0894915 -1.3970940
H -0.3825376 1.3007499 2.4636839
H -0.9045623 1.9464409 0.8334346
H -0.4004765 -1.0943102 -2.4754864
H -0.7908016 -1.9603589 -0.8853716
H 0.4086116 1.0881624 -2.4750960
H 0.7893205 1.9588829 -0.8844977
H 0.3795148 -1.3002118 2.4603099
H 0.9110036 -1.9423098 0.8285699
14
E = -233.049042
C -0.0024367 0.0001431 0.8177239
C -0.0073457 0.0015743 -0.6708166
C -0.3473606 1.2094612 1.3910411
C 0.3511311 -1.2085222 1.3867969
C 0.4176014 1.0911896 -1.3963077
C -0.4236218 -1.0909228 -1.3969807
H -0.3827971 1.2988123 2.4641696
H -0.9020242 1.9477670 0.8334560
H -0.3995094 -1.0979753 -2.4753124
H -0.7924639 -1.9605789 -0.8841307
H 0.4086344 1.0909284 -2.4747725
H 0.7904174 1.9588258 -0.8830962
H 0.3788907 -1.2986744 2.4600859
H 0.9108845 -1.9420281 0.8281434
14
E = -233.048939

C -0.0028846 -0.0001455 0.8158458
C -0.0072758 0.0012546 -0.6725286
C -0.3492010 1.2077682 1.3910167
C 0.3522906 -1.2071980 1.3870570
C 0.4193775 1.0916820 -1.3958683
C -0.4227016 -1.0926605 -1.3970398
H -0.3832484 1.2972009 2.4641208
H -0.8991176 1.9488243 0.8327563
H -0.3993226 -1.1009331 -2.4753250
H -0.7933709 -1.9610272 -0.8833049
H 0.4045675 1.0955487 -2.4742474
H 0.7924307 1.9584458 -0.8814795
H 0.3780690 -1.2966817 2.4603581
H 0.9103875 -1.9420786 0.8286386
14
E = -233.048829
C -0.0031399 -0.0003766 0.8140683
C -0.0067428 0.0009472 -0.6742168
C -0.3511402 1.2060102 1.3908751
C 0.3535072 -1.2058873 1.3873311
C 0.4212776 1.0922734 -1.3954816
C -0.4215753 -1.0943676 -1.3971094
H -0.3834408 1.2957209 2.4640448
H -0.8969082 1.9494452 0.8319868
H -0.4006625 -1.1032313 -2.4753567
H -0.7937328 -1.9616616 -0.8825838
H 0.4016934 1.0997222 -2.4736779
H 0.7939246 1.9585222 -0.8800344
H 0.3774847 -1.2946094 2.4606621
H 0.9094550 -1.9425075 0.8294924
14
E = -233.048709
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C -0.3532878 1.2042983 1.3908529
C 0.3550702 -1.2045882 1.3874315
C 0.4223032 1.0932603 -1.3950992
C -0.4215905 -1.0956543 -1.3970109
H -0.3838643 1.2940554 2.4640179
H -0.8957422 1.9496746 0.8314936
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H -0.7950527 -1.9618381 -0.8815814
H 0.4021904 1.1026030 -2.4732846
H 0.7948266 1.9589297 -0.8784176
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H 0.9085280 -1.9429792 0.8296184
14
E = -233.048590
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C -0.0060794 0.0007786 -0.6775855
C -0.3552421 1.2026192 1.3909391
C 0.3567114 -1.2032668 1.3874344
C 0.4227981 1.0945240 -1.3947776
C -0.4222577 -1.0964920 -1.3967720
H -0.3826464 1.2927829 2.4641627
H -0.8956706 1.9490139 0.8311953
H -0.4012070 -1.1088506 -2.4749650
H -0.7976190 -1.9612468 -0.8802956
H 0.4037550 1.1049290 -2.4729592
H 0.7941513 1.9600278 -0.8773344
H 0.3788760 -1.2903927 2.4610256
H 0.9069477 -1.9438696 0.8294632
14
E = -233.048457
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C 0.3582838 -1.2019855 1.3874224
C 0.4232050 1.0957971 -1.3943524
C -0.4229170 -1.0974732 -1.3965780
H -0.3810197 1.2917521 2.4641391
H -0.8955336 1.9487772 0.8308050
H -0.4007291 -1.1120131 -2.4746951
H -0.8001256 -1.9607622 -0.8791115
H 0.4056605 1.1072717 -2.4724988
H 0.7938966 1.9609373 -0.8757616
H 0.3799713 -1.2884638 2.4611117
H 0.9055910 -1.9445341 0.8291509
14
E = -233.048306
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C -0.0055616 0.0006136 -0.6809614

C -0.3608829 1.1987947 1.3908344
C 0.3606914 -1.2003864 1.3874458
C 0.4237253 1.0969867 -1.3939896
C -0.4231528 -1.0986842 -1.3963471
H -0.3817287 1.2901377 2.4641423
H -0.8951135 1.9488319 0.8302302
H -0.4033092 -1.1140848 -2.4744341
H -0.8018505 -1.9606932 -0.8776400
H 0.4084765 1.1091726 -2.4721706
H 0.7948985 1.9612576 -0.8739471
H 0.3813872 -1.2864816 2.4611436
H 0.9050225 -1.9447882 0.8286945
14
E = -233.048143
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C -0.0045236 0.0003179 -0.6826462
C -0.3640449 1.1967559 1.3905173
C 0.3629473 -1.1988652 1.3877102
C 0.4251686 1.0979807 -1.3934989
C -0.4224113 -1.1001150 -1.3961914
H -0.3817596 1.2890411 2.4637318
H -0.8954041 1.9482260 0.8293237
H -0.4069542 -1.1154872 -2.4742684
H -0.8032310 -1.9605492 -0.8763820
H 0.4099932 1.1117641 -2.4717451
H 0.7962010 1.9615361 -0.8720374
H 0.3825898 -1.2842201 2.4614572
H 0.9039361 -1.9454873 0.8287720
14
E = -233.047956
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C -0.0032095 0.0003342 -0.6843275
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C 0.3670889 -1.1969055 1.3876571
C 0.4254799 1.0994634 -1.3935819
C -0.4236645 -1.1007171 -1.3954550
H -0.3863467 1.2858247 2.4636924
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H -0.8073288 -1.9586751 -0.8739771
H 0.4121782 1.1139031 -2.4717403

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H 0.3862784 -1.2814916 2.4615828
H 0.9003340 -1.9478582 0.8280202
14
E = -233.047754
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C -0.0021081 0.0002381 -0.6860049
C -0.3679909 1.1929780 1.3903138
C 0.3700973 -1.1954987 1.3876626
C 0.4262495 1.1007228 -1.3934161
C -0.4251299 -1.1011623 -1.3949622
H -0.3937435 1.2817142 2.4636887
H -0.8937426 1.9476285 0.8281464
H -0.4072948 -1.1206251 -2.4730531
H -0.8127202 -1.9565363 -0.8716526
H 0.4159054 1.1153673 -2.4716077
H 0.7983105 1.9626165 -0.8697814
H 0.3950361 -1.2763991 2.4621210
H 0.8963027 -1.9504463 0.8267199
14
E = -233.047559
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C 0.3724781 -1.1937040 1.3875300
C 0.4262092 1.1020837 -1.3930683
C -0.4260418 -1.1021284 -1.3947435
H -0.3945782 1.2800230 2.4636675
H -0.8926767 1.9481014 0.8281986
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C 0.4417856 1.1353863 -1.3850604
C -0.4398871 -1.1360921 -1.3850575
H -0.4201067 1.2182136 2.4591670

H -0.8519428 1.9600536 0.8267887
H -0.4249815 -1.2097382 -2.4597684
H -0.8493625 -1.9604583 -0.8290909
H 0.4215031 1.2112460 -2.4594941
H 0.8504290 1.9601580 -0.8291806
H 0.4183169 -1.2185259 2.4595106
H 0.8529035 -1.9600232 0.8277558
14
E = -233.038025
C 0.0005297 0.0000407 0.7446912
C 0.0008661 -0.0001566 -0.7413426
C -0.4372265 1.1394227 1.3847799
C 0.4370125 -1.1396047 1.3852040
C 0.4412089 1.1361311 -1.3849877
C -0.4404485 -1.1360140 -1.3850122
H -0.4183010 1.2184234 2.4590032
H -0.8535011 1.9592041 0.8269071
H -0.4227288 -1.2112991 -2.4595640
H -0.8502212 -1.9600425 -0.8287571
H 0.4206396 1.2125702 -2.4593731
H 0.8518778 1.9597281 -0.8287928
H 0.4187709 -1.2178674 2.4594834
H 0.8515215 -1.9605360 0.8277607
14
E = -233.037889
C -0.0003547 -0.0004571 0.7440208
C 0.0012502 -0.0006691 -0.7420206
C -0.4398094 1.1381542 1.3841613
C 0.4369517 -1.1390361 1.3853406
C 0.4431753 1.1357568 -1.3844149
C -0.4393602 -1.1371454 -1.3851862
H -0.4241747 1.2158221 2.4586370
H -0.8522382 1.9596172 0.8260079
H -0.4294311 -1.2096795 -2.4600648
H -0.8514738 -1.9599049 -0.8287977
H 0.4244199 1.2129239 -2.4585948
H 0.8531973 1.9590702 -0.8273420
H 0.4266856 -1.2137810 2.4599009
H 0.8511620 -1.9606712 0.8283525
14
E = -233.037859

C 0.0008492 0.0001158 0.7437478
C 0.0006043 -0.0001185 -0.7422588
C -0.4383112 1.1384721 1.3846476
C 0.4391955 -1.1382003 1.3848243
C 0.4418304 1.1364277 -1.3849747
C -0.4408855 -1.1367092 -1.3846154
H -0.4268291 1.2138471 2.4592378
H -0.8508824 1.9601392 0.8269740
H -0.4276542 -1.2114244 -2.4592943
H -0.8528333 -1.9591431 -0.8275769
H 0.4261862 1.2119958 -2.4593451
H 0.8521939 1.9597967 -0.8282235
H 0.4250450 -1.2147422 2.4593081
H 0.8514911 -1.9604567 0.8275493
14
E = -233.037815
C 0.0000089 -0.0000037 0.7429956
C -0.0000089 0.0000037 -0.7429956
C -0.4403087 1.1374152 1.3846696
C 0.4403420 -1.1374292 1.3846475
C 0.4416850 1.1368954 -1.3846581
C -0.4417183 -1.1368814 -1.3846590
H -0.4219610 1.2151603 2.4589930
H -0.8497527 1.9608720 0.8273592
H -0.4234905 -1.2146487 -2.4589828
H -0.8521453 -1.9598421 -0.8273388
H 0.4234314 1.2146735 -2.4589808
H 0.8521254 1.9598505 -0.8273395
H 0.4220201 -1.2151851 2.4589706
H 0.8497726 -1.9608804 0.8273190

13.3 TCNQ-TME dyad, $S_{0,min}$ geometry

42
E = -2731.394002
S 6.9069569 -1.5357885 0.00000000
C 8.4692033 -0.7461856 0.00000000
C 8.4995341 0.5769835 0.00000000
S 6.9744367 1.4364203 0.00000000
C 5.9735806 -0.0278051 0.00000000

H 9.3390003 -1.3781631 0.00000000
H 9.3974215 1.1682558 0.00000000
C 4.6398969 0.0001434 0.00000000
S 3.7153050 1.5270570 0.00000000
C 2.1720014 0.7350040 0.00000000
C 2.1356086 -0.6041526 0.00000000
S 3.6477757 -1.4828504 0.00000000
C 0.7840657 1.2873659 0.00000000
C 0.7617222 -1.0514821 0.00000000
C -0.0565081 0.0276378 0.00000000
H 0.5873813 1.9026141 0.87715200
H 0.5873813 1.9026141 -0.87715200
C -1.4862483 0.0202086 0.00000000
C -2.3097541 1.1013727 0.00000000
C -3.6766736 0.6586509 0.00000000
C -3.7203492 -0.7157851 0.00000000
C -2.3126189 -1.2314083 0.00000000
H -2.0998808 -1.8489455 0.87047423
H -2.0998808 -1.8489455 -0.87047423
C -4.9080388 1.4428663 0.00000000
C -6.1490289 0.6857692 0.00000000
C -6.1667365 -0.6488964 0.00000000
C -4.9437771 -1.4433327 0.00000000
C -4.9701696 2.8052032 0.00000000
C -6.2278393 3.4864790 0.00000000
N -7.2348067 4.0388491 0.00000000
C -3.8452248 3.6821656 0.00000000
N -2.9663587 4.4216447 0.00000000
C -5.0505015 -2.8148431 0.00000000
C -3.9315888 -3.6932943 0.00000000
N -3.0347529 -4.4132019 0.00000000
C -6.3200737 -3.4617239 0.00000000
N -7.3455526 -3.9818479 0.00000000
H -7.1095361 -1.1677379 0.00000000
H -7.0766979 1.2305476 0.00000000
H -1.9849405 2.1216569 0.00000000
H 0.4555716 -2.0830643 0.00000000