Boundary values for the charge transferred during an electronic transition
Insights from matrix analysis

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In this contribution we start by proving and generalizing a conjecture that has been established few decades ago, relating the value of the integral of the detachment/attachment density in two pictures — one accounting for transition-induced basis relaxation and one which does not account for such a relaxation. To this end, we combine an extension to Cauchy’s interlacing theorem and a brother theorem that we bring in the paper. Using this result together with a corollary to Lidskii-Wielandt theorem allows us to derive an upper bound for the electronic charge that is effectively displaced during the molecular electronic transition from one quantum state to another. This quantity can be regarded as the neat charge that has been transferred during the transition. Our derivations show that this boundary value can be determined from a simple singular value decomposition and two trace operations.

Keywords: Molecular electronic transitions; electronic-structure theory; one-body reduced density matrices.

I. Introduction

Providing quantitative insights into the light-induced molecular electronic-structure reorganization is a challenging task that has seen plenty of contributions providing complementary pieces of information [1–15]. Those approaches are often coupled with a qualitative analysis for visualization purposes [6, 15–24]. In this regard, two pairs of one-electron reduced density functions have attracted the attention of the molecular electronic excited states community: the depletion/accumulation densities [15], obtained by separating the positive-valued and negative-valued density contributions to the one-electron reduced difference density — note that “depletion” and “accumulation” names are not really used, but the density functions are —, and the detachment/attachment densities [19, 24], obtained by a similarity transformation of the one-electron reduced difference density matrix performed in order to obtain and separate the negative and positive “transition occupation numbers” and to collect the related information into two one-electron reduced density matrices: the detachment and attachment density matrices — rigorous foundations for this methodology is provided in ref. [24]. The densities corresponding to these density matrices are visualized and analyzed as a “departure/arrival” density map, so that they can show some overlap, hence it is a picture different from the depletion/accumulation one. However, some relationships exist between the two pictures — for instance, the detachment/attachment density integral value is an upper bound to the depletion/accumulation density integral value.

When introducing the detachment/attachment picture [19], Head-Gordon and co-workers gave an interesting comment relatively to the integral of the detachment/attachment densities: when using the configuration interaction singles (CIS) method [25] — for which this integral is equal to unity in the native picture —, a post-transition account for a transition-induced basis relaxation results in adding a relaxation matrix to the “unrelaxed” one-body reduced difference density matrix. The authors postulated that incorporating such a relaxation effect into the analysis will, when producing the eigenvalues of the one-electron reduced difference density matrix, make “all negative eigenvalues becoming more negative and all positive eigenvalues becoming more positive. From the inter-leaving theorem of symmetric matrices, (...) the promotion number for the relaxed CIS density rigorously satisfies $p \geq 1$”. In the previous quote, $p$ is the detachment/attachment density integral — called promotion number; we will use another symbol for it in this contribution —, and the “interleaving theorem” is another name for Cauchy’s interlacing theorem (vide infra). This result has, to our knowledge, never been actually proved. Therefore, we would like to show in this contribution that not only this statement is true, but it can be extended to more sophisticated electronic excited-state calculation methods such as time-dependent Hartree-Fock theory [26], time-dependent density-functional theory [27], or the Bethe-Salpeter method [28].

On the other hand, in a paper published almost two decades ago [29], Furche and co-workers have mentioned, about the time-dependent density-functional theory one-electron reduced difference density matrix, that “in analogy to the ground state KS scheme, $P$ would yield the exact density difference if the exact (time-dependent) exchange-correlation functional were used. This follows from the fact that the density computed from $P$ is identical to the functional derivative of the excitation energy with respect to a local external potential.” Here, “KS” stands for “Kohn-Sham”, and $P$ is the relaxed one-electron reduced difference density matrix — another symbol for it will be used

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in this contribution.
According to some relationships that will be derived in this paper, we will see that it is possible to provide a theoretical value for the upper bound to the detachment/attachment density integral, hence the light-induced transferred density integral — the charge transferred during the molecular electronic transition.

Proofs to the linear-algebraic propositions that are reported in this contribution without providing a proof directly in the text can be found in refs [30–35].

II. Hypotheses, notations, and background

In what follows, \( n, m, N, M \) and \( L \) are non-zero natural numbers. In this contribution, we will deal with \( N \)-electron systems in the field of real numbers. In this contribution, we will deal with \( N \)-electron systems in the field of real numbers. In this contribution, we will deal with \( N \)-electron systems in the field of real numbers. In this contribution, we will deal with \( N \)-electron systems in the field of real numbers. In this contribution, we will deal with \( N \)-electron systems in the field of real numbers.

We start this section by recalling two important lemmas:

**Lemma II.1** Let \( A \) be an \( m \times n \) complex matrix and \( A^\dagger \) its adjoint. Then,

\[
A A^\dagger \geq 0,
\]

\[
A^\dagger A \geq 0,
\]

i.e., the product of a matrix by its adjoint is a positive semi-definite matrix.

**Lemma II.2** Let \( A \) and \( B \) be two \( n \times n \) complex matrices. Then,

\[
(A \geq 0 \text{ and } B \geq 0) \implies (A + B \geq 0),
\]

i.e., the sum of two positive semi-definite matrices is a positive semi-definite matrix.

In the case of excited-state calculation methods mentioned in the introduction section, the relaxed one-body reduced difference density matrix \( \gamma_{rlx} \) elements read, in the \( R \) basis,

\[
\forall \psi_0 \in [1, L]^2,
\]

\[
\gamma_{rlx}^{\underline{r}, \underline{s}}(\underline{r}, \underline{s}) := \left\langle \psi_0 \left| \left[ \hat{T}^{\underline{r}}, \left[ s_{\underline{r}}^{\underline{s}}, \hat{T} \right] \right] + \left[ s_{\underline{r}}^{\underline{s}}, \hat{Z} \right] \right| \psi_0 \right\rangle
\]

where \( \psi_0 \) is a wavefunction whose rows are single Slater determinants, and

\[
\hat{T} = \sum_{a=1}^{N} \sum_{a=N+1}^{L} x_{ia} a^{\dagger} a - y_{ia}^{\dagger} \hat{a},
\]

\[
\hat{Z} = \sum_{i=1}^{N} \sum_{a=N+1}^{L} z_{ia}^{\dagger} a - z_{ia} a^{\dagger} \hat{a}.
\]

In the expression of \( \hat{Z} \), the \( \ast \) symbol denotes complex conjugation; in the expression of \( \hat{T} \) and \( \hat{Z} \), we have that \( \hat{a}^{\dagger} \) and \( \hat{i}^{\dagger} \) respectively denote the second-quantization creation operators corresponding to the \( a^{\dagger} \) and \( i^{\dagger} \) spinorbitals, while \( \hat{a} \) and \( \hat{i} \) denote the second-quantization annihilation operators corresponding to the \( a^{\dagger} \) and \( i^{\dagger} \) spinorbitals. The \( x \), \( y \), and \( z \) vectors all belong to \( \mathbb{C}^{(N \times (L-N))} \). We can recast their vector components into matrices as follows: For every \((i, a)\) in \([1, N] \times [N + 1, L] \), we set

\[
X_{i,a} := x_{ia},
\]

\[
Y_{i,a} := y_{ia},
\]

\[
Z_{i,a} := z_{ia},
\]

and, using rules recapitulated in refs. [24, 36], we find that the relaxed one-body reduced difference density matrix is the simple sum of two matrices

\[
\gamma = \gamma_{rlx} \in \mathbb{C}^{L \times L}
\]

where the unrelaxed one-body reduced difference density matrix, \( \gamma_{rlx} \), is block-diagonal:

\[
\gamma := (C_1 \oplus C_2)
\]

with

\[
C_1 := XX^{\dagger} + YY^{\dagger},
\]

\[
C_2 := X^{\dagger}X + Y^{\dagger}Y.
\]
The case of CIS mentioned in the introduction of this article is simply obtained when the $Y$ matrix vanishes — a complete comment on the spectral decomposition of the unrelaxed CIS one-electron reduced difference density matrix in terms of the singular values and singular vectors of $X$ is given in Appendix VI.B. The $\gamma^Z$ matrix in (1) is an $L \times L$ Hermitian matrix, sum of two nilpotent matrices:

$$\gamma^Z = \begin{pmatrix} 0_o & Z \\ Z^\dagger & 0_o \end{pmatrix}$$

(3)

where $0_o$ and $0_o$ are the $(L - N) \times (L - N)$ and the $N \times N$ zero matrices, respectively. In (2), the $C_1$ and $C_2$ matrices are sums of normal matrices, hence they are both Hermitian. According to lemmas II.1 and II.2, $C_1$ is negative semidefinite while $C_2$ is positive semidefinite:

$$0 \geq C_1 \in \mathbb{C}^{N \times N},$$

(4)

$$0 \geq C_2 \in \mathbb{C}^{(L-N)\times(L-N)}$$

(5)

and $C_1$ and $C_2$ have the same trace, up to a sign:

$$-\text{tr}(C_1) = \text{tr}(C_2).$$

(6)

This number is of such importance in this contribution that it will be given a label using the $\vartheta$ symbol.

**B. Electronic transitions: density pictures**

Let $\Delta$ be a one-body reduced difference density matrix depicting the passage of an $N$-electron system from a given “state” to another, with a fixed geometry of the $M$ nuclei, and in the same basis. This passage can, for instance, be the electronic transition in the nuclei, and in the same basis. This passage from the unrelaxed to the relaxed picture of the electronic transition, and $\Delta$ is then simply equal to $\gamma^Z$. In any of these three cases, the $\Delta$ matrix is Hermitian.

Once this “departure/arrival” one-body reduced difference density matrix has been identified, one can use the reduced density matrix theory to find the basis in which the matrix representation of this “passage” is the most compact (i.e., diagonal). Such a unitary diagonalization produces the eigenvalues of the chosen $\Delta$ matrix:

$$\exists (P, Q) \in \mathbb{C}^{L \times L} \times \mathbb{R}^{L \times L}, P^\dagger \Delta P = Q$$

with

$$Q = \text{diag}(\lambda_i(\Delta))_{i \in [1,L]}.$$  

The diagonal entries of $Q$ are seen as the transition occupation numbers and are then sorted into two arrays of positive numbers:

$$Q_+ := \text{diag} \left( \frac{\lambda_i(\Delta) + \lambda_i(\Delta)}{2} \right)_{i \in [1,L]},$$

$$Q_- := \text{diag} \left( \frac{\lambda_i(\Delta) - \lambda_i(\Delta)}{2} \right)_{i \in [1,L]}.$$  

The original transformation matrix $P$ is then used to backtransform $Q_-$ and $Q_+$ in $F$, respectively producing the so-called one-particle detachment ($Q_D$) and attachment ($Q_A$) density matrices in $F$:

$$Q_D := PQ_- P^\dagger,$$

$$Q_A := PQ_+ P^\dagger,$$

with the property that

$$Q_A - Q_D = \Delta.$$  

(7)

Such a procedure applied to the unrelaxed difference density matrix produces the unrelaxed detachment and attachment density matrices that will be written $\gamma^d$ and $\gamma^a$, respectively. If the relaxed one-body reduced difference density matrix is diagonalized instead, the relaxed detachment and attachment density matrices will be written $\gamma^d_{rlx}$ and $\gamma^a_{rlx}$, respectively. Finally, if we are interested in deriving a detachment/attachment picture of the unrelaxed-relaxed transition process, it is the $\gamma^Z$ matrix that should be diagonalized to produce the relaxation detachment ($\gamma^d_{rlx}$) and attachment ($\gamma^a_{rlx}$) density matrices.

According to equations (4) and (5), we have that

$$\gamma^d = (-C_1) \oplus 0_o,$$

$$\gamma^a = 0_o \oplus C_2.$$  

We also notice that, according to equation (6), $\gamma^\Delta$, $\gamma^\Delta_{rlx}$, and $\gamma^Z$ are all zero trace. Therefore,

$$S_{\gamma}(\Delta) = -S_{\gamma}(\Delta).$$

Using equation (7), we deduce that, for a given $\Delta$ (i.e., either $\gamma^\Delta$, $\gamma^\Delta_{rlx}$, or $\gamma^Z$), the trace of the corresponding detachment density matrix will be equal to the trace of the corresponding attachment density matrix:

$$\text{tr}(Q_D) = \text{tr}(Q_A),$$  

(8)

and this number is equal to $S_{\gamma}(\Delta)$. For $\gamma^\Delta$, this number has already been labeled $\vartheta$; for $\gamma^\Delta_{rlx}$, this number will be labeled $\vartheta_{rlx}$; for $\gamma^Z$, this number will be labeled $\vartheta^Z$.

Any of these detachment/attachment density matrices, or the one-body reduced difference density matrix, can be used to build the corresponding detachment/attachment one-particle density function or one-electron difference density function, respectively. For instance, in the unrelaxed picture, these three functions read, in the $F$ basis,

$$n_\omega : S_4 \rightarrow I$$

$$s \mapsto n_\omega(s) = \sum_{r=1}^L \sum_{s=1}^L \langle \gamma^\omega \rangle_{r,s} \varphi_r(s) \varphi_s^\dagger(s)$$

where

$$S_4 := \mathbb{R}^3 \times \{|\uparrow\rangle, |\downarrow\rangle\},$$

and $\omega$ can be either $d$ or $a$ (in which case $I = \mathbb{R}_+$) or $\Delta$ (in which case $I = \mathbb{R}$).
The integral over $S_4$ of $n_d$ and $n_a$ is equal to the trace of the unrelaxed detachment and attachment density matrices:

$$\int_{S_4} ds \; n_d(s) = \int_{S_4} ds \; n_a(s) \quad \text{and} \quad \int_{S_4} ds \; n_a(s) = \vartheta,$$

and similarly for $\vartheta^{tx}$ and $\vartheta^z$. Note that this is not exactly a definite integral, but rather a sum over the discrete values for the spin projection, and an integral over $\mathbb{R}^3$.

We introduce another pair of density functions:

$$\begin{align*}
n_+ : S_4 & \longrightarrow \mathbb{R}_+ \\
s & \longmapsto n_+(s) = \frac{|n_\Delta(s)| + n_\Delta(s)}{2}, \\
n_- : S_4 & \longrightarrow \mathbb{R}_+ \\
s & \longmapsto n_-(s) = \frac{|n_\Delta(s)| - n_\Delta(s)}{2},
\end{align*}$$

The first one keeps only the positive entries of $n_\Delta$ on $S_4$ — i.e., the positive contributions to the difference density —, while the second one keeps only the absolute value of the negative contributions to the difference density. We have

$$\int_{S_4} ds \; n_+(s) = \int_{S_4} ds \; n_-(s).$$

Two numbers have then been introduced to assess the locality of the electronic transition in the unrelaxed and relaxed pictures. In the unrelaxed picture first, the dimensionless, normalized spatial overlap between the unrelaxed detachment and attachment densities is computed as [13]

$$\phi_S := \vartheta^{-1} \int_{S_4} ds \; \sqrt{n_d(s)n_a(s)},$$

while the neat displaced charge, i.e., the charge transferred during the electronic transition when one takes the before/after “bilingual” picture, reads [15]

$$0 \leq q^{CT} := \int_{S_4} ds \; n_+(s).$$

Due to (7), one can show that $(n_a - n_d = n_\Delta)$, hence

$$\forall s \in S_4,$$

$$n_a(s) \geq n_d(s) \iff |n_\Delta(s) = n_+(s)| \leq n_+(s) \leq n_a(s),$$

$$n_a(s) \leq n_d(s) \iff [-n_\Delta(s) = n_-(s)] \leq n_-(s) \leq n_d(s),$$

leading to

$$0 \leq q^{CT} \leq \vartheta. \quad (9)$$

Extension of $q^{CT}$ to the relaxed picture is straightforward, and gives what we will write $q^{CT}_{rlx}$, with

$$0 \leq q^{CT}_{rlx} \leq \vartheta^{rlx}, \quad (10)$$

due to the fact that (9) was established from the structure of the objects used for the construction of the $q^{CT}$ and $\vartheta$ numbers, independently from the nature of these objects.

III. Lower bound to the relaxed detachment/attachment density integral

In this section we will prove that the trace of the relaxed detachment/attachment density matrices, hence the integral over all the space of the relaxed detachment/attachment one-particle densities, is superior or equal to the unrelaxed one for the methods considered in this paper. We start by recalling that

**Lemma III.1 (Grassmann)** Let $U$ and $V$ be two linear subspaces of $\mathbb{C}^n$. Then,

$$\dim(U) + \dim(V) > n \iff \dim(U \cap V) > 0.$$ 

The following theorem is a known result from matrix analysis, and corresponds to the “interleaving theorem” invoked by Head-Gordon and co-workers in their conjecture [19]:

**Theorem III.1 (Cauchy’s interlacing)** Let $A$ be a bordered $n \times n$ Hermitian matrix partitioned as

$$A = \begin{pmatrix} B & y \\ y^\dagger & a \end{pmatrix},$$

where $a$ is real,

$$B \in \mathbb{C}^{(n-1)\times(n-1)},$$

and

$$y \in \mathbb{C}^{(n-1)\times 1}.$$ 

Let $\alpha^\dagger = (\alpha_1^\dagger, \ldots, \alpha_n^\dagger)$ and $\beta^\dagger = (\beta_1^\dagger, \ldots, \beta_{n-1}^\dagger)$ be the $n$–tuple of the eigenvalues of $A$ sorted in the increasing order and the $(n-1)$–tuple of the eigenvalues of $B$ sorted in increasing order, respectively. Then,

$$\alpha_1^\dagger \leq \beta_1^\dagger \leq \alpha_2^\dagger \leq \cdots \leq \beta_{n-1}^\dagger \leq \alpha_n^\dagger.$$ 

Its first natural extension, also known in the literature as “Cauchy’s generalized interlacing theorem”, is named “Eigenvalue embedding theorem I” below — see theorem III.2 —, for the eigenvalues do not really “interlace” anymore. Theorem III.2 is one of the two brother theorems we will introduce here. Both can be seen as a consequence of the separation theorem by Poincaré [31], i.e., in terms of compressions of Hermitian operators. For theorem III.2 we are interested in the compression of Hermitian operators to a given subspace; for theorem III.3 we are interested in the compression of the same Hermitian operator to the complementary space relatively to theorem III.2. The complementarity of the two theorems will then be used for deriving our first boundary value for the relaxed detachment/attachment trace. Note that since “Cauchy’s generalized interlacing theorem” is always introduced and proved in the shape of III.2, for pedagogical purposes we provide here a proof for the second compression, i.e., for theorem III.3 instead.

The restriction of the one-body reduced unrelaxed and relaxed difference density operators [24] to the linear span of the $L$–dimensional

$$F = (F_\sigma, F_\varphi)$$
basis of one-particle-state functions, $\hat{\gamma}^\Delta$ and $\hat{\gamma}_r^{\Delta_b}$, respectively have the $\gamma^\Delta$ and the $\gamma_r^{\Delta_b}$ matrix representations in $F$. Here,

$$ F_0 := (\varphi_i)_{i \in [1,N]}, $$

and

$$ F_v := (\varphi_i)_{i \in [N+1,N]}, $$

If we write $F, F_o, and F_v$ the linear span of $F, F_o, and F_v$, respectively, we have that $\hat{\gamma}^\Delta$ and $\hat{\gamma}_r^{\Delta_b}$ are two $F \rightarrow F$ maps, and that the compression of these two operators to $F_o$ using

$$ R_o : (F \rightarrow F) \rightarrow (F_o \rightarrow F_o), $$

$$ \hat{A} \mapsto \hat{C}_o \hat{A} \hat{C}_o, $$

has identical matrix representation ($C_1$) in $F_o$ when $\hat{A}$ is either $\hat{\gamma}^\Delta$ or $\hat{\gamma}_r^{\Delta_b}$ if the compression operator $\hat{C}_o$ has the rectangular

$$ M(\hat{C}_o, F, F_o) = \begin{pmatrix} I_o & \end{pmatrix}, $$

matrix representation, where $I_o$ is the $N \times N$ identity matrix, and $0_o$ is the $(L - N) \times N$ zero matrix.

The following theorem corresponds to this first compression:

**Theorem III.2 (Eigenvalue embedding I)** Let $A$ be an $n \times n$ Hermitian matrix partitioned as

$$ A = \begin{pmatrix} B & D \\ D^t & C \end{pmatrix}, $$

with

$$ B \in \mathbb{C}^{m \times m}, $$

assuming that $(m < n)$. Let $\alpha^\downarrow = (\alpha_1^\downarrow, \ldots, \alpha_n^\downarrow)$ and $\beta^\downarrow = (\beta_1^\downarrow, \ldots, \beta_m^\downarrow)$ be the $n$–tuple of the eigenvalues of $A$ sorted in the increasing order and the $m$–tuple of the eigenvalues of $B$ sorted in increasing order, respectively. Then,

$$ \forall i \in [1,(n - m)], \alpha_i^\downarrow \geq \beta_i^\downarrow \geq \alpha_i^\downarrow. $$

**Proof.** For the sake of readability, we set $V := [1,(n - m)]$.

We also rewrite the eigenvalues in a more convenient way for our proof:

$$ \forall i \in [1,n], \lambda_i^\downarrow(A) := \alpha_i^\downarrow, $$

and

$$ \forall i \in V, \lambda_i^\downarrow(C) := \gamma_i^\downarrow. $$

Let

$$ V^\downarrow := (v_1^\downarrow, \ldots, v_{n-m}^\downarrow) $$

be the $(n - m)$–tuple of eigenvectors of $C$ with their corresponding eigenvalues sorted in decreasing order

$$ \lambda_1^\downarrow(C) \geq \ldots \geq \lambda_{n-m}^\downarrow(C). $$

For every $p$ in $V$ we set

$$ V_p^\downarrow := \text{span}(v_1^\downarrow, \ldots, v_p^\downarrow) $$

and

$$ W_p^\downarrow := \left\{ \begin{pmatrix} 0_m \\ v \end{pmatrix} \in \mathbb{C}^{n \times 1} : v \in V_p^\downarrow \right\}, $$

where $0_m$ denotes the $m \times 1$ zero column vector. We have that

$$ \forall p \in V, \dim(W_p^\downarrow) = p. $$

If we denote by

$$ R^\downarrow := (r_1^\downarrow, \ldots, r_n^\downarrow) $$

the $n$–tuple of eigenvectors of $A$ with their respective eigenvalues being sorted in decreasing order

$$ \lambda_1^\downarrow(A) \geq \ldots \geq \lambda_n^\downarrow(A), $$

Theorem III.3 (Eigenvalue embedding II) Let $A$ be an $n \times n$ Hermitian matrix partitioned as

$$ A = \begin{pmatrix} B & D \\ D^t & C \end{pmatrix}, $$

with

$$ B \in \mathbb{C}^{m \times m}, $$

assuming that $(m < n)$. Let

$$ \alpha^\downarrow = (\alpha_1^\downarrow, \ldots, \alpha_n^\downarrow) $$

and

$$ \gamma^\downarrow = (\gamma_1^\downarrow, \ldots, \gamma_{n-m}^\downarrow) $$

be the $n$–tuple of the eigenvalues of $A$ sorted in the decreasing order and the $(n - m)$–tuple of the eigenvalues of $C$ sorted in decreasing order, respectively. Then,

$$ \forall i \in [1,(n - m)], \alpha_i^\downarrow \geq \gamma_i^\downarrow \geq \alpha_i^\downarrow. $$

**Proof.** For the sake of readability, we set $V := [1,(n - m)]$.
Then for every $p$ in $V$ we can set
\[ R_p := \text{span} \{ r_p^1, \ldots, r_p^n \}, \]
consider
\[ \lambda_p^1(A) = \max_{r \in R_p} R_A(r), \quad (13) \]
as well as
\[ \dim(R_p) = n - p + 1, \]
and conclude that
\[ \dim(R_p) + \dim(W_p^\perp) > n. \]
Therefore, according to lemma III.1, we find that, for every $p$ in $V$,
\[ \dim(R_p \cap W_p^\perp) \neq 0, \]
i.e.,
\[ \exists y \in R_p \cap W_p^\perp \ni y \neq 0_n. \]
With this in hand, we can conclude that for every $p$ in $V$ there exists at least one non-zero vector $y$ belonging simultaneously to $R_p$ and $W_p^\perp$. Since we know that
\[ \forall p \in V, y \in R_p \cap W_p^\perp \Rightarrow y \in W_p^\perp, \]
we know that such a $y$ vector must have the structure of the vectors belonging to $W_p^\perp$, i.e.,
\[ y := \begin{pmatrix} 0_m \\ y_v \end{pmatrix} \]
with $y_v \in V_p^\perp$, implying that, for such a $y$,
\[ R_A(y) = R_C(y_v). \]
Since, for any $p$ in $V$, we have that
\[ \forall y_v \in V_p^\perp, R_C(y_v) \leq \max_{v \in V_p^\perp} R_C(v) = \lambda_p^1(C), \]
and, according to equation (13),
\[ \forall y \in R_p, \lambda_p^1(A) = \max_{r \in R_p} R_A(r) \geq R_A(y) \]
we conclude that
\[ \forall p \in V, \lambda_p^1(A) \geq \lambda_p^1(C). \quad (14) \]
Applying (14) to $-\mathbf{A}$ instead of $\mathbf{A}$ gives
\[ \forall p \in V, \lambda_p^1(-\mathbf{A}) \geq \lambda_p^1(-\mathbf{C}), \]
i.e.,
\[ \forall p \in V, -\lambda_{n-p+1}(-\mathbf{A}) \geq -\lambda_{n-p+1}(-\mathbf{C}). \quad (15) \]
For every $p$ in $V$, setting
\[ t_p := \frac{n - m - p + 1}{n} \in V \]
turns (15) into
\[ \forall i_p \in V, \lambda_{i_p+m}^1(\mathbf{A}) \leq \lambda_{i_p}^1(\mathbf{C}). \quad (16) \]
Combining (14) with (16) gives
\[ \forall p \in V, \lambda_{p+m}^1(\mathbf{A}) \leq \lambda_p^1(\mathbf{C}) \leq \lambda_p^1(\mathbf{A}), \]
which completes the proof. \qed

For the sake of completeness, we introduce and prove the dual of theorem III.3:

**Theorem III.4 (Eigenvalue embedding II, dual)**

Let $\mathbf{A}$ be an $n \times n$ Hermitian matrix partitioned as
\[ \mathbf{A} = \begin{pmatrix} \mathbf{B} & \mathbf{D} \\ \mathbf{D}^\dagger & \mathbf{C} \end{pmatrix} \]
with
\[ \mathbf{B} \in \mathbb{C}^{m \times m}, \]
assuming that $(m < n)$. Let $V$ denote $[1, (n - m)]$. Let
\[ \alpha^1 := (\alpha^1_1, \ldots, \alpha^1_n) \]
and
\[ \gamma^1 := (\gamma^1_1, \ldots, \gamma^1_{n-m}) \]
be the $n$-tuple of the eigenvalues of $\mathbf{A}$ sorted in the increasing order and the $(n-m)$-tuple of the eigenvalues of $\mathbf{C}$ sorted in increasing order, respectively. Then,
\[ \forall i \in V, \alpha_{i+m} \geq \gamma_i \geq \alpha_i. \]
**Proof.** Without loss of generality, we insert
\[ \mathbf{A} = \mathbf{A}_1 \text{ and } \mathbf{A}_1 := -\mathbf{A}_2 \]
in theorem III.3, and write $\alpha^1_i, \alpha^2_i, \gamma^1_i, \text{ and } \gamma^2_i$ the tuples of eigenvalues defined in the same way as before but relatively to $\mathbf{A}_1$ and $\mathbf{A}_2$ and their respective blocks, i.e., with $i \in \{1, 2\}$. We have that
\[ \alpha^2_i = -\alpha^1_i, \]
\[ \gamma^2_i = -\gamma^1_i. \]
We now turn to the notations from (11) and (12), and write theorem III.3 for $\mathbf{A}_1$, i.e.,
\[ \forall i \in V, \lambda_i^1(\mathbf{A}_1) \geq \lambda_i^1(\mathbf{C}_1) \geq \lambda_{i+m}^1(\mathbf{A}_1). \]
Plugging the definition of $\mathbf{A}_1$ gives
\[ \forall i \in V, \lambda_i^1(-\mathbf{A}_2) \geq \lambda_i^1(-\mathbf{C}_2) \geq \lambda_{i+m}^1(-\mathbf{A}_2), \]
which, using the relationships between the tuples of eigenvalues above, gives
\[ \forall i \in V, -\lambda_i^1(\mathbf{A}_2) \geq -\lambda_i^1(\mathbf{C}_2) \geq -\lambda_{i+m}^1(\mathbf{A}_2), \]
i.e.,
\[ \forall i \in V, \lambda_i^1(\mathbf{A}_2) \leq \lambda_i^1(\mathbf{C}_2) \leq \lambda_{i+m}^1(\mathbf{A}_2), \]
which completes the proof. \qed

We now turn to the comparison of the trace of the relaxed and unrelaxed detachment/attachment density matrices. For this sake, we provide the following corollary to theorems III.2 and III.3:

**Corollary III.1** Let $\mathbf{A}_\pm$ be a zero-trace $n \times n$ Hermitian matrix partitioned as
\[ \mathbf{A}_\pm = \begin{pmatrix} \mathbf{B} & \mathbf{D} \\ \mathbf{D}^\dagger & \mathbf{C} \end{pmatrix} \]
with

\[ 0 \geq B \in \mathbb{C}^{n \times m} \text{ and } 0 \preceq C \in \mathbb{C}^{(n-m) \times (n-m)}, \]

assuming that \( m < n \). Let \( A_- \) be the zero-trace \( n \times n \) Hermitian matrix defined as

\[ A_- = B \oplus C. \]

Then,

\[ S_+(A_+) \geq S_+(A_-). \]

**Proof.** For the sake of readability, we will denote again \([1, (n-m)]\) by \( V \). Let

\[ v^+ := \left(v^+_p\right)_{p \in V} \]

denote the \((n-m)\)-tuple of the eigenvalues of \( C \) listed in decreasing order. Let

\[ t^+ := \left(t^+_p\right)_{p \in [1, n]} \]

denote the \( n\)-tuple of the eigenvalues of \( A_+ \) listed in decreasing order, and let

\[ k^+ := \left(k^+_p\right)_{p \in [1, n]} \]

denote the \( n\)-tuple of the eigenvalues of \( A_- \) listed in decreasing order. Then, according to the hypotheses and to theorem III.3, we have that

\[ (\forall p \in V, v^+_p \geq 0) \implies (\forall p \in V, t^+_p \geq 0). \]

Since \( \forall p \in [1, m], t^+_p = t^+_m = 0 \), we find that, according to the hypotheses and to theorem III.2,

\[ \forall p \in [n - m + 1, n], t^+_p \leq 0, \]

and we deduce that

\[ \sum_{p=1}^{n-m} t^+_p \geq \sum_{p=1}^{n-m} v^+_p, \]

can be used to conclude that

\[ \sum_{p=1}^{n} \max(t^+_p, 0) \geq \sum_{p=1}^{n} \max(k^+_p, 0), \]

i.e.,

\[ S_+(A_+) \geq S_+(A_-), \]

which completes the proof. \( \square \)

Applying corollary III.1 to \( \gamma^{rlx} \) as \( A_+ \) and \( \gamma^A \) as \( A_- \), we finally obtain the following important inequality

\[ \vartheta^{rlx} \geq \vartheta, \quad (17) \]

proving that the unrelaxed detachment/attachment density integral is a lower bound to the relaxed one. In particular, setting \( Y \) equal to zero in \( C_1 \) and \( C_2 \) proves the conjecture of Head-Gordon and co-workers relatively to the configuration interaction singles method.

### IV. Upper bound to the relaxed detachment/attachment density integral

In this section we will first prove that the sum of the positive eigenvalues of \( \gamma^Z \) is equal to the sum of the singular values of \( Z \), and we will combine this result to an important result from linear algebra to derive the upper boundary value to the charge transferred during a molecular electronic transition.

Setting

\[ q := \min(N, L - N), \]

the singular value decomposition of the \( Z \) matrix from \( \gamma^Z \) reads

\[ Z = SHU^\dagger \in \mathbb{C}^{N \times (L - N)} \]

where \( S \in \mathbb{C}^{N \times N} \) and \( U \in \mathbb{C}^{(L-N) \times (L-N)} \). The matrix containing the singular values,

\[ H \in \mathbb{R}^{N \times (L - N)}, \]

is diagonal, i.e.,

\[ \forall (i, j) \in [1, N] \times [1, (L - N)], (i \neq j) \implies (H)_{i,j} = 0. \]

We set

\[ \forall p \in [1, q], h_p := (H)_{p,p}. \]

We also have that \( S^\dagger = S^{-1} \) and \( U^\dagger = U^{-1}. \)

The square of \( \gamma^Z \) can also be written

\[ (\gamma^Z)^2 \gamma^Z = ZZ^\dagger \oplus Z^\dagger Z \]

and commutes with \( \gamma^Z \). One eigenvalue decomposition of \( (\gamma^Z)^2 \gamma^Z \) reads

\[ G^\dagger (ZZ^\dagger \oplus Z^\dagger Z) G = HH^\dagger \oplus H^\dagger H, \]

with

\[ G = S \oplus U, \]

so the squared \( \ell^2 \)-norm of \( z \) is simply

\[ z^2 = \sum_{i=1}^{q} h_i^2. \]

Each non-zero eigenvalue in \( HH^\dagger \oplus H^\dagger H \) is degenerate with a multiplicity of two.

Since \( ZZ^\dagger \oplus Z^\dagger Z \) commutes with \( \gamma^Z \), it is possible to build an orthonormal basis with vectors that are eigenvectors of both matrices. To construct this basis, we first reconsider \( S \) as an \( N \)-tuple of orthonormalized vectors

\[ S := (s_i)_{i \in [1, N]}, \]

\( U \) as an \( (L - N) \)-tuple of orthonormalized vectors

\[ U := (u_a)_{a \in [1, (L - N)]}, \]

so we can finally rewrite \( G \) as an \( L \)-tuple of vectors:

\[ G := (s'_1, \ldots, s'_N, u'_1, \ldots, u'_{L-N}), \]
with
\[ \forall p \in [1, N], \ s_p := \left( \frac{s_p}{L-N} \right), \]
and
\[ \forall p \in [1, (L-N)], \ u_p^* := \left( \frac{0_{N}}{u_p} \right). \]

The vectors in \( S \) and \( U \) are ordered so that
\[ \forall p \in [1, q], \ Z u_p = h_p s_p, \quad Z^T s_p = h_p u_p. \]

We now construct
\[ C = (c_1^+, \ldots, c_q^+, c_1^-, \ldots, c_q^-) \]
with
\[ \forall p \in [1, q], \quad c_p^+ = \frac{s_p^+ + u_p^*}{\sqrt{2}}, \]
\[ c_p^- = \frac{s_p^+ - u_p^*}{\sqrt{2}}, \]
and, for the last \( L - 2q \) columns,
\[ C' := (c_1^+, \ldots, c_{L-2q}^+) \]
with,
\[ \forall p \in [1, L-2q], \quad c_p' \in \{ c \in C^{L \times 1} : c \in \ker (\gamma^Z), \ c \cdot C = 1 \}. \]

We can see that the first \( 2q \) vectors in \( C \) are eigenvectors of both \( \gamma^Z \) and of its second power.

We define five matrices:
\[ H_2 := \text{diag} (h_1^2, \ldots, h_q^2), \quad H_+ := \text{diag} (h_1, \ldots, h_q), \quad H_- := -H_+, \]
as well as \( 0_q \) and \( 0_{L-2q} \) that are \( q \times q \) and the \((L-2q) \times (L-2q)\) zero matrices, respectively.

Case 1. \((L - N \neq N)\)

We have
\[ C^\dagger (\gamma^Z)^\dagger \gamma^Z C = H_2 \oplus H_2 \oplus 0_{L-2q}, \]
and
\[ C^\dagger \gamma^Z C = H_+ \oplus H_- \oplus 0_{L-2q}. \]

Case 2. \((L - N = N)\)

We have
\[ C^\dagger (\gamma^Z)^\dagger \gamma^Z C = H_2 \oplus H_2, \]
and
\[ C^\dagger \gamma^Z C = H_+ \oplus H_- \]

In Case 1 as well as in Case 2, we see that the singular value decomposition of the \( Z \) matrix fully determines the detachment and attachment density matrices related to the relaxation process: in Case 1 we have
\[ \gamma^d_c = C (0_q \oplus H_+ \oplus 0_{L-2q}) C^\dagger, \]
\[ \gamma^a_c = C (H_+ \oplus 0_q \oplus 0_{L-2q}) C^\dagger, \]
while in Case 2 we have
\[ \gamma^d_c = C (0_q \oplus H_+) C^\dagger, \]
\[ \gamma^a_c = C (H_+ \oplus 0_q) C^\dagger, \]
The integral of the \( Z \)-vector-related detachment/attachment densities is then
\[ \varrho^Z = \sum_{p=1}^{q} h_p. \]

We introduce the Lidskii-Wielandt theorem:

**Theorem IV.1 (Lidskii-Wielandt)** Let \( A \) and \( B \) be two \( n \times n \) Hermitian matrices. Let \( C \) be their sum. Let
\[ \alpha^+ = (\alpha_1^+, \ldots, \alpha_n^+), \quad \beta^+ = (\beta_1^+, \ldots, \beta_n^+), \quad \gamma^+ = (\gamma_1^+, \ldots, \gamma_n^+), \]
be the decreasing-order \( n \)-tuples of the eigenvalues of \( A, B, \) and \( C \) respectively. Then, for any choice of \( 1 \leq i_1 < \cdots < i_k \leq n, \)
\[ \sum_{j=1}^{k} (\gamma_j^{i_j} - \alpha_j^{i_j}) \leq \sum_{j=1}^{k} \beta_j^{i_j}. \]

One corollary to the Lidskii-Wielandt theorem is

**Corollary IV.1** Let \( A, B, C, \alpha^+, \beta^+, \) and \( \gamma^+ \) be the matrices and \( n \)-tuples from theorem IV.1. Then, the \( \gamma^+ \) \( n \)-tuple is majorized by \( (\alpha^+ + \beta^+) \), i.e.,
\[ \forall k \in [1, n], \sum_{j=1}^{k} \gamma_j^i \leq \sum_{j=1}^{k} (\alpha_j^i + \beta_j^i) \]
and
\[ \sum_{j=1}^{n} \gamma_j^i = \sum_{j=1}^{n} (\alpha_j^i + \beta_j^i). \]

A rapid case study \([N < (L-N)]: N = (L-N); N > (L-N)\] shows that, if we consider the \( \gamma^Z \) eigenvalues matrix with eigenvalues sorted in decreasing order, i.e., in Case 1,
\[ \lambda^i (\gamma^Z) = H_+^i \oplus 0_{L-2q} \oplus H_-^i, \]
(where the definition of \( H_+^i \) and \( H_-^i \) is obvious), and in Case 2
\[ \lambda^i (\gamma^Z) = H_+^i \oplus H_-^i, \]
the following propositions are always true:
\[
\forall i \in [1, L - N], \lambda_i^A(\gamma^Z) \geq 0, \\
\forall i \in [1, N], \lambda_i^{L-N+1}(\gamma^Z) \leq 0.
\]
Therefore, choosing \( A = \gamma^A, B = \gamma^Z, C = \gamma^{L-N+1} \), and setting \( (k = L - N) \) in corollary IV.1 gives, together with (17),
\[
\vartheta \leq \vartheta^{RLx} \leq \vartheta + \vartheta^Z,
\]
hence, according to (10),
\[
0 \leq \varrho_{RLx}^{CT} \leq \vartheta + \varrho^Z,
\]
which is our final result.

V. Conclusion

We have first revisited, proved and extended a long-lived conjecture related to the relationship between the relaxed and unrelaxed detachment/attachment density integral values. Then, we have applied the detachment/attachment picture to the basis relaxation transformation, picturing such a transformation using a departure/arrival situation, where the unrelaxed excited state is the departure state, and the relaxed excited state is the arrival state — leaving the ground state unperturbed by the basis relaxation, so that the basis-relaxation departure/arrival picture is then extended to the ground/excited state difference since the ground state will remain identical in the relaxed and unrelaxed picture, canceling out when considering the relaxed/unrelaxed difference. The detachment/attachment density matrices related to the unrelaxed-to-relaxed passage are then derived, and their trace is shown to be simply equal to the sum of the singular values of the basis-relaxation matrix. Finally, we have derived the upper boundary value to the exact value of the charge transferred during a light-induced molecular electronic transition as the sum of the unrelaxed detachment/attachment density matrix trace and the basis-relaxation detachment/attachment density matrix trace, using matrix analysis. The final inequality shows that the excitation and relaxation processes do not result in simply additive contributions to the fully relaxed detachment/attachment picture of the electronic state transition.

VI. Appendices

A. Basic results from linear algebra

Lemma VI.1 Let \( A \) be an \( m \times n \) complex, normal matrix (i.e., commuting with its conjugate transpose \( A^\dagger \)). Let \( p \) be any strictly positive integer lower or equal to \( \min(m,n) \). Then, the \( p \)th left-singular vector \( u_p \in \mathbb{C}^{m\times 1} \) of \( A \) corresponds to the same singular value \( \sigma_p \) as the \( p \)th right-singular vector \( v_p \in \mathbb{C}^{n\times 1} \) of \( A \).

Lemma VI.1 also reads
\[
A^\dagger u_p = \sigma_p v_p.
\]

This lemma simply states the existence of a singular value decomposition for any normal rectangular matrix, and is given here without proof. We also give

Lemma VI.2 Let \( A \) be an \( m \times n \) complex matrix and \( A^\dagger \) its adjoint. Let \( q \) be the strictly positive integer equal to \( \min(m,n) \). Then, \( AA^\dagger \) and \( A^\dagger A \) share \( q \) eigenvalues. Those eigenvalues are the squared singular values of \( A \).

Proof. Let \( p \) be any strictly positive integer lower or equal to \( g = \min(m,n) \). Then, multiplying equation (18) to the left by \( A^\dagger \) leads, according to equation (19), to
\[
A^\dagger A v_p = \sigma_p^2 v_p,
\]
while multiplying equation (19) to the left by \( A \) leads, according to equation (18), to
\[
A A^\dagger u_p = \sigma_p^2 u_p,
\]
\( \square \)

B. Comments on the CIS case

In the case of the CIS method (i.e., when \( Y \) is the \( N \times (L - N) \) zero matrix), the spectral decomposition of the one-body reduced unrelaxed difference density matrix can be rewritten using singular vectors and singular values of \( X \). We write \( K \) the general diagonal matrix of eigenvalues of the CIS one-body reduced unrelaxed difference density matrix, and \( M \) its general matrix of eigenvectors. Lemmas VI.1 and VI.2 are prerequisites for this appendix. We will discuss how we can sort the eigenvalues and eigenvectors in three cases \( |N < (L - N)|; N = L - N; N > (L - N)| \), but before that, we write the singular value decomposition of \( X \):
\[
\exists (O, V, \lambda) \in \mathbb{C}^{N \times N} \times \mathbb{C}^{(L-N) \times (L-N)} \times \mathbb{R}_+^{N \times (L-N)},
\]
\( O X V = \lambda \),
with
\[
\forall (i,j) \in [1, N] \times [1, (L - N)], (i \neq j) \implies (\lambda)_{i,j} = 0.
\]

Before dealing with the ordering of the eigenvalues in \( K \), we re-define the \( O \) matrix as an \( N \)-tuple of vectors
\[
O = (o_1, \ldots, o_N)
\]
and \( V \) as an \( (L - N) \)-tuple of vectors
\[
V = (v_1, \ldots, v_{L-N}).
\]
In this comment we will discuss three possibilities that can be encountered:
Case 1: $N < (L - N)$

We define
\[ \Lambda := \lambda \lambda^t \in \mathbb{R}_+^{N \times N}, \]
(i.e., an $N$–tuple of column vectors in which the column vectors are ordered such that \( \lambda \in \mathbb{R}_+ \)).

We have that
\[ \lambda^t \lambda = \Lambda \oplus 0_N \]
with $0_N$ being the $(L - 2N) \times (L - 2N)$ zero matrix. According to
\[ \forall p \in [N + 1, L - N], \ v_p \in \ker(\mathbf{X}), \]
where the kernel of $\mathbf{X}$ is defined as
\[ \ker(\mathbf{X}) = \left\{ \mathbf{w} \in \mathbb{C}^{(L - N) \times 1} : \mathbf{Xw} = \mathbf{0}_N \right\}, \]
with $0_N$ being the $N \times 1$ zero column vector, we define two matrices: the first one is
\[ \mathbf{V}_1^\uparrow := (\mathbf{v}_1^\uparrow, \ldots, \mathbf{v}_N^\uparrow), \]
i.e., an $N$–tuple of vectors in which the column vectors are ordered such that
\[ \mathbf{X}^\uparrow \mathbf{X} \mathbf{V}_1^\uparrow = \mathbf{V}_1^\uparrow \Lambda^\uparrow, \]
where
\[ \Lambda^\uparrow := \text{diag} (\lambda_1^2, \ldots, \lambda_N^2). \]
The second matrix is
\[ \mathbf{V}_{\ker} := (\mathbf{v}_{N+1}, \ldots, \mathbf{v}_{L-N}). \]
While we identify $\mathbf{V}$ as $(\mathbf{V}_O, \mathbf{V}_{\ker})$ where $\mathbf{V}_O$ is the $N$–tuple of column vectors
\[ \mathbf{V}_O = (\mathbf{v}_i)_{i \in [1,N]}, \]
we can permute columns of $\mathbf{V}$ so that the column vectors belonging to $\ker(\mathbf{X})$ appear first
\[ \mathbf{V}_\triangledown := \left( \mathbf{V}_{\ker}, \mathbf{v}_1^\uparrow \right). \]
We then have that
\[ \left( \mathbf{X}^\uparrow \mathbf{X} \right) \mathbf{V}_\triangledown = \mathbf{V}_\triangledown (0_N \oplus \Lambda^\uparrow). \]
We also define $\Omega^\downarrow$ and $\Omega^\uparrow$ such that
\[ \forall \omega \in \{\uparrow, \downarrow\}, \ \mathbf{X}^\uparrow \mathbf{X} \Omega^\omega = \Omega^\omega (\Lambda \oplus 0_N) \]
and we finally define $\mathbf{V}_\downarrow^\uparrow$ such that
\[ \mathbf{X}^\uparrow \mathbf{X} \downarrow^\uparrow = \mathbf{V}_\downarrow^\uparrow (\Lambda^\downarrow \oplus 0_N). \]
This allows us to write the matrix of eigenvalues of $\gamma_{\mathbf{X}}^{\uparrow}$ sorted in a decreasing order as
\[ \mathbf{K}_1^\uparrow := \Omega^\uparrow \oplus 0_N \oplus (-\Lambda^\uparrow) \]
with the corresponding matrix of eigenvectors:
\[ \mathbf{M}_1^\uparrow := \begin{pmatrix} 0_N & \Omega^\uparrow \\ \Lambda^\uparrow & 0_N \end{pmatrix}. \]
On the other hand, the matrix of eigenvalues of $\gamma_\mathbf{X}$ sorted in an increasing order reads
\[ \mathbf{K}_1^\downarrow := (-\Lambda^\downarrow) \oplus 0_N \oplus \Lambda^\uparrow \]
with the corresponding matrix of eigenvectors:
\[ \mathbf{M}_1^\downarrow := \Omega^\downarrow \oplus \mathbf{V}_\triangledown. \]

Case 2: $N > (L - N)$

We define
\[ \Omega := \lambda \lambda^t \in \mathbb{R}_+^{(L-N) \times (L-N)}, \]
i.e.,
\[ \text{diag}(\lambda_1, \ldots, \lambda_N) = \text{diag}(\lambda_1^2, \ldots, \lambda_N^2). \]
We have that
\[ \lambda^t \lambda = \Omega \oplus 0_2 \]
with $0_2$ being the $(2N - L) \times (2N - L)$ zero matrix. According to
\[ \forall p \in [L - N + 1, N], \ o_p \in \text{coker}(\mathbf{X}), \]
where the cokernel of $\mathbf{X}$ is defined as
\[ \text{coker}(\mathbf{X}) = \left\{ \mathbf{w} \in \mathbb{C}^{N \times 1} : \mathbf{x}^\uparrow \mathbf{w} = \mathbf{0}_{L-N} \right\}, \]
with $0_{L-N}$ is the $(L-N) \times 1$ column vector, we define two matrices:
\[ \mathbf{O}_2 := (\mathbf{o}_1^\downarrow, \ldots, \mathbf{o}_{L-N}^\uparrow), \]
i.e., an $(L-N)$–tuple in which the column vectors are ordered such that
\[ \mathbf{X}^\uparrow \mathbf{X} \mathbf{O}_2^\downarrow = \mathbf{O}_2^\downarrow \Omega^\uparrow, \]
where
\[ \Omega^\uparrow = \text{diag} (\Omega_1^\downarrow, \ldots, \Omega_{L-N}^\uparrow). \]
The second matrix is
\[ \mathbf{O}_{\text{coker}} := (\mathbf{o}_{L-N+1}, \ldots, \mathbf{o}_N). \]
We now permute the columns of $\mathbf{O}$ such that the column vectors belonging to the cokernel of $\mathbf{X}$ appear first
\[ \mathbf{o}_\triangledown := \left( \mathbf{O}_{\text{coker}}, \mathbf{O}_2^\downarrow \right). \]
We then have that
\[ \left( \mathbf{X}^\uparrow \mathbf{X} \right) \mathbf{o}_\triangledown = \mathbf{o}_\triangledown (0_2 \oplus \Omega^\uparrow). \]
We also define $\mathbf{V}_2^\downarrow$ and $\mathbf{V}_2^\uparrow$ such that
\[ \forall \omega \in \{\uparrow, \downarrow\}, \ \mathbf{X}^\uparrow \mathbf{X} \mathbf{V}_2^\omega = \mathbf{V}_2^\omega \Omega^\omega \]
and $O_2^\dagger$ such that
\[
XX^\dagger O_2^\dagger = O_2^\dagger (\Omega^\dagger \oplus 0_3).
\]
(54)
This allows us to write the matrix of eigenvalues of $\gamma_{\text{CIS}}$ sorted in a decreasing order as
\[
K_2^\dagger = \Omega^\dagger \oplus 0_2 \oplus (-\Omega^\dagger)
\]
(55)
with the corresponding matrix of eigenvectors:
\[
M_2^\dagger := \begin{pmatrix}
0_o & O_2^\dagger \\
V_2^\dagger & 0_v
\end{pmatrix}.
\]
(56)
On the other hand, the matrix of eigenvalues of $\gamma_{\text{CIS}}$ sorted in an increasing order reads
\[
K_2^\dagger = (-\Omega^\dagger) \oplus 0_2 \oplus \Omega^\dagger
\]
(57)
with the corresponding matrix of eigenvectors:
\[
M_2 := O_2^\dagger \oplus V_2^\dagger.
\]
(58)
Case 3: $N = L - N$
We have that
\[
\Gamma := \lambda \lambda^\dagger = \lambda^\dagger \lambda,
\]
(59)
with
\[
\Gamma = \text{diag}(\Gamma_1, \ldots, \Gamma_N) = \text{diag}(\lambda_1^2, \ldots, \lambda_N^2).
\]
(60)
We have
\[
\forall \omega \in \{\uparrow, \downarrow\}, XX^\dagger O_3^\omega = O_3^\omega \Gamma^\omega
\]
(61)
and
\[
\forall \omega \in \{\uparrow, \downarrow\}, X^\dagger XX^\dagger V_3^\omega = V_3^\omega \Gamma^\omega
\]
(62)
so we can write
\[
K_3^\dagger = (-\Gamma^\dagger) \oplus \Gamma^\uparrow
\]
(63)
with the corresponding matrix of eigenvectors
\[
M_3^\dagger := O_3^\dagger \oplus V_3^\dagger.
\]
(64)
We can also write
\[
K_3^\dagger = \Gamma^\uparrow \oplus (-\Gamma^\dagger)
\]
(65)
with the corresponding matrix of eigenvectors:
\[
M_3 := \begin{pmatrix}
0_o & O_3^\dagger \\
V_3^\dagger & 0_v
\end{pmatrix}.
\]
(66)

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